July 28-August 2, 2019 Shanghai China

FEMTO 14

The 14th Femtochemistry Conference
- Dynamics of the Complexity in Chemistry, Biology, and Physics

CONFERENCE PROGRAM

MAPS

锦北楼 Cathay Building

1F	梦咖啡	Dream Cafe
1F	自选商场	Supermaket
2F-10F	客房	Guest Room
11F	锦江厅	JinJiang Ballroom
11F	老夜上海餐厅	Old shanghai Moon Restaurant
14F	锦福会中餐厅	Fortune Palace

贵宾楼 Grosvenor House

G	茗苑	Tea House
1F-16F	客房	Guest Room
17-18F	贵宾俱乐部	VIP Club

锦江小礼堂 Jin Jiang Grand Hall

1F 锦江健身中心 Jin Jiang Health Centre

峻岭楼 Junling Building

锦楠楼 Cathay Garden

G	大堂酒廊	Lobby Lounge
G	百事丽西餐厅	Cathay Brasserie
G	锦江礼品店	Jin Jiang Gift Shop
1F-5F	客房	Guest Room

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ORGANIZATION & COMMITTEE

Hosts

Center for Ultrafast Science and Technology, SJTU
Key Laboratory for Laser Plasmas (MOE), SJTU

State Key Laboratory of Precision Spectroscopy, ECNU

Conference Honorary Chairs

Jie Zhang
Shanghai Jiao Tong University, China

Ruxin Li
Shanghai Institute of Optics and Fine Mechanics, CAS, China

Zhenrong Sun
East China Normal University, China

Conference Chairs

Dongping Zhong
The Ohio State University, USA and Shanghai Jiao Tong University, China

Jian Wu
East China Normal University, China

Local Chair

Jie Chen
Shanghai Jiao Tong University, China

GENERAL INFORMATION

The 14th Femtochemistry Conference (FEMTO 14) - Dynamics of the Complexity in Chemistry, Biology, and Physics - which will be jointly organized by Shanghai Jiao Tong University (SJTU) and East China Normal University (ECNU), will take place **in Shanghai, China from July 28 to August 2, 2019.**

The series of biennial FEMTO meetings started in 1993 and the first conference was held in Berlin, Germany. Previous editions of this meeting have been held in eleven different countries, and after 10 years, the meeting is coming back to China again. Therefore, FEMTO 14 builds on the very rich history of previous Femtochemistry meetings.

FEMTO 14 will celebrate the twentieth anniversary of the Nobel Prize to Femtochemistry in memory of the late Nobel Laureate Ahmed H. Zewail. The conference will bring together scientists from all over the world to present and discuss the most recent advances in understanding the dynamics of the complexity using ultrafast photons and electrons, including various complex processes in chemistry, biology, materials, and physics with contributions from both theory and experiment. The detailed program could be found on the conference website (http://femto14.htcis.net).

Conference Venue: Jin Jiang Grand Hall of Jin Jiang Hotel
Address: 59 South Maoming Road, Shanghai 200041, China

Poster Preparation

Poster sessions are scheduled to provide an opportunity for selected papers to be presented in greater visual detail. Presenters should display paper titles, lists of authors and author affiliations on their posters. Authors must remain in the vicinity of their bulletin boards for the duration of the session (60–90 minutes) to answer questions. Poster presenters are responsible to remove their posters, and the conference staff will not collect the posters left at the end of the poster session.

Bulletin board size: 0.95 m (width) * 2.47 m (height), recommended poster size: 0.8 m (width) * 1.2 m (height) .
Poster session: 19:30-22:00, 29 July for Poster No. 1 to No. 77;
19:30-22:00, 30 July for Poster No. 78 to No. 156
Setup time: 08:00-18:00, 29 July for Poster No. 1 to No. 77;
08:00-18:00, 30 July for Poster No.78 to No. 156
Setup address: Jin Lan Room & Jin Yu Room (2F, Jin Jiang Grand Hall)

Best Poster Awards will be selected to recognize the outstanding posters. Awardees will be acknowledged at the FEMTO 14 Conference Dinner.

Tips

WIFI: Jinjianghotel
WIFI Password: hotel guest: room number+family name
non-in-house guest: mobile phone number

In accordance with the recently published government policy, effective July 1st, 2019, hotels in Shanghai will no longer pro-actively provide single-use products such as toothbrushes, combs, shower loofahs, razors, nail files and shoe brushes. If necessary, please call the hotel reception service. Thank you for your kind understanding and cooperation in making our Earth greener!

GENERAL CONFERENCE PROGRAM

28 July, 2019

Time	Event	Location
10:00-20:00	*Conference Registration*	Cathay Buliding
14:00-17:25	*Zewail Alumni Symposium*	Jin Lan Room & Jin Yu Room (2F, Jin Jiang Grand Hall)
18:00-21:30	*FEMTO 14 Reception ALL WELCOME*	Jin Jiang Grand Hall

29 July, 2019

Time	Event	Location
08:00-08:30	*Opening Ceremony*	Jin Jiang Grand Hall
08:30-12:10	*Technical Session*	Jin Jiang Grand Hall
12:10-13:45	*Lunch*	Cathay Buliding& Cathay Garden
13:45-18:00	*Technical Session*	Jin Jiang Grand Hall
18:00-19:30	*Dinner*	Cathay Buliding & Cathay Garden
19:30-22:00	*Poster Session 1*	Jin Lan Room & Jin Yu Room (2F, Jin Jiang Grand Hall)

30 July, 2019

Time	Event	Location
08:00-12:15	*Technical Session*	Jin Jiang Grand Hall
12:15-13:45	*Lunch*	Cathay Buliding& Cathay Garden
13:45-18:00	*Technical Session*	Jin Jiang Grand Hall
18:00-19:30	*Dinner*	Cathay Buliding & Cathay Garden
19:30-22:00	*Poster Session 2*	Jin Lan Room & Jin Yu Room (2F, Jin Jiang Grand Hall)

31 July, 2019

Time	Event	Location
08:00-12:50	*Technical Session*	Jin Jiang Grand Hall
12:50-14:00	*Lunch*	Cathay Buliding & Cathay Garden
14:00-18:00	*Free Discussion*	
18:00-22:00	*Conference Dinner*	Shanghai Tower

1 August, 2019

Time	Event	Location
08:00-12:15	*Technical Session*	Jin Jiang Grand Hall
12:15-13:45	*Lunch*	Cathay Buliding
13:45-18:00	*Zewail Prize Symposium*	Jin Jiang Grand Hall
18:00-19:30	*Dinner*	Cathay Buliding

2 August, 2019

Time	Event	Location
08:00-12:15	*Technical Session*	Jin Jiang Grand Hall
12:15-12:45	*Concluding Remarks*	Jin Jiang Grand Hall
12:45-14:00	*Lunch*	Cathay Buliding

SCIENTIFIC PROGRAM

Location: Jin Lan Room & Jin Yu Room (2F, Jin Jiang Grand Hall)

14:00-17:25, 28 July, 2019

Zewail Alumni Symposium

Chaired by Theis Ivan Sølling, King Fahad University for Petroleum and Minerals

14:00-14:10	Introduction
14:10-14:30	**Klaus Braagaard Møller, Technical University of Denmark** *Femtosecond mapping of photo-induced bond formation and accompanying relaxation dynamics*
14:30-14:50	**Thorsten Bernhardt, University of Ulm** *Ultrafast analysis of adsorbate photoreactions on magnesia and titania*
14:50-15:10	**Eric Wei-Guang Diau, National Chiao Tung University** *Ultrafast carrier dynamics for understanding of stability of tin perovskites*
15:10-15:30	**Qing-Bin Lu, University of Waterloo** *Application of femtochemistry to medicine: Discovery of a new class of antitumor molecules for targeted chemotherapy and radiotherapy of cancers*
15:30-15:45	Tea Break

Chaired by Bin Chen, Shanghai Jiao Tong University

15:45-16:05	**Omar F. Mohammed, King Abdullah University of Science & Technology** *Real-space imaging of ultrafast surface carrier dynamics in solar cell materials using 4D electron microscopy*
16:05-16:25	**Oh-Hoon Kwon, Ulsan National Institute of Science and Technology** *Visualization of acoustic vibration of plasmonic single nanorods using an ultrafast electron microscope coupled to a direct detection camera*
16:25-16:45	**Jongweon Cho, Myongji University** *Visualization of electrons movement in materials*
16:45-17:05	**Giovanni Maria Vanacore, Ecole Polytechnique Federale de Lausanne** *Ultrafast coherent manipulation of a free-electron wave function by electron-light quantum interaction*
17:05-17:25	**Ding-Shyue Yang, University of Houston** *Visualizing structures and photoinduced dynamics of materials and interfaces*

18:00-21:30, 28 July, 2019

18:00-21:30	Welcome Reception

Location: Banquet Hall (1F, Jin Jiang Grand Hall)

08:00-12:10, 29 July, 2019	
08:00-08:30	**Opening Ceremony**
Strong Field and Ultrafast Physics	
Chaired by Majed Chergui, Ecole Polytechnique Fédérale de Lausanne	
08:30-08:40	Introduction
08:40-09:15	**Jie Zhang, Shanghai Jiao Tong University** *Studies on an advanced ignition scheme in inertial confinement fusion*
09:15-09:50	**Margaret Murnane, University of Colorado and NIST, Boulder** *Imaging at the wavelength limit using high harmonic sources*
09:50-10:10	**Jochen Küpper, DESY, CFEL and Universität Hamburg** *Strong-field physics in the molecular frame: Toward recording the 'quantum molecular movie'*
10:10-10:25	Tea Break
Chaired by Liejia Qian, Shanghai Jiao Tong University	
10:25-11:00	**Qihuang Gong, Peking University** *Ultrahigh spatiotemporal-resolved research on surface plasmon modes in metal and carriers in semiconductors using femtosecond time resolved PEEM system*
11:00-11:30	**Jörn Manz, Shanxi University** *How femtosecond nuclear dynamics rules attosecond electronic motions: De- and recoherences of charge migration*
11:30-11:50	**Hans Jakob Wörner, ETH Zurich** *Probing molecular chirality on femtosecond and attosecond time scales*
11:50-12:10	**Peixiang Lu, Huazhong University of Science and Technology** *Precision measurement of transient processes in atoms and molecules*
12:10-13:45	Lunch

Location: Banquet Hall (1F, Jin Jiang Grand Hall)

13:45-18:00, 29 July, 2019	
Optical Control and Gas-Phase Dynamics	
Chaired by Niels Engholm Henriksen, Technical University of Denmark	
13:45-13:55	Introduction
13:55-14:30	**Ruxin Li, Shanghai Institute of Optics and Fine Mechanics, CAS** *Femtosecond ultra-high peak power lasers: 10 PW and beyond*
14:30-15:00	**Marcos Dantus, Michigan State University** *Unexpected dynamics and control of electron recollision initiated chemistry*
15:00-15:20	**Shuyun Zhou, Tsinghua University** *Laser induced band gap engineering revealed by ultrafast time- and angle-resolved photoemission spectroscopy*
15:20-15:40	**Takayoshi Kobayashi, University of Electro-Communications** *Sub-10-fs DUV laser for ultrafast dynamics*
15:40-16:00	**Shaobo Fang, Institute of Physics, CAS** *High-energy sub-cycle waveform manipulation and potential applications*
16:00-16:15	Tea Break
Chaired by Jian Wu, East China Normal University	
16:15-16:50	**Henry Kapteyn, University of Colorado and NIST, Boulder** *Capturing the fastest charge and spin dynamics in materials using high harmonic sources*
16:50-17:20	**Albert Stolow, University of Ottawa** *Nuclear-driven electronic coherences in polyatomic molecules*
17:20-17:40	**Bing Zhang, Wuhan Institute of Physics and Mathematics** *Probing ultrafast relaxation dynamics in photoexcited fluorophenols*
17:40-18:00	**Luis Banares, Universidad Complutense de Madrid** *Strong ultrafast laser field shaping of chemical reactions*
18:00-19:30	Dinner
19:30-22:00, 29 July, 2019	
19:30-22:00	Poster Session 1

Location: Banquet Hall (1F, Jin Jiang Grand Hall)

08:00-12:15, 30 July, 2019

Reaction Dynamics of Complex Molecules in Solution

Chaired by Yi Luo, University of Science and Technology of China

08:00-08:10	Introduction
08:10-08:45	**Weihai Fang, Beijing Normal University** *Quantum trajectory mean-field approach and its implementation for exploring the dynamics of the photoinduced ring-opening of 2-thiophenone*
08:45-09:15	**Yiqin Gao, Peking University** *Dynamic electric field complicates chemical reactions in solutions*
09:15-09:35	**Stephen Bradforth, University of Southern California** *Photoelectron processes in liquids: Probing elementary reactivity and excited state dynamics*
09:35-09:55	**Andong Xia, Institute of Chemistry, CAS** *Ultrafast ground-state intramolecular proton transfer in diethylaminohydroxyflavone resolved with pump-dump-probe spectroscopy*
09:55-10:10	Tea Break

Chaired by Bei Ding, Shanghai Jiao Tong University

10:10-10:45	**Yi Luo, University of Science and Technology of China** *Energy and electron transfer in condensed phase mediated by molecular structure and its environment*
10:45-11:15	**Hiroshi Miyasaka, Osaka University** *Ring-opening reactions in highly excited states of 6-π photochromic systems as revealed by femtosecond double-pulse excitation*
11:15-11:35	**Martina Havenith, Ruhr-University Bochum** *Studying solvation of photoexcited pyranine by nonlinear THz spectroscopy*
11:35-11:55	**Ehud Pines, Ben Gurion University of the Negev** *Direct experimental evidence and exact numeric simulations of diffusion-assisted geminate-recombination reaction coupled to reversible intramolecular proton transfer over water bridges*
11:55-12:15	**Zhi-Heng Loh, Nanyang Technological University** *Capturing transient species in ionized liquid water and aqueous solutions*
12:15-13:45	Lunch

Location: Banquet Hall (1F, Jin Jiang Grand Hall)

13:45-18:00, 30 July, 2019	
Biological Dynamics in Proteins and DNA/RNA	
Chaired by Dongping Zhong, The Ohio State University and Shanghai Jiao Tong University	
13:45-13:55	Introduction
13:55-14:30	**Graham Fleming, University of California, Berkeley** *Two dimensional electronic vibrational (2DEV) spectroscopy: A new method to observe the conspiracy of electrons and nuclei in molecular dynamics*
14:30-15:00	**David Jonas, University of Colorado Boulder** *Insights into photosynthetic light harvesting from 2D femtosecond spectroscopy*
15:00-15:20	**Jiali Gao, University of Minnesota and Shenzhen Bay Laboratory** *Allosteric regulation of light-harvesting and quenching*
15:20-15:40	**Yuxiang Weng, Institute of Physics, CAS** *Protein dynamical structural change in LHCII of higher plants in switching from light harvesting to photoprotection: A protein machine in response to the environmental change*
15:40-16:00	**Fei Ma, Institute of Botany, CAS** *Identification of coherent charge separation in primary photosynthesis by two-dimensional wavelet analysis*
16:00-16:15	Tea Break
Chaired by Jinquan Chen, East China Normal University	
16:15-16:50	**Thomas Elsaesser, Max-Born-Institute** *Ultrafast vibrational probes of electric fields and ions interacting with hydrated DNA and RNA*
16:50-17:20	**Dimitra Markovitsi, CNRS** *UV-induced processes in DNA multimers: From femtoseconds to milliseconds*
17:20-17:40	**Hongmei Su, Beijing Normal University** *G-quadruplex DNA-mediated ultrafast photodynamics*
17:40-18:00	**Friedrich Temps, Christian-Albrechts-University Kiel** *Electronic relaxation of 2-aminopurine-thymine base pairs by femtosecond time-resolved transient vibrational absorption spectroscopy*
18:00-19:30	Dinner
19:30-22:00, 30 July, 2019	
19:30-22:00	Poster Session 2

Location: Banquet Hall (1F, Jin Jiang Grand Hall)

08:00-12:50, 31 July, 2019

Structural Dynamics of Biological Systems in Space and Time

Chaired by Thomas Elsaesser, Max-Born-Institute

08:00-08:10	Introduction
08:10-08:45	**Ilme Schlichting, Max Planck Institute for Medical Research** *Ultrafast time-resolved protein crystallography–recent insights*
08:45-09:15	**John Zhang, NYU Shanghai & East China Normal University** *Free energies in protein-protein and protein-ligand bindings*
09:15-09:35	**Feng Gai, University of Pennsylvania** *Ultrafast dynamics in amyloid fibrils*
09:35-09:55	**Chong Fang, Oregon State University** *Characterization and functionalization of the GFP core: From excited state proton transfer to color tuning*
09:55-10:10	Tea Break

Chaired by Jianping Zhang, Renmin University of China

10:10-10:45	**Majed Chergui, Ecole Polytechnique Fédérale de Lausanne** *Structural dynamics of biosystems and materials using ultrashort optical and X-ray pulses*
10:45-11:15	**Hyotcherl Ihee, Korea Advanced Institute of Science and Technology** *Visualizing chemical reactions in space and time using time-resolved X-ray solution scattering*
11:15-11:35	**Michael Schuurman, National Research Council Canada** *Nonadiabatic dynamics probed via time-resolved X-ray spectroscopy*
11:35-11:55	**Josh Vura-Weis, University of Illinois at Urbana-Champaign** *What did the metals know, and when did they know it? Femtosecond M-edge XANES reveals short-lived states in transition metal complexes and organohalide perovskites*
11:55-12:15	**Kristina F. Chang, University of California, Berkeley** *Direct mapping of photochemical dynamics across conical intersections by XUV transient absorption spectroscopy*
12:15-12:35	**Wei Zhuang, Fujian Institute of Research on the Structure of Matter, CAS** *Ion effect on hydrogen bonding network in water*

FEI Talk

12:35-12:50	**Eric Van Cappellen, Thermo Fisher Scientific** *UltraFast TEM, a manufacture's perspective*
12:50-14:00	Lunch

14:00-18:00, 31 July, 2019

14:00-18:00	Free Discussion

18:00-22:00, 31 July, 2019

18:00-22:00	Conference Dinner

Location: Banquet Hall (1F, Jin Jiang Grand Hall)

08:00-12:15, 1 August, 2019

Structural Dynamics of Molecules and Solids in Space and Time

Chaired by Jianming Cao, Florida State University and Shanghai Jiao Tong University

08:00-08:10	Introduction
08:10-08:45	**Martin Wolf, Fritz Haber Institute of the Max Planck Society** *Beyond the molecular movie: The ultrafast electronic structure view of surface dynamics*
08:45-09:15	**R. J. Dwayne Miller, The Max Planck Institute for the Structure and Dynamics of Matter and University of Toronto** *Mapping atomic motions with ultrabright electrons: Fundamental space-time limits to imaging chemistry*
09:15-09:35	**David Flannigan, University of Minnesota** *Revealing the influence of local structure on lattice dynamics with ultrafast electron microscopy*
09:35-09:55	**Jianqi Li, Institute of Physics, CAS** *Development and application of UTEM in material science*
09:55-10:10	Tea Break

Chaired by Zhenrong Sun, East China Normal University

10:10-10:45	**Ferenc Krausz, Max Planck Institute of Quantum Optics & Ludwig-Maximilians-Universität München** *Attosecond science: From tracing electrons to cancer detection*
10:45-11:15	**Kenji Ohmori, National Institutes of Natural Sciences** *Ultrafast many-body electron dynamics in a strongly correlated ultracold rydberg gas*
11:15-11:35	**Chong-Yu Ruan, Michigan State University** *Advances in femtosecond electron imaging and spectroscopy with high-brightness beam*
11:35-11:55	**Sascha Schäfer, University of Oldenburg** *Ultrafast nanoscale dynamics probed by time-resolved electron microscopy*
11:55-12:15	**Mihaela Zigman, Ludwig-Maximilians-Universität München, Max Planck Institute of Quantum Optics** *Field-resolved infrared molecular fingerprinting for cancer detection*
12:15-13:45	Lunch

Location: Banquet Hall (1F, Jin Jiang Grand Hall)

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<tr><td colspan="2">Chaired by Keli Han, Dalian Institute of Chemical Physics, CAS</td></tr>
<tr><td>13:55-14:45</td><td>George Schatz, Northwestern University
<i>Plasmonic arrays</i></td></tr>
<tr><td>14:45-15:20</td><td>Xueming Yang, Dalian Institute of Chemical Physics, CAS/SUSTech
<i>Geometric effects in chemical reaction</i></td></tr>
<tr><td>15:20-15:55</td><td>Vartkess A. Apkarian, University of California, Irvine
<i>A femtosecond trajectory over a few decades</i></td></tr>
<tr><td>15:55-16:10</td><td>Tea Break</td></tr>
<tr><td>16:10-16:45</td><td>Lasse Jensen, The Pennsylvania State University
<i>Understanding tip-enhanced Raman scattering single molecule images</i></td></tr>
<tr><td>16:45-17:20</td><td>David Nesbitt, University of Colorado Boulder and National Institute of Standard and Technology
<i>Controlling electron photoemission from plasmonic nanostructures: New tools and insights from ultrafast spectroscopy</i></td></tr>
<tr><td colspan="2">Chaired by Bin Chen, Shanghai Jiao Tong University</td></tr>
<tr><td>17:20-17:40</td><td>Xiaochun Gong, East China Normal University
<i>Strong-field dynamics of molecules: Electron-nuclear correlation</i></td></tr>
<tr><td>17:40-18:00</td><td>Yangyi Lu, The Ohio State University and Shanghai Jiao Tong University
<i>Modeling nonequilibrium dynamics of photoinduced ultrafast forward and backward electron transfer: A quantum-classical master equation approach</i></td></tr>
<tr><td>18:00-19:30</td><td>Dinner</td></tr>
</table>

Location: Banquet Hall (1F, Jin Jiang Grand Hall)

08:00-12:45, 2 August, 2019

Ultrafast Dynamics at Surfaces, Interfaces, and Nanostructures

Chaired by Manho Lim, Pusan National University

08:00-08:10	Introduction
08:10-08:45	**Tony Heinz, Stanford University** *Ultrafast electron dynamics in two-dimensional materials and heterostructures*
08:45-09:15	**James Hynes, University of Colorado, Boulder and ENS, Paris** *Water at electrified graphene interfaces: Structure, dynamics, and vibrational SFG spectroscopy*
09:15-09:35	**Carlos Silva, Georgia Institute of Technology** *Many-body elastic scattering of exciton polarons in organic-inorganic hybrid perovskites*
09:35-09:55	**Keli Han, Dalian Institute of Chemical Physics, CAS** *Carrier dynamics of lead-free perovskite nanocrystals*
09:55-10:10	Tea Break

Chaired by Jie Chen, Shanghai Jiao Tong University

10:10-10:45	**Shaul Mukamel, University of California, Irvine** *Ultrafast spectroscopy and imaging of molecules with classical, quantum, and noisy X-ray pulses*
10:45-11:15	**Hongfei Wang, Fudan University** *Nonlinear optical spectroscopy at the frequency and time limit: Lineshape mattters*
11:15-11:35	**Robert Baker, The Ohio State University** *Achieving surface sensitivity in ultrafast XUV spectroscopy*
11:35-11:55	**Shengye Jin, Dalian Institute of Chemical Physics, CAS** *Long-distance carrier transport beyond the limit of exciton in layered 2D perovskite quantum* wells
11:55-12:15	**Chunfeng Zhang, Nanjing University** *Suppressing bimolecular recombination in organic photovoltaic blends with non-fullerene acceptors*
12:15-12:45	Concluding Remarks
12:45-14:00	Lunch

Poster Session 1

Time: 19:30-22:00, 29 July

Location: Jin Lan Room & Jin Yu Room (2F, Jin Jiang Grand Hall)

Note: ＊: Corresponding Author
Bold: Presenter

No.	Title/Authors/Affiliations
Topic: Structural Dynamics of Molecules, Solids, and Biological Systems in Space and Time	
1	*Visualizing chemical reactions in space and time using time-resolved X-ray solution scattering* **Eun Hyuk Choi**[1,2], Jong Gu Kim[1,2], Doo-Sik Ahn[1,2], Hyotcherl Ihee[1,2,*] 1. KAIST; 2. Center for Nanomaterials and Chemical Reactions, Institue for Basic Science (IBS)
2	*Femtosecond study of water in a biomolecular hydrogel* **Eliane van Dam**[1,*], Huib Bakker[1] 1. AMOLF
3	*Capturing the electronic quantum coherence in Fenna-Matthews-Olson complex* **Hong-Guang Duan**[1] 1. Max Planck Institute for the Structure and Dynamics of Matter, Hamburg
4	*Ultrafast relaxation dynamics and optical nonlinear properties of porphyrin-based surface-supported metal-organic frameworks* **Chun Gu**[1], Hang Zhang[1], Jianbo Hu[1,*] 1. Institute of Fluid Physics, China Academy of Engineering Physics
5	*The role of meta- and para-phenylene bridges in charge separation of perylene diimide dimers* **Yuanyuan Guo**[1,*] 1. Institute of Chemistry, Chinese Academy of Sciences
6	*Ultrafast carrier dynamics in two-dimensional Bi_2O_2Se with variable layer numbers* **Yadong Han**[1], Chun Gu[1], Hang Zhang[1], Jianbo Hu[1,*] 1. Institute of Fluid Physics, China Academy of Engineering Physics
7	*Ligands effects on structural dynamics in Fe(II) spin crossover revealed by ultrafast electron diffraction* **Yifeng Jiang**[1,*], Lai Chung Liu[2], Antoine Sarracini[2], Kamil M. Krawczyk[2], Jordan S. Wentzell[2], Cheng Lu2, Ryan L. Field[2], Samir F. Matar[3], Wojciech Gawelda[4], Henrike M. Müller-Werkmeister[5], R. J. Dwayne Miller[1,2] 1. Max Planck Institute for the Structure and Dynamics of Matter; 2. Departments of Chemistry and Physics, *University* of Toronto; 3. Lebanese German University, LGU, Sahel-Alma Campus; 4. European XFEL; 5. Institute of *Chemistry,* University of Potsdam
8	*Spin and excited-state charge dynamics in myoglobin* **Dominik Kinschel**[1,*], Camila Bacellar[1], Oliviero Cannelli[1], Giulia F. Mancini[1], Boris Sorokin[1], Frederico A. Lima[2], Tetsuo Katayama[3], Wojciech Gawelda[2], Dmitry Khakhulin[2], Christian Bressler[2], Chris Milne[4], Toshinori Suzuki[5], Kazuhiko Misawa[6], Majed Chergui[1] 1. EPFL; 2. European XFEL; 3. SACLA; 4. PSI; 5. Kyoto University; 6. Tokyo University of Agriculture and Technology
9	*Unravelling ultrafast dynamics in photoexcited azaindoles* **Iker Lamas**[1], Raul Montero[2], Asier Longarte[1] 1. Spectroscopy Laboratory, Departamento Química Física, Facultad de Ciencia y Tecnología, Universidad del País Vasco, UPV/EHU, Apartado 644, 48080 Bilbao, Spain; 2. SGIKER Laser, Universidad del País Vasco, UPV/EHU, Apartado 644, 48080 Bilbao, Spain
10	*The flexibility of enzyme probed with 2D-IR* **Yunliang Li**[1,*] 1.Institue of Physics, Chinese Acedmic of Sicences

11 *Hydrogen bond dynamics in flavin mononucleotide unveiled by femtosecond stimulated Raman spectroscopy*

Yingliang Liu[1], Andrikopoulos Prokopios[1], Nils Lenngren[2], Alessandra Picchiotti[2], Miroslav Kloz[2], Chaudhari Aditya Suresh[1], Precek Martin[2], Mateusz Rebarz[2], Jakob Andreasson[2,3], Janos Hajdu[2,4], Bohdan Schneider[1], Gustavo Fuertes[1,*]

1. Institute of Biotechnology of the Czech Academy of Sciences; 2. ELI Beamlines, Institute of Physics of the Czech Academy of Sciences; 3. Condensed Matter Physics, Department of Physics, Chalmers University of Technology; 4. Department of Cell and Molecular Biology, Molecular Biophysics, Uppsala University

12 *Ultrafast photoreduction of solvated iron compounds probed by VUV photoelectron spectroscopy*

Luca Longetti[1,*], Thomas R. Barillot[1], Michele Puppin[1], José Ojeda[1], Christopher Arrell[1,2], Frank van Mourik[1], Majed Chergui[1,*]

1. Laboratotry of Ultrafast Spectroscopy (LSU), LACUS, EPFL,1015 Lausanne, Switzerland; 2. Laboratory for Advanced Photonics, PSI, 5232 Villigen, Switzerland

13 *A single-shot ultrafast electron diffraction approach based on compressive sensing*

Dalong Qi[1,*]

1. State Key Laboratory of Precision Spectroscopy, East China Normal University, Shanghai, China

14 *Structural phase transition and dynamics in MoTe$_2$ and WTe$_2$ studied by femtosecond electron diffraction*

Yingpeng Qi[1]

1. Shanghai Jiao Tong University

15 *Cycloreversion reactivity of photochromic diarylethene derivatives: Negative correlation between the S$_1$ state and higher state excited by stepwise two-photon absorption*

Hikaru Sotome[1], Tatsuhiro Nagasaka[1], Seiya Kobatake[2], Hiroshi Miyasaka[1,*]

1. Osaka University; 2. Osaka City University

16 *Polaronic species in polymers for solar cells distinguished by ultrafast dynamics of their infrared activated vibrational fingerprint*

Klara Stallhofer[1], Matthias Nuber[1], Reinhard Kienberger[1], Volker Körstgens[1], Peter Müller-Buschbaum[1,2]

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17 *RF compressed femtosecond electron microscope*

Shuaishuai Sun[1], Xiaoyi Sun[1], Joseph Williams[1], Faran Zhou[1], Chong-yu Ruan[1,*]

1. Michigan State University

18 *Ultrafast liquid dynamics revealed by solute-pump/solvent-probe spectroscopy*

Xiang Sun[1,2,*]

1. NYU Shanghai; 2. NYU-ECNU Center for Computational Chemistry at NYU Shanghai

19 *pH dependence, kinetics and light-harvesting regulation of nonphotochemical quenching in Chlamydomonas*

Lijin Tian[1,*]

1. Institute of Botany, CAS

20 *Ultrafast coherent phonon dynamics in Bi$_3$Se$_2$Te*

Atsushi Yabushita[1,*], Ying-Kuan Ko[1], Kobayashi Takayoshi[1], Chih-Wei Luo[1], Huu Phuoc Le[2]

1. National Chiao Tung University, Taiwan; 2. Can Tho University in Medicine and Pharmacy

21 *Femtosecond electronic relaxation and real-time vibrational dynamics in 2'-hydroxychalcone*

Yoshihiro Yamakita[1], Nanae Yokoyama[1], Bing Xue[1], Naoyuki Shiokawa[1], Yu Harabuchi[2], Satoshi Maeda[2], Takayoshi Kobayashi[1,*]

1. University of Electro-Communications; 2. Hokkaido University

22 *Densities of vibrational states of low-frequency internal motions for flexible and stiff*

Feng Zhang[1,*]

1. Molecular Photoscience Research Center, Kobe University

23 *Light conversion in bacterial photosynthesis*

Jianping Zhang[1,*]

1. Renmin University of China

24 *The importance of conformational change in excited states for efficient thermally activated delayed fluorescence*

Wei Zhang[1,2]

1. Institute of Chemistry, Chinese Academy of Sciences; 2. University of Chinese Academy of Sciences

25 *Two-dimensional spectroscopy study of unnatural amino acid probes*

Wenkai Zhang[1,*]

1. Beijing Normal University

26 *Energy transfer mechanism of light-harvesting complex II revealed by two-dimensional electronic spectroscopy*

Ruidan Zhu[1,2,*], Jiading Zou[1,2], Xuan Leng[1,2], Zhuan Wang[1], Hailong Chen[1], Yuxiang Weng[1,2,*]

1. Institute of Physics, Chinese Academy of Sciences, Beijing; 2. University of Chinese Academy of Sciences, Beijing

Topic: Strong Laser Field and Optical Control of Light-Matter Interactions

27 *Ultrafast laser-assisted stabilization of ionized adenine*

Fabio Covito[1,*], Angel Rubio[1]

1.Max Planck Institute for Structure and Dynamics of Matter

28 *Controlling the dynamic processes in rare-earth ion doped luminescent materials by phase-shaped femtosecond laser field*

Lianzhong Deng[1], Ye Zheng[1], Jianping Li[1], **Shian Zhang**[1,*]

1. State Key Laboratory of Precision Spectroscopy, East China Normal University, Shanghai, China

29 *Frequency-resolved photon-electronic spectroscopy for excited state population detection*

Hui Dong[1,*]

1. Graduate School of Chinese Academy of Engineering Physics

30 *Detecting electronic coherences by multidimensional high-harmonic spectroscopy*

Konstantin Dorfman[1,*]

1. State Key Laboratory of Precision Spectroscopy, East China Normal University, Shanghai, China

31 *Dynamics Stark effect on the photodissociation*

Wei Gao[1], Yongchang Han[1,*]

1. Dalian University Of Technology

32 *Molecular gases for low energy hollow core fiber pulse compression*

Elissa Haddad[1], Reza Safaei[1], Ojoon Kwon[1], Adrien Leblanc[1], Vincent Cardin[1], Bruno Eugen Schmidt[2], Philippe Lassonde[1], **Heide Ibrahim**[1,*], François Légaré[1]

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33 *Investigating the probability of photoassociation with random phase thermally two-dimensional wavefunctions*

Jinwei Hu[1], Yongchang Han[1,*]

1. Dalian University of Technology

34 *Size-dependent biexciton binding in perovskite nanocrystals studied by two-dimensional electronic spectroscopy*

Xinyu Huang[1,*], Lan Chen[1], Zhengyuan Qin[1], Chunfeng Zhang[1], Min Xiao[1]

1. School of Physics, Nanjing University

35 *Timing dissociative ionization of H_2 using a polarization-skewed femtosecond laser pulse*

Qinying Ji[1], **Shengzhe Pan**[1], Feng He[2,*], Jian Wu[1,*]

1. State Key Laboratory of Precision Spectroscopy, East China Normal University, Shanghai, China; 2. Center for Ultrafast Science and Technology, SJTU Key Laboratory for Laser Plasmas (MOE), SJTU

36 *Coherent control of electronic charge migration via quantum control of nuclear motions*

Dongming Jia[1], Jörn Manz[1,2,3,4], Yonggang Yang[1,4]

1. Institute of Laser Spectroscopy, Shanxi University; 2. International Center for Chemical Theory, University of Science and Technology of China; 3. Institut für Theoretische Chemie und Biochemie, Freie Universität Berlin; 4. Collaborative Innovation Center of Extreme Optics, Shanxi University

37 *Ultrafast dynamics of single femtosecond laser-induced periodic ripples*
Tianqing Jia[1,*]
1. State Key Laboratory of Precision Spectroscopy, East China Normal University, Shanghai, China

38 *Photochemical by-products of femtosecond filamentation in air*
Jingjing Ju[1,*], Liang Ran[2], Zhaoze Deng[2], Haiyi Sun[1], Jiansheng Liu[1], Ruxin Li[1]
1. Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Sciences; 2. Institute of Atmospheric Physics, Chinese Academy of Sciences

39 *Following macroscopic effects in gas phase experiments using XUV–UV spectroscopy*
Gabriel Karras[1]
1. Central Laser Facility, STFC Rutherford Appleton Laboratory, Didcot, Oxfordshire OX11

40 *Anisotropic order-disorder-order phase transition and strong light-matter coupling in black phosphorus visualized by ultrafast electron microscopy*
Ye-Jin Kim[1,2], Oh-Hoon Kwon[1,2,*]
1. Department of Chemistry, School of Natural Science; 2. Center for Soft and Living Matter, Institute for Basic Science (IBS)

41 *Dielectric properties manipulated by a shaped ultrashort laser pulse*
Xiaoshuang Kong[1,2], Feng Wang[1,*], Liangyou Peng[2,*]
1. Beijing Institute of Technology University; 2. Peking Univeristy

42 *Creating, imaging, and controlling chiral molecules with electric fields*
Jochen Küpper[1,*]
1. CFEL, DESY and Universität Hamburg

43 *Phase-matching-free pulse retrieval based on transient absorption in solids*
Adrien Leblanc[1], Philippe Lassonde[1], Stephane Petit[2], Jean-Chrisophe Delagnes[2], Elissa Haddad[1], Guilmot Ernotte[1], Mina Bionta[1], Vincent Gruson[1], Bruno Eugen Schmidt[3], **Heide Ibrahim[1,*]**, Eric Cormier[2], François Légaré[1]
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44 *Superradiance from nitrogen ions pumped by femtosecond laser filed*
Qingqing Liang[1], Yi Liu[1,*]
1. Shanghai Key Lab of Modern Optical System, University of Shanghai for Science and Technology

45 *Manipulating momentum and angular distributions of photoemission from hydrogen atoms by few-cycle super-intense and ultra-short laser pulses*
Aihua Liu[1,*], **Genliang Li[1]**
1. Jilin University

46 *Improving super-resolution nanoscopy by actively controlling the fluorescence of nanoparticles*
Congyue Liu[1,*], Wei Liu[1], Ju Wang[1], Xue Cheng[1], Shufeng Wang[1]
1. State Key Laboratory for Mesoscopic Physics, Department of Physics, Peking University

47 *Surface heterogeneity of perovskite thin films observed with PEEM*
Wei Liu[1,*], Ju Wang[1], Congyue Liu[1], Xue Cheng[1], Shufeng Wang[1]
1. State Key Laboratory for Mesoscopic Physics, Department of Physics, Peking University

48 *Coherent control of the cavity-free lasing of nitrogen ions pumped by femtosecond laser pusles*
Yi Liu[1,*]
1. University of Shanghai for Science and Technology

49 *The propagation effects in the absorption spectra of Rb*
Zuoye Liu[1,*], Yu He[1], Bitao Hu[1]
1. Lanzhou University

50 *Ultrafast dissociation of OCS influenced by neighbor ions* $OCS^{(1,2)+}$ *in a dimer*
Sizuo Luo[1,*]
1. Institute of Atomic and Molecular Physics, Jilin University

51 *Coincidence velocity map imaging using TPX3Cam, a time stamping optical camera with 1.5 ns timing resolution*
Erik Maddox[1]
1. Amsterdam Scientific Instruments

52 *Subcycle transfer of photon momentum in nondipole tunneling ionization*
Hongcheng Ni[1,2], Stefan Donsa[1], Joachim Burgdörfer[1]
1. Institute for Theoretical Physics, Technische Universität Wien; 2. State Key Laboratory of Precision Spectroscopy, East China Normal University

53 *Femtosecond EUV beamline for ultrafast materials research and development*
Z. Nie[1], I. C. E. Turcu[1,2,*], Y. Li[1], X. Zhang[1], L. He[1,*], J. Tu[1], H. Xu[1], Y. Chen[1], X. Ruan[1], F. Frassetto[3], P. Miotti[3], N. Fabris[3], L. Poletto[3,*], J. Wu[4], Q. Lu[5], C. Liu[5], T. Kampen[6], Y. Zhai[7], W. Liu[8], C. Cacho[9], X. Wang[1], F. Wang[1], R. Zhang[1] and Y. Xu[1,10,*]
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54 *Transient alignment of liquid water*
Fabio Novelli[1], Luis Ruiz Pestana[2], Federico Sebastiani[1], Claudius Hoberg[1], Kochise Bennett[2], Nick Stavrias[3], Lex Van der Meer[3], Gerhard Schwaab[1], Teresa Head-Gordon[2], Martina Havenith[1]
1. Department of Physical Chemistry II, Ruhr-Universität Bochum, 44780 Bochum, Germany; 2. Chemical Sciences Division, Lawrence Berkeley National Laboratory; 3. Radboud University, Institute for Molecules and Materials, FELIX Laboratory, Nijmegen, Netherlands

55 *Vibrational echo in argon dimer*
Junjie Qiang[1]
1. State Key Laboratory of Precision Spectroscopy, East China Normal University, Shanghai, China

56 *Femtosecond 100 W-level OPCPAs from 800 nm to 2 μm*
Robert Riedel[1,*], Michael Schulz[1], Ivanka Grguraš[1], Torsten Golz[1], Jan Buß[1], Mark Prandolini[1,*]
1. Class 5 Photonics GmbH

57 *Quantum tomography of ultrafast photochemical reactions in single molecules*
Chuancun Shu[1,*]
1. Central South University

58 *Spatiotemporal observation of electron dynamics in femtosecond laser manufacturing*
Jingya Sun[1,*]
1. Beijing Institute of Technology

59 *Excitation-dependent emission-color switch from individual Mn-doped perovskite microcrystals*
Qi Sun[1,2], Shiping Wang[1,2], Chunyi Zhao[1,2], Jing Leng[1], Wenming Tian[1], Shengye Jin[1,*]
1. State Key Laboratory of Molecular Reaction Dynamics, Dalian Institute of Chemical Physics; 2. University of Chinese Academy of Sciences

60 *Mid-infrared supercontinuum using Ge-Sb-S fiber with high laser damage threshold*
Kangzhen Tian[1,*]
1. Jiangsu Normal University

61 *Tunneling site resolved double ionization in strong laser fields*
Jihong Tong[1], Xiaochun Gong[1,*], Jian Wu[1,*], Wenyu Jiang[1], Wenbin Zhang[1]
1. State Key Laboratory of Precision Spectroscopy, East China Normal University, Shanghai, China

62 *Optical resonance shifts in two-dimensional cold atomic ensembles with positional correlations*
Boxiang Wang[1], Changying Zhao[1]
1. Shanghai Jiao Tong University

63 *The ultrafast spin-flip dynamics of transition metal complexes triggered by X-ray light*

Huihui Wang[1], Sergey Bokarev[2], Oliver Kühn[2]

1. Institute of Laser Spectroscopy, Shanxi University, Taiyuan 030006, China; 2. Institute for Physics, Rostock University, Albert-Einstein Street 23-24 Rostock 18059, Germany

64 *Unveiling the stimulated robust carrier lifetime of surface-bound excitons in InSe*

Rui Wang[1,2], Xiantao Jiang[1], Zhengqian Luo[2,*], Han Zhang[1,*]

1. College of Optoelectronic Engineering, Shenzhen University, Shenzhen 518060, P.R. China.; 2. Department of Electronic Engineering, Xiamen University, Xiamen 361005, P.R. China.

65 *Improving super-resolution nanoscopy by ultrafast coherent control*

Shufeng Wang[1,*], Congyue Liu[1], Wei Liu[1]

1. State Key Laboratory for Artificial Microstructure and Mesoscopic Physics, Department of Physics, Peking University, Beijing 100871, China

66 *N_2^+ lasing by bicircular two-color fields*

Jian Wu[1,*], Heping Zeng[1], Ya Cheng[1], Huailiang Xu[1], Boqu He[1], Junjie Qiang[1], Kang Lin[1], Qiying Song[1], **Hanxiao Li[1]**, Bo Xu[2], Jinming Chen[2], Zhaoxiang Liu[2], Jinping Yao[2,*]

1. State Key Laboratory of Precision Spectroscopy, East China Normal University, Shanghai, China; 2. State Key Laboratory of High Field Laser Physics, Shanghai Institute of Optics and Fine Mechanics

67 *Non-dipole effects in strong-field double ionization of atom*

Jian Wu[1], **Fenghao Sun[1]**, Feng He[2]

1. State Key Laboratory of Precision Spectroscopy, East China Normal University, Shanghai, China; 2. Shanghai Jiao Tong University

68 *Attosecond time resolved photoionization in argon atom*

Jian Wu[1], Jihong Tong[1], Xiaochun Gong[1,*], **Wenyu Jiang[1]**

1. State Key Laboratory of Precision Spectroscopy, East China Normal University, Shanghai, China

69 *Nonsequential double ionization of Ne subject to few-cycle femtosecond laser pulses*

ZhiLei Xiao[1,2], Wei Quan[1,*]

1. Wuhan Institute of Physics and Mathematics, Chinese Academy of Sciences; 2. University of Chinese Academy of Sciences

70 *Long-range Coulomb effect in above-threshold ionization of Ne subject to few-cycle and multi-cycle laser fields*

SongPo Xu[1,2], Wei Quan[1,*]

1. Wuhan Institute of Physics and Mathematics, Chinese Academy of Sciences; 2. School of Physics, University of Chinese Academy of Sciences

71 *Ultrafast excitation dynamics for noble gas atoms subject to intense femtosecond laser fields*

SongPo Xu[1], MingQing Liu[2], ShiLin Hu[3], Zheng Shu[2], Wei Quan[1,*], Jing Chen[2], XiaoJun Liu[1]

1. Wuhan Institute of Physics and Mathematics, Chinese Academy of Sciences; 2. Institute of Applied Physics and Computational Mathematics; 3. School of Physics and Astronomy, Sun Yat-Sen University (Zhuhai Campus)

72 *Dissociation ionization of halogenated alkanes under an intense femtosecond laser fields*

Yang Yan[1,*]

1. State Key Laboratory of Precision Spectroscopy, East China Normal University, Shanghai, China

73 *Disentangle the multielectron dissociative ionization dynamics using above-threshold ionization spectroscopies*

Yizhang Yang[1], Chuncheng Wang[1], Jing Chen[2], Weidong Li[3], Xiaolei Hao[3], Dajun Ding[1,*]

1. Institute of Atomic and Molecular Physics, Jilin University; 2. Institute of Applied Physics and Computational Mathematics,Beijing; 3. Institute of Theoretical Physics and Department of Physics, Shanxi University

74 *Imaging of molecular structure with high spatial and temporal resolutions*

Chunyang Zhai[1,*], Lixin He[1], Xiaosong Zhu[1], Pengfei Lan[1], Peixiang Lu[1]

1. Huazhong University of Science and Technology

75 *Optimal regulation of crystal orientation for cascaded four-wave mixing in ZnO single crystal*

Hang Zhang[1], Chun Gu[1], Jianbo Hu[1,*]

1. Institute of Fluid Physics, China Academy of Engineering Physics (CAEP)

76 *Electron-nuclear correlated multiphoton-route to Rydberg fragments of molecules*

Wenbin Zhang[1], Jian Wu[1,*]

1. State Key Laboratory of Precision Spectroscopy, East China Normal University, Shanghai, China

77 *Attosecond photoelectron holography in strong field tunneling ionization*

Yueming Zhou[1,*], Peixiang Lu[1]

1. Huazhong University of Science and Technology

Poster Session 2

Time: 19:30-22:00, 30 July

Location: Jin Lan Room & Jin Yu Room (2F, Jin Jiang Grand Hall)

Note: * : Corresponding Author
Bold: Presenter

No.	Title/Authors/Affiliations
	Topic: Reaction Dynamics of Complex Molecules in Gas Phase and in Solution
78	*Tracking a photocatalytic decarboxylation reaction over picosecond to millisecond timescales using ultrafast infrared transient absorption spectroscopy* **Aditi Bhattacherjee**[1,*] 1. University of Bristol
79	*Chemical dynamics of the reactions of tricarbon with propene – toward the formation of resonantly stabilized free radicals* **Chih-Hao Chin**[1,*], Tong Zhu[1,*], John Zhang[1,2] 1. State Key Laboratory of Precision Spectroscopy, East China Normal University, Shanghai, China; 2. New York University
80	*Hydrated excess protons in acetonitrile/water mixtures: Solvation species and ultrafast proton motions* **Benjamin Fingerhut**[1,*] 1.Max-Born-Institut für Nichtlineare Optik und Kurzzeitspektroskopie, Berlin 12489, Germany
81	*Ultrafast chemical dynamics of energetic molecules* **Jayanta Ghosh**[1], Atanu Bhattacharya[2,*] 1. Indian Institute of Science, Bangalore, India; 2. Indian Institute of Science, Bangalore,India
82	*Femtosecond photoexcitation dynamics of atoms and dimers inside a quantum solvent* **Pascal Heim**[1,*], Bernhard Thaler[1], Leonhard Treiber[1], Miriam Meyer[1], Wolfgang E. Ernst[1], Markus Koch[1] 1. Graz University of Technology
83	*Generation of near-UV 5-fs pulse laser to clarify charge transfer dynamics* **Sena Hshimoto**[1], Atsushi Yabushita[1], Kotaro Okamura[1], Takayoshi Kobayashi[1], Izumi Iwakura[1,*] 1. Kanagawa University
84	*Delayed relaxation at complex resonances studied by wavelength-selected XUV pulses* Evgenii Ikonnikov[1], Johan Hummert[2], Nicola Mayer[2], Geert Reitsma[2], Martin Eckstein[2], **Oleg Kornilov**[2,*] 1. Max-Born-Instituite, Berlin; 2. Max-Born-Institute, Berlin
85	*UV 10-fs spectroscopy elucidating the α substitution effect on photodeprotection of o-nitrobenzyl acetate* **Izumi Iwakura**[1,*], Sena Hashimoto[1], Kenta Hamada[1], Atsushi Yabushita[1], Takayoshi Kobayashi[1], Hirotaka Fujita[1], Yuta Ono[1], Noriko Kasuga[1], Kazuo Yamaguchi[1] 1. Kanagawa University
86	*Strong isotope effect for branching ratios in the vacuum ultraviolet photodissociation of carbon monoxide* Pan Jiang[1], Xiaoping Chi[1], **Min Cheng**[1,*], Hong Gao[1,*], Qihe Zhu[1] 1. Institute of Chemistry, Chinese Academy of Sciences
87	*Ultrafast transient absorption investigation on the photochemical reaction mechanisms of selected aromatic carbonyl compounds* **Jiani Ma**[1,*], Yan Guo[1], David Lee Phillips[2] 1. Northwest University; 2. The University of Hong Kong
88	*A research on isolation and application of aflatoxinoxidase based on magnetic protein label* **Meng Ma**[1] 1. East China Normal University, Shanghai, China

89 *Ultrafast photoionization dynamics of aromatic compounds as revealed by femtosecond stepwise two-photon excitation*

Koga Masafumi[1], Yusuke Yoneda[1], Naoki Ide[1], Tomomi Kawakami[1], Hikaru Sotome[1], Hiroshi Miyasaka[1,*]

1. Osaka University

90 *Ultrafast dynamics of phenol and its methyl substituted derivatives*

Yanjun Min[1]

1. Dalian Institute of Chemical Physics

91 *Intramolecular charge transfer and solvation dynamics of push-pull dyes with different π-conjugated linkers*

Xinmiao Niu[1]

1. Institute of Chemistry, Chinese Academy of Sciences

92 *Probing photoinduced reaction mechanisms of sulfur-containing molecules via ultrafast X-ray spectroscopy*

Miguel Ochmann[1,*], Abid Hussain[1], Jessica Harich[1], Rory Ma[1,2], Amy A. Cordones[3], Jae Hyuk Lee[4], Kiryong Hong[5], Robert W. Schoenlein[6], Tae Kyu Kim[2], Nils Huse[1]

1. INF, University of Hamburg; 2. Pusan National University; 3. PULSE Institute, Stanford University; 4. Pohang Accelerator Laboratory; 5. Korea Research Institute of Standards and Science; 6. SLAC National Accelerator Laboratory

93 *Ultrafast excited-state relaxation of quinazolinone family*

Jae-Heon Park[1,2], Ye-Jin Kim[1,2], Saikat Maiti[3,4], Sung You Hong[3], Oh-Hoon Kwon[1,2,*]

1. Department of Chemistry, Ulsan National Institute of Science and Technology; 2. Center for Soft and Living Matter, Institute for Basic Science; 3. School of Energy and Chemical Engineering, Ulsan National Institute of Science and Technology; 4. Center for Genomic Integrity, Institute for Basic Science

94 *Metal-cation enhanced deprotonation of water by photobases*

Dina Pines[1], Ehud Pines[1,*]

1. Department of Chemistry, Ben Gurion University of the Negev, Beer Sheva

95 *Electronic deactivation and reaction dynamics of the potential photoactive protoncrane 7-hydroxyquinoline-8-carbaldehyde (HQC)*

Sebastian Schatz[1], Michal Rode[2], Andrzej Sobolewski[2], Friedrich Temps[1,*]

1. Christian-Albrechts-University Kiel; 2. Polish Academy of Science

96 *Complete photodissociation dynamics of CF_2I_2 in solution*

Juhyang Shin[1], **Seongchul Park[1]**, Cheongha Lim[1], Hojeong Yoon[1], Manho Lim[1]

1. Pusan National University

97 *Time-resolved molecular dynamics in Rydberg states of NH_3 induced by low-order-harmonic radiation*

Vit Svoboda[1], Chuncheng Wang[1], Hans Jakob Wörner[1]

1. ETH Zurich

98 *The femtosecond time scale of radiation damage: Formation and decay of solvated electrons in neutral water clusters*

Vit Svoboda[1], Hans Jakob Wörner[1], Frank Stienkemeier[2], Rupert Michiels[2], Aaron C. LaForge[2], Jakub Med[3], Petr Slaví ek[3]

1. ETH Zurich; 2. University of Freiburg;3.UCT Prague

99 *Time-resolving molecular chirality in the gas phase with high-harmonic radiation*

Vit Svoboda[1], Hans Jakob Wörner[1], Niraghatam Bhargava Ram[1], Jürgen Stohner[2]

1. ETH Zurich; 2. ZHAW

100 *Ultrafast dynamics of laser dye DCM in the excited state*

Rikako Takagi[1], Sena Hashimoto[1], Atsushi Yabushita[1], Takayoshi Kobayashi[1], Izumi Iwakura[1,*]

1. Kanagawa University

101 *H-elimination dynamics of photoexcited fluorophenols revealed with velocity map imaging*

Ying Tang[1]

1. Wuhan Institute of Physics and Mathematics, Chinese Academy of Sciences

102 *Ab initio dynamics simulations of UV damage to the green fluorescent protein chromophore*

Wutthinan Thongyod[1,2], Chutintorn Punwong[2], William James Glover[1,3,4,*]

1. NYU-ECNU Center for Computational Chemistry at NYU Shanghai, 3663 Zhongshan Road North, Shanghai;
2. Department of Physics, Prince of Songkla University, Hatyai; 3. NYU Shanghai, 1555 Century Avenue, Shanghai;
4. Department of Chemistry, New York University, New York

103 *Bimolecular reactions from van der Waals complexes: A quantum dynamical study of Br + I_2 —> BrI + I*

Alexandre Voute[1], Klaus Braagaard Møller[1], René Wugt Larsen[1], Niels Engholm Henriksen[1,*]

1. Technical University of Denmark, Department of Chemistry

104 *Experimental observation of the vibrational fingerprint of the cis-trans isomerisation through a 2^1B-S_0 conical intersection in cis-stilbene*

Chuncheng Wang[1,2], **Max Waters**[1], Vit Svoboda[1], Conaill Perry[1], Hans Jakob Wörner[1,*]

1. ETH Zürich; 2. Jilin University

105 *Highly efficient reduction of graphene oxide (GO) in GO/MXene hybrid films revealed by ultrafast spectroscopy*

Hai Wang[1], **Lei Wang**[1,*]

1. Jilin University

106 *The role of nonradiative processes in photodissociation dynamics of S_1 state of propanal*

Yanmei Wang[1,*], Xulan Deng[1], Zhenfei Gu[1,2], Bing Zhang[1]

1. Wuhan Institute of Physics and Mathematics, CAS; 2. School of Physics and Astronomy, Yunnan University

107 *Ultrafast decay dynamics of higher-lying excited states of water*

Guorong Wu[1,*]

1. State Key Laboratory of Molecular Reaction Dynamics, Dalian Institute of Chemical Physics, CAS

108 *Photoexcitation dynamics of Roussin's red ester as nitric oxide donor*

Hojeong Yoon[1], Seongchul Park[1], Juhyang Shin[1], Cheongha Lim[1], Manho Lim[1,*]

1. Pusan National University, Busan 46241, Korea

109 *Li^+ ion solvation in organic electrolytes determined by ultrafast vibrational spectroscopy*

Kaijun Yuan[1,*]

1. Dalian Institute of Chemical Physics, Chinese Academy of Sciences

110 *Photoinduced charge migration in anthraquinone pharmaceutical product*

Song Zhang[1,*], Lian Wang[1], Ye Wang[1], Bing Zhang[1]

1. Wuhan Institute of Physics and Mathematics, Chinese Academy of Sciences

111 *Solvent effects on triplet-triplet annihilation upconversion of perylene with C_{60}-Bodipy as photosensitizer*

Xiaoguo Zhou[1,*]

1. University of Science and Technology of China

Topic: Biological Dynamics in DNA/RNA and Proteins

112 *Ultrafast dynamics in methylated cytosine and its derivatives*

Jinquan Chen[1,*]

1. State Key Laboratory of Precision Spectroscopy, East China Normal University, Shanghai, China

113 *Relaxation of excited states in DNA nucleobase adenine studied by sub-picosecond XEOL*

Natalia Gerasimova[1,*], Siarhei Dziarzhytski[2], Harald Redlin[2], Taras Malyi[2]

1. European XFEL; 2. DESY

114 *Mapping the intermolecular excited state proton transfer channel between biliverdin dimethyl ester and solvent molecules by ultrafast spectroscopy*

Yangyi Liu[1]

1. State Key Laboratory of Precision Spectroscopy, East China Normal University, Shanghai, China

115 *Structural characters and radical propagation in Escherichia Coli class la Ribonucleotide reductase from multistate technique*

Haisheng Ren[1,*]

1. Sichuan University, China

116 *How epigenetic DNA nucleosides regulate DNA photodamage: Photoproperties of epigenetic nucleosides revealed by ultrafast laser spectroscopy*

Xueli Wang[1,*]

1. State Key Laboratory of Precision Spectroscopy, East China Normal University, Shanghai, China

117 *Observation of electronic relaxation in thymine to its ground state after UV excitation through VUV photoelectron spectroscopy*

Max Waters[1,*], Chuncheng Wang[1,2], Vit Svoboda[1], Miguel Angel Silva Toledo[3], **Hans Jakob Wörner**[1]

1.ETH Zürich; 2. Jilin University; 3. CFEL

118 *Fluorescence quenching dynamics of AMND in abasic site-containing DNA for nucleobase recognition*

Chunfan Yang[1]

1. Beijing Normal Univeristy

119 *Ultrafast dynamics of short-range protein electron transfer in anabaena flavodoxin*

Jie Yang[1], Yifei Zhang[1], Tingfang He[2], Bei Ding[1,*], Dongping Zhong[1,2,*]

1. School of Chemistry and Chemical Engineering, Shanghai Jiao Tong University; 2. Department of Physics, Department of Chemistry and Biochemistry, Programs of Biophysics, Chemical Physics, and Biochemistry,The Ohio State University

120 *Ultrafast vibrational energy relaxation of membrane-bound peptide and the water in ion channel*

Shuji Ye[1,*]

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121 *Theoretical exploitation of acceptors for organic NIR-II fluorophores*

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Topic: Chemical Dynamics at Surfaces, Interfaces, and Nanostructures

122 *Ultrafast charge transfer and energy dissipation in 2D materials*

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1.Institute of Physics, Chinese Academy of Sciences; 2. Beijing National Laboratory for Molecular Sciences, Peking University; 3. Department of Materials Science and Nano Engineering, Rice University

123 *Investigating the dynamics of excitons in monolayer WSe_2 before and after organic super acid treatment*

Xin Chen[1], Lei Wang[1,*], Hai-Yu Wang[1,*]

1. Jilin University

124 *Visualization of electrons movement in materials*

Jongweon Cho[1,*]

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125 *Cooperative action of laser-induced thermal effects and ionic coordination on order of TPPA0 porphyrin derivative self-assembled interface probed via real-time second harmonic generation*

Bin Dong[1], Ruipeng Bai[1], Man Xue[1], Caihe Liu[1], Minghua Liu[1], Yuan Guo[1], **Zhen Zhang**[1,*]

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126 *Visualizing ultrafast dynamics of nanoparticles in liquid by 4D electron microscopy*

Xuewen Fu[1,2,*], Chen Bin[2], Tang Jau[2]

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127 *Probing atomic-scale photoexcitation and transient carrier dynamics of rutile TiO_2(110)*

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128 *Many-body effects in photocatalytic dynamics*

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129 *Interracial energy transfer on MXene from ultrafast dynamics investigation to clinical application*

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130 *Intramolecular singlet fission in a spiro-conjugated dye solution*
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131 *Excited-state proton transfer of a cationic superphotoacid in an AOT reverse micelle*
Hak-Won Nho[1], Jae-Heon Park[1,2], Oh-Hoon Kwon[1,2]
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132 *Pressure manipulation of exciton dynamics in few-layer* MoS_2
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133 *Real-time observation of amyloid formation at biomembrane surface by ultra-sensitive femtosecond sum frequency generation vibrational spectroscopy*
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134 *Heterogenous dynamics in self-assembled materials revealed by transient VSFG microscopy*
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135 *Ultrafast charge generation and nongeminate recombination in non-fullerene OPV blends*
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136 *Accelerated exciton decay behavior in Mn-doped* $CsPbCl_3$ *nanocrystals revealed by modified femtosecond transient absorption spectroscopy*
Shiping Wang[1], Jing Leng[1,*], Yanfeng Yin[1], Junxue Liu[1], Shengye Jin[1,*]
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137 *Charge carrier dynamics on graphitic-*C_3N_4 *with copper phosphide modification*
Wenchao Wang[1], **Ruixue Zhu**[2,*], David Lee Phillips[1,*]
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138 *Triplet energy transfer across the inorganic/organic interface for photochemical photon upconversion*
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139 *Ultrabright 300 keV photoelectron gun designed for single-shot ultrafast electron diffraction*
Jing Yang[1], Jiangtao Li[2], Jianbo Hu[2,*]
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140 *Ultrafast spectroscopic study on the luminescence mechanism of metal nanoclusters*
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141 *Femtosecond SFG visualized hydroxide ion in macroscopically hydrophobic teflon matrix*
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142 *Direct observation of local and charge-transfer exciton hybridization in high-efficiency organic solar cells*
Jiangbin Zhang[1,2,*], Bin Kan[3], Xiangjian Wan[3], Yongsheng Chen[3], Richard H. Friend[1], Artem A. Bakulin[2,*]
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143 *How MXene (*Ti_3C_2*) convert different wavelength photon into thermal energy –an ultrafast view*
Qi Zhang[1]
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144 *Pressure-induced tunable electron transfer and Auger recombination rates in CdSe/ZnS quantum dot-anthraquinone complexe*
Huifang Zhao[1], Hang Yin[1], Xiaochun Liu[1], Hui Li[1], **Ying Shi[1,*]**, Cailong Liu[1,*], Mingxing Jin[1], Jianbo Gao[2,*], Yi Luo[3], Dajun Ding[1,*]
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Topic: Dynamics of Strongly Correlated Materials and Energy Conversion

145 *Capturing coherent evolution of excitons and polarons in $CH_3NH_3PbI_3$*
Ajay Jha[1]
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146 *Femtosecond laser driven terahertz current and terahertz driven local spin-wave modes*
Zuanming Jin[1,*], Xian Lin[1], Guohong Ma[1]
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147 *Charge transport in 2D layered perovskites mediated by molecular conformation of its organic cation*
Chuanzhao Li[1]
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148 *Coherent formation of polaron pairs in all-polymer photovoltaic blends*
Qian Li[1], Chunfeng Zhang[1], Rui Wang[1], Min Xiao[2]
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149 *Ultrafast dynamics of MoS_2 sheet studied by time-resolved spectroscopic photoemission electron spectroscopy*
Yu Liang[1], Bo-han Li[1], Ziling Li[2], Guanhua Zhang[1], Julong Sun[1], Chuanyao Zhou[1], Yu Ye[2], Xueming Yang[1], **Zefeng Ren[1,*]**
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150 *Photoexcitation dynamics in perovskite solar cells*
Run Long[1,*]
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151 *Carrier dynamics in CdSe nanorods and hybrid metal-CdSe nanodumbbells: Implication for photocatalysts*
Won-Woo Park[1], Choi Ji Yong[2], Hyunjoon Song[2], Oh-Hoon Kwon[1,*]
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152 *Coherent singlet fission in pentacene driven by low-frequency intermolecular vibrations*
Vandana Tiwari[1,2]
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153 *Femtosecond transient absorption spectroscopy study on excited state dynamics in different phases of perovskite nanocrystals*
Chao Wang[1], **Guangjiu Zhao[1,*]**
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154 *Room temperature “hidden state” in $La_{0.175}Ca_{0.125}MnO_3$ observed by time-resolved XRD*
Haijuan Zhang[1], Yuanyuan Zhang[2], Jie Chen[1,*]
1. Shanghai Jiao Tong University; 2. East China Normal University

155 *Trap-assisted hole transport in layered hybrid perovskite*
Chunyi Zhao[1,2], Qi Sun[1,2], Wenming Tian[1,*], Shengye Jin[1,*]
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156 *Ultrafast charge and energy transfer dynamics at 2D ultimate proximity*
Haiming Zhu[1,*]
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Poster Session 1 - Late Poster

No content here.

Poster Session 2 - Late Poster

Time: 19:30-22:00, 30 July

Location: Jin Lan Room & Jin Yu Room (2F, Jin Jiang Grand Hall)

Note: * : Corresponding Author
Bold: Presenter

No.	Title/Authors/Affiliations
Topic: Biological Dynamics in DNA/RNA and Proteins	
L2-1	*Application of fluorescence Stokes shift on protein solvation dynamics and the validity of linear response theory* **Tanping Li**[1,*] 1. School of Physics and Optoelectronic Engineering, Xidian University

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Abstracts for Invited Presentations

14:10-14:30 28 July 2019

Femtosecond mapping of photo-induced bond formation and accompanying relaxation dynamics

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X-ray free-electron lasers (XFELs) may be used for investigations of structural dynamics on the femtosecond time scale. However, analysis and interpretation of the experimental outcomes require support from theory and detailed atomistic simulations. In this contribution, we will build on our developments of the theory behind time-resolved X-ray scattering [1,2] and illustrate how we have used Born-Oppenheimer Molecular Dynamics (BOMD) computer simulations to assist XFEL scattering measurements for mapping of excited-state, adiabatic bond formation and accompanying solute and solvent relaxations in solvated $[Ir_2(Dimen)_4]^{2+}$ [3,4] and $[Pt_2(P_2O_5H_2)_4]^{4-}$ [5,6].

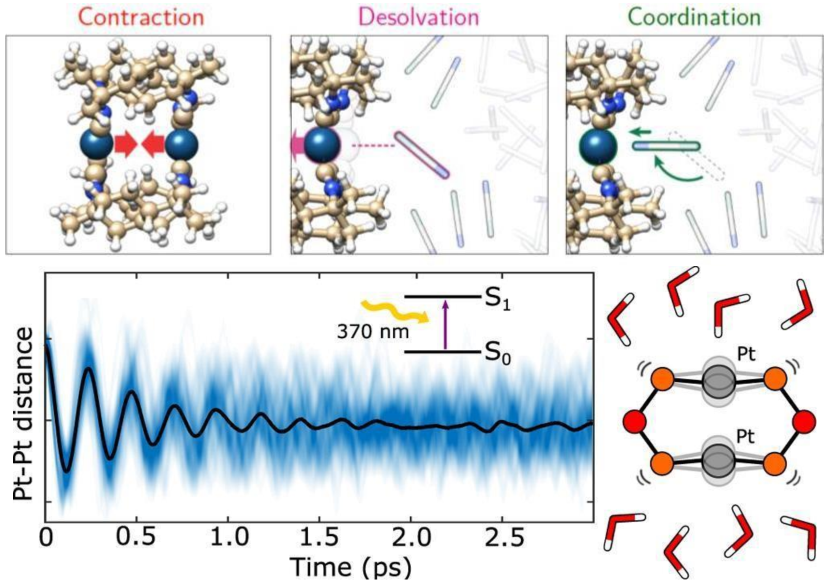

Figure 1. Our QM/MM BOMD code was used to model ground-state properties as well as the ultrafast photo- induced bond formation between the metals and the subsequent intra- and inter-molecular relaxation dynamics in solvated $[Ir_2(Dimen)_4]^{2+}$ (top) and $[Pt_2(P_2O_5H_2)_4]^{4-}$ (bottom).

References:

[1] K.B. Møller, N.E. Henriksen, Struct. Bond. **142**, 185 (2012).
[2] M. Simmermacher et al., Phys. Rev. Lett. **122**, 073003 (2019).
[3] A.O. Dohn et al., J. Phys. Chem. Lett. **5**, 2414 (2014).
[4] T.B. van Driel et al., Nat. Commun. **7**, 13678 (2016).
[5] G. Levi et al., J. Phys. Chem. C **122**, 7100 (2018).
[6] K. Haldrup et al., Phys. Rev. Lett. **122**, 063001 (2019).

14:30-14:50 28 July 2019

Ultrafast analysis of adsorbate photoreactions on magnesia and titania

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The photo-induced reaction dynamics of adsorbate molecules on well-defined ultra-thin magnesium oxide films is investigated using femtosecond laser pump-probe mass spectrometry with resonance enhanced multi-photon ionization detection.

It is demonstrated that this technique is able to reveal the real-time dynamics of the transition state and the product formation in a surface-aligned photoreaction as a function of oxide film thickness. In this experiment, methyl halide molecules adsorbed on a magnesia thin film serve as a photochemical model system.

Furthermore, titania, i.e. titanium dioxide, is a widely applicable semiconductor with interesting photochemical properties. To optimize self-cleaning surfaces, e.g., a fundamental understanding of the reaction dynamics of photooxidation reactions is necessary. We employ the same method of femtosecond laser pump-probe mass spectrometry to obtain new insights into the molecular reaction dynamics of the photooxidation of small ketone molecules (especially acetone and butanone) on TiO_2(110). Additionally, two-photon-photoemission-spectroscopy-measurements provide insight into the transient electronic surface structure upon photoexcitation.

14:50-15:10 28 July 2019

Ultrafast carrier dynamics for understanding of performance of tin perovskites

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Perovskite solar cells (PSC) have attracted much attention in recent years due to the rapid progress on the performance of PSC to attain PCE over 24 % within 10 years. Our group follows this trend working on PSC but with emphasizing on three major directions (i) lead-free PSC, (ii) carbon-based mesoscopic PSC and (iii) planar heterojunction (PHJ) PSC with the p-contact layer using graphene oxide (GO) and reduced GO (rGO) derivatives. In this presentation, I will focus on femtosecond (fs) transient absorption spectral (TAS) investigations on tin perovskites with additive Tin Fluoride (SnF_2) and co-additive Ethylenediammonium Diiodide ($EDAI_2$) to understand the effect of additives for the enhancement of the performance and stability of the $FASnI_3$ perovskite solar cells. The additive $EDAI_2$ plays a key role to cause slow passivation of the surface and relaxation of crystal strain such that the device performance increases gradually with increasing duration of storage. Three samples, pristine $FASnI_3$, $FASnI_3$ + 10 % SnF_2 and $FASnI_3$ + 10 % SnF_2 + 1 % $EDAI_2$, were used for the fs TAS studies and the results were fitted using the method of Singular Value Decomposition (SVD) with a suitable consecutive kinetic model. We found that the additive and co-additive can retard the carrier cooling time by one order of magnitude to overcome the so-called “hot-carrier phonon bottleneck”. Moreover, the defect states can be significantly reduced by adding the additive/co-additive, which reasonably explain the high performance and stability of tin-based perovskite solar cells.

15:10-15:30 28 July 2019

Application of femtochemistry to medicine: Discovery of a new class of antitumor molecules for targeted chemotherapy and radiotherapy of cancers

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The conquest of cancer continues to pose great challenges to medical science. There is a compelling need for innovative cancer research integrating biomedical sciences with physical sciences in order to ultimately conquer cancer. In our research, femtochemistry has been integrated with biomedical sciences to advance fundamental understanding and therapies of human diseases notably cancer [1]. Our studies have led to the discoveries of a new molecular mechanism (the reductive damaging mechanism) of DNA [2,3] and of the molecular mechanisms of action of existing anti-cancer agents [4-6]. These findings have provided new insight into tumorigenesis and offered unique opportunities to develop new effective drugs for high-performance therapy of cancer. We have thus found a new class of non-platinum-based anticancer compounds, called *femtomedicine (FMD) compounds*, for natural targeted chemotherapy and radiotherapy of a variety of cancers, e.g., cervical cancer, ovarian cancer, head and neck cancer, breast cancer, lung cancer, pancreatic cancer, etc [7-10]. Treatments of various cancer cells in vitro and in vivo mouse models with FMD compounds led to effective chemotherapy and enhanced radiotherapy, while the compounds themselves induced no or little toxicity toward normal cells/tissues. These compounds are therefore a new class of potent antitumor agents, which are expected to be translated into clinical trials for targeted chemotherapy and radiotherapy of multiple types of cancer. The results also show that application of femtochemistry to biology and medicine can bring breakthroughs in understanding fundamental biological processes and lead to advances in cancer therapy. This talk will discuss some progress in this exciting frontier.

References:

[1] Q.-B. Lu, Mutat. Res.: Rev. Mutat. Res. **704**, 190-199 (2010).

[2] J. Nguyen et al., Proc. Natl. Acad. Sci. USA (PNAS) **108**, 11778-11783 (2011).

[3] L. Y. Lu, N. Ou, Q.-B. Lu, Sci. Rep. **3**, 3169(1-11) (2013).

[4] Q.-B. Lu, J. Med. Chem. **50**, 2601-2604 (2007).

[5] Q.-B. Lu et al., Mol. Pharmaceutics **4**, 624-628 (2007).

[6] C.-R. Wang, Q.-B. Lu, J. Am. Chem. Soc. **132**, 14710–14713 (2010).

[7] T. Luo et al., Proc. Natl. Acad. Sci. USA (PNAS) **109**, 10175-10180 (2012).

[8] Q.-B. Lu et al., EBioMedicine **2**, 544-553 (2015).

[9] C.-R. Wang et al., Mol. Cancer Ther. **15**, 640-650 (2016).

[10] R. G. Goetze et al., EBioMedicine, (2019).

15:45-16:05 28 July 2019

Real-space imaging of ultrafast surface carrier dynamics in solar cell materials using 4D electron microscopy

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Direct visualization of charge carrier dynamics including carrier diffusion lengths and carrier trapping selectively on semiconductors-material surfaces and at their interfaces has been a key component and one of the major challenges for the development of real- world solar-cell devices. Real-space imaging of such dynamical processes can be obtained using the one-of-a-kind methodology of four-dimensional scanning ultrafast electron microscopy (4D S-UEM). In this presentation, I will show how the thickness, native oxide layers, surface morphology and surface orientation of the absorber layer can significantly influence the charge carrier dynamics and their carrier diffusion lengths. We explore these effects in several photoactive materials commonly applied for optoelectronic and solar cells devices including Silicon, CdSe, CdTe single crystals and InGaN nanowires before and after surface treatment. This work provides a foundation for potential applications of (4D S-UEM) to a wide range of devices in the fields of chemical and materials research including the real-space visualization of the surface and interface of photoactive layers at nanoscale level. [1-6]

References:

[1] M. El-Zohry, B. S. Shaheen, V. M. Burlakov, J. Yin, M. N. Hedhili, S. Shikin, B. S. Ooi, O. M. Bakr, O. F. Mohammed**,** Chem, **5,** 706-718 (2019).

[2] F. Mohammed, D.-S. Yang, S. Pal, A. H. Zewail, J. Am. Chem. Soc. **133,** 7708 (2011).

[3] J. Sun, V. A. Melnikov, J. I. Khan, O. F. Mohammed, J. Phys. Chem. Lett. **6,** 3884 (2015).

[4] R. Bose, J. Sun, J. I. Khan, B. S. Shaheen, A. Adhikari, T. K. Ng, V. M. Burlakov, M. P. Parida, D. Priante, A. Goriely, B. S. Ooi, O. M. Bakr, O. F. Mohammed, Adv. Mater. **28,** 5106 (2016).

[5] S. Shaheen, J. Sun, D-S Yang, and O. F. Mohammed, J. Phys. Chem. Lett. **8,** 2455 (2017).

[6] R. Bose, A. Bera, M. R. Parida, A. Adhikari, B. S. Shaheen, E. Alarousu, J. Sun, T. Wu, O. M. Bakr, O. F. Mohammed, Nano Lett. **16**, 4417 (2016).

16:05-16:25 28 July 2019

Visualization of acoustic vibration of plasmonic single nanorods using an ultrafast electron microscope coupled to a direct detection camera

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The use of fs photoelectron packets suffers from significant Coulomb repulsion. This space-charge effect limits the formation and propagation of fs-long electron pulses considerably because ultrashort electron pulses broaden in space and lengthen in time [1]. To circumvent the limitation in ultrafast electron microscopy (UEM), the approach has been the optimization of a gun region [2-4]. In addition, to build up an image of affordable quality integrated in seconds, stroboscopic measurements at the repetition rate of MHz are typical for detecting at least tens of electrons per pixel on a conventional CCD camera. This high repetition, however, limits the choice of a specimen because the specimen upon photoexcitation must fully relax to its original configuration in less than microseconds, the case of which is practically rare for typical TEM specimens.

Here, we push the limit of the spatial resolution of UEM by an-order-of-magnitude improvement with demonstrating the direct visualization of the sub-nm plasmonic acoustic vibration of a single gold nanorod with the period of picoseconds. This was made possible by uniquely integrating a direct electron-detection camera, which poses much higher sensitivity to incident electrons, to our UEM [5]. This allows for a less number of electrons to record an image, *i.e.*, the lower repetition rate for stroboscopic imaging and the minimal space-charge effect for a high- resolution imaging are possible. This enabled the unprecedented spatiotemporal imaging of characteristic vibration of each nanorod interfaced with a substrate, which has been veiled in optical ensemble measurements in solution [6].

References:

[1] Z. Tao, et al., J. Appl. Phys. **111**, 044316 (2012).

[2] K. Bücker, et al., Ultramicroscopy **171**, 8 (2016).

[3] E. Kieft, et al., Struct. Dyn. **2**, 051101 (2015).

[4] S. Ji, et al., Struct. Dyn. **4**, 054303 (2017).

[5] Y. M. Lee, et al., Struct. Dyn. **4**, 044023 (2017).

[6] Y.-J. Kim, et al., Matter, in press.

16:25-16:45 28 July 2019

Visualization of electrons movement in materials

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The dynamics of charged carriers such as electrons and holes largely accounts for various material's physical and chemical properties spanning from carrier transport to chemical reactivity. Real-space and real-time visualization offers intuitive picture of such dynamics. I will present advances of imaging and capturing non-equilibrium charge carrier dynamics taking place on materials surfaces and interfaces. The use of time-resolved scanning electron microscopy techniques allows us to elucidate dynamical steps of carrier excitation, relaxation and diffusion processes occurring in materials in real space and time. The results of these experiments will be exemplified by such dynamics in conventional semiconductors, such as gallium arsenide [1], and 2D materials, such as single- layer graphene [2].

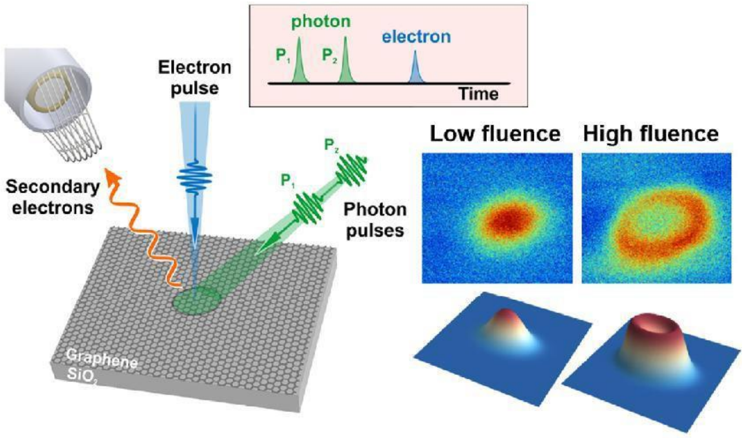

Figure 1. Carrier dynamics in single-layer graphene [2].

References:

[1] J. Cho, T. Y. Hwang, and A. H. Zewail, PNAS **111**, 2094 (2014).

[2] J. Cho, J. Tang, T. Y. Hwang, and A. H. Zewail, Nanoscale **10**, 10343 (2018).

16:45-17:05 28 July 2019

Ultrafast coherent manipulation of a free-electron wave function by electron-light quantum interaction

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The interaction between light and electrons can be exploited for generating radiation, such as in synchrotrons and free electron lasers, or for controlling electron beams for the dynamical investigation of materials and molecules. Using electromagnetic fields, the coherent control of an electron wave function can be pushed to unexplored timescales, enabling new applications in light-assisted quantum devices and diagnostics at extremely small timescales, such as those governing intramolecular electronic motions and nuclear processes.

In this contribution, I will describe a novel method for the coherent longitudinal and transverse phase manipulation of a free-electron wave function. Using appropriately synthesized optical light fields I will demonstrate how to modulate the energy, linear momentum and orbital angular momentum (vorticity) of the electron wave function with attosecond precision.

A relativistic pulsed electron beam was made to interact with an appropriately synthesized electromagnetic field. The field was generated either by a sequence of two fs laser pulses reflected at the surface of a mirror (semi-infinite field), or by the coherent superposition of the surface plasmon polaritons (SPPs) optically-generated from nanofabricated structures (near field). The energy- momentum exchange resulting from the electron-field interaction was directly mapped via momentum- resolved ultrafast electron energy-loss spectroscopy in an ultrafast transmission electron microscope (UEM).

First, we demonstrated that it is possible to coherently manipulate the longitudinal phase of a free-electron wave function using a semi-infinite light field configuration composed of a sequence of two mutually phase-locked light pulses impinging on a mirror and delayed in time by fractions of the optical cycle [1]. Then, we have shown that also the transverse electron's phase profile can be efficiently manipulated when using localized fields coupled to light-induced collective electronic modes (surface plasmon polaritons, SPPs). To demonstrate this effect, we have generated an ultrafast vortex electron beam by means of a spatially-confined optical field carrying Orbital Angular Momentum (OAM) as generated by the excitation of chiral SPPs [2]. The formation of chiral plasmons relies on the spin-to- OAM conversion from circularly-polarized light in non-paraxial scattering from a nanoscale cavity. The quantized inelastic coupling between a free-electron and a chiral SPP is thus responsible for an efficient transfer of a nonzero topological charge and helical phase distribution from the near-field to the electron wave function. The ability to access the phase profile of low-energy quantized plasmonic excitations directly resulted from the development of a new ultrafast time-domain holographic imaging technique [3]. With this method we were able to capture attosecond/nanometer-resolved phase-sensitive movies of rapidly evolving localized electromagnetic fields in plasmonic structures.

References

[1] Vanacore, Madan, et al., Nature Communication **9**, 2694 (2018)

[2] Vanacore, Berruto, et al., Nature Materials https://doi.org/10.1038/s41563-019-0336-1

[3] Madan, Vanacore et al., Science Advances **5**, eaav8358 (2019)

17:05-17:25 28 July 2019

Visualizing structures and photoinduced dynamics of materials and interfaces

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In the last two decades, developments of time-resolved electron- and xray-based imaging and spectroscopic techniques have led to various breakthroughs in visualizing and understanding light–matter interactions and photoinduced phase transitions at nonequilibrium conditions. With ultrafast electron diffraction (UED) and microscopy, most studies were conducted in a transmission geometry for the best temporal resolution possible. In this presentation, I will discuss the benefits of using UED in a reflection geometry to visualize structures and dynamics at materials surfaces and interfaces. A few examples will be given to examine the impacts due to surface strain or the existence of an interfacial structure, including (*i*) the ultrafast carrier, structural, and energy-transport dynamics in sapphire- supported monolayer MoS_2 [1], (*ii*) the behavioral differences between ultrathin supported films and strain-free specimens for the photoinduced transformation of VO_2 [2], and (*iii*) the structures and photoinduced nonequilibrium dynamics of selected interfacial molecular assemblies [3,4]. I will conclude the talk with an outlook for a broader use of reflection UED.

*We acknowledge the support from the R. A. Welch Foundation, Samsung Global Research Outreach Program, and a National Science Foundation CAREER Award.

References:

[1] X. He, M. Chebl, & D.-S. Yang (in preparation).

[2] X. He, N. Punpongjareorn, W. Liang, Y. Lin, C. Chen, A. J. Jacobson, & D.-S. Yang, Sci. Rep. **7**, 10045 (2017).

[3] X. He, C. Wu & D.-S. Yang, J. Chem. Phys. **145**, 171102 (2016).

[4] D.-S. Yang & X. He, Chem. Phys. Lett. **683**, 625–632 (2017).

08:40-09:15 29 July 2019

Studies on an advanced ignition scheme in inertial confinement fusion

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The laboratory demonstration of thermonuclear ignition has been the primary pursuit for decades for the research of inertial confinement fusion. While major progress has been made, significant challenges remain in the pursuit of ignition. In order to tackle the challenges, great international efforts have been made and various ignition schemes have been proposed and intensively investigated. In this talk, we shall review the current status and analyse intrinsic difficulties of traditional central ignition approach. We shall present an advanced ignition scheme and detailed simulation results based on this scheme, with a plan to upgrade Shenguang II laser facility to carry out an experimental campaign.

09:15-09:50 29 July 2019

Imaging at the wavelength limit using high harmonic sources

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Laser-like beams at very short wavelengths (1-50nm) can now be routinely generated using high harmonic up-conversion (HHG) of tabletop femtosecond lasers. These new quantum light sources are providing powerful new tools for probing and understanding nanoscale material properties and function. The short wavelength of HHG beams are well suited to advanced spectroscopies and imaging with high spatial resolution.[1-4] while the femtosecond-to-attosecond duration of HHG pulses is fast enough to capture the fastest spin, charge and phonon dynamics in materials. This talk will discuss the first sub-wavelength imaging using short wavelength sources, as well as new capabilities for tuning the polarization, spin and orbital angular momentum of HHG beams by sculpting the driving laser beams.

References:

[1] L. Rego et al., Science, in press (2019).

[2] K. Dorney et al., Nature Photonics **13**, 123 (2019).

[3] D. Gardner et al., Nature Photonics **11**, 259 (2017).

[4] R. Karl et al., Science Advances **4**, eaau4295 (2018).

09:50-10:10 29 July 2019

Strong-field physics in the molecularframe: Toward recording the ‘quantum molecularmovie’

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The interaction of strong laser fields with matter intrinsically provides powerful tools, such as laser-induced electron diffraction, to image transient dynamics with an extremely high spatiotemporal resolution. In strong-field physics, the initial conditions of this interaction are generally considered a weak perturbation. We investigated strong-field ionization of state- selected and strongly laser- aligned molecules and showed, for the first time, a full real-time picture of the photoelectron dynamics in the presence of the laser field and the molecular interaction. We demonstrated that the molecular potential defines the initial conditions of the photoelectron at birth and has a dramatic impact on the overall strong-field recollision dynamics: it sets the clock for the emission of electrons with a given rescattering kinetic energy. This result represents a new benchmark for the seminal statements of molecular- frame strong-field physics. Our findings have strong impact on the interpretation of self- diffraction experiments, where the photoelectron momentum distribution is used to retrieve molecular structures. Furthermore, the resulting encoding of the time-energy relation in molecular- frame photoelectron distributions shows the way of accessing a deeper understanding of electron transport during strong-field interactions and probing the molecular potential in real-time.

References

https://www.controlled-molecule-imaging.org/publication/scientific

10:25-11:00 29 July 2019

Ultrahigh spatiotemporal-resolved research on surface plasmon modes in metal and carriers in semiconductors using femtosecond time resolved PEEM system

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The ultrafast dynamics at the nanoscale is the most important physical mechanism that determines the physical phenomena and material properties. Therefore, it has always been the research hotspot in various fields such as optoelectronics, materials science and chemistry. However, a large number of existing detection methods do not have both the ultrasmall spatial resolution and the ultrafast temporal resolution. It greatly restricts the direct detection and regulation of ultrafast dynamics at the nanoscale in experiments. Here, the ultrafast time-resolved photoelectron microscopy system is established and utilized to realize the imaging of the ultrafast dynamic processes of surface plasmon modes in metal and carriers in semiconductors. Meanwhile, novel mechanisms are proposed to manipulate their spatiotemporal characteristics. The talk includes following parts:

1. The characteristics of surface plasmon modes in the near field are investigated directly by PEEM. In the frequency domain, the wavelength tunable laser is integrated with PEEM to measure the near-field spectra and image the mode distributions. In the time domain, the 7fs pulsed laser is integrated with PEEM to record the ultrafast dynamics. The dephasing time of the surface plasmon mode can be obtained by fitting the photoemission intensity curves in the time domain. Moreover, the active modulation of the dephasing time can be realized based on the strong coupling and weak coupling, respectively. For the strong coupling, the multilayer structure is designed to achieve the strong coupling between the localized and the propagating mode. With the change of detuning, the proportion of the localized and propagating mode in the coupled mode can be regulated. Therefore, the active modulation of time-domain characteristics can be achieved by the strong coupling. For the weak coupling, a stacked structure is designed to achieve the weak coupling between the surface plasmon bright mode and the dark mode. By adjusting the detuning, the loss channel of the dark mode can be modulated. Thus, the active regulation of the dephasing time of the dark mode can be achieved by the weak coupling.

2. The ultrahigh spatiotemporal-resolved imaging of carrier dynamics at the graphene/gallium arsenide interface can be realized by PEEM integrated with two-color pump and probe technique. Experimental results show that the existence of gallium arsenide would arouse the mutual effect on carrier dynamics between graphene and gallium arsenide. In this way, the carrier dynamics in graphene would be changed and excited electrons in graphene would have a longer lifetime of 14.7 ps than previously reported ones (2.4 ps) on a SiO_2 substrate. In addition, the bending direction of energy band of gallium arsenide at the interface can be changed by moving the Fermi level of gallium arsenide. It would switch the direction of carrier transport at the interface and arouse that the lifetime of exited electrons in graphene decreases to 2.6 ps. Therefore, the ultrafast carrier dynamics at graphene/gallium arsenide interface can be modulated in both time and space dimensions.

11:00-11:30 29 July 2019

How femtosecond nuclear dynamics rules attosecond electronic motions: *De- and recoherences* of charge migration

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Femtoseconds and attoseconds are the natural times scales of electronic and nuclear motions. Prominent examples are the motions during molecular vibrations and during charge migration from one molecular site to another, respectively. Charge migration may be initiated by an ultrashort laser pulse which generates a superposition of interfering non-degenerate electronic states. Constructive interferences depend, however, on good overlap of the associated nuclear wave functions. If the nuclear wave functions in different electronic states run away from each other, their overlap decays, and this causes *decoherence* of charge migration, on typical time scales of few femtoseconds. Nuclear motions may, however, cause (partial) revivals of the overlaps on much longer time scales, typically several hundred femtoseconds. As a consequence, this induces *recoherence* of charge migration. The new phenomenon opens new ways to laser control of electronic motions *via* coherent control of nuclear motions. It is demonstrated by quantum dynamics simulations of the concerted time evolutions of the nuclear and electronic densities and fluxes in ionized iodoacetylene; for the methods, see Refs. [1,2]. This application is for the same system and for the same experimental scenario for which Kraus et al. achieved the first joint experimental and theoretical reconstruction of charge migration [3,4].

References:

[1] T. Bredtmann, D. J. Diestler, S.-D. Li, J. Manz, J. F. Pérez-Torres, W.-J. Tian, Y.-B. Wu, Y. Yang and H.-J. Zhai, Phys.Chem.Chem.Phys. **17**, 29421 (2015).

[2] H. Ding, D. Jia, J. Manz and Y. Yang, Mol. Phys. **115**, 1813-1825 (2017).

[3] P. M. Kraus, B. Mignolet, D. Baykusheva, A. Rupenyan, L. Horný, E. F. Penka, G. Grassi, O. I. Tolstikhin, J. Schneider, F. Jensen, L. B. Madsen, A. D. Bandrauk, F. Remacle and H. J. Wörner, Science **350**, 790-795 (2015).

[4] H. J. Wörner, C. A. Arrell, N. Banerji, A. Cannizzo, M. Chergui, A. K. Das, P. Hamm,

[5] U. Keller, P. M. Kraus, E. Liberatore, P. Lopez-Tarifa, M. Lucchini, M. Meuwly, C. Milne, J.-E. Moser, U. Rothlisberger, G. Smolentsev, J. Teuscher, J. A. van Bokhoven and O. Wenger, Struct. Dyn. **4**, 061508 (2017).

11:30-11:50 29 July 2019

Probing molecular chirality on femtosecond and attosecond time scales

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In the first part, I will discuss high-harmonic spectroscopy driven by two-color counter-rotating circularly-polarized pulses, called "bi-circular high-harmonic spectroscopy" (BHHS). This technique is sensitive to the electronic structure and symmetry of atoms and molecules, in particular to symmetry breaking [1,2]. Applying BHHS to chiral molecules, we have observed large chiral dichroism, reaching up to 13 % [3]. This chiral dichroism (CD) originates from laser-driven charge-migration dynamics. Comparison of experiment and calculations suggests the possibility of reconstructing attosecond chiral hole dynamics from the experimental results. We have further applied BHHS in a pump-probe scheme using ultraviolet pump-pulses in a transient-grating geometry to study the photodissociation of chiral 2-iodobutane molecules. Following photoexcitation, the CD is observed to change its sign, increase and then decrease to zero as a function of the pump-probe delay [4]. Comparison to calculations suggests that the observed time evolution traces the coordinate dependence of the magnetic transition-dipole moment between the lowest electronic states of the cation. In the second part, I will discuss time-resolved photoelectron-circular dichroism (TR-PECD) based on circularly-polarized VUV radiation. Such measurements have been performed on chiral CHBrFI as well as 2-iodobutane molecules, following selective C-I bond dissociation. In both cases a clear PECD signal is observed during temporal overlap of pump and probe pulses and is found to rapidly decay to zero. Interestingly, the decay of the TRPECD signal in 2-iodobutane is much faster than that observed using BHHS, confirming the different physical origins of the chiral responses in the two measurement schemes.

References:

[1] D. Baykusheva, M. S. Ahsan, N. Lin, H. J. Wörner, Phys. Rev. Lett. **116**, 123001 (2016)

[2] D. Baykusheva, S. Brennecke, M. Lein, H. J. Wörner, Phys. Rev. Lett. **119**, 203201 (2017)

[3] D. Baykusheva, H. J. Wörner, Phys. Rev. X **8**, 031060 (2018)

[4] D. Baykusheva, D. Zindel, E. Bommeli, M. Ochsner, A. Tehlar, H. J. Wörner, PNAS, in revision

11:50-12:10 29 July 2019

Precision measurement of transient processes in atoms and molecules

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Atoms, molecules and electrons are the building blocks of matter. The control and measurement of these particles are the foundation of physics, chemistry, materials and information science, which determines our capability of understanding and transforming nature. These microscopic particles possess extreme scales in both spatial and temporal domain. For example, femtosecond pulse is capable of observing the motion of nuclei in molecules and controlling the chemical reactions. For exploring exceedingly fast electronic motion, the attosecond pulses are generally required. The harnessing of these particles thus ask for advanced detection techniques with unprecedented time and spatial resolution. Here, I will introduce several advanced tools that the Ultrafast-Optics group in Huazhong Univercity of Science and Technology has developed and demonstrate their applications for high precision measurement of transient processes in atoms and molecules.

References:

[1] M Li, H Xie et al, Physical Review Letters **122**, 183202 (2019)

[2] J Tan, Y Zhou et al, Physical review letters **121**, 253203 (2018)

[3] L He, P Lan et al, Physical review letters **121**, 163201 (2018)

[4] M He, Y Li et al, Physical review letters **120**, 133204 (2018)

[5] L He, Q Zhang et al, Nature communications **9**, 1108 (2018)

[6] P Lan, M Ruhmann et al, Physical review letters **119**, 03320 1(2017)

13:55-14:30 29 July 2019

Femtosecond ultra-high peak power lasers: 10PW and beyond

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The birth and fast development of ultra-high peak power femtosecond ultrafast lasers with peak powers at the level of petawatt or even higher have led to the generation of unprecedented extreme physical conditions in laboratories, which pave new ways to the long-standing pursuit of fundamential sciences and the promising applications for the better life of humanbeing. In this presentation, we will report the latest progress of the Shanghai Super-intense Ultrafast Laser Facility (SLUF). The SULF project was started in the beginning of 2016 and would opened to users in 2019, which is located in Zhangjiang comprehensive national scientific center in Shanghai. The 100 PW laser will be in the Station of Extreme Light (SEL), which is one of the stations at Shanghai High repetition rate XFEL and Extreme light facility (SHINE). The implementation of the project began in early 2018 and would opened to users 5-6 years later. Finally, we will outline some potential applications by using the 10 PW and 100 PW lasers.

14:30-15:00 29 July 2019

Unexpected dynamics and control of electron recollision initiated chemistry

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In a recent study [1], we provided experimental and theoretical evidence for the existence of two reaction pathways for the formation of H_3^+ from methanol under strong-field ionization. In brief (see Fig. 1a), both reaction pathways are initiated by the ultrafast double ionization of the parent molecule and proceed through prompt formation of a roaming neutral H_2 moiety from the methyl site. The roaming H_2 fragment abstracts a third proton from the methyl carbon or from the hydroxyl oxygen leading to the formation of H_3^+. We have examined the involvement of H_2 roaming mechanisms in ionic species in the formation of H_3^+ from a series of alcohols and thiols [2,3] under strong-field excitation. Through systematic pulse shaping we were able to control the yield of H_3^+ fragments by almost an order of magnitude (see Fig. 1b) for pulses with identical spectra and peak intensities [4]. This presentation will present the latest results from our group.

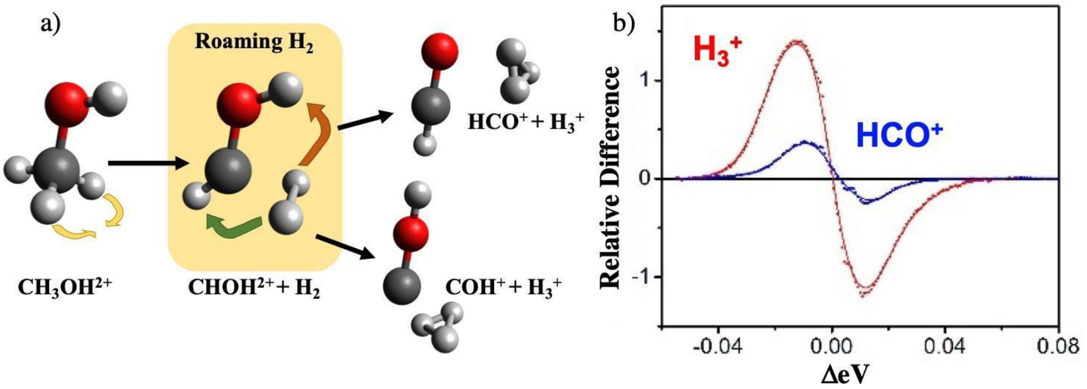

Figure 1. a) The strong-field electron recollision triggered fragmentation of methanol produces H_3^+ and either HCO^+ or CHO^+. Following nonsequential double ionization of the oxygen atom, a neutral H_2 molecule is formed, which then abstracts a proton from either end of the molecule. b) When scanning a ¾π step across the spectrum of the laser pulse the yield (positive-negative phase step) changes significantly.

References:

[1] N. Ekanayake, et al. Sci. Rep. **7**, 4703 (2017).

[2] N. Ekanayake, et al. Nat. Commun. **9**, 5186 (2018).

[3] N. Ekanayake, et al. J. Chem. Phys. **149**, 244310 (2018).

[4] M. J. Michie, et al. J. Chem. Phys. **150**, 044303 (2019).

15:00-15:20 29 July 2019

Laser induced band gap engineering revealed by ultrafast time- and angle- resolved photoemission spectroscopy

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By combining ultrafast pump-probe with angle-resolved photoemission spectroscopy, time- and angle-resolved photoemission spectroscopy (TrARPES) provides unique capability for resolving the ultrafast dynamics of electronic structure with both energy, momentum and temporal resolution. Here I will present our experimental results on band gap engineering of layered materials induced by ultrafast laser excitation.

15:20-15:40 29 July 2019

Sub-10-fs DUV laser for ultrafast dynamics

T. Kobayashi[1,*], Y. Kida[1], B. Xue[1], Y. Yabushita[1]

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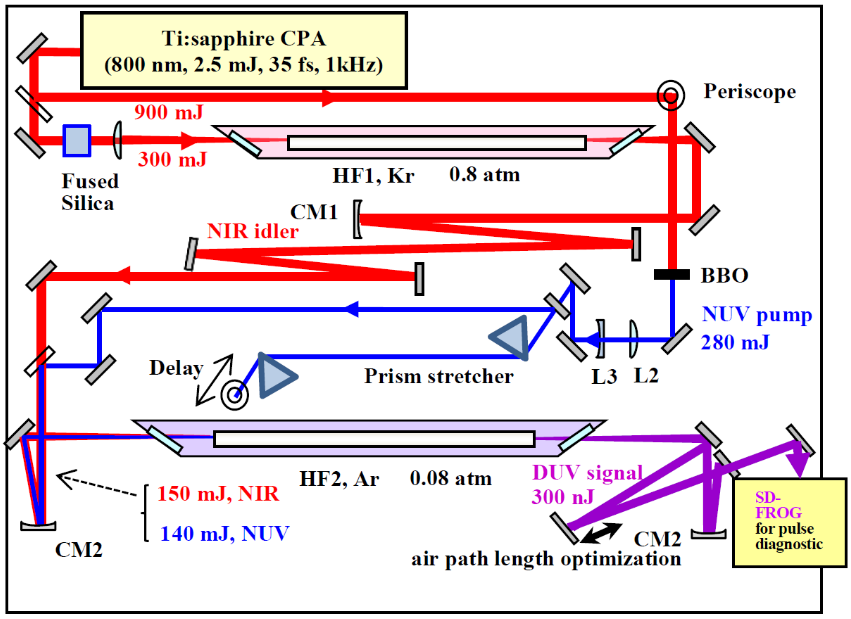
Ultrafast spectroscopy of several molecules has been performed with a sub-10-fs deep ultraviolet (DUV) laser system shown in the figure [1,2]. The ultrafast time- resolved difference absorption/emission spectra [$\Delta A(\lambda,t_d)$] were recorded using a polychromator coupled to a 128-channel lock-in amplifier (MCLIA) and a piezo- controlled optical delay stage. Two-dimensional (2D) spectra $\Delta A(\lambda,t_d)$ of real-time vibrational spectra (RTVS) were obtained as contours on the 2D map. The ΔA spectra in the Visible (Vis) or DUV regions monitor ultrafast electronic relaxations and real-time amplitudes of vibrations coupled to the corresponding electronic states at t_d.



A 798 nm fundamental-frequency output from a Ti:sapphire regenerative amplifier is split into 2. In experiment A, DUV pump (260–280nm)/Vis broadband probe (500–650 nm). In experiment B, DUV pump/DUV probe.

One of the two split fundamental-frequency pulses is used to generate the DUV pump pulse. It is separated into two beams again. One beam is frequency doubled in a BBO crystal to produce 399 nm pulse and negatively chirped by diffraction with a grating pair two times. The other beam is focused into a Kr-filled hollow-core fiber and converted to positively-chirped NIR supercontinuum by the self-phase modulation. The chirp is adjusted with chirped mirrors. Both of the beams are finally overlapped spatially and temporally in an Ar- filled hollow core fiber, and converted to a chirped DUV pump pulse based on the parametric four-wave mixing. The negative group-velocity dispersion of the DUV pump pulse is compensated by travelling through air, resulting in nearly FT-limited FWHM duration of 9 fs. The other separated fundamental pulse is spectrally broadened by focusing into a sapphire plate.

References:

[1] T. Kobayashi, Y. Kida, Phys. Chem. Chem. Phys., **14**, 6200-6210 (2012).

[2] B. Xue, A. Yabushita, T. Kobayashi, Phys. Chem. Chem. Phys., **18**, 17044-17053 (2016).

15:40-16:00 29 July 2019

High-energy sub-cycle waveform manipulation and potential applications

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We present a new scheme to support high energy sub-cycle waveforms of sub-femtosecond duration, which aim to study and control the nonlinear interactions of matter with custom-tailored sub-optical-cycle waveforms.

The generation of complicated and arbitrarily tailored optical waveforms opens new prospects for the studies of extreme light-matter interactions [1-3]. Optimization of the synthesized waveforms demands complete spatiotemporal overlap of few-optical-cycle light pulses with different spectra. In parallel synthesis schemes, one of the key enabling technologies for coherent optical waveform synthesizers is the full control of the complete phase between multiple few-cycle sub-pulses. In this letter, we demonstrate a scheme to stabilize complete phase for waveforms synthesizer, which open up new horizons for controlling strong-field interactions in atoms, molecules, solids, and novel applications in life sciences.

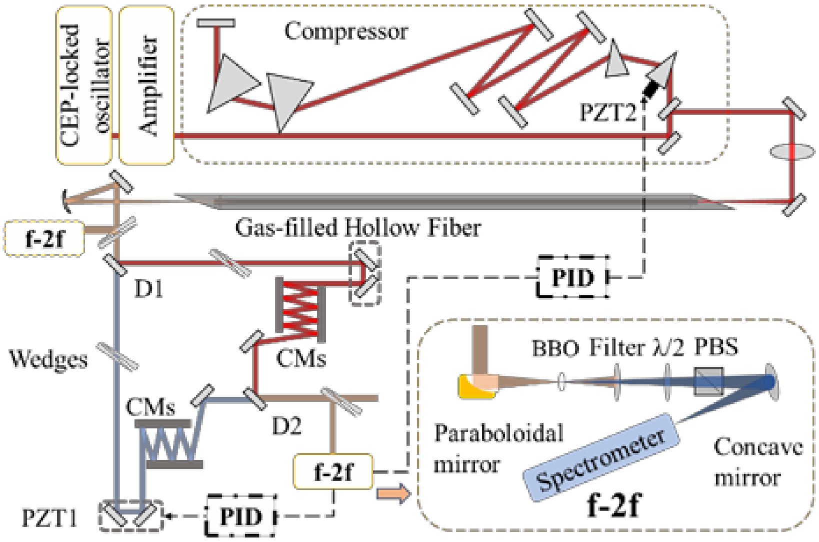

Figure 1. Experimental setup

References:

[1] F. Krausz, and M. Ivanov, Rev. Mod. Phys. **81**, 163 (2009).

[2] D. Mucke, S. Fang, G. Cirmi, G. M. Rossi, S. H. Chia, H. Ye, Y. D. Yang, R.Mainz, Manzoni, P. Farinello, G. Cerullo, and F. X. Kärtner, IEEE J. Sel. Top. Quanturn Electron. **21** (2015).

[3] C. Manzoni, O. D. Mucke, G. Cirmi, S. Fang, J. Moses, S. W. Huang, K. H. Hong, G. Cerullo, and F. X. Kartner, Laser Photon. Rev. **9**, 129 (2015).

16:15-16:50 29 July 2019

Capturing the fastest charge and spin dynamics in materials using high harmonic sources

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In recent work, we developed a new technique called ultrafast electron calorimetry, which allows us to measure the dynamic electron temperature in laser excited materials.[1-3] Because the heat capacity of the electrons is much less than that of the lattice, any change in state or couplings in a material manifests itself as an abrupt change in the electron temperature. In the case of laser excited magnetic or quasi-2D materials, the timescales for subsequently exciting the phonon baths will be different. This means that ultrafast laser excitation is a unique way for isolating the interactions between the charges, spins and phonons. In quasi-2D materials, this allows us to drive a material in to new metastable phases, or to tune the electron-phonon coupling - essentially by using a laser to induce unique periodic lattice distortions that cannot be accessed using static pressure/doping. In magnetic materials, we can directly transfer spin-polarization in materials using light, on femtosecond-to-attosecond timescales.[2, 4]

References:

[1] X. Shi et al., Science Advances **5**, eaav4449 (2019).

[2] P. Tengdin et al., Science Advances **4**, 9744 (2018).

[3] X. Shi et al., submitted (2019).

[4] P. Tengdin et al., submitted (2019).

16:50-17:20 29 July 2019

Nuclear-driven electronic coherences in polyatomic molecules

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Electronic coherences in molecules has emerged as a ‘grand challenge’ in molecular sciences due to the role that electronic correlations and dynamics play in structure and bonding[1]. In the field of ultrafast science, electronic coherences can be prepared by attosecond or few-fs pulses, producing purely electronic wavepackets which persist while the atoms are ‘frozen’ (i.e. a few femtoseconds). Once the atoms unavoidably start to move, the wavepacket could dephase and the electronic coherence might be lost. However, some suggested that nuclear motion could modify or even induce electronic coherences.

To date, theoretical investigations of nuclear-driven electronic coherences were all based on Molecular Frame (MF) calculations and proposed observables based on nonlinear coherent X-ray techniques. Here we make a very simple but important point. For gas phase molecules, the full Hamiltonian necessarily includes the rotational degrees of freedom which is defined only in the Lab Frame (LF), not in the MF. This leads to significant new insights and opportunities, as well as dramatic technical simplifications, for the probing of ultrafast electronic coherences in gas phase molecules. Here we present our study of electronic coherences in a polyatomic molecule (NH_3), non-adiabatically driven by nuclear (electronic-Coriolis) dynamics.

We emphasize that any MF electronic coherences (whether produced by ultrafast pulses, electronic-nuclear coupling, or both) initiated by polarized laser pulses will necessarily create a net anisotropy of the electronic probability distribution in the LF. As a LF anisotropy, it can be directly measured in the LF by any ultrafast angle-resolved scattering technique which is sensitive to electron probability distributions, such as X-ray or electron scattering, or as we show here: angle-resolved photoelectron spectroscopy.

References:

[1] U.S. Department of Energy. Basic Energy Sciences Roundtable Report: “Opportunities for Basic Research at the Frontiers of XFEL Ultrafast Science” (2018).

17:20-17:40 29 July 2019

Probing ultrafast relaxation dynamics in photoexcited fluorophenols

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As one of the most important chemical and biological building blocks, the relaxation dynamics of photoexcited phenol have received a lot of attention, and H-atom tunneling dynamics are found to be the dominant relaxation channel for population decay from the photoprepared S_1 state under an $S_1(\pi\pi^*)/S_2(\pi\sigma^*)$ conical intersection (CI) [1-3]. In this work, ultrafast time-resolved velocity map ion imaging and time-resolved ion yield methods are used to investigated the electronic state relaxation dynamics in photoexcited m-fluorophenol and o-fluorophenol. As for m-fluorophenol, following excitation to S_1 state below the S_1/S_2 CI, we have observed H atoms with high kinetic energy which we assign to yielding from tunneling beneath the S_1/S_2 CI, followed by coupling onto the dissociative S_2 state. This tunneling mechanism is confirmed by studies on the selectively deuterated species, m-fluorophenol-d1, an remarkable increase in the tunneling time is observed. Meanwhile, our quantum chemical calculations have also provided relevant evidence for the consequence. However, the photodissociation dynamics is quite different for photoexcited o-fluorophenol, an extra photodissociation channel is observed, which is absent for the relaxation of photoexcited m-fluorophenol. Our results highlight the effect that the presence of additional functional group F, and more specifically the precise location of the functional group, can have on the dissociation dynamics of phenols.

References:

[1] Richard N. Dixon, Thomas A. A. Oliver, and Michael N. R. Ashfold, The Journal of Chemical Physics **134**, 194303 (2011).

[2] Gareth M. Robert, Adam S. Chatterley, Jamie D. Young, and Vasilios G. Stavros, The Journal of Physical Chemistry Letters **3**, 348 (2012).

[3] Adam S. Chatterley, Jamie D. Young, Dave Townsend, Justyna M. Zurek, Martin J. Paterson, Gareth M. Roberts and Vasilios G. Stavros, Phys. Chem. Chem. Phys., **15**, 6879 (2013).

17:40-18:00 29 July 2019

Strong ultrafast laser field shaping of chemical reactions

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Ultrafast laser radiation can easily achieve electric fields that are intense enough to induce changes and dramatically modify the potential energy landscapes of molecules. This way strong laser fields can be regarded as additional reagents in chemical processes, capable of steering reactions towards desired targets [1].

We have studied ultrafast molecular photodissociation processes taking place under the influence of strong laser fields. We have shown that it is possible to modify observables such as quantum yields [2,3], lifetimes [2], translational energies [3], or spatial distributions of the ejected fragments (i.e. the reaction stereodynamics) [4], by using strong picosecond or femtosecond near-infrared pulses. The control is achieved by opening new strong-field-induced reaction channels [2], or by creating light-induced conical intersections and modulating the potentials around them by light-induced potentials [3]. These control scenarios and the outlook for future work will be the subject of this presentation.

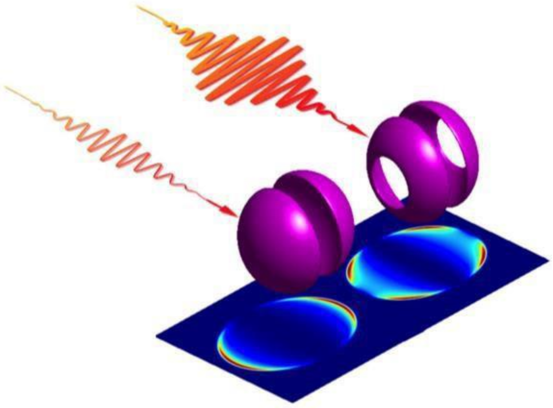

Figure 1. Control of spatial distributions of ejected fragments in photodissociation [4].

References:

[1] R. Solá, J. González-Vázquez, R. de Nalda, and L. Bañares, Phys. Chem. Chem. Phys. **17**, 13183 (2015).

[2] M. E. Corrales, G. Balerdi, V. Loriot, R. de Nalda, and L. Bañares, Faraday Discuss. **163**, 447 (2013).

[3] M. E. Corrales, J. González-Vázquez, G. Balerdi, I. R. Solá, R. de Nalda, L. Bañares, Nature Chem. **6**, 785 (2014).

[4] M. E. Corrales, R. de Nalda, and L. Bañares, Nature Comm. **8**, 1345 (2017).

08:10-08:45 30 July 2019

Quantum trajectory mean-field approach and its implementation for exploring the dynamics of the photoinduced ring-opening of 2-thiophenone

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Recently, we proposed a quantum trajectory mean-field (QTMF) approach for simulation of non-adiabatic processes[1,2], which was numerically implemented for direct *ab initio* dynamical simulation of photodissociation of diazirinone[2] and acetylacetone[3]. The combined QTMF/MM simulation has been performed for exploring the dynamics of the photoinduced ring-opening of 2-thiophenone in acetonitrile. The ring-opening reaction is predicted to be an ultrafast process with a time constant of ~230 fs and proceeds mainly along the diabatic $S_2(^1\pi\pi^*)$ pathway, while the $^1\pi\pi^* \rightarrow {}^1\pi\sigma^*$ transition takes place with little probability via the $^1\pi\sigma^*/^1\pi\pi^*$ conical intersection and the $^1\pi\sigma^*$ excited state plays a minor role in the ring-opening reaction, which is quite different from those reported for many heterocyclic molecules. All of these will be discussed in this presentation.

References:

[1] W. Feng, L. T. Xu, X. Q. Li, W-H. Fang, and Y. J. Yan, AIP Adv., **4**, 077131 (2014).
[2] B. B. Xie, L. Liu, G. Cui, W-H. Fang, X. Q. Li, J. Chem. Phys., **143**, 194107 (2015).
[3] B. B. Xie, G. Cui, W-H. Fang, J. Chem. Theory Comput., **13**, 2717 (2017).

08:45-09:15 30 July 2019

Dynamic electric field complicates chemical reactions in solutions

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Chemical reactions can be strongly influenced by external electric field (EEF). But as EEF is often time-dependent and in case that it doesn't adapt quickly enough to reaction progress especially during fast barrier crossing processes, dynamic effects could be important. In this talk, we report the finding that electrostatic interactions can reduce the height of reaction barrier for a Claisen Rearrangement reaction in water. On the other hand, strong electrostatic interactions modify the barrier into an effective potential well and confine the reactant at the reaction barrier until solvents re-adjust to provide an appropriate EEF for charge redistribution. As a result, instead of a concerted mechanism a stepwise one. Consequently, the motion of solvents modulates the reaction dynamics and leads to heterogeneous reaction paths even in a seemingly homogenous aqueous solution. We thus conclude that excessive stabilization of TS retards the barrier crossing dynamics, making the thermodynamically favorable pathway dynamically less favorable.

09:15-09:35 30 July 2019

Photoelectron processes in liquids: Probing elementary reactivity and excited state dynamics

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In the condensed phase, intermolecular interactions often control chemical reactivity and photochemistry but are complex to quantify [1]. Understanding how the electronic structure of an aqueous solute is intricately bound up with the arrangement of the host liquid provides insight into how reactions are influenced by the environment in which they take place. We have exploited liquid-jet photoelectron spectroscopy to learn about the orbital energies and shapes for the solvated electron in liquid ammonia as well as the electronic structure of the liquid itself [2]. With femtosecond time resolution, we have performed optically resonantly enhanced multi-photon ionization of indole, tryptophan, phenol and tyrosine also tracking the non-equilibrium evolution of the valence orbitals involved in excited state reactions [3,4].

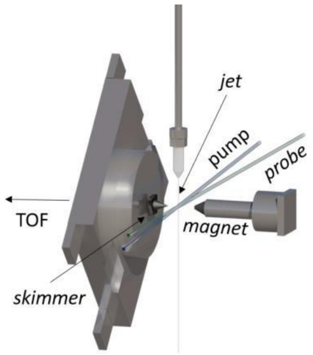

Figure 1, Liquid jet photoelectron spectroscopy

References:

[1] R. Seidel, B. Winter, and S. Bradforth, Annu. Rev. Phys. Chem. **67**, 283 (2016).

[2] T. Buttersack, P.E. Mason, R.S. McMullen, T. Martinek, K. Brezina, D. Hein, H. Ali, C. Kolbeck, C. Schewe, S. Malerz, B. Winter, R. Seidel, O. Marsalek, P. Jungwirth, and S.E. Bradforth, J. Am. Chem. Soc. **141**, 1838 (2019).

[3] G. Kumar, A. Roy, R.S. McMullen, S. Kutagulla, and S.E. Bradforth, Farad. Discuss. **212**, 359 (2018).

[4] A. Roy, R. Seidel, G. Kumar, and S.E. Bradforth, J. Phys. Chem. B **122**, 3723 (2018).

09:35-09:55 30 July 2019

Ultrafast ground-state intramolecular proton transfer in Diethylaminohydroxyflavone resolved with pump-dump- probe spectroscopy

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4'-*N,N*-Diethylamino-3-hydroxyflavone (DEAHF), due to excited-state intramolecular proton transfer (ESIPT) reaction, exhibits two solvent-dependent emission bands. Because of the slow formation and fast decay of the ground- state tautomer, its population does not accumulate enough for its detection during the normal photocycle. As a result, the details of the ground-state intramolecular proton-transfer (GSIPT) reaction have remained unknown. The present work uses femtosecond pump-dump-probe spectroscopy to prepare the short-lived ground-state tautomer and track this GSIPT process in solution. By simultaneously measuring femtosecond pump-probe and pump-dump- probe spectra, ultrafast kinetics of the ESIPT and GSIPT reactions are obtained. The GSIPT reaction is shown to be a solvent-dependent irreversible two-state process in two solvents, with estimated time constants of 1.7 ps in toluene and 10 ps in the more polar tetrahydrofuran. These results are of great value in both fully describing the photocycle of this four-level proton transfer molecule, and for providing a deeper understanding of dynamical solvent effects on tautomerization.

References:

[1] Zhuoran Kuang, Qianjin Guo,Xian Wang,Hongwei Song,Mark Maroncelli*, Andong Xia*. J.Phys.Chem.Lett., **9**, 4174 (2018)

10:10-10:45 30 July 2019

Energy and electron transfer in condensed phase mediated by molecular structure and its environment

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Energy and electron transfer play a key role in determining the functionalities of the materials, which could be regulated by the structure of the system and the environment surrounded it. Ultrafast spectroscopy has demonstrated its power to monitor such changes over the years. Here, I will add two more cases related to photocatalytic and biomolecular systems by pump-probe and sum-frequency generation techniques. The first example is the electron transfer dynamics of semiconductor g-C_3N_4 in methanol and water environments. We identified the elusive reverse hole transfer process and determined its timescale (a few hundred picoseconds). The critical role of interfacially chemisorbed methoxy (instead of methanol) as the dominant hole-scavenging species was verified thanks to its favorable energy-level alignment at the interface. In the case of biomolecular system, the vibrational energy transfer in proteins at cell membrane interface has been examined. The relaxation time of the amide I modes shows a very strong dependence on the H_2O exposure, but not on D_2O exposure, indicating the relaxation dynamics of amide I mode in H_2O environment is heavily controlled by a direct resonant channel that is energetically coupled with protein- bound water molecules. These examples highlight the importance of the specific interaction on the energy/electron transfer provided by the surroundings. The truth could only be revealed by ultrafast measurements.

References:

[1] Z. Chen, Q. Zhang, and Y. Luo. Angew. Chem. Int. Ed. **57**, 5320 (2018).
[2] J. Tan, J. Zhang, C. Li, Y. Luo, and S.J. Ye. Nat. Commun. **10**, 1010 (2019).

10:45-11:15 30 July 2019

Ring-opening reactions in highly excited states of 6- π photochromic systems as revealed by femtosecond double-pulse excitation

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Photochromism is a photoinduced reversible transformation in a chemical species between two isomers. Instant property changes by photoirradiation have raised a possible application to various systems [1]. Also from the basic viewpoints of photochemistry, these reactions have been attracting much attention as representative photo-isomerization processes. Diarylethene and fulgide derivatives are typical photochromic molecular systems where photoinduced ring-closing and opening reactions take place in 6-π electron systems. For these systems, we reported that the cycloreversion (ring-opening) reaction in highly excited states attained by the stepwise visible two-photon absorption of the closed ring-isomer of these molecules takes place with a very large reaction yield (< 50 %) even in the derivative with a small ring-opening reaction yield (< 1%) by the one-photon visible excitation to the lowest excited state. Interestingly, one-photon UV excitation, whose pump energy is comparable with the visible two-photon excitation, does not induce such enhancement of the reaction. This specific response is one of distinct examples of reactions beyond Kasha’s rule and is potentially applicable to the gated-function leading to the realization of the optical memory system fulfilling both the photo-reading and photo-erasing abilities.

In this presentation, we will discuss the mechanisms of the multiphoton-gated reaction in diarylethene [2-5] and fulgide [6] derivatives, depending on the character of the electronic state and the geometry of the Franck-Condon state in higher excited states, by introducing the experimental results using the femtosecond double-pulse excitation method with variable time intervals between the two pump pulses.

References:

[1] M. Irie, T. Fukaminato, K. Matsuda and S. Kobatake, Chem. Rev., **114**, 12174 (2014)

[2] H. Miyasaka, S. Ito, Y. Ishibashi, in “Photon-Working Switches”, Y. Yokoyama, K. Nakatani eds, Springer, p.225 (2017).

[3] H. Sotome, T. Nagasaka, K. Une, S. Morikawa, T. Katayama, S. Kobatake, M. Irie, H. Miyasaka, J. Am. Chem. Soc., **139** 17159 (2017).

[4] H. Sotome, T. Nagasaka, K. Une, C. Okui, Y. Ishibashi, K. Kamada, S. Kobatake, M. Irie, H. Miyasaka, J. Phys. Chem. Lett., **8** 3272 (2017).

[5] T. Nagasaka, T. Kunishi, H. Sotome, M. Koga, M. Morimoto, M. Irie, H. Miyasaka, Phys.Chem. Chem. Phys., **20** 19776 (2018).

[6] T. Nagasaka, H. Sotome, Y. Yoshida, Y. Yokoyama, H. Miyasaka, J. Phys. Chem. C, **122** 24987 (2018).

11:15-11:35 30 July 2019

Studying solvation of photoexcited pyranine by non-linear THz spectroscopy

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We present results of non-linear THz experiment to study changes in the solvation dynamics upon photo-excitation. By using two color plasma filaments, we generate strong and broadband single- cycle THz pulses with pulse energies of ~ 10nJ and more than 10THz bandwidth. Thereby we are able to probe the hydrogen bond frequency range which is sensitive to the hydrogen bond stretch mode around 200 $cm^{-1} \approx$ 6THz. Generation of two-color plasma filaments is done in ambient air with an inline frequency doubling geometry by a two color conversion including a fundamental (800nm) and second harmonic (400nm) beam High field strengths are obtained by THz generation via two-color plasma filamentation in combination with high imaging quality of custom elliptical mirrors.

Using a novel rapid scanning technique, a single measurements dynamic range exceeds 60dB in less than one minute of integration time and enables a live-view of the THz pulse as well as the pump- probe signal with up to 4Hz update rate. r. During an OPTP experiment at each pump-delay position the difference of the transmitted THz field for the optically excited and the non-excited sample is measured. In the talk I will present results on the changes in solvation dynamics upon photoexcitation of the photoacid pyranine, which are indicative of the instantaneous changes in the hydrogen bond network.

ACKNOWLEDGEMENTS

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11:35-11:55 30 July 2019

Direct experimental evidence and exact numeric simulations of diffusion-assisted geminate-recombination reaction coupled to reversible intramolecular proton transfer over water bridges

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Time-resolved fluorescence is used to study the excited-state proton transfer (ESPT) from 2-naphthol-6, 8-disulfonate (2N68DS) in water. Unlike in our previous studied bifunctional photoacid systems [1], the observed kinetics deviate from the exact numeric solution of a diffusion-assisted geminate recombination reaction [2] and conforms to a reaction where the geminate-recombination process is coupled to a non-diffusional over-water-bridge intramolecular PT. The two protonation targets following the photoacids dissociation are the initial O^- dissociation cite and the nearby SO_3^- side-group of the photoacid. The observation in neat water is corroborated in acetonitrile and PC solutions containing small amounts of water where PT to the solvent does not occur and a slow intramolecular ESPT process from the OH group via a pre-existing H-bonding water bridge to the SO_3^- groups is observed. The fluorescence of the proton-transferred hydrogen-bonding complex is shifted by about 2000 cm^{-1} from the R*OH fluorescence band in neat water. As the water content in the mixture increases, the R^*O^- fluorescence band shifts noticeably to the blue reaching for $\chi_{water} > 0.3$ the band position observed in pure water for R^*O^-. This marks the transition to reaction conditions where intermolecular proton transfer to the solvent prevails over the intramolecular ESPT process along pre-existing water bridges.

References:

[1] J. Ditkovich, D. Pines and E. Pines, Phys. Chem. Chem. Phys. **18**, 16106 (2016).

[2] R. Simkovitch, D. Pines, N. Agmon, E. Pines, and D. Huppert, J. Phys. Chem. B **120**, 12615 (2016).

11:55-12:15 30 July 2019

Capturing transient species in ionized liquid water and aqueous solutions

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The ionization of liquid water serves as the principal trigger for a myriad of phenomena that are relevant to radiation chemistry and radiation biology. The earliest events that follow the ionization of water, however, remain relatively unknown. By employing few-cycle pulses in the visible to near-infrared (500 – 900 nm) and the short-wave infrared (0.9 – 1.7μm), we have performed a comprehensive probe of the fate of the electron that is initially injected into the conduction band by ionization. The results suggest that the relaxation of the conduction band electron to the hydrated s electron proceeds via an intermediate state, whose lifetime is found to be 77 ± 6 fs (118 ± 7 fs) in H_2O (D_2O). In complementary experiments, femtosecond soft X-ray free-electron laser probing of ionized liquid at the oxygen K edge furnished the X-ray absorption spectrum and a formation time of 181 ± 19 fs for the highly reactive OH radical species. Our studies of ionized liquid water have also been extended to anion photodetachment in aqueous solution. For example, in the case of phenoxide, which serves as a model for the redox-active amino acid tyrosine, photodetachment launches vibrational wave packet motion along multiple vibrational modes of the phenoxyl radical product. Analysis of the vibrational wave packet dynamics reveals the normal modes that drive structural reorganization upon photodetachment. Our results shed light on the elementary ultrafast dynamics that accompany the interaction of ionizing radiation with molecules of biological relevance.

13:55-14:30 30 July 2019

Two dimensional electronic vibrational (2DEV) spectroscopy: A new method to observe the conspiracy of electrons and nuclei in molecular dynamics

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Two-dimensional electronic-vibrational (2DEV) spectroscopy correlates the evolution of the nuclear (vibrational) degrees of freedom with that of the electronic degrees of freedom. This leads to multiple applications in ultrafast chemistry and biology. In this talk I will show a number of the initial applications to illustrate the potential range of the method. In the simplest case, the presence of an acceptor vibration can signal the presence of electronic energy transfer to that site, less obviously the 2DEV line shape, in particular the center line slope (CLS) can signal which states are energy transfer partners. In strongly mixed systems the sensitivity of the vibrational transition moments to the coupling can produce rather complex spectra with prominent, if short-lived, vibronic coherences. In this case, vibrations that are very weak or even dark in the electronic spectrum can give strong 2DEV signals providing new insight into the roles of dark charge transfer states, for example. Returning to the CLS it can signal rapid changes in structure during large amplitude motion such as motion to, and passage through a conical intersection. Combined with spectro-electrochemistry 2DEV spectroscopy can give new insights into one-and multiple-proton coupled electron transfer reactions, this work being a collaboration with T. Moore, A. Moore and D. Gust.

14:30-15:00 30 July 2019

Insights into photosynthetic light harvesting from 2D femtosecond spectroscopy

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In photosynthesis, antenna proteins harvest light and efficiently transfer the electronic excitation energy to reaction center proteins that initiate chemical transformations. The standard Forster framework for understanding electronic energy transfer is based on the adiabatic approximation of slow vibrations and fast electrons. Femtosecond two-dimensional spectra of many antenna proteins exhibit quantum beats with signatures indicative of nonadiabatic vibrational-electronic coupling. The longest lived beats originate from a nonadiabatic enhancement of Raman pathways for excitation of delocalized vibrations on the electronic ground state. These vibrations characterize the initial coupling between excited states and reveal nested funnels in which the adiabatic approximation fails spectacularly over entire vibrational coordinate spaces, driving energy transfer outside the Forster framework. The nonadiabatic dynamics in nested funnels is efficient at low vibrational energies and does not require high vibrational velocities or conical intersections.

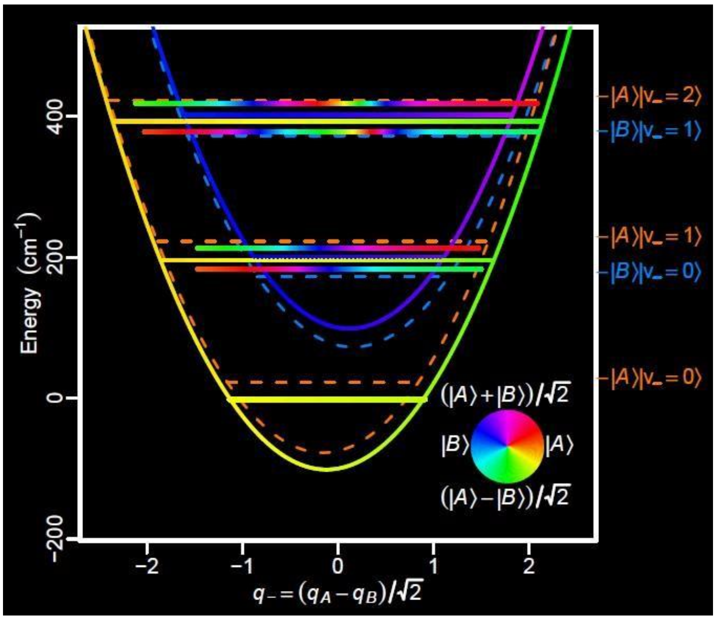

Figure 1. A nested funnel for nonadiabatic energy transfer showing: the uncoupled states in which the donor (B) and acceptor (A) pigments are electronically excited (dashed potential curves and energy levels); the adiabatic excitons (solid potential curves and energy levels); and the nonadiabatic energy levels (multicolored energy levels). Color indicates electronic character of each eigenfunction at every coordinate.

References:

[1] David M. Jonas, Annu. Rev. Phys. Chem. **69**, 327 (2018).

15:00-15:20 30 July 2019

Allosteric regulation of light-harvesting and quenching

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Major light-harvesting complex of photosystem II (LHCII) is a photoreceptor protein that regulates energy transfer and dissipation in response to rapid fluctuations of light intensity, directly affecting the efficiency of photosynthesis. In this presentation, I will describe an investigation combining molecular dynamics simulation and temperature-jump time-resolved IR spectroscopy to understand the mechanism of energy dissipation in LHCII. An allosteric regulation of the global protein conformational changes induced by local conformational transitions will be discussed. The dynamic motions induce close contacts between the associated chromophores to facilitate fluorescence quenching. In addition, I will discuss a multistate density functional theory designed to model photochemical and charge transfer processes.

15:20-15:40 30 July 2019

Protein dynamical structural change in LHCII of higher plants in switching from light harvesting to photoprotection: A protein machine in response to the environmental change

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Green plants have to cope with the drastic light intensity change during the daytime and they have successfully developed a mechanism of switching between high efficient light energy collection and energy dissipation known as non-photochemical quenching (NPQ). It has been proved that the light-harvesting complexes of photosystem II of green plants play such a dual role. It is generally accepted that this regulatory switch is induced by increased acidity or temperature due to conformational change. However, the molecular origin of the protein dynamics at the atomic level is still unknown.

In this talk, I will briefly present some of the results related to the efficient energy transfer in major light-harvesting complexes of photosystem II (LHCII) trimer by 2D electronic spectroscopy, then some of the results about the temperature-jump time-resolved infrared spectroscopic and molecular dynamics simulations study of the NPQ mechanism of LHCII trimer in view of protein dynamical structural changes in response to the acidity and elevated temperature. We found that the spontaneous formation of a pair of local α-helices from 310-helix E and the C-terminal coil, in response to increased environmental temperature or acidity, induces a scissoring motion of transmembrane helices A and B, shifting the conformational equilibrium to a more open state. The dynamical allosteric conformation change leads to close contacts between the carotenoid lutein 1 and photoactivated chlorphyll pigments, facilitating energy quenching. The results reveal a unified mechanism by which LHCII controls the dissipation of excess excited energy in response to increased temperature and acidity, as a result of intense Sun light in plants photosynthesis.

15:40-16:00 30 July 2019

Identification of coherent charge separation in primary photosynthesis by two-dimensional wavelet analysis

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Charge separation is the primary and essential step for photosynthesis [1] and has been recently studied by coherent two-dimensional (2D) electronic spectroscopy. In plant photosystem II reaction center (RC), electronic and vibronic coherences were found assisting the charge separation process [2]. For purple bacteria, we identified short-lived electronic and long-lived vibrational coherences [3]. To reveal evolution dynamics of the coherences, we performed wavelet analysis [4] for three mutant RCs, which is able to provide both frequency and dynamic information of the oscillatory signatures. The obtained scalogram and time-resolved 2D frequency map explicitly show a ~100 fs electronic coherence between the excited special pair (P*) and the charge separation precursor, $P_A^+P_B^-$ state, and vibrational coherences lasting for hundreds of fs. Comparing the three RCs, we conclude that the electronically coherent electron transfer inside the P dimer is responsible for the super-high charge separation efficiency. We believe that the delicate quantum coherence mechanism in the RC could provide guidance for new artificial energy conversion systems that exploit coherence phenomena to achieve maximum efficiency.

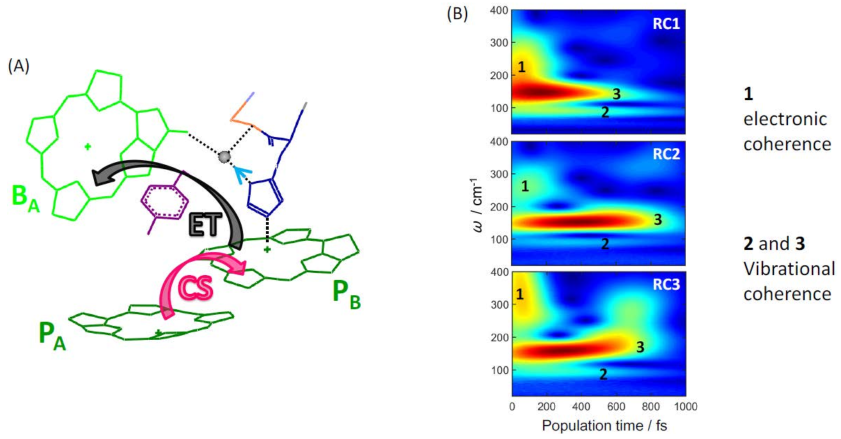

Figure 1. (A) Scheme of primary charge separation in bacterial RC. (B) Scalogram of the (P, $P_A^+P_B^-$) cross peaks of the three mutant RCs.*

References:

[1] S. Rafiq and G. Scholes, J. Am. Chem. Soc. **141**, 708 (2019).

[2] E. Romero, et al. Nat. Phys. **10**, 676 (2014).

[3] F. Ma, et al. Nat. Commun. **10**, 933 (2019).

[4] E. Romero, et al. Sci. Rep. **7**, 1 (2017).

16:15-16:50 30 July 2019

Ultrafast vibrational probes of electric fields and ions interacting with hydrated DNA and RNA

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The structure and function of charged biomolecules are strongly influenced by electric interactions with their fluctuating water shell and ion atmosphere [1]. The strength, range, and dynamics of electric forces are barely characterized at the molecular level, a fact calling for new noninvasive probes which map electric fields and their fluctuation amplitudes on the ultrafast time scale. In this talk, new results on electric interactions of double-helical DNA and RNA with their water and ion environment are presented. Backbone vibrations located at the interface to the water shell serve as probes of noncovalent interactions. Our work is based on a combination of two-dimensional infrared (2D-IR) spectroscopy and molecular dynamics and quantum mechanical molecular mechanics simulations.

Experiments with native salmon testes DNA show pronounced changes of the 2D-IR lineshapes of backbone vibrations upon changing the water level from essentially dehydrated conditions up to full hydration, i.e., upon building the solvation shell layer by layer [2]. The electric field acting on the backbone oscillators originates mainly from the dipolar water molecules of the environment, with a spatial range that is confined to 2-3 water layers. The electric field amplitudes felt by the backbone phosphate groups are on the order of 100 MV/cm with fluctuation amplitudes of ±25 MV/cm. In comparison to DNA, short double-helical RNA structures interact with a more structured first water layer at the interface, generating electric fields of similar magnitude [3].

DNA and RNA helices are studied in an aqueous environment with a tailored excess concentration of magnesium ions (Mg^{2+}) for identifying particular ionic arrangements and their interaction strength. A proof-of-principle experiment with the model system dimethyl-phosphate reveals a blue-shifted phosphate stretching band in both the linear and 2D-IR spectra as hallmark of phosphate- magnesium contact ion pairs [4]. A similar behavior is observed with RNA in an environment with Mg^{2+} excess. The blue-shift is caused by the repulsive interaction of phosphate oxygens in contact with Mg^{2+}.

References:

[1] D. Laage, T. Elsaesser, and J. T. Hynes, Chem. Rev. **117**, 10694 (2017).

[2] T. Siebert, B. Guchhait, Y. Liu, B. P. Fingerhut, and T. Elsaesser, J. Phys. Chem. Lett. **7**, 3131 (2016).

[3] E. M. Bruening, J. Schauss, T. Siebert, B. P. Fingerhut, and T. Elsaesser, J. Phys. Chem. Lett. **9**, 583 (2018).

[4] J. Schauss, F. Dahms, B. P. Fingerhut, and T. Elsaesser, J. Phys. Chem. Lett. **10**, 238 (2019).

16:50-17:20 30 July 2019

UV-induced processes in DNA multimers: From femtoseconds to milliseconds

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Characterization of the UV-induced processes in DNA is important in respect to the damage of the genetic code and the development of DNA-inspired devices. Excited state relaxation and reactivity of systems with various secondary structures has been studied by time-resolved optical spectroscopy [1]. These processes will be illustrated using as examples the adenine single strand $(dA)_{20}$, the first DNA multimer to be studied by fs spectroscopy [2], and a telomeric G-quadruplex, in which collective effects are particularly pronounced [3].

The low fluorescence anisotropy determined by fluorescence upconversion evidenced that ultrafast energy transfer takes place among bases via exciton states [3,4]. The dynamics of this process (<100 fs) was resolved recently by transient absorption spectroscopy with resolution of 30 ns (coll. with Politecnico di Milano) [5]. The same study, performed for $(dA)_{20}$, revealed that excited charge transfer (CT) states, involving neighboring adenines ($A^{+}A^{-}$), are formed progressively, within 3 ps, in line with the time-resolved fluorescence spectra [4]. The fluorescence decays are strongly non-exponential, extending from fs to ns [6]. Deviations from classical kinetic models are due to the anisotropic structure of the systems and the occurrence of conformational motions. This is also reflected in the dynamics of base radicals studied on the μs/ms time-scales. The latter are formed with a quantum yield of 10^{-3} upon absorption of single photons whose energy is lower than the ionization potential of free bases [7,8]. The mechanism responsible for such an unexpected ionization could involve CT states.

References

[1] D. Markovitsi, Photochem. Photobiol. **92**, 45 (2016).

[2] D. Markovitsi, A. Sharonov, D. Onidas, T. Gustavsson, ChemPhysChem **3**, 303 (2003).

[3] P. Changenet-Barret, Y. Hua, D. Markovitsi, Top. Curr. Chem. **356**, 183 (2015).

[4] A. Banyasz, T. Gustavsson, D. Onidas, P. Changenet-Barret, D. Markovitsi, R. Importa, Chem. Eur. J. **19**, 3762 (2013).

[5] R. Borrego-Varillas, G. Cerullo, D. Markovitsi, J. Phys. Chem. Lett. **10** 1639 (2019).

[6] D. Markovitsi, F. Talbot, T. Gustavsson, D. Onidas, E. Lazzarotto, S. Marguet, Nature **441**, E7 (2006).

[7] A. Banyasz, T. Ketola, A. Muñoz-Losa, S. Rishi, A. Adhikary, M.D. Sevilla, L. Martinez-Fernandez, R. Imprората, D. Markovitsi, J. Phys. Chem. Lett. **7**, 3949 (2016).

[8] A. Banyasz, L. Martinez-Fernandez, C. Balty, M. Perron, T. Douki, R. Improta, D. Markovitsi, J. Am. Chem. Soc. **139**, 10561 (2017).

17:20-17:40 30 July 2019

G-quadruplex DNA-mediated ultrafast photodynamics

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DNA G-quadruplexes are noncanonical four-stranded structures formed by specific guanine-rich DNA sequences, by stacking of planar G-tetrads comprising of four guanines *via* Hoogsteen hydrogen bonds. G-quadruplex motifs are prevalent in genomes. G-quadruplex DNA have attracted intense research interests in terms of their biological role, as targets for anticancer therapy and, more recently, of their potential applications in the field of molecular electronics. Here we report our studies on the ultrafast photodynamics involving DNA G- quadruplex by means of transient absorption spectroscopy and time-resolved fluorescence spectroscopy. We will present the results of the direct observation of hole transfer process along vertical direction via G-stacks (38-130 ps), or along horizontal direction via G-tetrad (60-240 ps), from which a mechanism of multitude pathways involving electron coupling of tetrads and stacks has been established to understand the effective conduit of positive charge for G- quadruplex DNA. Moreover, we will present the results of the light-switch and ultrafast photodynamics of the transition metal compound $[Ru(phen)_2(dppz)]^{2+}$ in G-quadruplex distinct from that in ds-DNA, featuring a depopulation process from ^{3}MLCT state to a dark state in G-quadruplex. The inhibited dark state channel in ds-DNA is open in G-quadruplex, but with a slower rate of formation (~ 10 ps) compared to that in water (~ 2.7 ps), indicating that the local bound microenvironment in G-quadruplex is only partially hydrophobic and can allow dynamic entering of water molecules. These ultrafast photodynamics studies provide in-depth mechanistic understandings of the striking hole transfer and structural properties of G-quadruplex DNA.

References:

[1] Blackburn, E. H. Nature, **350**, 569 (1991)

[2] Choi, J.; Park, J.; Tanaka, A.; Park, M. J.; Jang, Y. J.; Fujitsuka, M.; Kim, S. K.; Majima, T. Angew Chem Int Edit, **52**, 1134 (2013)

[3] Arun K. Thazhathveetil, Michelle A. Harris, Ryan M. Young, Michael R. Wasielewski and Frederick D. Lewis, J. Am. Chem. Soc,**139**,1730 (2017)

[4] Kumar, C. V.; Barton, J. K.; Turro, N. J., J. Am. Chem. Soc., **107**,5518 (1985)

[5] Cardin, C. J.; Kelly, J. M.; Quinn, S. J., Chem. Sci., **8**, 4705. (2017)

[6] Wu, L. D.; Liu, K. H.; Jie, J. L.; Song, D.; Su, H. M., J. Am. Chem. Soc. **137**, 259. (2015)

17:40-18:00 30 July 2019

Electronic relaxation of 2-aminopurine-thymine base pairs by femtosecond time-resolved transient vibrational absorption spectroscopy

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The fluorescent analogue 2-aminopurine (2AP) of the canonical nucleobase adenine (6-aminopurine) finds frequent application in molecular biology for probing DNA and RNA structure and conformational dynamics, because it base- pairs with thymine (T) without disrupting the helical structure of DNA. However, the processes that are responsible for the quenching of the fluorescence of the 2AP chromophore in the DNA/RNA microenvironment have remained elusive. Although attempts have been made to ascribe decreased excited-state lifetimes to intra-strand charge-transfer and stacking interactions, possible effects by inter- strand H-bonding have been widely ignored.

We have investigated the electronic relaxation dynamics of the 2AP monomer and H-bonded 2AP·T pairs in Watson−Crick (WC) and Hoogsteen (HS) conformations after UV photoexcitation by means of femtosecond time-resolved transient vibrational absorption spectroscopy (TVAS), which has the advantage over transient electronic absorption spectroscopy (TEAS) of being chemical structure-sensitive. The measurements led to the identification of numerous cha- racteristic vibrational marker bands of the $^1\pi\pi^*$, $^1n\pi^*$ and $^3\pi\pi^*$ electronic states that are involved in the relaxation dynamics of the excited states. The vibrational band assignments were aided by ab initio quantum chemical calculations. The observed time profiles were analyzed by global fitting. The observation of the ground-state recovery of 2AP allowed us to determine the triplet quantum yield [1]. In 2AP·T, the WC conformation features slowed-down, monomer-like elec- tronic relaxation in $\tau \sim 1.6$ ns towards ground-state recovery and triplet formation. In stark contrast, the dynamics associated with 2AP·T in the HS motif exhibits much faster electronic deactivation in $\tau \sim 70$ ps, likely via a conical intersection (CoIn) involving double proton transfer in the excited state [2].

References:

[1] H. Böhnke, K. Röttger, R. A. Ingle, H. J. B. Marroux, M. Bohnsack, A. J. Orr-Ewing, F. Temps, Phys. Chem. Chem. Phys. **20**, 20033 (2018).

[2] H. Böhnke, K. Röttger, R. A. Ingle, H. J. B. Marroux, M. Bohnsack, N. K. Schwalb, A. Orr-Ewing, F. Temps, J. Phys. Chem. B **123**, 2904 (2019).

08:10-08:45 31 July 2019

Ultrafast time-resolved protein crystallography – recent insights

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Light is important for organisms from all domains of life, serving as an energy resource or carrier of information initiating intra- or intercellular signaling. Photosensitive proteins, endowed with a light-absorbing chromophore, enable this. Detailed insight into the ultrafast events has been obtained by various forms of spectroscopy and computation. However, direct structural information necessary to understand the underlying molecular mechanisms has been inaccessible until recently. The unique properties of X-ray free electron lasers open the sub-ps time domain for time-resolved crystallography using small crystals that can be efficiently photolyzed, thus providing access to the long sought-after excited state and intermediate structures. This is not only important for the fundamental understanding of light-driven processes but has practical impact on future developments of e.g. fluorescent proteins for optical nanoscopy or retinal proteins for optogenetics. We present recent insight on the initial events in photodissociation and photoisomerization reactions in proteins obtained by time-resolved serial femtosecond crystallography.

08:45-09:15 31 July 2019

Free energies in protein-protein and protein-ligand bindings

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Theoretical calculation of protein-protein and protein-ligand binding free energies is a grand challenge in computational biology. Accurate prediction of critical residues along with their specific and quantitative contributions to protein-protein binding free energy is extremely helpful to reveal binding mechanisms and identify drug-like molecules that alter protein-protein interactions. In this talk, we discuss methods to quantitatively analyze residue-specific contributions of binding free energies in protein-protein and protein-ligand bindings. Predictions of hot stops for some important protein-protein interactions are discussed.

09:15-09:35 31 July 2019

Ultrafast dynamics in amyloid fibrils

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Accumulation of amyloid fibrils in various locations of the human body is either the symptom or cause of many diseases. Therefore, a large body of research has focused on elucidating their structures, kinetics of formation, assembly mechanisms, and interactions with other biological components. In comparison, much less effort has been given to assessing the conformational dynamics of amyloid fibrils, especially those occurring at a specific sidechain location and on ultrafast time scales. Such dynamics could play an important role in stabilizing the underlying protofibrils as they increase the ‘hidden’ entropy of the system, hence deserving more attention. However, investigating ultrafast fibrillar dynamics in a site-specific manner using optical or vibrational spectroscopy is difficult since native spectroscopic signals often lack the needed specificity due to spectral overlapping and degeneracy. In this talk, we will show how site- specific structural and dynamic information of amyloid assembly can be obtained by measuring the linear and nonlinear infrared spectroscopic signals of a specifically incorporated unnatural amino acid-based vibrational probe. In particular, we will discuss how this strategy can be employed to interrogate the hydrogen-bonding dynamics and the existence of α-sheets in amyloid fibrils.

09:35-09:55 31 July 2019

Characterization and functionalization of the GFP core: From excited state proton transfer to color tuning

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Since green fluorescent protein (GFP) revolutionized molecular and cellular biology about 25 years ago [1], new mechanistic insights into fluorescence mechanisms of the GFP chromophore and its derivatives have been enabled by advanced characterization techniques. Using wavelength-tunable femtosecond stimulated Raman spectroscopy (FSRS) [2,3], aided by femtosecond transient absorption spectroscopy and quantum calculations, we tracked key structural motions along the ultrafast excited state proton transfer (ESPT) coordinate in a series of newly developed super-photoacids [4,6] and red-emitting fluorophores[5] in solution. The site-specific modification of GFP core chromophore (Figure 1) with various electron-donating or withdrawing groups [5,6] at two opposite ends exerts an additive effect on the frontier molecular orbitals, achieving a dramatic redshift of emission with a unique "double-donor-one-acceptor" structure.

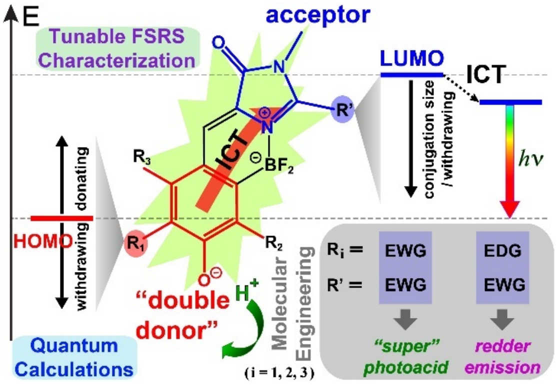

Figure 1. Synergistic tuning of the ground and excited electronic states of the GFP chromophore, aided by femtosecond spectroscopy and semi-empirical calculations, enables the rational design of new biomimetic fluorophores with desirable functions.

References:

[1] M. Chalfie, Y. Tu, G. Euskirchen, W. W. Ward, and D. C. Prasher, Science **263**, 802 (1994).

[2] C. Fang, R. R. Frontiera, R. Tran, and R. A. Mathies, Nature **462**, 200 (2009).

[3] C. Fang, L. Tang, B. G. Oscar, and C. Chen, J. Phys. Chem. Lett. **9**, 3253 (2018).

[4] C. Chen, W. Liu, M. S. Baranov, N. S. Baleeva, I. V. Yampolsky, L. Zhu, Y. Wang, A. Shamir, K. M. Solntsev, and C. Fang, J. Phys. Chem. Lett. **8**, 5921 (2017).

[5] C. Chen, M. S. Baranov, L. Zhu, N. S. Baleeva, A. Y. Smirnov, S. Zaitseva, I. V. Yampolsky, K. M. Solntsev, and C. Fang, Chem. Commun. **55**, 2537 (2019).

[6] C. Chen, L. Zhu, M. S. Baranov, L. Tang, N. S. Baleeva, A. Y. Smirnov, I. V. Yampolsky, K. M. Solntsev, and C. Fang, J. Phys. Chem. B **123**, 3804 (2019).

10:10-10:45 31 July 2019

Structural dynamics of biosystems and materials using ultrashort optical and X-ray pulses

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With the advent of X-ray free electron lasers delivering intense femtosecond pulses of soft to hard X-rays, new insight can be gained into photo-induced processes in matter. In this talk, I will report on recent transient optical and X-ray absorption, as well as transient X-ray emission studies carried on ferric and ferrous hemoproteins that monitor in detail the return to the initial state after photoexcitation (with or without ligand dissociation) of the heme.

Transition metal oxides are an important class of materials due to their potential applications in photocatalysis and photovoltaics. These applications are determined by the generation of charge carriers and their subsequent fate either as free charges or by trapping at defects. I will show recent optical and X-ray studies that allow one to selectively determine the fate of either the electrons or the holes after photoexcitation above the band gap.

10:45-11:15 31 July 2019

Visualizing chemical reactions in space and time using time-resolved X-ray solution scattering

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Chemical reaction is a course of events involving the bond breaking and bond formation, which results in the transformation of reactants to products. To understand the dynamics of chemical reactions, therefore, it is highly desirable to monitor the structures of reacting species during the reaction. In this regard, time-resolved X-ray solution scattering (TRXSS) is a powerful technique to directly capture the reaction dynamics. In TRXSS, temporally short optical laser and X-ray pulse are used as pump and probe, respectively, which allows us to track the reaction with sub-angstrom spatial resolution by virtue of the structural sensitivity of X-ray as well as high temporal resolution. Here, we present two representative works in which bond formation and transient isomer formation have been directly observed using TRXSS. In particular, direct observation of Au- Au bond formation in the photoreaction of a gold trimer complex [1] and formation of transient isomer of bismuth complex during a photoreaction of bismuth triiodide in solution [2] will be described. Detailed kinetics and structures of intermediates involved in both of the reactions will also be presented.

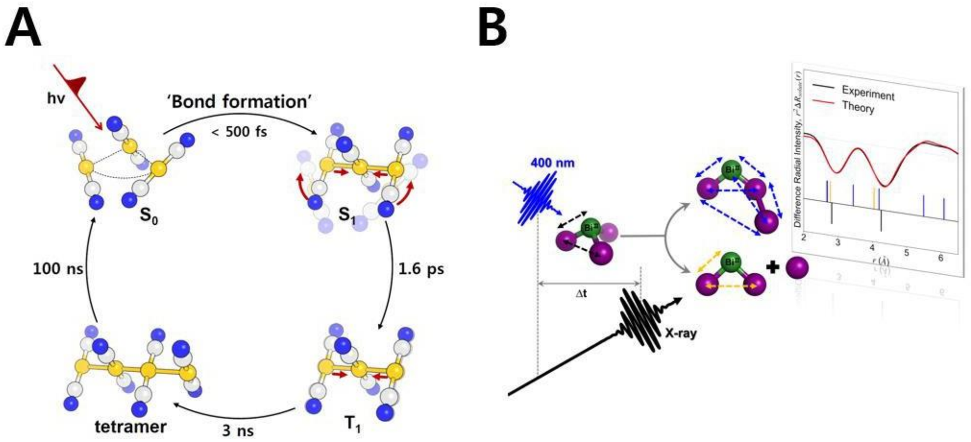

Figure 1. Structural dynamics of gold trimer (A) and bismuth triiodide (B).

References:

[1] K. H. Kim and J. G. Kim et al. Nature **518**, 385 (2015).

[2] E. H. Choi and D. –S. Ahn et al. J. Phys. Chem. Lett **10**, 1279 (2019).

11:15-11:35 31 July 2019

Nonadiabatic dynamics probed via time-resolved X-ray spectroscopy

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Developments in ultrafast X-ray sources offer powerful new means of probing the complex nonadiabatic dynamics in photoexcited molecules. In particular, the electronic structures associated with conical intersections are predicted to have unique spectral signatures when probed using X-ray spectroscopies[1]. Using high level *ab initio* quantum dynamics simulations and novel electronic structure methods, we have simulated time-resolved X-ray spectroscopy (TRXAS and TRXPS) of a number of prototypical unsaturated organic chromophores (including ethylene, allene, and butadiene). The techniques are highly sensitive to all aspects of the ensuing dynamics and can distinguish between the different nuclear dynamical pathways that result in electronic relaxation. The ability of X- ray spectroscopies to provide a clear picture of the wavepacket dynamics near conical intersections is related to transient charge localization at specific atomic sites that is driven by the nuclear dynamics. Given the ubiquity of this type of charge localization in excited state dynamics, ultrafast X-ray spectroscopies offer a unique and powerful route to the direct observation of dynamics around conical intersections.

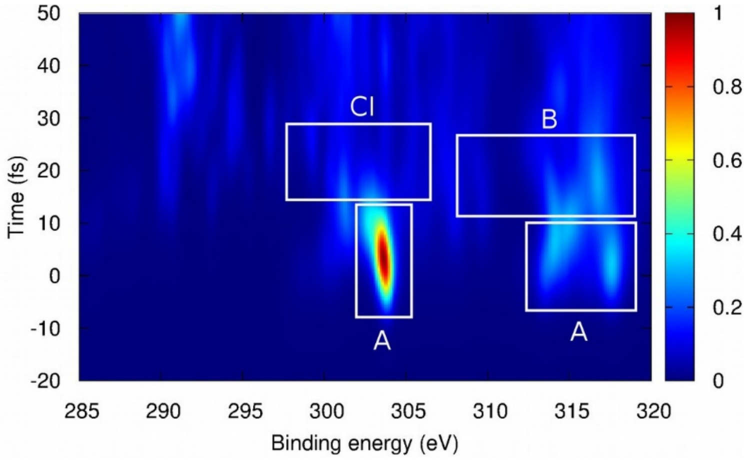

Figure 1. Simulated TRXAS of ethylene following VUV photo-excitation.

References:

[1] S. P. Neville, M. Chergui, A. Stolow, M. S. Schuurman, Phys. Rev. Lett., **120**, 243001 (2018).

11:35-11:55 31 July 2019

What did the metals know, and when did they know it? Femtosecond M-edge XANES reveals short-lived states in transition metal complexes and organohalide perovskites

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X-ray absorption near edge spectroscopy (XANES or NEXAFS) is a powerful technique for electronic structure determination. However, widespread use of XANES is limited by the need for synchrotron light sources with tunable x-ray energy. Recent developments in extreme ultraviolet (XUV) light sources using the laser-based technique of high-harmonic generation have enabled core-level spectroscopy to be performed on femtosecond to attosecond timescales. We have extended the scope of tabletop XUV spectroscopy and demonstrated that $M_{2,3}$-edge XANES, corresponding to 3p→3d transitions, can reliably measure the electronic structure of first-row transition metal coordination complexes with femtosecond time resolution. We use this ability to track the excited-state relaxation pathways of photocatalysts and spin crossover complexes. In semiconductors such as $CH_3NH_3PbI_3$, distinct signals are observed for photoinduced electrons and holes, allowing the dynamics of each carrier to be tracked independently. This work establishes extreme ultraviolet spectroscopy as a useful tool for mainstream research in inorganic, organometallic, and materials chemistry.

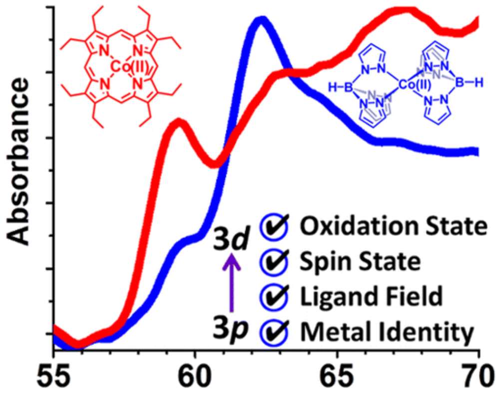

Figure 1. Tabletop M-edge XANES spectroscopy probes 3p→3d transitions and measures the electronic structure of transition metal complexes at fs timescales.

11:55-12:15 31 July 2019

Direct mapping of photochemical dynamics across conical intersections by XUV transient absorption spectroscopy

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XUV transient absorption spectroscopy is utilized to map the transit of a wave packet across a conical intersection in the photodissociation of *tert*-butyl iodide (t-C_4H_9I) and *iso*-propyl iodide (i-C_3H_7I). UV excitation of the alkyl iodides at 277 nm launches a wave packet on the repulsive $n\sigma^*$ triplet state $^3Q_0$, initiating rapid dissociation along the C-I bond (Fig. 1(a)). As the molecule begins to fragment, a conical intersection crossing with the $n\sigma^*$ singlet $^1Q_1$ state allows the wave packet path to be redirected from $^3Q_0$ to $^1Q_1$ [1]. Since C-I bond-breaking along $^1Q_1$ releases I atoms whereas dissociation along $^3Q_0$ yields I*, a nearly complete transfer of the wave packet across the conicalintersection results in the selective formation of atomic I as the primary photoproduct. In the case of t-C_4H_9I and i-C_3H_7I the observed I/I* product ratios are ~13:1 and ~2:1, respectively [2]. To track the dissociation in real time, gaseous alkyl iodide molecules are excited by a UV pump (277 nm, 50 fs) and probed by XUV pulses (40-70 eV, ~200 as) targeting I(4*d*) core-to-valence absorptions. In the measured transient absorption spectra of both t-C_4H_9I and i-C_3H_7I (Fig. 1(b)), a prominent resonance at ~47.1 eV assigned to the initial $^3Q_0$ wave packet appears within the pump duration and decays within 100 fs. Concomitant with the $^3Q_0$ decay, a synchronized rise of a ~45.6 eV resonance assigned to $^1Q_1$ is observed, signifying non-adiabatic wave packet transfer across the $^3Q_0$-$^1Q_1$ conical intersection. Analysis of the $^3Q_0$ decay in t-C_4H_9I reveals a35± 7 fs time constant for the wave packet to navigate the conical intersection. For i-C_3H_7I, this time constant is measured to be greater than 65 ± 5 fs. In both molecules, a decay of the $^1Q_1$ resonance into rising atomic I resonances confirms that, once the crossing is complete, the majority of C-I dissociation resumes along the $^1Q_1$ surface.

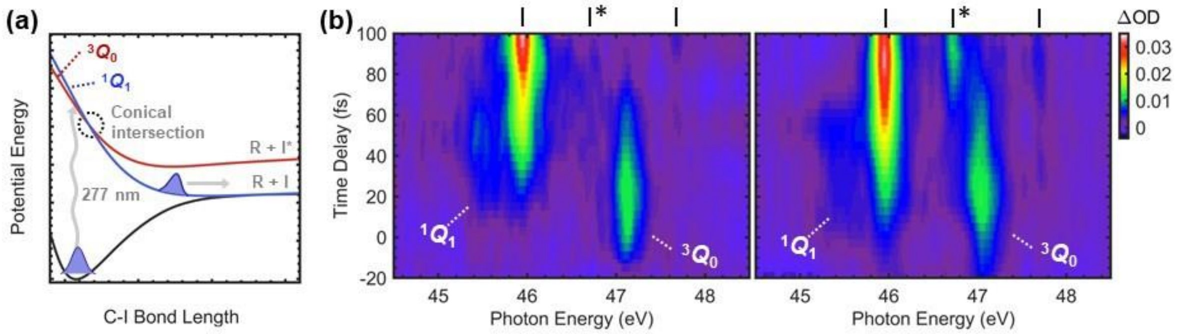

Figure 1. (a) *Potential energy curves of the alkyl iodides (R-I) plotted along the C−I bond.* ***(b)****Time-dependent XUV absorption spectrum of tert-butyl iodide (left) and iso-propyl iodide (right).*

References:

[1] M. E. Corrales, V. Loriot, G. Balerdi, J. González-Vázquez, R. De Nalda, L.Bañares, and A.H. Zewail. Phys. Chem. Chem. Phys. **19**, 8812 (2014).

[2] X. P. Zhang, W. B. Lee, and K. C. Lin. J. Phys. Chem. A. **113**, 35 (2009).

12:15-12:35 31 July 2019

Ion effect on hydrogen bonding network in water

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The effect of hydrated ions on the dynamics of surrounding water have significant specificity which is far from being fully comprehended. Various vibrational spectroscopies are among the most powerful experimental tools to explore this issue. The interpretation of these spectroscopy signals is, however, usually non-trivial and requires the help from theoretical studies. We've developed a series of theoretical approaches to simulate and analyze the vibrational spectroscopies of the ionic solution, which reproduce nicely the spectra including THZ, Raman, fsIR, 2DIR, IRPD and Raman-THZ. Based on these simulations, we attempt to address several important issues about the ion effect on water hydrogen bonding network, including 1) ion specificity in their effects on water dynamics and 2) spatial range of ion effects. Novel techniques including complex network recognition and gaussian field model are employed to assist the analysis.

12:35-12:50 31 July 2019

UltraFast TEM, a manufacture’s perspective

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For FEI (the legacy company acquired by Thermo Fisher Scientific in 2016) UltraFast TEM began as a request from Prof. Zewail for a modified TEM. The first system delivered to Caltech was a Tecnai 12 and referred to as UEM-1 and it was soon followed by UEM-2 a modified Tecnai F20. These prototypes would later be followed by a commercial offering the Tecnai Femto. Now that the Tecnai product line was discontinued Thermo Fisher Scientific is working in partnership with IDES to develop the Talos Femto, basis for the next generation of UltraFast TEM’s. The Talos Femto which will come in two dedicated flavors stroboscopic and single shot will be presented.

08:10-08:45 1 August 2019

Beyond the molecular movie: The ultrafast electronic structure view of surface dynamics

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In a Born-Oppenheimer description, atomic motions evolve across a potential energy surface determined by the occupation of electronic states as a function of nuclear coordinates. A key goal of structural dynamics is therefore to record a “molecular movie” of the atom positions in a dynamically evolving system. Ultrafast photo-induced phase transitions provide a test case for how the forces and resulting nuclear motion along the reaction co-ordinate originate from a non- equilibrium population of excited electronic states. Moreover, small changes of nuclear positions in solids can correlate with dramatic electronic structure changes (e.g. conductivity). In this talk I discuss recent advances in time- resolved spectroscopy allowing for direct probing of the underlying fundamental steps of ultrafast non-equilibrium dynamics of solids and surfaces like ultrafast phase transitions, coherent lattice excitations or chemical reactions at surfaces. Utilizing photoemission with fs XUV laser pulses at 500 kHz (trARPES) [1], we obtain direct access to the transient electronic structure during the ultrafast phase transition in indium nanowires on Si(111) and lay out a detailed reaction pathway including temporally separated transitions of electronic and atomic structure [2]. While such spatially periodic systems imply that all unit cells react collectively, ultrafast surface reactions are often induced by quasi-thermal activation and fluctuations. An appropriate probe of local electronic structure changes is time-resolved resonant inelastic x-ray scattering (trRIXS) providing direct insight into chemical bond formation [3]. Finally I will briefly discuss some recent attempts to access the space-time limit in surface dynamics using scanning probe techniques.

References:

[1] M. Puppin et al., Rev. Sci. Instrum. **90**, 023104 (2019)

[2] C.W. Nicholson et al., Science **362**, 821 (2018)

[3] H Öström et al., Science **347**, 978 (2015)

08:45-09:15 1 August 2019

Mapping atomic motions with ultrabright electrons: Fundamental space-time limits to imaging chemistry

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One of the dream experiments in science has been to watch atomic motions on the primary timescales of chemistry. This prospect would provide a direct observation of the reaction forces, the very essence of chemistry, and the central unifying concept of transition states that links chemistry to biology. This experiment has been referred to as "making the molecular movie" with respect to observing net rms atomic motions during structural changes. There are not only extraordinary requirements for simultaneous spatial-temporal resolution but equally important, due to sample limitations, also one on source brightness. Taking the problem to be an imaging problem, as one makes the shutter speed shorter and shorter, a brighter and brighter source is needed to maintain image contrast. With the development of ultrabright electron capable of literally lighting up atomic motions, this experiment was first realized (Siwick et al Science 2003) and efforts accelerated with the onset of XFELs (Miller, Science 2014). A number of different chemical reactions will be discussed from electrocyclization with conserved stereochemistry (Jean-Ruel et al JCP B 2013), intermolecular electron transfer for organic systems (Gao et al Nature 2013; Liu et al Chem Phys Lett 2017), metal to metal electron transfer (Ishikawa et al, Science 2015), to the recent observation of coherently directed bond formation using the classic I_3^- system, in a process analogous to a quantum Newton's cradle (Xian et al Nature Chem 2017). These studies have discovered that these nominally 100+ dimensional problems, representing the number of degrees of freedom in the system, distilled down to atomic projections along a few principle reaction coordinates. The specific details depend on the spatial resolution to these motions. The most dramatic example will be shown for the simplest form of chemistry, electron transfer, which is the heart of redox chemistry. No bonds are made or broken. The reaction coordinate is dictated by medium repolarization that stabilizes the charge transfer state. This motion was captured with sub-Å (.01 Å) and 100 fs timescale resolution (Ishakawa, Hayes et al Science 2015) – the fundamental space-time resolution to following the primary processes of chemistry. Without any detailed analysis, the key large-amplitude modes can be identified by eye from the molecular movie. This reduction in dimensionality appears to be general, arising from the very strong anharmonicity of the many body potential in the barrier crossing region. We now are beginning to see the underlying physics for the generalized reaction mechanisms that have been empirically discovered over time. The "magic of chemistry" is this enormous reduction in dimensionality in the barrier crossing region that ultimately makes chemical concepts transferrable. How far can this reductionist view be extended with respect to complexity? The ultimate goal in scaling system complexity is to obtain atomically resolved protein functions to understand how nature tamed chemistry over all conceivable length scales. New approaches based on the prinicples used for femtosecond electron diffraction, both in terms of source technology and image reconstruction, hold promise for real space studies of single biomolecules. This prospect is within reach and will provide a definitive test of the collective mode coupling model (Miller Acc. Chem. Research 1994) to bridge chemistry to biology, which will be discussed as the driving force for this work.

09:15-09:35 1 August 2019

Revealing the influence of local structure on lattice dynamics with ultrafast electron microscopy

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Spatially-localized dynamics that contribute to ensemble-averaged signals are influenced by ever-present lattice discontinuities, nanoscale morphological structures, and interfaces, the distribution of which is heterogeneous over nanoscale volumes. Thus, probing of local responses can provide a richer, more detailed picture of the formation, evolution, and decay of ultrafast non-equilibrium energy transport and conversion in functional materials. Here, I will discuss how we have used fs electron imaging with an ultrafast electron microscope (UEM) to visualize phonon dynamics in a variety of materials, with particular emphasis placed on understanding the influence of lattice discontinuities and structural anisotropies. After a brief overview of UEM [1], I will describe how the concepts of conventional bright-field imaging can be extended to UEM to visualize local coherent phonon dynamics. In TMDs we have found that fs photoexcitation leads to the generation of phonon wavetrains preferentially at vacuum-crystal interfaces and crystal step edges [2-4]. This arises *via* an initial impulsive expansion along the *c*-axis van der Waals stacking direction occurring within the first few picoseconds, as indicated by local oscillatory bend-contour motions. Impulsive excitation of this *interlayer* low-frequency breathing mode induces the launch of coherent *ab*-plane phonon wavefronts due to the rapid, picosecond development of a phase lag between the neighboring layers owing to varying total transit times of the speed-of-sound *c*-axis phonons. As with the stacking direction, the coherent intralayer modes propagate at the speed of sound and initially along a single wavevector oriented perpendicular to the defect nucleation sites prior to the first scattering events. Aspects of this behavior are in contrast to those in strongly-photoexcited Ge, in which a number of remarkable responses have been observed with UEM [5]; including the launch of highly-coherent phonon wavefronts propagating with hypersonic phase velocities, the significantly-delayed (*i.e.*, 10s of picoseconds or more) generation of phonon wavetrains relative to the precise moment of fs photoexcitation, and the *time- varying* phase-velocity dispersions displaying single-exponential relaxation to the bulk speed of sound. This survey of recent results will serve to illustrate the rich and detailed information obtainable with fs electron imaging.

References:

[1] D. Plemmons, P. Suri, D. Flannigan, Chem. Mater. **27**, 3178 (2015).

[2] D. Cremons, D. Plemmons, D. Flannigan, Nat. Commun. **7**, 11230 (2016).

[3] A. McKenna, J. Eliason, D. Flannigan, Nano Lett. **17**, 3952 (2017).

[4] D. Cremons, D. Plemmons, D. Flannigan, Struct. Dyn. **4**, 044019 (2017).

[5] D. Cremons, D. Du, and D. Flannigan, Phys. Rev. Mater. **1**, 073801 (2017).

09:35-09:55 1 August 2019

Development and application of UTEM in material science

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Structural dynamics and transient states after a pulsed femtosecond laser excitation in new quantum materials attract considerable attention. Recent advances in the ultrafast transmission electron microscope (UTEM), with high spatiotemporal resolution, have made it possible to directly visualize the rapid changes of lattice, electron, and magnetic structures in materials [1]. In the past years, we successfully developed UTEMs at the Institute of Physics, Chinese Academy of Sciences (IOP, CAS), based on a JEOL-2000EX and a JEOL-2100F microscope. Then these UTEMs have been extensively improved for photoemission imaging and time-resolved observation in several experimental measurements, including ultrafast electron diffraction/imaging, Lorentz UTEM, time-resolved electron energy-loss spectroscopy and photoinduced near-field electron microscopy (PINEM)).

Based on the UTEM platform in our laboratory, we have also performed a variety of investigations on the structural dynamics of functional materials. For instance, the notable anisotropic lattice dynamics in multi-walled carbon nanotubes (MWCNTs) has been studied in a full reversible cycle with the time scale ranging from picoseconds to hundreds of microseconds [2]; The martensitic transition in the Heusler alloy $Mn_{50}Ni_{40}Sn_{10}$ has also been investigated using the time- resolved images and diffraction patterns [3]. The photoinduced magnetic processes in half-Heusler alloy NiMnGa have been directly observed using time- resolved Lorentz TEM imaging. Very recently, the nonequilibrium phase transition between CDW states and the relevant hidden quantum states has been studied and analyzed in 1T-$TaS_{2-x}Se_x$ at low temperatures [4].

References:

[1] H. Yang, S. Song, M. Zhang, et al., Chin. Phys. B, **27**, 70703 (2018).

[2] Z. Li, S. Sun, Z.-A. Li, et al., Nanoscale, **9**, 13313 (2017).

[3] M. Zhang, G. Cao, H. Tian, et al., Physical Review B, **96**, 174203 (2017).

[4] K. Sun, S. Sun, C. Zhu A. et al., Science Advances, **4,** eaas9660 (2018).

10:10-10:45 1 August 2019

Attosecond science: From tracing electrons to cancer detection

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attoworld.de, ferenckrausz.de

Born around the turn of the new millennium, attosecond metrology has provided real-time insight into atomic-scale electron motions and light field oscillation, previously inaccessible to human observation. Until recently, this capability has relied on attosecond extreme ultraviolet pulses, generated and measured in complex vacuum systems. Next-generation attosecond metrology is now about to change this state of matters profoundly. Sub-femtosecond current injection into wide-gap materials can directly probe ultrafast electron phenomena in condensed matter systems and can also be used for sampling the electric field of light up to ultraviolet frequencies. Petahertz field sampling draws on a robust solid-state circuitry and routine few-cycle laser technology, opening the door for complete characterization of electromagnetic fields all the way from the far infrared to the vacuum ultraviolet. These fields, with accurately measured temporal evolution, serve as a unique probe for the polarization response of matter. Field-resolved spectroscopy will access valence electronic as well as nuclear motions in all forms of matter and constitutes a generalization of pump-probe approaches. Its implementation with a solid-state instrumentation opens the door for real-world applications, such as early cancer detection by measuring miniscule changes of the molecular composition of blood via field-resolved vibrational molecular fingerprinting.

10:45-11:15 1 August 2019

Ultrafast many-body electron dynamics in a strongly correlated ultracold rydberg gas

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Many-body correlations govern a variety of important quantum phenomena including the emergence of superconductivity and magnetism in condensed matter as well as chemical reactions in liquids. Understanding quantum many-body systems is thus one of the central goals of modern sciences and technologies. Here we demonstrate a new pathway towards this goal by generating a strongly correlated ultracold Rydberg gas with an ultrashort laser pulse. We have applied our ultrafast and ultrahigh-precision coherent control with attosecond precision [1-8] to this strongly correlated Rydberg gas, and have successfully observed and controlled its ultrafast electron dynamics [9-11]. Our approach will offer a new platform for the quantum simulation of strongly correlated quantum many-body dynamics on the ultrafast timescale [12].

References:

[1] K. Ohmori et. al., Phys. Rev. Lett. **91**, 243003 (2003).

[2] H. Katsuki et. al., Science **311**, 1589 (2006).

[3] K. Ohmori et. al., Phys. Rev. Lett. **96**, 093002 (2006).

[4] H. Katsuki et. al., Phys. Rev. Lett. **102**, 103602 (2009).

[5] K. Hosaka et al., Phys. Rev. Lett. **104**, 180501 (2010) (Highlighted by Nature **465**, 138 (2010); Physics **3**, 38 (2010)).

[6] H. Goto et al., Nature Physics **7**, 383 (2011)

[7] (Highlighted by Nature Physics **7**, 373 (2011); Nature Photonics **5**, 382 (2011)).

[8] H. Katsuki et al., Nature Commun. **4**, 2801 (2013).

[9] H. Katsuki et al., Phys. Rev. B **92**, 094511 (2015).

[10] N. Takei et al., Nature Commun. **7**, 13449 (2016) (Highlighted by Science **354**, 1388 (2016); IOP PhyscisWorld.com (2016)).

[11] H. Katsuki et al., Acc. Chem. Res. **51**, 1174 (2018). [11] C. Liu et al., Phys. Rev. Lett. **121**, 173201 (2018).

[12] Patent Publication Number: US 2018/0292786 A1; JAPAN 2018-180179, “Quantum simulator and quantum simulation method,” H. Sakai (Hamamatsu Photonics K.K.), K. Ohmori (NINS) et al., Publication date: Oct. 11, 2018 (US); Nov. 15, 2018 (JAPAN).

11:15-11:35 1 August 2019

Advances in femtosecond electron imaging and spectroscopy with high-brightness beam

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The advances of ultrafast electron-based imaging and spectroscopy technologies in the last decades have enabled a new perspective in understanding the fundamental processes at far from equilibrium[1]. Ranging from molecular assemblies to systems on the nanometer scale, visualizing the transient evolution involving coupling between photon field, charge carrier and the lattice degrees of freedom allows the interaction hierarchy to be analyzed [2-5]. These photoinduced nonequilibrium dynamics are not only central for understanding the complex interactions, but also may yield new ways for controlling the processes. An emerging area is to directly resolve the relevant processes in an ultrafast electron microscope where the local properties and morphology can be distinguished from the ensemble averages, relevant for more precise material design and integration with many technical implications. I will discuss the progresses using electron- based approaches to image the bond breaking, charge carrier flow, and electronic switching in photo-induced phase transitions in nanostructures and 2D materials. I will also discuss methods to combine diffraction, imaging, and spectroscopy in a single setup to follow many key degrees of freedom at once as a next step to resolve complex dynamics in the development of high-intensity ultrafast electron microscopy systems [6,7,8].

References:

[1] H. Zewail, Science **328**, 187 (2010).

[2] Z. Tao et al., Phys. Rev. Lett. **109**, 166406 (2012).

[3] T.R. T. Han et al., Sci. Adv. **1**, e1400173 (2015).

[4] Z. Tao et al., Sci. Rep. **6**, 38514 (2016).

[5] F. Zhou et al., arXiv 1904.07120 (2019).

[6] F. Zhou, J. Williams, C.-Y. Ruan, Chem. Phys. Lett. **683**, 488 (2017).

[7] J. Williams et al., Struc. Dyn. **4**, 044035 (2017).

[8] Zerbe et al. Phys. Rev. Accel. Beams **21**, 064201 (2018).

11:35-11:55 1 August 2019

Ultrafast nanoscale dynamics probed by time-resolved electron microscopy

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Ultrafast transmission electron microscopy (UTEM) is an emerging technique, enabling the investigation of ultrafast dynamics on nanometer length scales [1,2]. In UTEM, a pulsed electron beam with sub-picosecond bunch duration is utilized to stroboscopically probe optically triggered processes. Dynamics in structural, electronic and spin degrees of freedom are generally accessible in UTEM by utilizing the versatile imaging and diffraction capabilities of state-of-the-art electron microscopes. However, up to now, the broad applicability of UTEM was limited by the coherence properties of available pulsed electron sources.

In the Göttingen UTEM project, we developed nanoscale laser-driven photocathodes, which allow for the generation of electron pulses with largely improved coherence properties. With this approach, we achieve, at the sample position, electron focal spot sizes down to below one nanometer and pulse durations of about 200 fs [2,3].

In the talk, our methodological developments in the Göttingen UTEM project will be summarized, covering applications of coherent optical phase modulation of electron pulses in optical near-fields [4] for the formation of attosecond electron pulse trains [5], and the demonstration of the nanoscale mapping of optically induced ultrafast structural and magnetic dynamics [6,7]. Furthermore, the current progress in the Oldenburg UTEM project will be reported, with first experiments on the coupling of free-electron states with high-quality whispering gallery modes.

References:

[1] A. H. Zewail, Science **328**, 187–93 (2010).
[2] A. Feist et al., Ultramicroscopy **176**, 63-73(2017).
[3] N. Bach et al., Structural Dynamics **6**, 014301 (2019).
[4] A. Feist et al., Nature **521**, 200–203 (2015).
[5] K. E. Priebe et al., Nature Photonics **11**, 793 (2017).
[6] A. Feist et al., Structural Dynamics **5**, 014302 (2018).
[7] N. Rubiano et al., Phys. Rev. X **8**, 031052 (2018).

11:55-12:15 1 August 2019

Field-resolved infrared molecular fingerprinting for cancer detection

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The molecular composition of biofluids, such as blood, is an indicator of human physiological states. Broadband mid-infrared spectroscopy carries great potential for biological and biomedical applications, as it provides fast and label-free access to the molecular composition of biofluids. One remaining challenge is the complexity of such biofluids. While physiological phenotypes are driven by minor changes in the concentration of thousands of different molecules and many low- abundance molecules are very informative for disease detection, they are often not detectable with conventional Fourier-Transform Infrared (FTIR) spectroscopy and quantum-cascade laser (QCL) based approaches, due to the lack of sensitivity and specificity.

Here we show how field-resolved spectroscopy (FRS) of few-cycle-excited molecular vibrations can be utilised to address the above shortcomings. Direct sampling of the infrared field oscillations, emerging from the excited sample, permits temporal separation of the molecular-composition-specific signal. We have developed an FRS approach for infrared molecular fingerprinting of liquid biopsies and for the first time apply it in a clinical study comparing blood sera of lung cancer, prostate cancer, and breast cancer patients with sera of well-matched cohorts of control individuals. Our preliminary analysis of correlated spectral changes of time-domain sampled field oscillations with a machine learning classifier reveals a high accuracy for cancer detection, especially for lung cancer.

Even though FRS is in an early stage of development with limited spectral coverage in comparison to FTIR, it has the ability to detect cancer with similar efficiency. This suggests that next generation FRS holds promise for future infrared molecular fingerprinting in molecular medicine.

13:55-14:45 1 August 2019

Plasmonic arrays

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This talk will overview the interplay between optics, plasmonics, and excitonics for structures that consist of arrays of gold, silver or aluminum nanoparticles in 1D, 2D and 3D. Particular emphasis will be on arrays whose spacing satisfies a diffraction condition as this produces hybrid plasmon-polariton modes known as lattice plasmons. These modes combine two desirable properties, narrow and controllable spectral lines, and strong local field enhancements, and this has resulted in a number of plasmon-enhanced effects (absorption, scattering, SERS, stimulated emission). The talk will focus on the unusual extinction and scattering properties of the lattices including quadrupole resonance effects for aluminum lattices, and the recent discovery of optical modes that are turned on by dipole- quadrupole coupling between particles. In addition, several examples of emission properties that are mediated with lattice plasmons are presented, including examples of both weak and strong coupling. We also describe the properties of lattice plasmon lasers, including the influence of quadrupole effects, of lattice symmetry, and of lattice stretching. An emphasis will be on the theory of lattice plasmons, which for lasers requires combining electrodynamics with a quantum description of the dye photophysics. I will also talk about recent work on 3D superlattices of plasmonic particles, and on exciton transfer between dye molecules that is mediated by plasmonic nanoparticles.

14:45-15:20 1 August 2019

Geometric effects in chemical reaction

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It was long predicted theoretically that geometric phase effect could have a significant effect in the dynamics of chemical reactions with conical intersections. However, experimental observation of such effect proves to be extremely difficult and fruitless. Recently, we have developed a new crossed beams imaging machine and observed the geometric phase effect in the $H+HD \rightarrow H_2+D$ reaction at collision energy above the conical intersection of this reaction using the high resolution threshold ionization - imaging technique. In addition, we have also detected experimental evidence of geometric phase effect, through quantum interference between topological pathways, at collision energies significantly lower than the conical intersection energy using high resolution H-atom Rydberg tagging technique. These new experiments in combination with accurate quantum dynamics calculations allows us to probe and understand this interesting dynamics effect in chemical reaction at the most fundamental level.

15:20-15:55 1 August 2019

A femtosecond trajectory over a few decades

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Our foray into femtochemistry started with a measurement of bond formation through photo-induced harpooning in liquid xenon [1]. I submitted the paper to CPL, and it was Zewail, the editor, who suggested the change in the original submission to include "femtochemistry" in the title. It was during the 1st Femtochemistry conference that we succeeded in recording the first time-resolved measurements of bond-breaking, remaking and vibrational dynamics in what was to become a model system for some years to follow – I_2 in rare gas solids. There were bets against this possibility, since it was obvious from the absorption spectra that the collective quantum phases in the condensed phase did not survive a vibrational period. Seeing oscillatory signals was all the rage at the time, since they implied coherence, which carried an almost magical connotation then. The fluorescence detected pump-probe signals, including measurements with chirped-pulses as some of the earliest demonstrations of coherence control in condensed matter, could be simulated classically. I reported these results at the 2nd Femtochemistry conference in Lausanne, to declare that what was prepared and measured was classical coherence – nothing quantum mechanical about it. A challenge raised in the Q&A that followed was to consider Raman processes. Indeed, explicit treatments of Raman requires bilinear algebra, for which we resorted to semiclassical dynamics using ensembles of trajectories propagated on multiple electronic surfaces and correlated according to time-circuit diagrams (double-sided Feynman diagrams). Experimentally, Q-coherences could be prepared and time-resolved through 4-wave mixing processes, with the most incisive experiments realized through non-collinear measurements using three independently tunable incident pulses. Preparation of Schrodinger cat states, environment induced recoherence, tomographic state reconstruction and direct observation of the Wigner hole, were among the deeper quantum thought experiments that were realized in related model systems. These remained outside the reach of theory up until very recently, where quantum atomistic treatments of systems as large as I_2/Kr_{18} embedded in a solid environment was made possible through multi-configurational time-dependent Hartree (MCTDH) methods [2,3]. Fundamentals of decoherence and emergence of classicality are among the concepts that can now be scrutinized by closing the loop between experiment and theory. Applications, such as the implementation of quantum algorithms using nonlinear optics and molecular Hamiltonians, was another direction that we pursued [4]. These suffered from the inherent classicality of ensembles. To reach utility, implementations on individual molecules was deemed essential, and this was one of the motivations to pursue time [5] and frequency [6] domain spectroscopy within individual molecules. That effort lead to the first experimental observation of vibrational normal modes in real space, through Raman spectromicroscopy with Å-scale spatial resolution [7]. This opens a new chapter in light-matter interactions, where atomically confined light allows the preparation and interrogation of nonlocal superpositions of extended states, such as phonons and charge density waves in real space. The trajectory of femtochemistry takes a new turn in this limit, as I will expand.

References:

[1] Zadoyan, R.; Apkarian, V. A. Femtochemistry of the Liquid Phase Laser-Induced Harpoon Reaction between Cl2 and Xe. Observation of Bond Formation in Real Time. *Chemical Physics Letters* **1993**, *206*.

[2] Picconi, D.; Cina, J. A.; Burghardt, I. Quantum Dynamics and Spectroscopy of Dihalogens in Solid Matrices. I. Efficient Simulation of the Photodynamics of the Embedded I 2 Kr 18 Cluster Using the G-MCTDH Method. *Journal of Chemical Physics* **2019**, *150*, 1–16.

[3] Picconi, D.; Cina, J. A.; Burghardt, I. Quantum Dynamics and Spectroscopy of Dihalogens in Solid Matrices. II. Theoretical Aspects and G-MCTDH Simulations of Time-Resolved Coherent Raman Spectra of Schrödinger Cat States of the Embedded I 2 Kr 18 Cluster. *Journal of Chemical Physics* **2019**, *150*, 1–15.

[4] Zadoyan, R.; Kohen, D.; Lidar, D. A.; Apkarian, V. A. The Manipulation of Massive Ro- Vibronic Superpositions Using Time–Frequency-Resolved Coherent Anti-Stokes Raman Scattering (TFRCARS): From Quantum Control to Quantum Computing. *Chemical Physics* **2001**, *266*, 323–351.

[5] Yampolsky, S.; Fishman, D. a.; Dey, S.; Hulkko, E.; Banik, M.; Potma, E. O.; Apkarian, V. a. Seeing a Single Molecule Vibrate through Time-Resolved Coherent Anti-Stokes Raman Scattering. *Nature Photonics* **2014**, *8*, 650–656.

[6] Lee, J.; Perdue, S. M.; Rodriguez Perez, A.; Apkarian, V. A. Vibronic Motion with Joint Angstrom-Femtosecond Resolution Observed through Fano Progressions Recorded within One Molecule. *ACS nano* **2014**, *8*, 54–63.

[7] Lee, J.; Crampton, K. T.; Tallarida, N.; Apkarian, V. A. Visualizing Vibrational Normal Modes of a Single Molecule with Atomically Confined Light. *Nature* **2019**, *568*, 78–82.

16:10-16:45 1 August 2019

Understanding tip-enhanced Raman scattering single molecule images

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Tip-enhanced Raman spectroscopy (TERS) exhibits new selection rule and sub- nanometer spatial resolution, which is attributed to the plasmonic near-field confinement. Despite recent advances in simulations of TERS spectra under highly confined fields, a simply physical mechanism has remained elusive. Here we will show that single- molecule TERS images can be explained by local sub-molecular density changes induced by the near-field during the Raman process. The local sub-molecular density changes determine the spatial resolution in TERS and the gradient-based selection rule.

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References:

[1] X Chen, P Liu, Z Hu, L Jensen, ChemRxiv, 2019

16:45-17:20 1 August 2019

Controlling electron photoemission from plasmonic nanostructures: New tools and insights from ultrafast spectroscopy

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Chemists have been amazingly successful with the manipulation of complicated molecular systems, whereas to a physicist, even simple molecules can seem overwhelmingly complex. This talk will provide an overview of work in my labs that attempts to address complex molecular systems but with a physical chemist’s eye toward finding the underlying simplicity. Collective electron oscillation in noble metal Au, Ag, and Cu nanostructures (i.e., “plasmons”) leads to intense absorption strengths and offers enormous potential for real-world applications in medicine, solar energy and photonics. In this talk, I will describe how the novel combination of ultrafast OPO lasers, in-vacuo microscopy, electron photoemission, and velocity map imaging (VMI) provides a novel and remarkably sensitive experimental platform for exploring the fundamental chemical physics and spectroscopy of nanoplasmonic materials. In particular, I will focus on recent experimental results from our labs involving ultrafast scanning photoionization imaging microscopy (SPIM), whereby ultrafast lasers are used to excite a variety of shaped “designer” plasmonic nanomaterials (nanostars, nanorods, spheres, cubes, shells, etc.) via dual beam near IR-UV pump-probe (n=2) or near IR multiphoton (n=3,4) photon excitation. One key result from these VMI studies is the demonstration of *all-optical control* of electron photoemission *intensity* and *directionality* by time dependent control of laser frequency and polarization state, which suggests extremely bright, time dependent sources for ultrafast electron microscopy and reaction dynamics on/near shaped plasmonic nanostructures.

17:20-17:40 1 August 2019

Strong-field dynamics of molecules: Electron-nuclear correlation

X. Gong[1,*], P. Lu[1], W. Zhang[1], J. Ma[1], H.-X. Li[1], F. Sun[1], J. Qiang[1], H. Li[1], and J. Wu[1,*]

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As the primary stage of the light-molecule interaction, the photon energy deposited into the electrons and nuclei governs the photoionization process. Until recently, the electron-nuclear sharing of the absorbed photon energy in strong- field multiphoton dissociative ionization of molecules was revealed in the simplest molecule [1]. However, the experiments showed negligible photon energy sharing between the emitted electrons and ionic fragments in the double ionization of polyatomic hydrocarbon molecule [2,3]. By coincidently measuring the momenta of photoelectron and dissociative fragments, we clarified the generation picture of molecular Rydberg fragments in strong-laser fields [4], and observed the coexistence of high-order above-threshold dissociation [5] and high-order above- threshold ionization in molecular dissociative ionization as sketched in Fig. 1. Our results give a perspective to disentangle the complex electron–nuclear correlation in molecules.

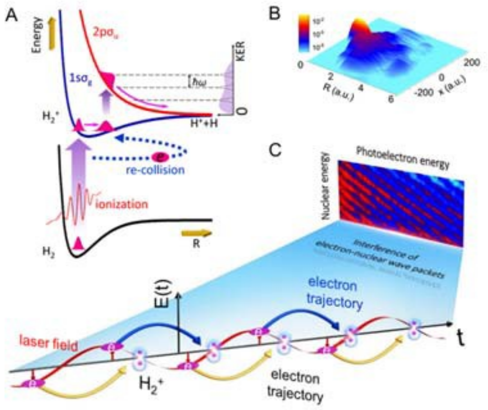

Figure 1. Interference of periodically emitted electron-nuclear wave packets.

References:

[1] J. Wu et al., Phys. Rev. Lett.**111**, 023002 (2013).
[2] X.Gong et al., Phys. Rev. Lett. **112**, 243001 (2014).
[3] X.Gong et al., Phys. Rev. Lett. **114**, 163001 (2015).
[4] P. Lu et al., Phys. Rev. A **95**, 033404 (2017).
[5] W. Zhang et al., Nat. Commun. **10**, 757 (2019).
[6] P. Lu et al., PNAS **115**, 2049 (2018).

17:40-18:00 1 August 2019

Modeling nonequilibrium dynamics of photoinduced ultrafast forward and backward electron transfer: A quantum-classical master equation approach

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Photoinduced electron transfer reactions in biological systems display a variety of dynamics that deviate from the predictions of the Marcus theory. These ultrafast reactions happen in a timescale that overlaps with that of their local environmental relaxations. Based on our analyses, these reaction dynamics are nonequilibrium and heavily depend on the relative timescales of the ET reaction and its corresponding local relaxations [1]. Significantly, the driving force and reorganization energy of such reaction can greatly differ from their values at equilibrium [2]. Because of the nonequilibrium interplay between the forward ET reaction (FET) and its local environment, the dynamics of the backward ET (BET) is deeply correlated with that of FET, as well as its local relaxations. Based on the quantum master equation (QME), a three-state model is proposed to address the FET and BET simultaneously. Using the three-state model, diverse ET systems with reaction timescales ranging from femtoseconds to nanoseconds are analyzed within a consistent picture.

References:

[1] Y. Lu, D. Zhong, J. Phys. Chem. Lett., **10**, 3, 346-351 (2019).

[2] Y. Lu, M. Kundu, and D. Zhong, submitted.

08:10-08:45 2 August 2019

Ultrafast electron dynamics in two-dimensional materials and heterostructures

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Complementing graphene, the transition metal dichalcogenides (TMDCs) in the family MX_2 (M = Mo, W; X = S, Se, Te) have emerged as fascinating examples of atomically thin semiconductors. These 2D materials are characterized by direct optical band gaps, with unusually strong excitonic interactions associated with their reduced dimensionality and reduced dielectric screening. In addition, the materials have two degenerate valleys at the extrema of the valence and conduction bands, with a valley circular dichroism providing direct access to one valley or the other by virtue of the helicity of circularly polarized light. These systems have been shown to host not only strongly bound excitons, but also charged excitons, biexcitons, and even charged biexcitons, correlated 5-body states. In addition to identifying the different elementary excitations in the system, the community has made considerable progress in understanding key aspects of the associated electron dynamics.

In this paper, we will illustrate progress in the field through a discussion of the dynamics of excitons. In addition to their rapid intrinsic radiative decay, excitons in some of the material systems can also be formed in triplet-like states for which radiative decay is forbidden. We will describe the dynamics of these states as monitored using a magnetic brightening technique. In addition, at increased exciton concentration, exciton-exciton interactions become significant, leading to the observation of a rapid exciton-exciton annihilation process, as has been characterized by ultrafast pump-probe measurements. We will further describe how control of the polarization of light can be used both to create exciton in a given valley and even to control its ultrafast valley state using a circularly polarized control pulse through the optical Stark effect.

With the ability to stack atomically thin 2D van-der-Waals layers comes the possibility of creating many new layered materials. We will describe in particular how TMDC monolayers can be combined to form atomically thin heterostructures and how these systems support new interlayer exciton states. The dynamics of the formation of interlayer excitons through charge transfer between the adjacent monolayers occurs on the femtosecond time scale. We will discuss how this process can be monitored both through ultrafast pump-probe spectroscopy and through the THz emission associated with the ultrafast charge transfer. This approach provides a new spectroscopic tool for the direct characterization of charge transfer on the femtosecond time scale over distances as short as a fraction of a nanometer.

08:45-09:15 2 August 2019

Water at electrified graphene interfaces: Structure, dynamics, and vibrational SFG spectroscopy

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Water's properties at electrified interfaces are central in e.g. electrochemistry. We will present the main results of our theoretical study of liquid water/electrified semi-metallic graphene sheet interfaces, employing classical molecular dynamics simulations at different constant potentials. We find that – surprisingly – the interfacial water molecules' spatial structural and dynamical properties vary non- monotonically with the applied potential. We show that this behavior can be directly understood within the extended jump-model of water hydrogen-bond exchanges. We also indicate how simulated vibrational sum-frequency generation spectra of the water OH stretch depend on the contributions from the interfacial and the subsequent water layers. Finally, several key consequences of adding ions – present in almost all electrochemical contexts – are discussed.

References:

[1] D. Laage and J.T. Hynes Science **311**, 832 (2006).

[2] G. Stirnemann, S. R.-V. Castrillon, J. T. Hynes, P. G. Debenedetti, D. Laage PhysChemChemPhys **13**, 19911 (2011).

09:15-09:35 2 August 2019

Many-body elastic scattering of exciton polarons in organic-inorganic hybrid perovskites

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Owing to both electronic and dielectric confinement effects, two-dimensional organic-inorganic hybrid perovskites sustain strongly bound excitons at room temperature. The intrinsic optical lineshape reflects multiple excitons with distinct binding energies, each dressed differently by the hybrid lattice. Given this complexity, a fundamentally far-reaching issue is how Coulomb-mediated many-body interactions --- elastic scattering such as excitation-induced dephasing, inelastic exciton bimolecular scattering, and multi-exciton binding --- depend upon the specific exciton-lattice coupling. We report the intrinsic and density-dependent exciton pure dephasing rates and their dependence on temperature by means of a coherent nonlinear spectroscopy. We find exceptionally strong screening effects on multi-exciton scattering relative to other two-dimensional single-atomic-layer semiconductors. Importantly, the exciton-density dependence of the dephasing rates is markedly different for distinct excitons. These findings establish the consequences of particular lattice dressing on exciton many-body quantum dynamics, which critically define fundamental optical properties that underpin photonics and quantum optoelectronics in relevant exciton density regimes.

09:35-09:55 2 August 2019

Carrier dynamics of lead-free perovskite nanocrystals

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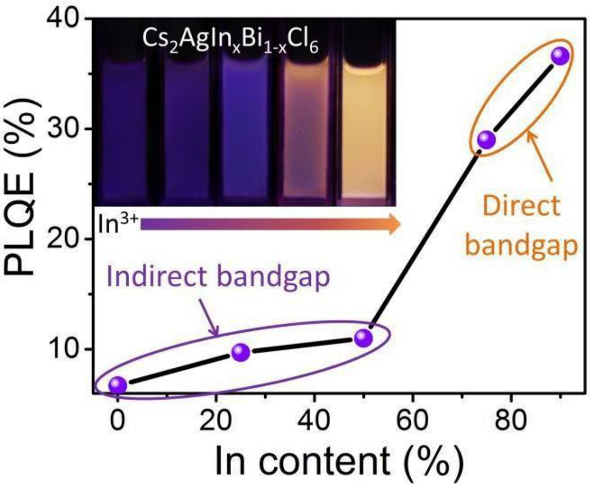
Lead-based perovskite nanocrystals (NCs) have demonstrated outstanding optical properties and cheap synthesis methods conferring them a tremendous potential in the field of optoelectronic devices. However, two critical problems are still unresolved and hindering their commercial applications: one is the fact of being lead-based and the other is the poor stability. Thus, searching for low toxic and highly stable perovskite NCs is on the cutting-edge of the research. We report the synthesis and optical properties of air-stable lead-free perovskite NCs. Femtosecond transient absorption study indicate that charge-carrier trapping is prominent in lead-free perovskite NCs. The charge-carrier trapping processes include surface trapping (50~100 ps) and intrinsic self-trapping respectively. The photoluminescence quantum yield can be improved by over 100 times by better controlling the trap states.



References:

[1] Bin Yang, Junsheng Chen, Feng Hong, Xin Mao, Kaibo Zheng, Songqiu Yang, Yajuan Li, Tõnu Pullerits, Weiqiao Deng, Keli Han*, Lead-Free, Air-Stable All-Inorganic Cesium Bismuth Halide Perovskite Nanocrystals. Angew. Chem. Int. Ed. **56**, 12471-12475 (2017).

[2] Bin Yang, Xin Mao, Feng Hong, Weiwei Meng, Yuxuan Tang, Xusheng Xia, Songqiu Yang, Weiqiao Deng, Keli Han*, Lead-Free Direct Bandgap Double Perovskite Nanocrystals with Bright Dual-Color Emission. J. Am. Chem. Soc. **140**, 17001– 17006 (2018).

10:10-10:45 2 August 2019

Ultrafast spectroscopy and imaging of molecules with classical, quantum, and noisy X-ray pulses

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We demonstrate how stimulated Raman detection of an X-ray probe may be used to monitor the phase and dynamics of the valence electronic state wavepacket created by e.g. photoexcitation, photoionization and Auger processes. Conical intersections (CoIn) dominate the pathways and outcomes of virtually all photophysical and photochemical molecular processes. Short X-ray pulses can directly detect the passage through a CoIn with a high temporal and spectral sensitivity. Applications will also be made for X ray sum frequency generation, and detecting molecular chirality.

Quantum light opens up new avenues for spectroscopy by utilizing parameters of the quantum state of light as novel control knobs and through the variation of photon statistics by coupling to matter. Nonlinear optical signals induced by quantized light fields and entangled photon pairs will be presented. Quantum phase-sensitive diffraction and imaging using entangled photons is presented. A novel quantum diffraction-based far- field imaging technique whereby one photon of an entangled pair is diffracted of a sample and detected in coincidence with its twin is proposed. When a molecule interacts with an external field, the phase information is imprinted in the state of the field in a detectable way. A Schmidt decomposition of the state of the field and the resulting diffraction signal can be used for image enhancement by reweighing the Schmidt modes. Imaging with weak quantum fields is made possible, avoiding damage to delicate biological samples.

Time- and frequency-resolved ultrafast diffraction of noisy X-ray pulses is presented. A frequency-dependent stochastic phase can be used as a frequency marker. Separation of different frequency contributions to the diffraction pattern can be achieved. Fine-tuned correlation functions can control specific charge density contributions. These results are applicable for inherently noisy sources such as SASE free electron lasers, as well as to coherent sources with intentionally added randomness.

References

[1] Lyuzhou Ye, Jérémy R. Rouxel, Daeheum Cho, and Shaul Mukamel. “Imaging electron- density fluctuations by multidimensional X-ray photon-coincidence diffraction”, PNAS, **116**, 395-400 (2019)

[2] Shahaf Asban, Daeheum Cho, and Shaul Mukamel. “Frequency-resolved ultrafast single- molecule diffraction of noisy X-ray pulses ''. JPCL (Submitted, 2019)

[3] Shahaf Asban, Konstantin E. Dorfman and Shaul Mukamel. “Quantum phase-sensitive diffraction and imaging using entangled photons”. PNAS, In press (2019)

10:45-11:15 2 August 2019

Nonlinear optical spectroscopy at the frequency and time limit: Lineshape mattters

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In nonlinear optical spectroscopy, if pulses with extremely different pulse widths are used, it is possible to obtain broadband spectroscopic measurement with both excellent frequency and time resolution through the measurement of accurate spectral lineshapes [1,2]. Here we would like to explore the effects of the temporal and chirping properties in the pulses of the spectroscopic measurement using broadband sum-frequency vibrational spectroscopy (BB-SFG-VS) as an example.

References:

[1] L. Velarde, and H. F. Wang, Phys. Chem. Chem. Phys. **15**, 19970 (2013).

[2] H. F. Wang, L. Velarde, W. Gan, and L. Fu, Annu. Rev. Phys. Chem. **66**, 189-216 (2015).

11:15-11:35 2 August 2019

Achieving surface sensitivity in ultrafast XUV spectroscopy

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The ability to follow electron dynamics at surfaces is necessary to identify the material properties and surface states, which mediate carrier lifetime and determine energy conversion efficiency. Toward this goal we have developed extreme ultraviolet reflection-absorption (XUV-RA) spectroscopy as a surface-specific analog of XUV transient absorption. [1, 2] This technique is used to study ultrafast electron trapping and defect-mediated recombination at the surface of NiO [3] as well as charge transfer and exciton dissociation dynamics in Fe_2O_3/NiO heterostructures [4]. Direct observation of ultrafast electron trapping and recombination in NiO shows that grain boundaries rather than oxygen vacancies are responsible for fast electron-hole pair recombination. This result clarifies design parameters for this water oxidation catalyst by showing that oxygen vacancies have no detrimental effect on carrier lifetime. Rather, carrier lifetime can be dramatically extended by the elimination of near-surface grain boundaries even in the presence of chemically active oxygen vacancies. In Fe_2O_3/NiO heterostructures, transient XUV-RA measurements reveal the formation of Ni^{3+} following selective photoexcitation of the underlying Fe_2O_3 substrate. This indicates that hole transfer occurs to NiO valence band states composed of significant Ni 3d character. We find that hole injection process proceeds via a two-step sequential mechanism where exciton dissociation occurs in Fe_2O_3 in 470 ± 220 fs, followed by subsequent hole injection to NiO in 880 ± 560 fs. These results reveal the chemical nature of the hole acceptor state in NiO hole transport layers and provides a direct observation of exciton dissociation and interfacial hole transfer in this system.

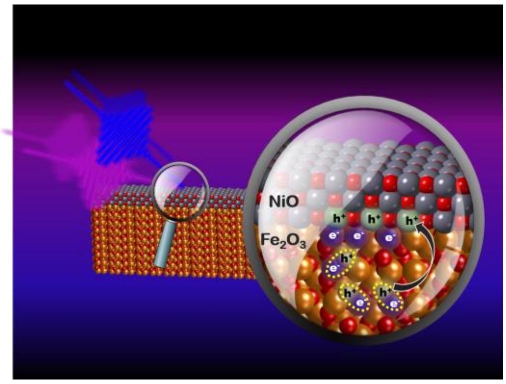

Figure 1. Ultrafast XUV-RA spectroscopy provides direct observation of exciton dissociation and interfacial hole transfer at a Fe_2O_3/NiO heterostructure.

References:

[1] Cirri, A., Husek, J., Biswas, S., Baker, L.R. Achieving Surface Sensitivity in Ultrafast XUV Spectroscopy: M2,3-Edge Reflection–Absorption of Transition Metal Oxides. The Journal of Physical Chemistry C **121**, 15861 (2017).

[2] Biswas, S., Husek, J., Baker, L.R. Elucidating ultrafast electron dynamics at surfaces using extreme ultraviolet (XUV) reflection–absorption spectroscopy. Chemical Communications **54**, 4216 (2018).

[3] Biswas, S., Husek, J., Londo, S., Baker, L.R. Ultrafast Electron Trapping and Defect- Mediated Recombination in NiO Probed by Femtosecond Extreme Ultraviolet Reflection–Absorption Spectroscopy. The Journal of Physical Chemistry Letters **9**, 5047 (2018).

[4] Biswas, S., Husek, J., Londo, S., Fugate, E.A., Baker, L.R. Identifying the acceptor state in NiO hole collection layers: direct observation of exciton dissociation and interfacial hole transfer across a Fe_2O_3/NiO heterojunction. Physical Chemistry Chemical Physics **20**, 24545 (2018).

11:35-11:55 2 August 2019

Long-distance carrier transport beyond the limit of exciton in layered 2D perovskite quantum wells

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Semiconductor quantum wells are a type of material consisting of an extremely thin semiconductor with small bandgap sandwiched by materials with large bandgap. Due to the low dielectric screen and geometric confinement effects, excitons in quantum wells possess a larger banding energy than in bulk counterparts, and their diffusion length is limited by the short lifetime (typically < a few ns) and low diffusivity. Here, we report an observation of surprisingly long distance carrier transport beyond the limit of excitons in $(BA)_2(MA)_{n-1}Pb_nI_{3n+1}$ (BA= $CH_3(CH_2)_3NH_3^+$, MA = $CH_3NH_3^+$) layered 2D perovskite (n = 2~ 4) quantum wells. We observed that in 2D layered perovskites photoinduced excitons undergo a hole trapping process and thus dissociate into free electrons and trapped holes. We confirmed that these trapped holes and free electrons can transport, in different pathways, over a long distance of a few micrometers. This result implies that the transport of photoinduced energy carriers in 2D perovskite quantum wells is beyond the limit of exciton, making 2D perovskites a unique material for applications relying on long-distance energy delivery.

11:55-12:15 2 August 2019

Suppressing triplet loss channel in organic photovoltaic blends with fluorinated acceptors

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The landscape of solar cell technology has recently been changed by implanting non-fullerene acceptors [1]. In particular, most highly efficient devices have been demonstrated by using fluorinated acceptors. Nonetheless, the underlying mechanism remains elusive. Here, we conduct ultrafast spectroscopic study on the charge generation dynamics in a model system with polymer donor PTQ10 and small molecular acceptor of MO-IDIC (Fig. 1a) demonstrated in our early work [2]. We observe the triplet loss channel [3] in the PTQ10/MO-IDIC blend (Fig. 1b) is strongly suppressed in the PTQ10/MO-IDIC-4F blend (Fig. 1c) as confirmed by triplet sensitization measurement (Fig. 1d). Quantum chemical calculation suggests that triplet suppression is enabled by the improved gap between triplet energy of acceptor and triplet charge-transfer states.

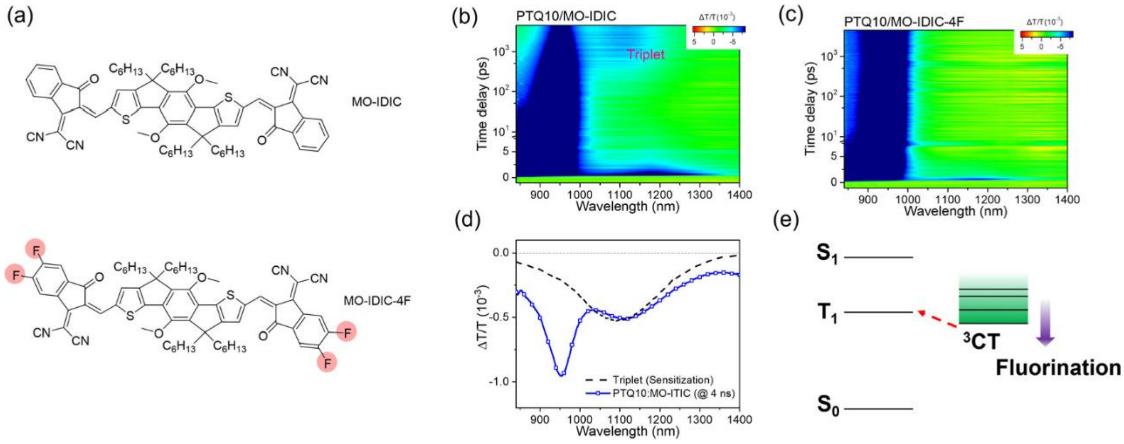

Figure 1. (a) Molecule structures of donor PTQ10 and acceptors of MO-IDIC, MO-IDIC-2F and MO-IDIC-4F. TA data of blends with acceptors of (b) MO-IDIC, and (c) MO-IDIC-4F, respectively. (d) TA spectrum at 4 ns is compared with spectral feature of triplet states of MO-IDIC. (e) Proposed mechanism of triplet loss channel. In the blends with fluorinated acceptors, the triplet channel is suppressed by increased endothermicity.

References:

[1] L. Meng, Y. Zhang, X. Wan et al., Science **361**, 1094 (2018).

[2] X. Li, F. Pan, C. Sun et al., Nature Commun. **10**, 519 (2019).

[3] L. Xue, Y. Yang, J. Xu et al., Adv. Mater. **29**, 1703344 (2017).

Abstracts for Poster Presentations

Poster No.1

Visualizing chemical reactions in space and time using time-resolved X-ray solution scattering

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Chemical reaction is a course of events involving the bond breaking and bond formation, which results in the transformation of reactants to products. To understand the dynamics of chemical reactions, therefore, it is highly desirable to monitor the structures of reacting species during the reaction. In this regard, time-resolved X-ray solution scattering (TRXSS) is a powerful technique to directly capture the reaction dynamics. In TRXSS, temporally short optical laser and X-ray pulse are used as pump and probe, respectively, which allows us to track the reaction with sub-angstrom spatial resolution by virtue of the structural sensitivity of X-ray as well as high temporal resolution. Here, we present two representative works in which bond formation and transient isomer formation have been directly observed using TRXSS. In particular, direct observation of Au- Au bond formation in the photoreaction of a gold trimer complex [1] and formation of transient isomer of bismuth complex during a photoreaction of bismuth triiodide in solution [2] will be described. Detailed kinetics and structures of intermediates involved in both of the reactions will also be presented.

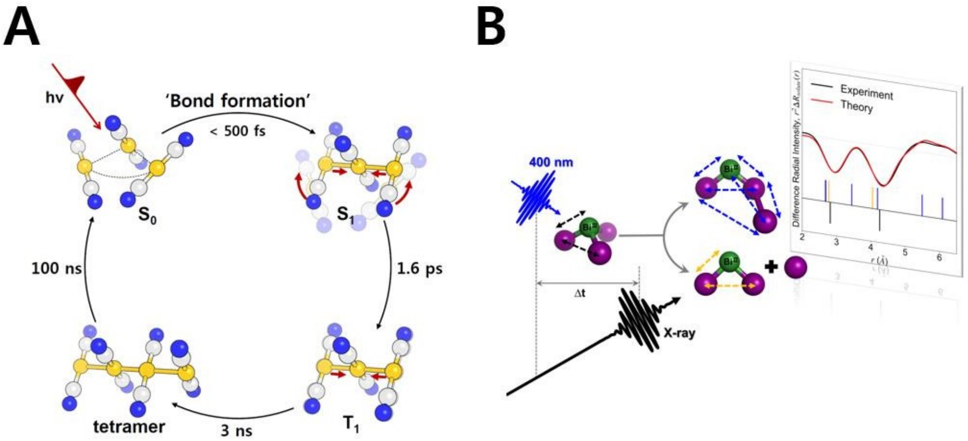

Figure 1. Structural dynamics of gold trimer (A) and bismuth triiodide (B).

References:

[1] K. H. Kim and J. G. Kim et al. Nature **518**, 385 (2015).

[2] E.H. Choi and D. –S. Ahn et al. J. Phys. Chem. Lett **10**, 1279 (2019).

Poster No.2

Femtosecond study of water in a biomolecular hydrogel

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Hyaluronic acid is a ubiquitous biopolymer with an intriguing tunable macroscopic response: it changes from a liquid aqueous solution to an elastic hydrogel in a narrow pH range around pH 2.5 [1]. We study the role of water in the formation of this elastic state using polarization-resolved infrared pump-probe spectroscopy, using pulses with a pulse duration of 200 femtoseconds, an energy of ~10 μJ and a central wavelength of 4 μm.

We observe that in solutions of hyaluronic acid, a fraction of the water molecules shows much slower reorientation dynamics (τ_{or} >10 picoseconds) than in bulk water (τ_{or}=2.5 picoseconds). These slow water molecules are located at the hydrophobic parts of the polymers [2]. Interestingly, the fraction of slow water molecules has a strong pH dependence (Fig. A and B), which strongly correlates with that of the macroscopic viscoelastic behavior (Fig. C) [3]. This observation shows that the formation of the elastic state involves the expulsion of a large fraction of the hydration water from the surfaces of the hyaluronic acid polymers.

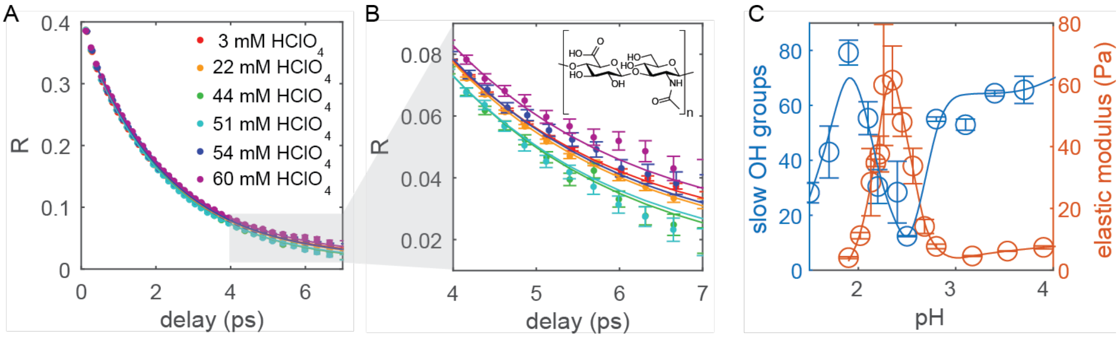

Figure1. (A) Anisotropy decay as function of delay time of hyaluronic acid solutions with different $HClO_4$ concentration. (B) Detail of the anisotropy decay. (C) Number of slowly OH orienting groups per disaccharide unit of hyaluronic acid as a function of pH (blue) and rheology of hyaluronic acid solutions as a function of pH (red).

References:

[1] E.A. Balazs, J. Cui, Bioactive Carbohydrates and Dietary Fibre **2**, 143 (2013).

[2] S.T. van der Post, S. Scheidelaar, and H.J. Bakker, J. Phys. Chem. B. **117**, 15101 (2013).

[3] G. Giubertoni, et al. J. Phys. Chem. B. **123**, 3043 (2019).

Poster No.3

Capturing the electronic quantum coherence in Fenna- Matthews-Olson complex

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Photosynthesis captures sunlight energy to convert carbon dioxide into biomass and it almost provides the whole energy for life on earth. During the initial step of photosynthesis, the energy of solar photon is transformed into electronic excitation energy of the light-harvesting complexes with unity efficiency. Previous studies proposed the long-lived electronic coherence for the efficient energy transport. Here, we revisit the two-dimensional electronic spectroscopy of excitation energy transfer in Fenna-Matthew-Olson (FMO) complex at different temperatures. In the Markovian region, the consistency of electronic decoherence and dephasing demonstrates the validity of direct judging lifetime of electronic coherence from anti-diagonal bandwidth in 2D spectrum at high temperature [1]. Here, we perform spectroscopic measurements at different temperatures to seek lifetimes of electronic quantum coherences. At low temperature, the evidence of electronic coherence is clearly identified. A theoretical modeling is developed to calculate 2D spectra, which captures the lifetime of electronic quantum coherence at different temperatures.

Reference:

[1] H. -G. Duan, et al. Proc. Natl. Acad. Sci. (USA) **114**, 8493 (2017).

Poster No.4

Ultrafast relaxation dynamics and optical nonlinear properties of porphyrin-based surface-supported metal- organic frameworks

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Porphyrin-based surface-supported metal-organic frameworks (SURMOFs) have potential applications in photonics and optoelectronics. [1-3] In this work, we fabricate monolithic, oriented, crystalline and highly porous porphyrin MOF coatings (Co-SURMOF) on quartz substrate by step-by-step liquid-phase epitaxy, and then investigate ultrafast carrier dynamics and optical nonlinearity by using femtosecond time-resolved transient absorption spectroscopy and Z-scan technique. Transient transmission spectra at the pump of 400 nm show that the photoexcited carriers are relaxed through three channels on the time scales of 200fs, 5ps, and 45ps with a component ratio of 33%, 16%, and 51%, respectively. The first two channels are ascribed to the thermalization of hot carriers through the exciton-exciton scattering and non-radiative exciton recombination. The third channel is ascribed to the fast energy hopping to adjacent porphyrin molecules according to Förster energy transfer model. [4-6] Z-scan measurements of Co-SURMOF, on the other hand, exhibit a significant saturated absorption response under the 400nm excitation. The high energy hopping ratio and super high third- order nonlinear absorption coefficient indicate that Co-SURMOF is a promising optical and optoelectronic material. Our results thus provided important photophysical support for the future applications.

References:

[1] V. Stavila, A. Talin, and M. Allendorf, Chemical Society Reviews **43**, 5994 (2014).

[2] J. Liu, C. Wöll, Chemical Society Reviews **46**, 5730 (2017).

[3] W. Gao, M. Chrzanowski, and S. Ma, Chemical Society Reviews **43**, 5841 (2014).

[4] C. Lee, O. Farha, et al, Journal of the American Chemical Society **133**, 15858 (2011).

[5] H. Son, S. Jin, et al, Journal of the American Chemical Society **135**, 862 (2013).

[6] Q. Zhang, C. Zhang, et al, Journal of the American Chemical Society **138**, 5308 (2016).

Poster No.5

The role of meta- and para-phenylene bridges in charge separation of perylene diimide dimers

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Generally, the possibility and rate of charge separation (CS) in Donor-Bridge-Acceptor molecules mainly depend on two factors: electronic coupling and solvent effects. [1] Bridge unit definitely plays a crucial role in electronic coupling. To study the bridge dependent charge separation reaction, the isomeric N-Annulated perylene diimide dimers (**p-BDNP** and **m-BDNP**) with different bridge structure were employed as the research model. By means of transient absorption combined with quantum calculation, the rate of charge separation of **m-BDNP** is more than an order of magnitude slower than through the **p-BDNP**. Because of the destructive interference of the wave function in meta-substituted bridge, weak coupling can be appreciable in **m-BDNP**, and thus leading to the decreased charge separation rate. [2] In addition, the charge separation dynamic of the two isomeric dimers in solvents with different polarity were also tracked by transient absorption. The charge separation is disfavored in weak polar solvent, while clear ionic signal can be observed in polar solvents due to solvent reaction field. Furthermore, the rate of charge separation increases as the solvent polarity increasing. The role of solvent effects in free energy of activation of charge separation has also been identified with quantum calculation.

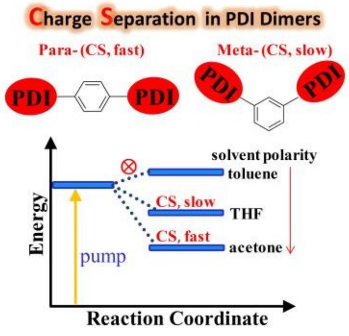

Figure 1. Kinetic scheme for photoinduced charge separation in PDI dimers.

References:

[1] Michael W. Holman, Carlos Silva, J. Phys. Chem. A, **109**, 8548 (2005).

[2] Annie Butler Ricks, Michael R. Wasielewski, J. Am. Chem. Soc., **132**, 15427 (2010).

Poster No.6

Ultrafast carrier dynamics in two-dimensional Bi_2O_2Se with variable layer numbers

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Two-dimensional (2D) materials with new functions are of interest in the field of optoelectronics. As a new member of 2D materials, Bi_2O_2Se has attracted much attention due to the high electron mobility [1-3]. Here, we report on the photo-induced carriers dynamics in Bi_2O_2Se with variable layer numbers by femtosecond time-resolved absorption spectroscopy. We find that the lifetime of excited carriers changes slightly when the pump fluence increases but decreases rapidly as the layer number decreases. The lifetime varies from hundreds of picosecond in the bulk to tens of picosecond in few-layer samples. A tentative model implies that the effect of the surface recombination may play an important role in the carrier decay.

References:

[1] J. Wu, H. Yuan, M. Meng, et al, Nature Nanotechnology **12**,530(2017).

[2] J. Wu, Y. Liu, Z. Tan, et al, Advanced Materials **29**: 1704060(2017).

[3] X. Tian, H. Luo, R. Wei, et al, Advanced Materials **30**:1801021(2018).

[4] H. Wang, C. Zhang, F. Rana, Nano Letters **15**: 8204-8210(2015).

[5] M. Seo, H. Yamaguchi, A. D. Mohite, et al, Scientific Reports **6**: 21601(2016).

Poster No.7

Ligands effects on structural dynamics in Fe(II) spin crossover revealed by ultrafast electron diffraction

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Photoinduced spin crossover (SCO) compounds have huge potential for applications arising from their spin transition and electronic dynamics [1]. Here, single-crystal $[Fe^{II}(bpy)_3](PF_6)_2$ — the prototypical system for SCO dynamics — was studied for the first time using ultrafast electron diffraction (UED) in the single-photon excitation regime. Due to the unique capability of UED, a complete molecular movie of photoinduced SCO — particularly with regard to the ligand motions — were resolved by using a refinement model based on principal component analysis. The structural dynamics of two SCO compounds with different ligands were explained as the result of the metal ligand bond distribution during spin transition and vibrational cooling [2]. Our findings provide key information to understand the role of ligands in the structural dynamics and the vibrational energy dissipation process of photoexcited SCO.

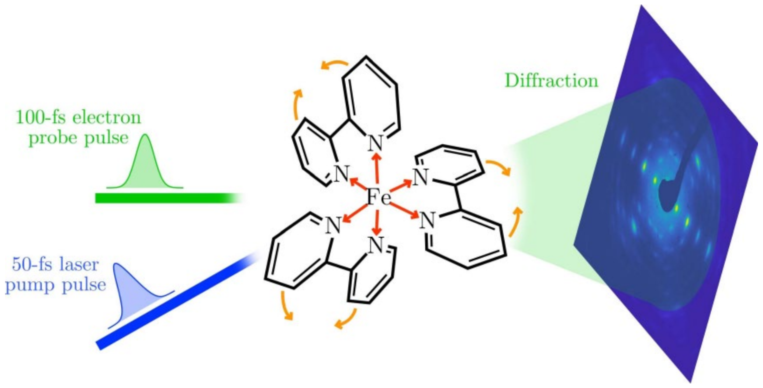

Figure 1: Schematic of the experiment. $[Fe^{II}(bpy)_3](PF_6)_2$ in single crystal was investigated by ultrafast electron diffraction.

References:

[1] McCusker, J. K. Science **363**, 484-488 (2019).

[2] Jiang, Y. et al. Angew. Chemie Int. Ed. **56**, 7130-7134 (2017).

Poster No.8

Spin and excited-state charge dynamics in myoglobin

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Nitric oxide (NO) bound to Myoglobins (Mb) exhibits several remarkable physiological functions that are still being explored, e.g. as a neurotransmitter, in signal transduction, etc. The dissociation from and rebinding of ligands to the Fe in the heme (respiration) can be mimicked by photo-dissociating them with a pulse of light and monitoring the recombination with a probe pulse. While several studies of these processes have been carried out using visible, infrared and Raman probes, none is capable of element-specificity and structural-sensitivity at the Fe site. In recombining to the Fe atom, the NO binds in a domed geometry of the heme, as suggested by previous time-resolved resonance Raman studies. [2]. We will present results of our study on MbNO using both X-ray absorption and emission spectroscopy (XAS and XES), which we complemented by X-ray diffuse scattering (XDS), carried out at the EXFEL, SACLA and SwissFEL.

The current understanding of respiration in heme proteins is, dissociation and doming occur in a single step, followed by the so called “protein quake” (structural changes in the protein). This serves as the basic mechanism for respiration in all mammals. Contrary to this, our study reveals a 2-step process for the dissociation and doming. XAS and XES show that upon prompt dissociation (< 70 fs), the NO ligand is dissociated, after which doming occurs going through an ^{3}T state into the final quintet state, to which NO recombines in a multiexponential fashion over time scales of a few tens of ps to ~200 ps, which is in agreement with our previous study [1]. Both the XES Kα and the Kβ show a clear signature of the triplet and quintet state and combination with XAS enables correlation to the structural changes around the Fe-atom. XDS instead is sensitive to global structural changes of the protein and the combination of these three techniques can potentially reveal a unique picture of electronic, local and global structural changes of Mb.

References:

[1] Silatani, M. et al. PNAS **112**, 12922–12927 (2015).

[2] Kruglik, S. G. et al. PNAS **107**, 13678–13683 (2010).

Poster No.9

Unravelling ultrafast dynamics in photoexcited azaindoles

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Since ultrashort lifetimes were observed in the past decade for all nucleobases implicated in the replication process, the understanding of DNA excited state dynamics has become a hot topic. Nowadays, it is widely accepted that ultrafast internal conversion (IC) implies an enhanced photostability, because a short excited state lifetime prevents molecules from photochemical reactions leading to the eventual formation of harmful photoproducts. [1] The ultrafast IC processes are ultimately linked to the molecular structure. From this perspective, subtle structural changes modify the potential energy surfaces, and consequently, alter the accessibility to the conical intersections that control the different relaxation pathways. Therefore, the question arises whether there is a common pattern in the deactivation dynamics of nucleobases and other life related molecules. The present work focuses on the relaxation dynamics of azaindoles in the isolated and condensed phase. The dynamics of these species, which are a good starting point for a bottom-up approach to more complex systems, has been tracked by femtosecond time-resolved ionization spectroscopy (TR-MPI), transient absorption spectroscopy and fluorescence up-conversion techniques. The electronic excitation region explored (304-239 nm) covers three electronic excited states: the two ππ*, L_a and L_b states, and the dark nπ* state. [2] The collected set of data provides new insights on the photophysical and photochemical events following excitation in the aforementioned energy interval, allowing us to balance the relative importance of the involved relaxation channels.

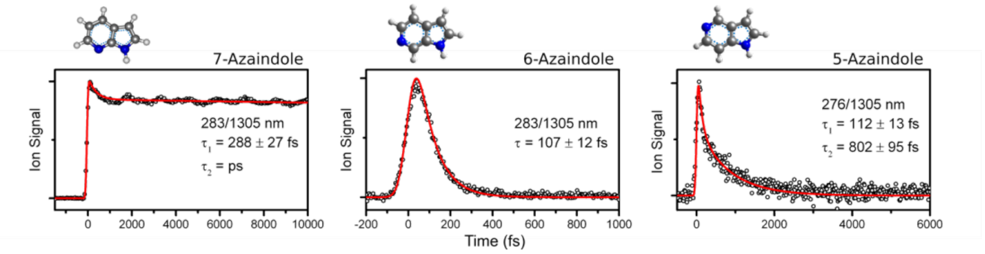

Figure 1. TR-MPI transients of 7AI, 6AI and 5AI.

References:

[1] K. Kleinermanns, D. Nachtigallová, and S. de Vries, Int. Rev Phys. Chem. **32**, 308 (2013).

[2] L. Serrano-Andrés, M. Merchán, A. C. Borin, J. Stålring, and S. Paulo, Int. J. Quantum Chem. **84**, 181 (2001).

Poster No.10

The flexibility of enzyme probed with 2D-IR

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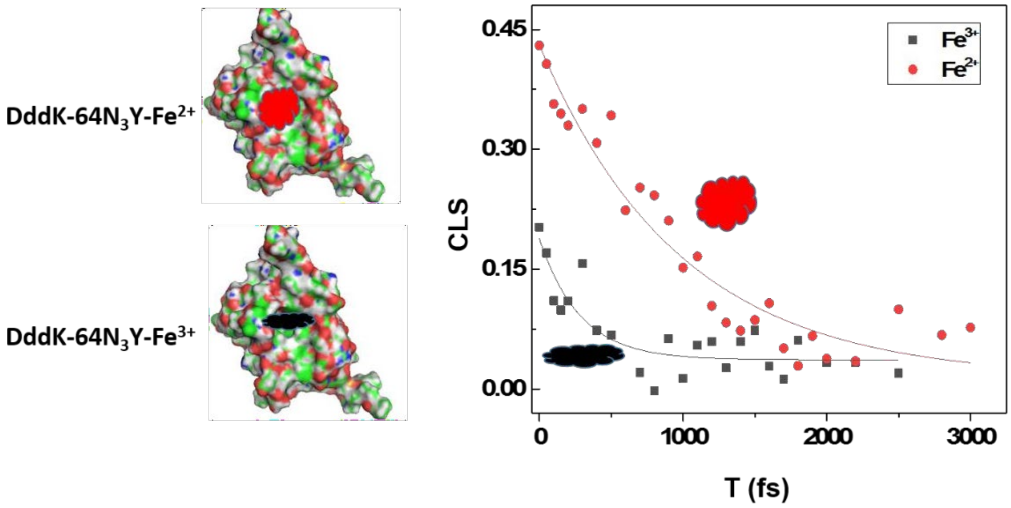
The flexibility of Enzyme at active site is vital for the catalytic reaction. But it is difficult to quantify. By exploring the characteristic of fast dynamic measurement of 2D-IR, we successfully quantify the active site flexibility. Firstly, tyrosine analog m-azido-tyrosine (N_3Y) was genetically incorporated to the active site 64 of metalloenzyme DddK as the IR probe. Then, our 2D-IR results reveal that the more flexible the active site is, the slower decay curve of FFCF will have.



References:

[1] Tsou, C. L., Conformational flexibility of enzyme active site. Science **1993**, 262 (5132), 380-381.

[2] Schnicker, N. J.; De Silva, S. M.; Todd, J. D.; Dey, M., Structural and Biochemical Insights into Dimethylsulfoniopropionate Cleavage by Cofactor-Bound DddK from the Prolific Marine Bacterium Pelagibacter. Biochemistry-Us **2017**, 56 (23), 2873-2885.

Poster No.11

Hydrogen bond dynamics in flavin mononucleotide unveiled by femtosecond stimulated Raman spectroscopy

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The yellow-colored pigment flavin mononucleotide (FMN) is a cofactor found in photoreceptor proteins containing the light–oxygen–voltage (LOV) domain [1]. Absorption of blue-light by the FMN moiety bound to the LOV domain induces a conformational change that ultimately triggers a biological output, e.g., gene activation. Therefore LOV proteins represent a novel platform for optogenetic applications [2]. We have deciphered the ultrafast changes in the hydrogen bond network of FMN upon blue- light illumination by femtosecond stimulated Raman spectroscopy (FSRS). To this end, the solvation structure of the flavin group free in solution was studied systematically in different DMSO/water mixtures.

A state-of-the-art FSRS setup has been developed at the European Extreme Light Infrastructure Beamlines (ELI Beamlines). We employed the watermarking technique [3, 4] by using a double-pass compressor grating and a customized chopper, coupled with home-made software to deconvolve the spectra. Global analysis of UV–visible transient absorption and FSRS spectra of FMN allowed us to extract the contribution from the singlet (S_1) and triplet (T_1) states in the different solvents. In order to assign the bands and shifts observed in the experimental Raman spectra of FMN in the excited states, we have carried out extensive quantum chemistry simulations.

Our results pave the way for the investigation of the hydrogen bond dynamics of FMN embedded in LOV proteins by FSRS in order to unveil the structural dynamics of flavin-binding photosensors.

References:

[1] S. Crosson, S. Rajagopal, K. Moffat. Biochemistry **42**, 2 (2003).

[2] A. Losi, K.H. Gardner, A. Möglich. Chem. Rev. **118** (21), 10659 (2018).

[3] M. Kloz, R. van Grondelle, J.T.M. Kennis. Phys. Chem. Chem. Phys. **13**, 18123 (2011).

[4] M. Kloz, J. Weißenborn, T. Polívka, H.A. Frank, J.T.M. Kennis. Phys. Chem. Chem. Phys. **18**, 14619 (2016).

Poster No.12

Ultrafast photoreduction of solvated iron compounds probed by VUV photoelectron spectroscopy

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Transition metal complexes can exist in high-spin (HS) or low-spin (LS) configuration [1], whose equilibrium depends on oxidation state, nature of the ligands and coordination number. Light interaction triggers ultrafast electronic and structural dynamics such as bond cleavage, symmetry rearrangement, spin-crossover (SCO), and charge transfer [2]. These compounds can hardly be isolated, hence photochemistry of solutions is studied, where solvent plays a relevant role [3]. A good example is ferric trisoxalate $[Fe^{III}(ox)_3]^{3-}$ (ferrioxalate), that upon UV excitation experiences ligand dissociation because of forerunner processes yet to be clarified. The final photoproduct lacks of a ligand and the iron is reduced [4]. This implies a ligand-to-metal charge transfer (LMCT) process, a ligand-field strength reduction and a possible SCO. Recent X-ray studies at the iron K-edge concluded fragmentation within 3 ps [5]; other IR measurements demonstrated ligand bond breakage within the first ps [6]. Though, it is not yet clarified whether the metal reduction is a consequence of the cleavage or not, and how structural and electronic dynamics interplay at early times. We present a time-resolved study of photoexcited ferrioxalate aqueous solution by means of photoelectron spectroscopy (PES) [7-8], which is the only technique that provides the whole accessible electronic structure of the sample [9]. This allows us to simultaneously track electronic and structural dynamics of photoexcited solvated molecules. Upon 266 nm-wavelength excitation, we observe the Fe 3d photoelectron signal to shift towards the ferrous spectral position (see figure), i.e. metal photoreduction is prompt (<50 fs), which ascertains the LMCT as primary and driving mechanism. Furthermore, the transient photoelectron iron signal does not undergo peak narrowing whithin our probe range (5 ps), otherwise expected in case of HS to LS SCO [10]. These results, together with previous infrared [6] and X-ray studies [5], yield a complete picture of the photoinduced dynamics.

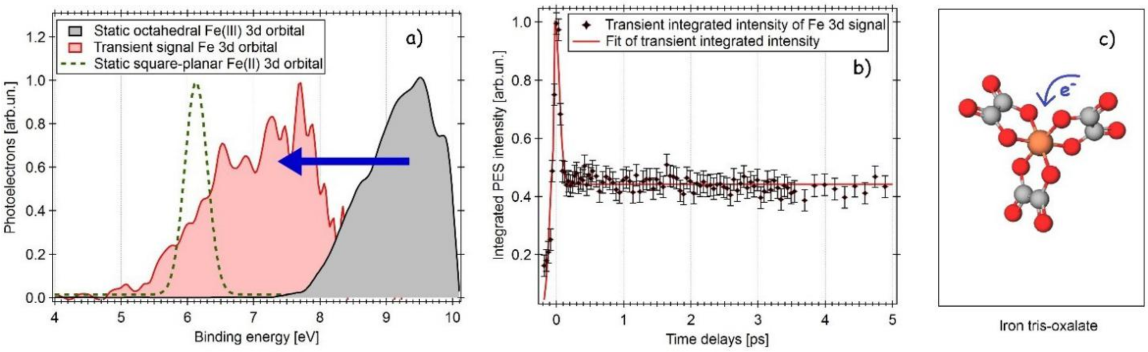

Figure 1. a) Static PES signal of Fe(III)oxalate iron 3d orbitals (black), and transient Fe(II) upon 266 nm excitation (red) shifting towards the final product Fe(II) spectral signature (dashed); b) the integrated PES Fe(II) transient intensity rises and remains constant; c) sketch of ferrioxalate with LMCT mechanism.

References:

[1] J. Beattie, Advances in Inorganic Chemistry **32**, 1-53 (1988)

[2] J. Ojeda et al., Phys. Chem. Chem. Phys. **19**, 17052-17062 (2017)

[3] T. Penfold et al., J. Phys. Chem. A, **117**, 4591–4601 (2013)

[4] Hatchard and Parker, Proc. Royal Society of London Series A, **235**, 518-536 (1956)

[5] Y. Ogi et al., Structural Dynamics **2**, 34901 (2015)

[6]S. Straub et al., Phys. Chem. Chem. Phys. **20**, 21390 (2018)

[7] J. Ojeda et al., Structural Dynamics **3**, 023602 (2016)1 (2006)

[8] C. Arrell et al., Chimia **71**, 268-272 (2017)

[9] B. Winter and M. Faubel, Chem. Rev. **106**, 1176−121

[10] R. Seidel et al., J. Phys. Chem. B **115**, 11671-11677 (2011)

Poster No.13

A single-shot ultrafast electron diffraction approach based on compressive sensing

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Ultrafast electron diffraction (UED) has been proposed as a useful technique to resolve real-time structural dynamics of matter [1, 2]. However, it still suffers from problems of repetitive acquisition and timing jitter. We propose a new technique called compressed ultrafast electron diffraction imaging (CUEDI). As shown in Figure 1, a time-resolved electron diffraction data cube is encoded by an electron encoder, deflected by a time-varying electric field and recorded by a detector, and finally the two-dimensional superimposed data are decoded by a CS algorithm to recover the time-evolving electron diffraction pattern. To validate the feasibility of our proposed scheme, a detailed electron trajectory is simulated using a charged particle tracing software, and a time-resolved electron diffraction pattern for the polycrystalline gold melting process is reconstructed. The results indicate that CUEDI is capable of resolving the structural dynamics in reciprocal space with one shot, and holds great potential to replace the traditional pump-probe scheme [3].

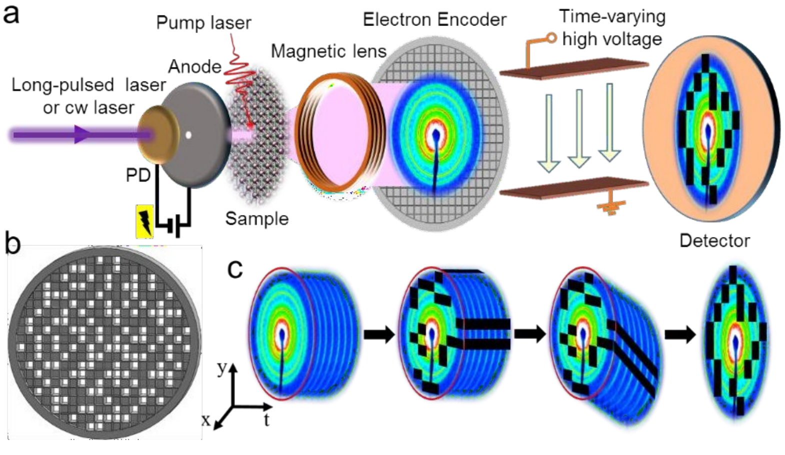

Figure 1. The schematic diagram of the CUEDI (a); A detailed illustration of the electron encoder (b); The data acquisition for the CUEDI (c).

References:

[1] A. H. Zewail, Annu. Rev. Phys. Chem. **57**, 65 (2006).

[2] R. J. D. Miller, Science **343**, 1180 (2014).

[3] D. Qi et al., Phys. Rev. Appl. **10**, 054061 (2018).

Poster No.14

Structural phase transition and dynamics in $MoTe_2$ and WTe_2 studied by femtosecond electron diffraction

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Recently, the semimetallic transition metal tellurides, WTe_2 and $MoTe_2$, have sparked broad research interest due to unique electronic structures and crystal configurations, such as Weyl fermions, ferroelectricity and in-plane anisotropy. The photoexcitation can potentially trigger structural transition and the corresponding electronic properties therein in an ultrafast fashion [1, 2], which offers further prospects in future applications as optically controlled switching devices. Here we report the research on femtosecond laser induced structural phase transition and transient structural dynamics in WTe_2 and $MoTe_2$ by ultrafast electron diffraction [3]. We observe the visible and IR femtosecond laser excitations invoke coherent shear mode in WTe_2 and $MoTe_2$, indicating a potential structural transition from topological semimetal Td phase to trivial semimetal 1T' phase. Another several coherent optical and acoustic phonon modes are also found which we will discuss to map a complete atomic motions during the structural transition. In both 1T' and Td phase, a significant in-plane anisotropic electron-phonon interaction is presented. We compare the results here with optical measurements reported before.

References:

[1] E. J. Sie et al., Nature **565**, 61 (2019).

[2] M. Y. Zhang et al., arXiv:1806.09075 (2018).

[3] L. Waldecker et al., J. Appl. Phys. **117**, 044903 (2015).

Poster No.15

Cycloreversion reactivity of photochromic diarylethene derivatives: Negative correlation between the S_1 state and higher state excited by stepwise two-photon absorption

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Photochromic diarylethene derivatives undergo the efficient ring-opening (cycloreversion) reaction in higher excited states (S_n states) attained by stepwise/simultaneous two-photon absorption of visible light [1,2]. In the present study, to comprehensively investigate the cycloreversion reactivity in the S_1 and higher excited states, we have applied stepwise two-photon excitation to various diarylethene derivatives with femtosecond multicolor laser pulses. Figure 1a shows the two-photon cycloreversion reaction amount of **PT** plotted as a function of the time interval between two excitation pulses. Upon the sequential excitation, the permanent bleaching signal due to the cycloreversion reaction, which corresponds to the reaction amount, increases compared with the one-photon reaction level. This result indicates that **PT** undergoes the cycloreversion reaction with higher quantum efficiency in the Sn state than in the S_1 state. On the other hand, **BT**, whose the cycloreversion reaction yield is ca. 30%, shows an opposite signal, as shown in Figure 1b. This result indicates that the higher excited state of **BT** attained by stepwise two-photon excitation leads to the suppression of the cycloreversion reaction. The results in the present study indicate that the reactivity in the S_n state is negatively correlated with that in the S_1 state.

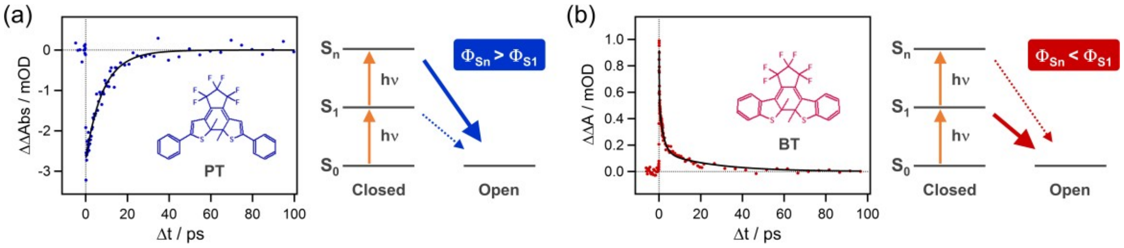

Figure 1. Two-photon cycloreversion reaction amounts of (a) ***PT*** *and (b)* ***BT*** *excited with two sequential femtosecond pulses. The horizontal axis indicates the time interval between the two excitation pulses. The corresponding reaction pathways and yields are also shown.*

References:

[1] H. Sotome, T. Nagasaka, K. Une, S. Morikawa, T. Katayama, S. Kobatake, M. Irie, H. Miyasaka, J. Am. Chem. Soc. **139**, 17159 (2017).

[2] H. Sotome, T. Nagasaka, K. Une, C. Okui, Y. Ishibashi, K. Kamada, S. Kobatake, M. Irie, H. Miyasaka, J. Phys. Chem. Lett. **8**, 3272 (2017).

Poster No.16

Polaronic species in polymers for solar cells distinguished by ultrafast dynamics of their infrared activated vibrational fingerprint

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Several distinct photoexcited species play a role in the process of charge generation in polymer photovoltaics. A clear understanding of their dynamics is fundamental for the design of efficient organic solar cells. Besides excitons, polaron pairs have been identified as another type of bound charges alongside free polarons. To study their diffusion and recombination characteristics, contributions from distinct species need to be assigned unambiguously. So far, this distinction has mostly been based on the excitation density dependence of their respective dynamics or modeling of the underlying spectra. We show that charge- induced infrared activated vibrations - that are specific to the nature of the respective charged species - can be employed to directly identify them in femtosecond visible-pump/mid-infrared-probe spectroscopy. Generation and recombination of polaronic species on ultrafast time-scales are investigated in bulk heterojunctions formed of polythiophene derivatives with TiO_2 nanoparticles as an inorganic acceptor, as well as in films of the pure polymer. Vibrational modes sensitive to the presence of a charge promise access to polaron dynamics on a femtosecond timescale in other emerging materials, such as perovskites, as well. Here, coupling between electronic excitation and molecular vibrations is believed to play a fundamental role.

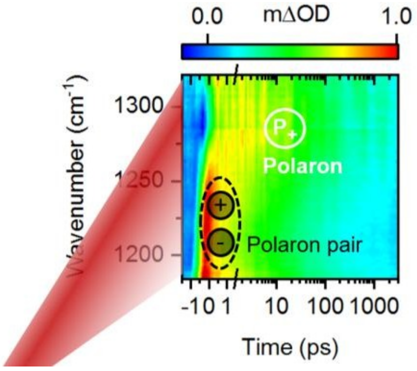

Figure 1. Illustration of direct distinction between polarons and polaron pairs by the dynamics of their specific infrared activated vibrational modes on a femtosecond to picosecond time scale.

Poster No.17

RF compressed femtosecond electron microscope

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Ultrafast electron diffraction and microscopy has been proved to be a powerful tool for studying ultrafast phenomena in materials. However, significant challenges exist in reaching adequate sensitivities at the femtosecond temporal scale due to the space charge effect at high-charge densities. Here we report the development of new ultrafast electron microscope that utilizes RF cavities as the longitudinal lens to reach time and energy compression [1]. With this key concept, we demonstrate the first prototype RF compressed ultrafast electron microscope based on Hitachi H800 TEM and combined with 25keV photo DC gun. We show that by tuning the longitudinal lens, 100 fs time resolution, 15 nm imaging resolution, and sub-eV energy resolution are reached using electron pulse within 1kHz, 25keV, and 10^4 e-/pulse. Furthermore, the imaging resolution can be easily improved to sub-nm by upgrade the beam energy of electron gun. The current level of performance clearly suggests that the collective space charge limits presented in the earlier ultrafast electron microscope experiments can be overcome, and it opens the prospect of performing ultrafast electron energy loss spectroscopy with new RF optical components in a commercial TEM system [2].

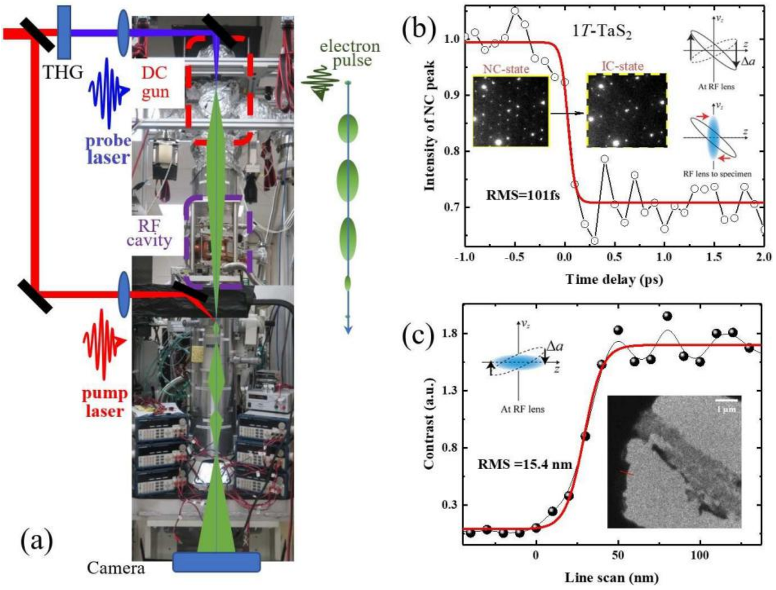

Figure 1. RF compressed UEM. (a) Setup; (b) Time resolution; (c) Spatial resolution.

References:

[1] J. Williams, F. Zhou, T. Sun, et al., Struct. Dyn. **4**, 044035 (2017).

[2] F. Zhou, J. Williams, and C.-Y. Ruan, Chem. Phys. Lett. **683**, 488 (2017).

Poster No.18

Ultrafast liquid dynamics revealed by solute-pump/solvent-probe spectroscopy

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The most common spectroscopic ways of studying solvation dynamics in liquids such as time-dependent fluorescence spectroscopy, share a feature of probing the solute directly after a resonant excitation. The solvent structure/dynamics has to be inferred from the concomitant evolution of the solute-solvent interaction energy. By combining a resonant solute-pump with nonresident 4-wave-mixing light scattering, we can track solvation dynamics by looking at the structure- sensitive many-body polarizability, as in recently developed solute-pump/solvent- probe spectroscopy. With this novel spectroscopy, it is now possible to directly measure how that solvent dynamics can be modified by electronically excitation of the solute. In this work, I will show a molecular theory and hybrid computational approaches involving the instantaneous normal mode theory and equilibrium and nonequilibrium molecular dynamics simulations for computing the novel spectra. The sensitivity to the changes in local solvent structural dynamics makes the solute-pump/solvent-probe spectroscopy a great alternative for solvation dynamics.

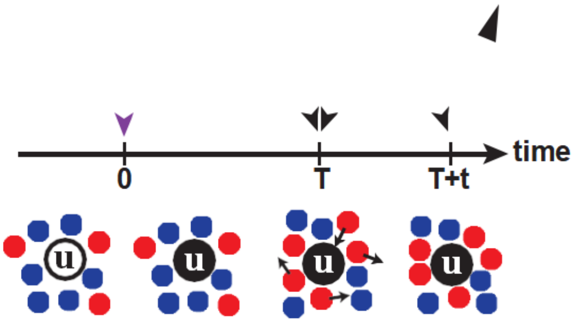

Figure 1. The sequence of events in solute-pump/solvent-probe spectroscopy. Initial resonant solute (u) electronic excitation at time 0 is followed by a 4-wave- mixing non-resonant light scattering at time T and T+t.

References:

[1] X. Sun, B. M. Ladanyi, and R. M. Stratt, J. Phys. Chem. B, **119**, 9129-9139 (2015).

[2] X. Sun and R. M. Stratt, J. Chem. Phys. **139**, 044506 (2013).

[3] X. Sun and R. M. Stratt, Phys. Chem. Chem. Phys. **14**, 6320-6331 (2012).

Poster No.19

pH dependence, kinetics and light-harvesting regulation of nonphotochemical quenching in Chlamydomonas

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Sunlight drives photosynthesis but can also cause photodamage. To protect themselves, photosynthetic organisms dissipate the excess absorbed energy as heat, in a process known as nonphotochemical quenching (NPQ) [1]. In green algae, diatoms, and mosses, NPQ depends on the light-harvesting complex stress-related (LHCSR) proteins [2]. Here we investigated NPQ in Chlamydomonas reinhardtii using an approach that maintains the cells in a stable quenched state. We show that in the presence of LHCSR3, all of the photosystem (PS) II complexes are quenched and the LHCs are the site of quenching, which occurs at a rate of ~150 ps^{-1} and is not induced by LHCII aggregation. The effective light-harvesting capacity of PSII decreases upon NPQ, and the NPQ rate is independent of the redox state of the reaction center. Finally, we could measure the pH dependence of NPQ, showing that the luminal pH is always above 5.5 in vivo and highlighting the role of LHCSR3 as an ultrasensitive pH sensor [3].

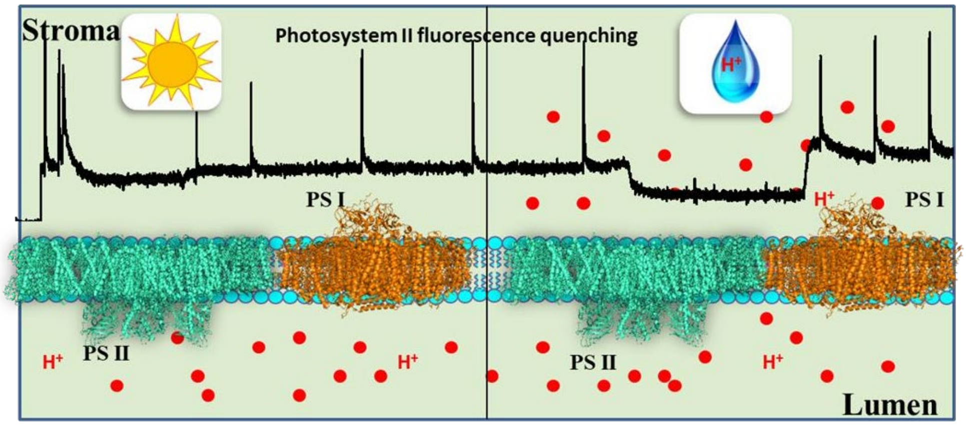

Figure 1. A TOC graphic presentation

References:

[1] G. Peers, T. B. Truong, E. Ostendorf, A. Busch, D. Elrad, A. R. Grossman, M. Hippler, and K. K. Niyogi. Nature **462**, 518 (2009).

[2] J. D. Rochaix, Annu Rev Plant Biol **65**, 287 (2014).

[3] L. Tian, W. Nawrocki, X. Liu, I. Polukhina, I. H. M. van Stokkum, and R. Croce. Proc Natl Acad Sci U S A, **116**, 8320 (2019).

Poster No.20

Ultrafast coherent phonon dynamics in Bi_3Se_2Te

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Bi_3Se_2Te has been recently studied suggesting the existence of the topological surface state [1]. In the present work, we have performed transient absorption spectroscopy of the Bi_3Se_2Te film to study its ultrafast dynamics after photo-excitation. The measurement result of transient absorption (Fig. 1) shows periodical modulation reflecting the coherent optical photon mode at the period of ~500 fs.

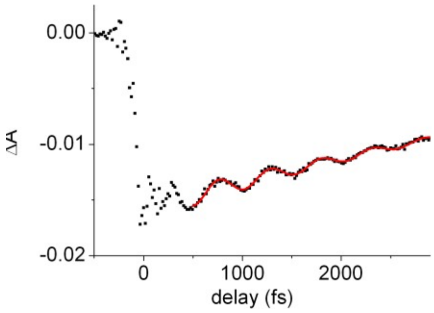

Figure 1. Transient absorption trace of Bi_3Se_2Te

The relaxation lifetime of ~2 ps was assigned to the lifetime of this phonon mode. The cosine-like phase of the signal modulation and the pump power dependence of the signal indicates that the observed dynamics of the phonon mode can be assigned to that of the electronic excited state. [2]

References:

[1] P.H. Le, S.-P. Chiu, S.-R. Jian, C.W. Luo, J.-Y. Lin, J.-J. Lin, K.H. Wu, M. Gospodinov, J. Alloys Compd. **679**, 350–357 (2016).

[2] Y.-K. Ko, Y.-H. Huang, P. H. Le., C.-W. Luo, M. M. Gospodinov, T. Kobayashi, A. Yabushita, Chem. Phys. **512**, 62-67 (2018)

Poster No.21

Femtosecond electronic relaxation and real-time vibrational dynamics in 2’-hydroxychalcone

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Femtosecond ultrafast electronic relaxation and vibrational dynamics in 2’-hydroxychalcone after deep ultraviolet (DUV) excitation were observed by two types of pump-probe spectroscopy experiments, i.e., DUV-pump pulse and visible-broadband-probe pulse (DUV/Vis) experiments and DUV-pump and DUV-probe (DUV/DUV) pulses experiments. Time-dependent density functional theory (TDDFT) calculations were performed to elucidate relaxation dynamics from the third singlet electronic excited state S_3. The DUV/Vis experiments and TDDFT calculations have disclosed the ultrafast dynamics of internal conversion from the initial S_3 state ($\tau_1 \approx 35$ fs) towards the S_1 state via a rapid process through the S_3/S_2 conical intersection and proton transfer [OH: $\tau_2(\mathrm{H}) \approx 93$ and OD: $\tau_2(\mathrm{D}) \approx 164$ fs] before deactivation through the S_1/S_0 conical intersection ($\tau_3 \approx 690$ fs). Thanks to the ultrashort pump and probe pulses, real-time observation of vibrational modes coupled to the electronic excitation was realized providing both amplitudes and phases. Spectrogram analyses were performed based on the real-time spectra obtained by the DUV/Vis experiments, in which instantaneous vibrational frequencies reflecting molecular structural change after the impulsive excitation were visualized. The vibrational frequency of central C=C bond stretch decreases from ~1600 cm^{-1} to ~1560 cm^{-1} in about 200–500 fs and recovers by ~550 fs. Normal mode analyses along the decay path support the observed variation of the C=C stretching frequency. The temporal weakening of the central C=C bond is connected with the angle of the two aromatic rings which flip back to the initial conformation.

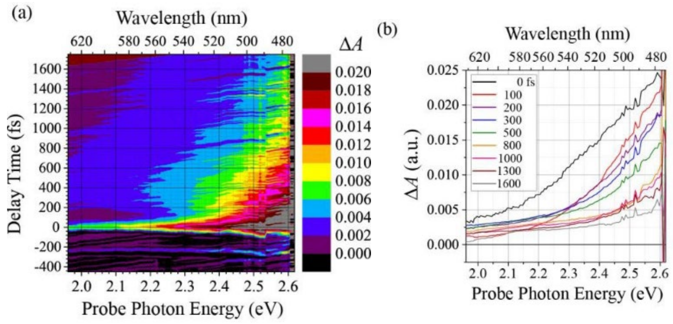

Figure 1. (a) Two-dimensional spectrum and (b) spectral slices of $\Delta A(\omega,t_d)$ for trans-2’-hydroxychalcone (2’HC) in methanol.

References:

[1] Y. Yamakita, N. Yokoyama, B. Xue, N. Shiokawa, Y. Harabuchi, S. Maeda, T. Kobayashi, Phys. Chem. Chem. Phys. **21**, 5315-5846 (2019).

Poster No.22

Densities of vibrational states of low-frequency internal motions for flexible and stiff molecules

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The functionality of biomolecules have been assumed being intimately related to the low-frequency internal vibrations. Revealing information on the density of state (DOS) of the low-frequency internal vibrations is therefore of importance. So far, detecting unambiguous experimental signals of the low-frequency vibrations in aqueous solution is still formidable owing to the strong damping effect induced by solvent. It is nevertheless feasible to obtain sharp peaks of the low-frequency modes if we prepare the biomolecules at crystalline state. We employed THz spectroscopy to analyze the DOS of internal vibrations of several oligoglycine and polycyclic aromatic samples. In Figure 1, we show the cumulative growth curves of internal vibrations of (Gly)4 and tetracene in the THz frequency region. Obviously, (Gly)4 has a much faster growth rate of internal vibrations than tetracene does. This fact implies that, at room temperature, there are a greater fraction of internal modes becoming thermally active in the oligoglycine systems compared with that in the polycyclic aromatic systems.

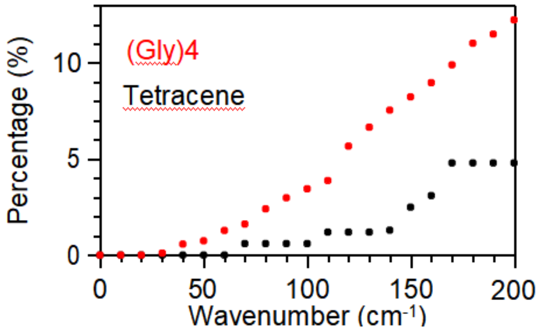

Figure 1. Cumulative growth curves of internal vibrations of $(Gly)_4$ and tetracene in the low-frequency region.

References:

[1] F. Zhang, H-W. Wang, K. Tominaga, M. Hayashi, WIREs Comput. Mol. Sci. **6**, 386-409 (2016)

[2] F. Zhang, M. Hayashi, H-W. Wang, K. Tominaga, O. Kambara, J-I Nishizawa, and T. Sasaki, J Chem Phys. **140**, 174509 (2014)

Poster No.23

Light conversion in bacterial photosynthesis

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In bacterial photosynthesis, light harvesting antennae and reaction centers are responsible for harnessing sunlight. The photosynthetic bacterium Thermochromatium (Tch.) tepidum is a thermophile growing at optimal temperature of 48~50 ºC. Its light harvesting antennae exhibit remarkable thermal stability and near infrared absorption. It is important to study the primary excitation dynamics of both isolated and membrane-embedded light harvesting complexes of Tch. tepidum, so as to understand the mechanisms of light absorption, thermal stability, photoprotection and resistance to environmental stresses.

We have investigated, by the use of triplet excitation profiles (TEPs),[1] the roles of multi-compositional carotenoids (Cars) in the core light harvesting complexes (m-LH1-RCs) from a mutant strain of Rhodobacter (Rba.) sphaeroides.[2] Transient absorption kinetics revealed the triplet excitation transfer from spheroidene (Spe, major composition ~85%) to spirilloxanthin (Spx, minor composition ~8%), implying that the two different kinds of Cars coexist in individual m-LH1-RC complexes. TEP results showed that Spx is involved in photoprotection by quenching ^{3}BChl*, whereas Spe does so merely for BChls of relatively low site energy. The Spe-to-Spx triplet excitation transfer and their inequivalence in quenching ^{3}BChl* constitute a mechanism of cooperative photoprotection.

We have also investigated, by the using femtosecond time-resolved absorption spectroscopy, the uphill excitation energy transfer (EET) from the core antennae (LH1s) to the reaction centers (RCs) by comparing the m-LH1-RC to the native LH1-RC of Tch. tepidum. The former exhibits a substantially large RC-LH1 energy difference (ΔE = 630 cm^{-1}, ~$3k_BT$). The semilogarithmic plot of the EET rate is found to be inversely proportional to ΔE, which consolidates a thermal activation mechanism for the uphill EET. The results are discussed on the basis of the newly reported LH1-RC structure of Tch. tepidum,[3] which allows us to propose the presence of specific doorway BChls in LH1 in promoting the uphill EET.

References:

[1] Yu J., Fu L.-M., Yu L.-J., J. Am. Chem. Soc. **139**, 15984 (2017).

[2] Nagashima, K. V., Sasaki, M., Hashimoto, K., Proc. Natl. Acad. Sci. U. S. A. **114**, 10906 (201 7).

[3] Niwa, S.; Yu, L.-J.; Takeda, K., Nature, **508**, 228 (2014).

Poster No.24

The importance of conformational change in excited states for efficient thermally activated delayed fluorescence

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A novel donor-acceptor (D-A) type (monomer) and its twin D-A type (dimer) molecules with geometries pretwisted conformations have been synthesized to investigate the molecular conformational-dependent mechanism of efficient thermally activated delayed fluorescence (TADF). Chemical structure or conformation of TADF molecules, which intrinsically determines theexcited states dynamics of intramolecular charge transfer (ICT), are crucial for obtaining an efficient reverse intersystem crossing (rISC). In this work, the monomer and dimer were investigated to see what the decisive factor is for efficient TADF, conformations and/or ambient environment (solvent polarity and/or viscosity)? Theoretical calculations suggest that the ΔE_{st} of dimer at S_0 geometry is smaller than monomer, but the ΔE_{st} of dimer at S_1 geometry is larger than that of monomer, which means conformational change in the electronic excited process have more significant effect on monomer. As results, monomer shows a shorter-lifetime and higher-efficiency relative to dimer via steady state spectroscopy and viscosity- dependent control experiments. It is found that suppressing conformational change will lead to a lower delayed fluorescence yield, which is agreement with the calculated results about ΔE_{st} , indicating an excited states conformational change are required for both monomer and dimer to show higher delayed fluorescence yield. Femtosecond and nanosecond transient absorption measurements reveal that both two molecules emit delayed fluorescence after a solvation and conformational stabilized process in low polar toluene and high polar tetrahydrofuran, while the overall photoluminescence quantum yield reduced and lifetime of TADF decreased in more polar THF. The transient spectroscopic measurements also reveal that the radiative process and ISC process of monomer are faster than dimer. The results reveal that the stabilized conformation from S_0 to S_1, and to T_1 state to achieve the smaller orbital overlap value in the excited state is the determining factor controlling the ΔE_{st}. To control geometrical changes between ground state and excited states by designing the large D-A dihedral angel and steric hindrance or segment rigidly geometrical structural molecules is of great importance to controlling the efficient rISC processes for TADF. The work presented herein can expand our understanding of the effect of conformational change of excited states of ICT molecules considered as TADF emitters.

Poster No.25

Two-dimensional spectroscopy study of unnatural amino acid probes

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2D spectra have played a crucial role in the study of structure and dynamics of various proteins and peptides. But their low selectivity and sensitivity limited their application to larger proteins. In many cases, it is difficult or impossible to rely on natural vibration mode of biological molecules to display the structure and environmental information in a site-specific manner. To overcome these difficulties, we have developed various extrinsic vibrational probes that can be incorporated into biological molecules and used to site-specifically study their structural and environmental properties.

Poster No.26

Energy transfer mechanism of light-harvesting complex II revealed by two-dimensional electronic spectroscopy

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Light-harvesting antenna systems such as the light-harvesting complex II (LHCII) in higher plants, are essential for the high-efficiency energy transfer process in photosynthesis [1]. Using two-dimensional electronic spectroscopy (2DES) [2], we explored the possible coherent energy transfer in LHCII trimer from spinach at room temperature. The ultrafast energy transfer pathways are observed through the dynamics of cross peaks between different excitonic states. The recent simulations of 2D electronic spectra provide the comparable results in the identifications of these energy transfer process [3]. In order to reveal the possibility of coherent energy transfer directly, we also introduced the control of polarization in our previous 2DES setup [4] and demonstrated the suppression of incoherent processes in 2D spectra of Oxazine 720 in methanol solution.

References:

[1] T. Mirkovic, E. E. Ostroumov, J. M. Anna, R. van Grondelle, Govindjee, and G. D. Scholes, Chem. Rev. **117**, 249 (2017).

[2] R. D. Zhu, S. Yue, H. Li, X. Leng, Z. Wang, H. L. Chen, and Y. X. Weng, Opt. Express (accepted for publication).

[3] X. Leng, Y. M. Yan, R. D. Zhu, K. Song, Y. X. Weng, and Q. Shi, J. Phys. Chem. B. **122**, 4642 (2018).

[4] S. Yue, Z. Wang, X. C. He, G. B. Zhu, and Y. X. Weng, Chin. J. Chem. Phys. **28**, 509 (2015).

Poster No.27

Ultrafast laser-assisted stabilization of ionized adenine

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Radiation damage induced in DNA is a crucial subject in the study of e.g. cancer therapy [1]. Tissue damage is correlated to the response of DNA building blocks to ionizing radiation and is mostly attributed to secondary electrons carrying an energy comparable with the one transferred through extreme ultraviolet (XUV) attosecond pulses [2,3]. This induces ultrafast charge dynamics dominated by electronic correlations. Here, we present an experimental and theoretical investigation of the photo-fragmentation of the nucleobase adenine following ionization by an isolated attosecond XUV pulse (20–35 eV). The experiment shows the possibility of stabilizing doubly ionized adenine by sending a NIR pulse delayed by 2-3 fs from the XUV, see Fig. 1b. Without NIR or for shorter and much longer delays singly or doubly ionized adenine undergoes fragmentation. We relate this delay to the time it takes for correlation induced charge dynamics to excite the molecule to a state essential for the further ionization through the NIR pulse, see Fig. 1a. This additional ionization removes excess energy from the molecule, hence stabilizing it. We obtain qualitatively agreeing timescales from both a simple Fermi's golden rule approach and ab-initio many-body time- dependent dynamics based on Green's function theory (NEGF). We recognize the relevant mechanism to be a shake-up process, initiated by the XUV pulse, which excites electrons towards shallow molecular states, energetically accessible through 2 NIR photons, see Fig. 1c. To conclude, we find that it is possible to hinder the fragmentation of adenine ions by sending an additional properly timed laser pulse after shake-up processes take place [4].

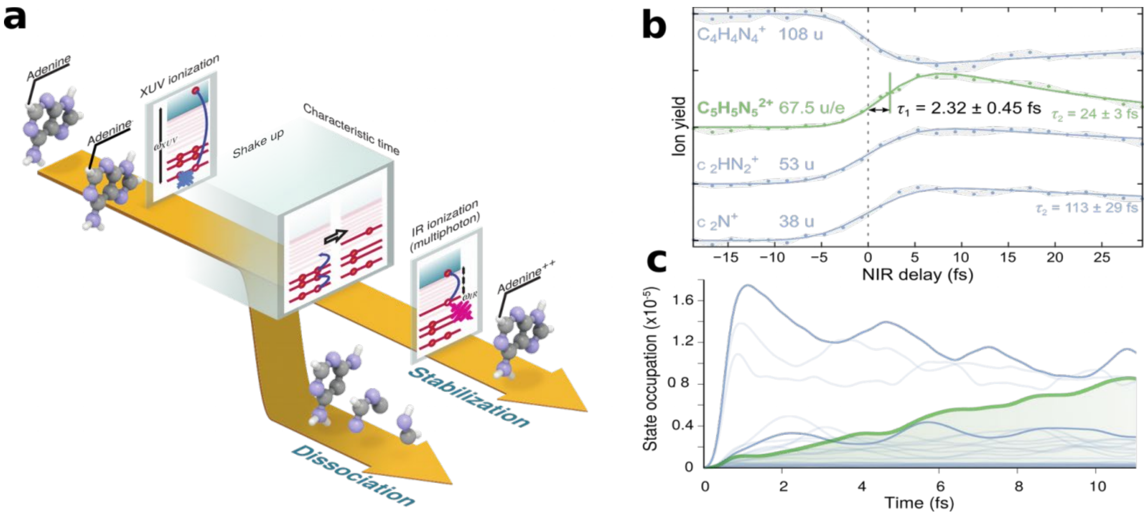

Figure 1. (a) Schematics of the process. (b) Mass spectrometry yields of fragments and adenine dication as a function of NIR delay: the former is delayed by 2-3 fs compared to the latter. (c) Simulated occupations of shake-up states with NEGF.

References:

[1] M. K. Shukla & J. Leszczynski (Eds.), Springer (2008).

[2] F. Calegari et al., Science **346**, 336 (2014).

[3] E. P. Månsson et al., Phys. Chem. Chem. Phys. **19**, 19815 (2017).

[4] F.Covito and co-workers, in preparation.

Poster No.28

Controlling the dynamic processes in rare-earth ion doped luminescent materials by phase-shaped femtosecond laser field

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The ability to control the dynamic processes in the rare-earth ion doped luminescent materials is crucial for their applications in color display, optoelectronic devices, laser sources, bio-labeling and optical memory, etc. Femtosecond laser field, featuring its ultrahigh intensity, ultrashort pulse duration and broad frequency band, can serve as an ideal excitation source for the luminescence of the rare-earth ion doped luminescent materials. In this lecture, I will introduce our recent experimental works on controlling the dynamic processes in rare-earth ion doped luminescent materials by phase-shaped femtosecond excitation laser field, including the valence state conversion manipulation in the doping Sm^{3+} ion, color tuning in the doping Er^{3+} ion and energy transfer control between the co-doping Tm^{3+} to Yb^{3+} ions.

References:

[1] Y. Zheng, Y.H. Yao, L.Z. Deng, W.J. Cheng, J.P. Li, T.Q. Jia, J. R. Qiu, Z. R.Sun, and S.A. Zhang. Photonics Research **6**(2), 144 (2018)

[2] L.Z. Deng, Y.H.Yao, Li. Deng, H.Y. Jia, Y. Zheng, C. Xu, J.P. Li, T.Q. Jia, J. R. Qiu, Z. R.Sun, and S.A. Zhang. Front. Phys.**14**(1), 13602 (2019)

[3] Y. Zheng, L.Z. Deng, J.P. Li, T.Q. Jia, J. R. Qiu, Z. R. Sun, and S.A. Zhang. Photonics Research **7**(4), 486 (2019)

Poster No.29

Frequency-resolved photon-electronic spectroscopy for excited state population detection

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Atomic excitation to excited states in a strong laser field is the key to high-order harmonic generation below the ionization threshold, yet it remains unclear mainly due to the lack of proper detection methods. We propose a frequency-resolved photon-electron spectroscopy technique to reconstruct a population of excited states with the second delayed laser pulse. The technique utilizes Fourier transformation to separate ionization from different excited states to different positions on the spectrum. With the advantage of separation, we provide a scheme to reconstruct populations on different excited states after the first pulse. The scheme is validated by a high-precision population reconstruction of helium and hydrogen atoms.

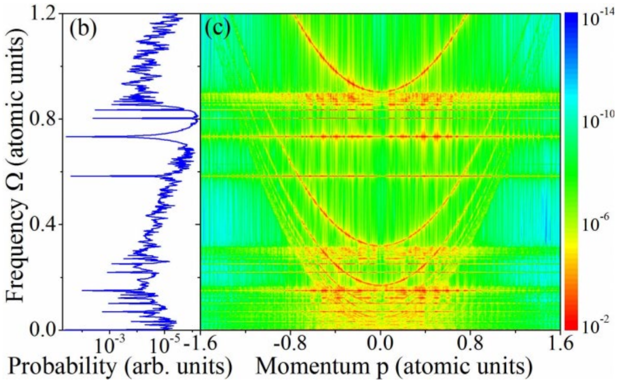

Figure 1. The 2-pulse frequency-resolved photon-electron spectroscopy.

References:

[1] L. Xu, H. Dong, and L.B. Fu, Opt. Lett. **43**, 5725-5728 (2018).

Poster No.30

Detecting electronic coherences by multidimensional high-harmonic spectroscopy

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Multidimensional pump-probe techniques aim to study the dynamics of electronic populations and coherences in molecular systems. To study coherent superposition of electronic bound states with energies more than few eV apart, X-ray light source is needed [1]. High harmonic generation (HHG) using strong IR pulses only can be used to generate electronic coherence via three-step process [2] ending with the recombination with excited bound states. The whole process is presented in figure 1(a) and (b). Using multiple pulses with well controlled delays the variety of multidimensional high harmonic signals are produced in various phase-matching directions. The 2D HHG spectra can be thus obtained by taking double Fourier transformation as shown in figure 1(c). Information about electronic coherences can be extracted from the 2D HHG spectra. To have our proposal carried out theoretically, a complete semi-perturbative model based on Liouville space diagrammatic techniques has been also developed.

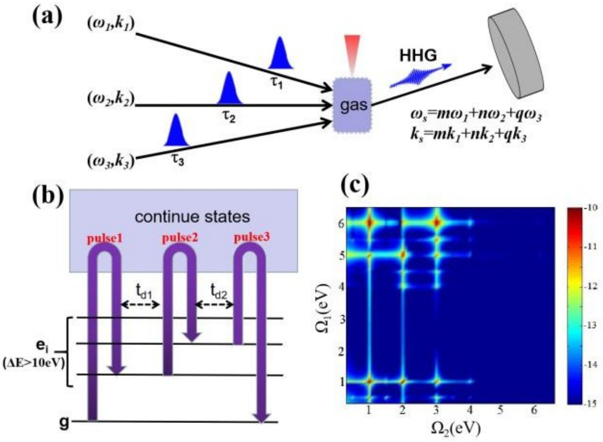

Figure 1. (a) Schematic of the multi-dimensional HHG detection; (b) Three-step model for HHG with multiple pulses; (c) 2D HHG spectra calculated by semi-perturbative model.

References:

[1] K. E. Dorfman, P. Wei, J. Liu, and R. Li, Optics Express **27**, 7147 (2019).

[2] M. Lewenstein, P. Balcou, M. Y. Ivanov, A. L'huillier, and P. B. Corkum, Physical Review A **49**, 2117 (1994).

Poster No.31

Dynamics stark effect on the photodissociation

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To the best our knowledge, the research works pay attention to interference signals of light-induced conical intersections from either excited state or from sum of two electronic states, while ignore interference signal from ground state all the time. Because most studies chose the non-polar diatomic molecules as research system, it is difficult to correlate the dissociation fragments with specific electronic state. However, the HD^+ system can overcome this difficult. The polar diatomic molecule HD^+ has a small energy gap of 3.7 meV between the ground state and the first excited state in the dissociation asymptotic region. The excited state produces the fragments D^++H, while the ground state produces the fragments H^++D.

By taking the HD^+ system as an example, the first-order dynamic Stark effect (DSE) and the intrinsic nonadiabatic coupling (INAC) effect on the photodissociation dynamics have been investigated theoretically. The energy difference between two dressed-state potential energy curves around R=10 a.u. can be equal to zero due to the first-order Stark shift, which results in a new passage connecting the excited state with the ground state. Moreover, the population transfer passage resulting from the INAC effect can be s uppressed by the DSE which enlarges the energy gap between two dressed states at appropriate laser intensity and pulse duration.

Poster No.32

Molecular gases for low energy hollow core fiber pulse compression

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Many femtosecond dynamic studies require a temporal resolution better by a factor of 2-5 compared to the standard pulse duration of many Ti:Sapphire lasers or optical parametric amplifiers, in combination with high repetition rates to acquire data with sufficient statistics. The latter is however often linked to µJ-level pulse energies. This limits the otherwise very efficient technique of spectral broadening in hollow core fibers (HCF) filled with rare gases. Xenon depicts the exception of a gas with very high ionization potential that is capable to broaden such energy level pulses. Here, we show that also hydrofluorocarbons can be used for efficient hollow core fiber pulse broadening at such energies. Fivefold compression, 45 fs down to ~9 fs, of low-energy Ti:Sapphire laser pulses (~16 µJ) is achieved in 1-1 difluoroethane ($C_2H_4F_2$, R152a) and compared to other molecular gases, as well as to conventionally used Argon and Krypton. Additionally, the influence of the group velocity to the spectral broadening dispersion and its pressure dependence are numerically discussed.

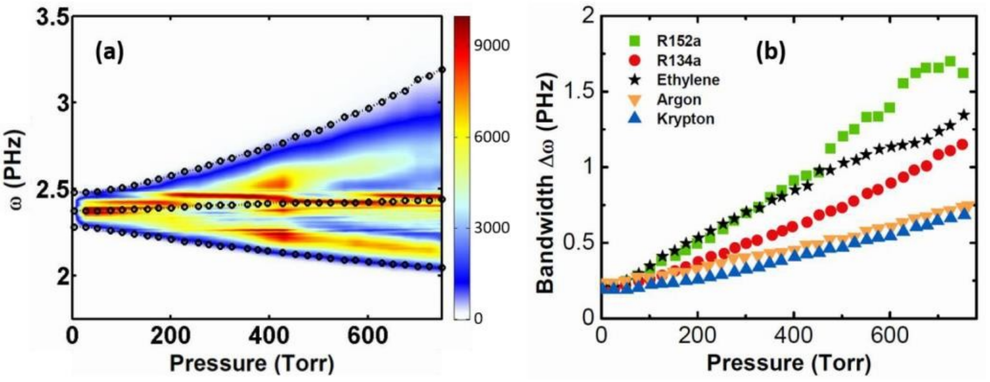

Figure 1. (a) Spectral broadening of a 120 µJ pulse vs. pressure in R134a; (b) Spectral broadening in all investigated gases for a pulse of 120 µJ (except in argon, 240 uJ).

References:

[1] E. Haddad, R. Safaei, A. Leblanc, R. Piccoli, Y.-G. Jeong, H. Ibrahim, B. E. Schmidt, R. Morandotti, L. Razzari, F. Légaré, and P. Lassonde, Opt. Express **26**, 25426 (2018).

[2] R. Safaei, O. Kwon, P. Lassonde, V. Cardin, E. Haddad, A. Leblanc, B. E. Schmidt, H. Ibrahim, and F. Légaré, OSA continuum **2**, 1488 (2019).

Poster No.33

Investigating the probability of photoassociation with random phase thermally two-dimensional wavefunctions

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A method for computing thermally averaged probability of femtosecond two-photon photoassociation (PA) of magnesium atoms is proposed. The method is based on two-dimensional wavefunctions with random phase [1] expansion of unity in the vibrational and rotational degree of freedom. For comparison, the PA probabilities are obtained by other two methods at the temperature of 1 K. For one method, the thermally averaged expectation value is got by Boltzmann thermal averaging all propagated two-dimensional energy eigenfunctions. The other is similar to the method presented by us, whose random phase expansion of unity merely in the vibrational degree of freedom [2]. Compared to other two methods, the new method become more and more favorable in computation effort as the temperature rises.

References:

[1] D. Gelman and R. Kosloff, Chem. Phys. Lett., **381**, 129 (2003).

[2] S. Amaran, R. Kosloff, M. Tomza, W. Skomorowski, F. Paw lowski, R. Moszynski, L. Rybak, L. Levin, Z. Amitay, J. Martin Berglund, D. M. Reich and C. P. Koch, J. Chem. Phys., **139**, 164124 (2013).

Poster No.34

Size-dependent biexciton binding in perovskite nanocrystals studied by two-dimensional electronic spectroscopy

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Colloidal lead halide perovskite (LHP) nanocrystals (NCs) have recently emerged with excellent performances in optoelectronic applications [1]. And the optical gain in LHP NCs, which plays a significant role in the lasing, is strongly related to biexcitons [2]. However, the biexciton binding changed with NC sizes is averaged by inhomogeneity of NCs in one-dimensional spectroscopies. Here, we use two- dimensional electronic spectroscopy [3] to probe the biexciton state in LHP NCs for the first time. We measured the nonlinear optical response of $CsPbBr_3$ NCs at 10 K in different polarization configurations. The cross-circularly polarized ($\sigma^+\sigma^-\sigma^+\sigma^-$) rephasing spectrum (Figure 1(b)) distinctly shows a negative excited state absorption (ESA) from exciton to biexciton, beneath the diagonal, compared to co-circularly polarized ($\sigma^-\sigma^-\sigma^-\sigma^-$) spectrum (Figure 1(c)). Furthermore, the difference between excitation and emission energies of the ESA signal is equal to the biexciton binding energy (Δ_{XX}) [4]. Since the ESA signal is tilt to the diagonal (Figure 1(b)), we can propose that as the size of NC decreases and the emission energy increases, the Δ_{XX} will increase. These results help to understand the biexcitons in LHP NCs further.

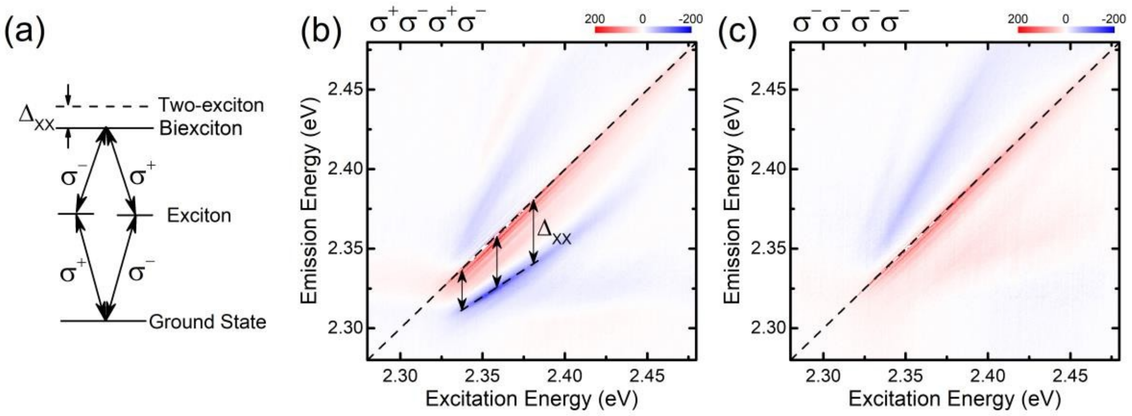

Figure 1. (a) The energy level scheme. (b) Cross-circularly polarized rephasing spectrum. The time delay between the 2nd and 3rd pulse is 150 fs. The dash lines mark the diagonal and the ESA from exciton to biexciton. (c) Co-circularly polarized rephasing spectrum.

References:

[1] Q. A. Akkerman et al., Nat. Mater. **17**, 394 (2018).

[2] W. Zhao et al., J. Phys. Chem. Lett. **10**, 1251 (2019).

[3] W. Zhu et al., Opt. Express **25**(18), 21115 (2017)

[4] G. Moody et al., Phys. Rev. B **87**, 041304 (2013).

Poster No.35

Timing dissociative ionization of H_2 using a polarization-skewed femtosecond laser pulse

Q. Ji[1], S. Pan[1, *], P. He[2], J. Wang[2], P. Lu[1], H. Li[1], X. Gong[1], K. Lin[1], W. Zhang[1], J. Ma[1], H. Li[1], C. Duan[1], P. Liu[3], Y. Bai[3], R. Li[3], F. He[2], J. Wu[1]

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We experimentally time different pathways of the dissociative ionization of H_2 driven by a polarization-skewed laser pulse [1,2], which encodes the ionization instants of H_2 and photon-coupled dipole-transition instants in stretching H_2^+ to the emission directions of the photoelectrons and nuclear fragments, respectively. Our results show that the one-photon and net-two-photon dissociation pathways of H_2^+ spend the same time to reach the one-photon transition internuclear separation after the photoionization of H_2.

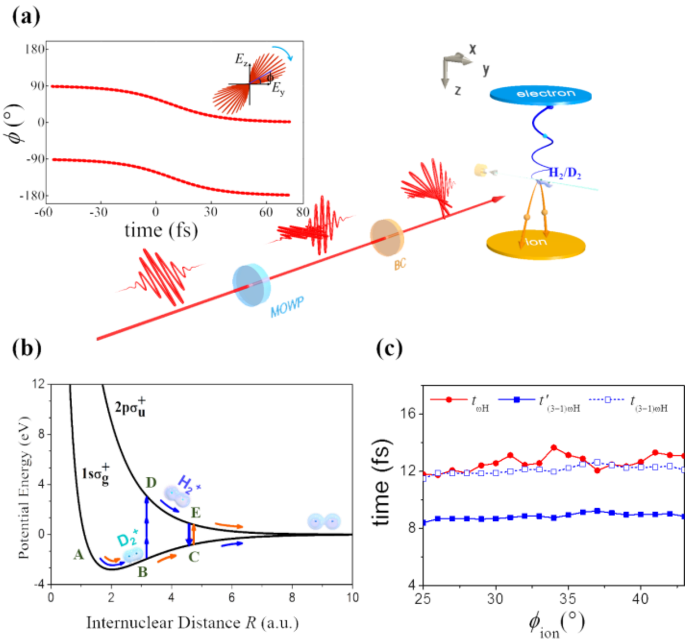

Figure 1. (a) Scheme of experimental setup. (b) The potential energy curves of H_2^+. (c) Experimentally extracted propagation time of different dissociation pathways of H_2.

References:

[1] Q. Ji, J. Wang, P. Lu et al, Phys. Rev. A **96**, 053423 (2017).

[2] Q. Ji, S. Pan, P. He et al, (in submission).

Poster No.36

Coherent control of electronic charge migration via quantum control of nuclear motions

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Recently we have shown that nuclear motions may cause decoherence of electronic charge migration followed by recoherence, on rather different time scales: hundreds of attoseconds for the charge migration, few femtoseconds for the decoherence, and more than one hundred femtoseconds for the recoherence [1]. Here we show how this new phenomenon can be used for a new approach to coherent control of electronic motion via quantum control of nuclear motion. For this purpose, we carry out quantum dynamical model simulations of the concerted nuclear and electronic motions driven by ultrashort pump-and-dump laser pulses which prepare a coherent superposition of two wave functions in the electronic ground and excited states. Proper choice of the time delay between the laser pulses allows control of the time evolution of the nuclear wave functions and their overlap in the different electronic states. Good nuclear overlaps may be achieved in pre-determined time windows, and this switches on (recoherence) and off (decoherence) electronic charge migration in these time windows, even well after the laser pulses. As a proof-of-principle, we present model applications to the iodoacetylenic cation - the same system for which Kraus et al. achieved the first joint experimental and theoretical reconstruction of charge migration [2,3].

References:

[1] D. Jia, J. Manz and Y. Yang, Lecture presented at Femto 14 Shanghai, July 28-Aug. 02, 2019.

[2] P. M. Kraus, B. Mignolet, D. Baykusheva, A. Rupenyan, L. Horný, E. F. Penka, G. Grassi, O. I. Tolstikhin, J. Schneider, F. Jensen, L. B. Madsen, A. D. Bandrauk, F. Remacle and H. J. Wörner, Science **350**, 790-795 (2015).

[3] H. J. Wörner, C. A. Arrell, N. Banerji, A. Cannizzo, M. Chergui, A. K. Das, P. Hamm,U. Keller, P. M. Kraus, E. Liberatore, P. Lopez-Tarifa, M. Lucchini, M. Meuwly, C. Milne, J.-E. Moser, U. Rothlisberger, G. Smolentsev, J. Teuscher, J. A. van Bokhoven and O. Wenger, Struct. Dyn. **4**, 061508 (2017).

Poster No.37

Ultrafast dynamics of single femtosecond laser-induced periodic ripples

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The ultrafast dynamics of periodic ripples are studied during their formation on the surface of gold film with a prefabricated nanogroove. These transient ripples are induced by a single 800-nm, 50-fs laser pulse and are observed by a collinear pump-probe imaging method. When the laser polarization is parallel to the nanogroove, transient ripples begin appearing after an elapse time of 25–80 ps, and become clear and regular at 400–600 ps. The ripple period increases from 685 nm to 770 nm as the laser fluence *F* increases from 0.73 to 3.42 J/cm^2. The evolution of temperature and lattice temperature are theoretically studied using the two-temperature model.

When the laser fluence F is above 0.73 J/cm^2, the electron temperature rises to several 10^4 K, and the collision frequency rises above 10^{16} /s, which further causes the localization of hot electrons. Moreover, the d-band electrons can be excited through two-photon absorption and become free electrons. Using the dielectric constant of the excited states, which includes the effects of hot-electron localization and d-band transitions, the period predicted by the surface-plasmon-polariton (SPP) model accords well with experimental results. Both theory and experiment give support to SPP excitations playing a prominent role in the formation of periodic ripples induced by femtosecond laser pulses.

Poster No.38

Photochemical by-products of femtosecond filamentation in air

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Femtosecond filamentation stems from a dynamic balance between nonlinear Kerr self-focusing effect and plasma/high order Kerr effect-induced self-defocusing effect, which clamps the intensity inside each filament of 5×10^{13}W/cm^2 and plasma density 1×10^{16}/cm^3 [1]. By collision, plasmas and neutral molecules interact with each other and produce photochemical through complex photoxidation reactions. Previous studies have shown that in air the photochemical by-products mainly are ozone (O_3) and nitrogen oxides (NO_x) [2,3]. They together with H_2O/NH_3 form binary chemicals of HNO_3/NH_4NO_3, which are considered as an important contributor to laser-based weather modification [4,5]. To well understand and control the femtosecond filamentation-induced photochemical generation, dedicated study bridges the plasma generation and chemical production is desired.

In our study, a 1 kHz 10 mJ/30fs 800nm femtosecond laser was employed. By manipulating laser parameters, such as laser energy, pulse repetition rate etc., and components of surround air, we find that production of NO_x climbs up with increase of laser energy and surrounding relative humidity. However, O_3 production is suppressed under relative high laser energy employed in the present experiment. Competition occurs between NO_x and O_3 generation and NO_x ultimately is generated preferentially. Underlying physical mechanism was also analyzed.

References:

[1] A. Couairon, A. Mysyrowicz, Phys. Rep. **441**, 47-189 (2007).

[2] Y. Petit, S. Henin, J. Kasparian et al., Appl. Phys. Lett. **97**, 021108 (2010).

[3] L. Ran, Z. Z. Deng, J. J. Ju et al., Atmospheric Environment **206**, 271-279 (2019).

[4] J. P. Wolf, Rep. Prog. Phys. **81**(2), 026001 (2018).

[5] J.J. Ju, J.S. Liu, C. Wang et al., Opt. Lett. **37**, 1214-1216 (2012).

Poster No.39

Following macroscopic effects in gas phase experiments using XUV- UV spectroscopy

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A novel experimental approach employing XUV and UV femtosecond pulses is presented and applied to the study of ultrafast physical processes in atomic targets in the gas phase. In particular, we report on the resonant pulse propagation of XUV pulses in He atoms employing photoelectron spectroscopy.

The propagation of light pulses with a spectrum containing frequencies that are resonant with the hosting medium has been studied extensively in the literature [1, 2] and it has been shown that, when the pulse duration is shorter than the lifetime of the excited states, the pulse envelope can be significantly reshaped along its propagation. Here, we discuss the macroscopic propagation of a resonant XUV attosecond pulse train, APT, by performing photoelectron spectroscopy in He atoms. In particular, we show that the phase accumulated by the resonant propagation of the XUV pulse leads to a chirped electron wave-packet, EWP, with characteristic signatures both in the attosecond and the femtosecond timescales. The use of an APT provides a good compromise between time and spectral resolution, which is essential to explore quantum systems [3]. The use of the 2nd harmonic, which lies in the UV, instead of the often used fundamental frequency, which is in the IR here, has multiple benefits. Specifically, it avoids the spectral congestion, increases the energy range of the accessible bound states and projects the spectral properties of the emitted EWPs to the spatial coordinates. Our results demonstrate the feasibility of studying real atomic and potentially molecular states with attosecond resolution, how the characteristics of matter in the frequency domain could be projected in the real space and how one could gain control on the directionality EWPs.

References:

[1] G. L. Lamb, Rev. Mod. Phys. **43**, 99 (1971).

[2] U. van Bürck, Hyperfine Interactions **123**, 483 (1999).

[3] M. Isinger, et al Science, **358**, 893-896 (2018).

Poster No.40

Anisotropic order-disorder-order phase transition and strong light-matter coupling in black phosphorus visualized by ultrafast electron microscopy

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Similar to graphite and transition metal dichalcogenides, black phosphorus (BP) has a layered structure but uniquely with an in-plane puckered geometry. This structural (nuclear and electronic) anisotropy of BP has attracted explosive research to reveal anisotropic in-plane properties, optical, electrical, thermal, and mechanical, promoting it as a promising platform for application in (opto)electronics and thermoelectrics [1-3]. Despite its fundamental and technical interest, however, experimental investigation of structural dynamics, elementary to all the physical properties, in BP has been overlooked.

Here, we report the in-plane structural dynamics of photoexcited thin-film BP flakes by the time-resolved, structure-sensitive probe of ultrafast electron microscopy [4,5], with its thickness, the polarization and duration of excitation pulse, and the dynamic range being controlled. For the first time, we report the thermally-driven pre-melt phase of BP upon nanosecond photoexcitation with in-plane anisotropy, follow its evolution and relaxation, and discuss the impact of the pre-melt phase on electron transport. In the non-thermal melting regime, dense plasmon- polaritons were generated by intense femtosecond excitation, inducing the patterned deformation in plane with the period of the polaritons' wavelength. Our observation offers insights into the non-equilibrium structural dynamics as a consequence of light-matter interactions in anisotropic two-dimensional materials for tailoring promising optoelectronic, photonic, and thermoelectric devices.

References:

[1] L. Li, Y. Yu, G. J. Ye, Q. Ge, X. Ou, H. Wu, D. Feng, X. H. Chen, and Y. Zhang, Nat. Nanotech. **9**, 372 (2014).

[2] M. Massicotte, P. Schmidt, F. Vialla, K. G. Schädler, A. Reserbat-Plantey, K. Watanabe,T. Taniguchi, K. J. Tielrooij, and F. H. L. Koppens, Nat. Nanotech. **11**, 42 (2016)

[3] X. Ling, H. Wang, S. Huang, F. Xia, and M. S. Dresselhaus, Proc. Natl. Acad. Sci. USA **112**, 4523 (2015).

[4] Y. M. Lee, Y. J. Kim, Y.-J. Kim, and O.-H. Kwon, Struct. Dyn. **4**, 044012 (2017).

[5] Y.-J. Kim, H. Jung, S. W. Han, and O.-H. Kwon, Matter **In press** (2019).

Poster No.41

Dielectric properties manipulated by a shaped ultrashort laser pulse

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Based on the time-dependent density functional theory, we theoretically investigate that a shaped ultrashort laser pulse with high intensity may realize the nondestructive and reversible manipulation of the optical responses in wide band gap material. Taking the insulator cubic boron nitride (cBN) crystal for example, we show the dielectric functions of material irradiated by purposely designed ultrashort single- and double-pulse light. Tuning the parameters of pulse light, the cBN can be switched between isotropic and anisotropic for a particular light frequency during the pulse. The anisotropic properties even reaches the level of hyperbolic dispersion. The reversible and highly nonlinear modification provides a great potential for all-optical signal processing and ultrafast optical-field-effect devices.

Poster No.42

Creating, imaging, and controlling chiral molecules with electric fields

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Chirality is one of the hot topics in the fields of physics, chemistry, and biology as well as pharmacology, with its importance emphasized by the fact that life on earth is based on chiral biomolecules all naturally selected with a single handedness. Chirality is a fundamental and important property of molecules and materials leading to very interesting optical-activity effects in linear and nonlinear optics. Accurate experimental characterization of the enantiomeric excess and absolute handedness in mixtures of chiral molecules, efficient chiral purification and discrimination remain very challenging and highly demanding tasks for a broad scope of applications. Recently, a number of novel experiments have been developed for measuring the enantiomeric excess [1,2] or absolute handedness [3,4].

We will present novel robust techniques devised in our group for (i) producing dynamic chirality from samples of statically achiral molecules [5], (ii) efficient spatial separation of chiral molecules, and (iii) detecting the enantiomeric excess [6]. The results are based on robust and highly accurate variational simulations of molecular ro-vibrational dynamics in the presence of electric fields [7]. We will also present challenges and perspectives for experimental realizations of the proposed techniques.

References:

[1] M. H. M. Janssen, I. Powis, Phys. Chem. Chem. Phys. **16**, 856 (2014)

[2] D. Patterson, M. Schnell, J. M. Doyle, Nature **497**, 475 (2013)

[3] P. Herwig, K. Zawatzky, M. Grieser, O. Heber, B. Jordon-Thaden, C. Krantz, O. Novotný, R. Repnow, V. Schurig, D. Schwalm, Z. Vager, A. Wolf, O. Trapp, H. Kreckel, Science, **342**, 1084 (2013)

[4] M. Pitzer, M.Kunitski, A. S. Johnson, T.Jahnke, H. Sann, F. Sturm, Lothar Ph. H. Schmidt, H. Schmidt-Böcking, R. Dörner, J. Stohner, J. Kiedrowski, M. Reggelin, S. Marquardt, A. Schießer, R. Berger, M.S. Schöffler, Science **342**, 1096 (2013)

[5] A. Owens, A. Yachmenev, S. Yurchenko, J. Küpper, Phys. Rev. Lett. **121**, 193201 (2018)

[6] A. Yachmenev, S. N. Yurchenko, Phys. Rev. Lett. **117**, 033001 (2016)

[7] A. Owens, A. Yachmenev, J. Chem. Phys. **148**, 124102 (2018)

Poster No.43

Phase-matching-free pulse retrieval based on transient absorption in solids

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We report a novel metrology tool to characterize femtosecond pulses. It is free of phase matching constraints, enabling to measure pulses with ultra-broadband spectra and very low energy at the spectrometer detection limit. Transient absorption in solids is used to switch the transmissivity of a thin sample upon photoexcitation with a pump pulse. The drop in transmission is probed with the pulse to be characterized in function of the pump-probe delay. From this frequency resolved optical switching dataset the temporal amplitude and phase profiles of both the optical switch and the probe pulse can be extracted with a phase retrieval algorithm. The only spectral limitation of this technique is the transmissivity of the solid sample. For instance, zinc selenide, provides the capability to characterize pulses with a spectrum spanning from 0.5 to 20 µm, denoting more than 4 octaves. This approach was demonstrated experimentally by measuring the profiles of pulses centered at 0.8, 1.5, 1.75, 4.0, and 10 µm. Furthermore, the robustness of this technique was shown by measuring identical pulses with different solid samples and with different pump pulse durations.

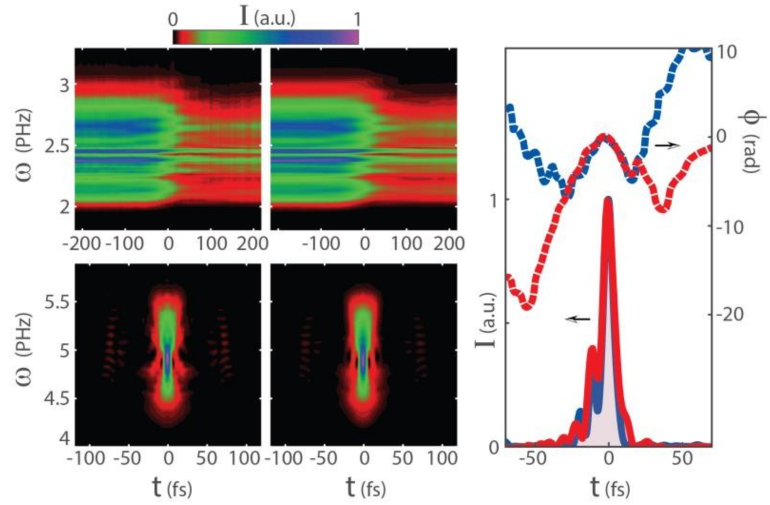

Figure 1: 1st row: frequency resolved optical switching measurement of a few-cycle (7.5 fs) 800 nm pulse. – 2nd row: Reference SHG-FROG measurement of the same pulse. – 1st column: experimental dataset. –2nd column: reconstructed dataset with a ptychographic algorithm. – Last column: extracted temporal amplitude and phase profiles, using FROG (blue) and the optical switching technique (red).

Poster No.44

Superradiance from nitrogen ions pumped by femtosecond laser filed

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The so called “air lasing” is a typical cooperative quantum effect due to the superradiance of highly concentrated atoms, molecules or ions[1-3]. Because of its simple implementation and high optical gain in short wavelength, it holds a great potential in remote detection and ultraviolet laser source.Here, we study this superradiance by injecting a pair of resonant seeding pulses at different intensity ratios inside the nitrogen gas plasma. Strong modulation of the 391.4 nm superradiant emission with a period of 1.3 fs is observed when the delay between the two seeding pulses is finely tuned. Modulation contrast as a function of the time delay is showed in the below figure for different ratio of intensity between the two seeding pulses. Experimental observations confirmed the key role of the coherence in the N_2^+ "air lasing" and provided a new perspective for the clarification of the dynamics of superradiance.

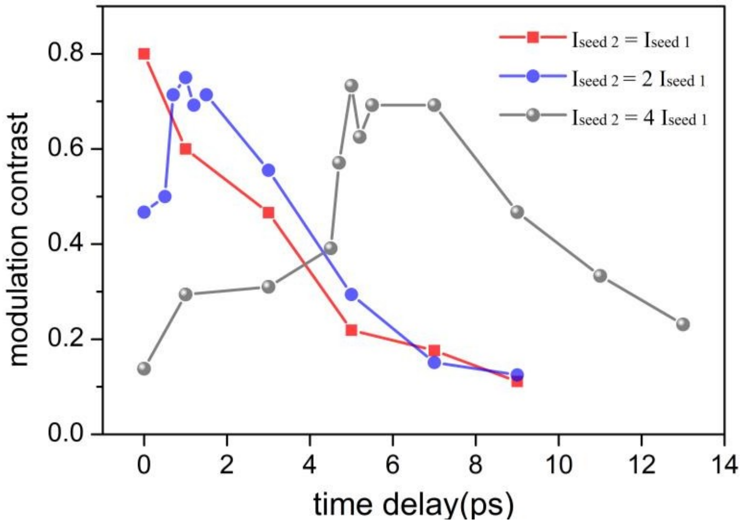

Figure 1. Superradiance modulation contrast as a function of the time delay.

References:

[1] Y. Liu, et al, Recollision induced superradiance of ionized nitrogen molecules, Phys. Rev. Lett. **115**, 133203 (2015).

[2] Y. Liu, et al, Unexpected sensitivity of nitrogen ions superradiant emission on pump laser wavelength and duration, Phys. Rev. Lett. **119**, 203205 (2017).

[3] H. Xu, et al, “Sub-10-fs population inversion in N2+ in air lasing through multiple state coupling,” Nat. Commun. **6**, 8347 (2015).

Poster No.45

Manipulating momentum and angular distributions of photoemission from hydrogen atoms by few-cycle super-intense and ultra-short laser pulses

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With the rapid development of laser light sources, especially the free electron laser, it is expecting soon to generate super-intense laser intensities of the order of 10^{23}-10^{24} W/cm^2 [1]. Meanwhile, the ultra-short pulses with only about 50 as have been reported [2,3]. It is interesting to know what the new effect is when the laser pulse is extremely short and intense.

By numerically solving the time-dependent Schrödinger equations, we investigated the momentum and angular distributions of ground sate hydrogen atoms irradiated by few-cycle super-intense and ultra-short laser pulses. We discussed numerical results with various laser intensities and carrier-envelop phases (CEPs), and studied the instantaneous emission of photoelectrons. By analyzing the momentum and the angular distributions (see Figure 1), CEP and ionization yield, we found that the instantaneous amplitude of electric field play the decisive role in dynamic process. By varying the laser intensities and CEPs, we can manipulate the instantaneous electric field and therefore control the momentum and angular distributions of photoelectrons.

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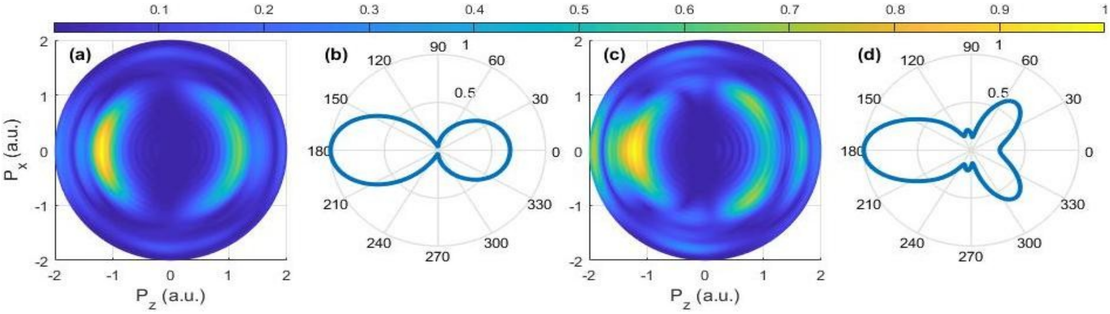

Figure 1. (color online) Photoelectron momentum and angular distributions of hydrogen atoms under pulses of 5 o.c. with central carrier frequency 1 a.u. and peak electric field E_0=3 (a,b) and 6 (c,d) a.u..

References:

[1] The ELI project, https://eli-laser.eu. Webpages were pulled at April. 10, 2019.

[2] J. Li et al., Nat. Commun. **8**, 186 (2017).

[3] T. Gaumnitz et al., Opt. Express **25**, 27506 (2017).

Poster No.46

Improving super-resolution nanoscopy by actively controlling the fluorescence of nanoparticles

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The super-resolution nanoscopy circumvents the Abbe's diffraction limit and extends the optical resolution down to nanometer scale. For all super resolution methods, the most fundamental requirement is to generate or to make use of the fluorescent variation of individual emitters. Some single-molecule localization microscopies (SMLM), such as STORM [1] and SOFI [2], are based on the extraction of stochastic optics or spontaneous blinking fluctuations to achieve super-resolution. However, there is not an active way to effectively manipulate individual the behavior of emitters fluorescence. This research introduces an active control strategy based on the principle of excited state coherent control [3], femtosecond pulse shaping, multiphoton microscopy and periodic modulation. We extract the independent fluctuations obtained by active control by periodic enhancement, and input them into the localization algorithm to obtain super- resolution results. This proof-of-principle study has pushed the resolution of the wide field nanoscopic imaging towards the range of diffraction unlimited technique. It can resolve twin emitters with 50~70nm apart and continuous linear structure with localization accuracy of ~30nm, which both for simulation and experiments with ZnCdS quantum dots (QD) films and QD625 labelled COS7 cells. Besides, the technique is a general method for various probing systems including dyes, proteins, and nanoparticles etc. In addition, since it is purely optical and modifies only the temporal properties of the femtosecond pump laser, it is also feasible to incorporate into other (super-resolution) microscope systems.

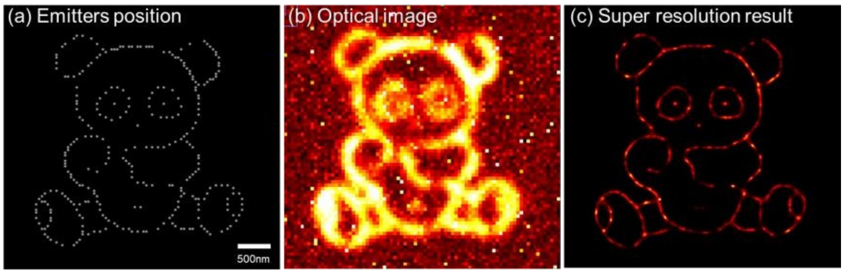

Figure 1. Super resolution resolving imaging of super-resolution nanoscopy by actively controlling (SNAC).

References:

[1] Rust, M. J., Bates, M. & Zhuang, X. Nature Methods **3**, 793-796 (2006)

[2] Dertinger, T., Colyer, R., Iyer, G., Weiss, S. & Enderlein, J. Proceedings of the National Academy of Sciences of the United States of America **106**, 22287-22292 (2009).

[3] Brif, C., Chakrabarti, R. & Rabitz, H. New Journal of Physics **12**, 075008 (2010).

Poster No.47

Surface heterogeneity of perovskite thin films observed with PEEM

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In recent years, lead halide perovskite has been paid a tremendous attention as a promising optoelectronic material since its first innovative application in dye-sensitized solar cells with efficiency 3.8% and the PCE record of perovskite solar cells has achieved 24.2% [1]. However, the charge transportation of this kind of material remains ambiguous, especially the influence of surface traps [2]. Here, we combine the pump-probe technique and photoemission electron microscopy (PEEM) to investigate the surface carrier dynamics of $CsPbBr_3$ thin films directly [3]. Our result shows the surface heterogeneity of perovskite thin films, in which there are two types region including dark state represents large grains and bright state represents grain boundary, cluster of little grains. Time resolved experiment shows that the charge carrier generates at the dark state and diffuses to the bright state within picoseconds, which confirms the trapping nature of the bright state. We anticipate that our result and further experiment would disclose the mechanism hiding in perovskite materials and enhance the efficiency of optoelectronic devices based on lead halide perovskite.

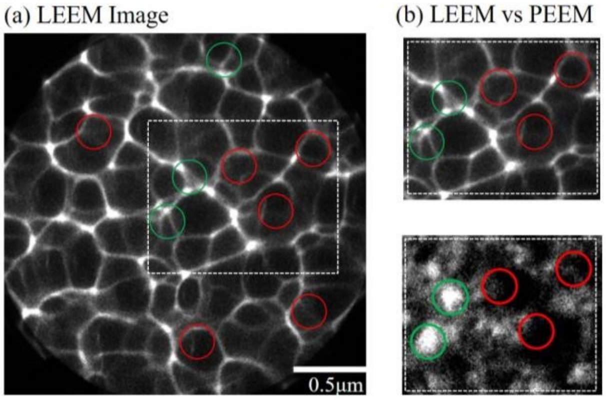

Figure 1. LEEM imaging of perovskite thin films (a) and the imaging compare of LEEM (b, top) and PEEM (b, bottom).

References:

[1] https://www.nrel.gov/pv/cell-efficiency.html

[2] X. Wu, M.T. Trinh, D. Niesner, et al. Journal of the American Chemical Society **137**, 2089 (2015).

[3] Michael K.L. Man, Athanasios Margiolakis, Skylar Deckoff-Jones, et al. Nature Nanotechnology **12**, 36 (2017)

Poster No.48

Coherent control of the cavity-free lasing of nitrogen ions pumped by femtosecond laser pulses

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Air exposed to intense femtosecond laser pulse turns into plasma, which radiates in a wide range of electromagnetic spectrum. In 2011, coherent emission from atomic Oxygen and singly ionized Nitrogen from air plasma in both backward and forward directions was reported individually by two groups [1, 2], which was later coined as “air laser”. This concept has attracted much attention since the backward emission provides a concept-new light source for remote sensing application, which is expected to improve the sensitivity of remote sensing tremendously [1]. The “air laser” emission at 391.4 and 427.8 nm, corresponding to the transition between $B^2\Sigma_u^+$ and $X^2\Sigma_g^+$ levels of the singly ionized nitrogen molecule, was reported when 800 nm femtosecond pulses were used to pump ambient air. Pumped with the widely available high power femtosecond laser pulses, lasing of nitrogen molecules holds great potential for remote sensing application. Up to now, interpretation of this emission remains controversial and several different models have been proposed [3-5].

In this talk, we first present time-resolved measurements of this 391.4nm emission showing that the emission is of the nature of superradiance [6,7], a cooperative emission process enabled by macroscopic coherence between the relevant energy levels. To further verify the presence of macroscopic coherence and reveal its temporal evolution, we performed two coherent control experiments, addressing respectively the $B^2\Sigma_u^+ - X^2\Sigma_g^+$ and $A^2\Pi_u - X^2\Sigma_g^+$ transitions of the nitrogen ions[8,9]. Strong intensity modulation of the 391.4nm emission was observed in both cases, indicating the crucial role of macroscopic coherence in the lasing process of nitrogen ions.

In the first experiment, we injected a pair of resonant seeding pulses at different intensity ratios inside the nitrogen gas plasma [8]. Strong modulation with a period of 1.3 fs which corresponding to the $B^2\Sigma_u^+ - X^2\Sigma_g^+$ transition is observed

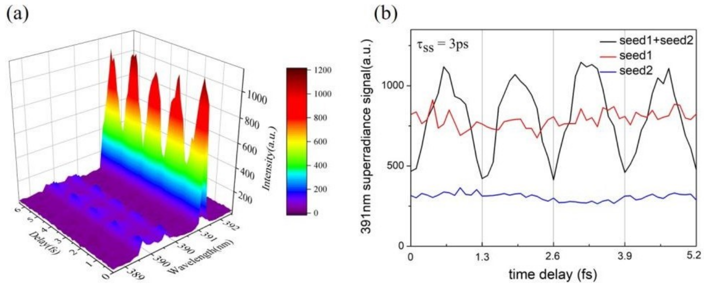

Fig. 1. Coherent control with a pair of seeding pulses addressing the B-X transition. (a) Spectrum of the forward emission for tuning of time delay, the delay between the two seeding pulses is about 3 ps. (b), Intensity of the 391.4nm spectrum component versus time delay.

when the delay between the two seeding pulses is finely tuned, shown in Fig. 1 (a)-(b). The modulation contrast increases and then decreases with the delay time when the second seed pulse is stronger than the first one, and the maximum modulation contrast occurs at longer delay time when the second seeding pulse is stronger. This reveals the increase of the macroscopic polarization with time after the seeding pulse. In the second experiment, we launched a second weaker femtosecond control pulse at 800 nm after the main 800 nm pump pulse, and measured the forward emission at 391.4 nm and sideway fluorescence as a function of the delay between the pump and control pulses [9]. A pronounced modulation with a period $X^2\Sigma_g^+ - A^2\Pi_u$ transition was observed by fine tuning the delay around several fixed values up to 5 ps, as shown in Fig. 2 (a) and (b). We note that the observed modulation is not due to field interference between the pump and control pulses because their time separation largely exceeds the pulse duration. Rather, it arises from the coherent interaction of the second injected pulse field with the long-lived polarization generated by the first pulse. At the same time, it was noticed that the sideway fluorescence, which is proportional to the population in the upper $B^2\Sigma_u^+$ state, does not present any visible change, see Fig.2(c).

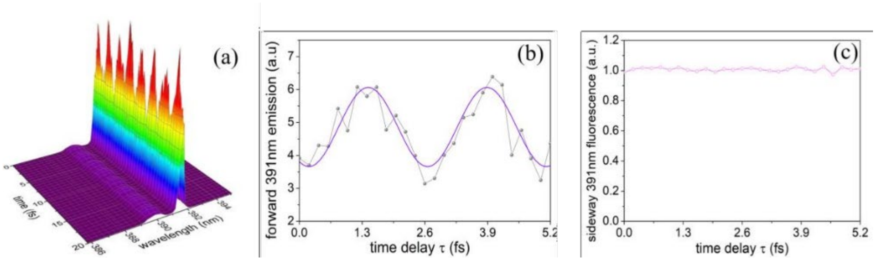

Fig. 2. Coherent control with a pair of pump-control pulses addressing the A-X transition. (a), Spectrum of the forward emission as a function of the pump-control time delay in the vicinity of 1 ps. (b), Intensity of the 391.4 nm spectral component for fine tuning of the time delay around 1 ps. (c) Sideway fluorescence at 391.4 nm measured in the same condition as in (b).

In summary, two coherent control experiments addressing respectively the B-X and A-X optical transitions of nitrogen ions show the crucial role of macroscopic coherence in this lasing action. Moreover, these observations provide a new level of control on the "air lasing" based on nitrogen ions, which can find potential applications in optical remote sensing. A theoretical model based on the Maxwell-Bloch equations, taking into consideration of the macroscopic coherence, has been developed and will also be discussed.

References:

[1] A. Dogariu, J. B. Michael, M.O. Scully, R.B. Miles, “High-gain backward lasing in air”, Science **331**, 442-445 (2011).

[2] J. Yao, *et al*, “High-brightness switchable multi wavelength remote laser in air,” Phys. Rev. A **84**, 051802(R) (2011).

[3] J. Yao, *et al*, “Population redistribution among multiple electronic states of molecular nitrogen ions in strong laser fields,” Phys. Rev. Lett. **116**, 143007 (2016).

[4] H. Xu, E. Lotstedt, A. Iwasaki, K. Yamanouchi, “Sub-10-fs population inversion in N2+ in air lasing through multiple state coupling,” Nat. Commun. **6**, 8347 (2015).

[5] A. Azarm, P. Corkum, and P. Polynkin, “Optical gain in rotationally excited nitrogen molecular ions,” Phys. Rev A **96**, 051401(R) (2017).

[6] Y. Liu, *et al*, “Recollision-induced superradiance of ionized nitrogen molecules,” Phys. Rev. Lett. **115**, 133203 (2015).

[7] Y. Liu, et al, “Unexpected sensitivity of nitrogen ions superradiant emission on pump laser wavelength and duration,” Phys. Rev. Lett. **119**(20), 203205 (2017).

[8] A. Zhang, et al, “Coherent modulation of superradiance from nitrogen ions pumped with femtosecond pulses,” Opt. Express, **27**, 12638 (2019).

[9] A. Mysyrowicz, *et al*, “Lasing without inversion in air,” Arxiv 1806.05818 (2018).

Poster No.49

The propagation effects in the absorption spectra of Rb

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The exponential decay of a discrete excited state with a finite lifetime gives rise to the well-known Lorentzian line shape in the absence of external fields. Here, we observe and model the breakdown of the underlying weak- excitation approximation by single-pulse absorption spectroscopy. The intensity-dependent profiles suggest the dipole response is phase-modulated by the intense excitation pulse itself. Further, as the atomic density increases, the Fano- like shapes are broadened, with novel substructures emerging therein for sufficiently large densities. These are clear signatures of propagation effects in which collective interactions need to be taken into account, and linear Beer's law is no longer valid. Laser-modified spectral profiles are explored and identified as being the combined contribution of self-modulation and propagation effects. Afterwards, spectral line-shape modification of the V-type system excited and coupled by two pump and probe pulses with a variable time delay is investigated, from the single-atom level into deep-density cases. The coherence of the upper levels and pulse propagation are uncovered to account for the dynamical behaviors and their sensitive dependence on pulse intensity and atomic density. In knowledge of these fundamental phenomena in single-pulse and time-resolved absorption spectra will enable a closer scrutiny of strong-field dynamics in more complex systems. In addition, the strong dependence of the resulting lines on intensity and density also points to the possibility of shaping light field by the synergic action of intense-field and propagation effects, paving the way towards controlling quantum processes on ultrafast timescales.

We first measured line-shape evolution as a function of pulse intensity in Rb at 70 ℃, in the dilute-gas limit. With the increase of pulse intensity, the absorption lines no longer stay as natural, symmetric Lorentzian shapes. Instead, asymmetric Fano lines emerge for increasing intensity. The intensity-dependent line profiles indicate the dipole responses are phase modulated by the single excitation pulse itself [1].

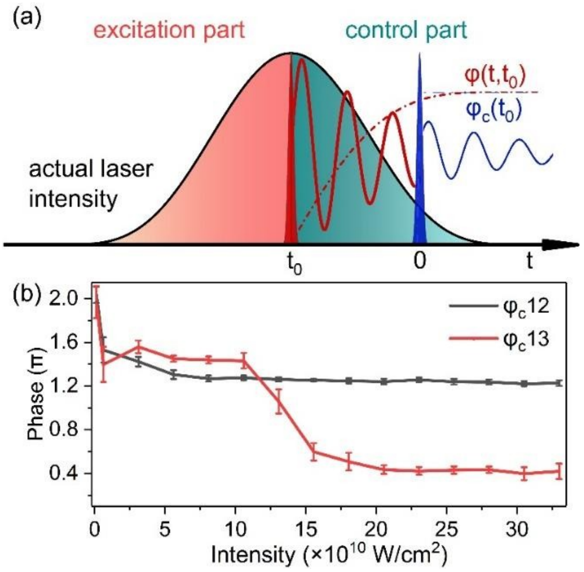

Figure1. (a) Analytical model. The actual laser is divided into excitation and control parts artificially. (b) Phases extracted from measured spectra.

For a straightforward interpretation of the phenomenon and to include the self-modulation effects for simplicity, we divide the laser pulse into excitation and control parts, as is shown in Fig. 1. In this way, an intuitive physical picture is presented. We model the polarization response to be initialized at the peak of laser pulse envelope, and the subsequent control part to Stark-shift the excited states. It thus imprints an additional phase, and the disturbed dipole response function is described as

$$\mathrm{d}(\mathrm{t},\mathrm{t}_0) \propto \begin{cases} 0 & t < t_0 \\ ie^{\left(-i\omega_0-\frac{\Gamma}{2}\right)(t-t_0)} & t_0 \leq t < 0. \\ ie^{\left(-i\omega_0-\frac{\Gamma}{2}\right)(t-t_0)}e^{i\varphi_c(t_0)} & 0 \leq t \end{cases}$$

Results in Fig. 1(b) is the Phases extracted from measured spectra as a function of pulse intensity. These artificial phases reflect the interaction strength. The modulation effect typically increases with a stronger laser pulse, showing as approximately monotonous decrease for both resonances. A more remarkable intensity dependence is displayed for φ_c13, as the stronger |1→|3⟩ transition is more susceptible to external light fields. However, complex higher-order (e.g. Raman) processes between the resonances and further excitation/ionization of Rb can lead to deviations from the simple monotonous behavior. The near-constant plateaus suggest the electronic dipoles become more and more insensitive to the laser pulse with the increase of pulse intensity.

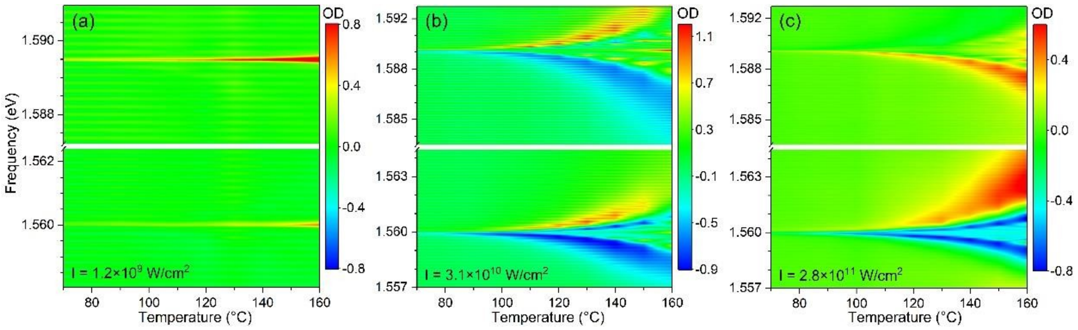

Figure2. Experimental absorption spectra as a function of temperature with different fixed pulse intensities.

To go beyond the single-atom response, the analytical formula for the polarization response when taking consideration of macroscopic effects is expressed as:

$$\mathrm{d}(\omega, \mathrm{x}, \mathrm{t}_0) = \chi(\omega)\mathrm{E}_\mathrm{e}(\omega, \mathrm{x}, \mathrm{t}_0) + \mathrm{i}\chi(\omega)\frac{(\mathrm{L}-1)\mathrm{Q}(\mathrm{x}, \mathrm{t}_0)}{\sqrt{8\pi\mathrm{N}\mu^2}}.$$

We use the following wave equation for the pulse propagation:

$$\frac{\partial}{\partial x}\mathrm{E}_\mathrm{e}(\omega, \mathrm{x}, \mathrm{t}_0) = -2\pi\mathrm{i}\frac{\omega}{\mathrm{c}}\mathrm{d}(\omega, \mathrm{x}, \mathrm{t}_0).$$

The space-dependent absorbance is readily calculated as

$$\mathrm{OD}(\omega, \mathrm{x}, \mathrm{t}_0) = \ln\left[\frac{|\mathrm{E}_\mathrm{e}(\omega, 0, \mathrm{t}_0)|^2}{|\mathrm{E}_\mathrm{e}(\omega, \mathrm{x}, \mathrm{t}_0)|^2}\right].$$

The experimental measured absorption spectra were then reproduced, and the results are displayed in Fig. 3.

Time-delay scans of optical density for different atomic densities was also performed with different cell temperatures. The intense pump pulse renders the conspicuous dynamical beats for both positive and negative time delays, and the~140 fs oscillation period corresponds to the spin-orbit spitting of 0.03 eV. Spectral profiles are broadened with substructures emerging in the center of lines, and hyperbolic curves are observed for negative delays. At time delays close to zero, the absorption spectrum is strongly modified.

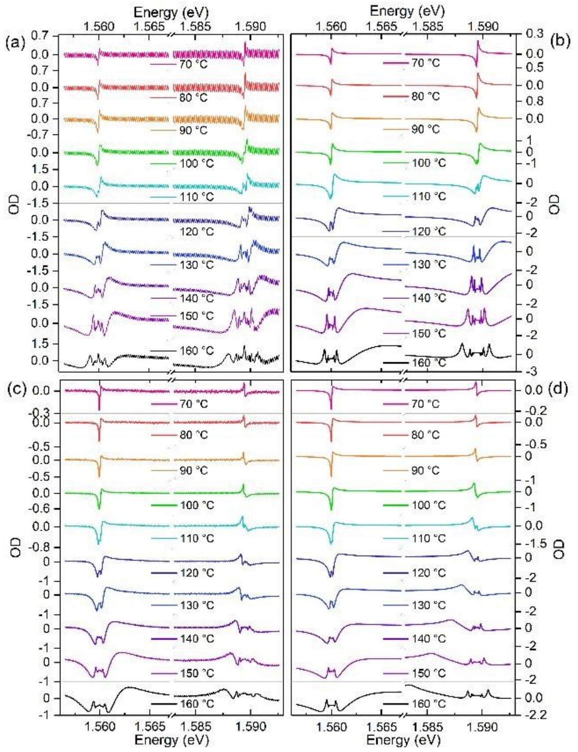

Figure 3 (a), (c) Experimental line shapes shown in contrast to (b),(d) simulations at fixed pulse intensities [3.1×10^{10} W/cm^2 in (a) and (b), 2.8×10^{11} W/cm^2 in (c) and (d)] with varying temperature.

In conclusions, spectral lines in single-pulse absorption spectra are governed by the combined action of self-modulation and propagation effects. The coherence of the upper levels and pulse propagation account for the quantum beats and the rich spectral features on line center in time-resolved transient absorption spectra.

References：

[1] Y. He, Z. Liu, Z. Cui, Y. Zhang, A. N. Pfeiffer, T. Pfeifer, J. Ding, and B. Hu, Signatures of self-modulation effects during pulse propagation in single-pulse absorption spectra, Phys. Rev. A **99**, 053418(2019).

Poster No.50

Ultrafast dissociation of OCS influenced by neighbor ions $OCS^{(1,2)+}$ in a dimer

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We have experimentally observed two Coulomb explosion (CE) channels $(OCS)_2^{(3,4)+} \rightarrow OCS^{(1,2)+} + OC^+ + S^+$ of $(OCS)_2$ in strong laser fields, and studied the three-body dissociation dynamics based on coincidence measurement. Two indirect and one direct pathways are assigned and shown in Fig. 1. Three channels can be distinguished in two-dimensional kinetic energy correlation diagram as shown in Fig. 1(a). The KER for S+ from three channels are shown in Fig. 1(b). The indirect dissociation process of OCS^{2+} in the dimer is confirmed to be the same as the two-body dissociation from monomer $OCS^{2+} \rightarrow OC^+ + S^+$, which have two different channels as shown in Fig. 1(c) and (d). The corresponding three-body dissociation process and the influences of neighbor ions in the dimer have been discussed [1,2]. The neighbor ions $OCS^{(1,2)+}$ enhance the yield of low KER ions for the indirect pathways, and create a new excited state of OCS^{2+} for generate the direct pathways.

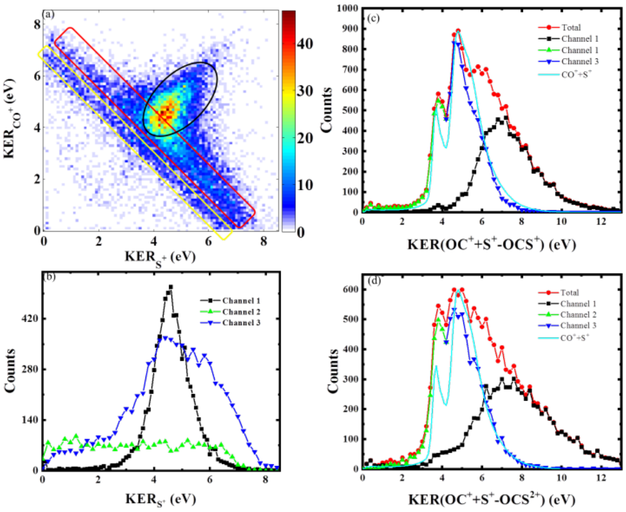

Figure 1. (a) KER correlation distribution of three channels, (b) the corresponding KER spectra for S^+, (c-d) the KER of $OC^+ + S^+$-$OCS^{(1,2)+}$ which represent the KER of the second step.

References:

[1] X Ding et al., Phys. Rev. Lett. **118**, 153001 (2017).

[2] A. Méry et al., Phys. Rev. Lett. **118**, 233402 (2017).

Poster No.51

Coincidence velocity map imaging using TPX3Cam, a time stamping optical camera with 1.5 ns timing resolution

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We demonstrate a coincidence velocity map imaging apparatus equipped with a novel time-stamping fast optical camera [1], TPX3Cam, whose high sensitivity and nanosecond timing resolution allow for simultaneous position and time-of-flight detection. This single detector design is simple, flexible, and capable of highly differential measurements. We show detailed characterization of the camera and its application in strong field ionization experiments.

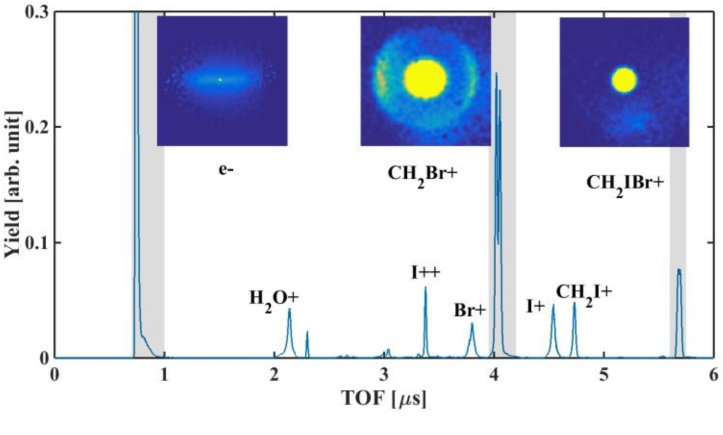

Figure 1. TOF mass spectrum for CH_2IBr following strong field ionization. Momentum distribution are shown for selected fragments.

References:

[1] A. Nomerotski, arXiv: 1902.01357v2 (2019).

Poster No.52

Subcycle transfer of photon momentum in nondipole tunneling ionization

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Up to date, the studies of photon momentum transfer in nondipole tunneling ionization focus on the integral momentum transfer for the entire pulse. We, instead, simulate the subcycle transfer of photon momentum for a typical attoclock setup. The rotating vector potential of the elliptically polarized laser pulse acts as the clock of momentum transfer in the laser propagation direction. We find that the average transferred photon momentum shows a pronounced minimum near the pulse peak where the emission probability is maximal, as illustrated in Figure 1. The (overall) nonzero shift of the longitudinal momentum is predicted to be $p_z = E/c + I_p/3c$ [1] (for the entire pulse), where the first term comes from the propagation in the field after ionization while the second term comes from the tunneling process. We use the backpropagation method [2] to explicitly separate these two contributions in order to test the expected shift, as well as to resolve these contributions on the subcycle scale.

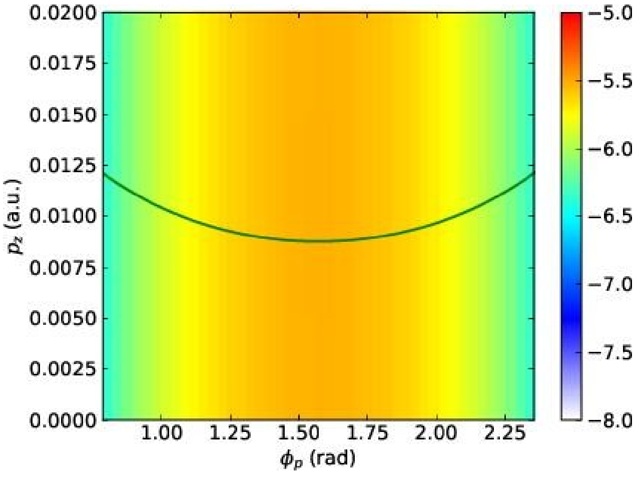

Figure 1. Electron momentum distribution around the maximal emission direction (logarithmic scale). The ϕ_p axis is the electron emission angle within the polarization plane and p_z is the longitudinal momentum in the laser propagation direction. The green solid line is the average longitudinal momentum as a function of emission angle $\langle p_z \rangle(\phi_p)$.

References:

[1] M. Klaiber et al., Phys. Rev. Lett. **110**, 153004 (2013).

[2] H. Ni et al., Phys. Rev. Lett. 117, 023002 (2016); Phys. Rev. A **97**, 013426 (2018); Phys. Rev. A **98**, 013411 (2018).

Poster No.53

Femtosecond EUV beamline for ultrafast materials research and development

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A new femtosecond, Extreme Ultraviolet, time resolved Spin-ARPES beamline was developed for ultrafast materials research and development. The 50fs laser-driven, table-top beamline is an integral part of the “Ultrafast Spintronic Materials Facility”, dedicated to engineering new ultrafast materials. This facility will provide a fast in-situ analysis and development of new materials. The high harmonic generation EUV source emits a flux of 2.3×10^{11} photons/second (2.3×10^{8} photons/pulse) at H23 (35.7 eV). The photon energy ranges from 10 eV to 70 eV enabling surface sensitive studies of the electronic structure. Ultrafast surface photovoltaic effect with ~650 fs rise-time was measured in p-GaAs (100) from time-resolved ARPES spectra. The data acquisition time could be reduced by over two orders of magnitude by scaling the laser driver from 1 kHz, 4W to 1 MHz, kW repetition rate and average power respectively.

References:

[1] Z. Nie, et al., Femtosecond EUV Beamline for Ultrafast Materials Research and Development. Appl. Sci. **9,** 370 (2019).

[2] Petersen, et al. Clocking the melting transition of charge and lattice order in 1T-TaS2 with ultrafast extreme-ultraviolet angle-resolved photoemission spectroscopy. Phys. Rev. Lett. **107,** 177402 (2011).

[3] Gierz, et al., Snapshots of non-equilibrium Dirac carrier distributions in graphene. Nat. Mat. **12,** 1119-1124 (2013).

Poster No.54

Transient alignment of liquid water

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The solvation of molecules in water is pivotal for a myriad of molecular phenomena and is of crucial importance to understand diverse issues such as chemical reactivity and biomolecular function. It has been shown that laser techniques in the infrared (IR) and Terahertz (THz) frequency ranges offer fundamental insights into hydration from small solutes to proteins.

In bulk liquid, motions of water molecules lead to ultrafast fluctuations at femto- to pico-second time scales. Underlying molecular processes range from diffusional motions spanning nanoseconds, rattling modes of anions and cations within their solvation shells on a ps time scale, the breaking and reformation of hydrogen bonds, and includes sub-100 fs librational motions.

Here we report unprecedented non-linear THz experiments on water molecules in the liquid phase. This novel finding shows that molecular alignment in bulk liquids is feasible.

Poster No.55

Vibrational echo in argon dimer

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Echo is a ubiquitous phenomenon found in many physical systems, ranging from particle beams in hadron accelerators [1] to a gas of rotating molecules [2].In 1950, Erwin Hahn reported [3] that if a spin system is irradiated by two properly timed and shaped pulses, a third pulse appears at twice the delay between the first two. In the above spin echo system, the mirror is played by a second, time delayed pulse which is able to reverse the flow of time and recreate the original impulsive event.

In our experiments [4], using an ultrashort femtosecond laser pulse, we create a localized vibrational wave packet in an argon dimer cation. This wave packet oscillates back and forth in the ion potential well and disperses completely due to the spread of the oscillation frequencies within the wave packet. Next, after a time delay of t_k, by applying a second pulsed excitation and waiting a time period equal to the delay between the two excitations, a pulsed coherent signal emerges again, which is referred to as "echo".

Here, by utilizing pump (kick) and probe pulses of mutually detuned wavelengths and COLTRIMS methodology, we identify the right events from the background which allow to follow the vibrational dynamics with femtosecond resolution. Experimental results unambiguously show the change in KER yield signal periodicity at t ≈ 2tk, corresponding to the predicted time of echo appearance. The results of numerical simulations support the observation. In conclusion, the classical in nature echo effect is inter wined with quantum revivals and this novel kind of echo may provide opportunities for further experimental and theoretical research related to quantum/classical correspondence in molecular systems.

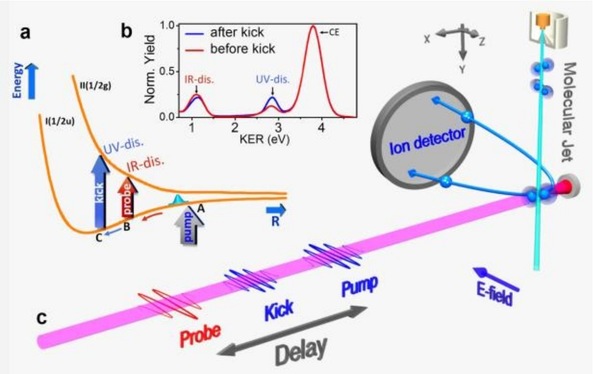

Figure 1. a The field-free potential energy curves of Ar_2 cation. b Dissociation channel induced by 790 nm and 395 nm pulses with different KER ranging from 0.7 eV to 3.2 eV, together with the Coulomb explosion (CE) double ionization channel close to 4eV. c Schematic illustration of the experimental setup.

References:

[1] Spentzouris L K et al., Phys. Rev. Lett. 76620(1996)

[2] K. Lin et al., Phys. Rev. X **6**, 041056 (2016).

[3] E. L. Hahn, Phys. Rev. **80**, 589 (1950).

[4] Submitted to Nature Physics

Poster No.56

Femtosecond 100 W-level OPCPAs from 800 nm to 2 μm

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A review of high power (100 W-level), femtosecond OPCPAs at near IR and short-wave IR wavelengths pumped by Yb-based solid-state lasers is presented.

Femtosecond, high power and high repetition rate lasers are critical for many applications in the physical, chemical, and biological sciences. Previously, laser sources from x-ray to THz were driven from Ti:Sapphire lasers at 800 nm with limited bandwidth (Fourier limited pulse of ~20 fs), and more importantly limited power levels – power levels ~40 W and above require large complex cooling systems. Optical parametric chirped-pulse amplification (OPCPA) together with bulk crystal white-light-generation (WLG) opens up the possibility high power lasers (well above 100 W), with wavelength tunable and broadband (for example, < 10 fs at 800 nm), requiring no complex cooling with a compact design [1]. Previous thermal studies of nonlinear crystal – BBO, LBO [2] and KTA [3] – demonstrated the possibility of using these crystals for high power applications at 800 nm and 1.5 μm. Recently, compact, 100 W-level OPCPAs are now commercially available from Class 5 Photonics GmbH using BBO at 800 nm [4] and KTA with a tunable range 1.45 – 2 μm [5].

References:

[1] M.J. Prandolini, R. Riedel, M. Schulz, and F. Tavella, in [Proceedings: Proc. 36th Int. Free-Electron Laser Conf. Basel, Switzerland], **TUA02** (2014).

[2] R. Riedel, J. Rothhardt, K. Beil, B. Gronloh, A. Klenke, H. Höppner, M. Schulz, U. Teubner, C. Kränkel, J. Limpert, A. Tünnermann, M.J. Prandolini, and F. Tavella, Opt. Express **22**, 17607 (2014).

[3] K. Mecseki, H.T. Olgun, M. Windeler, M. Schulz, R. Riedel, F. Tavella, M.J. Prandolini, Proc. of SPIE **10082**, 100821C (2017).

[4] K. Mecseki; M.K.R. Windeler, A. Miahnahri, J.S. Robinson, J.M. Fraser, A.R. Fry, F. Tavella, Opt. Lett. **44**, 1257 (2019).

[5] K. Mecseki, M.K.R. Windeler, M.J. Prandolini, J.S. Robinson, J.M. Fraser, A.R. Fry, F. Tavella, Proc. of SPIE **11033**, 110330I (2019).

Poster No.57

Quantum tomography of ultrafast photochemical reactions in single molecules

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We demonstrate photoluminescence-detected ultrafast phase-only coherent control on single organic molecules in a disordered matrix at room temperature, see Fig. 1. Combined with a quantum dynamics identification procedure, we reconstruct multiphoton processes and energy landscapes for each molecule [1]. We find strong phase dependencies of the corresponding transitions into highly excited states. Importantly, also transitions into hidden states, which are not connected to photoluminescent channels, are monitored and controlled. Our combined approach provides a general toolbox to manipulate and understand ultrafast photoinduced processes in single quantum systems [1, 2], which is a prerequisite to quantum tomography of ultrafast photochemical reactions in single molecules.

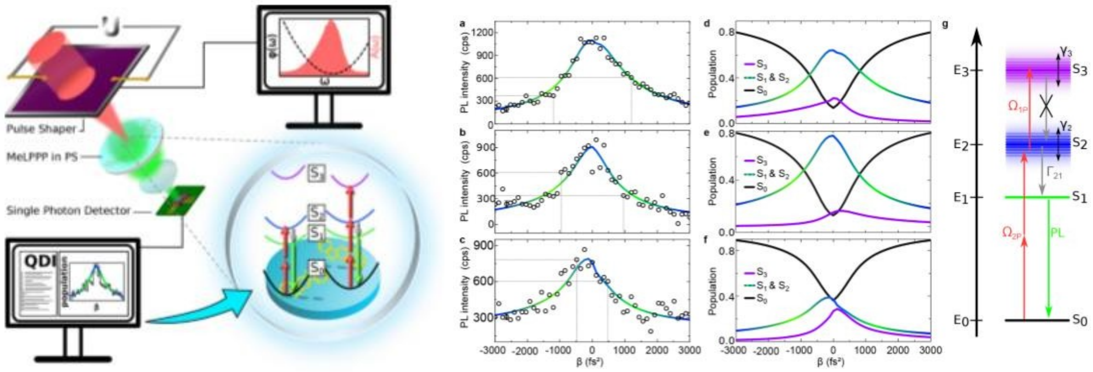

Figure 1: Left: Concept of photoluminescence-detected multiphoton coherent control and quantum dynamics identification of single organic molecules. Right: (a−c) Chirp-dependent PL signals for three different single molecules (open circles) and the theoretical QDI simulations (blue−green lines); (d−f) Simulated chirp-dependent populations of all involved electronic states. (g) Schematic illustration of the extended four-level model used for the QDI simulations.

References:

[1] K. Wilma, C.C. Shu, U. Scherf, R. Hildner, J. Am. Chem. Soc., **140**, 15329 (2018).

[2] K. Wilma, C.C. Shu, U. Scherf, R. Hildner, New J. Phys., **21**, 045001 (2019).

Poster No.58

Spatiotemporal observation of electron dynamics in femtosecond laser manufacturing

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Femtosecond pulse durations are shorter than many physical/chemical characteristic times, which permits manipulating, adjusting, or interfering with electron dynamics, thereby further improving femtosecond laser fabrication. The controlling localized transient electron dynamics poses a challenge for measuring at the electron level during fabrication processes. Unlike the traditional long-pulse laser processing, femtosecond laser essentially changes the mechanism of laser- material interactions, which is a non-linear, non-equilibrium, and complicated physical-chemical process. So far, many fundamental questions remain concerning the physical origin of the material removal process under the extreme non-equilibrium conditions imposed by femtosecond laser irradiation, affecting the quality, accuracy, efficiency, and controllability of femtosecond laser processing and limiting the further extension of femtosecond laser micro/nano fabrication techniques and applications. To acquire the plasma dynamics, several diagnostic techniques have been developed, among which time-resolved shadowgraphy pump-probe technique with femtosecond temporal resolution and micrometer spatial resolution has been widely employed. we mainly focused on (I) the evolution of laser-induced plasma and shockwave expansion controlled by femtosecond laser pulse train. The experimental results reveal that during the second pulse irradiation on the crater induced by the first pulse, the expansion of plasma and shockwave are enhanced in the longitudinal direction (see Fig.1). This can be attributed to the crater with concave lens-like morphology can tremendously reflect and refocus the latter part of laser pulse [1,2]. (II) structure-assisted filament splitting during ultrafast multiple pulse laser ablation. This is attributed to the emergence of transient high-reflective surface induced by the first pulse. The electron density analysis showed that the total deposition energy of double pulses case was generally higher than that of single pulse case [3,4]. The results of experiment demonstrated optimization of energy deposition efficiency by temporally shaping laser pulses, which will benefit the fabrication process.

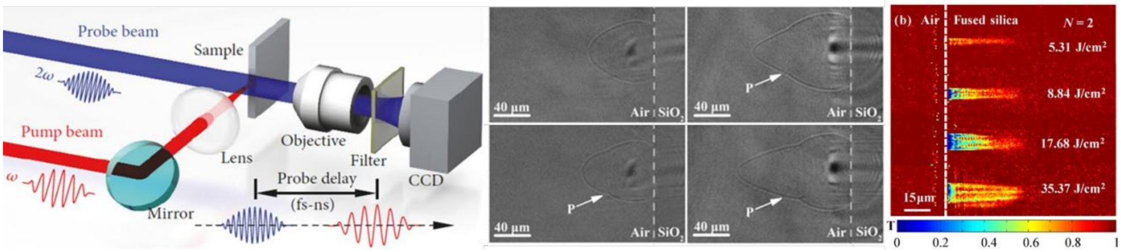

Figure 1. Schematic of the time-resolved shadowgraphy pump-probe setup

References:

[1] Qingsong Wang, Lan Jiang, Jingya Sun, Changji Pan, Weina Han, Guoyan Wang, Hao Zhang, Costas P. Grigoropoulos, Yongfeng Lu, Enhancing the expansion of a plasma shockwave by crater-induced laser refocusing in femtosecond laser ablation of fused silica,Photonics Research, 2017, **5**(5): 488-493

[2] Qingsong Wang, Lan Jiang, Jingya Sun, Changji Pan, Weina Han, GuoyanWang, Feifei Wang, Kaihu Zhang, Ming Li, Yongfeng Lu, Structure-Mediated Excitation of Air Plasma and Silicon Plasma Expansion in Femtosecond Laser Pulses Ablation. Research, 2018, 5709748

[3] Changji Pan, Lan Jiang, Jingya Sun, Qingsong Wang, Feifei Wang, and Yongfeng Lu，The temporal-spatial evolution of electron dynamics induced by femtosecond double pulses, Japanese Journal of Applied Physics **58**, (2019)030901

[4] Feiei Wang, Changji Pan, Jingya Sun, Qingsong Wang, Yongfeng Lu, Lan Jiang, Direct observation of structure-assisted filament splitting during ultrafast multiplepulse laser ablation. Optics Express, 2019, Vol **27**, 100050

Poster No.59

Excitation-dependent emission-color switch from individual Mn-doped perovskite microcrystals

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Mn^{2+} doped $CsPbCl_3$ perovskites have attracted many researchers' attention due to their dual-color photoluminescence (PL) from both exciton and Mn^{2+} dopant [1- 3]. In this work, unlike most previous reports focusing on the development of Mn- doped perovskite nanocrystals, we for the time reported the synthesis of Mn-doped $CsPbCl_3$ microcrystals (MCs) with a doping ratio of ~3 atom%. The dopant PL intensity in relative to the total emitting photons (I_{Mn}/I_{total}) can be tuned from ~0.9 to ~0.1 by changing the repetition rate or pulse intensity of excitation laser, leading to the reversible switch of emission color from orange/red (Mn^{2+}) to blue (exciton) from an individual MC. A trap-mediated energy transfer mechanism (exciton-to- trap-to-Mn) is proposed to explain the saturation of Mn emission which enables the tunability of PL emission of individual Mn-doped $CsPbCl_3$ MCs.

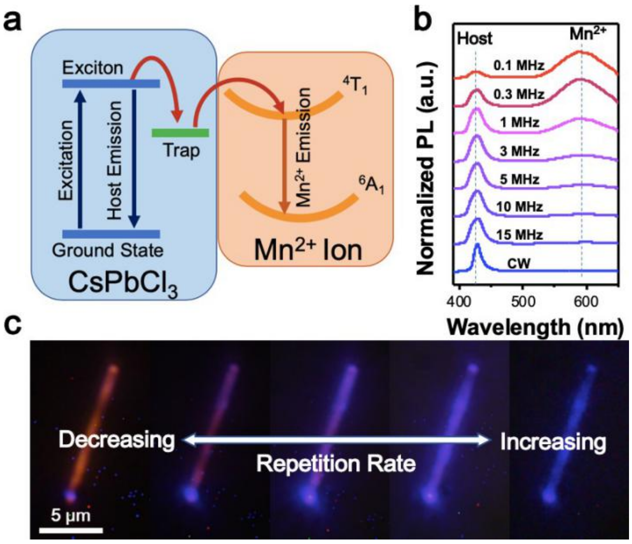

Figure 1. (a) Schematic diagram of the trap-mediated energy transfer mechanism from $CsPbCl_3$ host to Mn^{2+} dopant. PL spectra (b) and the corresponding PL images (c) showing the evolution of the emission color of an individual Mn-doped MC from orange/red to violet and to blue with increasing of repetition rate.

References:

[1] D. Chen, G. Fang, and X. Chen, ACS Appl. Mat. Interfaces **9**, 40477 (2017).

[2] H. Liu, Z. Wu, and J. Shao, ACS Nano **11**, 2239 (2017).

[3] T. He, J. Li, and C. Ren, Appl. Phys. Lett **111**, 211105 (2017).

Poster No.60

Mid-infrared supercontinuum using Ge-Sb-S fiber with high laser damage threshold

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Chalcogenide glass fibers have excellent mid-infrared transmittance and high nonlinearity. In recent years, the generation of mid-infrared supercontinuum (SC) using chalcogenide glass fibers as non-linear media has attracted much attention [1-3]. At present, the laser damage threshold of chalcogenide glass fibers is generally quite low, which is the key problem to limit the acquisition of mid- infrared SC with high average power and ultra-wideband. The simulation results show that chalcogenide glass fiber pumped by high repetition frequency femtosecond laser is an ideal way to realize high average power ultra-wideband mid-infrared SC. Compared with Se-based and Te-based chalcogenide glasses, S-based chalcogenide glasses have higher glass transition temperature (Tg) and larger average chemical bond strength. Therefore, sulfide glasses exhibit higher laser damage threshold and better mechanical properties. In addition, the optical band gap of sulfide glass is relatively large and the zero group velocity dispersion wavelength of the material is relatively short. These characteristics are conducive to adjusting the zero-dispersion wavelength of the optical fiber to short-wave direction, which makes it easier to pump the optical fiber in the anomalous dispersion region and greatly broadening the pulse spectrum. In order to explore chalcogenide glass fibers suitable for producing high brightness and broadband mid-infrared SC, the third-order nonlinearity, laser damage characteristics and mid-infrared optical properties of Ge-Sb-S glass were investigated. The small- core diameter step-index fibers were fabricated and the supercontinuum generation in the fibers was studied.

References:

[1] C. R. Petersen, U. Møller, I. Kubat, B. Zhou, S. Dupont, J. Ramsay, T. Benson, S. Sujecki, N. Abdel-Moneim, Z. Tang, D. Furniss, A. Seddon and O. Bang, Nature Photonics **8**, 830 (2014).

[2] Y. Yu, X. Gai, P. Ma, K. Vu, Z. Yang, R. Wang, D. Y. Choi, S. Madden and B. Luther-Davies, Optics Letters **41**, 958 (2016).

[3] B. Zhang, Y. Yu, C. Zhai, S. Qi, Y. Wang, A. Yang, X. Gai, R. Wang, Z. Yang and B. Luther-Davies, Journal of the American Ceramic Society **99**, 2565 (2016).

Poster No.61

Tunneling site resolved double ionization in strong laser fields

J. Tong[1, *], X. Gong [1], W. Jiang[1], W. Zhang[1], J. Ma[1], H-X. Li[1], F. Sun[1], J. Qiang[1], F. Chen[1], P. Lu[1], H. Li[1], and J. Wu[1]

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In strong-field light-matter interactions, the strong laser field and the Coulomb force [1, 2] dominate the photoelectron dynamics, presenting a modified tunneling site and a controversial ionization velocity, especially on the polyatomic Coulomb potential [3, 4]. To examine the tunneling site effect, we conceive a distinctive two- steps and trajectory resolved double ionization measurement in ArKr by completely recording the three-dimensional momenta of ejected electrons and ions coincidently, which allows us to unambiguously identify the second electron as illustrated in Fig.1. We find a sensitive dependence of Coulomb effect on the initial transverse momentum distribution of the second released electron.

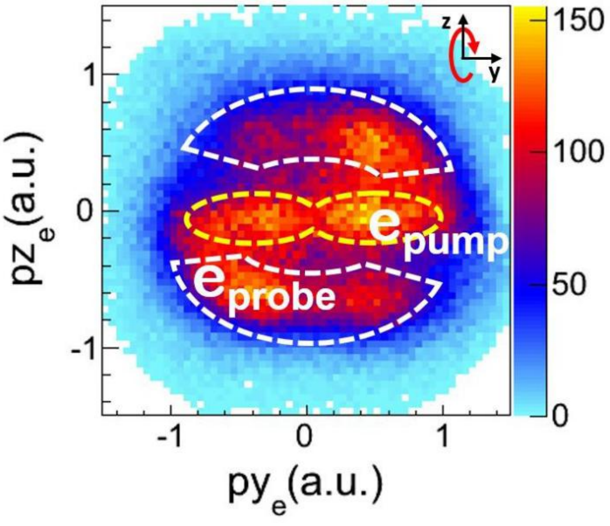

Figure 1. Trajectory resolved Photoelectron momentum distribution.

References:

[1] K. Liu and I. Barth, Phys. Rev. Lett. **119**, 243204 (2017).

[2] D. Shafir *et al.*, Phys. Rev. Lett. **111**, 023005 (2013).

[3] X.Gong *et al.*, Phys. Rev. Lett. **112**, 243001 (2014).

[4] J. Wu *et al.*, Nat. Commu. **3**, 1113 (2012).

Poster No.62

Optical resonance shifts in two-dimensional cold atomic ensembles with positional correlations

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Engineering light-matter interactions in cold atomic ensembles is one of the central topics in modern quantum and atomic optics. The collective response of dense atomic gases under light excitation, which crucially depends on the spatial distribution of atoms and the geometry of the ensemble, has important impacts on quantum technologies like quantum sensors, atomic clocks and quantum information storage. Here we analyze near-resonant light transmission in two- dimensional, ultracold and dense atomic ensembles with positional correlations. Based on the coupled-dipole simulations under different atom number densities and correlation lengths, we show that the collective effects are strongly influenced by those positional correlations, manifested as significant shifts and broadening or narrowing of transmission resonance lines. Since mean-field theories like Lorentz-Lorenz relation are not capable of describing such collective effects, we develop a theory that takes the complicated interplay between light- induced dipole-dipole interactions and positional correlations into account, which exhibits a good agreement with exact simulations. This work may provide profound implications on collective and cooperative effects in cold atomic ensembles as well as the study of mesoscopic physics concerning light transport in strongly scattering disorder media.

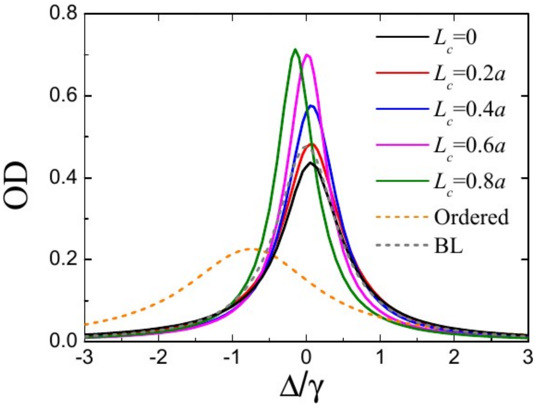

Figure 1. Optical depth (OD) of 2D atomic gas with different levels of correlation lengths.

References:

[1] S. D. Jenkins, J. Ruostekoski, J. Javanainen, R. Bourgain, S. Jennewein, Y. R. P. Sortais, and A. Browaeys. Phys. Rev. Lett. **116**, 183601 (2016).

[2] T. Peyrot, Y. R. P. Sortais, A. Browaeys, A. Sargsyan, D. Sarkisyan, J. Keaveney, I.G. Hughes, and C. S. Adams, Phys. Rev. Lett. **120**, 243401 (2018).

Poster No.63

The ultrafast spin-flip dynamics of transition metal complexes triggered by X-ray light

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The rapid development of X-ray sources and advances in attosecond spectroscopy have provided powerful tools to study more and more intricate effects on the atomic scale, such as, the correlation between valence and core electrons [1]. Most electronic spin dynamics are considered for excitation of valence electrons, the processes are usually driven by nuclear motions [2,3]. However, excitation of core electrons will trigger spin-flip dynamics in much shorter timescale, which is due to the strong spin-orbit coupling [4]. Here we investigate the ultrafast spin dynamics of transition metal compounds to understand the behavior and mechanism of ultrafast spin-flip dynamics driven by the inner electron ionization or excitation. Such processes may involve up to thousand coupled electronic states at the same time. We develop a method named density matrix based time-dependent restricted active space configuration interaction to treat many-electron problems, which well describe the electron correlation and spin-orbit coupling. The ultrafast laser field can be used to excite a core electron to prepare a complex superposition of quantum states and then steer the ultrafast spin dynamics. Interestingly, modest variation of carrier frequency, amplitude and pulse duration can lead to substantial changes in the spin-state composition, suggesting its control by soft X-ray light. The theoretical simulation will provide guidance to ultrafast attosecond experiments and we expect the experimental verification of the ultrafast elelctronic spin-flip dynamics.

References:

[1] G. Sansone, T. Pfeifer, K. Simeonidis, and A. I. Kuleff, Chem. Phys. Phys. Chem. **13**, 661-680 (2012).

[2] W. Zhang, K. S. Kjær, R. A. Mori, U. Bergmann, M. Chollet, L. A. Fredin, R. G. Hadt, R. W. Hartsock, T. Harlang, T. Kroll, K. Kubiček, H. T. Lemke, H. W. Liang, Y. Liu, M. M. Nielsen, P. Persson, J. S. Robinson, E. I. Solomon, Z. Sun, D. Sokaras, T. B. Driel, T. Weng, D. Zhu, K. Wärnmark, V. Sundström and K. J. Gaffney, Chem. Sci. **8**, 515-523 (2017).

[3] G. Auböck, M. Chergui, Nat. Chem. **7**, 629 (2015).

[4] H. Wang, S. I. Bokarev, S. G. Aziz and O. Kuhn, Phys. Rev. Lett. **118**, 023001 (2017).

Poster No.64

Unveiling the stimulated robust carrier lifetime of surface-bound excitons in InSe

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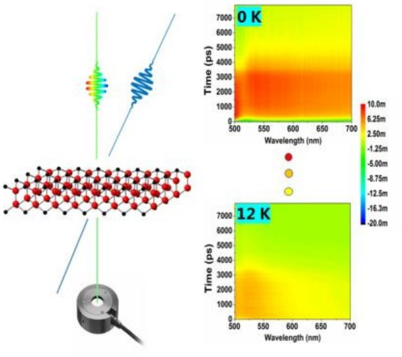

Figure 1. Schematic illustration of the measurements of pump-probe and temporally and spectrally resolved transient absorption signals of 0k -12k InSe.

References:

[1] A.G. W. Mudd, B. S. A. Svatek, and C. A. I. Dmitriev, A. Advanced Materials **25**, 5714 (2013).

[2] A. W. Feng, B. W. Zheng, and C. P. A. Hu, A. Advanced Materials **26**, 6587 (2014).

[3] A. S. D. Lei, B. L. H. Ge, and C. P. M. Ajayan, A. Acs Nano **8**, 1263 (2014).

[4] A. D. A. Bandurin, B. A. V. Tyurnina, and C. Y. Cao, A. Nature Nanotechnology **12**, 223 (2017).

Poster No.65

Improving super-resolution nanoscopy by ultrafast coherent control

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For wide field super-resolution nanoscopy, the most fundamental requirement is to observe the personalized fluorescent emission of each individual emitter, which closely locate beyond the optical diffraction limited. Some single-molecule localization microscopy (SMLM) methods, are based on the extraction of stochastic optics or spontaneous blinking fluctuations to achieve super-resolution [1,2]. However, there is not an active way to effectively manipulate individual the behavior of emitters fluorescence. This contribution introduces an active control strategy based on the principle of excited state coherent control [3], femtosecond pulse shaping, multiphoton microscopy and periodic modulation. This proof-of-principle study has pushed the resolution of the wide field nanoscopic imaging towards the range of diffraction unlimited technique. It can resolve twin emitters with 50~70nm apart and continuous linear structure with localization accuracy of ~30nm, which both for simulation and experiments with ZnCdS quantum dots (QD) films and QD625 labelled COS7 cells. Besides, since it is purely optical and modifies only the temporal properties of the femtosecond pump laser, it is also feasible to incorporate into other (super-resolution) microscope systems.

References:

[1] Rust, M. J., Bates, M. & Zhuang, X. Nature Methods **3**, 793-796 (2006)

[2] Dertinger, T., Colyer, R., Iyer, G., Weiss, S. & Enderlein, J. Proceedings of the National Academy of Sciences of the United States of America **106**, 22287-22292 (2009).

[3] Brif, C., Chakrabarti, R. & Rabitz, H. New Journal of Physics **12**, 075008 (2010).

Poster No.66

N^{2+} lasing by bicircular two-color fields

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Air lasing emission by launching intense ultrashort laser pulses in atmosphere has been intensively investigated for its promising applications. However, the physical mechanism of N_2^+ lasing is under hot debate [1-3], including the proposed electron recollision induced excitation scenario [3,4]. Here, we perform a straightforward experiment to examine the role of the electron recollision on the lasing emission from singly ionized N_2 driven by bicircular two-color (BTC) laser fields. No noticeable enhancement of the lasing emission was observed when switching the BTC laser fields from co-rotating to counter-rotating. It indicates a minor contribution of electron recollision on the N_2^+ lasing generation. Interestingly, the polarization of the newborn lasing closely follows that of the seed, providing a route for remote creation of the circularly polarized air lasing.

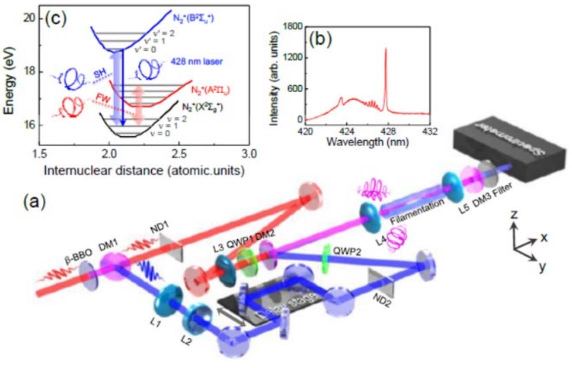

Figure 1. (a) Schematic diagram of the experimental setup. (b) Spectrum of the forward lasing emission peaked at 428 nm. (c) Energy-level diagram of ionized nitrogen molecules in which the transition between $B^2\Sigma_u^+$ and $X2\Sigma_g^+$ states corresponding to the 428 nm wavelength is indicated.

References:

[1] J. Yao, S. Jiang, and W. Chu et al, Phys. Rev. Lett. **116**, 143007 (2016).

[2] H. Xu, E. Lötstedt, and A. Iwasaki et al, Nat. Commun. **6**, 8347 (2015).

[3] Y. Liu, P. Ding, and G. Lambert et al, Phys. Rev. Lett. **115**, 133203 (2015).

[4] M. Britton, P. Laferrière, and D. H. Ko et al, Phys. Rev. Lett. **120**, 13320 (2018).

Poster No.67

Non-dipole effects in strong-field double ionization of atom

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Electric dipole approximation is generally adopted theoretically to understand the strong-field ionization dynamics. However, it was recently demonstrated that the photoelectron can acquire a non-zero momentum opposite to the laser propagation direction [1-2]. The previous measurement performed in the velocity map imaging (VMI) apparatus without coincidence is hard to identify the photoelectrons from the double ionization and thus the non-dipole effects induced momentum deflects of the photoelectrons is yet lacking of experimental observation. By precisely determining the absolute zero momentum using Rydberg atoms, we find that the sum momentum shift of the two photoelectrons produced in double ionization even reach 0.074 a.u.. The amount of shift of the two-photoelectron sum-momentum depends on detailed rescattering scenarios, which register in distinct areas of their correlated photoelectron-photoelectron momentum distribution as revealed by our classical-trajectory numerical simulations.

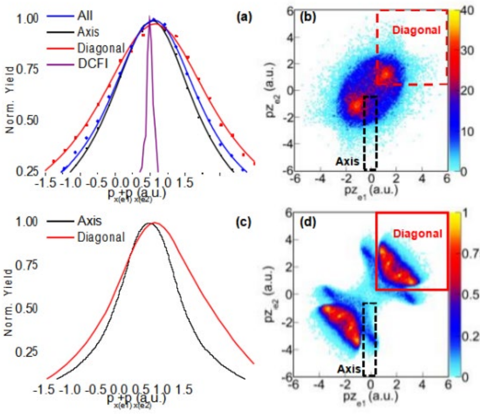

Fig. 1. (a) Experimentally measured projections of all electrons (blue curve) and different parts (black curve and red curve) of EMDs onto the beam propagation. (b) Experimentally measured correlated EMDs of Ar at 2.0 μm and an intensity of 1.4 × 10^{14} W/cm^2. (c) Simulated projections of different parts of EMDs onto the beam propagation (d) Simulated EMDs of Ar at 2.0 μm and an intensity of 2.0 × 10^{14} W/cm^2.

References:

[1] A. Ludwig et al 2014 Phys. Rev. Lett. **113**, 243001

[2] J. Daněk et al 2018 J. Phys. B: At. Mol. Opt. Phys. **51**, 114001

Poster No.68

Attosecond time resolved photoionization in Argon atom

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The resonant population of the intermediate Rydberg states leads to significant enhancement of photoelectron emissions, i.e., the Freeman resonance ionization as it is firstly observed about 30 years ago [1]. By using a phase- controlled orthogonal two-color (OTC) femtosecond laser pulse, we proposed a novel ultrafast photoelectron emission timing approach. A Freeman Resonance time delay of 140±40 attoseconds between photoelectrons emitted via the 4f and 5p Rydberg states of argon is observed in strong laser field ionization [2].

The OTC field spatiotemporally steers the emission dynamics of the photoelectrons and meanwhile allows us to unambiguously distinguish the main and sideband peaks of the above-threshold ionization spectrum. As schematically illustrated in Fig. 1, the initial phase ϕ_{PP} retrieved from laser phase dependent PADs reveals ultrashort time delays in photoelectron emissions. As compared to the well-known photon transition time delay [3-5] and the recently predicted absorption time delay in two-photon ionization of Helium, here we experimentally observed a Freeman resonance time delay in multiphoton ionization of Argon atoms

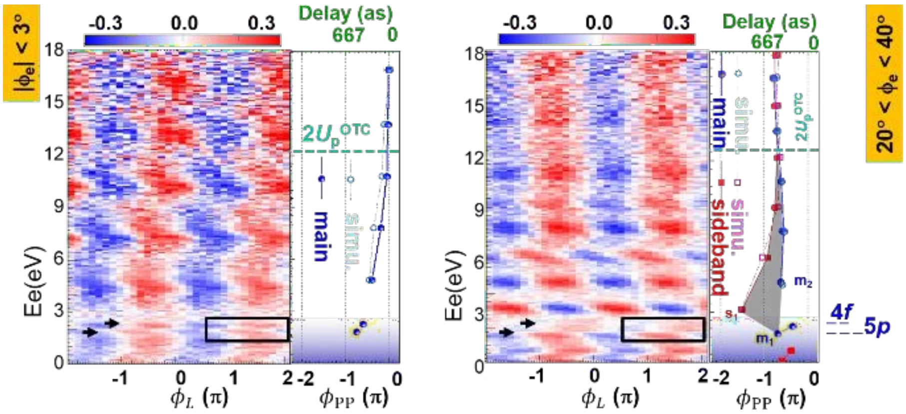

Figure 1. Freeman resonance emission time delay measured in strong laser fields.

References:

[1] R. R. Freeman et al., Phys. Rev. Lett. **59**, 1092 (1987).

[2] X. Gong et al., Phys. Rev. Lett. **118**, 143203 (2017).

[3] M. Schultze et al., Science **328**, 1658 (2010).

[4] K. Klünder et al., Phys. Rev. Lett. **106**, 143002 (2011).

[5] M. Huppert et al., Phys. Rev. Lett., **117**, 093001 (2016).

Poster No.69

Nonsequential double ionization of Ne subject to few- cycle femtosecond laser pulses

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We investigate experimentally and theoretically the evolution of Ne^{2+} ion momentum distributions from Nonsequential Double Ionization (NSDI) in few- cycle laser pulses with respect to the Carrier Envelope Phase (CEP). The distributions depend strongly on the CEP and exhibit an asymmetric double- hump structure at some CEPs. A semiclassical calculation can reproduce the experimental observations qualitatively, where the rescattering scenario is identified as the main mechanism of double ionization. Analysis based on this semiclassical model reveals that double ionization happens a little later than the collision. Depending on the time difference between the ionization moment and the collision moment, we can distinguish two ionization pathways involved, i.e., Recollision Impact Ionization (RII) and Recollision-Excitation and Subsequent Ionization (RESI) pathways. The asymmetric pattern results from both RII and RESI: RII leads to the asymmetric double-hump structure, while RESI contributes to the low momentum part and makes the dip of the double-hump structure shallower. Furthermore, Ne^{2+} ion momentum distributions from RII depend strongly on the CEP and the distributions from RESI seem to be not very sensitive to the CEP. To demonstrate the significant role of long range Coulomb potential in NSDI process, we replace the Coulomb potential with a short-range model potential in the calculation and the results show appreciable distinctions from the case of Coulomb potential. It is clarified that the Coulomb potential raises the yields of RESI process, which, in turn, makes the dip shallower.

Poster No.70

Long-range Coulomb effect in above-threshold ionization of Ne subject to few-cycle and multi-cycle laser fields

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The long-range Coulomb effect (LRCE) is demonstrated experimentally and theoretically by investigating the pulse duration dependence of low-energy structure (LES) in above-threshold ionization of Ne. It is found experimentally that at 800 nm the LES shows itself as a double-hump structure (DHS) in momentum distribution of singly charged ion for Ne, and moreover, this structure is more prominent for multicycle laser fields than for few-cycle cases. This result can be reproduced and explained qualitatively with a semiclassical model and attributed to the paramount role of LRCE. That is to say, after the laser field vanishes, the electrons decelerate while flying away from the core by the long-range tail of Coulomb potential, which eventually makes DHS less notable.

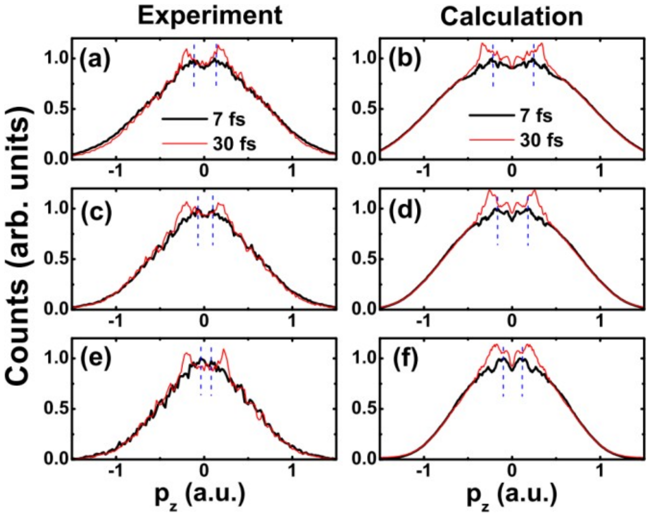

Figure 1. The measured [(a), (c), (e)] and calculated [(b), (d), (f)] longitudinal momentum distributions (LMDs) integrated over the transverse momentum of Ne$^+$ for the pulse durations of 7 fs (thick black line) and 30 fs (thin red line). The laser intensities are 5.0×10^{14} W/cm^2 for panels (a) and (b), 3.8×10^{14} W/cm^2 for panels (c) and (d), and 2.9×10^{14} W/cm^2 for panels (e) and (f).

Poster No.71

Ultrafast excitation dynamics for noble gas atoms subject to intense femtosecond laser fields

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The ionization dynamics for atoms subject to an intense laser field can be comprehended with the physical picture between two limiting cases, i.e., multiphoton ionization and tunneling ionization. A pivotal role is given to the Keldysh parameter γ to indicate the transition between these two limits, which has already been elucidated by plenty of documented works (see, e.g., [1-3]). In contrast to the atomic ionization dynamics in strong laser field, atomic excitation dynamics has not yet been well understood. The consensus on the mechanism of atomic excitation in an intense laser field hasn't been reached so far. More importantly, the formation of a complete picture of this strong field phenomenon is still hindered by the limited experimental works and, meanwhile, the rather coarse measurement precision so far.

In this Letter, we report a comprehensive investigation of the atomic excitation dynamics and successfully observe a transition from MPI to TI regime. We show a clear transition of strong field atomic excitation from the regime of multiphoton to tunneling ionization by investigating the intensity dependence of the yields of neutral excited atoms (Ar^*) and singly charged ions (Ar^+) of argon. Our results indicate that, for 400 nm, the multiphoton resonance mechanism plays a significant role at low intensity and coherent recapture mechanism becomes important at high intensity. While for 800 nm, distinctive out-of-phase oscillations of Ar^* and Ar^+ yields are identified in experiment for the first time, implying that coherent recapture mechanism dominates. Our work provides a more comprehensive understanding of the atomic excitation mechanism in an intense laser field [4].

References:

[1] B. Walker et al., Phys. Rev. A **48**, R894 (1993)

[2] E. Mevel et al., Phys. Rev. Lett. **70**, 406 (1993).

[3] J. L. Chaloupka et al., Phys. Rev. Lett. **90**, 033002 (2003).

[4] S. P. Xu et al., (submitted)

Poster No.72

Dissociation ionization of halogenated alkanes under an intense femtosecond laser fields

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In recent works, we experimentally demonstrate the photo-dissociation process of halogenated alkanes molecules in the intense femtosecond laser fields by TOF mass spectrum and dc-sliced VMI technique [1-4]. Our results show the low kinetic energy components are from the dissociative ionization process of singly charged molecular ions, and the positive charge assignment are greatly influenced by the appearance energy of the fragment ions. The high kinetic energy components result from Coulomb explosion of multi-charged molecular ions. Finally, it is inferred that how to take place under intense laser fields about the dynamic evolution process of halogenated alkanes molecules.

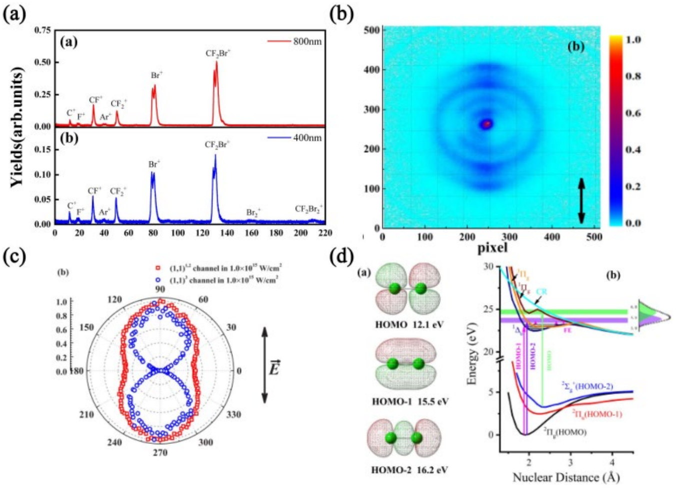

Figure 1. (a) TOF mass spectra of molecular CF_2Br_2, (b) the dc-sliced image of Cl^+ ion, (c) the relate channel ion's angular distribution, (d) the corresponding vertical ionization energy and calculated PESs of ground and several excited states

References:

[1] J. Zhang, Y. Yan, Z. Li, H. Sun, and Z. Sun. Physical Review A **98**, 4 (2018).

[2] B. Liu, Y. Yang, H. Sun, Z. Sun, Molecules **23**, 3096 (2018).

[3] J. Zhang, Y. Yang, Z.R. Sun, Chemical Physics Letters **667**, 238-243(2017).

[4] J. Zhang, S. Zhang, Y. Yang, S. Sun, Physical Review A **90**, 053428(2014).

Poster No.73

Disentangle the multielectron dissociative ionization dynamics using above-threshold ionization spectroscopies

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The multielectron effect in strong-field induced dissociative ionization of polyatomic molecule has attracted many attentions [1]. However, identifying the ionization yield ratio from different orbitals to the certain dissociative ionization channel is still challenging. We established that above-threshold ionization (ATI) electron spectroscopy can be directly used to quantitatively identify the multielectron ionization contribution in strong field-induced dissociative ionization of polyatomic molecules. The dominant role of symmetry dependent constructive and destructive interference in line profile of ATI spectrum has been approved by comparing the measured and calculated spectra for C_2H_4 and C_2H_6. For the fragment channel ($C_2H_4^+ + H_2$) of C_2H_6, we derived that the contribution from HOMO and HOMO-1 ionization is about 0.2 and 0.8, respectively, by fitting the ATI line profile. This allows us to directly present the orbital dependent behavior of debating strong field phenomenon, such as the very low energy electron and the node structure. As shown in Fig.1, dramatically orbital dependent momentum distributions in low energy electrons region have been observed, which provides new evidence to elucidate their underlying dynamics.

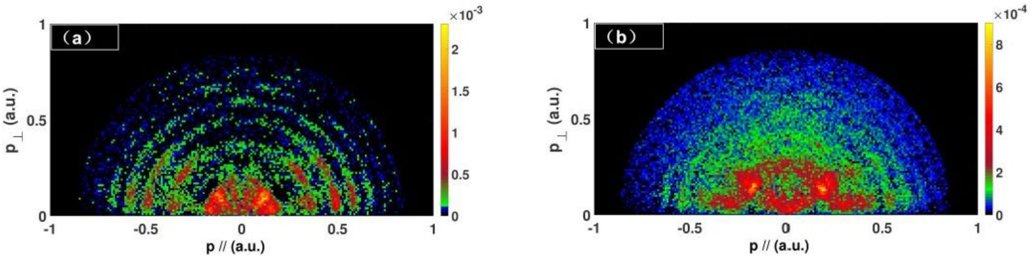

Figure 1. The electron momentum distributions of pure HOMO (a) and HOMO-1 orbitals (b).

References:

[1] C. Wang, M. Okunishi, X. Hao, Y. Ito, J. Chen, Y. Yang, R. R. Luccheses, M. Zhang, B. Yan, W. D. Li, D. Ding, K. Ueda, Phys. Rev. A. **93**, 043422 (2016).

Poster No.74

Imaging of molecular structure with high spatial and temporal resolutions

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Molecular orbital tomography (MOT) based on high-harmonic generation opens a way to track the molecular electron dynamics or even follow a chemical reaction [1]. However, it represents a formidable challenge to measure the full amplitude and phase maps as a function of the harmonic order and molecular alignment angle. In our work, we report a scheme, called diffractive MOT, to retrieve the molecular orbital solely from the amplitude of high harmonics without measuring any phase information [2]. We have applied this method to image the molecular orbitals of several different molecules in experiment. To further improve the quality of the retrieved orbital, we consider taking account of the correction of the Columbic potential [3]. Moreover, we have also experimentally demonstrated a single-shot MOT scheme which heralds the advent of the potential to get the real-time evolution of the molecular orbital [4].

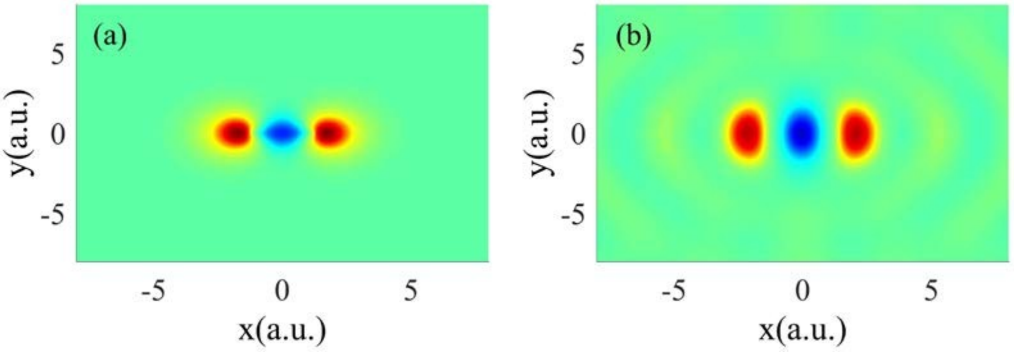

Figure 1. (a) Calculated molecular orbital of N_2 with the ab initio method. The orbital is projected to the polarization plane. (b) Reconstructed orbital from the experimental data.

References:

[1] J. Itatani, J. Levesque, D. Zeidler, et al., Nature **432**, 867 (2004).

[2] C. Zhai, X. Zhu, P. Lan, et al., Phys. Rev. A **95**, 033420 (2017).

[3] C. Zhai, L. He, P. Lan, et al., Sci. Rep. **6**, 23236 (2016).

[4] C. Zhai, X. Zhang, X. Zhu, et al., Opt. Express **26**, 2775 (2018).

Poster No.75

Optimal regulation of crystal orientation for cascaded four-wave mixing in ZnO single crystal

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Frequency converted femtosecond laser pulses through cascaded four-wave mixing (CFWM) have advantages of self-matched phase condition, broadband conversion, and relatively high efficiency. Generally, CFWM was investigated in isotropic third-order nonlinear media, such as BK7 glass [1], sapphire plate [2]. For a noncentrosymmetric medium, the mechanism for CFWM generation would be more complex. In this work, we achieve CFWM generation in two pieces of ZnO crystals with different crystal orientations. We discover that CFWM generation is extremely sensitive to the polarization of two incident femtosecond laser pulses in the case of ZnO (0001), in contrast to the case of ZnO (11-20). An explanation of cascaded second-order contribution to the third-order nonlinearities is proposed to understand the condition of CFWM generation even when the two incident pulses were perpendicularly polarized. Experiments results show that the cascaded second-order contribution should be another significant way of controlling third-order nonlinear processes.

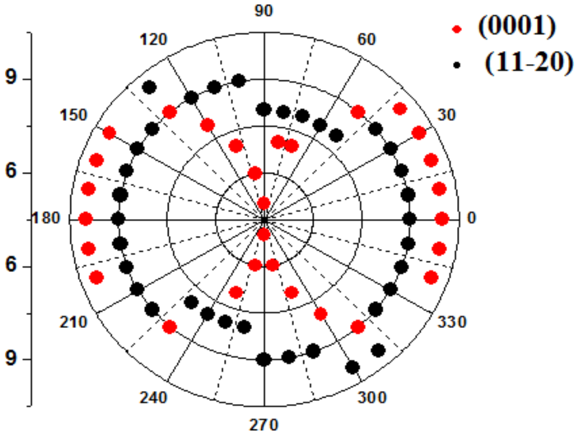

Figure 1. Polarization dependence of order numbers of CFWM in ZnO single crystals with orientations of (0001) and (11-20).

References:

[1] H. Crespo, J. T. Mendonça, and A. Dos Santos, Opt. Letters **25**, 829 (2000).

[2] J. Liu, T. Kobayashi, Opt. Express **16**, 22119 (2008).

Poster No.76

Electron-nuclear correlated multiphoton-route to Rydberg fragments of molecules

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Atoms and molecules exposed to strong laser fields can be excited to the Rydberg states with very high principal quantum numbers and large orbitals. It allows to accelerate neutral particles, generate near-threshold harmonics, and reveal multiphoton Rabi oscillations and rich photoelectron spectra. However, the physical mechanism of Rydberg state excitation in strong laser fields is yet a puzzle via the multiphoton resoant excitation [1] or frustrated tunneling ionization [2, 3, 4] scenarios. While the electron is much lighter than the nuclei, their motions are indeed strongly correlated in molecules [5]. Thereby, a complete measurement of the ejected electrons and nuclear fragments is desired to fully understand the strong-field dynamics of molecules.

Here [6], by fully measuring the freed electron, charged and neutral nuclear fragments in coincidence using a reaction microscope, we identify the electron-nuclear correlated multiphoton resonant excitation as the general mechanism in producing Rydberg fragments of a breaking H_2 molecule. By resonantly populating the repulsive Rydberg states at a smaller internuclear distance, the Rydberg fragmentation channel gains a larger nuclear kinetic energy than that of the double ionization channel for which the excess photon energy above the ionization threshold is taken by the freed electron. Due to the ac-Stark shift of the potentials, the internuclear separation for resonant Rydberg state excitation increases with the laser intensity. It alters the photon energy partition between the ejected electrons and nuclei and thus leads to distinct nuclear kinetic energy spectra of the Rydberg fragmentation channel, which explains the experimental observations driven by different laser intensities and wavelengths. The full understanding of the physical mechanism makes it possible to produce Rydberg states with desired characteristics, and thus create the coherent quantum systems for various applications. Our findings open new possibilities to manipulate the dynamics of the electrons or nuclei via one of them, and thus determine the ultimate fate of the molecules.

References:

[1] M. P. de Boer et al., Phys. Rev. Lett. **68**, 2747(1992).

[2] T. Nubbemeyer et al., Phys. Rev. Lett. **101**, 233001(2008).

[3] B. Manschwetus et al., Phys. Rev. Lett. **102**, 113002(2009).

[4] W. Zhang et al., Phys. Rev. Lett. **119**, 253202(2017).

[5] W. Zhang et al., Phys. Rev. Lett. **117**, 103002(2016).

[6] W. Zhang et al., Nat. Commun. **10**, 757(2019).

Poster No.77

Attosecond photoelectron holography in strong field tunneling ionization

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Watching the valence electron move in molecules on its intrinsic timescale has been one of the central goals of attosecond science and it requires measurements with subatomic spatial and attosecond temporal resolutions. The time-resolved photoelectron holography in strong-field tunneling ionization holds the promise to access this realm. In this time-resolved photoelectron holography, tunneling ionization of atoms or molecules by a strong laser field generates an electron wavepacket, and this electron wavepacket could reach the detector directly or undergoes rescattering by the parent ion, giving rising to the interference in the photoelectron momentum distribution (PEMD), referred as photoelectron holography in strong field tunneling ionization. We will show how the structure information was encoded in the hologram and proposed a procedure to extract the important structure information, the phase of the elastic scattering amplitude [1]. We will also show how the information of valence electron motion is encoded in the hologram of the PEMD [2]. Moreover, this photoelectron holography could provide information about the tunneling ionization process itself. Adding a weak perturbation in orthogonal to the strong fundamental field, the hologram is shifted. By analyzing the response of the hologram to the perturbation, the real part of the tunneling ionization time, which denotes the instant when the electron exits the potential barrier, and the associated rescattering time are precisely retrieved. Moreover, the imaginary part of the tunneling ionization time, which has been interpreted as a quantity related to electron motion under the potential barrier, is also precisely determined [3].

References:

[1] Y. Zhou, O. Tolstikhin and T. Morishita, Phy. Rev. Lett. **116**, 173001 (2016).

[2] M. He, Y. Li, Y. Zhou et al, Phy. Rev. Lett. **120**, 133204 (2018).

[3] J. Tan, Y. Zhou, M. He et al, Phy. Rev. Lett. **121**, 253203 (2018).

Poster No.78

Tracking a photocatalytic decarboxylation reaction over picosecond to millisecond timescales using ultrafast infrared transient absorption spectroscopy

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The photocatalytic decarboxylation of carboxylic acids is a powerful workhorse in the field of organic chemistry, enabling the synthesis of complex molecular structures via the formation of new carbon-carbon bonds [1]. We track the mechanism of such a reaction using phenanthrene (Phen, Figure 1) and 1,4-dicyanobenzene (DCB) as the starting reagents. The reaction is triggered using an ultrashort 256 nm pulse and probed using ultrashort mid-infrared pulses in a transient absorption experiment [2]. A high repetition-rate (100 kHz) mid-infrared laser allows the probing of the elementary steps from sub-picosecond to milliseconds. The mechanism includes two single electron transfer steps following photoexcitation and the unimolecular decay of a short-lived carboxyl radical. The results show that ultrafast spectroscopy using a high repetition-rate probe can be applied for a joint kinetic and mechanistic understanding of complex reaction mechanisms.

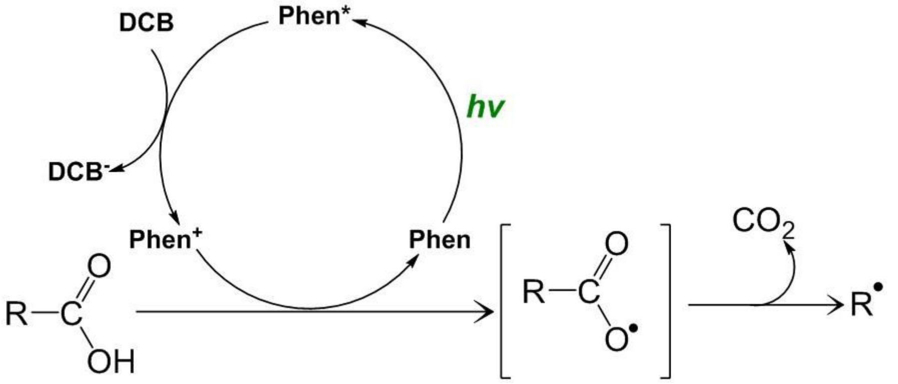

Figure 1. Elementary steps of a photocatalytic decarboxylation reaction probed by ultrafast infrared transient absorption spectroscopy.

References:

[1] J. Xuan, Z. G. Zhang, and W. J. Xiao, Angewandte Chemie-International Edition **54**, 15632 (2015).

[2] A. Bhattacherjee, M. Sneha, L. Lewis-Borrell, O. Tao, I. P. Clark and A. J. Orr-Ewing, submitted (2019).

Poster No.79

Chemical dynamics of the reactions of tricarbon with propene – toward the formation of resonantly stabilized free radicals

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Ab initio calculations of the singlet and triplet potential energy surfaces for the $C_3(X^1\Sigma_g^+/a^3\Pi_u)$ with propene reaction have been performed at the CCSD(T)/CBS//B3LYP/6-311G(d,p) + ZPE[B3LYP/6-311G(d,p)] level for key intermediates and products. These calculations have been followed by statistical calculations of reaction rates and product branching ratios in the C_3 + C_3H_6 reaction under single-collision conditions. The results show that the reaction can produce mostly $H_2CCCCCH$ + CH_3 and $H_2CCHCCCCH$ + H, and up to 3% of various acyclic C_6H_5 (phenyl/fulvene radical) isomers + H. The product branching ratios are sensitive to the initial reaction adduct (C_6H_6) formed in the entrance channels via a slight barrier ~1 kcal/mol insertion of the C_3 radical into the C-C double bond of propene. The results allowed us to conclude that the C_3 + C_3H_6 reaction result in major amounts in the direct growth of the carbon-skeleton to six-carbon C_6H_5 products via the C_3-for-H exchange because C-H bond cleavages in C_6H_6 radicals are generally more preferable than C-C bond cleavages.

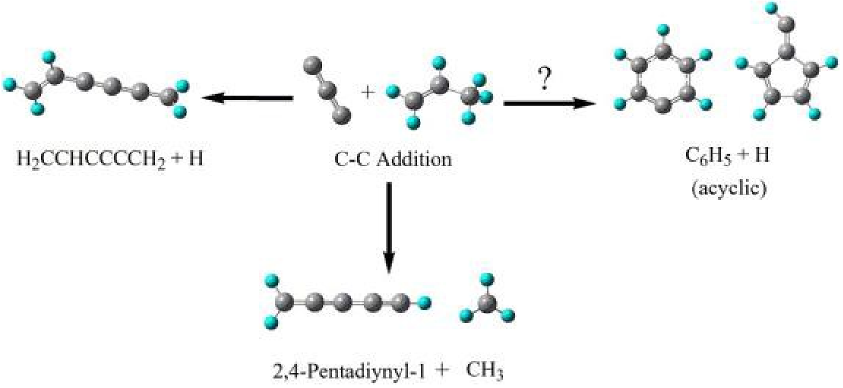

Figure 1. Distinct C_6H_5 isomers formed via H loss in the reactions of C_3 with propene.

References:

[1] A. M. Mebel, G.-S. Kim, V. V. Kislov, R. I. Kaiser, J. Phys. Chem. A **111**, 6704 (2007).

Poster No.80

Hydrated excess protons in acetonitrile/water mixtures: Solvation species and ultrafast proton motions

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Solvation structures of excess protons in aqueous surrounding are highly relevant to electric properties and an understanding of proton transport in liquids and membranes. Despite substantial effort the prevailing molecular structures that determine the microscopic mechanisms of proton transport under thermal equilibrium conditions remain debated. By combining nonlinear two-dimensional infrared spectroscopy with ab-initio molecular dynamics simulations we investigate ultrafast proton dynamics in acetonitrile/water mixtures of varying relative concentrations. The strategy of building the water environment step-by- step provides unique benchmark of aqueous solvation geometries via a spectroscopic comparison to excess protons selectively prepared with only a few water molecules in neat acetonitrile [1,2].

We show that for all investigated mixing ratios, including neat water, protons are confined within a low barrier double-minimum potential, with large amplitude displacements driven by fluctuating electric fields [3]. The results demonstrate that protons in water are predominantly solvated within dimeric water structures, i.e., within a $H_5O_2^+$ Zundel motif. The observations are rationalized by in depth simulations of the excess proton dynamics where the proton is hosted by a varying number of water molecules, thus directly resembling the experimental conditions. The simulations reveal how the dimeric water solvation structure interacts with its closest water neighbor in an $H_7O_3^+$ unit without persistent localization of the excess proton on a single water molecule. The results suggest that a rearrangement of the hierarchical hydrogen bond structure, accompanied by translocation of the solvation geometry are key elements in the microscopic mechanism of proton translocation in aqueous solution via the von Grotthuss mechanism.

References:

[1] F. Dahms, R. Costard, E. Pines, B. P. Fingerhut, E. T. J. Nibbering, and T. Elsaesser, Angew. Chem. Int. Ed. **55**, 10600−10605 (2016).

[2] F. Dahms, B. P. Fingerhut, E. T. J. Nibbering, E. Pines, and T. Elsaesser, Science **357**, 491− 495 (2017).

[3] A. Kundu, F. Dahms, B. P. Fingerhut, E.T.J. Nibbering, E. Pines, and T. Elsaesser, J. Phys. Chem. Lett. **10**, 2287−2294 (2019).

Poster No.81

Ultrafast chemical dynamics of energetic molecules

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The release of stored chemical energy from energetic molecules can be initiated by means of different initiation events, such as laser ignition and shock waves. These initiation events can easily excite energetic molecules to various electronically excited states [1]. That is why a complete understanding of the initial steps of the chemical dynamics of energetic molecules from the excited electronic states is highly desirable. In general, conical intersection (CI), plays an important role in the initial steps of chemical dynamics following their electronic excitations. In this context, our effort is to understand the different aspects of the ultrafast unimolecular relaxation dynamics of energetic molecules through CIs. Our goal is to predict molecular level understanding of the release of stored chemical energy from energetic compounds which represents a major challenge for physical chemistry [2]. For this task, by employing ab initio multiple spawning (AIMS) simulation using the complete active space self-consistent field (CASSCF) electronic wavefunction, we have investigated the relaxation dynamics [3-5] and fragmentation dynamics [6] of different homologous energetic molecules from electronically excited states. Recently, we have shown the relevance of $\pi\sigma^*$ state in the photodissociation or fragmentation dynamics of 2H-1,2,3-triazole($C_2H_3N_3$) and furazan molecules ($C_2H_2N_2O$) [6]. We have also deposited Pyrrole and 2H-1,2,3-triazole on the well-characterized single crystal TiO_2(110) surface under ultrahigh vacuum (UHV). The adsorption of pyrrole has been investigated using temperature-programmed desorption (TPD). Our final motivation is to study surface catalyzed decomposition reactions of energetic molecules with the help of femtosecond two-pulse correlation spectroscopy. Furthermore, to mimic laser ignition conditions, we have monitored the response of energetic molecules towards the strong laser field through the high harmonic generation spectroscopy. Thus, decomposition of electronically excited energetic molecules will be discussed combining AIMS simulation, two pulse correlation spectroscopy and High Harmonic Spectroscopy (HHG).

References:

[1] R. Bernstein, Overview Recent Res. Energ. Mater. **16**, 000161 (2005).

[2] R. W. Shaw, T. B. Brill, D. L. Thompson, Adv. Ser. Phys. Chem. **16**, 532 (2005).

[3] J Ghosh, S Banerjee, A. Bhattacharya, Chemical Physics, **494,** 78-89 (2017).

[4] J Ghosh, H Gajapathy, A Konar, GM Narasimhaiah, A Bhattacharya, The Journal of Chemical Physics, **147**, 204302 (2017).

[5] J Ghosh, S Bhaumik, A Bhattacharya, Chemical Physics, **513**, 221-229 (2018).

[6] J Ghosh, H Gajapathy, A. Jayachandran, E. R. Bernstein, A Bhattacharya, The Journal of Chemical Physics, **150**, 164304-11 (2019).

Poster No.82

Femtosecond photoexcitation dynamics of atoms and dimers inside a quantum solvent

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Superfluid helium nanodroplets (He_N) represent a promising approach to study femtosecond dynamics in previously inaccessible systems. Here, we present first time-resolved investigations of single indium (In) atoms and In_2 molecules located inside He_N, obtained by combining time-resolved photoelectron/-ion spectroscopy and time-dependent helium density functional theory modelling.

In the case of In atoms, photoexcitation triggers an expansion of the He bubble within 600 fs, which is represented by a 300 meV shift of the photoelectron kinetic energy (Fig. 1a). Simulations reveal that In excited-state electronic energy is converted into He kinetic energy (pressure waves) during this process [1]. The bubble expansion is followed by an oscillation of the He bubble with a period of (28±1) ps, and ultimately leads to ejection of the dopant from the droplet after ~60 ps. In the case of In_2, photoexcitation initiates a very similar response of the He solvent (Fig. 1b). Superimposed we find a strong periodic modulation of the photoelectron signal, indicating coherent nuclear wave packet (WP) motion of In_2 with a 0.42 ps period. The slowly decaying periodic signal reappears after 150 and 300 ps, representing the half and full revivals of the WP, respectively. Appearance of these revivals demonstrates that the He-induced influence on coherent nuclear motions can be lower by a factor of 10-100 compared to conventional solvents.

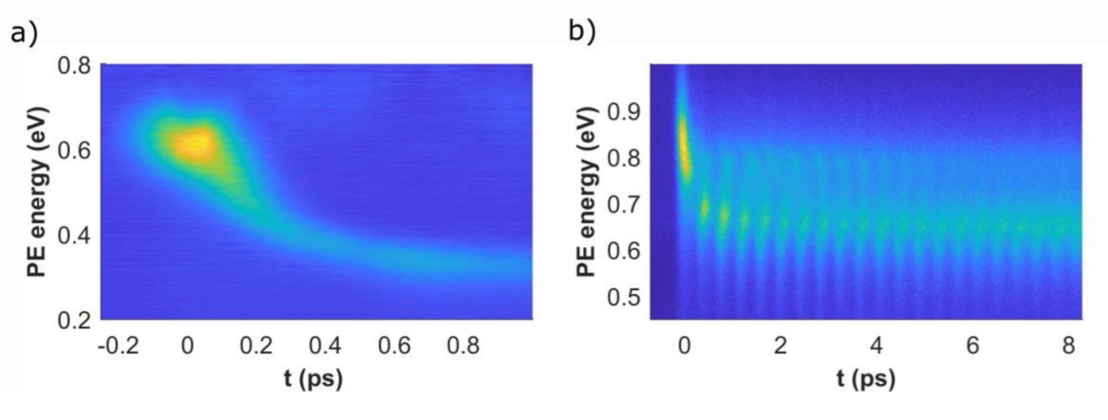

Figure 1. Transient photoelectron kinetic energy for In (a) and In_2 (b) inside He_N.

References:

[1] B. Thaler, S. Ranftl, P. Heim, S. Cesnik, L. Treiber, R. Meyer, A. W. Hauser, W. E. Ernst, and M. Koch, Nat. Commun **9**, 4006 (2018).

Poster No.83

Generation of near-UV 5-fs pulse laser to clarify charge transfer dynamics

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We have developed near ultraviolet (NUV) 5-fs pulse laser to study ultrafast dynamic process in photoreaction including transition state. Laser pulse generated by Ti: sapphire regenerative amplifier (center wavelength 800 nm, average power 2.5 W, pulse duration 35 fs) was introduced to a BBO crystal to generate intense NUV laser pulse at 400 nm. The NUV laser pulse transmitted through a hollow fiber filled with Ar gas to broaden its spectral bandwidth. Group delay dispersion of the broadband NUV pulse was compensated using a chirped-mirror pair and a diffraction grating resulting in the pulse duration of ~5 fs proved by the SD-FROG.

The NUV pulse was used to study dynamics of charge transfer (CT) in 9,9'- bianthryl (BA). The NUV pulse irradiation excites BA to produce locally excited (LE) state. The LE state relaxes to the electronic ground (EG) state in non-polar solvent. However, in polar solvent, the LE state generated by photo-excitation relaxes to CT state by intra-molecular CT, then further relaxes to the EG state.

Transient absorption of BA in non-polar solvent, hexane, was measured by pump- probe method. Spectrogram analysis of the measurement result shows that frequency of C-C stretching mode is modulated at the period of 500 fs reflecting torsion between the two anthracene rings of BA (Fig. 1a-b). Next, transient absorption of BA in polar solvent, acetonitrile, was analyzed to show that frequency of C-C stretching mode is modulated at the same period of 500 fs (Fig. 1c). Thus, BA in the electronic excited state was found to show molecular structure change of torsion between the two anthracene rings independent whether or not contribution of CT exists [1].

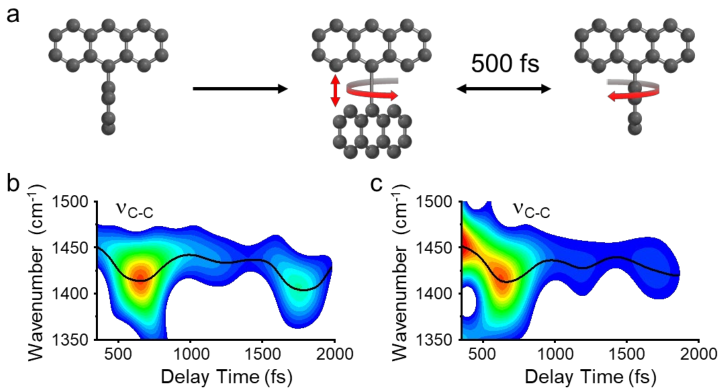

Figure 1. Molecular structure change induced by photo excitation (a). Spectrogram traces after photo excitation of BA in hexane solution (b) and in acetonitrile solution (c).

Reference:

[1] S. Hashimoto, A. Yabushita, T. Kobayashi, K. Okamura, I. Iwakura, *Chem. Phys.*, **512** 128 (2018).

Poster No.84

Delayed relaxation at complex resonances studied by wavelength-selected XUV pulses

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In the last three decades the development of the ultrafast pulsed lasers and high- order harmonic generation open access to time-dependent studies in XUV photon energy range. Excited electronic states accessible in this energy region often have a complex character combining several coupled electronic states, such as multi-state Fano resonances, and undergo ultrafast relaxation dynamics such as autoionization, dissociation or relaxation via conical intersections on nanosecond to attosecond timescales. In this contribution we consider time-resolved molecular dynamics studies carried out using wavelength-selected XUV pulses [1] in the general framework of quasi-bound states coupled to one or several continua representing relaxation pathways. Using XUV pulses we prepare i) highly excited neutral N_2 molecules [2,3] and ii) naphthalene cations [4] and follow quantum-mechanically delayed relaxation by ionization with moderately strong NIR pulses. Related time-domain effects are predicted for dynamics at conical intersections in a molecule coupled to environment [5]. To address this problem we extend our methods to photoelectron spectroscopy in solution opening route to study relaxation at conical intersections perturbed by solvents [6].

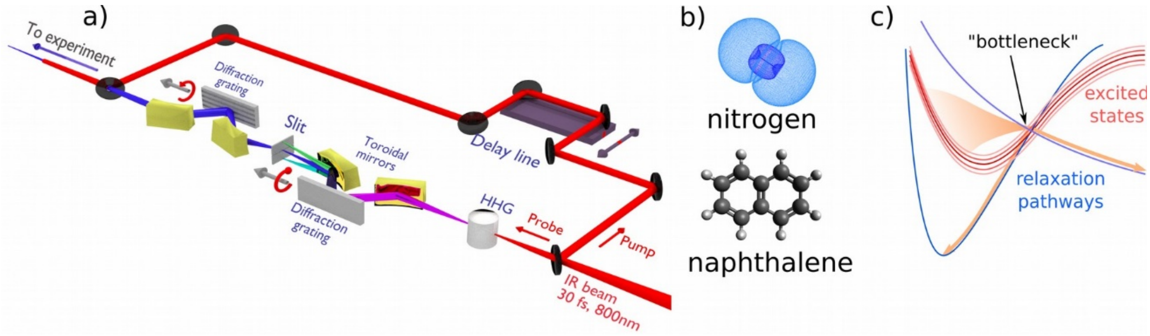

Figure 1. a) Experimental pump-probe setup with wavelength-selected XUV pulses. b) Structures of N_2 and naphthalene molecules. c) Schematic representation of the "bottleneck effect".

References:

[1] M. Eckstein et al., J. Phys. Chem. Lett. **6**, 419 (2015).

[2] M. Eckstein et al., Phys. Rev. Lett. **116**, 163003 (2016).

[3] M. Eckstein et al., Faraday Discuss. **194**, 509 (2016).

[4] G. Reitsma et al., J. Phys. Chem. A **123**, 3068 (2019).

[5] L. Chen et al., Faraday Discuss. **194**, 61 (2016).

[6] J. Hummert et al., J. Phys. Chem. Lett. **9**, 6649 (2018).

Poster No.85

UV 10-fs spectroscopy elucidating the α substitution effect on photodeprotection of *o*-nitrobenzyl acetate

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o-Nitrobenzyl derivatives are widely used as model compounds to study photodeprotection mechanism of caged compounds. Introduction of two methoxy groups to an *o*-nitrobenzyl ring induces redshift of π-π^* absorption. This is expected to provide useful applications in material chemistry, however its low quantum yield of deprotection (ϕ=0.01) still prevents a real application. To clarify the mechanism of low quantum yield inevitably related to ultrafast relaxation of the excited state, a UV 10-fs laser (350~450 nm) [1] was used to investigate the dynamics of 4,5-dimethoxy-2-nitrobenzyl acetate (compounds **1**-**4**) in acetonitrile solution [2].

From the exponential fitting analysis of the observed ΔA trace (Fig. 1), it was found that π-π^* excitation of compounds **1-3** is followed by transition from S*(π-π^*) to S*(n-π^*) in ~500 fs, and the S*(n-π^*) decays in ~1000 fs. On the other hand, S*(n-π^*) of compound 4 has relatively long lifetime of ~1500 fs, which is explained as follows. Via S*(n-π^*), electron density of lone pair of O atom in NO_2 decreases to proceed α-hydrogen abstraction via six-membered transition state. The α substituent enlarged as large as tertiary butyl group inhibits the formation of six-membered transition state, resulting in the elongation of the lifetime of S*(n-π^*).

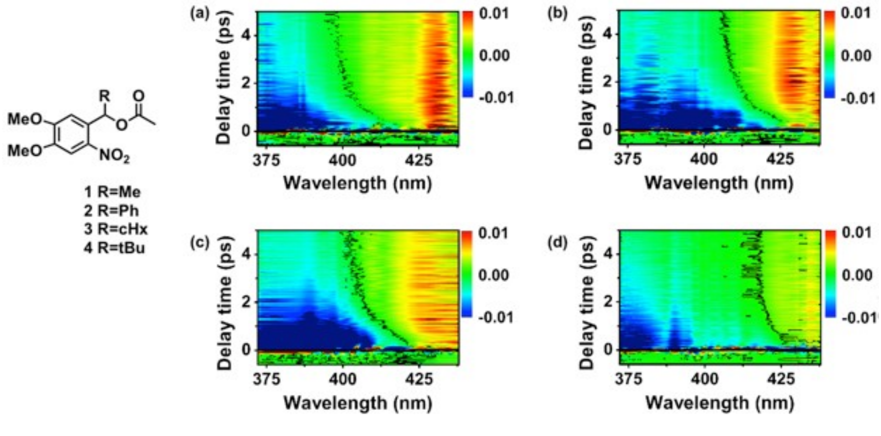

*Figure 1. Two-dimensional map of ΔA in the delay time range from -600 to 5000 fs probed in 370-435nm spectral region. (a) Compound **1**, (b) compound **2**, (c) compound **3**, (d) compound **4**.*

References:

[1] S. Hashimoto, et al., Chem. Phys. **512** 128 (2018).

[2] S.Hashimoto, et al., Chem. Phys. https://doi.org/ 10.1016/j.chemphys.2019.04.031.

Poster No.86

Strong isotope effect for branching ratios in the vacuum ultraviolet photodissociation of carbon monoxide

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Carbon monoxide (CO) is the second most abundant molecular species in the universe just after H_2 and the main gas phase reservoir of interstellar carbon and oxygen atoms [1,2]. The photodissociation of CO is believed to be the key photochemical reaction for understanding the very large oxygen isotopic heterogeneities observed in the solar system. The isotope effect of the CO photodissociation would deeply influence the key input parameters for the self-shielding model [3]. For ^{13}CO, we systemically demonstrate that the isotope substitution would greatly change the branching ratios of the photodissociation which depend on specific quantum states of CO excited. This new isotope effect could have great impact on the current photochemical modelings.

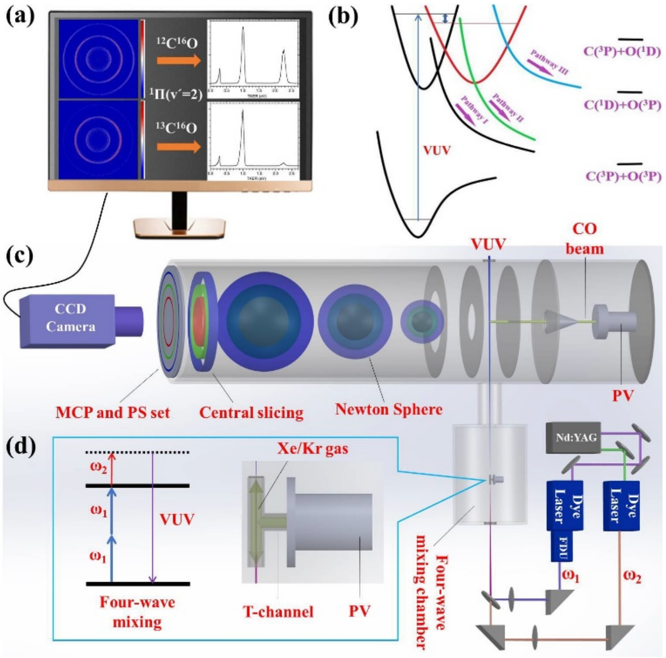

Figure 1. Schematic diagram of experimental setup and photodissociation process

References:

[1] Qing-Zhu Yin, Science, **305**, 1729 (2004).

[2] E. D. Young, Science, **317**, 211 (2007).

[3] J.R. Lyons, E.D. Young, Nature, **435**, 317 (2005).

Poster No.87

Ultrafast transient absorption investigation on the photochemical reaction mechanisms of selected aromatic carbonyl compounds

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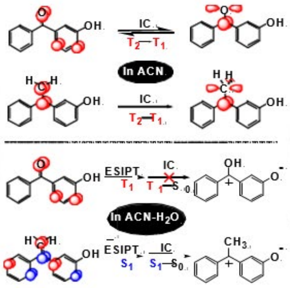
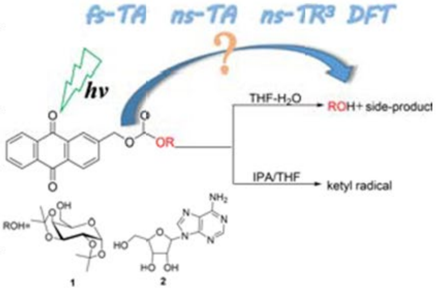
In our recent years' study, femtosecond time-resolved transient absorption spectroscopy experiments and density functional theory computations are done for the mechanistic investigation of selected aromatic carbonyl compounds, especially for some photochemical reactions in water containing solutions. We will reported some of our investigated systems, like the photoredox reaction mechanism study on benzophenone and anthraquinone compounds [1-9], the photodeprotection reaction mechanism on selected photoliable protecting groups [10-12] and the photoinduced generation of quinone methides species [13,14].





Reference:

[1] Jiani Ma, Ming-De Li, David Lee Phillips, Peter Wan. Journal of Organic Chemistry,**76**, 3710-3719(2011).

[2] Jiani Ma, Tao Su, Ming-De Li, Wei Du, Jinqing Huang, Xiangguo Guan, David Lee Phillips. Journal of the American Chemical Society.**134**, 14858-14868(2012).

[3] Jiani Ma, Tao Su, Ming-De Li, Xiting Zhang, Jinqing Huang, David Lee Phillips. Journal of Organic Chemistry.**78**, 4867-4878(2013).

[4] Jinqing Huang, Jiani Ma, Mingde Li, Mingyue Liu, Xiting Zha g, and David Lee Phillips, Journal of Organic Chemstry, **80**, 9425-9436(2015).

[5] Jiani Ma, Huai Li, Xiting Zhang, Mingde Li and David Lee Phillips. Journal of Organic Chemistry, **81**, 9553−9559(2016).

[6] Xiting Zhang, Jiani Ma, Songbo Li, Mingde Li, Xiangguo Guan, Ruixue Zhu and David Lee Phillips. Journal of Organic Chemistry, **81**, 5330−5336(2016).

[7] Xiting Zhang, Jiani Ma, David Lee Phillips. J. Phys. Chem. Lett., **7**, 4860−4864(2016).

[8] Qingqing Song, Xiting Zhang, Jiani Ma, Yan Guo and David Lee Phillips. Scientific Reports, **7**, doi:10.1038/s41598-017- 09192-3(2017).

[9] Jiani Ma, Xiting Zhang and David doi:10.1021/acs. Accounts .8b00619. Lee Phillips. Accounts of Chemical

[10] Jiani Ma, Adam C. Rea, Huiying An, Chensheng Ma, Xiangguo Guan, Ming-De Li; Tao Su, Chi Shun Yeung, Kyle T. Har is, Yue Zhu, Jameil L. Nganga, Olesya D. Fedoryak, Timothy M.Dore, David Lee Phillips. Chemistry-A European Journal.**18**, 6854-6865(2012).

[11] Jiani Ma, Jan-Michael Mewes, Kyle T. Harris, Timothy M. Dore, David Lee Phillips, and Andreas Dreuw. Physical C emistry Chemical Physics. doi:10.1039/C6CP05499D(2016).

[12] Yan Guo, Qingqing Song, Jialing Wang, Jiani Ma, Xiting Zhang and David Lee Phillips. Journal of Organic Chemistry, **83**, 13454–13462(2018).

[13] Jiani Ma, Xiting Zhang, Nikola Basarić, Peter Wand and David Lee Phillips. Physical Chemistry Chemical Physics, **17**, 9205-9211(2015).

[14] Jiani Ma, Xiting Zhang, Nikola Basarić and David Lee Phillips. Journal of American Chemical Society, **139**, 18349−18357(2017).

Poster No.88

A research on isolation and application of aflatoxin-oxidase based on magnetic protein label

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In this study, the separation of aflatoxin-oxidase (AFO) based on pigeon magnetic protein (clMagR) label [1], and the aflatoxin B1(AFB1) scavenging system were established. The AFO was co-encoded with the clMagR gene, then inserted into pET28a(+) and transformedinto *E.coli* BL21(DE3) for heterologously expression of fusion protein [2]. the purified AFO was applied to the detoxification assay of AFB1. The results showed that the magnetic protein label slightly increased the soluble protein expression of AFO [3,4]. At the same time, the magnetic protein-labeled AFO has good magnetic induction and was efficiently separated by adsorption with magnetic nanoparticles. The detoxification rate of AFB1 was 23.20 % using purified AFO, this can be reused for 8 times. This study developed a novel strategy for the separation of aflatoxin-oxidase, and its application in biological detoxification of aflatoxin [5]. This strategy provides promising utilization for further protein separation research and applications.

Key words: magnetic protein; aflatoxin-oxidase; protein separation; magnetic nanoparticles; detoxification

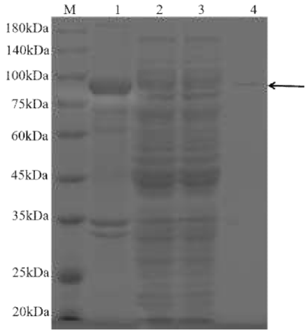

Figure 1. SDS-PAGE of the effect of magnetic beads adsorbing AFO-MagRprotein complex

References:

[1] Qin S , Yin H , Yang C , et al. Nature Materials, **15**(2):217-226(2015).

[2] Wen S, Guan M, Zhou T, et al. WQei Sheng Wu Xue Bao Acta Microbiologica Sinica, **51**(9): 1212-1221(2011).

[3] Wu Y Z, Lu F P, Jiang H L, et al. Food and Chemical Toxicology, **76**: 125-131(2015).

[4] Liu D L, Yao D S, Liang R, et al. Food and Chemical Toxicology, **36**(7): 563-574(1998).

[5] Xu T, Xie C, Yao D, et al. Biochemical and Biophysical Research Communications, **494**(3-4): 621-625(2017).

Poster No.89

Ultrafast photoionization dynamics of aromatic compounds as revealed by femtosecond stepwise two-photon excitation

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Photoionization is one of the elementary processes in photochemical reactions. Ionization proceeds when the excitation photon energy is beyond the ionization potential (Ip) of molecules. Ip in solution phase is, however, much lower than that in the gas phase and this low energy ionization was attributed to the dielectric interaction between the solvent and the solute in higher excited (S_n) state. In order to directly elucidate the dynamics relating to this dielectric interaction, we have applied femtosecond transient absorption (TA) spectroscopy under sequential two- photon excitation (Figure 1a, [1]). Figure 1b shows TA spectra of *N,N,N',N'*-tetramethyl-*p*-phenylenediamine (TMPD) in ethanol under the stepwise excitation with two pump pulses (pump1: 340 nm, pump2: 680 nm). Delay time shown at the right side of each spectrum was the time interval between pump2 and probe pulse. The pump1 was irradiated at -10 ps. Immediately after the pump2 excitation, the bleaching of S_1 state as well as the S_n state absorption are observed. The positive band due to the S_n state absorption exhibits a rapid decay with a time constant below 70 fs. On the other hand, the new absorption band due to the cation radical gradually increases around 500-600 nm with a time constant of ~5 ps. This delayed generation of the cation clearly indicates that the ionization does not directly take place from the S_n state, but through a specific intermediate state. Moreover, the solvent dependences of the temporal evolutions of the TA spectra showed that the rate of the radical cation formation is comparable to the timescale of the solvation. This result strongly suggests that solvation of the intermediate species regulates the ionization process in solution phase. At the conference site, we will discuss the dependence of the excitation energy and substituents on the photoionization dynamics.

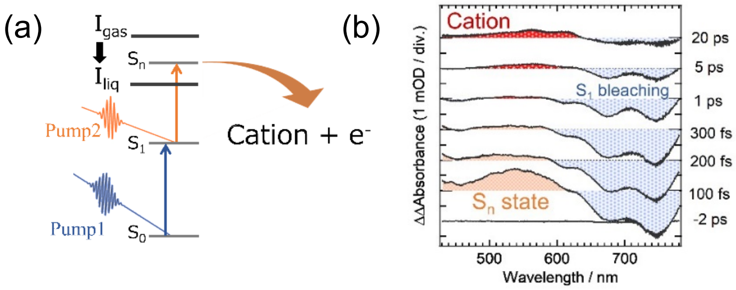

Figure 1. (a) A schematic diagram of the photoionization induced by stepwise two-photon excitation. (b) TA spectra of TMPD in ethanol excited with pump1 and pump2.

References:

[1] M. Koga, Y. Yoneda, H. Sotome, and H. Miyasaka, Phys. Chem. Chem. Phys. **21**, 2889 (2019)

Poster No.90

Ultrafast dynamics of phenol and its methyl substituted derivatives

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Phenol is key component of the chromophore in aromatic amino acids, such as tyrosine, which dominate the ultraviolet absorption spectra of many biological molecules [1]. The understanding of its photochemistry and photophysics may aid the development of simple models of such dynamics extendable to larger molecules.

We use a combination of femtosecond pump-probe experiment and velocity map imaging technology to obtain the time-resolved photoelectron spectra (TRPES) [2] for studying the excited-state nonadiabatic dynamics of phenol and methyl substitution effects in its methyl substituted derivatives.

References:

[1] M.N.R. Ashfold, B. Cronin, A. L. Devine, R. N. Dixon, and M. G. D. Nix, Science **312**, 1637 (2006)

[2] Zhigang He, Zhichao Chen, Dongxu Dai, Guorong Wu, and Xueming Yang, Chin. J. Chem. Phys **30**, 247 (2017).

Poster No.91

Intramolecular charge transfer and solvation dynamics of push-pull dyes with different π-conjugated linkers

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The solvation-dependent excited state dynamics of two push-pull fluorophores with donor-π-acceptor (D-π-A) structures were investigated using steady-state and ultrafast transient absorption (TA) spectroscopy, and backed by theoretical calculations. Identical D and A groups were present in both dyes, which differed only in the structure of their central π-conjugated linkers. Dye 1 features a p-phenylenediethynyl linker, while dye 2 contains a 2,5-diethynylthiophene linker. From the steady-state spectra, no appreciable shifts in absorption bands were observed, whereas large red-shifts in emission were seen with increasing solvent polarity, which indicated that the excited states were more polar than the ground state. Theoretical calculations support charge transfer from the triphenylamine (TPA) donor to the pentafluorosulfanyl (SF_5) acceptor via π-conjugated linkers to form an intramolecular charge transfer (ICT) state. TA spectra revealed that a solvation-stabilized conformationally relaxed intramolecular charge transfer (ICT') state was formed in polar solvents, but only an ICT state was observed in nonpolar solvent. The SE band was quenched within 1 ps in high-polarity solvent, which corresponds to the low fluorescence quantum yield. It can be concluded that dye with the p-phenylenediethynyl π-linker (i.e., dye 1) exhibits a larger degree of ICT than the thiophene analogue (i.e., dye 2). These findings demonstrate how solvation can fine-tune the photophysical properties of push- pull dyes, and this study highlights the importance of π-conjugated linkers in the excited state ICT process.

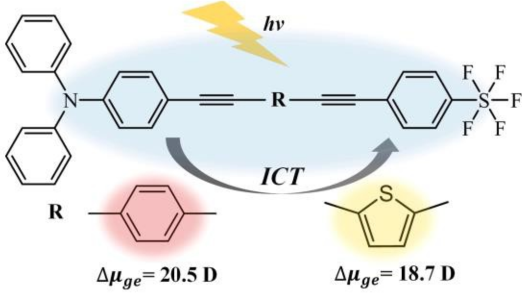

Figure 1. Molecular structures of two dyes.

Poster No.92

Probing photoinduced reaction mechanisms of sulfur-containing molecules via ultrafast X-ray spectroscopy

Miguel Ochmann[1,*], Abid Hussain[1], Jessica Harich[1], Rory Ma[1,2], Amy A. Cordones[3], Jae Hyuk Lee[4], Kiryong Hong[5], Robert W. Schoenlein[3,7], Tae Kyu Kim[2], Nils Huse[1]

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The elemental specificity of X-ray spectroscopy provides unique complimentary information to vibrational and electronic spectroscopy or non-resonant scattering techniques, when studying chemical reaction mechanisms. This can be especially useful for so-called heteroatoms in functional groups of organic molecules [1,2] which govern most of the chemistry of the respective organic molecule. Two structurally related examples of sulfur containing molecules and their photo-induced chemical reactions probed with time-resolved X-ray absorption spectroscopy are presented to highlight the prospects of time-resolved X-ray spectroscopy: (i) An aromatic thiol, which undergoes S-H bond cleavage upon UV illumination. (ii) The simplest aliphatic disulfide, which undergoes competitive C-S and S-S bond cleavage upon UV illumination. Both structural motifs play important roles in biochemistry and the presented results serve as model studies.

Preliminary data from PAL-XFEL will provide an outlook how time-resolved X-ray spectroscopy of heteroatoms can identify transient excited states and new species in sulfur-containing molecules using femtosecond and picosecond X-ray spectroscopy.

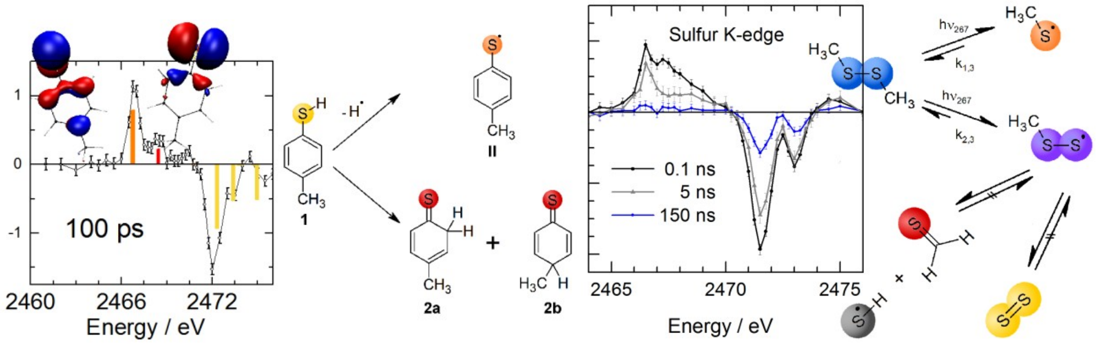

Figure 1: Left: Transient X-ray absorption spectrum at 100 ps of UV excited aroamtic thiol and scheme of the two observed reaction pathways. Right: Transient X-ray absorption spectrum at 0.1, 5 and 150 ns of UV excited dimethyl disulfide and scheme of the proposed reaction pathways.

References:

[1] M. Ochmann et al., J. Am. Chem. Soc. **139**, 4789 (2017).

[2] M. Ochmann et al., J. Am. Chem. Soc. **140**, 6554 (2018).

Poster No.93

Ultrafast excited-state relaxation of quinazolinone family

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By the noble reacting route of the trifluoroetylation of the 2-arylquinazolin-4(*3H*)-ones (**1a**), C-, O-, and N-functionalized quinazolinone subclasses were obtained: **3a**, **4a**, and **5a**, respectively (Figure 1). Because of the pharmaceutical value of the quinazolinone alkaloids, it is important for these chemicals to be photochemically inert. Here, we reveal the outstanding photostability of the quinazolin-4(*3H*)-one family. First, the steady-state measurements were performed in a series of solvents, methanol, acetonitrile, n-hexane, and chloroform. All the quinazolinone family barely emit and therefore, their quantum yields are found to be as low as $0.6{-}3.1 \times 10^{-4}$. For the in- depth investigation of the photostability, the fs-resolved fluorescence kinetic profiles are obtained by utilizing the fluorescence-upconversion technique. For **1a**, **3a**, and **5a**, the ultrafast lifetime components on the time scales of hundreds of fs to a few ps result from the nonradiative relaxation via the conical intersections from the electronic state of the amide tautomer form. The lifetime of the iminol-based/O-trifluoroethylated **4a** in nonpolar solvents is significantly elongated to several tens of picosecond. The elongation of the lifetime is attributed to the raising of the energy barrier by O-trifluoroethylation. In the polar solvents, the charge-transfer state becomes stabilized with the conical intersection being accessible. This resulted in the reduction of the lifetime to 1−10 ps.

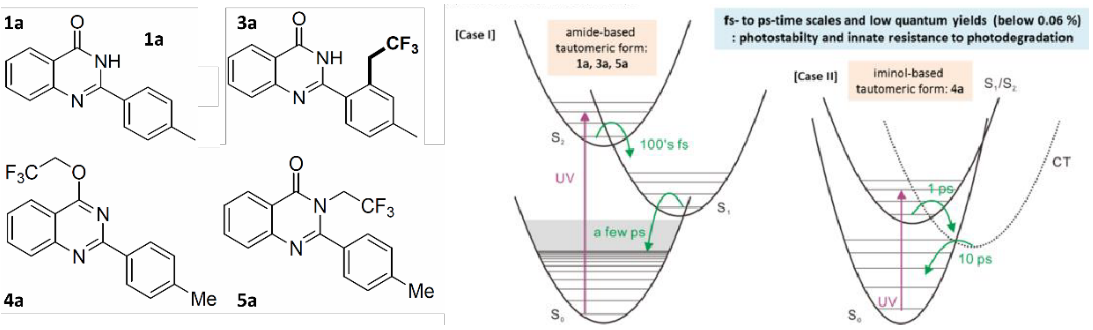

Figure 1. The molecular structure of the quinazolinone family and their photophysics with schematic energy: [Case Ⅰ] Amid-based 1a, 3a, and 5a, and [Case Ⅱ] iminol-based 4a.

References:

[1] S. Maiti, J. Kim, J.-H. Park, D. Nam, J. B. Lee, Y.-J. Kim, J.-M. Kee, J. K. Seo, K. Myung, J.-U. Rohde, W. Choe, O.-H. Kwon, and S. Y. Hong, J. Org. Chem., in press.

Poster No.94

Metal-cation enhanced deprotonation of water by photobases

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Picosecond fluorescence spectroscopy was used to study the photo-initiated proton-transfer reaction from water to a family of quinolines photobases in presence of metal cations. The ps-resolved on-contact proton transfer rate from water to the photobase follows the Kiefer-Hynes free-energy correlation which rationalizes the semi-empiric Marcus BEBO correlation for proton transfer [1, 2]. By analyzing the contact proton transfer rates we find water acidity to increase when solvating metal cations according to the following order: $Zn^{2+} > Mg^{2+} > Ca^{2+} > Li^{+} > Na^{+} > K^{+}$ which generally agrees with the Hofmeister series of cation reactivities [3].

References:

[1] D. Pines, J. Ditkovich, T. Mukra, Y. Miller, P.M. Kiefer, S. Daschakraborty, J. T. Hynes, and E. Pines, J. Phys. Chem. B **120**, 2440 (2016).

[2] N. Munitz, Y. Avital, D. Pines, E. T. J. Nibbering, and E. Pines, E. Isr. J. Chem. **49**, 261 (2009).

[3] M. G. Cacace, E.M. Landau, and J. J. Ramsden, Quart. Rev. Biophys. **30**, 241 (1997).

Poster No.95

Electronic deactivation and reaction dynamics of the potential photoactive protoncrane 7-hydroxyquinoline-8- carbaldehyde (HQC)

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A design attempt for photoreversible proton transfer switches features crane-like substituents that are capable of transferring protons from one side of the molecular scaffold to the other. Suggested by quantum chemical calculations, 7-hydroxyquinoline-8-carbaldehyde (HQC) has been proposed to exhibit such a reaction pathway between two stable tautomers (Figure 1A, [1]). In solution, those two forms are already present in the electronic ground state, with the equilibrium between them being solvent dependent. To shed light onto the ensuing dynamics, we used transient electronic absorption spectroscopy (Figure 1B) and fluorescence up-conversion spectroscopy. A global analysis of the observed transient absorption signals in acetonitrile provided one short time constant of τ_1 = 1.7ps, one intermediate constant of τ_2 = 11ps and a long-lived component of τ_3 > 1ns. The obtained time constants are discussed in terms of dynamic contributions towards electronic deactivation and reactive intramolecular proton transfer of HQC.

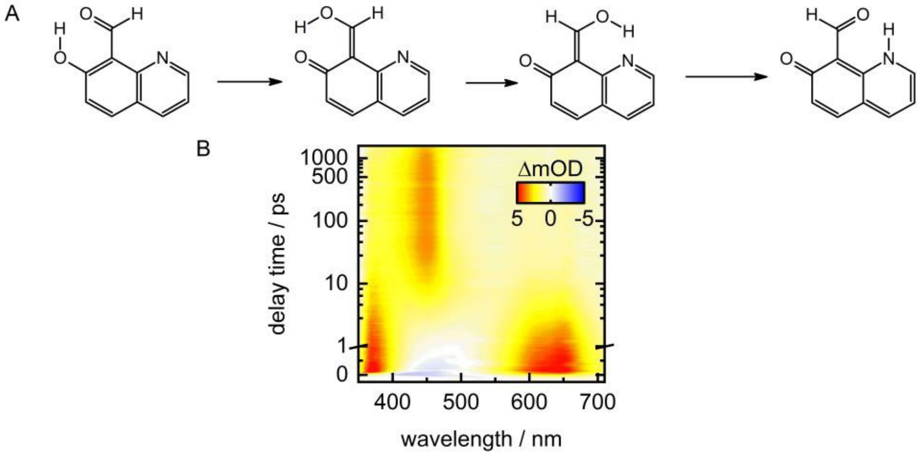

Figure 1. A) Postulated reaction scheme of HQC. B) Spectro-temporal transient absorption map of HQC in MeCN after photoexcitation at λ = 325nm.

References:

[1] V. Vetokhina, J. Nowacki, M. Pietrzak, M. F. Rode, A. L. Sobolewski, J. Waluk, J. Herbich, J. Phys. Chem. A **117**, 9127 (2013).

Poster No.96

Complete photodissociation dynamics of CF_2I_2 in solution

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Photodissociation dynamics of CF_2I_2 in cyclohexane were evaluated by probing the C–F stretching mode over a wide time range after ultraviolet excitation using femtosecond infrared spectroscopy. After the ultrafast ($<$ 0.2ps) state-selective photodissociation of CF_2I_2 as in the gas phase (267 nm excitation led to exclusive three-body dissociation (CF_2 + I + I), 350 nm to exclusive two-body dissociation (CF_2I + I), and 310 nm to a mixture of three- and two-body dissociations), various secondary reactions were observed. Once produced, some nascent CF_2 radicals immediately formed a complex with the departing I atom ($I\cdots CF_2$), which produced either CF_2I or CF_2 radicals. The produced CF_2I geminately recombined with the I atom, whereas the CF_2 radical reacted bimolecularly to produce C_2F_4 with a diffusion-limited rate constant of 8.1×10^9 M^{-1} s^{-1}. Some nascent CF_2I radicals were produced with sufficient excess energy to further dissociate into CF_2 and I, or immediately reacted with the dissociated I atom to form the I_2–CF_2 isomer that rapidly dissociated into CF_2 and I_2. Other nascent CF_2I radicals geminately recombined with the I atom with various time constants. Thus, the nascent photoproducts, CF_2 and CF_2I take various reaction paths: complex formation, secondary dissociation, isomer formation, and fast and slow germinate rebindings. The ensuing reaction path of the nascent photoproduct is dictated by its internal energy as well as solvent environment, which leads to different interactions between the photoproduct and solvent. Measurement over a broad time range with a structure-sensitive probe could reveal the fate of all the reaction intermediates, which allows evaluation of the complete reaction dynamics in solution.

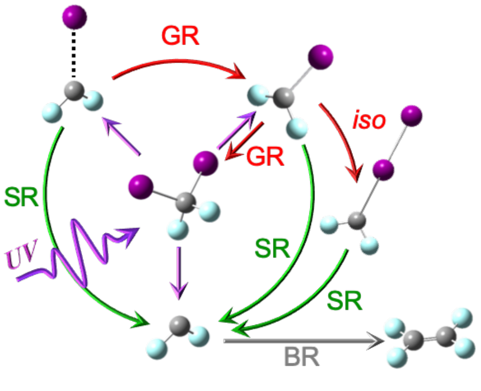

Figure 1. Photoexcited CF_2I_2 in c-C_6H_{12} undergoes various secondary reactions (SR) including complex and isomer formation, after ultrafast two- or three-body dissociations

Poster No.97

Time-resolved molecular dynamics in Rydberg states of NH_3 induced by low-order-harmonic radiation

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A source of vacuum-ultraviolet (VUV) light is a highly desirable tool for studies of molecular dynamics involving high-lying excited or Rydberg states. With such sources, one-photon excitation of these states is feasible which leads to dramatic simplification of the photo-induced dynamics. We have realized a femtosecond VUV source based on low-order harmonic generation in a semi-infinite gas cell. A 400 nm driver beam generates the third (133 nm, 9.3 eV) and fifth (80 nm, 15.5 eV) harmonics [1], which are used in an interferometer together with 266 nm femtosecond pulses to implement a two-color single-photon pump-probe scheme. The interferometer is combined with a velocity-map-imaging spectrometer to measure excited-state dynamics via energy and angular distributions of photoelectrons.

In this contribution, we present the time-resolved Rydberg-state dynamics in ammonia (NH_3), which has been extensively studied in the literature [2], but its ultrafast dynamics in high-lying (around 9.3 eV) Rydberg states, excited in the single-photon regime has not yet been addressed. After photo-excitation, two main features appear in the photoelectron spectrum with vertical binding energies of 1.7 eV and 2.9 eV, and with distinctly different anisotropy parameters of $\beta \approx 2.0 \pm 0.1$ and 0.5 ± 0.1, respectively. This information assisted the assignment of the respective Rydberg states. The two features also exhibit different dynamics characterized by decay times of 170 ± 20 fs and 260 ± 20 fs, respectively. The decay times are extracted using a global-analysis approach, revealing a population transfer between the states agreeing with a sequential kinetic model.

References:

[1] V. Svoboda et al. J. Chem. Phys., **146**, 084301 (2017).

[2] S. R. Langford et al. J. Chem. Phys., **108**, 6667 (1998).

Poster No.98

The femtosecond time scale of radiation damage: Formation and decay of solvated electrons in neutral water clusters

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The solvated electron has attracted a lot of attention for more than two centuries [1]. The solvated electron is created during radiolysis of liquid water and/or water clusters [2]. It influences the nature of several processes; in molecular biology, in radiation chemistry, and in electron-transfer processes. Therefore, it is essential to understand its role within the dynamics of the aforementioned processes [3].

In this contribution, we will present a table-top experiment using a low-order harmonic source to reveal the photo-induced ultrafast dynamics of the solvated electron in large water clusters ($\langle n \rangle \sim 400$), where the photoelectron images are detected employing a time-resolved velocity-map-imaging setup [4]. The solvated electron is studied by means of a (1 + 1') photon pump-probe scheme, where 80 or 133 nm pulses are used to create the solvated electron and a subsequent 266 nm pulse probes both its creation and solvation dynamics. The cross-correlation that defines the temporal resolution of the experiment has been determined to be less than 85 fs.

In our experiments, we observed an increase of the vertical binding energy with time together with a narrowing of the photoelectron band of the solvated electron. This is the first direct observation of the solvation dynamics in water clusters taking place on a ~200 fs time-scale following our earlier observation of their formation using a free-electron laser [2]. The photoelectron data are accompanied with a map of the anisotropy parameter $\beta_2(E, \Delta t)$ which shows a small, persistent anisotropy of the solvated electron. The results are interpreted by comparison with ab initio calculations.

References:

[1] J. M. Herbert, and M. P. Coons, Annu. Rev. Phys. Chem., **68**, 20.1 (2017).

[2] A. C. LaForge et al., PRL, **122**, 133001 (2019).

[3] A. E. Bragg et al., Science, **306**, 669 (2004).

[4] V. Svoboda et al., J. Chem. Phys., **146**, 084301 (2017).

Poster No.99

Time-resolving molecular chirality in the gas phase with high-harmonic radiation

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Chirality is an important concept in chemistry and other natural sciences. Recently, molecular chirality in isolated molecules, i.e. gas-phase, has become accessible using photoionization of a chiral molecule and subsequent angle-resolved photoelectron detection, revealing the phenomenon of photoelectron circular dichroism (PECD) [1]. PECD experiments have already evolved from synchrotron-based static measurements to laser-based sources utilizing femtosecond time-resolution [2-4].

In this contribution, we will present the first TR-PECD experiments with circularly polarized high harmonics, thereby introducing a general experimental scheme for femtochirality. Photoelectrons are ionized in a (1 + 1')-photon scheme involving linearly polarized 266 nm and circularly polarized 133 nm pulses. By employing ultrafast laser pulses, femtosecond temporal resolution is achieved.

Our work focuses on one of the simplest chiral molecules – CHFBrI and discusses its dynamics and time-dependent chirality during the photo-dissociation along the C-I bond. The data reveals a rapid change of the chirality at early time delays later leading to a coherent population of one stereoisomer only. Experimental results are supported by high-level ab initio and electron-molecule scattering calculations, providing an explanation of all observed phenomena.

The broad applicability of the time-resolved PECD measurement scheme is demonstrated by studying another chiral molecule, 2-iodobutane. Whereas CHFBrI displays a non-vanishing PECD at long pump-probe delays, the PECD decays to zero after the photo-dissociation of 2-iodobutane, reflecting the effective chirality of the product radicals on long timescales. This chirality is explained using the concept of a stereomutation time calculated via a DVR approach in a 1D potential along the respective inversion coordinate.

References:

[1] I. Powis, J. Chem. Phys., **112**, 301 (2000).

[2] Ch. Lux et al., Angew. Chem. Int. Ed., **51**, 5001 (2012).

[3] S. Beaulieu et al., Faraday Discuss., **194**, 325 (2016).

[4] S. Beaulieu et al., Science, **358**, 1288 (2017).

Poster No.100

Ultrafast dynamics of laser dye DCM in the excited state

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Intramolecular charge transfer proceeds in a laser dye, DCM（Dicyanomethylene-2-methyl-6-(p-dimethylaminostyryl)-*4H*-pyran, Fig.1a） after photo-excitation. In the present work, we have studied ultrafast dynamics of DCM in DMSO solution using ultraviolet 10-fs laser and pump-probe measurement system [1]. The absorbance of the solution was adjusted to be 1.4 in a glass cell with optical path length of 1mm.

The solvent DMSO cannot be single-photon excited by the UV 10-fs pulse with 350-450 nm band due to its absence of absorption longer than 340 nm (Fig. 1b). Time- resolved difference absorption (ΔA) of neat DMSO solvent shows the signal oscillating around ΔA = 0 (Fig. 1c), which indicates that DMSO is not excited into electronic excited state. The ΔA trace before zero delay (negative time [2]) shows a coherent artifact caused by pump-perturbed free-induction decay. On the other hand, DMSO solution of DCM absorbs light at wavelength shorter than 600 nm, which can be excited by single photon absorption of the UV 10-fs pulse laser. The measurement result of ΔA shows negative signal (Fig. 1c), which is thought to be assigned to the signal of DCM. Exponential fitting has estimated that it transfers to charge transfer state in ~30 fs.

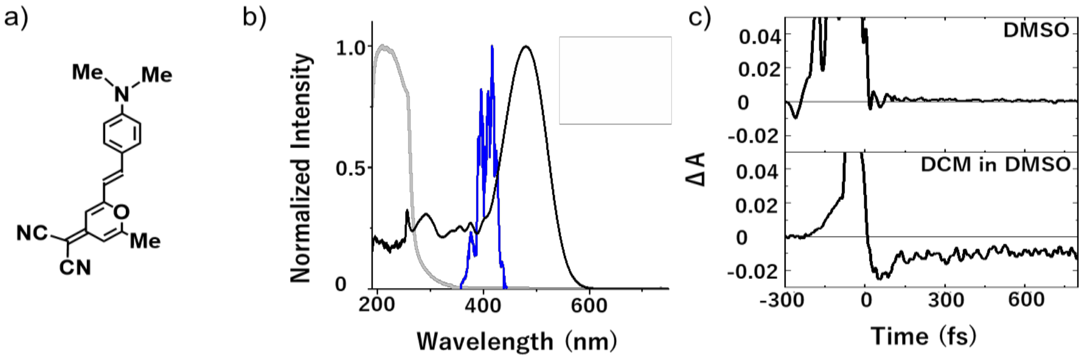

Figure 1. a) Molecular structure of DCM, b) Absorption spectrum of DMSO (gray curve), DMSO solution of DCM (black curve), and laser spectrum (blue curve). c) Transient difference absorption signal of DMSO (probed at 373 nm) and of DCM in DMSO solution (probed at 373 nm)

References:

[1] S. Hashimoto, A. Yabushita, T. Kobayashi, K. Okamura, I. Iwakura, Chem. Phys., **512** 128 (2018).

[2] J.–P. Likforrman, M. Joffre, G. Cheriaux, D. Hulin Opt. Lett. **20** 2006 (1995).

Poster No.101

H-elimination dynamics of photoexcited fluorophenols revealed with velocity map imaging

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Quantum tunneling effect refers to the quantum mechanical phenomenon where a particle, such as electron, hydrogen atom, tunnels through a barrier that it classically could not surmount. It plays a fundamental role in a myriad of chemical and biological processes, particularly in enzyme activity which could improve the reaction rate dramatically. As an ultraviolet chromophore in tyrosine, investigation on relaxation dynamics of phenol is important for understanding the active mechanism of tyrosine. Previous research has confirmed experimentally that photodissociation of phenol is correlated with the H atom on O-H bond tunneling through the barrier under S1/S2 conical intersection [1]. In this work, using femtosecond pump-probe technique, time of flight mass spectroscopy, and ion velocity map imaging, we have investigated the photodissociation dynamics of the derivatives of phenol at a series of wavelengths. With the help of quantum chemical calculations, we try to understand the role of tunneling which plays in the H-elimination dynamics. Furthermore, by introducing a third femtosecond infrared laser on the photoexcited molecules, we are able to modify the H-elimination process, and we observe the change of the tunneling time and also the different H-elimination channels.

References:

[1] Gareth M. Robert, Adam S. Chatterley, Jamie D. Young, and Vasilios G. Stavros, The Journal of Physical Chemistry Letters **3**, 348 (2012).

Poster No.102

Ab initio dynamics simulations of UV damage to the green fluorescent protein chromophore

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Green Fluorescent Protein (GFP) has revolutionized many fields of biology due its use in imaging organisms at the sub-cellular level. However, upon exposure to UV or intense visible light, GFP undergoes an irreversible reaction, altering its photocycle. A detailed understanding of the pathways that lead to GFP's irreversible damage following excitation to high-energy states will aid greatly in the development of new FPs with improved imaging and lifetime characteristics. Yet the mechanism of GFP's UV-initiated photodamage is currently the subject of controversy, with competing interpretations of photoelectron experiments on the isolated chromophore arguing for either nonadiabatic decay [1] or autoionization [2] from the initial S_3 excited state being dominant.

To address the controversy in GFP's UV-initiated excited-state decay pathways, we performed ab initio multiple spawning excited-state dynamics simulations on the GFP chromophore with on-the-fly multiconfigurational electronic structure using our recently developed dynamically-weighted complete active space self-consistent field (DW-CASSCF) method [3]. DW-CASSCF improves the description of Potential Energy Surfaces (PES) from the workhorse state-averaged (SA) CASSCF method, in particular by removing discontinuities in the PES where states not included in the state averaging cross with those that are. Our simulations of the GFP chromophore reveal that ultrafast non-adiabatic decay of the initial excited state outcompetes autoionization.

References:

[1] A. Bochenkova, C. R. S. Mooney, et al, Chem. Sci. **8**, 3154 (2017).

[2] C. W. West, J. N. Bull, et al, J. Phys. Chem. B **119**, 3982 (2015).

[3] W. Glover, J. Chem. Phys. **141**, 171102 (2014).

Poster No.103

Bimolecular reactions from van der Waals complexes: a quantum dynamical study of $Br + I_2 \rightarrow BrI + I$

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A large body of experimental and theoretical work done so far in time resolved pump-probe spectroscopy concerns unimolecular reactions, whereas bimolecular reactions remain less explored [1-3]. Indeed, molecular collisions are complex to comprehend, since one needs to consider all impact parameters and all possible orientations two reactants may have with respect to each other as they collide. In order to simplify the problem, one can constrain the dynamics of such a collision by putting two reactants in the conditions of forming a weakly bound complex, i.e. aggregates of two molecules kept together by weak interactions such as Van der Waals forces or hydrogen bonds. The initial geometry of the two-molecule system would then be well-defined and represents an ideal initial condition for triggering the reaction with a laser pulse.

Pioneering experimental work by the groups of Sims et al. [2] followed by Tuchler et al. [3] has been carried out on the complex H–Br···I–I, where they could follow the unfolding of the exchange reaction $Br + I_2 \rightarrow BrI + I$ upon dissociation of the HBr moiety by a laser pulse. In this work we analyze their results by a nuclear wavepacket propagation on a model potential energy surface fitted to selected ab initio points.

References:

[1] N. E. Henriksen, Chem. Phys. **442**, 2–8 (2014).

[2] I. R. Sims, M. Gruebele, E. D. Potter, A. H. Zewail, J. Chem. Phys. **97**(6), 4127–4148 (1992)

[3] M. F. Tuchler, S. Wright, J. D. McDonald, J. Chem. Phys. **106**(7), 2634–2645 (1997)

Poster No.104

Experimental observation of the vibrational fingerprint of the cis-trans isomerisation through a 2^1B-S0 conical intersection in cis-stilbene

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A hallmark of many photodynamics lectures, cis-stilbene exhibits both a photoinduced pericyclic ring-closure and photoinduced cis-trans isomerisation. Many experiments have been devoted to studying the photophysics of stilbene, however the experimental difficulties have left the debate on the precise dynamics somewhat open-ended. Much of the theoretical calculations in the existing literature on the cis-trans isomerisation has been consistent in that the 2^1B state is coupled to the ground state in part by torsional motion around the C-phenyl bonds [1,2].

Utilising low-order harmonic generation in a semi-infinite gas cell in combination with a VMI photoelectron spectrometer, we observe the presence of clear oscillatory motions in the time-resolved photoelectron spectrum (TRPES) which can be associated with the C-phenyl torsional motion. Additionally, by examining the time evolution of the oscillatory motion, we see a signature of intramolecular vibrational relaxation for the low frequency components in the time-frequency analysis of the TRPES. By Fourier analysis of the time-resolved photoelectron spectrum, the components can be associated with the overtones of the torsional motion between an ethylenic carbon and a phenylic carbon in trans-stilbene. This is rationalised by the motion of the system through a conical intersection between the 2^1B state and the ground state. This results in the population of highly excited vibrational modes that correspond to the trajectory of the nuclei across the potential-energy surfaces, i.e., the appearance of overtones for these C-phenyl torsional motions in the TRPES.

In this contribution, we demonstrate that we can follow the ultrafast time-resolved dynamics of the 2^1B state of cis-stilbene as it undergoes a photoisomerisation to trans-stilbene. To do this, we measured and assigned the low-frequency vibrational fingerprint of the conical intersection that couples the ground state to the 2^1B state along the C-phenyl torsional motion, which presents itself in the TRPES as overtones of the low-frequency torsional motion.

References:

[1] G. Orlandi et al., Phys. Chem. Chem. Phys., **19**, 25095 (2017).

[2] Y. Dou, and R. Allen, J. Chem. Phys., **119**, 10658 (2003).

Poster No.105

Highly efficient reduction of graphene oxide (GO) in GO/MXene hybrid films revealed by ultrafast spectroscopy

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For graphene oxide (GO) and reduced GO (rGO), we have observed the quantum-confined graphene-like electronic states in insulator-like GO and semiconductor-like rGO by femtosecond selective excitation in broadband transient absorption spectroscopy [1], and then, investigated their fluorescence evolution processes [2]. Here, we further find that there is semimetal-like rGO, which possesses the bleaching features of both semiconductor-type band gap of rGO and semimetal-type graphene. This indicates a two-phase coexistence phenomenon in semimetal-like rGO, due to the nature of inhomogeneity of rGO. Finally, we find that MXene ($Ti_3C_2T_x$) can reduce GO to semimetal-like rGO which even possesses graphene-like transient spectral evolution. It demonstrates a highly efficient reduction of GO in GO/MXene hybrid films.

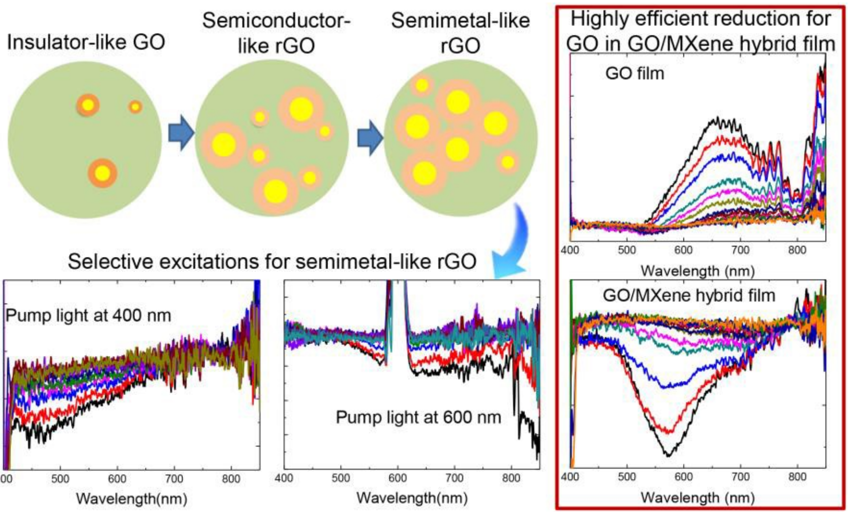

Figure 1. Illustration for the electronic structure of GO, rGO, and rGO/MXene.

References:

[1] L. Wang, H. Y. Wang, Y. Wang, S. J. Zhu, Y. L. Zhang, J. H. Zhang, Q. D. Chen, W. Han, H. L. Xu, B. Yang, and H. B. Sun, Adv. Mater. **25**, 6539 (2013).

[2] Y. Y. Yue, Y. Chen, Y. X. Zhang, L. Wang, and H. Y. Wang, Opt. Mater. Express **7**, 2519 (2017).

Poster No.106

The role of nonradiative processes in photodissociation dynamics of S_1 state of propanal

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The UV photochemistry of aliphatic aldehydes has been extensively studied because of the very important role in atmospheric chemistry [1-3]. Propanal, as a typical larger aliphatic aldehyde, its photodissociation dynamics has been discussed under different wavelengths [4-6]. However, the pioneering photophysics nonradiative processes, which are closely related to the final decomposition pathways, remains less explored and very little is known about the ultrafast dynamics in related excited states and the corresponding role in the photodissociation. The ultrafast nonradiative dynamics in S_1 electronic excited state and the corresponding photodissociation dynamics in propanal molecule have been investigated with time-resolved photoelectron imaging and time-of- flight mass spectrometry at 330 and 320 nm. For excitation to the S_1 state near the band origin at 330 nm, the population undergoes ultrafast internal conversion (IC) to S_0 on a 153 fs time scale and intersystem crossing (ISC) to T_1 within 774 fs, followed by the second slower ISC induced by $S_0 \leftarrow T_1$ on a 106 ps time scale, which favors the dissociation pathway of molecular channel into CH_3CH_3+CO products. As the S_1 state is populated following 320 nm excitation, only the IC to S_0 and the first ISC to T_1 was observed within 73 fs and 663 fs, respectively. Most of the molecules in T_1 state dissociate into CH_3CH_2+CHO radical products since the energy provided by 320 nm photons are sufficient enough to surmount the barrier on the triplet surface. The quantum yield of CHO product at 320 nm is significantly higher than that of 330 nm. It is of great interest to investigate the ultrafast nonradiative dynamics of electronic excited state in propanal and to discuss how the products pathways in photodissociation chemistry is affected.

References:

[1] H. Kato, and M. Baba, Chem. Rev. **95**, 2311 (1995).

[2] D. Shemesh, S. L. Blair, S. A. Nizkorodov and R. B. Geber, Phys. Chem. Chem. Phys. **16**, 23861 (2014).

[3] K. C. Lin, P. Y. Tsai, M. H. Chao, M. Nakamura, T. Kasai, A. Lombardi, F. Palazzetti, and V. Aquilanti, Int. Rev. Phys. Chem. **37**, 217 (2018).

[4] Y. Chen, and L. Zhu, J. Phys. Chem. A **105**, 9689 (2001).

[5] P. Y. Tsai, H. K. Li, T. Kasai and K. C. Lin, Phys. Chem. Chem. Phys. **17**, 23112 (2015).

[6] A. W. Harrison, and S. H. Kable, J. Chem. Phys. **148**, 164308 (2018).

Poster No.107

Ultrafast decay dynamics of higher-lying excited states of water

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Photochemistry of water in the gas phase is one of the most important, yet challenging topic in molecular reaction dynamics. It also serves as an ideal model system to study complicated excited-state dynamics of polyatomic molecules. Excited-state dynamics of water molecules has been intensively studied, both experimentally and theoretically, for more than four decades and rich information has been obtained from various studies, such as absorption spectra, resonance-enhanced multiphoton ionization (REMPI) spectra, OH product fluorescence excitation spectra, H product action spectra and photodissociation dynamics, time-resolved excited-state dynamics, etc.

Among these studies, experiments are mainly limited to the frequency domain, due to the lack of the wavelength-tunable fs laser source in the VUV range. In the last couple of years, we have studied the ultrafast decay dynamics of water molecules excited into the C, D, D’, D’’ and F states, by using two-photon excitation and time-resolved photoelectron imaging (TRPEI) techniques [1-3]. Extensive information on the ultrafast decay dynamics of these higher-lying excited states has been derived. In this talk, a detailed report on the experimental results will be given.

References:

[1] Z. He, Z. Chen, D. Yan, D. Dai, G. Wu and X. Yang, Chin. J. Chem. Phys. **30**, 247 (2017).

[2] Z. He, D. Yang, Z. Chen, K. Yuan, D. Dai, G. Wu and X. Yang, Phys. Chem. Chem. Phys. **19**, 29795 (2017).

[3] D. Yang, Y. Min, Z. Chen, Z. He, Z. Chen, K. Yuan, D. Dai, G. Wu and X. Yang, Chin. J. Chem. Phys. **32**, 59 (2019).

[4] Y. Min, D. Yang, Z. He, Z. Chen, K. Yuan, D. Dai, G. Wu and X. Yang, Phys. Chem. Chem. Phys., submited.

Poster No.108

Photoexcitation dynamics of Roussin's Red Ester as nitric oxide donor

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Femtosecond infrared spectroscopy was used to study photodissociation dynamics of nitric oxide (NO) from Roussin's Red Ester (RRE), $[\mathrm{Fe_2(\mu\text{-}RS)_2(NO)_4}]$ in water in broad time range spanning from 0.3ps to 10μs after excitation at various wavelengths (310, 360, and 400 nm). When excited, RRE either releases a NO producing a radical species deficient one NO (**RS**), $[\mathrm{Fe_2(\mu\text{-}RS)_2(NO)_3}]$ or relaxes into the ground state without the photodeligation via a metastable state (**MS**). The **RS** appears immediately after photoexcitation, suggesting that the photolysis of one NO is faster than 0.3 ps. Some fraction of **RS** geminately recombines with NO on the picosecond time scale and the remaining **RS** recombines bimolecularly with a rate constant of $7\times10^{7}\ \mathrm{M^{-1}s^{-1}}$[1,2]. For a given RRE molecule, the fractional yield of the **MS** depends on the excitation wavelength (λ_{ex}) but the relaxation time of the **MS** was independent of the λ_{ex}. Most efficient NO delivery condition was obtained by revealing the λ_{ex} with lowest GR yield as well as the highest primary quantum yield. Detailed photodissociation dynamics of RRE including the NO-producing quantum yield can be utilized in the pin-point usage of NO in chemistry and biology [3,4].

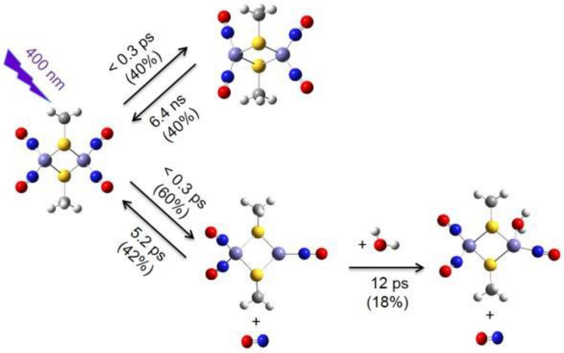

Figure 1. A scheme used to fit time-dependent population change after photoexcitation.

References:

[1] José Clayston Melo Pereira, A. V. I., Rui-Min Han, and Peter C. Ford, J. Am. Chem. Soc. **137**, 328 (2015).

[2] Christa L. Conrado, James L. Bourassa, Christian Egler, Stephen Wecksler, and Peter C. Ford* Inorganic Chemistry **42**, 2288 (2003).

[3] Alexis D. Ostrowski and Peter C. Ford, Dalton transactions **48**, 10660 (2009)

[4] Han-Hun Chang,a Hung-Jen Huang,a Yun-Lung Ho,b Yu-Der Wen,b Wei-Ning Huangc and Show-Jen Chiou* Dalton Transactions **32**, 6396 (2009).

Poster No.109

Li$^+$ ion solvation in organic electrolytes determined by ultrafast vibrational spectroscopy

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Lithium ion batteries are an attractive power source for a wide variety of applications. To expanding the performance limit of current Li-ion batteries requires ion-solvent interaction, which governs the ion transport behavior of the electrolytes, to be fully understood. We herein examine the coordination number of Li$^+$ ion in LiPF$_6$-organic electrolyte solutions by using femtosecond vibrational spectroscopy. In this abstract, we discussed Li+ ion solvation in CH$_3$CN, EC, PC, and DMC solutions.

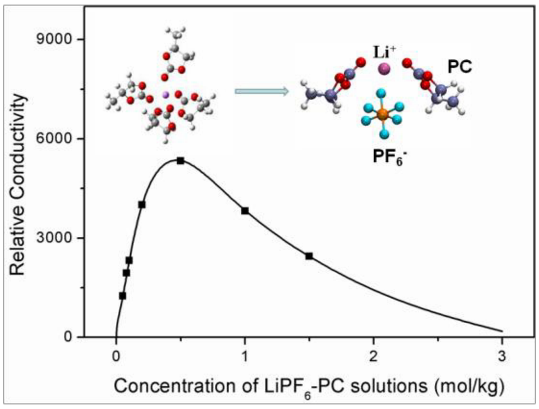

Figure 1. The LiPF$_6$ salt concentration dependent LiPF$_6$-PC solvation structures, in accord with the variation of the ion conductivity.

References:

[1] K. J. Yuan, H. T. Bian, Y. N. Shen, B. Jiang, J. B. Li, Y. F. Zhang, H. L. Chen and J. R. Zheng*, J. Phys. Chem. B, **118**, 3689(2014).

[2] Y. N. Shen, G. H. Deng, C. Q. Ge, Y. H. Tian, G. R. Wu, X. M. Yang, J. R. Zheng and K. J. Yuan*, Phys. Chem. Chem. Phys., **18**, 14867(2016).

[3] B. Jiang, V. Ponnuchamy*, Y. N. Shen, X. M. Yang, K. J. Yuan*, V. Vetere, S. Mossa,

[4] Skarmoutsos*, Y. F. Zhang and J. R. Zheng*, J. Phys. Chem. Lett., **7**, 3554(2016).

[5] H. Jiang, Q. Zhang, Y. Tong, L. Sui, G. Wu, K. J. Yuan*, X. M. Yang, Phys. Chem. Chem. Phys., ASAP(2019)

Poster No.110

Photoinduced charge migration in anthraquinone pharmaceutical product

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Over the past decade there has been a considerable amount of research toward understanding the unimolecular deactivation pathway of photoexcited pharmaceutical products in the presence of biological substrates [1,2]. Quinones are also widely employed as electron and hydrogen atom acceptors in thermal and photochemical processes by nature as well as in man-made systems [3]. The excited quinones react with DNA by two separate oxidative pathways: hydrogen atom abstraction from the deoxyribose component of the nucleic acid backbone and electron transfer from a nearby base. A substitution of anthraquinone has a pronounced effect on its electronic structure and photophysical properties. In this work, we investigated the mechanism of intramolecular charge transfer and the following radiationless dynamics of the excited states in a serial of anthraquinones [4-6]. Time-resolved spectra show a strong non-radiation transition processes in the prepared samples, and this results in very strong electron-photon interaction causing a charge transfer transition. The intramolecular charge transfer mechanism is observed and verified. Moreover, Intramolecular hydrogen bonding and dynamics of intramolecular proton transfer have been inspected and excluded in quinone systems. It is revealed that the molecules show distinct intramolecular and intermolecular charge transfer characters under different experimental conditions. The electronic structures of donors and acceptors obviously effect the rate of the charge transfer. Furthermore, the contribution of an electron transfer mechanism is clarified in human damage and the properties for electron transfer of pharmaceutical product have been discussed verifying the participation of the triplet states. Our results observed here have yield a physically intuitive picture of the photoinduced charge transfer and radiationless dynamics in anthraquinone pharmaceutical products.

References:

[1] Y. Cheng, H. Cheng, C. Jiang, X. Qiu, K. Wang, W. Huan, A. Yuan, J. Wu, and Y. Hu, Nat. Comm. **6**, 8785 (2015).

[2] F. Bergeron, V. K. Nair, and J. R. Wagner, J. Am. Chem. Soc. **128**, 14798 (2006).

[3] M. H. V. Huynh, and T. J. Meyer, Chem. Rev. **107**, 5004 (2007).

[4] S. Zhang, S. Sun, M. Zhou, L. Wang, and B. Zhang, Sci. Rep. **7**, 43419 (2017).

[5] M. Zhou, S. Zhang, L. Wang, and B. Zhang, Chem. Phys. Lett. **710**, 1 (2018).

[6] M. Zhou, L. Wang, S. Zhang, and B. Zhang, Chem. Phys. Lett. **717**, 1 (2019).

Poster No.111

Solvent effects on triplet-triplet annihilation upconversion of perylene with C_{60}-Bodipy as photosensitizer

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Solvent effects usually play an important role in triplet-triplet annihilation (TTA) upconversion process [1,2]. In this work, we investigated the kinetics of TTA upconversion fluorescence of perylene with C_{60}-Bodipy as triplet photosensitizers in five solvents, 1,4-dioxane, toluene, chlorobenzene, dichlorobenzene, and tetrahydrofuran (THF). The overall TTA upconversion quantum yields were measured, e.g. 14.1% in dichlorobenzene, 13.4% in chlorobenzene, 9.8% in dioxane, 9.2% in toluene, and 4.4% in tetrahydrofuran. The intramolecular energy transfer in the C_{60}-Bodipy dyad was confirmed using femtosecond transient absorption spectroscopy, while the triplet-triplet energy transfer (TTET) and TTA processes were analyzed using nanosecond transient absorption spectroscopy. Of special interest was that a nonsignificant dependence on solvents was found in the system, except for fluorescence emission of perylene itself.

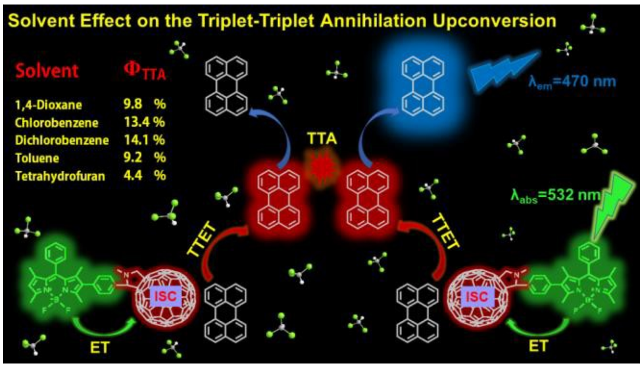

Figure 1. Solvent effect on TTA upconversion of perylene with C_{60}-Bodipy as photosensitizers.

References:

[1] Q. H Zhou, M. M. Zhou, Y. X Wei, X. G. Zhou, S. L. Liu, S. Zhang, and B. Zhang, Phys. Chem. Chem. Phys. **19**, 1516 (2017).

[2] Y. X. Wei, M. M. Zhou, Q. H. Zhou, X. G. Zhou, S. L. Liu, S. Zhang, B. Zhang Phys. Chem. Chem. Phys. **19**, 22049 (2017).

Poster No.112

Ultrafast dynamics in methylated cytosine and its derivatives

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DNA methylation is a stable epigenetic mark that has an important role in gene regulation.[1, 2] DNA methylation in the form of 5-methylcytosine (5mC) can be actively reversed to unmodified cytosine(C) through TET dioxygenase-mediated oxidation of 5mC to 5-hydroxymethylcytosine (5hmC), 5-formylcytosine (5fC) and 5-carboxylcytosine (5caC). Thus, 5-methylcytosine is often referred to "the fifth base of DNA". A comprehensive understanding of the electronic excited state relaxation in cytosine and its methylated derivatives is crucial for revealing UV-induced photodamage to the biological genome[3]. We used femtosecond time-resolved spectroscopy to comprehensively study the excited state dynamics of 5mC, 5hmC, 5fC and 5caC in solution[4, 5]. Two distinct nonradiative decay channels were directly observed in 5mC and 5hmC singlet states while long-lived triplet state was seen in 5fC but not in 5caC. Potential double proton transfer were also observed in 5hmC, 5fC and 5caC.

Reference

[1] Ito, S., et al., Tet Proteins Can Convert 5-Methylcytosine to 5-Formylcytosine and 5- Carboxylcytosine. Science,.**333**(6047): p. 1300-1303(2011).

[2] Pfaffeneder, T., et al., The Discovery of 5-Formylcytosine in Embryonic Stem Cell DNA. Angewandte Chemie, **50**(31): p. 7008-7012(2011).

[3] Crespo-Hernández, C.E., et al., Ultrafast excited-state dynamics in nucleic acids. Chem. Rev., **104**: p. 1977-2019(2004).

[4] Wang, X., et al., Excited State Decay Pathways of 2′-Deoxy-5-methylcytidine and Deoxycytidine Revisited in Solution: A Comprehensive Kinetic Study by Femtosecond Transient Absorption. The Journal of Physical Chemistry B, **122**(28): p. 7027- 7037(2018).

[5] Pollum, M., L. Martinez-Fernandez, and C.E. Crespo-Hernández, Photochemistry of nucleic acid bases and their thio- and aza-analogues in solution. Top. Curr. Chem. **355**: p. 245-327(2015).

Poster No.113

Relaxation of excited states in DNA nucleobase adenine studied by sub-picosecond XEOL

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Prominent photostability of nucleobases to UV radiation has been approached for the last two decades by various time-resolved techniques and theoretical modelling; although it is agreed that photostability results from ultra-fast relaxation of excited states, the exact pathways are still debated. Here we approach the problem of resistance of nucleobases to ionizing radiation, which has not been addressed on the level of energy relaxation pathways so far, by a novel technique: trXEOL (time-resolved X-ray Excited Optical Luminescence) with sub-picosecond time resolution. In trXEOL, the rise time reflects population of intermediate states, decay time – relaxation of this intermediate state (Fig.1). Reported here are the first to our knowledge measurements of XEOL in the femtosecond regime. The experiment has been performed at FLASH using 3rd harmonic of FEL radiation as excitation and optical laser pulses at 810 nm synchronized to FEL pulses to gate optical luminescence from sample in a BBO crystal. The adenine and 9-methyl-adenine have been studied under excitation below (380 eV) and above (430 eV) Nitrogen K-edge. The characteristic UV fluorescence of adenine typically observed under UV excitation has been detected. Typical temporal signature of XEOL in adenine is presented in Fig.1. It can be fitted by a hyperbolic decay law with characteristic 380 fs rise time and 600 fs decay time for 320 nm luminescence and 1.1 ps decay time for 340 nm luminescence. No difference in temporal profiles of luminescence for excitations below and above Nitrogen K edge has been observed, and slightly faster decay after first 2 ps in case of 9-methyl-adenine indicates an additional quenching channel of intermediate excited states in modified material. Altogether, our results demonstrate efficient pathways of energy relaxation in adenine to high energy excitations within sub- to few picoseconds and support the concept of redundant pathways of energy relaxation in adenine developed by nature.

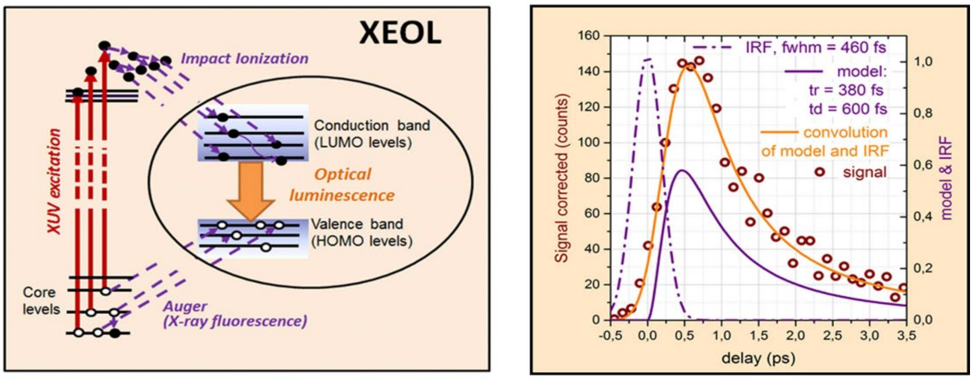

Figure 1. Schematic of XEOL processes (left); trXEOL in adenine (right).

Poster No.114

Mapping the intermolecular excited state proton transfer channel between biliverdin dimethyl ester and solvent molecules by ultrafast spectroscopy

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The deprotonation and protonation behavior of linear tetrapyrrole involves in many biological processes. Chromophore like biliverdin in protein surroundings tends to change its proton state and conformation to alter the luminescence state of the whole protein.[1] However, the specific dynamics involved have never been clearly described, to the best of our knowledge. Biliverdin dimethyl ester (BVE) are dissolved in a series of protic and aprotic solvents. A new luminous state with lifetime longer than 300 ps, for BVE in protonic solvent, are found with the help of picosecond time-correlated single photon counting (TCSPC) spectrometer and a time-resolved up-conversion fluorescence device. Transient absorption (TA) spectroscopy are used to indicates the switchability of BVE molecules between the light and dark states, under different solution conditions. An interesting proton transfer channel from the pyrrole ring to solvents is established. These study clearly describe the deexcitation behavior of a linear tetrapyrrole molecule in the solution phase and its interactions with surrounding environment. It has profound implications for understanding its role in proteins and other biological environments.

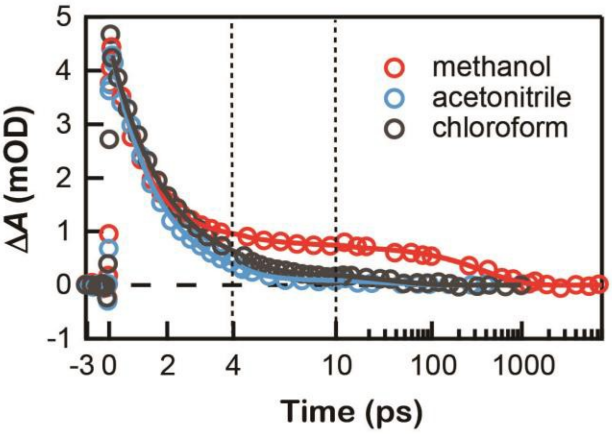

Figure 1. Excited state absorption kinetics of BVE in three solvents

References:

[1] A. Schmidt, L. Sauthof, M. Szczepek, M. F. Lopez, F. V. Escobar, B. M. Qureshi, N. Michael, D. Buhrke, T. Stevens, D. Kwiatkowski, D. von Stetten, M. A. Mroginski, N. Krauss, T. Lamparter, P. Hildebrandt, P. Scheerer, Nat Commun **9** (1), 4912(2018).

Poster No.115

Structural characters and radical propagation in Escherichia Coli class Ia ribonucleotide reductase from multistate technique

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RNR catalyzes the reduction of NDP to dNDP, requiring reversible radical transport across two homodimeric subunits (α2 and β2, which form an active α2β2 complex) over 35 Å via multiple PCET steps through conserved aromatic amino acid residues: Y122$^{\bullet}$↔W48?↔Y356 in β2, across the interface to Y731↔Y731↔C439 in α2. The transient C439$^{\bullet}$ is responsible for initiating nucleotide reduction. However, the structure of an active α2β2 complex of RNR is unknown due to lack of the interfacial information. We firstly constructed the entire structure of α2β2 complex including the missing 19 C-terminal residues (341-359) in the β2. Then MD simulations were employed to validate its rationality. The relative distances for key residues involved radical propagation pathway were obtained, which are in good agreement with the recent experimental observations. Based on constructed structure, the PCET reactions in α2 were investigated using QM/MM method.

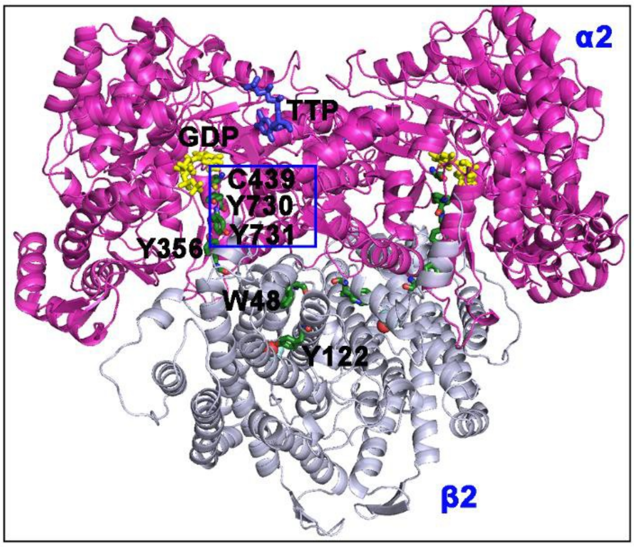

Figure 1. Entire structure of α2β2 complex

References:

[1] H.S. Sharon, and A. A. Stuchebrukhov. Chem. Rev. **110**(12), 6939 (2010).

Poster No.116

How epigenetic DNA nucleosides regulate DNA photodamage: Photo properties of epigenetic nucleosides revealed by ultrafast laser spectroscopy

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DNA methylation and demethylation are the crucial steps in epigenetics, which have an important role in gene regulation and mammalian development.[1-2] Methylated cytidine and its further oxidative products (5-hydroxymethylcytidine, 5-formylcytidine and 5-carboxylcytidine) are now called the "new four nucleosides of DNA". However, the appearance of these new epigenetic nucleosides changes the photochemical properties of natural DNA because they could have distinct excited state dynamics. For 2'-deoxy-5-methylcytidine and 2'-deoxy-5-hydroxymethylcytidine, several picosecond singlet excited state lifetimes can be found on the account of the appearance of energy barrier in ππ*. For 2'-deoxy-5-formylcytidine, we present direct observation of long-lived triplet state with a high triplet state quantum yield (69%), which eventually largely enhance DNA photodamage in solution. Meanwhile, 2'-deoxy-5-carboxylcytosine can dissipate most of the excess energy to the environment as heat within 1 ps. These results provide valuable information on how epigenetic DNA nucleosides regulate DNA photodamage in the initial stage.

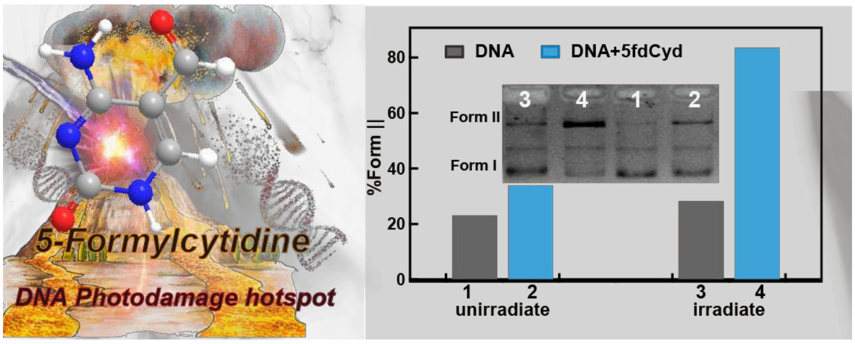

Figure 1. Agarose gel electrophoresis for formation of DNA form II upon irradiation of plasmid pBR322 in the presence or absence of 5fdCyd.

References:

[1] S. Ito, L. Shen, Q. Dai, S. C. Wu, L. B. Collins, J. A. Swenberg, C. He, Y. Zhang, science **333**, 1300-3(2011).

[2] Y.-F. He, B.-Z. Li, Z. Li, P. Liu, Y. Wang, Q. Tang, J. Ding, Y. Jia, Z. Chen, L. Li, Y. Sun, X. Li, Q. Dai, C. Song, K. Zhang, C.He, G. Xu, Science (**333**), 1303-1307(2011).

Poster No.117

Observation of electronic relaxation in thymine to its ground state after UV excitation through VUV photoelectron spectroscopy

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The DNA nucleobases famously show a surprising photostability upon UV excitation. It has previously been posited that this photostability is responsible for protecting the genetic code from UV damage - which in turn allowed life on Earth to evolve. Previous studies have indicated that the origin of this photostability is likely due to the excited-state wavepacket being efficiently directed to the electronic ground state via various conical intersection seams on the potential energy surfaces, however due to experimental limitations, the recovery time of the ground state population was previously impossible to observe.

There has been much debate over the precise photoinduced dynamics of thymine at 267 nm in the past. Previous studies have suggested that the decay from the initially prepared $S_2(\pi\pi^*)$ state undergoes ultrafast internal conversion to the $S_1(n\pi^*)$ state in less than 50 fs, which then decays further to the ground state after 2.4 ps. Contradictory to this, other work has suggested that the initial 50 fs decay is due to the relaxation of the molecule out of the highly vibronically excited Franck-Condon region to the S_2 minimum. Despite excellent and thorough work done by many groups around the world, there is still no strong consensus on the mechanism by which thymine relaxes to the ground state.

In this contribution, we present that the recovery time of the ground state upon UV excitation is observed using time-resolved vacuum-ultraviolet photoelectron imaging. By using a probe wavelength that is able to directly ionise the ground state, we are able to show that the ground state recovery time is significantly faster than what was previously measured. We find that there are two channels at play in the spectrum. The major channel decays to the ground state within the first 50 fs, whereas the minor channel displays a much longer ground-state recovery time.

References:

[1] Kang et al., J. Am. Chem. Soc., **124**, 12958 (2002).

[2] Hudock et al., J. Phys. Chem. A, **111**, 8500 (2007).

[3] Ulrich et al., Phys. Chem. Chem. Phys., **6**, 2796 (2004).

Poster No.118

Fluorescence quenching dynamics of AMND in abasic site-containing DNA for nucleobase recognition

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DNA is the main component of the chromosomes and the carrier of the genetic information. However, the DNA inside the cell is continuously interfered by external or internal factors. A cell produces about 10^4 purine or pyrimidine shedding sites every day, which are called abasic site (AP site). Accordingly, a DNA duplex containing AP site is commonly referred to AP-dsDNA. We have systematically studied the mechanism of nucleobase recognition by fluorescence quenching of 2-amino-7-methyl-1,8-naphthyridine (AMND) in the AP site-containing DNA duplexes. Our time-resolved fluorescence spectroscopy measurements reveal that such fluorescence quenching is originated from the photo-induced electron transfer from the flanking base guanine to the excited AMND in AP site-containing DNA duplexes, and the fluorescence responses of the excited AMND depend on the static π-stacking effects and dynamic conformation states caused by the interactions between AMND and the flanking bases. The triple-exponential function is applied for analyzing the fluorescence quenching dynamics. The electron transfer from guanine base to AMND* in hundreds of picoseconds and the fractional populations of different conformation states with different are both obtained. Our time-resolved measurements have point out that the electron transfer reactions between AMND and flanking bases is responsible for nucleobase recognition through the fluorescence quenching in AP site DNA. Meanwhile the conformational diversities caused by base motions plays an important role in fluorescence quenching of AMND moiety, which can be served as a sensitive probe of DNA structural dynamics. Therefore, AMND can act as a dual reporter of DNA structural sequence and nucleobase dynamics.

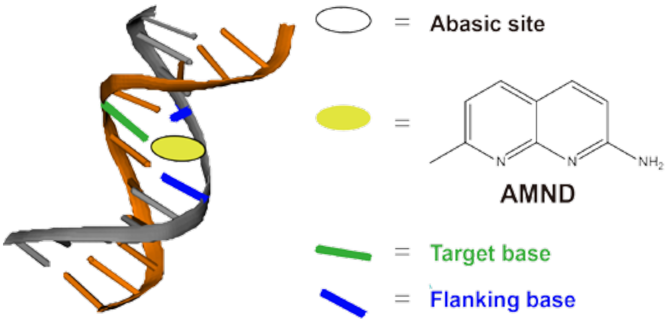

Figure 1. Schematic illustration of nucleobase recognition via interactions between AMND and the AP site-containing DNA duplexes.

References:

[1] K. Yoshimoto, S. Nishizawa, M. Minagawa, N. Teramae, Journal of the American Chemical Society,**125**, 8982-8983(2003).

[2] J. R. Lakowicz, Principles of Fluorescence Spectroscopy, Boston, MA: Springer US, Boston, MA, (2006).

[3] K. Yoshimoto, S. Nishizawa, H. Koshino, Y. Sato, N. Teramae, M. Maeda, Nucleic Acids Symposium Series, **49**, 255-256(2005).

[4] P. A. Sontz, N. B. Muren, J. K. Barton, Accounts of Chemical Research*,* **45**, 1792-1800(2012).

[5] R. K. Neely, D. Daujotyte, S. Grazulis, S. W. Magennis, D. T. F. Dryden, S. Klimašauskas, A. C. Jones, Nucleic Acids Research, **33**, 6953-6960*(*2005).

Poster No.119

Ultrafast dynamics of short-range protein electron transfer in anabaena flavodoxin

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Short-range electron transfer (ET) is ubiquitous in nature and is closely related to the functions of photosynthetic systems, photolyases, photoreceptors and so on. As short-range electron transfer occurs on an ultrafast timescale from femtoseconds to hundreds of picoseconds, which is similar to the timescale of the local environment relaxation of the protein and the solvent molecules, the dynamics is in nonequilibrium where the classical Marcus theory does not apply. Here, using oxidized Anabaena (Ana) flavodoxin as a model system, we have systematically studied the photoinduced redox cycle of the wildtype and six mutant proteins by means of the femtosecond fluorescence up-conversion and transient absorption spectroscopy. We observed ultrafast ET dynamics from the initial charge separation in 50-197 fs, charge recombination in 0.61-1.51 ps and the final cooling process of the products in 2.8-4.0 ps. The results have been compared with those obtained with D.vulgaris (DV) Flavodoxin.[1] The forward electron transfer from the photo-excited FMN to the adjacent W/Y in both species is much faster than the local environment relaxation, and thus falls in the 'frozen' region in the extended Sumi-Marcus two-dimensional model. In Ana flavodoxin, the forward electron transfer rate in the Y channel is faster than that in the W channel, while the trend is reversed in DV flavodoxin. This suggests the coupling constants between the donor and acceptor rather than the driving forces play a dominant role in determining the electron transfer rates in different channels in Ana flavodoxin.

References:

[1] He, T. F.; Guo, L.; Guo, X.; Chang, C. W.; Wang, L. and Zhong, D. Biochemistry, **52**, 9120−9128 (2013).

Poster No.120

Ultrafast vibrational energy relaxation of membrane-bound peptide and the water in ion channel

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Vibrational energy transfer (VET) of proteins at cell membrane plays critical roles in controlling the protein functionalities, but its detection is very challenging. By using a surface-sensitive femtosecond time-resolved sum frequency generation vibrational spectroscopy with infrared pump, the detection of the ultrafast VET in proteins at cell membrane has finally become possible. By pumping amide I band and probing amide I band, it is found that the relaxation time shows a very strong dependence on the H_2O exposure, but not on D_2O exposure. This observation clearly indicates that the exposure of amide I bond to H_2O opens up a resonant relaxation channel and facilitate direct vibrational energy transfer from the amide I mode to the H_2O bending mode. The protein backbone motions can thus be energetically coupled with protein-bound water molecules. Using this method, we measured the relaxation time of the amide I and amid A modes in the Influenza A M2 transmembrane domain (a pH-activated tetrameric proton channel embedded in the viral membrane), we verified that about 10%~20% amino acid residues of M2 protein are exposure to the water in the channel close state, indicating water molecules is still present inside the channel of M2 in the close state. The water molecules are coupling to C=O groups, rather than N-H group.

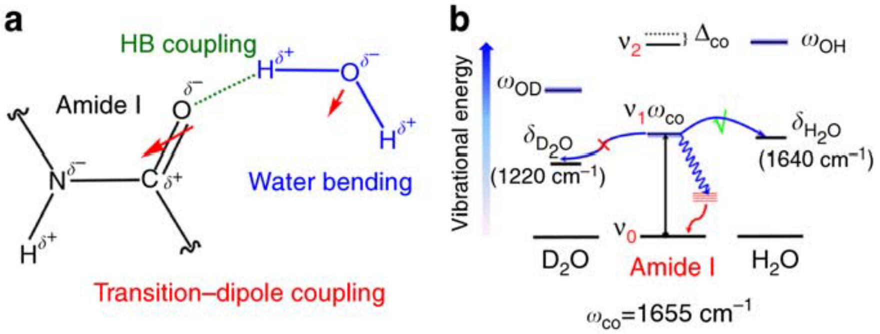

Figure 1. Coupling scheme and energy diagram. a Interaction between amide carbonyl and water through hydrogen bonding (HB) and transition–dipole coupling. b Energy-level depiction of the vibrational relaxation of the amide I mode to the water bending mode

References:

[1] J. Tan, J. Zhang, C. Li, Y. Luo, and S.J. Ye. *Nat. Commun.* **10**, 1010 (2019).

Poster No.121

Theoretical exploitation of acceptors for organic NIR-II fluorophores

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Small-molecule dyes with fluorescence emission in the NIR-II region (1000 - 1700 nm) have attracted considerable attention in the biomedical and bioimaging fields [1]. However, the currently reported organic NIR-II fluorophores are still limited and there is great demand to develop other novel NIR-II fluorophores besides benzobisthiadiazole (BBT)-based fluorophores [2]. More importantly, there is lack of the appropriate level of theory capable of providing both efficient and accurate predictions of the electronic structures of organic NIR-II fluorophores. In this work, successful application of time-dependent density functional theory (TDDFT) using optimally-tuned range-separated functionals [3] for calculations of both the absorption and fluorescence spectral properties has been demonstrated compared with the available experimental data. A series of thiadiazole-based acceptors (A) and derivatives based on the D-A-D skeleton are designed coupled with the triphenylamine donor (D). The structure-property relationships for these fluorophores are thus revealed by analyzing their ground (S_0) and excited (S_1) state geometries, frontier molecular orbitals (HOMO and LUMO), HOMO-LUMO energy gap, oscillator strength, hole-electron distribution and fluorescence wavelength. It is suggested that the existence of hypervalent structure leading to a much lower LUMO level and accompanying with significant hole-electron separation play a key role in the red-shift of the fluorescence emission in NIR-II region. In addition, the substitution of BBT oligomers and analogues as acceptor core is an efficient way to possess both a red-shifted fluorescence wavelength and an enhanced oscillator strength.

References:

[1] A. L. Antaris, H. Chen, K. Cheng, Y. Sun, G. Hong, C. Qu, S. Diao, Z. Deng, X. Hu, B. Zhang, Nat. Mater. **15**, 235 (2016).

[2] Q. Yang, Z. Ma, H. Wang, B. Zhou, S. Zhu, Y. Zhong, J. Wang, H. Wan, A. Antaris, R. Ma, Adv. Mater. **29**, 12 (2017).

[3] H. Sun, S. Zhang, C. Zhong, Z. Sun, J. Comput. Chem. **37**, 684 (2016).

Poster No.122

Ultrafast charge transfer and energy dissipation in 2D materials

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Two-dimensional (2D) materials, e.g. graphene, layered transition-metal dichalcogenides, have demonstrated a wide variety of unique optical, electrical, thermal and mechanical properties. Especially, van der Waals heterostructures realized by stacking various 2D materials give rise to fascinating photoelectric phenomena, providing the potential to fabricate atomic layered light-harvesting and optoelectronics device components. To reveal the ultrafast behaviors of various photo-induced carriers in 2D materials, we developed a novel ultrafast spectroscopy combining the femtosecond ultra-broadband infrared detection and microscopic technique. Based on the different response of photo-induced carriers on the detection wavelength ranging from visible, near-infrared to mid-infrared, dynamics of various kinds of excitons and free carriers in 2D materials can be specifically investigated. As a result, detailed pathways and mechanisms of some essential processes were well addressed, such as ultrafast formation of interlayer hot excitons in MoS_2/WS_2 heterostructures, ultrafast energy dissipation via coupling with internal and external phonons [1-2], etc.

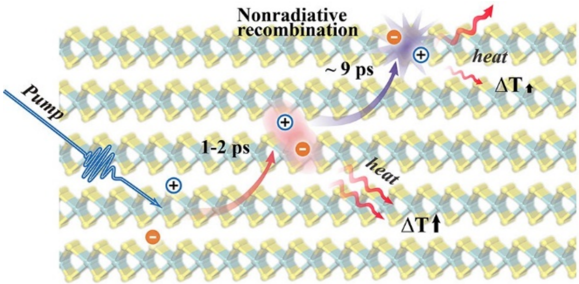

Figure 1. Two nonradiative energy dissipation channels after ultrafast excitation.

References:

[1] Z. Chi, H. Chen, Z. Chen, Q. Zhao, H. Chen, and Y. -X. Weng, ACS Nano **12**, 8961 (2018).

[2] H. Chen, X. Wen, J. Zhang, T. Wu, Y. Gong, X. Zhang, J. Yuan, C. Yi, J. Lou, and P. M. Ajayan, Nature communications **7**, 12512 (2016).

Poster No.123

Investigating the dynamics of excitons in monolayer WSe_2 before and after organic super acid treatment

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Due to the large photoluminescence quantum yield, high mobility and good stability, organic super acid treated two-dimensional WSe_2 has drown much attention. However, reports about the influence of organic super acid treatment on dynamic processes of excitons of monolayer WSe_2 are still rare. In this work, through broadband transient absorption (TA) spectra measured by femtosecond pump-probe system, we determine the dynamics of the A' and C excitons in monolayer and bulk WSe_2 at room temperature. Besides, we also observe the relaxation process of the holes between the two spin split states in valence band maximum in organic super acid treated monolayer WSe_2. We find that the organic super acid treatment on monolayer WSe_2 does not change the peak positions of exciton states, while those bleaching peaks' intensities increase significantly due to the enhancement of oscillator strength for exciton states, corresponding to stronger steady-state photoluminescence. Our work offers a useful approach to study the dynamics of excitons in two-dimensional layered semiconductors.

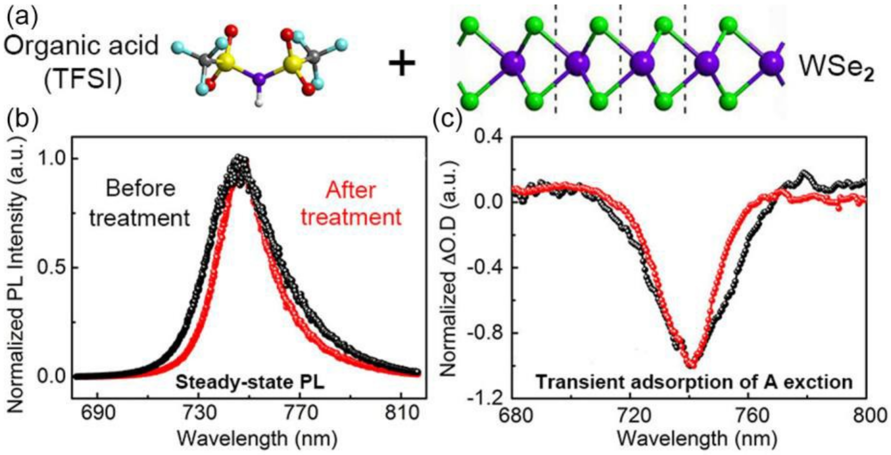

Figure 1. (a) Schematic diagram of organic super acid treated monolayer WSe_2 (osa-monolayer WSe_2); (b) The normalized PL spectra of monolayer WSe_2 before and after treatment with organic super acid; (c) The normalized TA spectra of monolayer WSe_2 before and after treatment with organic super acid.

References:

[1] A. X. Chen, B. L. Wang, and C. H. Y. Wang et al. Nanoscale, **10**, 9346 (2018).

[2] A. M. Amani, B. D. Lien, and C. D. Kiriya et al. Science, **350**, 1065 (2015).

Poster No.124

Visualization of electrons movement in materials

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The dynamics of charged carriers such as electrons and holes largely accounts for various material's physical and chemical properties spanning from carrier transport to chemical reactivity. Real-space and real-time visualization offers intuitive picture of such dynamics. I will present advances of imaging and capturing non-equilibrium charge carrier dynamics taking place on materials surfaces and interfaces. The use of time-resolved scanning electron microscopy techniques allows us to elucidate dynamical steps of carrier excitation, relaxation and diffusion processes occurring in materials in real space and time. The results of these experiments will be exemplified by such dynamics in conventional semiconductors, such as gallium arsenide [1], and 2D materials, such as single- layer graphene [2].

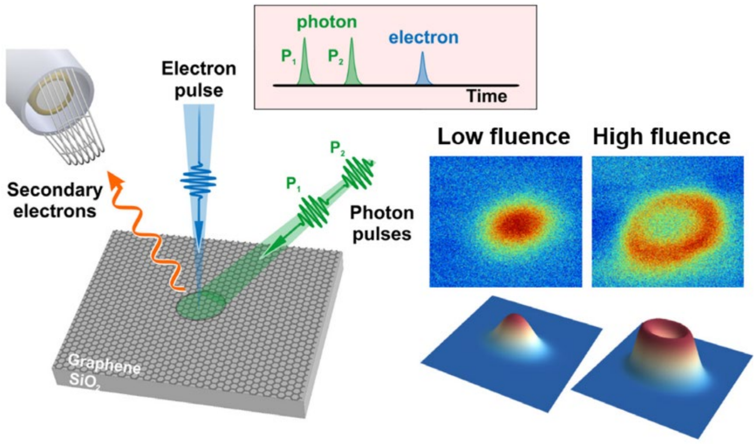

Figure 1. Carrier dynamics in single-layer graphene [2].

References:

[1] J. Cho, T. Y. Hwang, and A. H. Zewail, PNAS **111**, 2094 (2014).

[2] J. Cho, J. Tang, T. Y. Hwang, and A. H. Zewail, Nanoscale **10**, 10343 (2018).

Poster No.125

Cooperative action of laser-induced thermal effects and ionic coordination on order of TPPA0 porphyrin derivatives self-assembled interface probed via real-time second harmonic generation

Bin Dong [1,2], Ruipeng Bai [1, 2], Man Xue [1, 2], Caihe Liu [1, 2], Minghua Liu[1, 2], Yuan Guo [1, 2], Zhen Zhang [1,2] *

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In this report, we have performed a real-time second harmonic generation (SHG) to study the cooperative action of laser-induced thermal effects and ionic coordination on the order/disorder of TPPA0 (5,10,15,20-(tetrakis-((ethoxycarbonyl)methoxy) phenyl) porphyrin) monolayers on the interface of three aqueous solutions. For the pure water and $ZnCl_2$ aqueous interfaces, SH signals under the polarization of p-in/p-out declined with time; while on the $CuCl_2$ aqueous interfaces, the SH signals raised with time. It is found that the orientation angle with δ distribution cannot explain the ratios of the SH signal change, thus the gauss orientational distribution width has to be considered instead of δ distribution, which means the TPPA0 monolayers undergone the change process of the orientational order until reaching equilibrium. These results indicated that all of the SH signal changes with time are induced by local accumulated heat with repetitive laser excitation at 82 MHz. The cooperative action of laser-induced thermal effects and the ionic coordination cause the signal change with time, which means the order of the porphyrin derivatives self- assembled interface is affected by the combination of ions and heat. The finding in this work is important to the bioscience, as well as the functional molecular devices.

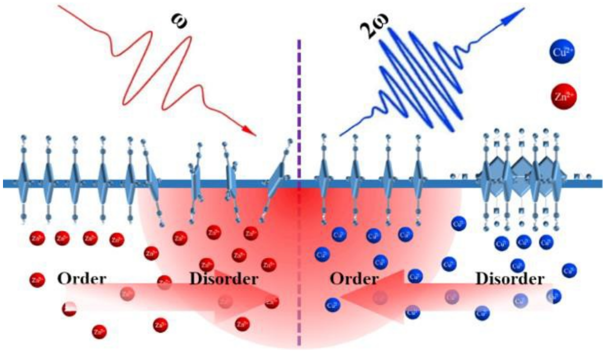

Figure 1. The order and disorder of TPPA0 at the interfaces.

References:

[1] Bin Dong, Ruipeng Bai, Wuming Zheng, Man Xue, Caihe Liu, Minghua Liu, Zhen Zhang, and Yuan Guo, J Phys Chem C, **123**, 11798 (2019).

Poster No.126

Visualizing ultrafast dynamics of nanoparticles in liquid by 4D electron microscopy

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In the past decade, four-dimensional electron microscopy (4D-EM), which enables the direct observation of transient structures, morphologies and carrier transport of materials in real time and space, has attracted increasing interest to the research community due to its powerful capability in the interdisciplines of physics, chemistry, material science, and biology [1-3]. However, the dynamical study by 4D-EM is only limited to solid systems. Here, we present our recent development of liquid-phase 4D-EM and its first application in imaging Brownian dynamics and photochemical reaction dynamics of nanoparticles in liquid on the nanometer-nanosecond time scale [4-6]. Both the translational and rotational dynamics of individual nanoparticles were imaged in both diffusion and ballistic regimes, and a full transition from diffusive to superdiffusive, and further to ballistic rotation was revealed with increasing the asymmetry of the particles. With increasing femtosecond laser fluence, photomorphic reaction dynamics such as agglomeration, coalescence, and fusion dynamics of plasmonic nanoparticles occur due to the surface plasmon excitation and photothermal effect, which were unraveled in real time and space. This advanced liquid-phase 4D EM opens a promising possibility for future study of numerous physical, chemical and biological dynamical processes in native environments.

References:

[1] A. Yurtsever and A. H. Zewail, Science **326**, 708-712 (2009).

[2] B. Barwick and A. H. Zewail et al., Nature **462**, 902-906 (2009) [3] AH. Zewail, Science **328**, 187-193 (2010).

[3] X. W. Fu and B. Chen et al., Science **355**, 494-498 (2017).

[4] X. W. Fu and B. Chen et al., Science Advances **3 (8)**, e1701160 (2017).

[5] X. W. Fu and B. Chen et al., J. Phys. Chem. Lett. 9, 4045–4052 (2018).

Poster No.127

Probing atomic-scale photoexcitation and transient carrier dynamics of rutile $TiO_2(110)$

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Understanding the photoexcitation and related charge dynamics of atomic-scale defects is of great importance for improving the performance of photocatalytic and solar-energy materials1–5. However, the local-environment effect of those atomic defects on the visible photoresponse and electron-hole recombination dynamics is yet to be established. The main challenge arises from that the conventional spectroscopic methods may easily smear out the subtle details of defect states due to the averaging effects. Using a pulsed-laser-combined scanning tunneling microscopy and spectroscopy, here we succeeded to probe the photoexcitation and transient relaxation dynamics of individual oxygen vacancies on rutile $TiO_2(110)$ surface, one at a time, with simultaneously high spatial and temporal resolution. Upon visible-laser illumination, the in-gap states derived from surface oxygen vacancies exhibit prominent downward energy shift accompanied with a suppression of valence tail states, which were attributed to photoexcited electronic transition between the defect levels and conduction band. We found that the photogenerated holes are localized at the defect sites and their lifetime is considerably decreased when the defects are brought close to each other. In contrast, the photogenerated electrons in the conduction band are quite delocalized and less sensitive to the local segregation of the oxygen vacancies. Instead, the lifetime of those electrons is dictated by the defect density fluctuation at a larger spatial scale, which is related to the electron diffusion length. This work can be easily extended to a wide range of photocatalytic and solar-energy materials, thus opening up a new avenue towards time-resolved-atom-resolved carrier dynamics and chemical reaction.

Poster No.128

Many-body effects in photocatalytic dynamics

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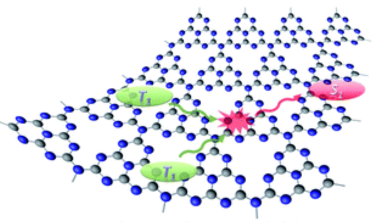
Recently low-dimensional materials hold great potential in the field of photocatalysis, whereas the concomitantly promoted many-body effects have long been ignored. These Coulomb interactions-mediated effects would lead to some intriguing, nontrivial band structures, thus promising versatile photocatalytic performance and optimizing strategies. Here, we give two results to illustrate the role of multibody effects in photocatalytic systems. The one is that ultrathin black phosphorus nanosheets exhibit an exotic, excitation-energy-dependent, optical switching effect in photocatalytic reactive oxygen species generation. The other one is that polymeric g-C_3N_4 matrix exhibit much more complicated excitation kinetics than inorganic materials, and the involved robust excitonic processes have to be taken into consideration



Excitonic Processes in Polymer Matrix of g-C_3N_4

References:

[1] J. Am. Chem. Soc., 2018, **140** (9), pp 3474–3480.

[2] Advanced Materials, 2016, **28**(32): 6940-6945.

[3] Chemical science, **8**(5), 4087-4092.

[4] Chemical Society Reviews, **39**(11), 4206-4219.

Poster No.129

Interracial energy transfer on MXene from ultrafast dynamics investigation to clinical application

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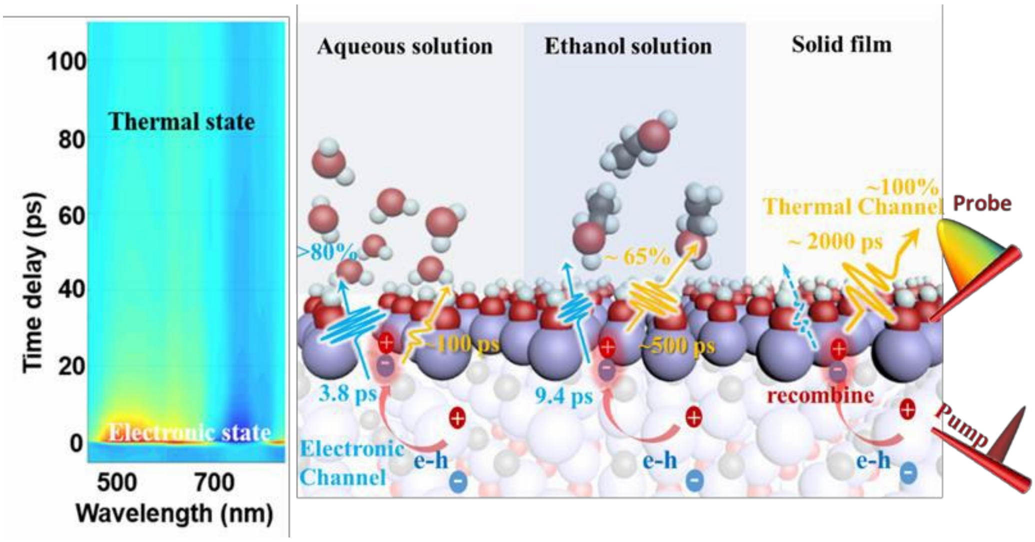
Photothermal conversion of MXene has many applications from biomedical therapy to energy harvesting. Utilization of the energy pathway from photons to surrounding molecules requires the understanding the ultrafast dynamics of absorbed photons. In this talk, for the first time, we present the light induced MXene for broad spectrum anti-bacterial applications. To understand the mechanism, we also performed the ultrafast investigation of the electronic energy dissipation of MXene ($Ti_3C_2T_x$) under plasmonic excitation in three different environments. Our data found that in water, energy dissipation is divided into electronic channel and thermal channel. After ultrafast excitation of MXene, >80% energy quickly dissipate into surrounding solvent within 3.8 picoseconds (ps) as electronic channel. The remaining energy as heat vanish with time constant ~100 ps as thermal channel. Tuning the solvent into ethanol could both narrow the electronic energy dissipation to 35% through electronic channel and slow down the thermal channel (400 ps). In solids, nearly 100% energy relaxation go through thermal channel with the remarkably slower rate (2000 ps). Our results suggested that interfacial interaction is crucial for fast electron-vibration coupling on MXene surface to channel the electronic excitation dissipation, providing important insights into the many biological related applications with MXene.



Poster No.130

Intramolecular singlet fission in a spiro-conjugated dye solution

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Singlet fission is a spin-allowed photophysical process that one singlet exciton can rapidly split into two triplet excitons [1]. The process has the potential to increase the power conversion efficiency of solar cells [2]. Recent years a series of molecules have been revealed to have singlet fission by experiments and Quantum calculations and the role of charge transfer state in the process has motivated significant research effort [3,4]. Here we synthesize a family of spiroconjugation molecules that can occur intramolecular singlet fission. Charge transfer state is observed in these spiroconjugation molecules and the singlet fission rates range from 1.5 ps to 446 ps due to different chemical structures. More importantly, we observe that singlet fission rates of these molecules is getting slower as the polarity of solution increases. These efficient molecules are unprecedented singlet fission materials and ideal model for explore the role of charge transfer state in singlet fission.

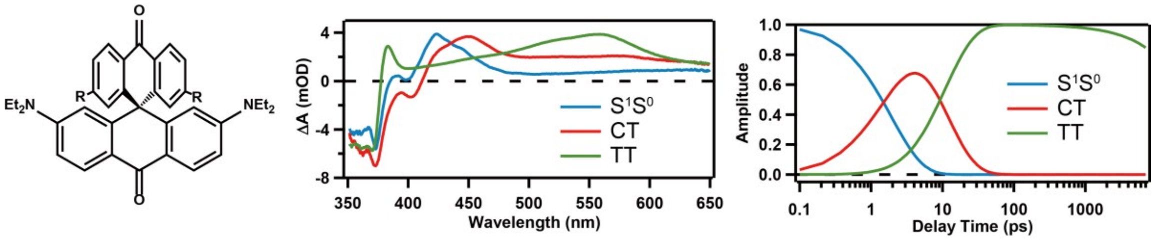

Figure 1. Structures of molecules and species associated spectra.

References:

[1] M. B. Smith and J. Michl, Chemical Reviews **110**, 6891-6936 (2010).

[2] M. C. Hanna and A. J. Nozik, Journal of Applied Physics **100**, 20074510-074510-074518 (2006).

[3] E. A. Margulies, C. E. Miller, Y. Wu, L. Ma, G. C. Schatz, R. M. Young and M. R. Wasielewski, Nature Chemistry, **8**, 1120-1125 (2016).

[4] E. A. Margulies, J. L. Logsdon, C. E. Miller, L. Ma, E. Simonoff, R. M. Young, G. C. Schatz and M. R. Wasielewski, Journal of the American Chemical Society, **139**, 663-671 (2017).

Poster No.131

Excited-state proton transfer of a cationic superphotoacid in an AOT reverse micelle

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Proton transfer is an omnipresent reaction in a wide range of scientific fields. Because the dynamics of proton-transfer reactions strongly depends on surroundings, it has been extensively studied in various systems spanning homogeneous to heterogeneous media. Among those, reverse micelles (RMs) are simplest experimental systems mimicking water pockets in biology to uncover reactions in biologically relevant environment. The RMs consist of a hydrophilic structure interfacing with a water nanopool in the core and the hydrophobic phase outside. The core is usually divided into two regions as bound water and free water. The interactions at the interface result in higher viscosity and rigidity in the bound water region than bulk water. The free water region is distant from the interface that its influence becomes weaker therein. This structural feature induces a heterogeneous polar confined environment with radial, dielectric and viscosity gradients in the nanopool.

Here, we chose a cationic probe, N-methyl-7-hydroxyquinolinium ($NM7HQ^+$), to investigate the confined water and study its effect on acid-base reactions. This probe changes its charge state from positive to neutral (or zwitterionic) upon photoexcitation via the proton transfer [1-4]. Therefore, ionic interaction at the interface may dramatically change during the photocycle. We measured time-resolved emission spectra (TRES) of $NM7HQ^+$ in AOT RMs using the time-correlated single photon counting and fluorescence upconversion techniques. Three bands originated from the excited cationic form, the keto form, and an Intermediate were effectively deconvoluted from TRES by fitting them with lognormal peak functions [5]. By following the time-dependent changes of peak intensity and position for each deconvoluted band, we studied the chemical kinetics of the ESPT and the effect of hydration in the confined environment on the ESPT dynamics.

References:

[1] Y.-J. Kim, et al., Chem. Eur. J. **23**, 17179 (2017).

[2] S.-Y. Park, et al., Chem. Eur. J. **22**, 4340 (2016).

[3] S.-Y. Park, et al., Phys. Chem. Chem. Phys. **18**, 24880 (2016).

[4] Y. M. Lee, et al., Methods Appl. Fluoresc. **4**, 024004 (2016).

[5] M. L. Horng, et al., J. Phys. Chem. **99**, 17311 (1995).

Poster No.132

Pressure manipulation of exciton dynamics in few-layer MoS_2

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[2]Institute of Atomic and Molecular Physics, Jilin University, Changchun, China

Transition metal dichalcogenides have recently attracted great interest as an important class of two-dimensional semiconductors because of their distinctive electronic, optical and catalytic properties. MoS_2 has a layered structure and different layers are combined by weak van der Waals forces. Theoretical and experimental studies on structure and bandgap changes of atomically thin few-layer MoS_2 under pressure have shown that when pressure is applied, the distance between different layers will decrease, which will enhance the interlayer coupling effect and thus significantly changes the structure and bandgap [1]. Here we demonstrate the first experimental study on ultrafast excited state dynamics of few-layer MoS_2 under high pressure by means of broadband transient absorption spectroscopy combined with diamond anvil cell devices (DAC). As shown in Fig 1, with the pressure increases, the A and B exciton peaks are dramatically blue shifted, and both of their decay dynamics get faster accordingly.

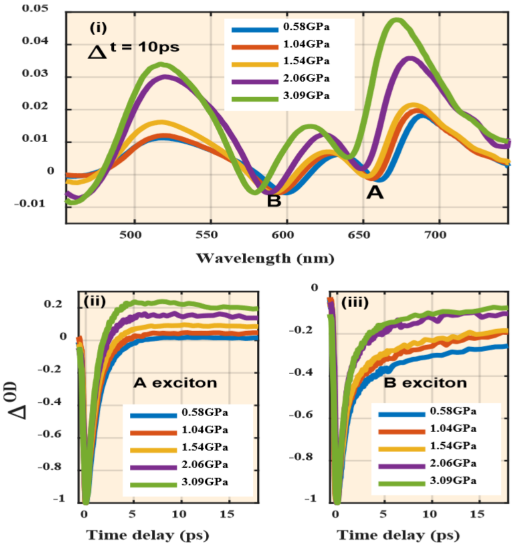

Figure 1. (i) Transient absorption (TA) spectra at time delay of 10ps under different pressures (ii) (iii) Normalized TA kinetics of A and B excitons at different pressures.

References:

[1] X. Dou, K. Ding, D. Jiang, X. Fan, B. Sun, ACS Nano **10**, 1619 (2016).

Poster No.133

Real-time observation of amyloid formation at biomembrane surface by ultra-sensitive femtosecond sum frequency generation vibrational spectroscopy

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Amyloid formation has been implicated in many fatal diseases, but its mechanism remains to be clarified due to a lack of effective methods that can capture the early formed intermediate species due to their transient nature. Especially, the aggregation process at negative charge phospholipid surface is very fast, which makes the identification of transient intermediates be more challenging. Resolving these problems requires developing the method that can quickly probe the time-dependent structural changes during the aggregation process with sufficient structural and temporal resolutions. In this study, we verify experimentally that the combination of interface-sensitive chiral amide I, achiral amide II and amide III spectral signals of the protein backbone generated in femtosecond sum frequency generation vibrational spectroscopy (SFG-VS) can provide a unique and powerful tool to capture amyloid intermediates at the interface during the aggregation process with sufficient structural and temporal resolutions. Such a combination can directly identify the formation of β-hairpin- like monomers, and β-sheet oligomers, and fibrils. A strong correlation between the amide II signals and the formation of β-sheet oligomers and fibrils was found. With this approach, the structural evolution of human islet amyloid polypeptides (hIAPP) at negative lipid bilayers was elucidated. It was firmly confirmed that hIAPP populates through β-sheet conformers without involving α-helical intermediates. The membrane-associated assembly of hIAPP proceeds by assembling with a β-hairpin-like monomer at the lipid bilayer surface, rather than by inserting the preassembled β-sheet oligomers in solution. This newly established protocol is ready to be utilized in revealing the mechanism of amyloid aggregation at the lipid membrane.

References:

[1] J.J. Tan, J.H. Zhang, Y. Luo, and S.J. Ye. *J. Am. Chem. Soc.* **141**, 1941(2019).

Poster No.134

Heterogenous dynamics in self-assembled materials revealed by transient VSFG microscopy

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We report, for the first time, observations of mesoscopically homogeneous but macroscopically heterogenous water dynamics in self-assembled materials by a new, spatially resolved infrared (IR) pump vibrational sum frequency generation (VSFG) probe microscope. Using this new technique, we spatially resolved dynamics of water bounded by host-guest, self-assembled sheets comprised of sodium dodecyl sulfate (SDS) and β-cyclodextrin (β-CD). We found that the strong hydrogen-bond interactions between β-CD and nearby water not only template nearby water networks to adopt the local ordering of the self-assemblies, but also allow resonant energy transfer from β-CD to nearby water. More interestingly, the resonant energy transfer dynamics are heterogeneous among domains, while remaining uniform within domains. This surprising result indicates that the water near self-assembled materials can be templated uniformly across micron domains. Because SDS@2β-CD is a synthetic capsid-like self-assembled material, similar mesoscopic ordering of water structure and dynamics could also exist in biological soft materials. The advancement of adding spatial resolution to ultrafast molecular vibrational spectroscopy opens a new way to probe mesoscopic molecular structure ordering and relaxation dynamics in biological systems, and hydro-responsive self-assembly materials for micro-optics and electronics.

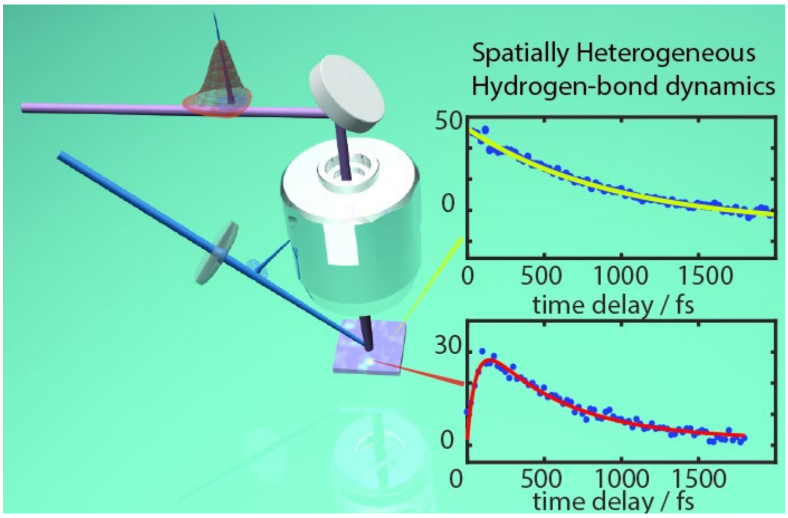

Figure 1. Schematic of the spatially resolved IR pump VSFG probe microscopy system and two representative types of vibrational dynamics from different domains.

Poster No.135

Ultrafast charge generation and nongeminate recombination in non-fullerene OPV blends

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Organic solar cells with non-fullerene acceptors develop rapidly in the last decade [1], and the PCE has exceeded 15 % recently [2]. Free charge generation is essential for photon current conversion in OPV devices [3]. Recent polymer/non- fullerene materials show efficient charge generation despite the low driving force, while the intrinsic mechanism is still unclear. Here, we study the free charge dynamics in the state-of-the-art system PM6/Y6 [2] utilizing transient absorption method (Fig. 1a). After exciting the acceptor, the ESA signal of free charges comes up within 50 ps, suggesting efficient charge separation after hole transfer (Fig. 1b). Furthermore, the charge generation and recombination processes strongly depend on the pump fluences even at excitation densities similar to solar illumination (Fig. 1c, d), showing the important role played by nongeminate recombination.

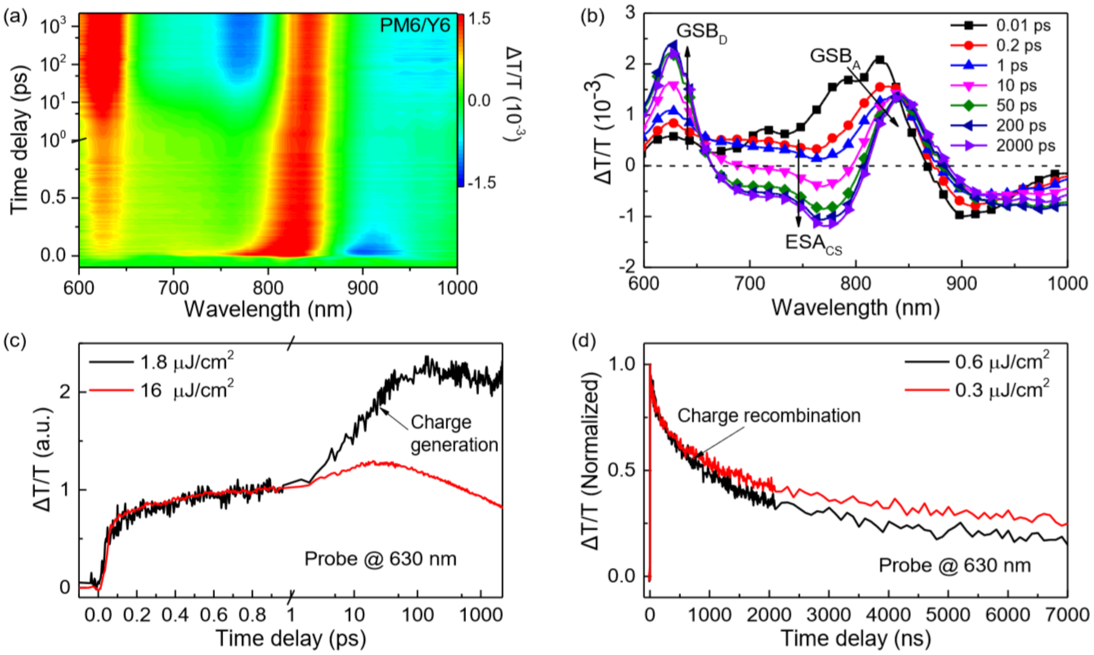

Figure 1. (a, b) TA signal recorded from a PM6/Y6 blend after pump at around 750 nm. (c, d) Charge dynamics at different pump fluences.

References:

[1] H. Bin, C. Zhang, Z. Zhang et al., Nature Communications **7**, 13651 (2016).

[2] J. Yuan, Y. Zhang, Y. Zou et al., Joule **3**, 1140 (2019).

[3] A. Bakulin, A. Rao, R. Friend et al., Science 335, 1340 (2012).

Poster No.136

Accelerated exciton decay behavior in Mn-doped $CsPbCl_3$ nanocrystals revealed by modified femtosecond transient absorption spectroscopy

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Cesium lead halide ($CsPbX_3$) nanocrystals (NCs) has been extensively studied by researchers for the excellent optical properties such as tunable emission color, narrow band width[1-2]. Furthermore, Mn-doped $CsPbCl_3$ has attracted most attention for its efficient energy transfer between host excitons and doped Mn ions which indicates some probable useful applications such as white light LED, solar concentrators[3-4]. Herein, we conducted pump-pump-probe transient absorption experiments with PL spectroscopy on the Mn-doped and undoped $CsPbCl_3$ NCs and find an accelerated recombination time of band-edge exciton about 10ps when the Mn ions is excited compared 3ns lifetime of host of excitons without Mn ions excited. This could shed some light of avoiding the disadvantage and improving the performance of the light emitting devices based on doping functional ions.

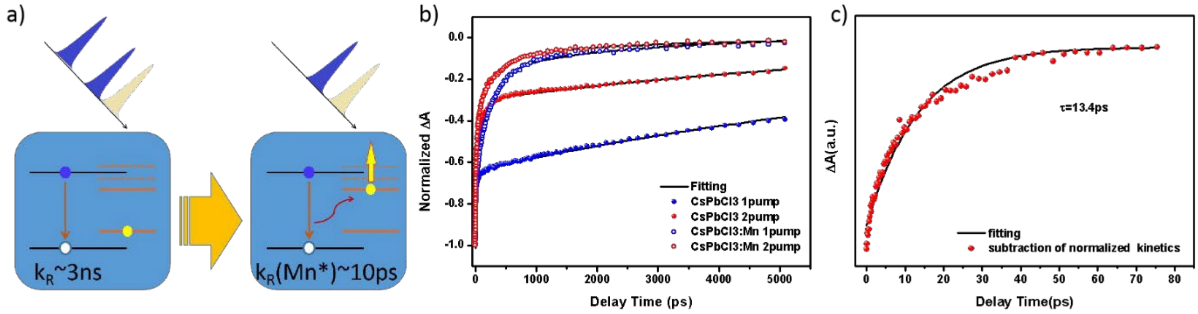

Figure 1. a) pump-pump-probe strategy illustration and exciton kinetics in Mn-doped NCs b) undoped and Mn-doped NC bleach peak kinetics of single and double pump-probe experiment. c) subtraction kinetics of double pump and single pump experiments

References:

[1] L Protesescu, M. V. Kovalenko. Nano Lett **15**, 3692 (2015).

[2] J Song, H Zeng. Adv Mater **27**, 7162 (2015).

[3] W Liu, V. I. Klimov. J Am Chem Soc **138**, 14954 (2016).

[4] D Parobek. D. H. Son. Nano Lett **16**, 7376 (2016).

Poster No.137

Charge carrier dynamics on graphitic-C_3N_4 with copper phosphide modification

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Photocatalytic H_2 production on Copper phosphide (Cu_3P) combined with graphitic carbon nitride (g-C_3N_4) under visible light ($\lambda > 420$ nm) has been investigated for the first time. The Cu_3P/g-C_3N_4 composite shows a superior visible- light-driven photocatalytic performance for H_2 evolution from water. Compared with pristine g-C_3N_4 (0.75 μmol h^{-1} g^{-1}) and Cu_3P (3.44 μmol h^{-1} g^{-1}), the photo-induced carrier separation efficiency of Cu_3P/g-C_3N_4 (277.2 μmol h^{-1} g^{-1}) was greatly enhanced. Cu_3P-induced electron redistribution promoted excited state electron transfer, and the trapping process can be suppressed effectively. Time-resolved spectroscopy experiments were carried out to study the photo-physical characteristics of g-C_3N_4 and Cu_3P hybrid from femtosecond to nanosecond time domains. The ultrafast time-resolved transient absorption results of the Cu_3P/g-C_3N_4 hybrids indicate an ultralow recombination process of the photoelectron-hole in an artificial photo-utilization system. The enhanced lifetime of the photo-induced active electrons was measured to be 209 ps by femtosecond time-resolved transient absorption experiments in the Cu_3P/g-C_3N_4 hybrid system. The possible photocatalytic mechanism for Cu_3P/g-C_3N_4 hybrid is proposed (Figure 1). Among these advantages, such new Cu_3P/g-C_3N_4 composite is a promising alternative material for solar-to-chemical energy conversion reactions.

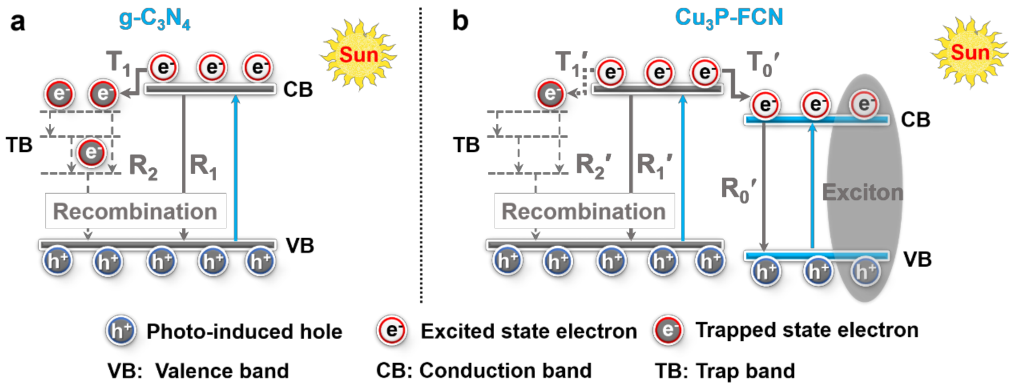

Figure 1. Mechanism of photo-induced electrons transfer and charge recombination routes: (a) g-C_3N_4; (b) Cu_3P-FCN under visible-light irradiation.

Poster No.138

Triplet energy transfer across the inorganic/organic interface for photochemical photon upconversion

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In recent years, sensitization of molecular triplets using inorganic semiconductor nanocrystals (NCs) *via* triplet energy transfer (TET) has attracted tremendous attention due to its potential application in photochemical photon upconversion, which can alleviate transmission losses in solar energy conversion. Yet the mechanism of TET across the inorganic/organic interface remains poorly understood. Many seemingly contradictory results have been reported, mainly because of the complicated trap states in NCs and the ill-defined relative energetics between NCs and molecules used in these studies.

Here we provide a unified picture for TET by performing ultrafast spectroscopic studies on a model system comprising lead halide perovskite NCs with very low surface trap densities as the triplet donor and polycyclic aromatic hydrocarbons (PAHs) which either favour or prohibit charge separated states as the triplet acceptors. Hole transfer from NCs to tetracene is energetically favoured, and hence TET proceeds via a charge separated state; In contrast, hole transfer to naphthalene is forbidden and TET is mediated by a virtual charge transfer state.[1] the second case, the TET rates approximately scales linearly with the square of exciton wavefunction at NC surfaces which increases with decreasing NC sizes.[2,3] Guided by these fundamental principles, we demonstrate a visible-to-UV upconversion efficiency of ~10% using a small-size NCs-naphthalene system.[4]

References:

[1] X. Luo, Y. Han, Z. Chen, Y. Li, G. Liang, and K. Wu, Submitted.

[2] X. Luo, R. Lai, Y. Li, Y. Han, G. Liang, X. Liu, T. Ding, and K. Wu, J. Am. Chem. Soc.**141**, 4186 (2019).

[3] Y. Han, X. Luo, R. Lai, Y. Li, G. Liang, and K. Wu, J. Phys. Chem. Lett. **10**, 1457 (2019).

[4] S. He, Y. Li, Y. Han, and K. Wu, Submitted.

Poster No.139

Ultrabright 300 keV photoelectron gun designed for single-shot ultrafast electron diffraction

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Due to the high scattering cross section of the electron-atom interaction, Ultrafast electron diffraction (UED) is a valuable technique to study ultrathin samples or surfaces at the atomic level [1,2]. So far, one of the major limitations of UED is challenging to reach a large flux and high temporal resolution simultaneously due to the of space charge effect [1,2]. To reduce such effect, MeV photoelectron RF gun have been available with a complicated and costly configuration [3,4]. As an alternative option, here we propose a 300 keV DC photoelectron gun which may effectively avoid the issues of MeV photoelectron gun, meanwhile, maximize the relativistic effect to eliminate the space charge effect, as shown in Figure 1(a). Simulations are performed with the GPT code. Results show that electron pulse duration is within 200 fs(FWHM) even if the electron number reaches 10^5, where the 70 μm(FWHM) laser spot size and the 150 fs (FWHM) laser pulse duration are used(shown in Figure 1(b)). With such an ultrabright UED technique, irreversible processes such as chemical reactions may become accessible.

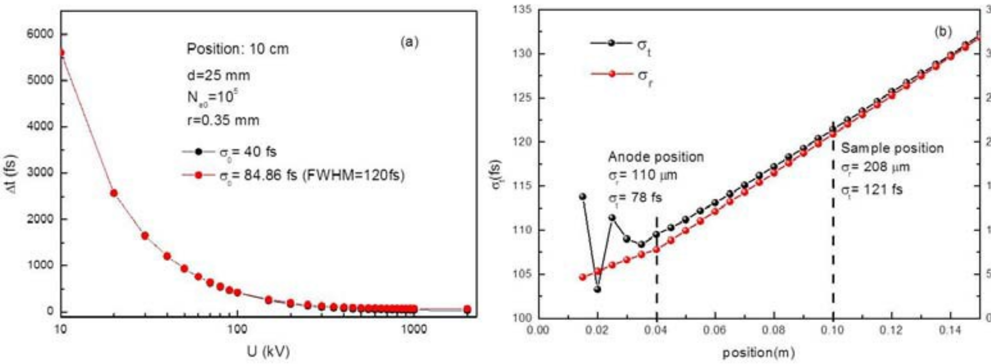

Figure 1. (a) duration of electron pulse vs energy of electrons, (b) spot size and duration of electron pulse vs distance from cathode (Simulations with GPT codes).

References:

[1] M. Chergui and A. H. Zewail, Chem. Phys. Chem. **10**, 28(2009).

[2] R. J. D. Miller, Science, **343**,1108–16(2014).

[3] X. J. Wang, D. Xiang, T. K. Kim and H. Ihee, J. Korean Phys. Soc. **48**, 390(2006).

[4] L. Zhao, Z. Wang, C. Lu, et al., Phys. Rev. X **8**,021061(2018).

Poster No.140

Ultrafast spectroscopic study on the luminescence mechanism of metal nanoclusters

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Metal nanoclusters (MNCs) with attractive properties including brightness, photostability and biological compatibility have been very actively investigated. However, a fundamental understanding about the origin of photoluminescence remains unclear which largely limits their further applications. Here, the excited state dynamics of MNCs was elaborately investigated with the help of time-resolved spectroscopy. The fluorescence/phosphorescence channel of MNCs could be regulated at room temperature by a simple solvent-stimulated strategy. The improvement of optic performance was assigned to the configuration optimization of the ligands on the interface of MNCs. The aggregation of surface carboxylate ligands on the metal core, that is, the cluster formation of carbonyl groups, activates the phosphorescence channel. These results should stimulate additional experimental and theoretical studies and could benefit the molecular- level design of luminescent MNC for optoelectronics and other applications.

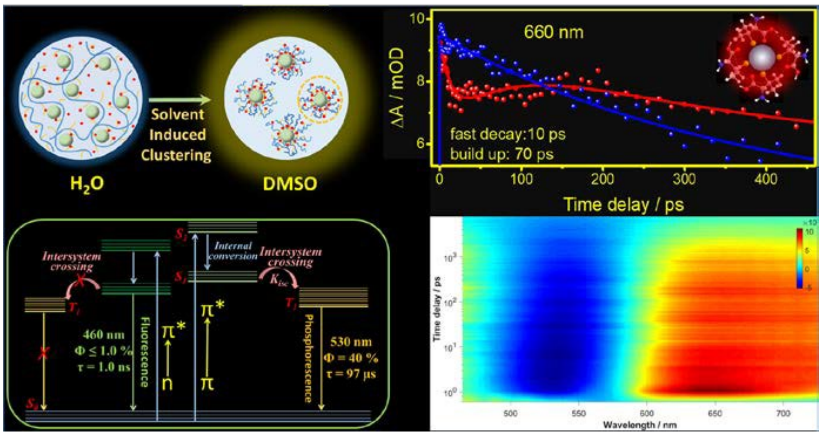

Figure 1. The dynamic behavior of excited states of MNCs.

References:

[1] Y. Chen, T. Yang, H. Pan, Y. Yuan, L. Chen, M. Liu, K. Zhang, S. Zhang, P. Wu, J. Xu, J. Am. Chem. Soc. **136** 1686-1689(2014).

[2] T. Yang, S. Dai, S. Yang, L. Chen, P. Liu, K. Dong, J. Zhou, Y. Chen, H. Pan, S. Zhang, J. Chen, K. Zhang, P. Wu, J. Xu, J. Phys. Chem. Lett. **8**, 3980-3985(2017).

Poster No.141

Femtosecond SFG visualized hydroxide ion in macroscopically hydrophobic Teflon matrix

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Surface charge of insulating materials is ubiquitous in our lives, yet its underlying mechanism is still poorly understood. Different debated mechanisms have been proposed for the dominating type of charge species that govern the charging behavior, ranging from electrons to ions, to radicals, to bits of material [1-4]. Recently, some people also hypothesized that the OH^- ions dissolved in the water adsorbed to the surface of polar solids would be reasonable charge carriers. In terms of this hypothesis, the interaction between water and insulating materials has been used to generate power [5,6] and water droplet transport [7]. However, to the best of our knowledge, there still lacks a molecular evidence to verify the existence of hydroxide ion on the surface or in the matrix of insulating materials. In this study, we visualized the hydroxide ion in the matrix of macroscopically hydrophobic Teflon for the first time by using femtosecond sum frequency generation vibrational spectroscopy. It is found that the hydroxide ion (OH^-) is present inhomogeneously both at the surface and in the matrix of Teflon. Femtosecond free induction decay experiments suggested that the free OH oscillators dephase on the time of ~100 fs, whereas the hydroxide ion dephases more slowly (~300 fs), indicating the interaction of hydroxide ion with Teflon is stronger than the interaction of free OH oscillators with Teflon. These findings shed light on the origination of the net negative charge on Teflon surface, which may help us better understand the basic principle of charging of insulating materials, and guide us to develop practical applications where this sort of charging plays a role.

References:

[1] D. J. Lacks, and R. M. Sankaran. J. Phys. D: Appl. Phys. **44**, 453001 (2011).

[2] L. S. McCarty, and G. M. Whitesides. Angew. Chem., Int. Ed. **47**, 2188 (2008).

[3] H. T. Baytekin, B. Baytekin, T. M. Hermans, B. Kowalczyk, and B. A. Grzybowski. Science **341**, 1368 (2013).

[4] H. T. Baytekin, B. Baytekin, J. T. Incorvati, and B. A. Grzybowski. Angew. Chem., Int. Ed. **51**, 4843 (2012).

[5] Y. Sun, X. Huang, and S. Soh. Chem. Sci. **6**, 3347 (2015).

[6] Z. H. Lin, G. Cheng, S. Lee, K. C. Pradel, and Z. L. Wang. Adv. Mater. **26**, 4690 (2014).

[7] Q. Sun, D. Wang, J. Zhang, Y. Li, S.J. Ye, J. Cui, L. Chen, Z. Wang, H.J. Butt, V. Doris, and X. Deng. arXiv preprint, arXiv:1901.02612 (2019).

Poster No.142

Direct observation of local and charge-transfer exciton hybridization in high-efficiency organic solar cells

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The recent efficiency boost in non-fullerene acceptor-based organic solar cells (OSCs) benefits from efficient charge generation at a small driving force together with high electroluminescence yield at donor-acceptor interfaces [1-4]. One major mystery in the community is whether and how charge-transfer (CT) states exist in OSCs with low interfacial energetic offsets, as highly sensitive external quantum efficiency does not distinguish the CT states from the local exciton (LE) states. Here, in benchmark low-offset donor: acceptor blends exhibiting a slow (picosecond) hole transfer, we observe ultrafast formation of free charges when directly exciting the absorption tail. Our experiment confirms the existence of CT states in state-of-the-art OSCs and provides direct evidence of the hybridization of LE and CT states, forming the paradigm of new generation low-offset OSCs.

References:

[1] D. Qian, Z. Zheng, H. Yao, W. Tress, T. R. Hopper, S. Chen, S. Li, J. Liu, S. Chen, J. Zhang, X.-K. Liu, B. Gao, L. Ouyang, Y. Jin, G. Pozina, I. A. Buyanova, W. M. Chen, O. Inganäs, V. Coropceanu, J.-L. Bredas, H. Yan, J. Hou, F. Zhang, A. A. Bakulin, F. Gao, Nat. Mater. **17**, 703(2018).

[2] F. D. Eisner, M. Azzouzi, Z. Fei, X. Hou, T. D. Anthopoulos, T. J. S. Dennis, M. Heeney, J. Nelson, J. Am. Chem. Soc. **141**, 6362(2019).

[3] G. Han, Y. Yi, J. Phys. Chem. Lett. 2911(2019).

[4] X.-K. Chen, V. Coropceanu, J.-L. Brédas, Nat. Commun. **9**, 5295(2018).

Poster No.143

How MXene (Ti_3C_2) convert different wavelength photon into thermal energy –an ultrafast view

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Efficient solar energy conversion through Nano-materials is a promising route to solve environment and energy issues. MXene, two-dimensional transition metal carbides has outstanding performances in the field of solar energy conversion. Ti_3C_2Tx as a type of MXene has been reported with internal 100% photo-to-thermal conversion efficiency [1]. Unique strong absorption in the UV (Ultraviolet) and NIR (near-infrared) region maybe can convert solar energy effectively. We results show MXene including two times constant are ~4ps and ~21ps by ultrafast spectroscopy. Contrasting with different excited wavelength indicate MXene have better energy conversion efficiency under UV than NIR. By comparing absorption differential spectra, we ensure UV would heat MXene to ~333K and NIR would heat MXene to ~313K at same laser intensity.

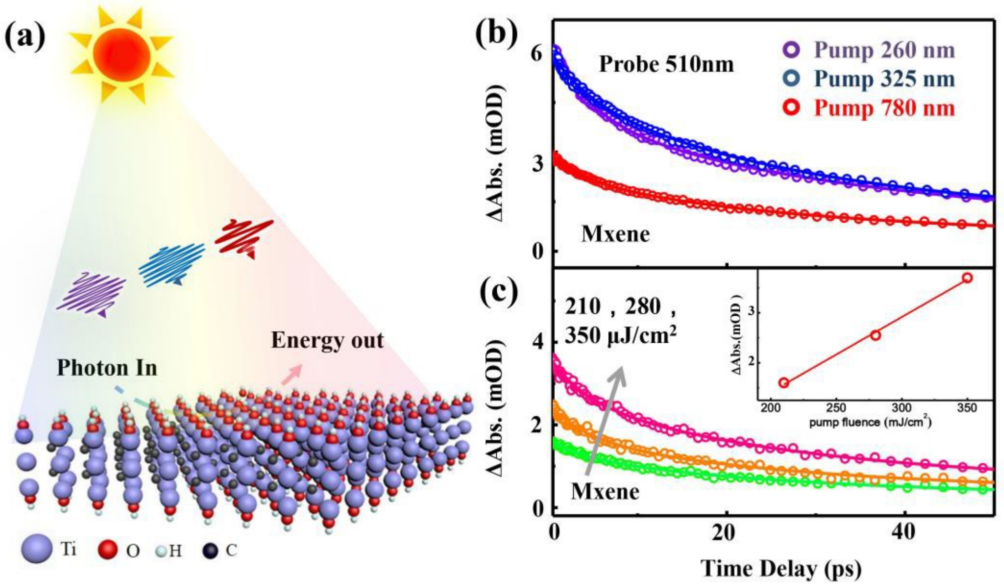

Figure 1. (a) Schematic diagram of energy conversion with different photo-excitation wavelength (b) Time-dependent photo-induce absorption change at 510nm under excitation at 260nm, 325nm, and 780nm. (c) Temporal evolution of MXene energy relaxation at 510nm with different pump fluence.

References:

[1] Li, R. Zhang, L. Shi, L. Wang, P, ACS Nano**, 11** (4), 3752-3759(2017).

Poster No.144

Pressure-induced tunable electron transfer and auger recombination rates in CdSe/ZnS quantum dot-anthraquinone complexe

Huifang Zhao[1], Hang Yin[1], Xiaochun Liu[1], Hui Li[1], Ying Shi[1,*], Cailong Liu[1], Mingxing Jin[1], Jianbo Gao[2], Yi Luo[3] and Dajun Ding[1]

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Promoting electron transfer (ET) process and suppressing Auger recombination (AR) in quantum dots (QDs) are crucial issues in the advance of technologies for QD-based devices. In this report, we use in situ high-pressure ultrafast transient absorption spectroscopy to explore the impact of pressure on the ET process between CdSe/ZnS and anthraquinone (AQ) and AR process in CdSe/ZnS dissolved in cyclohexane (CHX). Remarkably, it is observed that a novel saltation occurs in ET and AR lifetime at particular pressure points, corresponding to transformation of CHX crystal structure. Importantly, using this method, simultaneous shortening of ET process and prolongation of AR are achieved in a suitable ambient pressure range, which has a direct impact to develop the efficient solar-to-fuel conversion. This study highlights a brand-new approach for tuning ET and AR process in QDs and provides general guidelines for the development of new route towards the application of QDs.

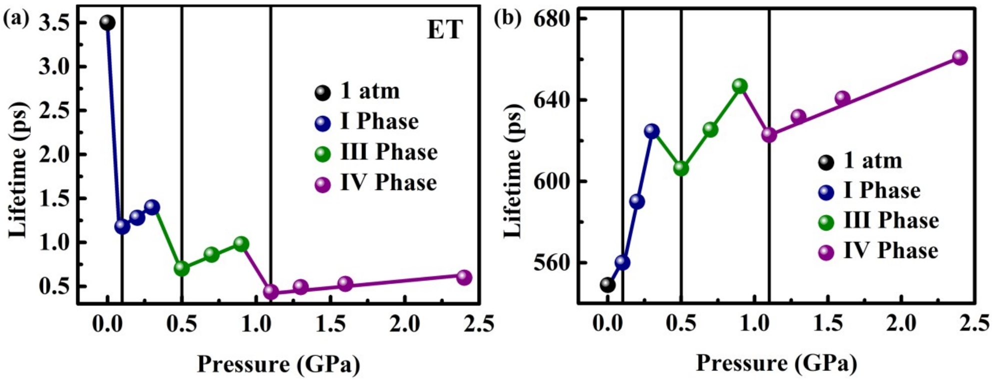

Figure 1. The dependence of pressure on the lifetime of ET process (a) and AR process (b).

Poster No.145

Capturing coherent evolution of excitons and polarons in $CH_3NH_3PbI_3$

Jha[1], H.G. Duan[1,2,3], V. Tiwari[1,4], G.R. Berdiyorov[5], A. Akimov[6], O. Vendrell[7], P.K. Nayak[8], H.J. Snaith[8], M. Thorwart[2,3], Z. Li[1], M.E. Madjet[5] & R.J.D. Miller[1,3,9]

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Hybrid organic-inorganic perovskites has gathered much attention owing to their unprecedented success in photovoltaics. To unravel the secrets to this success, we have studied the dynamics after primary excitation process of lead halide perovskites using heterodyne-detected transient grating and two-dimensional spectroscopy. We distinctly capture the excitons' signature, which are weakly coupled to phonons [1,2]. Additionally, we distinctly observe the ground and excited state vibrational modes corresponding to organic and inorganic sub-lattices. The interplay of strongly coupled dominant vibrational modes to ultrafast carrier generation process will be discussed.

References:

[1] A. Jha, H.-G. Duan, V. Tiwari, P. K. Nayak, H.J. Snaith, M. Thorwart, and R. J. D. Miller, ACS Photonics **5**, 852 (2017).

[2] V. Tiwari, H.-G. Duan, A. Jha, P. K. Nayak, M. Thorwart, H.J. Snaith, and R. J. D. Miller, EPJ Web of Conferences **205**, 06018 (2019).

Poster No.146

Femtosecond laser driven terahertz current and terahertz driven local spin-wave modes

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THz radiation falls between infrared and microwave radiation in the electromagnetic spectrum and can be used for a different types of material characterization. (1) Using terahertz emission spectroscopy, we probe the elementary spin dynamics in ferromagnetic metal on a femtosecond timescale. The interaction of light and matter on ultrafast time scales aims to develop new concepts for the generation of THz radiation. (2) Using terahertz time-domain spectroscopy, we report on the coherent control of terahertz spin waves in canted antiferromagnet rear-earth orthoferrites and observe a Dicke cooperativity in magnetic interactions.

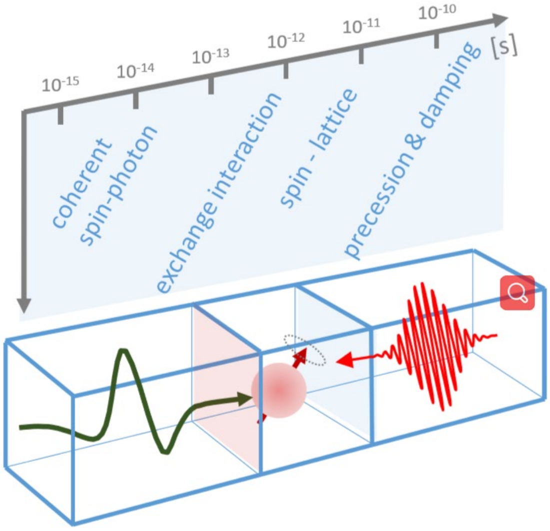

Figure 1. The schematic of femtosecond laser driven THz current and THz driven local spin-wave modes.

References:

[1] Zuanming Jin, et al., Nature Physics **11**, 761 (2015)

[2] Zuanming Jin, et al., Phys. Rev. B **87**, 094422 (2013)

[3] Shunnong Zhang, et al., Journal of Physics D: Applied Physics **51**, 034001 (2018)

[4] Xinwei Li, et al., Science **361**, 794 (2018)

[5] Shunnong Zhang, et al., Optics Letters **42**, 3080 (2017) Zuanming Jin, et al., Phys. Status Solidi RRL doi: 10.1002/pssr.201900057 (2019)

Poster No.147

Charge transport in 2D layered perovskites mediated by molecular conformation of its organic cation

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An in-depth understanding of the interplay between the microscopic structure and macroscopic optoeleronic properties of two-dimensional hybrid metal−halide perovskites (2DHPs) is essential to design new materials and improve device performance. Unfortunately, how exactly the sizes of organic cation impact their structure and their optoelectric function is not completely understood. Some studies suggest that the charge transport property is determined by the interlayer distance while others claim that it is controlled by both distance and barrier height/structure. Here, we investigate the charge carrier transport, interlayer distance and barrier structure of 2DHPs with various organic cation lengths using the combination of time-resolved optical-pump terahertz-probe (OPTP) spectroscopy, X-ray diffraction (XRD) and in situ sum frequency generation vibrational spectroscopy (SFG). Our results show that the high degree of molecular-level order for the $[PbI_6]^{4-}$ bound alkyl ligands given by SFG, leads to the increase of carrier mobility, demonstrating the significance of the structural order in enhancing charge transport and improving device performance in halide perovskites. In addition, we also found that the molecular conformation of P3ATs controls the hole-extraction ability from perovskites at the P3ATs/perovskites heterojunctions.

References:

[1] M. Yu, C. Yi, and N. Wang. Adv. Opt. Mater. **7**, 1801575 (2019).

[2] C. Quarti, N. Marchal, and D. Beljonne. J. Phys. Chem. Lett **9**, 3416 (2018).

[3] B. Yang, W. Ming, and M. Du. Adv. Mater **30,** 1705801 (2018).

[4] M. T. Frederick, J. L. Achtyl, and K. E. Knowle. J. Am. Chem. Soc. **133,** 7476 (2011).

Poster No.148

Coherent formation of polaron pairs in all-polymer photovoltaic blends

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N2200 has been regarded as a prominent polymer acceptor in all-polymer organic photovoltaic (OPV) devices [1]. Two excited states named polaron pair state (iPP) and exciton state (EX) can be generated simultaneously in N2200. However, only iPP mediated hole transfer is found in J51/N2200 all-polymer photovoltaic blends (Fig.1 a-c), while the EXs act as a spectator [2]. To control the iPP dynamics in the blend, it is of great significance to understand the formation process of iPP and EX in N2200. In this work, a superposition state of EX and iPP is observed utilizing ultrafast TA (temporal resolution <10 fs) in N2200 film, which then decoherent into EX and iPP states respectively in about 100 fs accompanied by vibrational oscillations (Fig.1 d&e). Furthermore, two-dimensional electronic spectroscopy (2DES) results of N2200 suggest that vibronic coupling between iPP and EX may play an important role in the initial photophysics of N2200 (Fig.1 f&g) [3].

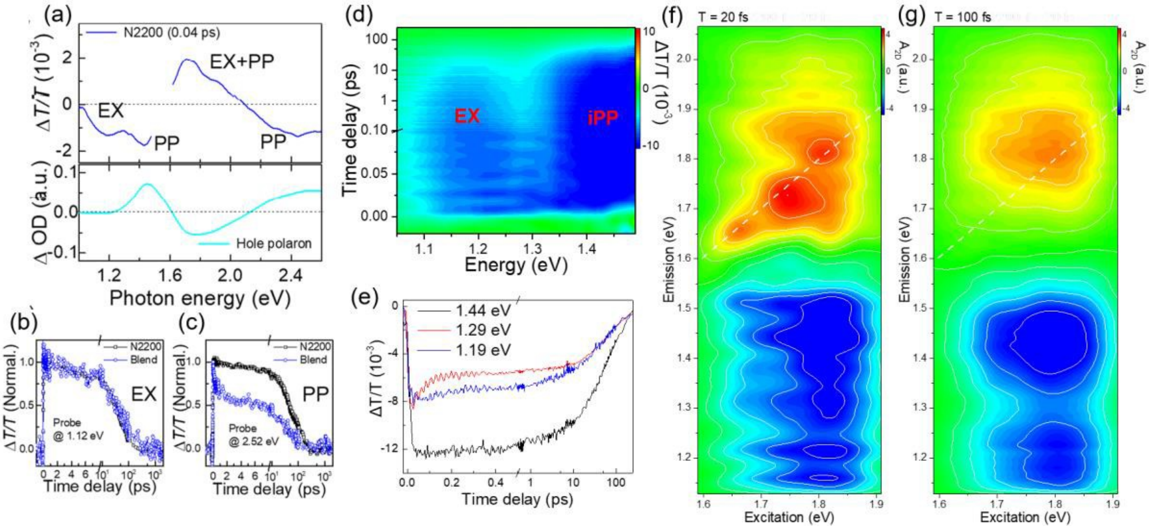

Figure 1. (a) TA spectra and spectro-electrochemistry measurements of neat N2200. (b, c) TA dynamics of the ESA bands in neat N2200 and blends. (d, e) Ultrafast TA of neat N2200. (f, g) 2DES results of neat N2200.

References:

[1] L. Gao, Z. Zhang, L. Xue et al., Advanced Materials **28**, 1884 (2016).

[2] R. Wang, Y. Yao, C. Zhang et al., Nature Communications **10**, 398 (2019).

[3] W. Zhu, R. Wang, C. Zhang et al., Optics Express **25**, 21115 (2017).

Poster No.149

Ultrafast dynamics of MoS_2 sheet studied by time-resolved spectroscopic photoemission electron spectroscopy

Yu Liang [1], Bo-han Li[1], Ziling Li[1], Guanhua Zhang[1], Julong Sun[1], Chuanyao Zhou[1], Yu Ye[2], Xueming Yang[1], Zefeng Ren[1, *]

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The advanced semiconductor technology is pushing the transistor to be smaller and faster, which demands us to understand carrier dynamics in semiconductor heterojunction in both space and time with high resolution. Recently, we developed ultrafast two-color pump-probe technique combining the spectroscopic photoemission electron microscopy (PEEM), and have realized both time resolution in femtosecond time scale and spatial resolution. Inhomogeneous ultrafast dynamics of electron yield and surface photovoltaic effect of MoS_2 sheet have been measured, and three different dynamical processes of carrier relaxation have been quantitatively analyzed. Energy-dependence, excitation power- dependence and MoS_2 size-dependence of ultrafast carrier dynamics have been systematically investigated. Our study will enhance our understanding on the photoelectric characters of 2D materials and shed light on the manufacture of high- performance optoelectronic devices.

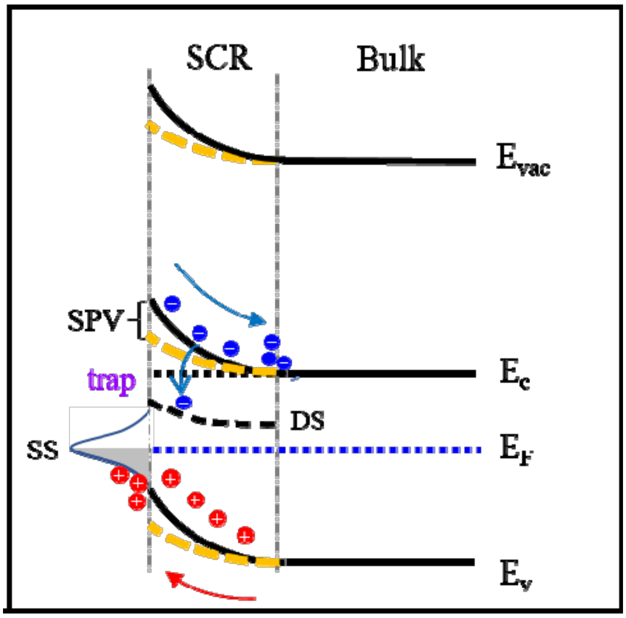

Figure. 1 schematic of ultrafast SPV dynamics and carrier relaxation process.

Poster No.150

Photoexcitation dynamics in perovskite solar cells

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Advancing organo halide perovskite solar cells requires understanding of carrier dynamics. Electron−hole recombination is a particularly important process because it constitutes a major pathway of energy and current losses. Non- adiabatic molecular dynamics combined with ab initio real-time time-dependent density functional theory enable us to model time-resolved laser experiments in real time and at the atomistic level, emphasizing realistic aspects of the materials, such as defects, dopants, boundaries, humidity, chemical bonding, etc. In this talk, we will discuss the factors influencing interfacial electron transfer and electron-hole recombination in hybrid inorganic-organic perovskites and all-inorganic perovskites. [1-10] The predicted changes in the charge and energy transfer and relaxation processes arise due to a complex interplay of charge localization, trapping, elastic and inelastic electron-phonon interactions, and thermal disorder. The reported dynamics studies formulate detailed conclusions which may further guide technological efforts towards design of optimal perovskite materials for photovoltaic purposes.

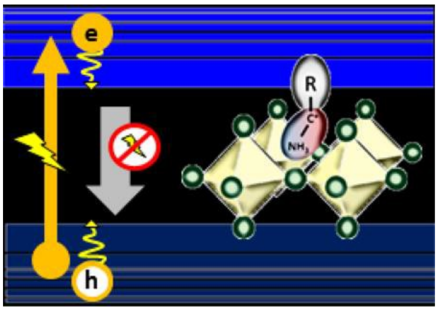

Figure 1. Photoexcitation Charge Dynamics.

References:

[1] R. Long, et al., ACS Nano **9**, 11143 (2015).
[2] R. Long, et al., J. Am. Chem. Soc. **138**, 3884 (2016).
[3] Z. S. Zhang, R. Long, et al., J. Am. Chem. Soc. **139**, 17327 (2017).
[4] R. Long, et al., J. Phys. Chem. Lett. **9**, 1164 (2018)
[5] J. L. H., R. Long, et al. J. Am. Chem. Soc. **141**, 5798 (2019).
[6] L. Q., R. Long, J. Phys. Chem. Lett. **10**, 672 (2019)
[7] Y. T. Wang, R. Long, J. Phys. Chem. Lett. **10**, 1617 (2019)

Poster No.151

Carrier dynamics in CdSe nanorods and hybrid metal-CdSe nanodumbbells: Implication for photocatalysts

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We explored photo-generated carrier dynamics of the CdSe nanorods using time-resolved absorption and photoluminescence (PL) spectroscopy spanning sub-ps to μs time domains. We controlled the fluence and wavelength of photoexcitation and the length of nanorods. The combined time-resolved spectroscopic study revealed the fast Auger recombination [1] and the shallow hole-trap [2] and band-edge relaxations in the bare-rod systems. For the semiconductor-metal hybrid systems, we performed metal domain-controlled experiments [3] and the majority of bleach recovery was found to follow ultrafast (<300 fs) channels. Additionally, minor slow bleach components were also observed in a μs time scale.

With the correlated time-resolved electronic spectroscopic methods, sensitive to both dark and luminescent states, covering a wide time window and broadband, we complete the carrier-relaxation mechanism of the CdSe nanorods and the CdSe-metal hybrid systems. From the comparison of the photocatalytic activities of defect-controlled metal tips on the hybrid semiconductor-metal nanostructures, it is revealed that the minor electron-transfer channels via metal trap sites (10's-of-μs relaxation components) are key to the photocatalytic activities.

References:

[1] V. I. Klimov, A. A. Mikhailovsky, D. W. McBranch, C. A. Leatherdale, and M. G. Bawendi, Science **287**, 5455 (2000).

[2] S. Taguchi, M. Saruyama, T. Teranishi, and Y. Kanemitsu, Phys. Rev. B **83**, 155324 (2011).

[3] J. Y. Choi, D. Jeong, S. J. Lee, D.-g. Kang, S. K. Kim, K. M. Nam, and H. Song, Nano Lett. **17**, 5688 (2017).

Poster No.152

Coherent singlet fission in pentacene driven by low-frequency intermolecular vibrations

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Singlet fission is one of the possible ways to circumvent the Shockley-Queisser limit on efficiency. Pentacene and its derivatives have shown great promise towards their application in devices to harness photons for carrier multiplication via singlet fission process. Here we study the singlet fission process in a pentacene thin film using a combination of transient-grating and two-dimensional electronic spectroscopy complemented by quantum chemical and nonadiabatic dynamics calculations. Our study reveals that the intermolecular rocking motions along the longitudinal molecular axis between the pentacene molecules play the essential role of an electronic bridge between the singlet and triplet-pair states. Along with high-frequency local vibrations acting as tuning modes, these rocking motions drive the ultrafast dynamics at the onical intersection in the singlet fission process.

Poster No.153

Femtosecond transient absorption spectroscopy study on excited state dynamics in different phases of perovskite nanocrystals

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Perovskite nanocrystals (PNCs) have gained much applications in LED, photodetectors and solar cells. In this talk, femtosecond transient absorption (FTA) spectroscopy is used to study photophysical properties of the different phase structures of PNCs. According to the singular value decomposition global fitting, the FTA dynamics of 1D orthomorphic NH_4PbI_3 PNCs is decomposed into three components and the 3D cubic phase $(NH_4)_{0.5}Cs_{0.5}Pb(I_{0.5}Br_{0.5})_3$ PNCs is decomposed into two components. It can be found the obvious ultrafast trap state capture process is a key factor to low photoluminescence properties. High photoluminescence quantum yields can be obtained by adjusting the optimal phase structure because of the large difference in bulk phase defects of different phase structures. These results suggest that the ammonium ions PNCs could be potential materials for optoelectronic applications.

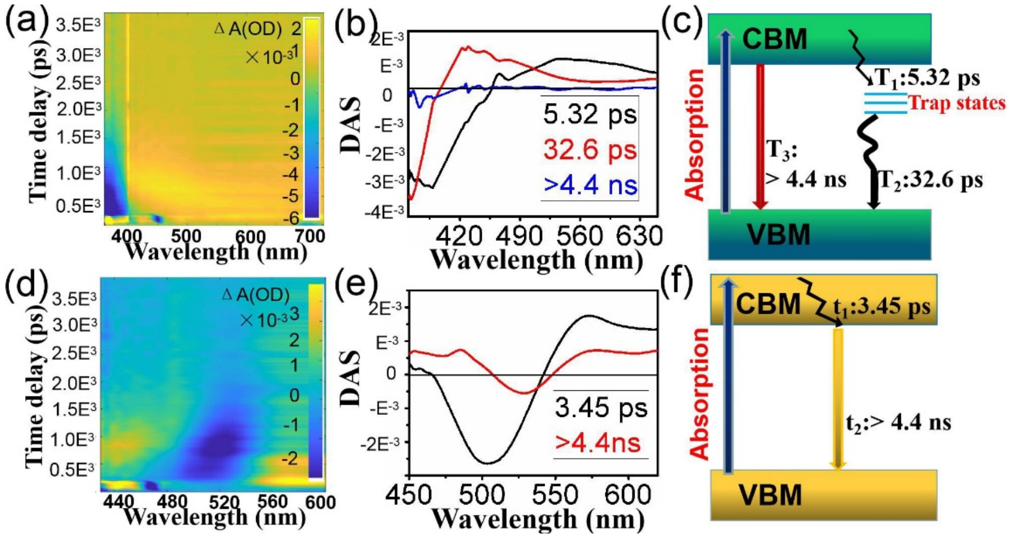

Fig.1 FTA spectra of (a) NH_4PbI_3 and (d) $(NH_4)_{0.5}Cs_{0.5}Pb(I_{0.5}Br_{0.5})_3$ PNCs. DAS for three fitting components from TA spectra of (b) NH_4PbI_3 of (e) $(NH_4)_{0.5}Cs_{0.5}Pb(I_{0.5}Br_{0.5})_3$ PNCs. Excited state dynamics mechanism of (c) NH_4PbI_3 of (f) $(NH_4)_{0.5}Cs_{0.5}Pb(I_{0.5}Br_{0.5})_3$ PNCs

References:

[1] Chao Wang, Guangjiu Zhao*, et al. Angew. Chem. Int. Ed, In Revision(2019).

Poster No.154

Room temperature "hidden state" in $La_{0.175}Ca_{0.125}MnO_3$ observed by time-resolved XRD

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Realizing active quantum control of materials near room temperature is one of the ultimate aims for their practical applications. Recent technological breakthroughs demonstrated that optical stimulation may lead to thermally inaccessible hidden states with unique properties [1-4]. However, most of the reported hidden states were induced around or below liquid nitrogen temperature [1, 2]. Here, we optically manipulated $La_{0.175}Ca_{0.125}MnO_3$ near its Curie temperature of 300 K, where typically complex phase competitions locate as well as opportunities for new functionality. A room temperature hidden state was formed with threshold behavior evidenced by a femtosecond paramagnetic (PM) to ferromagnetic (FM) order switching and a structural change distinct from thermal induced lattice expansion in tens of picoseconds accompanying with phonon softening. We propose that such a hidden state originates from the charge transfer between antiferromagnetic chains after strongly correlated spin-charge quantum excitation [1], which subsequently initiates an orbital polarization which subsequently initiates an orbital polarization rearrangement described as $Mn^{3+}_{3x^2-\frac{r^2}{3y^2}-r^2}Mn^{4+} \longrightarrow Mn^{4+}Mn^{3+}_{3z^2-r^2}$ and associated non-thermal lattice change. This study started from room temperature yet near a phase transition point, which suggests a new route to create or manipulate novel phases for practical purpose.

References:

[1] T. Li, A. Patz, and L. Mouchliadis *et al.*, Nature **496**, 69 (2013).

[2] R. Fukaya, Y. Okimoto, and M. Kunitomo *et al.*, Nat. Commun. **6**, 8519 (2015).

[3] J. Zhang, X. Tan, and M. Liu *et al.*, Nat. Mater. **15**, 956 (2016).

[4] V. R. Morrison, R. P. Chatelain, and K. L. Tiwari *et al.*, Science **346**, 445 (2018).

Poster No.155

Trap-assisted hole transport in layered hybrid perovskite

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Two-dimensional layered hybrid perovskites have been reported as absorbers in perovskite-based optoelectronic devices due to its better moisture tolerance than three-dimension ones [1,2]. Recent research has found the edge of layered single-crystals play the role of dissociation center where strongly bound excitons turn into free electrons and holes [3]. The edge of crystals is more likely 3D with n= ∞ compared with interior of n=2,3,4. However, the photogenerated carrier transport in layered single-crystals are unclear. In this paper, we systematically study the dynamics of photogenerated carriers in layered perovskite. We found that once excited, the excitons will generate and soon dissociate into free electrons and holes partly. The left excitons hardly transport and recombine around excitation spot. The dissociated electrons diffuse faster than holes because of hole traps. The electrons are captured when diffuse to edge of crystals.

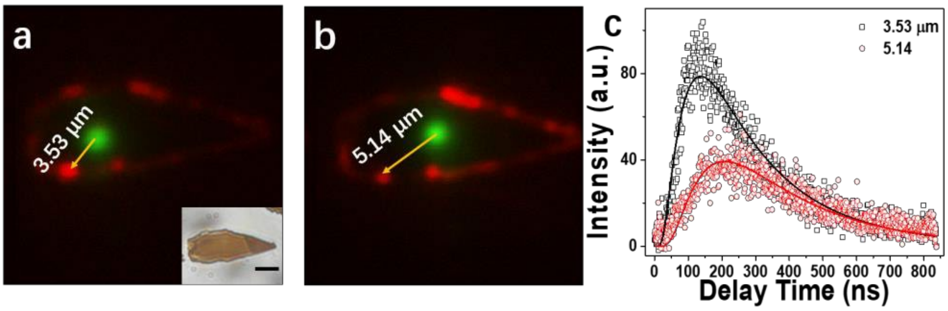

Figure 1. a, b are PL images collected at emission channels of 610 - 680 nm (interior) and > 710 nm (crystal edges) with different distance from excitation spot respectively. The scale bar is 5 μm. c is the dynamics same point (>710 nm) of a and b

References:

[1] D Cao, C. Stoumpos, O Farha, J Am Chem Soc **137**, 24, (2015).

[2] M Yuan, L Quan, R Comin, Nat Nanotechnol, **11**, 872, (2019).

[3] J Blancon, H Tsai, W Nie, Science, **355**, 1288, (2017).

Poster No.156

Ultrafast charge and energy transfer dynamics at 2D ultimate proximity

Haiming Zhu[1,*]

Because of their unique optical and electronic properties, graphene-like two-dimensional layered (2D) materials and their heterostructures coupled by van der Waals interactions have shown exciting potential in ultrathin and flexible optoelectronic, valleytronics, photovoltaic and light-emitting devices. As key steps, the photoexcitation dynamics including charge and energy transfer dynamics at Van der Waals interface determine the designing principles and overall performance of 2D optoelectronic devices. The rich and tunable properties and their atomically thin nature of 2D materials also provides an idea platform to investigate charge and energy transfer dynamics at ultimate proximity. In this talk, we will apply ultrafast laser spectroscopy to investigate the photoexcitation dynamics in 2D van der Waals heterostructures, to lay the foundation for designing novel and high-efficient van der Waals heterostructure devices.

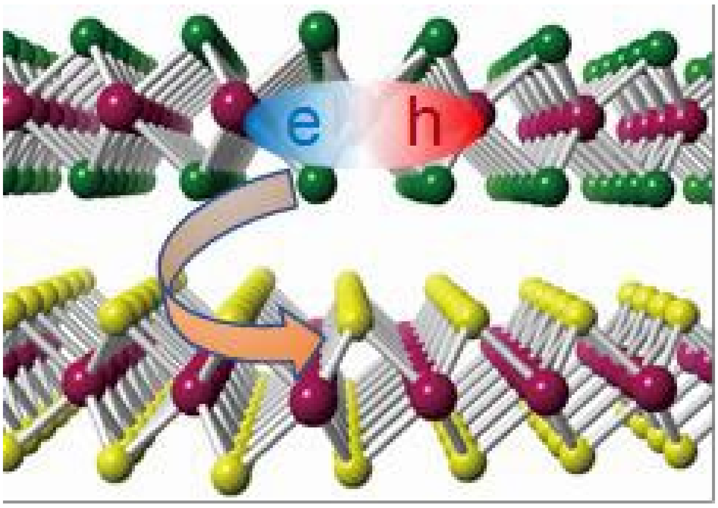

Figure 1. Charge and energy transfer process between two layered materials coupled by van der Waals force

Poster No.L2-1

Application of fluorescence Stokes shift on protein solvation dynamics and the validity of linear response theory

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We examine the water and protein dynamics using femtosecond-resolution fluorescence Stokes shift techniques. Molecular dynamics calculations reproduce the biphasic behavior of Stokes shift observed in experiments. We find that the initial dynamics in a few picoseconds represents fast local motions such as reorientations and translations of hydrating water molecules, followed by slow relaxation involving strongly coupled water-protein motions. The coupled water-protein motion is shown to be necessary for observation of the slow dynamics of Stokes shift occurring on the time scale of tens of picosecond.

The role of the Gaussian statistics on nonequilibrium relaxation dynamics upon the photon perturbation is also deeply explored. The linear response theory for the fluorescence Stokes shift is investigated. An analytical formulism is presented to recast Stokes shift into the contributions of the equilibrium time correlation functions of the solute-solvent interactions on the excited-state surface, and the latter is further reformed and depicted by the time relaxation of the moment. As the first application of the formulism in the molecular dynamics simulations, we find that the efficiency of the linear response theory relies on the Gaussian characteristics of the dominant moments in terms of the Stokes shift, which is identified by the same relaxation dynamics between those moments and the linear order one. Above observations develop an explicit criterion to measure the appropriateness of applying linear response theory.

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