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# USD 11

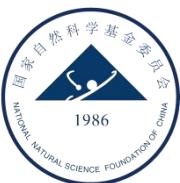
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Ultrafast Surface Dynamics

June 8-12, 2019  
Qiandao Lake, China



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SPECS-TII



## Organization and Contact

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## **WELCOME**

To the 11th International Symposium on Ultrafast Surface Dynamics. We wish you a pleasant stay at the Qiandao Lake and hope that you enjoy the scientific program of the USD 11. We are looking forward to stimulating talks and discussions.

Jin Zhao, Haiming Zhu and Hrvoje Petek

## **HISTORY**

The Ultrafast Surface Dynamics (USD) is an international symposium, which started in 1997 in Ascona Switzerland, and has since been held every 2-3 years alternating in various locations in Europe, the USA, and Japan.

- 2017 : USD10 in Inzell, Germany
- 2015 : USD9 in Lake Biwa, Japan
- 2013 : USD8 in Estes Park, U.S.A.
- 2010 : USD7 in Brijuni Islands, Croatia
- 2008 : USD6 in Kloster Banz, Germany
- 2006 : USD5 in Abashiri, Japan
- 2003 : USD4 in Telluride, U.S.A.
- 2001 : USD3 in San Sebastian, Spain
- 1999 : USD2 in Ringberg, Germany
- 1997 : USD1 in Ascona, Switzerland

## **SCOPE**

The conference focuses on the fundamental electronic excitations in solids, solid surfaces, and interfaces, which may ultimately couple to other degrees of freedom such atomic, molecular, spin, and other excitations, which evolve on time scales from attoseconds to picoseconds. The program of USD11 will encompass, but is not limited to, experiments on dynamics of interfacial electron transfer, the spin-dependent dynamics in topological materials, femtosecond relaxation during the photoemission of electrons, dynamical screening, collective excitations in solids, correlated electron phenomena, etc. A further focus will be the development and application of theoretical models that describe electronically excited states and their dynamics. New impulses are to be expected from the work on spin liquids and new developments in the field of time-resolved density function theory. The main objectives of the meeting are to exchange ideas on the current scientific results, the initiation and deepening of bilateral and multilateral cooperation among research groups, the discussion and development of visions on future research, and stimulation of ultrafast science research in China.

## **COVERED TOPICS**

- Fundamental Light-Matter interactions
- Electron Dynamics and Electronic Energy Transfer at Surfaces and Interfaces
- Electron Correlation
- Light Energy Conversion
- Surface and Interface Magnetisation Dynamics
- Ultrafast Plasmonics and Nano optics
- Dynamics in Low-dimensional materials

- Novel Phenomena / Materials
- Ultrafast Surface Science

#### **INTERNATIONAL STEERING COMMITTEE**

- Uwe Bovensiepen (University of Duisburg-Essen, Germany)
- Pedro Echenique (University of the Basque Country, Spain)
- Tony Heinz (Stanford University, USA)
- Ulrich Höfer (University of Marburg, Germany)
- Hrvoje Petek (University of Pittsburgh, USA)
- Martin Weinelt (Freie Universität Berlin, Germany)
- Martin Wolf (Fritz-Haber Institute, Germany)
- Xiaoyang Zhu (Columbia University, USA)
- Jin Zhao (University of Science & Technology of China, China)

## PROGRAM

Friday, June 7		
Session Chair: Hrvoje Petek		
19:30 - 19:40	<b>Welcoming Remarks</b>	
19:40 - 20:20	V. Ara Apkarian	Spectromicroscopy with Atomically Confined Light
20:20 - 21:00	Xiaohui Qiu	Cross-plane Thermal Conductance of Au/graphene/Au Heterojunctions
Saturday, June 8		
Session Chair: Ulrich Höfer		
09:00 - 09:40	Javier García de Abajo	Plasmonics in Atomically Thin Materials
09:40 - 10:20	Martin Aeschlimann	Mixing the Light-Spin with Plasmon-Orbit by Non-Linear Light Matter Interaction
10:20 - 10:40	<b>Coffee Break</b>	
10:40 - 11:20	Robin Huang	Revealing an Optical Spin Skyrmion in the Core of a Surface Plasmon Vortex
11:20 - 12:00	Markus B. Raschke	Ultrafast and Nonlinear Nanoscopy: Watching Quantum Dynamics in Time and Space
12:00 - 12:20	Zeno Schumacher	Ultrafast Nonlinear Optical Response Probed by Atomic Force Microscopy
12:20 - 14:40	<b>Lunch</b>	
Session Chair: Haiming Zhu		
14:40 - 15:20	Jigang Wang	Quantum Beats by Terahertz Symmetry Breaking: From Excitons, Polarons to Cooper Pairs (THz)
15:20 - 15:40	Oliver B. Wright	Imaging GHz Surface Phonons on Phononic Crystals, Acoustic Metamaterials and Microstructures
15:40 - 16:00	Timm Rohwer	Defect-mediated Control of Charge Density Waves
16:00 - 16:20	<b>Coffee Break</b>	
16:20 - 17:00	Cheng-Tien Chiang	Photoemission with High-order Harmonics at MHz
17:00 - 17:20	Henry Kapteyn	The Y-Fi VUV™ : A One-Box MHz rep-rate VUV Source for Surface

		and Chemical Dynamics Studies
17:20 - 19:00		<b>Dinner</b>
19:00 - 21:00		<b>Poster Session</b>
<b>Sunday, June 9</b>		
<b>Session Chair: Karl Martin Wolf</b>		
09:00 - 09:40	Xiaoyang Zhu	Excitons, phonons and Electrons in Two-dimensional Semiconductors and Heterojunctions
09:40 - 10:20	Uwe Bovensiepen	Local and Non-local Electron Dynamics in Layered Material Systems Analyzed by Time-resolved Photoelectron Spectroscopy
10:20 - 10:40		<b>Coffee Break</b>
10:40 - 11:20	Michael Bauer	Ultrafast Dynamics in 2D- and 3D-Dirac Materials
11:20 - 12:00	Shiwei Wu	Nonlinear Optical Response of Massless Dirac Fermions in Graphene Materials
12:00 - 12:20	Tzu-Yu Chen	Second Harmonic Generation in a Fully Centro-Symmetric Plasmonic Nanocircuit
12:20 - 14:40		<b>Lunch</b>
<b>Session Chair: Raschke Markus</b>		
14:40 - 15:20	Yihua Wang	Ultrafast Electronic and Magnetic Dynamics in Topological Materials
15:20 - 16:00	Kazuki Sumida	Surface Dirac Fermion Dynamics of Non-magnetic and Magnetic Topological Insulators
16:00 - 16:20		<b>Coffee Break</b>
16:20 - 17:00	Kaihui Liu	Ultrafast Optical Spectroscopy of Individual Nanomaterials
17:00 - 17:40	Haiming Zhu	Charge and Energy Transfer Dynamics at 2D Ultimate Proximity
17:40 - 18:00	Nan Pan	Magnetically-Sensitive Valley Polarization Reversal and Revival of Defect-Bound Excitons in WSe <sub>2</sub> -WS <sub>2</sub> Heterostructure
18:00 - 19:00		<b>Dinner</b>
19:00 - 21:00		<b>Poster Session</b>
<b>Monday, June 10</b>		
<b>Session Chair: Ryuichi Arafune</b>		
09:00 - 09:40	Christian Heide	Ultrafast Control of Electron Inside of Graphene and Across

		Solid-state Interfaces
09:40 - 10:00	Walter Pfeiffer	Final State Effective Mass Affects Attosecond Photoemission Delays in BiTeCl
10:00 - 10:20	Andreas Gebauer	Equivalence of RABBITT and Streaking Delays in Attosecond-Time-Resolved Photoemission Spectroscopy at Solid Surfaces
10:20 - 10:40	<b>Coffee Break</b>	
10:40 - 11:20	Sheng Meng	Ultrafast Carrier Relaxation and Pauli Drag Effect in Photoexcited Solids
11:20 - 12:00	Jin Zhao	Molecular Dynamics Investigations on the Excited Carriers in Condensed Matter Systems
12:00 - 14:00	<b>Lunch</b>	
14:00 - 18:00	<b>Conference Outing</b>	
18:00 - 21:00	<b>Conference Dinner</b>	

### Tuesday, June 11

#### Session Chair: Uwe Bovensiepen

09:00 - 09:40	Martin Wolf	Ultrafast Excited State Dynamics of Solids Probed by Time-resolved ARPES
09:40 - 10:20	Shijing Tan	Anisotropic Optical Response of Black Phosphorus Studied by Multiphoton Photoemission Spectroscopy
10:20 - 10:40	<b>Coffee Break</b>	
10:40 - 11:20	Fengqiu Wang	Excited-states Lifetime Control in Emerging Low-dimensional Materials
11:20 - 11:40	Takeshi Suzuki	Ultrafast Carrier Dynamics in a Twisted Bilayer Graphene Quasicrystal
11:40 - 14:40	<b>Lunch</b>	

#### Session Chair: Xiaoyang Zhu

14:40 - 15:20	Xuefeng Guo	Ultrafast Single Molecule Electrical Detection
15:20 - 15:40	Lisa Grad	Electron Dynamics on Cu <sub>2</sub> O(111) Probed with Time-Resolved Two-Photon Photoemission
15:40 - 16:00	Helmut Zacharias	Exciton-induced Chemical Reactions on Surfaces
16:00 - 16:20	<b>Coffee Break</b>	

16:20 - 16:40	Kiyoshi Miyata	Unique Dielectric Responses in Lead-halide Perovskites: Proposal of Ferroelectric Polaron		
16:40 - 17:00	Sebastian F. Maehrlein	Dialing into Dynamically Disordered Lattice Potentials in $\text{CsPbBr}_3$ via 2D Coherent Phonon Spectroscopy		
17:00 - 17:20	Kunie Ishioka	Charge Separation Dynamics at Lead Halide Perovskite Interfaces Monitored by Differential Transient Transmission Spectroscopy		
17:20 - 17:40	Daniel Niesner	Coupled Spin and Lattice Dynamics in Lead Halide Perovskites		
18:00 - 19:40	<b>Conference Banquet &amp; After-dinner Colloquium</b>			
<b>Session Chair: Shijing Tan</b>				
19:40 - 20:20	Hrvoje Petek	How to Dress a Metal		
20:20 - 21:00	Ulrich Höfer	Subcycle Time-resolved ARPES of THz-driven Dirac Currents in a Topological Surface Band		
21:00 - 21:20	<b>Poster Award</b>			
<b>Wednesday, June 12</b>				
<b>Session Chair: Kazuki Sumida</b>				
09:00 - 09:40	Martin Weinelt	Spin Mixing and Spin-wave Excitations in 3d and 4f Ferromagnetic Metals		
09:40 - 10:20	Ryuichi Arafune	Spectroscopic Investigation of Opto-spincurrent Control		
10:20 - 10:40	<b>Coffee Break</b>			
10:40 - 11:20	Zhensheng Tao	Critical Behaviors in 20fs Govern the Non-equilibrium Laser-induced Magnetic Phase Transition in Ni		
11:20 - 11:40	Stefan Mathias	Coherent Ultrafast Spin Transfer in Ferromagnetic Alloys		

# **ORAL CONTRIBUTIONS**

# Spectromicroscopy with Atomically Confined Light

**V. Ara Apkarian**

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Plasmonic nanojunctions can be used to effectively confine light on the Å scale, surpassing the diffraction limit of light by four orders of magnitude, and opening the atomistic near-field in optical microscopy. Seeing an atom, a single chemical bond, imaging the vibrational normal modes inside one molecule, and seeing sound and atomically resolved phonons are among the recent observations made in our laboratory under the rubric of tip-enhanced Raman spectro-microscopy (TERS). I will use these examples to expand on the unusual properties and multiple facets of atomistically confined plasmons and their applications. As light, the momentum uncertainty associated with spatial confinement gives visible photons the wavelength of x-rays, which allows the simultaneous energy and momentum matching conditions required to see dispersive acoustic phonons – to see sound. As fields, the dramatic enhancement through confinement accesses the strong-field limit and allows detection of the feeble Raman effect from individual atoms and molecules. This gives access to the electro-mechanical machinery of individual molecules for sensing and device applications, such as the recently demonstrated single molecule scanning electrometer and single molecule optoelectronics. As picocavity photons, superpositions of only two Fock states can be prepared and maintained. And in the quantum tunneling limit required for atomistic resolution, phototunneling current of confined electrons provides a more natural description of the local, time harmonic electro-magnetic fields and associated observables, such as the phase in Raman lines. There is much to be seen at the Å-scale opened up through atomically confined light.

# Cross-plane Thermal Conductance of Au/graphene/Au Heterojunctions

Junbo Zhou<sup>1,2</sup>, Rui Wang<sup>1</sup>, Xiaohui Qiu<sup>1,2</sup>

<sup>1</sup> Chinese Academy of Sciences Key Laboratory of Standardization and Measurement for Nanotechnology, Chinese Academy of Sciences Center for Excellence in Nanoscience, National Center for Nanoscience and Technology, Beijing 100190, China

<sup>2</sup> University of Chinese Academy of Sciences, Beijing 100049, China  
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2D materials provide an appealing platform for studying heat transport through atomically abrupt interface, allowing to reveal interesting phenomena such as thickness dependent thermal conductivity at the nanometer scale. Here, we employ the Frequency Domain Thermoreflectance (FDTR) technique to investigate the heat conduction across the heterojunction composed of few-layer graphene embedded in gold thin film. We measure the thermal conductance across this Au/graphene/Au sandwich structure as a function of graphene layer number at varying temperatures. It is found that electron transmission dominates the thermal conductance for monolayer graphene, and two-orders of magnitude decrease through bilayer graphene. For multilayer graphene sheets, phonon thermal conductivity is suggested to account for the heat transmission through the interfacial boundaries.

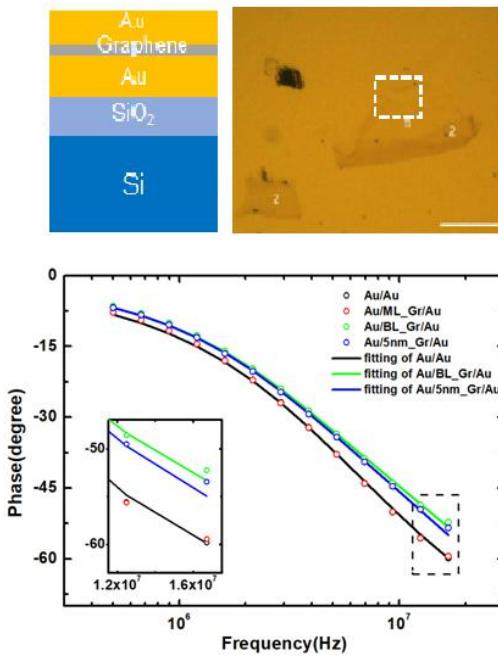


Fig. 1 Schematics of the Au/graphene/Au structure, and an optic image of graphene on Au with mono- and bi-layer graphene (scalebar 20um). The measured phase lags in FDTR and their fittings to a heat transfer model, through which the thermal conductivity of Au/graphene/Au heterostructures were derived.

# Nonlinear optical response of massless Dirac fermions in graphene

**Shiwei Wu<sup>1</sup>**

<sup>1</sup>*Department of Physics, Fudan University, China*

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Graphene exhibits extraordinarily strong coupling to light owing to its unique linear and gapless two-dimensional band structure that hosts massless Dirac fermions. In this talk, I will focus on its nonlinear optical response. For centrosymmetric monolayer graphene, the second-order optical nonlinearity is forbidden under the electric-dipole approximation. Yet, it possesses exceptionally strong third-order optical nonlinearity. Recently we show that the third-order nonlinearity including THG and FWM could be electrically tuned by orders of magnitude [1]. Furthermore, the electric-quadrupole response is also gate tunable, leading to strong doping-induced second harmonic generation (SHG) beyond the electric-dipole approximation [2]. In contrast to the electric dipole allowed THG, the electric quadrupole SHG strictly vanishes at zero doping, manifesting the electron-hole symmetry of massless Dirac fermions. We believe the findings in graphene are readily applicable to other related Dirac materials such as topological insulators, Dirac and Weyl semimetals. And the giant nonlinearity in graphene can be utilized to develop nonlinear optoelectronic devices.

[1] Tao Jiang,<sup>†</sup> Di Huang,<sup>†</sup> Jinluo Cheng, Xiaodong Fan, Zhihong Zhang, Yuwei Shan, Yangfan Yi, Yunyun Dai, Lei Shi, Kaihui Liu, Changgan Zeng, Jian Zi, J.E. Sipe, Yuen-Ron Shen, Wei-Tao Liu,\* Shiwei Wu\*, Gate tunable third-order nonlinear optical response of massless Dirac fermions in graphene. *Nature Photonics* 12, 430-436 (2018).

[2] Yu Zhang,<sup>†</sup> Di Huang,<sup>†</sup> Yuwei Shan, Tao Jiang, Zhihong Zhang, Kaihui Liu, Lei Shi, Jinluo Cheng, John E. Sipe, Wei-Tao Liu,\* Shiwei Wu\*, Doping induced second harmonic generation in centrosymmetric graphene from quadrupole response. *Physical Review Letters* 122, 047401 (2019).

# Mixing the Light-Spin with Plasmon-Orbit by Non-Linear Light Matter Interaction

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Transformation of light carrying spin angular momentum (SAM) to optical field vortices carrying orbital angular momentum (OAM) has been of wide interest in recent years. The interactions between two optical fields, each carrying one of those degrees of freedom, and furthermore, the transfer of the resulting angular momentum product to matter is seldom discussed. Here we measure the interaction between light carrying axial SAM and plasmon-polariton vortices carrying high order transverse OAM. The interaction is mediated by two-photon absorption within a gold surface, imprinting the resulting angular momentum mixing into matter by excitation of electrons that are photo-emitted into vacuum.

We demonstrate that, counter-intuitively, the spatial distribution of the emitted electrons carries the signature of a subtraction of the spin from the orbit angular momenta. We show experimentally and theoretically that the absorptive nature of this interaction leads to both single and double photon-plasmon angular momentum mixing processes by one- and two- photon interactions. The dynamics of two-photon absorption can be described by a three-level model system and its corresponding density matrix. The transition from the ground to the third excited state may occur through a population of an intermediate state, or through coherences only without passing through the diagonal elements. In this way one can discriminate between pathways consisting of a single angular momentum mixing in one absorption, with the two steps of the other absorption performed by pure photon or plasmon fields. Double mixing pathways occur when both transitions are performed by identical two-step sequences of mixing plasmon and light.

# Revealing an optical spin skyrmion in the core of a surface plasmon vortex

Yanan Dai<sup>1</sup>, Zhihang Zhou<sup>1</sup>, Atsushi Kubo<sup>2</sup>, Chen-Bin Huang<sup>3</sup>, and Hrvoje Petek<sup>1</sup>

<sup>1</sup> Department of Physics and Astronomy, University of Pittsburgh, USA

<sup>2</sup> Department of Physics, University of Tsukuba, Japan

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We report the first experimental evidence for an electric field spin angular momentum skyrmion at a plasmonic silver surface. Skyrmions are stable quasiparticles in magnetic materials with distinct topological spin textures that have been investigated for several years by electron microscopy and scan tunneling microscopy techniques. Our skyrmion represents a true topological spin texture, which is engendered by an Archimedean plasmonic coupling structure.

We employ interferometric time-resolved photoemission electron microscopy to record a movie of the plasmonic fields with <10 nm spatial resolution and <100 as pump-probe pulse delay/frame. Archimedean plasmonic coupling structures have been employed before to generate vortices, but they have never been explored from perspective of skyrmions. Unlike previous research, we employ linearly polarized light to generate the vortex, with the outcome that the skyrmion is generated by the geometrical charge of the coupling structure, rather than more complicated process involving the photon spin. We model the nanoscale fields, their polarization properties and plasmonic flow, which enable us to conclude that the skyrmion is a stable quasiparticle on the time scale of the pulse duration.

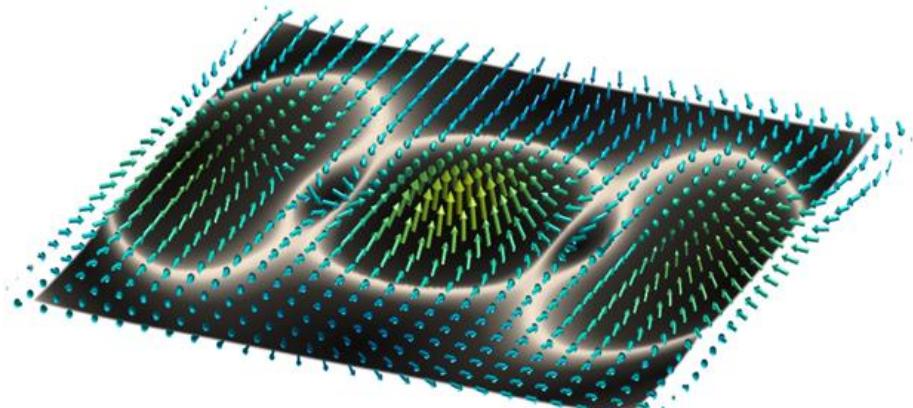


Fig. 1 The plasmonic Néel spin angular momentum (SAM) skyrmion. The colored arrows show the SAM pseudovectors that define the skyrmion texture created within a plasmonic vortex core; they are overlaid on the map of L-line singularities of SPP fields that define the skyrmion density.

# Ultrafast and Nonlinear Nanoscopy: Watching Quantum Dynamics in Time and Space

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We will present a new regime in nonlinear nano-optics and spatio-temporal nano-imaging for the study of fundamental coherent optical, plasmonic, and quantum dynamic phenomena with nanometer spatial and few-femtosecond temporal resolution. To gain the desired *nanometer spatial resolution* with *spectroscopic specificity* and *femtosecond temporal resolution* we combine plasmonic nano-focusing and optical antenna concepts with ultrafast and shaped laser pulses to precisely control optical excitation on fs-time and nm-length scales.

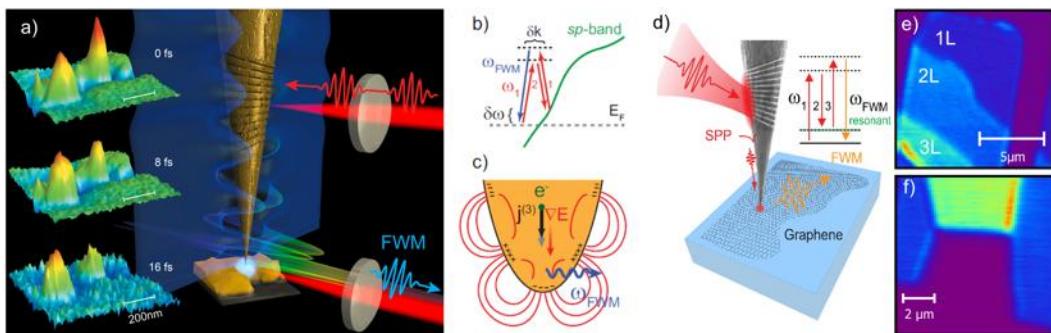


Fig. 1. a) Nonlinear four-wave mixing nano-focused imaging of coherent electron dynamics with few-fs time and few-nm spatial resolution [1]. b-c) New mechanism for enhanced nonlinear frequency conversion on the nanoscale by gradient field and evanescent wave phase matching [2]. d-f) Nonlinear nano-optics of graphene with layer dependent, enhanced edge contrast, and quantum Doppler nonlocal response exploring the unique electronic bandstructure and symmetries of graphene [3]. Examples include an ultrafast movie of the real time and real space coherent dynamics of coupled plasmonic nanostructures (Fig 1a) [1]. The underlying mechanism of four-wave mixing reveals a new regime of nonlinear frequency generation on the nanoscale based on a plasmonic gradient-field effect that provides for an efficient, conventionally dipole-forbidden nonlinear response (b,c) [2]. Similarly, in graphene, we discover new spatial, spectral, and temporal phenomena based on a bandstructure and tip nano-cavity enhanced nonlinear response (d-f) [3]. These results provide for localized and -enhanced nonlinear optical interactions with new possibilities for applications from nano-photonic circuits to quantum nano-optics, and -imaging.

1. V. Kravtsov, R. Ulbricht, R., J. M. Atkin, and M. B. Raschke, Plasmonic nanofocused four-wave mixing for femtosecond near-field imaging. *Nature Nanotechnol.* **11**, 459-464 (2016).

2. V. Kravtsov, S. AlMutairi, R. Ulbricht, A. Ryan Kutayiah, A. Belyanin, and M. B. Raschke, Enhanced third-order optical nonlinearity driven by surface-plasmon field gradients. *Phys. Rev. Lett.* **120**, 203903 (2018).
3. T. Jiang, V. Kravtsov, S. AlMutairi, A. Belyanin, and M. B. Raschke, Nonlinear nano-optics of graphene: symmetry, nonlocality, and coherent dynamics (submitted).

# Ultrafast nonlinear optical response probed by atomic force microscopy

Zeno Schumacher\*, Rasa Rejali, Andreas Spielhofer, Yoichi Miyahara and Peter Grutter

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Scanning probe technique such as non-contact Atomic Force Microscopy (nc-AFM) offer the ability to spatially resolve surfaces with sub-nanometer resolution. Recent technical advances, such as photo-induced force microscopy (PiFM) [1] and pump-probe style time-resolved nc-AFM [2,3], have shown the potential of light induced force measurements, resolving dynamics in the ps range. The high force sensitivity of AFM allows to resolve small variations in local optical induced forces and to decouple the probe volume from the focal size of the illumination.

Here, we present nanometer spatially resolved measurements of the ultrafast optical nonlinear response on a surface to a fs-pulsed laser. The induced polarization is detected as an additional force component in the nc-AFM measurement. Local variations on the nanometer scale of the induced polarization on a periodically poled lithium niobate surface can be resolved. The variation can be attributed to surface adsorbates and correlated to topographical nc-AFM measurements. Due to the high force sensitivity of this method, even monolayer samples can be detected. We will present recent results on a MoSe<sub>2</sub> monolayer detecting the ultrafast nanometer local nonlinear optical response. With this, nc-AFM adds to the toolbox of ultrafast surface science, whereas the force detection of ultrafast optical nonlinear processes is combined with nanometer spatial resolution.

- [1] Rajapaksa, I., Uenal, K., & Wickramasinghe, H. K. (2010). Image force microscopy of molecular resonance: A microscope principle. *Applied Physics Letters*, 97(7), 073121.
- [2] Jahng, J., et al. (2015). Ultrafast pump-probe force microscopy with nanoscale resolution. *Applied Physics Letters*, 106(8), 083113.
- [3] Schumacher, Z., Spielhofer, A., Miyahara, Y., & Grutter, P. (2017). The limit of time resolution in frequency modulation atomic force microscopy by a pump-probe approach. *Applied Physics Letters*, 110(5).

# **Quantum Beats by Terahertz Symmetry Breaking: From Exitons, Polarons to Cooper Pairs**

**Jigang Wang**

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Harnessing quantum coherence and quantum quench prethermalization has emerged as a cross-cutting theme for discovering and controlling emergent phases and collective modes, even thermodynamically forbidden. In this talk, I will discuss our recent progress towards applying these new tuning knobs using single- and few-cycle THz pulses to reveal quantum beats and ultrafast control in several systems of current focus including perovskite semiconductors and superconductors. Particularly, our results show that THz light-driven coherence and subcycle symmetry breaking allows the observation of forbidden collective modes and hidden many-body quantum phases that are difficult to be accessed by other tuning methods. Finally I will discuss far-reaching consequences of THz coherent and nano-optics methods on quantum matter discovery.

# Imaging GHz surface phonons on phononic crystals, acoustic metamaterials and microstructures

**Oliver B. Wright**

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Surface acoustic wave devices based on waves in resonators have found extensive application in high-frequency signal processing. In particular, phononic crystals, metamaterials and micron to sub-micron structures exhibit interesting physical properties, such as omnidirectional stop bands or tight wave confinement, that allow potential improvements to these devices. Here we present experiments and simulations of time-domain imaging of laser-excited surface phonons at frequencies from 100 MHz up to  $\sim$ 1 GHz on various novel micron-scale geometries, namely phononic crystals, phononic-crystal slab cavity structures, phononic crystal waveguides, metasurfaces, metamaterial extraordinary-transmission structures, and whispering gallery resonators. In addition to the time domain, we also present results in the frequency domain and in  $k$ - $t$  space, where  $k$  is the wavevector and  $t$  is the time. Applications include gigahertz phonon control.

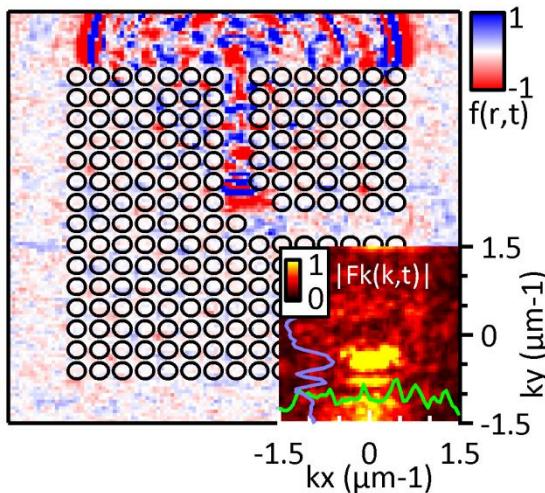


Figure 1. Time domain images of GHz ultrasonic waves propagating in a L-phononic crystal waveguide slab[1]. Imaged region  $125 \text{ } \mu\text{m}^2$ . Inset:  $k$ - $t$  space image. The green and blue lines are horizontal and vertical cross sections through the  $k$ -space origin, respectively. The time is 8.3 ns after excitation.

- [1] O. B. Wright and O. Matsuda, *Phil. Trans. Roy. Soc. A* **373**, 20140364 (2015).
- [2] O. Matsuda, S. Kaneko, O. B. Wright, and M. Tomoda, *IEEE Trans. Ultrason. Ferroelectr. Freq. Control* **62**, 584 (2015)
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- [4] S. Mezil, K. Fujita, P. H. Otsuka, M. Tomoda, M. Clark, O. B. Wright and O. Matsuda, *Appl. Phys. Lett.* **111**, 144103 (2017)
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# Defect-mediated Control of Charge Density Waves

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We investigated the light-induced melting of a unidirectional charge density wave (CDW) in LaTe<sub>3</sub> by a combination of time-resolved Angle Resolved Photo-Electron Spectroscopy (trARPES), Ultrafast Electron Diffraction (UED) and Transient Optical Spectroscopy (TOS) [1]. This enabled us to record and compare the correlated dynamics in both, the electronic and lattice subsystem, leading to an independent tracking of the CDW amplitude and phase dynamics. We find that a fast (approximately 1 picosecond) recovery of the CDW amplitude is followed by a slower re-establishment of phase coherence. This longer timescale is dictated by the presence of topological defects: long-range order is inhibited and is only restored when the defects annihilate.

On a different CDW compound a fully reversible on-off switching of domain-walls is demonstrated by means of UED. Both, the creation and annihilation can be realized by a single pulse of light [2]. Both studies provide a framework to understand and control the geometry of the ultrafast restored CDW's by utilizing defects and boundaries as a governing mechanism.

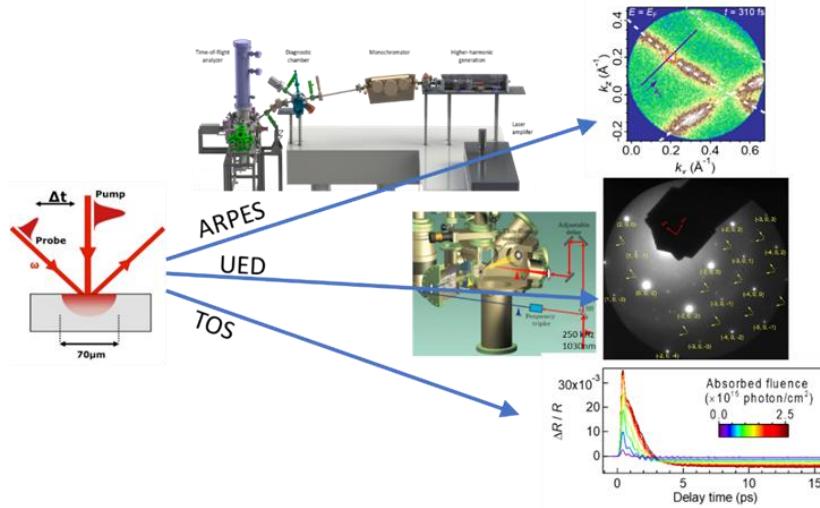


Fig. 1 Three time-resolved probes for combined detection of the transient electronic and lattice structure.

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# Photoemission with high-order harmonics at MHz

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With the recent progress of high-power femtosecond lasers, high-order harmonic generation (HHG) has evolved into a routine technique to convert the infrared laser light into a much higher photon energy ranging from vacuum-ultraviolet to x-ray. Due to the widely tunable photon energy range as well as the ultimate pulse duration below few femtoseconds, the application of HHG-based light source to photoelectron spectroscopy (PES) and microscopy have been pursued since decades [1]. However, these developments have been limited by the space-charge effects at low laser repetition rates of several kilohertz as well as the complexity of the HHG setups. By combining femtosecond lasers operating at megahertz repetition rates with a compact design of the HHG chamber, we have demonstrated efficient photoelectron spectroscopy experiments [2,3]. In this talk, the application of MHz-HHG light sources to time-resolved PES and the advanced double photoemission (DPE) spectroscopy will be presented.

In DPE experiments, a pair of photoelectrons are emitted due to the absorption of one single photon. Therefore DPE could provide a unique opportunity to analyze the interaction between electrons in solids. By using a pair of time-of-flight spectrometers in combination with the MHz-HHG light source [4,5], we analyze the two-dimensional energy distribution of correlated *sp-d* and *d-d* electron pairs from Ag(001) and Cu(111) [6]. These results will be compared with DPE on correlated materials such as NiO and discussed in terms of electron correlation.

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# The Y-Fi VUV<sup>TM</sup>: A One-Box MHz rep-rate VUV Source for Surface and Chemical Dynamics Studies

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Ultrashort-pulse light in the deep-UV-vacuum-UV spectral region, covering ~6-15 eV, is uniquely capable of probing physical and chemical processes, for example for Angle-Resolved Photoelectron Spectroscopy (ARPES) and Photoionization Mass Spectrometry PIMS. However, the ideal VUV light source for these applications—compact, with MHz repetition-rates and femtosecond-domain pulse duration—has not as-yet existed. High-order harmonic generation has proven useful for generating multiple harmonic orders in the VUV and EUV spectral regions; however, when attempting to scale to higher repetition rate, the much lower pulse energy from tabletop-scale MHz lasers requires a tight-focusing geometry with rapidly diminishing conversion efficiency.

Recently, we demonstrated a new architecture for VUV generation using cascaded four-wave mixing in a small-diameter, low-loss negative curvature (NC) fiber. We employ  $\omega+2\omega$  cascaded four-wave-mixing driven by an ultrafast fiber laser running at up to 5 MHz, to generate 1.2-eV-spaced harmonics extending to ~11 eV, with flux approaching synchrotron levels—all in a tabletop setup. This work is based on well-established four-wave mixing in gasses.[1-2] By using tight confinement of the driving laser mode over a long propagation length, we enable multi-step cascaded upconversion in a simple geometry.[3]

In this project, we started with a commercial (KMLabs YFi Ultra<sup>TM</sup>) ultrafast fiber laser producing ~10 J, 160 fs pulses @ 1040 nm with 1-10 MHz repetition rate. The output was frequency doubled with ~50-70% efficiency, recombined with 1040 nm light and retimed, then focused into a ~30 m diameter NC fiber backed with xenon gas. The first four-wave-mixing (FWM) step generates >200 mW at 345 nm, which then cascades to produce  $4\omega$ ,  $5\omega$ ,  $6\omega$ ,  $7\omega$ ,  $8\omega$ , and  $9\omega$  (Figure 1). The narrow ~20 meV spectral bandwidth of the laser broadens only modestly, by ~2-3x for the 10.8 eV emission and less for lower orders.

KMLabs is currently constructing a commercial version of this laser, the Y-Fi VUV<sup>TM</sup> (figure 2), which incorporates the fiber laser, the upconversion setup, and a monochromator into a single 0.75m x 1.5 m enclosure designed to replace conventional synchrotron and lamp-based light sources for a wide variety of VUV light applications. The monochromator is designed to accommodate applications both in high-resolution and in time-resolved studies, making it possible to either select a single harmonic order with minimal time-dispersion, or to spectrally narrow the output to ~1 meV. Further results characterizing the Y-Fi VUV<sup>TM</sup> will be presented, along with on-going experimental application results.

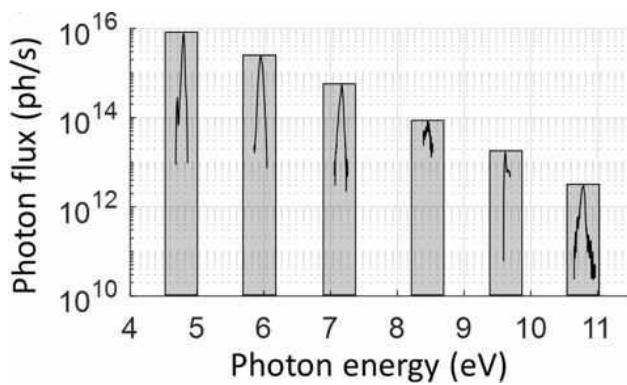


Figure 1. Using cascaded four-wave mixing to generate VUV harmonics of a 1 MHz 1040 nm fiber laser.  
 a) Photon flux and spectrum for 6 harmonics. Flux corresponds to intensity emitted by the source, measured by separating harmonic orders with a prism, using a power meter (up to 6 eV), current from a NIST-calibrated  $\text{Al}_2\text{O}_3$  photodiode (for 10.8 eV), and estimates for 7.2-9.6 eV based on relative fluorescence intensity.

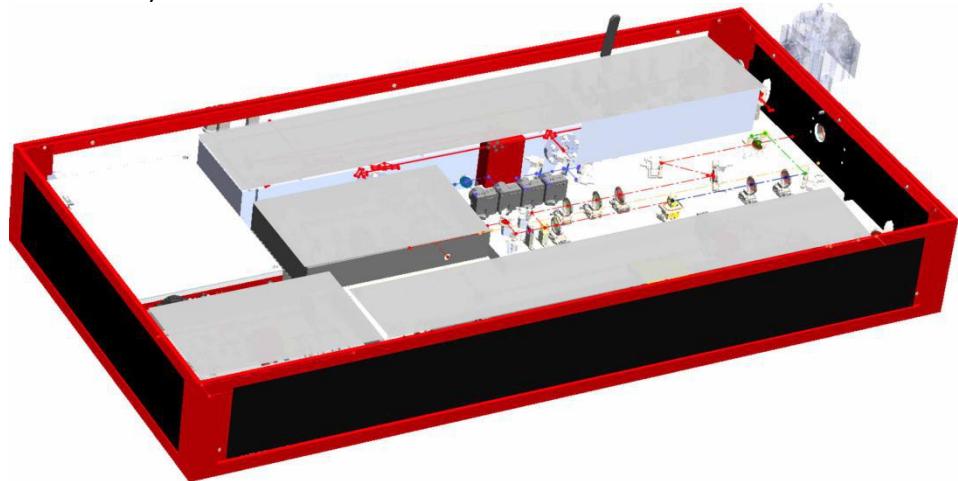


Figure 2. Y-Fi VUV design layout. Entire optical system including monochromator for energy selection occupies a footprint of 0.75 m x 1.5 m.

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# **Excitons, phonons, and electrons in two-dimensional semiconductors and heterojunctions**

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Two-dimensional (2D) semiconductors, such as transition metal dichalcogenides (TMDCs), are emerging platforms for exploring a broad range of electronic, optoelectronic, and quantum phenomena. These materials feature strong Coulombic interactions, making them ideal for studying highly correlated phenomena as a function of charge-carrier density. In this talk, I will discuss our recent effort in understanding dynamics at the 2D limit. Using a range of model systems, including TMDC hetero-bilayers, 2D superatomic semiconductors, and 2D magnetic materials, I will discuss how the strong Coulomb interaction is manifested in manybody dynamics, including spin-valley specific scattering, Mott transitions, and electron-phonon coupling. I will also discuss the prospects of quantum fluid phases, such as exciton condensates and 2D superconductivity.

# Local and non-local electron dynamics in layered material systems analyzed by time-resolved photoelectron spectroscopy

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An excited electron in condensed matter propagates with its momentum  $\mathbf{k}$  at an energy  $E(\mathbf{k})$  and experiences elastic and inelastic scattering processes, which lead to relaxation. Experiments employing femto-second time-resolved photoelectron spectroscopy exploited so far very successfully the surface sensitivity of photoelectron spectroscopy and probed such scattering processes locally at the surface or the surface near region in the time domain [1]. Here, we report on first experimental results which analyze the non-local dynamics of excited electrons in two-photon photoemission (2PPE). In these experiments one photon excites in a Au/Fe/MgO(001) heterostructure electrons in Fe. Electron propagation through the layer stack to the Au surface is detected in 2PPE in back-pump front-probe experiments, similar to pioneering femtosecond pump-probe experiments in condensed matter, which revealed time-dependent changes in the linear optical reflectivity of free standing Au films[2].

Electrons which propagate through the layer stack in the intermediate state are detected, similar to a time-of-flight analysis. We observe pronounced differences between front- and back-pumping the hetero-structure which are attributed to electron transport contributions through the layer stack. Given the investigated Au film thickness  $5 \text{ nm} \leq d_{\text{Au}} \leq 30 \text{ nm}$ , the Fermi velocity of Au of  $v_F=1.4 \text{ nm/fs}$ , and the 2PPE cross correlation width on the Au surface of 70 fs we are setup to distinguish ballistic and superdiffusive transport contributions. We identify elastically and inelastically scattered electrons which propagate in a superdiffusive regime [3]. Furthermore, absence of ballistic contributions at electron energies of 1-2 eV above the Fermi level for the thicker Au films are attributed to enhanced scattering at the Fe-Au interface.

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# Ultrafast Carrier Dynamics in 2D- and 3D-Dirac Materials

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In this contribution I will report on recent trARPES results on the relaxation of hot carriers in 2D- and 3D-Dirac cones. In HOPG we were able to follow different stages in the formation of a Fermi-Dirac distribution in a 2D-Dirac cone within the first 50 fs after excitation with 7 fs pump pulses [1]. The data allow discerning processes associated with electron-electron and electron-phonon interaction, which act differently on the energy and momentum relaxation of the nascent carrier distribution [2].

In the 3D-Dirac material Cd<sub>3</sub>As<sub>2</sub> the photoexcitation with 840 nm laser pulses results in an indirect population of the Dirac cone from higher-lying states. We observe a depopulation of the Dirac cone taking place on picosecond timescales (see Fig. 1). In contrast to HOPG the characteristic carrier dynamics is accompanied by a transient renormalization of background bands which form in the ground state in the presence of spin-orbit coupling [3].

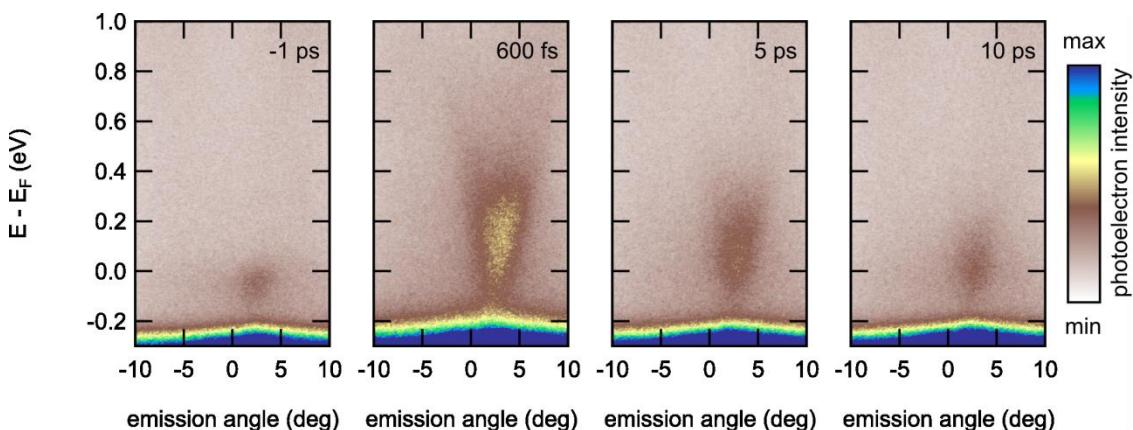


Fig. 1 TrARPES data of a 3D-Dirac cone in Cd<sub>3</sub>As<sub>2</sub>;

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# Plasmonics in atomically thin materials

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Two-dimensional polaritons have emerged as powerful tools to manipulate light at atomic scales in materials such as graphene, transition metal dichalcogenides, and atomically-thin metal films. In this talk, we will review recent experimental advances in the fabrication and performance of atomically-thin metallic films, as well as fundamental properties of these excitations, including their in/out-coupling to light and their potential for applications in sensing, nonlinear optics, and quantum physics.

# Second Harmonic Generation in a Fully Centro-Symmetric Plasmonic Nanocircuit

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In recent years, scientists have shown growing interest in surface plasma. Due to their distinct characteristics, surface plasma can demonstrate novel properties of the electromagnetic waves and thus provide the electric field enhancements with extremely high intensity. In previous works, it has been demonstrated that surface plasma can significantly enhance the optical signal which is highly dependent on the intensity of the electric field, especially in the case of nonlinear optical signals such as high order harmonic generations, Raman spectroscopy and two-photon fluorescence. Second harmonic generation (SHG) is one of the important project among them. Recent research has shown that the SHG signal generate by using surface plasma has many interesting features. However, almost all of the work on surface plasma generated second harmonic signal are study with localize surface plasmon (LSP), which although has very strong localized electric fields, the efficiency will be limited by the geometry of the structure due to the limitation of the SHG on symmetric environment. However, there has another type of surface plasma, which is called surface plasmon polariton (SPP). The primary aim of this paper is to analyze the effect of using SPP on a two-wire transmission line, which is a plasmonic waveguide to generate SHG signal and demonstrated that an optical mode of correct symmetry is sufficient to allow SHG even in centro-symmetric structure made of centro-symmetric material.

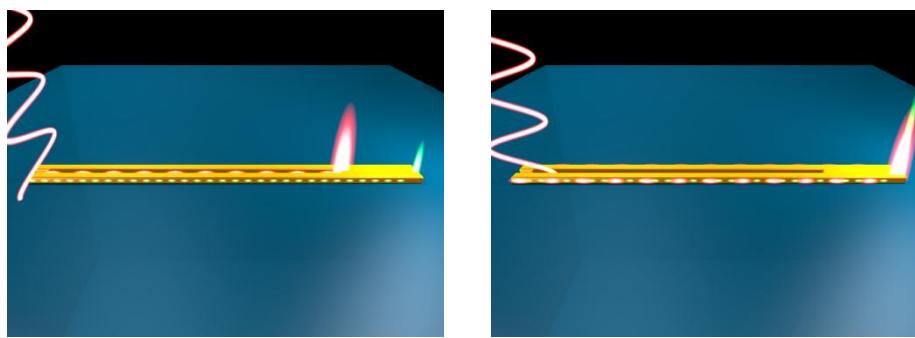


Fig 1. Schematic representation of using SPP to generate SHG signal on a two-wire transmission-line by both symmetric mode and anti-symmetric mode

# **Ultrafast electronic and magnetic dynamics in topological materials**

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Topological materials, for example topological insulators, Weyl semimetals and magnetic Weyl semimetals, have exotic bulk and boundary electronic states that are promising platforms for future spintronics and quantum computation. These states exhibit unique dynamics and even transform into new topological orders under femtosecond optical excitations. In this talk, I will present our recent work on probing the ultrafast electronic and magnetic dynamics of topological insulators, Weyl semimetals and magnetic Weyl semimetals using time-and-angle resolved photoemission spectroscopy (Tr-ARPES) and time-resolved magneto-optical Kerr effect (Tr-MOKE). I will discuss the interesting behavior of their electronic and magnetic order that are only accessible through optical excitations.

# Surface Dirac Fermion Dynamics of Non-magnetic and Magnetic Topological Insulators

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A three-dimensional (3D) topological insulator (TI) hosts spin-polarized massless Dirac states along its two-dimensional (2D) topological surface state (TSS) due to the topology of the bulk band structure. The Dirac-cone-like TSS exists as long as time-reversal symmetry is preserved and exhibits novel phenomena not found in conventional 2D metals. Thus, 3D TIs have attracted much attention as key materials to revolutionize electronic, spintronic and optoelectronic devices. Furthermore, magnetically doped TIs, possessing an energy gap created at the Dirac node through time-reversal symmetry breaking, are predicted to exhibit the quantized anomalous Hall effect (QAHE). Although several candidates of magnetic TIs were demonstrated to show long-range magnetic order, the realization of the QAHE is so far restricted to V- and Cr-doped  $(Sb,Bi)_2Te_3$  systems at extremely low temperature [1,2]. It is, therefore, very important to understand the interaction between the host and the magnetic dopants to raise the critical temperature of the QAHE. Nevertheless, the influence of the magnetic impurities to the TSSs remains unexplored.

Here, first, we investigate the ultrafast surface dynamics of bulk-carrier-tunable TIs  $(Sb_{1-x}Bi_x)_2Te_3$  by using time- and angle-resolved photoemission spectroscopy (TARPES). Our TARPES studies have disclosed several functions, such as >400-ps prolonged duration, population inversion, and surface photovoltage effect, on the nonequilibrated surface of TIs [3,4].

The second part of my talk will be focused on magnetic TIs. We systematically investigate the magnetic dopants induced modifications to electronic structures from the viewpoints of magnetic, structural and electronic properties and the ultrafast carrier dynamics in a series of  $Sb_{2-x}V_xTe_3$  ( $x = 0, 0.015, 0.03$ ) by a combination of x-ray magnetic circular dichroism (XMCD), scanning tunneling microscopy/spectroscopy (STM/STS) and TARPES. The XMCD experiment has revealed that the host Te 5p and Sb 5p electrons carry magnetic moments that mediate the localized V 3d spins. Furthermore, the STM and its differential conductance spectroscopy resolved two independent V-induced impurities, which are energetically overlapped with the TSS. More importantly, when the doping amount of V was increased, the electronic recovery time for the nonequilibrated state was drastically shortened to <500 fs. The observed shortened duration indicates an increased impurity scattering paths between the host and the magnetic dopants [5].

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# Ultrafast Optical Spectroscopy of Individual Nano-materials with Defined Atomic Structure

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When the characteristic length of a material shrink to 1 nm scale, many distinct physical phenomena, such as quantum confinement, enhanced many-body interactions, strong van der Waals inter-material couplings and ultrafast charge separation, will appear. To investigate the related fascinating low-dimensional physics, we need a tool to quantitatively link the atomic structures to the physical properties of these very small nano-materials. In this talk, I will introduce our recently developed in-situ TEM + high-sensitive ultrafast nano optics technique, which combines capability of structural characterization in TEM and property characterization in nano optics on the same individual nano-materials. Several examples of using this technique to study the interlayer quantum coupling and ultrafast dynamics in 1D carbon nanotube system and 2D atomic layered systems will be demonstrated.

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# Charge and Energy Transfer Dynamics at 2D Ultimate Proximity

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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.

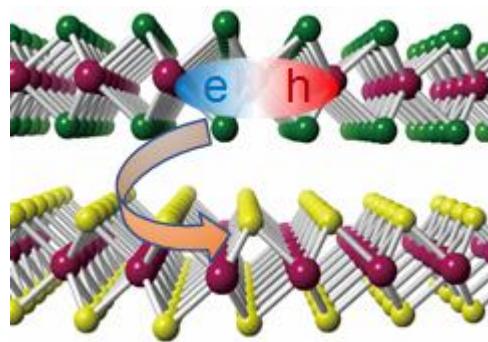


Fig. 1 Charge and energy transfer process between two layered materials coupled by van der Waals force.

# Magnetically-Sensitive Valley Polarization Reversal and Revival of Defect-Bound Excitons in WSe<sub>2</sub>-WS<sub>2</sub> Heterostructure

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A tunable, large valley polarization with a long valley lifetime is necessary to various valleytronic applications of the transition metal dichalcogenides (TMDs). However, the valley lifetime of free exciton(s) in monolayer TMDs is limited to a few picoseconds, and the valley properties can hardly be modulated by a magnetic field with moderate intensities. Therefore, constructing advanced architectures which possess low-field-tunable higher valley polarization and longer valley lifetime is exceptionally desirable. Very recently, it was found that certain defect-bound exciton could extend the valley lifetime to few microsecond in monolayer WSe<sub>2</sub>, which is exciting to the community. However, it remains challenging to tune the valley properties of defect-bound excitons. We find that the defect-bound interlayer (IX<sub>def</sub>) and intralayer (X<sub>def</sub>) excitons in an AB-stacked WSe<sub>2</sub>-WS<sub>2</sub> heterobilayer possess at least the microsecond valley lifetimes, and exhibit unprecedented magnetically-tunable valley properties: The valley polarization of IX<sub>def</sub> keeps reversed and can be surprisingly enhanced from -10% to -60% by a small out-of-plane magnetic field ( $\pm 0.4$  T), showing an anomalous and exceptionally-sharp  $\Lambda$ -shape as a function of the field. In contrast, the valley polarization of X<sub>def</sub> can revive after a fast decay and can be readily tuned under a moderate magnetic field ( $> \pm 1$  T), following the direction of the field and showing an X-shape. Our results demonstrate that the valley pseudospin of defect-bound excitons can be efficiently manipulated by a small magnetic field and the interlayer interactions, which will shed new light on both the understanding and the manipulating methods of the valley dynamics in TMDs and their heterostructures.

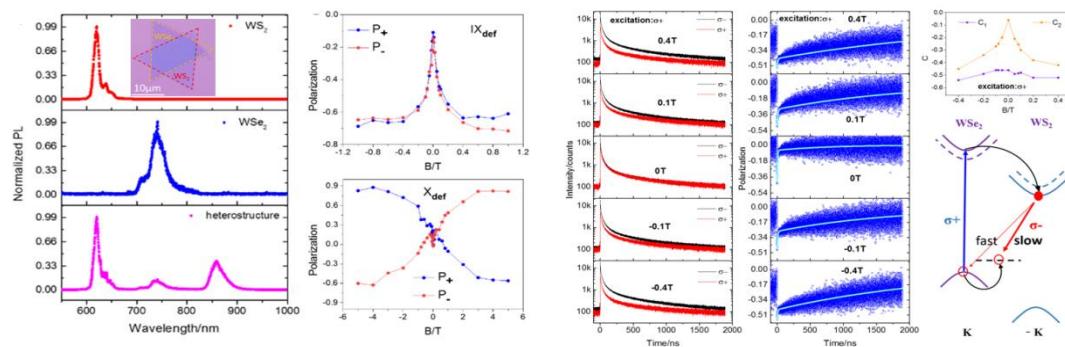


Fig. 1 Magnetically-sensitive valley properties of defect-bound excitons in WSe<sub>2</sub>-WS<sub>2</sub> heterobilayer.

# Ultrafast control of electron inside of graphene and across solid-state interfaces

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Graphene with its exceptional optical and electronic properties is a unique material for light-field controlled electron dynamics inside of a (semi-)metal. Its Dirac cone dispersion relation is ideally suited to drive intraband currents and interband transitions by the optical field of phase-controlled few-cycle laser pulses [1,2]. In particular, when the influence of the intraband dynamics to the interband transition cannot be neglected, this combined dynamics turn into a novel non-perturbative regime. Based on the coupled nature of the intraband and interband processes, we observe repeated coherent Landau-Zener transitions between valence and conduction band separated by around half an optical period of  $\sim 1.3$  fs. Because of the extremely fast dynamics, fully coherent Landau-Zener-Stckelberg interferometry takes place. Moreover, we could show complex electron trajectory control in the graphene plane using two perpendicular polarized light fields. That way we can turn on and off subsequent coherent ultrafast Landau-Zener transitions.

In the second part of the talk we combine graphene with the bulk material silicon carbide and show that the charge transfers across the graphene-SiC solid-state interface takes place within 300 attoseconds, which is the fastest charge transfer observed across a solid-state interface.

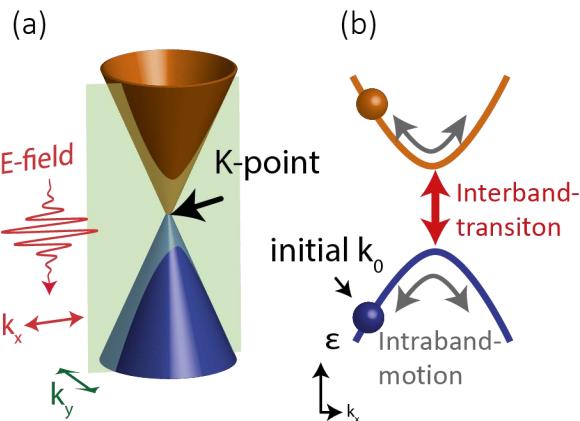


Fig. 1. (a) Electronic band structure of graphene around the Dirac point (K-point). The green area indicates a condition where both interband transition and intraband motion is present. (b) Combined interband and intraband dynamics.

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[2] C. Heide, T. Higuchi, H. B. Weber, P. Hommelhoff, Phys. Rev. Lett. 2018, 121, 207401.

# Final state effective mass affects attosecond photoemission delays in BiTeCl

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V. M. Silkin<sup>6,7,9</sup>, N.M. Kabachnik<sup>6,10,11</sup>, A.K. Kazansky<sup>6,7,9</sup>, P.M. Echenique<sup>6,7,12</sup>,  
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<sup>10</sup>Skobeltsyn Institute of Nuclear Physics, 119991 Moscow, Russia

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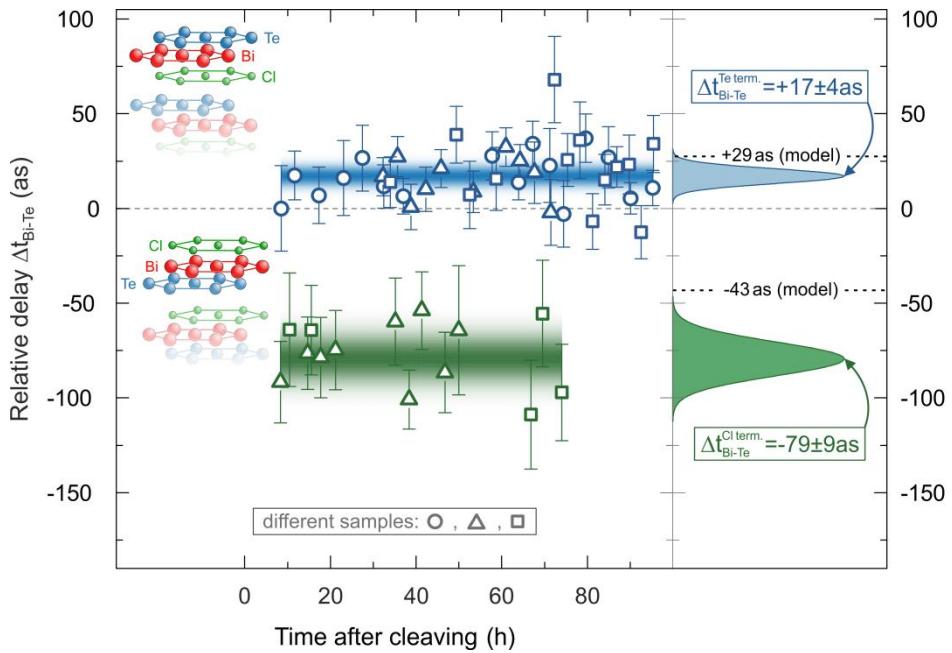
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Attosecond time-resolved photoemission based on the photoelectron streaking in a time-correlated strong IR field allows investigating temporal delays in the photoemission from different initial states. Discrepancies between experimental observations and existing theoretical models advance our understanding of mechanisms that determine the photoemission kinematics and, for example, allowed recently the identification of an intra-atomic delay as significant contribution to the total photoemission delay [1].

Here we report on attosecond time-resolved photoemission from the layered and non-centrosymmetric crystal BiTeCl. The lack of inversion symmetry allows studying relative photoemission delays for differently terminated but well-defined and inert surfaces with identical bulk properties. Furthermore, the different layer stacking enables the experimental determination of the inelastic mean free path (MFP). This reduces the ambiguities for electron trajectory calculations performed to model the observed relative photoemission delays and thus allows drawing qualitative conclusions from the experimental results.

The comparison with results from classical and quantum mechanical electron trajectory calculations results in a significant discrepancy for both terminated surfaces (Fig. 1). The modeling includes the experimentally observed MFP ( $\approx 3.5 \text{ \AA}$ ), the screened electron-hole interaction using a Yukawa potential, intra-atomic corrections within the Eisenbud-Wigner-Smith theory, and the dynamic atomic-scale screening of the IR-field for both surface terminations. The discrepancy between model and experiment is robust against the variation of model parameters within reasonable limits. This indicates that the only missing contribution in the model, i.e. the effective electron mass in the final states, is responsible for the observed discrepancies.



**Fig. 1.** Left panel: Experimentally observed relative streaking delays between the emissions from the core levels Bi-5d and Te-4d as a function of the time after cleaving for both surface terminations (Te-term.: blue, Cl-term.: green) and different samples ( $\circ$ ,  $\triangle$ ,  $\square$ ). Right panel: Comparison between the averaged delays from the left panel and the results from electron trajectory calculations (dashed lines).

1. F. Siek, S. Neb, P. Bartz, M. Hensen, C. Strüber, S. Fiechter, M. Torrent-Sucarrat, V. M. Silkin, E. E. Krasovskii, N. M. Kabachnik, S. Fritzsche, R. D. Muñoz, P. M. Echenique, A. K. Kazansky, N. Müller, W. Pfeiffer, U. Heinzmann, *Science*, **357** (2017)

# Equivalence of RABBITT and Streaking Delays in Attosecond-Time-Resolved Photoemission Spectroscopy at Solid Surfaces

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The dynamics of the photoelectric effect in solid-state systems can be investigated via attosecond-time-resolved photoelectron spectroscopy [1]. We provide a theoretical comparison of the delay information accessible by the two most important techniques, attosecond streaking spectroscopy and reconstruction of attosecond beating by interference of two-photon transitions (RABBITT) at solid surfaces, respectively.

The analysis is based on simulated time-resolved photoemission spectra obtained by solving the time-dependent Schrödinger equation in a single-active-electron approximation. The solid is described by a jellium-type potential including electron-hole interaction [2].

We show a continuous transition from the few-cycle RABBITT regime to the streaking regime as two special cases of laser-assisted photoemission (Fig. 1 and 2). The absolute delay times obtained by both methods agree with each other, within the uncertainty limits for kinetic energies  $>10$  eV. Moreover, for kinetic energies  $>10$  eV and for vanishing IR field inside the solid, the streaking delay time  $\tau_{\text{streaking}}$  and the RABBITT delay time  $\tau_{\text{RABBITT}}$  coincide with the classical time of flight  $t_{\text{PE}}$  for an electron propagating from the emitter atom to the surface, with only small deviations of less than 4 as due to quantum mechanical interference effects [3].

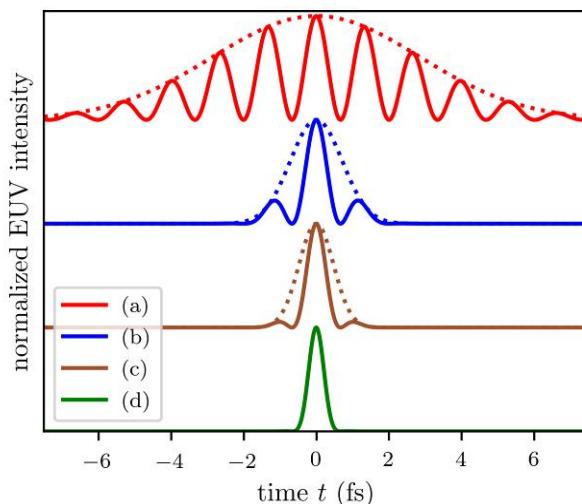


Fig. 1: EUV pulse profiles in the time domain corresponding to EUV spectra of fig. 2.

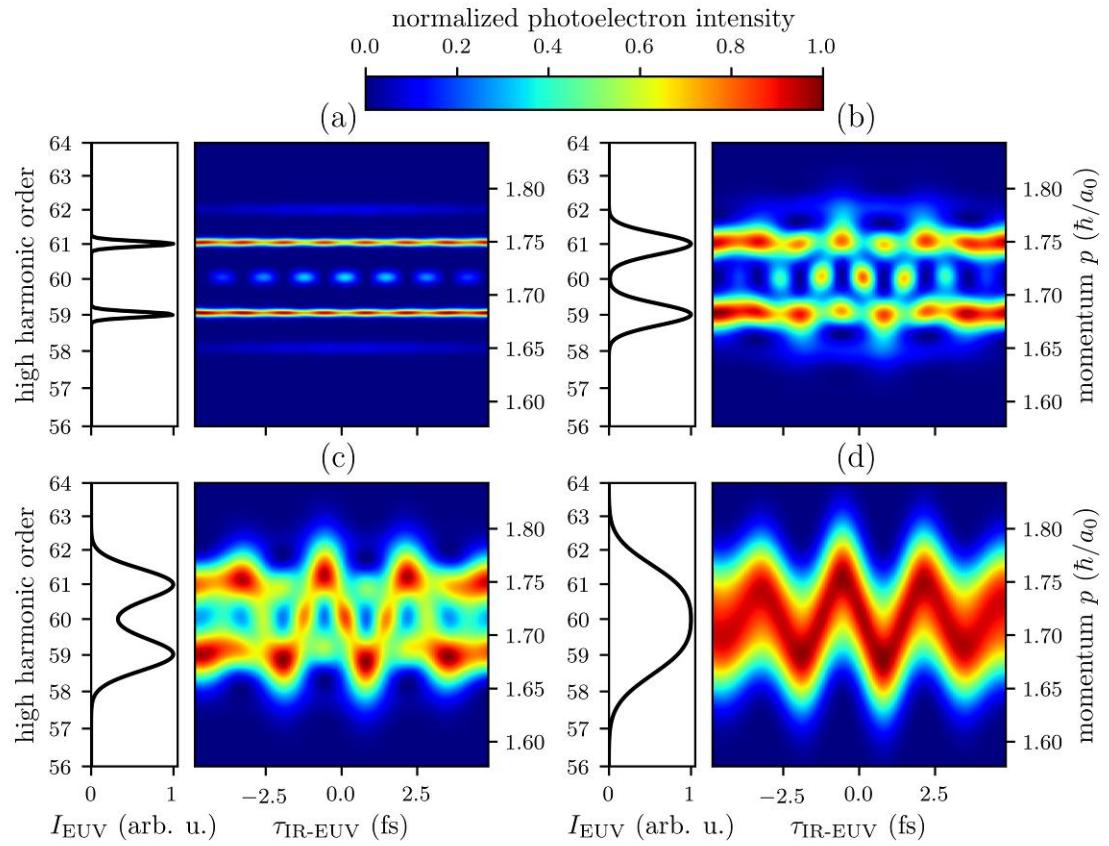


Fig. 2: Simulated photoelectron spectrograms (color plots) corresponding to four different EUV spectra (black lines). Each corresponding EUV excitation pulse (train) is given by the Fourier transform of the EUV spectra (Fig. 1). All spectra were normalized individually. IR pulses were treated independently with center wavelength at 800 nm, 5 fs pulse duration and 1 V/nm electric field peak amplitude.

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# Ultrafast carrier relaxation and Pauli drag effect in photoexcited solids

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Ultrafast light-matter interaction is a powerful tool for the study of solids. Upon laser excitation, carrier multiplication and lattice acceleration beyond thermal velocity can occur, as a result of far-from-equilibrium carrier relaxation. The roles of electron-electron and electron-phonon scatterings are identified by first-principles electron dynamic simulations, from which a unified phase diagram emerges (Fig. 1). It not only explains the experimentally-observed “inertial” melting, but also predicts an abnormal damping by Pauli Exclusion Principle with a new perspective on ultrahigh-intensity laser applications.

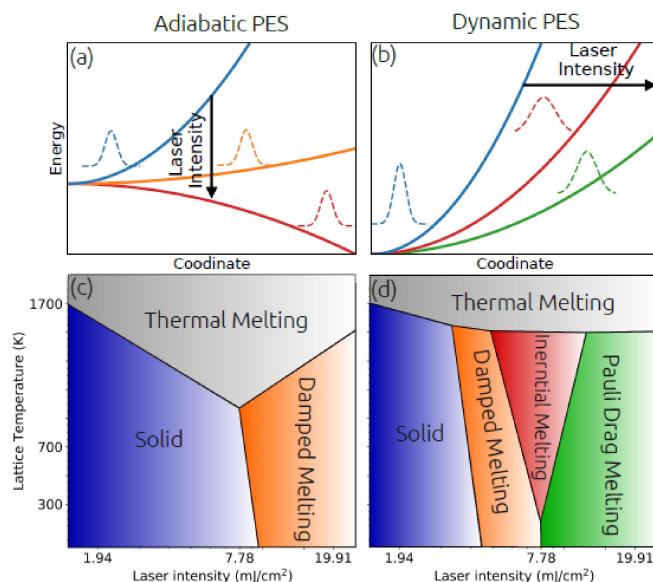


Fig. 1 Schematic phase diagrams for ultrafast laser melting under (a,c) adiabatic potential energy surfaces (PES) and (b, d) dynamic PES, respectively.

# ***Ab initio* Nonadiabatic Molecular Dynamics Investigations on the Excited Carriers in Condensed Matter Systems**

**Qijing Zheng<sup>1</sup>, Weibin Chu<sup>1</sup>, Chuanyu Zhao<sup>1</sup>, Lili Zhang<sup>1</sup>, Hongli Guo<sup>1, 2</sup>, Yanan Wang<sup>1</sup>, Xiang Jiang<sup>1</sup>, and Jin Zhao<sup>\*1, 3, 4</sup>**

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<sup>2</sup>*School of Physics and Technology, Center for Nanoscience and Nanotechnology, and Key Laboratory of Artificial Micro- and Nano-structures of Ministry of Education, Wuhan University, Wuhan 430072, China*

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The ultrafast dynamics of photo-excited charge carriers in condensed matter systems play an important role in optoelectronics and solar energy conversion. Yet it is challenging to understand such multi-dimensional dynamics at the atomic scale. Combining the real-time time-dependent density functional theory (RT-TDDFT) with fewest-switches surface hopping scheme, we develop time-dependent *ab initio* nonadiabatic molecular dynamics (NAMD) code Hefei-NAMD to simulate the excited carrier dynamics in condensed matter systems. Using this method, we have investigated the interfacial charge transfer dynamics, the electron-hole recombination dynamics and the excited spin-polarized hole dynamics in different condensed matter systems. The time-dependent dynamics of excited carriers are studied in energy, real and momentum spaces. In addition, the coupling of the excited carriers with phonons, defects and molecular adsorptions are investigated. The state-of-art NAMD studies provide unique insights to understand the ultrafast dynamics of the excited carriers in different condensed matter systems at the atomic scale.

# **Ultrafast Excited State Dynamics of Solids Probed by Time-resolved ARPES**

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Electronic excitations in solids may induce ultrafast dynamical processes at the atomic scale, including chemical surface reactions, coherent lattice excitations or ultrafast structural phase transitions. These processes are accompanied by pronounced changes of the electronic structure and occupation of electronic states. Recent advances in ultrafast spectroscopy allow direct probing of the underlying fundamental steps and provide a mechanistic understanding of transfer of energy from the electronic system into nuclear motions.

In my talk, I will discuss recent advances to probe such transient changes by a 500 kHz HHG based laser source enabling excited state band mapping throughout the complete Brillouin zone by time-resolved photoemission spectroscopy (trARPES) [1]. Such trARPES experiments provide detailed insights into excited states of black phosphorous, exciton dynamics in quasi-2D transition metal dichalcogenides (TMDCs) and photo-induced phase transitions in quasi-1D metal nanowires on In/Si(111). For the latter case we lay out a detailed reaction pathway including temporally separated transitions of electronic and atomic structure and demonstrate that the excited state population dynamics of the transient band structure is described by a thermalized electron distribution function [2].

Acknowledgements: This work was performed in collaboration with C.W. Nicholson, M. Puppin, S. Dong, R. Xiang, S. Beaulieu, M. Dendzik, L. Rettig and R. Ernstorfer (FHI Berlin) and A. Lücke, W.G. Schmidt (University of Paderborn).

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# Anisotropic Optical Response of Black Phosphorus Studied by Multiphoton Photoemission Spectroscopy

Shijing Tan,<sup>1</sup> Hongli Guo,<sup>2</sup> Min Feng,<sup>2</sup> Limin Cao,<sup>2</sup> Jin Zhao,<sup>1</sup> and Hrvoje Petek<sup>3\*</sup>

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Tunable direct bandgap and anisotropic carrier mobility are two main characteristics that make black phosphorus (BP) an excellent two-dimensional (2D) material for optoelectronic applications. We study the bandgap and the anisotropic optical response of BP by tuning the alignment between the optical electric field and the high symmetric axis of BP with a broadly tunable femtosecond laser from 350-900 nm. Multiphoton photoemission (mPP) spectroscopy with UV excitation images the distribution and dispersion of both the valence band maximum (VBM) and conduction band minimum (CBM), which form a direct bandgap of ~0.6 eV at the Z point. The intensity of CBM exhibits a mirror symmetric distribution with a maximum along the zigzag and a minimum along the armchair axis of the 2D crystalline structure. The intensity of VBM, however, presents a perplexing anisotropic distribution upon rotation by 180° (Fig. 1b). By tuning the photon energy from visible to near IR range, the image potential state (IPS) of BP appears prominently in the mPP spectra through resonant two- and three-photon excitation. The dependence of IPS distribution on crystalline structure, optical alignment and resonant excitation pathways will be discussed.

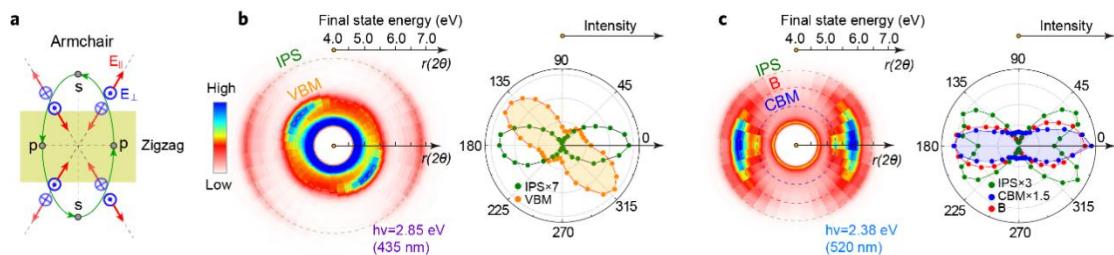


Fig. 1. **a**, A schematic drawing of the alignment between the optical electric field and the high symmetric axes of BP. **b,c**, polar map of polarization dependent mPP spectra excited with  $h\nu=2.85$  eV and 2.38 eV, respectively. The  $r$  denotes the  $E_{final}$  and the intensity is expressed by color and normalized at the secondary electron emission edge.

# **Excited-States Lifetime Control in Emerging Low-Dimensional Materials**

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Low-dimensional materials enabled photonics and optoelectronics are rapidly becoming a preferred avenue for next-generation information devices, promising both significant performance improvement and the potential for disruptive, novel-concept devices. Understanding the photocarrier dynamics, which is fundamentally linked to many of the figures of merits of photonic devices, is the key to develop high-performance devices and also one of the most fundamental tasks of ‘light-matter interaction’ research.

While it is highly desirable to be able to customize the photocarrier dynamics to adapt to various device applications, approaches developed for conventional materials are not directly applicable and such a task remains a significant experimental challenge. In this talk, recent progress in controlling the photocarrier lifetimes in three representative material classes (including graphene-like Dirac materials, Monolayer transition-metal dichalcogenides, as well as 2D semiconductor heterostructures) will be presented. Photonic applications of these emerging materials in ultrafast optical switches, high speed photodetectors will also be touched upon.

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- [9] Appl. Phys. Lett. 113, 061104 (2018)
- [10] Appl. Phys. Lett. 111, 091101 (2017)

# Ultrafast Carrier Dynamics in a Twisted Bilayer Graphene Quasicrystal

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<sup>3</sup>Research Institute of Electromechanical Communication, Tohoku University, Japan

<sup>4</sup>Department of Applied Quantum Physics and Nuclear Engineering, Kyushu University, Japan

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A twisted bilayer graphene has attracted broad attention owing to its exotic phases including superconductivity and Mott insulating controlled by tuning the twisting angle [1]. Furthermore, it has been demonstrated that a 2D quasicrystal was observed at twisting angle of 30 degrees, which offered new opportunities to investigate many properties characteristic to quasi-periodicities [2]. Here, we have studied non-equilibrium carrier dynamics in a twisted bilayer graphene quasicrystal by time- and angle-resolved photoemission spectroscopy (TARPES) using extreme ultraviolet (XUV) pulses obtained from high-harmonic generations (Fig. 1a). Figures 1b and 1e show the ARPES images for the upper- and lower-layer Dirac bands in equilibrium, respectively. After infrared (IR) photo-excitation (800 nm, 0.7 mJ/cm<sup>2</sup>), both bands exhibits immediate carrier transfer from occupied to unoccupied bands at the delay time ( $Dt$ ) of 0.05 ps (Figs. 1c and 1f), and excited carriers quickly relax to the original states at  $Dt = 0.16$  ps (Figs. 1d and 1g). In the presentation, I will show the further results with comparing replica bands and a non-twisted bilayer graphene.

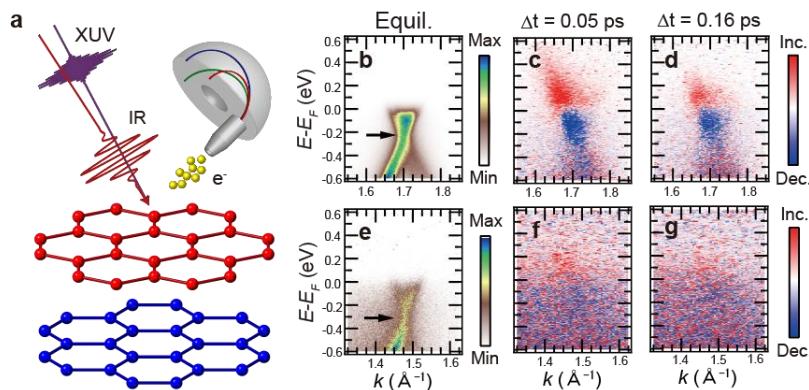


Fig. 1 a. Schematics of time- and angle-resolved photoemission spectroscopy (TARPES) for a twisted bilayer graphene quasicrystal. ARPES and difference TARPES images at the indicated delay times for the upper- (b-d) and lower-layer (e-g) Dirac bands.

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# Ultrafast Single-Molecule Electrical Detection

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A universal lithographic methodology for creating single-molecule devices based on carbon nanomaterials as point contacts has been developed. In this talk, I will detail our rational assay techniques by using bridge molecules with functional side groups capable of subsequent chemical/biocompatible assembly. We have tested this approach in chemical/biological systems, including DNA hybridization, aptamer-protein interaction, photocyclization, host-guest interaction, hydrogen-bond interaction, nucleophilic substitution ( $SN1$ ) and nucleophilic addition. Because it is constructed from a single molecule, each device can monitor individual binding events in real time. This methodology demonstrates a connection between electrical conduction and reaction dynamics that offers a glimpse into the future of integrated multifunctional sensors and devices.

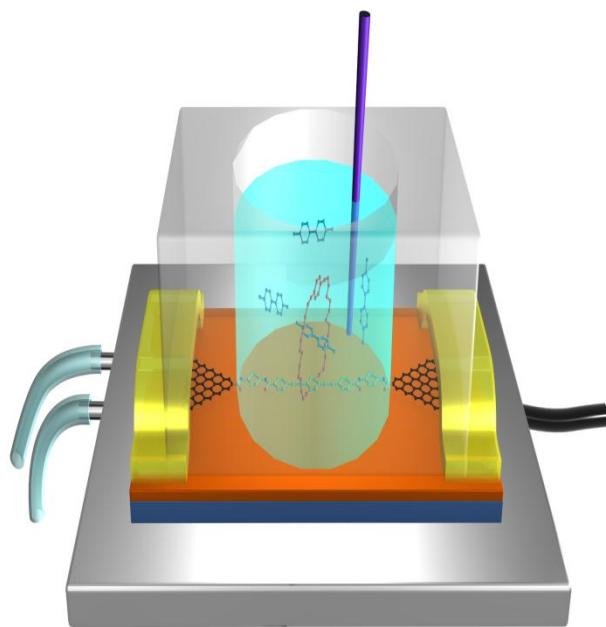


Figure1. Schematic of single-molecule electrical detection by using molecular electronic devices.

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# Electron Dynamics on Cu<sub>2</sub>O(111) Probed with Time-Resolved Two-Photon Photoemission

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Photocatalytic water splitting represents a sustainable way to store solar energy in the form of chemical bonds. For large-scale use, the combination of molecular catalysts supported on light-absorbing substrates is a viable approach. One material often used in such devices is cuprous oxide.

Due to its small direct band gap [1] it is an efficient absorber for solar light. Furthermore it is a p-type semiconductor with downward band bending towards the surface [2]. The related electric field leads to charge separation by accelerating photo-excited electrons towards the surface and holes into the bulk. Therefore the lifetime of free charge carriers is assumed to be long.

By means of time-resolved two-photon photoemission we have investigated the electronic structure and the electron dynamics of cuprous oxide on the (111) surface. On this particular surface two different reconstructions can be observed, a (1x1) and a (V3xV3)R30. For the (1x1) structure two electronic states of the conduction band could be observed at the  $\Gamma$ -point in good agreement with DFT calculations [3]. The lifetimes of photo-excited electrons in these two states is very different. A fast relaxation into the conduction band minimum (CBM) due to intraband and interband scattering takes place followed by a slow depopulation of the CBM. Moreover, a defect state located in the band gap was observed.

For the (V3xV3)R30 surface reconstruction, which is driven by defects in the surface layer, the properties change drastically. It seems that the band gap is completely filled with electronic states, probably with additional defect states. Therefore the electron dynamics is completely different in this case. Our current effort centres in understanding this change.

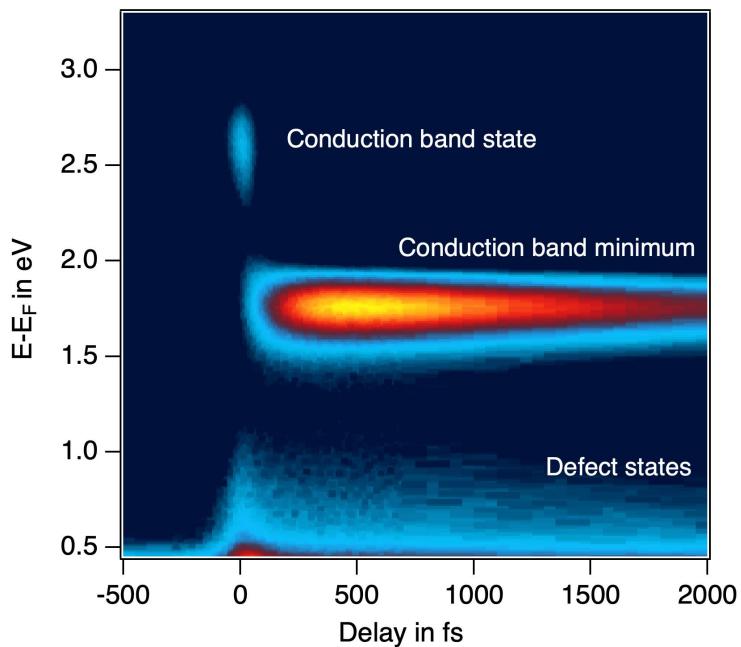


Fig. 1 Electronic structure of  $\text{Cu}_2\text{O}(111)$  at the  $\Gamma$ -point dependent on the delay between excitation pulse ( $h\nu=3\text{eV}$ , p-polarized) and probe pulse ( $h\nu=6\text{eV}$ , p-polarized). Two conduction band states and a defect state located in the band gap are observable with very different lifetimes.

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# Exciton-induced Chemical Reactions on Surfaces

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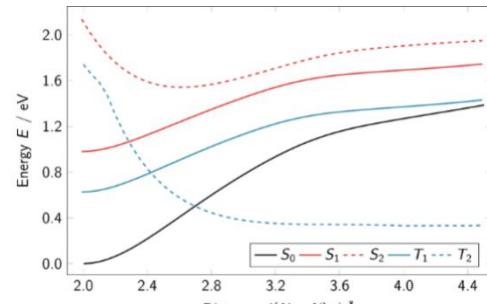
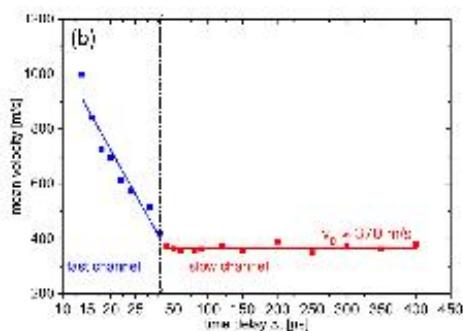
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In the last two decades hot carrier induced surface reactions initiated by femtosecond laser pulses have thoroughly been studied. The processes occurring after excitation of the system are typically described in a single hot carrier picture, which equilibrate via collisions with cold ones and eventually on a picosecond time scale by carrier-phonon interactions. Different dynamics and couplings may occur when long-lived excitonic states in the bandgap of semiconducting materials are populated. Then, energetic bound electron-hole pairs can interact across interfaces and with adsorbed molecules. High-energy bound excitons are present in fullerenes, carbon nanotubes, transition metal dichalcogenides, and other systems. We populated long-lived singlet and triplet excitons in a c(4x4) C<sub>60</sub> film on Cu(111). These excitons interacted with NO dimers adsorbed as cis-isomers, causing its dissociation and desorption from the surface. The desorbed NO molecules were analyzed with respect to their kinetic energy and with internal state selectivity.

Experimentally two different reaction dynamics are found, and supported by theoretical calculations. In a fast, prompt reaction channel the measured velocity of the NO molecules is correlated with the arrival time at the detection volume: fast molecules arrive first, slower later, as expected. This prompt channel is caused by charge transfer excitation from the HOMO centered at cis-(NO)<sub>2</sub> to singlet states of C<sub>60</sub> leads via the trans configuration to a low dissociation barrier of only 80 meV along the N – N bond.

In a slow channel to which the vast majority of the reacted molecules belong, all molecules show the same velocity, despite the fact that their arrival times at the detection volume changes from 40 μs to 400 μs. We show experimentally that the reaction is delayed over times which correspond to the average lifetimes of triplet excitons. Theoretically, the T<sub>2</sub> state is dissociative and shows a conical intersection with the T<sub>1</sub> state with a low barrier of 170 meV. Thus, triplet excitons of C<sub>60</sub> diffuse to the NO dimer adsorbate where the overall T<sub>1</sub> and T<sub>2</sub> states are formed causing the dissociative desorption of NO.



# Unique dielectric responses in lead-halide perovskites: Proposal of ferroelectric polaron

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Lead halide perovskites (LHPs) have become star materials for new-generation solar cells due to their exceptional tolerance to defects, which gives rise to long carrier lifetimes, long diffusion lengths, and low electron-hole recombination rates. Those remarkable optoelectronic properties can be explained by efficient charge screening along with polaron formation.<sup>1,2</sup> We detect the large polaron formation directly using Time-resolved optical Kerr effect (TR-OKE).<sup>3</sup> Dynamic responses from the inorganic sub-lattice detected in time domain uncovered that charge screening happens in less than 1 ps in both hybrid  $\text{CH}_3\text{NH}_3\text{PbBr}_3$  and all-inorganic  $\text{CsPbBr}_3$ . In the case of  $\text{CsPbBr}_3$ , coherent oscillations were clearly observed in non-resonant/pre-resonant pumping with changing the frequency of oscillation lower as getting closer to the band-gap.

Even more interesting, the oscillatory responses are smeared out in charge injection regime (Fig. 1a).

Such unusual responses motivated us to propose a new type of polaron: ferroelectric large polaron. Due to the huge anharmonicity of the lattice responses, the lattice surrounding an injected carrier might undergo ferroelectric phase transition due to the electric field exerted from the carrier, which results in much stronger screening than conventional Fröhlich polaron picture (Fig. 1b,c).<sup>4</sup>

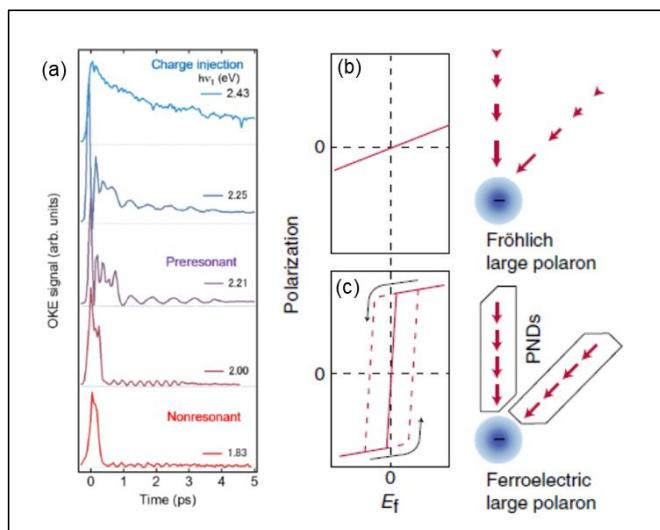


Fig. 1 (a) TR-OKE responses from a  $\text{CsPbBr}_3$  single crystal as a function of excitation photon energy. (b,c) Schematics of electric-field-polarization relationship for (b) Fröhlich polaron and (c) Ferroelectric polaron.

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# Dialing into dynamically disordered lattice potentials in CsPbBr<sub>3</sub> via 2D coherent phonon spectroscopy

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Lead halide perovskites (LHPs) have emerged as new darlings of materials science with broad potential for high performance solar cells and light emitting devices. The dynamically disordered and soft perovskite lattice is found to be a key component for their extraordinary optoelectronic properties<sup>1–5</sup>. The lattice potential is strongly anharmonic and fluctuates spatially and temporally, leading to a multitude of exciting phenomena such as (dynamic) Rashba effect<sup>6–8</sup>, ferroelectricity<sup>9,10</sup>, large polarons<sup>11,12</sup>, and ferroelectric polarons<sup>2,13</sup>. The unifying goal of these studies is finding the fundamental mechanism of charge carrier protection in LHPs, but so far, specific electronic state coupled lattice potentials have eluded experimental probes.

Here, we extend our time-resolved optical Kerr effect (TR-OKE) studies<sup>2</sup> of the all-inorganic CsPbBr<sub>3</sub> perovskite to two-dimensions (2D-OKE), which allows us to establish the phonon response with high excitation energy resolution. We use this time-resolved spectroscopy to investigate the coherent lattice response in the highly dilute charge injection regime, i.e. in the weak absorption tail below the electronic band gap. Additional discrimination against incoherent signals leads to purely coherent phonon sensitive detection of the charge induced polarization anisotropy.

We find a very effective displacive excitation of coherent phonons over a wide spectral range. Surprisingly, we witness an extreme pump energy-dependent shift in phonon frequency compared to the equilibrium normal mode analysis through ab-initio calculations. These results unveil the energy-dependent mapping of strongly modified non-equilibrium lattice potentials, which are likely coupled to localized electronic states. Such phonon response may form the basis for the design principle of defect tolerant semiconductors from efficient dynamic screening.

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# Charge Separation Dynamics at Lead Halide Perovskite Interfaces Monitored by Differential Transient Transmission Spectroscopy

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Lead halide perovskite solar cells have been developing rapidly in the past few years, with their power conversion efficiency exceeding 22%. The perovskite solar cells typically consist of a perovskite film sandwiched between thin layers of electron- and hole-transport materials, which allow ultrafast separation of the charges photoexcited in the perovskite. We systematically investigate the charge separation dynamics at the interfaces of  $\text{CH}_3\text{NH}_3\text{PbI}_3$  ( $\text{MAPbI}_3$ ) with three typical organic and inorganic hole transport materials (HTMs), PTAA, PEDOT:PSS and  $\text{NiO}_x$ , by means of pump-probe transmission measurements [1]. We photoexcite only near the  $\text{MAPbI}_3$ /HTM interface or near the back surface of the 250-nm thick perovskite film, and measure the differential transient transmission between the photoexcitation on the two sides to extract the carrier dynamics directly related to the hole injection (Fig. 1a). The differential transmission signals directly monitor the hole injection from  $\text{MAPbI}_3$  to PTAA and PEDOT:PSS within 1 and 2 ps, respectively, whereas that to  $\text{NiO}_x$  comprises two steps and takes 40 ps to be completed (Fig. 1b). The obtained injection dynamics are in contrast with the previous reports on the hole injection dynamics to PTAA and  $\text{NiO}_x$ , whose time scales were sub-nanosecond and sub-picosecond, respectively. We discuss the hole injection dynamics in comparison with the device performance of the solar cells containing the same  $\text{MAPbI}_3$ /HTM interfaces based on their current-voltage curves and quantum efficiencies.

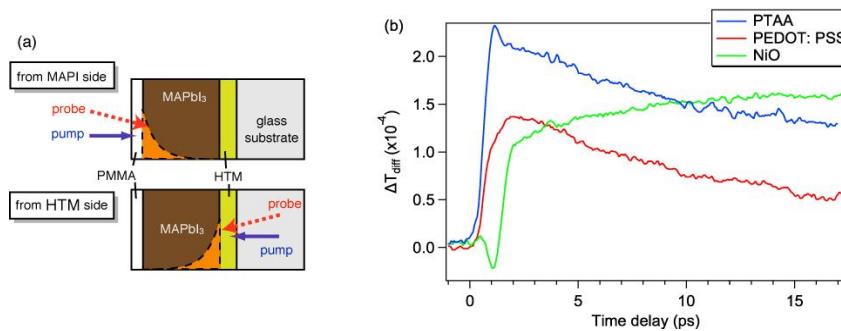


Fig. 1. (a) Schematics of the two configurations in the transient transmission measurements. (b) Differential transient transmission signals of  $\text{MAPbI}_3$  with different HTMs.

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# Coupled spin and lattice dynamics in lead halide perovskites

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Lead halide perovskites (LHPs) of the form APbHa<sub>3</sub> (A = organic CH<sub>3</sub>NH<sub>3</sub> or inorganic Cs; Ha = Br, I) exhibit long carrier lifetimes, despite being defect-rich direct-bandgap semiconductors, and high carrier mobilities despite their strongly ionic character. These properties make LHPs into promising novel materials for applications in thin-film solar cells and optoelectronics. The complex and unusual carrier dynamics in LHPs are supposedly related to the contained heavy elements and the associated strong spin-orbit coupling and low-frequency anharmonic phonon modes.

In spectroscopic [3] and transport [1] experiments using circularly polarized light, we demonstrated the occurrence of spin-polarized optical transitions below the direct band gap in the prototypical organic-inorganic LHP (CH<sub>3</sub>NH<sub>3</sub>)PbI<sub>3</sub>. Namely, the spin-polarized states give rise to a temperature-dependent circular photogalvanic effect at sufficiently high sample temperatures [1], i. e. above an order-disorder phase transition at 165 K. Our results imply a local and dynamical Rashba effect [2]. The symmetry-breaking field causing the dynamical Rashba effect arises from structural fluctuations, while the average bulk inversion symmetry is preserved.

To access the femtosecond spin and lattice dynamics at the surfaces of LHPs, epitaxial thin films are grown. We focus on purely inorganic CsPbHa<sub>3</sub> as model systems, complementing earlier studies on organic-inorganic LHPs [3, 4]. Static angle-resolved photoelectron spectroscopy is used to investigate the surface band structure of the films. The results are compared to the ones obtained from cleaved single crystals. Femtosecond dynamics are explored using time-resolved two-photon photoemission.

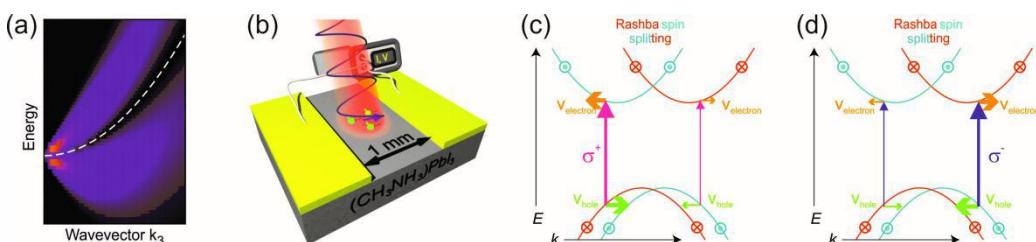


Fig. 1 (a) Dynamical Rashba effect in the conduction band of a semiconductor with strong spin-orbit coupling in the presence of thermal fluctuations [2]. (b) Experimental setup for identifying a circular photogalvanic effect, (c, d) illustrate the mechanism behind the effect [1].

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# Visualizing carrier transport in perovskites

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Slow cooling of hot polarons resulting from phonon bottleneck has been observed in hybrid organic-inorganic lead halide perovskites, pointing toward the potential for harvesting hot carriers to overcome the Shockley-Queisser limit. Open questions remain on whether the high optical phonon density from the bottleneck impedes the transport of these hot polarons. Here, we present a direct visualization of hot polaron transport in both single crystal nanoplate and polycrystalline lead halide perovskites utilizing ultrafast transient absorption microscopy with 300 femtosecond temporal resolution and 50 nanometer spatial precision. Remarkably, hot polaron diffusion does not suffer from but rather is enhanced by the presence of phonon bottleneck, opposite from what is observed in GaAs. Moreover, surface functionalization with phenethylammonium iodide increases the carrier diffusion coefficient of the near-surface carriers. We attribute the improved near-surface carrier diffusion to a more defect-tolerant character of the PEAI-terminated surface. Our work represents a leap forward in the understanding of surface chemistry and the underlying carrier transport properties in hybrid perovskites.

# How to Dress a Metal

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We rely on metals to perform veracious reflection of light. But what if light changes the electronic properties of a metal during the optical interaction? In fact, the electronic properties of solid state materials are not indelibly defined by the space-periodic interaction potentials of lattice ions as understood from the by Bloch theory, but can also be modified and manipulated by time-periodic potentials of strong optical fields, according to the Floquet-Bloch theory. Optical fields introduce a new degree-of-freedom to coherently manipulate, and turn-on transiently novel band properties of solids that do not exist under near-equilibrium conditions. Here we apply the coherent multidimensional photoelectron spectroscopy<sup>1</sup> to demonstrate the optical fields can manipulate the electronic structures of even the highest density optical materials, noble metals. We manipulate the surface bands of Cu(111) as a paradigm for how optical fields can tailor electronic band structures of solids. The optical field causes Autler-Townes splitting, deforming the surface electronic bands with a quasiparticle momentum-dependent resonance condition, such that modulating the effective band mass can transform electrons into holes on <10 fs time scale. More broadly, our research shows the interaction of electrons in a metal with optical fields. This is particularly poorly understood at epsilon near zero (ENZ) condition. We also examine the field-collective electronic response at ENZ. This research was performed by Marcel Reutzel and Andi Li.

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# Subcycle time-resolved ARPES of THz-driven Dirac currents in a topological surface band

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Lightwave electronics builds upon the idea that the electric field of intense light pulses can be used to accelerate electrons at terahertz to petahertz clock rates. Yet, in conventional semiconductors and dielectrics the finite band mass and ultrafast scattering limit the electron's ballistic excursion and velocity. Recently, we have shown that the unique band structure of topological insulators lifts these constraints [1]. In the first subcycle time-resolved ARPES experiment, we observe how the carrier wave of a THz pulse accelerates Dirac fermions in the topological surface state of Bi<sub>2</sub>Te<sub>3</sub>. While terahertz streaking of photo-emitted electrons traces the electromagnetic field at the surface, the acceleration of Dirac states leads to a strong redistribution of electrons in momentum space.

The experiments reveal that the electrons carrying the current react inertia-less on the accelerating field and travel with the Fermi velocity of 4.1 Å/fs or 410 nm/ps. The resulting surface currents reach peak densities as large as 2 A cm<sup>-1</sup>. Analysis with a semiclassical Boltzmann model shows that the relevant electron scattering times amount to at least 1 ps. This unique transport dynamics opens a realistic parameter space for lightwave electronics. Due to the large Fermi velocity, the low scattering rates, and the linear band structure, lightwave-driven Dirac fermions in TSSs may ballistically propagate in dispersion-free wavepackets over distances as large as several 100 nm. This distance easily exceeds the gate width of latest electronic transistors by orders of magnitude. Owing to spin-momentum locking, the ballistic Dirac currents also carry a spin current, which could enable spintronics up to optical clock rates.

As an experimental approach, we believe that our novel concept of subcycle THz-ARPES provides the most direct way of studying carrier transport in non-trivial band structures with high time resolution. We will briefly discuss perspectives as well as experimental difficulties of the technique.

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# Spin mixing and spin-wave excitations in 3d and 4f ferromagnetic metals

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Today's commonly accepted models of ultrafast magnetization dynamics are based on Elliot-Yafet spin relaxation [1] and spin transport [2]. In addition, recent works give evidence that magnons can drive ultrafast magnetization dynamics within 100 fs [3]. However, the nature of spin interactions and their dynamics in ferromagnetic metals is not well understood and only few studies could capture the coupling strength of electron-magnon interaction, which turned out to be surprisingly weak ( $\lambda \sim 0.2$ ) [4].

In this contribution we discuss various aspects of spin mixing and electron-magnon coupling in the 3d and 4f ferromagnetic metals, Fe, Gd and Tb. We utilized spin- and angle-resolved photoelectron spectroscopy to study the transient electronic structure, the temperature-dependent linewidths and the binding-energy renormalization of surface states interacting with magnons.

Ultrafast demagnetization in Gd depends on spin mixing of the valence bands. At low temperature and high spin polarization, we observe within the first 100 fs after 0.95 eV excitation an increase of the 5d exchange splitting in the bulk but its decrease at the surface. This is explained by optically-induced spin transfer [5] between the localized surface and delocalized bulk bands of Gd. For higher temperature spin mixing diminishes this ultrafast spin transfer consistent with an earlier onset of demagnetization. Comparing the subsequent magnetization dynamics in Gd and Tb we find clear evidence that sub-ps demagnetization in Tb is driven by coupling of 4f spins ( $S=3$ ,  $L=3$ ) to lattice excitations, i.e. spin-wave generation [6]. This channel of angular momentum transfer is missing in Gd ( $S=7/2$ , but  $L=0$ ). In a complementary spin-resolved photoemission study we investigated the magnetic phase transition following the 5d surface-state linewidth [7]. Surprisingly, contributions of electron-phonon and electron-magnon scattering to the linewidth are comparable for Gd and Tb.

For a thin iron film we studied the spin polarization and magnetic linear dichroism of a renormalized surface resonance. We find extraordinary strong electron-magnon coupling ( $\lambda \sim 4$ ), which separates dressed and undressed dispersion branches and manifests as magnetic dichroism rearranging the spectral weight between the two branches.

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# Spectroscopic investigation of Opto-spincurrent control

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Spincurrent plays one of the most substantial roles in spintronics. Both two classes of spincurrent, pure-spincurrent and spin-polarized current, are important. The former is a non-equilibrium distribution where electrons with spin up propagate in one direction whereas those with spin down propagate in the opposite direction and not accompanied by a net charge transfer. The latter is the charge transfer with the co-oriented spins. By using angle resolved high resolution one- and two- photon photoemission spectroscopy[1,2,3], we have investigated the opto-spin-current generated at surfaces via spin orbit interaction. In this talk, we have demonstrated that changing light polarization enable us to switch the opto-spin-current characteristics; opto-pure-spincurrent and spin-polarized photocurrent. The sample is Ir(111) covered by graphene. The Rashba-type band splitting has been found in the image potential states as well as the spin-split surface resonances in the occupied region. The optical direct transition between the spin-polarized states excited by the linear polarized light results in the opto-pure-spin-current, and the circularly polarized light excitation causes the spin-polarized photocurrent.

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# Critical behaviors in 20 fs govern the non-equilibrium laser-induced magnetic phase transition in Ni

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Manipulating material states with femtosecond laser illumination is a promising routine to create new states that are not accessible in equilibrium. To harness the power of possibility to engineer materials with light, it is key to understand how the energy transfers from photons to different degrees of freedom and how the hierarchy of the energy transfer shapes the behaviors of a material. In this talk, I will take the laser-induced ultrafast demagnetization in ferromagnetic metals as an example and show how the laser illumination leads to the loss of magnetic order in femtoseconds [1,2]. Here, by combining time- and angle-resolved photoelectron spectroscopy and ultrafast magneto-optical spectroscopy, we showed that the laser-induced ultrafast demagnetization in transition metal Ni is essentially of a *phase transition* of the coupled electron and spin system. In our results, we not only explained the fluence-dependent time constants, but also revealed that the critical phenomena, occurring in ~20 fs, play an important role in the phase transition, which includes the observation of the critical fluence to induce the phase transition and the divergence of the heat capacity at the critical point. To our knowledge, this is the first time such observations become available and set a new record for how fast the light and spin can be coupled in experiment. Surprisingly, our results indicate a strong connection between the non-equilibrium laser-induced dynamics and the material properties under thermal equilibrium, implying potential universality of our findings. By comparing with the ultrafast magneto-optical spectroscopy, we found possible coexistence of the transient ferro- and paramagnetic states at the critical point of the phase transition. This can be studied by the next-generation space-time-resolved spectroscopic technique.

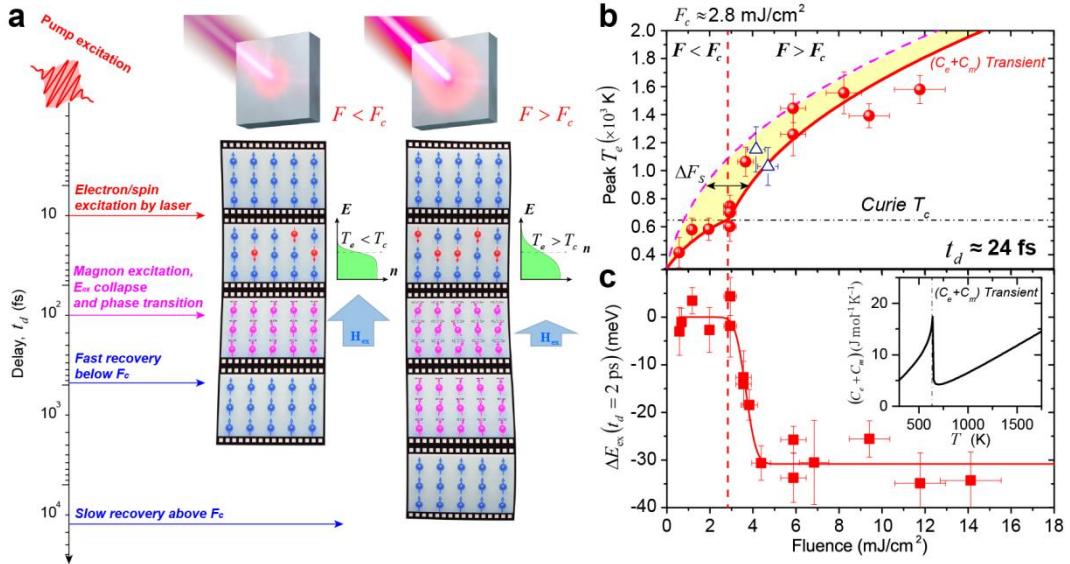


Fig. 1 a) Schematic of the critical behavior of ultrafast demagnetization in Ni. b) Peak electron temperature extracted  $\sim 24$  fs after excitation as a function of pump fluence, indicating a divergence of the heat capacity during the phase transition. c) Change of exchange splitting at 2 ps as a function of laser fluence at different sample temperatures. The agreement of the critical fluence ( $F_c$ ) in b) and c) indicates the connection of the electron dynamics to the magnetic properties of Ni.

- [1] P. Tengdin, W. You, C. Chen, X. Shi, D. Zusin, Y. Zhang, C. Gentry, A. Blonsky, M. Keller, P. M. Oppeneer, H. Kapteyn, Z. Tao, and M. Murnane, Sci. Adv. 4, eaap9744 (2018).
- [2] W. You, P. Tengdin, C. Chen, X. Shi, D. Zusin, Y. Zhang, C. Gentry, A. Blonsky, M. Keller, P. M. Oppeneer, H. Kapteyn, Z. Tao, and M. Murnane, Phys. Rev. Lett. 121, 77204 (2018).

# Coherent ultrafast spin transfer in ferromagnetic alloys

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The vision of using light to manipulate electronic and spin excitations in materials on their fundamental time and length scales requires new approaches in experiment and theory, to observe and understand these excitations. The ultimate speed limit for all-optical manipulation requires control schemes for which the electronic or magnetic sub-systems of the materials are directly manipulated on the timescale of the laser excitation pulse. In our work, we provide experimental evidence of such a direct, ultrafast and coherent spin transfer between two magnetic subsystems of an alloy of Fe and Ni. Our experimental findings are fully supported by time dependent density functional theory simulations and, hence, suggests the possibility of coherently controlling spin dynamics on sub-femtosecond timescales.

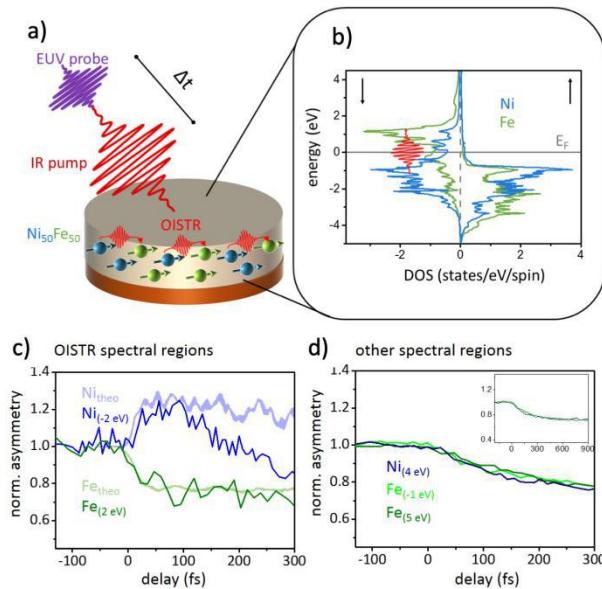


Fig. 1. a) Schematic overview of the optically-induced spin transfer (OISTR) experiment in FeNi. An EUV probe pulse investigates the element-specific magnetization dynamics in FeNi triggered by an IR pump. b) Projected density of states (pDOS) calculation for FeNi for Fe (green) and Ni (blue) demonstrating a favorable spin transfer from Ni to Fe in the minority channel. c) In the respective energy regions according to the calculations shown in b), the spin dynamics show a clear fingerprint of OISTR, on  $\approx 50$  fs time scales. d) In contrast, other spectral regions display only the conventional demagnetization.

# **POSTER CONTRIBUTIONS**

# Directional sub-femtosecond charge transfer dynamics and the dimensionality of 1T-TaS<sub>2</sub>

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For the layered transition metal dichalcogenide 1T-TaS<sub>2</sub>, we establish through a unique experimental approach and density functional theory, how ultrafast charge transfer in 1T-TaS<sub>2</sub> takes on isotropic three-dimensional character or anisotropic two-dimensional character, depending on the commensurability of the charge density wave phases of 1T-TaS<sub>2</sub>. [1] The X-ray spectroscopic core-hole-clock method prepares selectively in- and out-of-plane polarized sulfur 3p orbital occupation with respect to the 1T-TaS<sub>2</sub> planes and monitors sub-femtosecond wave packet delocalization. [2] Despite being a prototypical two-dimensional material, isotropic three-dimensional charge transfer is found in the commensurate charge density wave phase (CCDW), indicating strong coupling between layers. In contrast, anisotropic two-dimensional charge transfer occurs for the nearly commensurate phase (NCDW). In direct comparison, theory shows that interlayer interaction in the CCDW phase – not layer stacking variations – causes isotropic three-dimensional charge transfer. This is presumably a general mechanism for phase transitions and tailored properties of dichalcogenides with charge density waves.

[1] *Directional sub-femtosecond charge transfer dynamics and the dimensionality of 1T-TaS<sub>2</sub>*, D. Kühn, M. Müller, F. Sorgenfrei, E. Giangrisostomi, R. M. Jay, R. Ovsyannikov, N. Mårtensson, D. Sánchez-Portal & A. Föhlisch, *Scientific Reports*, 9:488 (2019).

[2] *Direct observation of electron dynamics in the attosecond domain*, A. Föhlisch, P. Feulner, F. Hennies, A. Fink, D. Menzel, D. Sanchez-Portal, P. M. Echenique & W. Wurth, *Nature*, 436:373–376 (2005).

# The Coherent Response of Ag/Cu(111) Surfaces to Optical Excitation

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Metal surfaces serve as nearly-perfect systems to study coherent light-matter interactions. Here, we apply the energy-resolved (ER) and interferometric time-resolved (ITR) multi-photon photoemission (mPP) technique to study the coherent nonlinear dynamics of Ag/Cu(111) surfaces excited by optical fields in the perturbative regime. We focus on the Shockley surface (SS) state of Ag/Cu(111) as a benchmark for spectroscopy.

By measuring the non-resonant 3- and 4-photon photoemission of the SS state, as well as its replica structures in the above-threshold photoemission (ATP), we correlate coherent polarizations and populations excited in the sample with final photoelectron distributions, where the interaction terminates. We also interpret the experimental results using an optical Bloch equation (OBE) model, which shows that the spectroscopic features of the 2D photoelectron spectra obeys a relation between the nonlinear orders of the coherent photoemission process  $m$  and the induced coherence  $n$ .

By measuring the resonant 4-photon photoemission between SS and image potential (IP) states, we observe the signature of optical dressing of surface band structures. By forming new eigenstates that are defined by the optical resonance condition and the external field strength, the AC Stark effect shifts the surface electronic band energies with quasiparticle momentum-dependent resonance condition and time-dependent optical field strength: the optical field distorts solid-state bands and can transform electrons into holes by modulating the effective band mass on <10 fs time scale.

# Electronic structure and ultrafast dynamics of $\alpha\text{-Fe}_2\text{O}_3$ (012) $(1\bar{1}02)$

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$\alpha\text{-Fe}_2\text{O}_3$  (or hematite) is the most stable iron oxide under standard conditions. Due to its vast abundance in earth's crust it plays a key role in geochemistry and corrosion processes. Moreover, as it catalyzes the water splitting reaction and has a favorable band gap of roughly 2.1 eV, it is considered as electrode material in photoelectrochemical cells. However, the probability of the photocatalytic reaction relies on the energy level alignment between excited states of hematite and water. In our study we investigate the (012) surface using static and time-resolved two-photon photoelectron spectroscopy. We discuss surface photovoltage effects and give first insights into electron dynamics of the clean hematite surface on an absolut energy scale.

# **Dynamics of Excited Hole at Au/TiO<sub>2</sub> Rutile(110) Interface**

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Photoinduced hot carriers transfer between gold nanoparticles (NPs) and semiconductor under plasmon excitation is an important phenomenon in photocatalysis. Electron-hole recombination velocity is the key factor of the photocatalytic device to drive redox reaction. In this study, we carried out nonadiabatic molecular dynamics calculations combined with real-time time-dependent density functional theory to investigate the effects of the number of Au atoms on hole transfer at Au/TiO<sub>2</sub> rutile(100) interface. It was found that Au<sub>3</sub> cluster was much better than single Au atom and one Au chain in hole trapping ability and electron-hole separation ability. This work provides insight into the rational design of plasmon-enhanced catalysts.

# Strong modification of the transport level alignment in molecular thin films after optical excitation

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Organic photovoltaic devices operate by absorbing light and generating current. These two processes are governed by the optical and transport properties of the organic semiconductor. Despite their common origin – the electronic band structure, their interplay between in organic materials is far from being understood.

In this work, we present new insights into the transient evolution of the valence band states of fullerene complexes on Ag(111) after optical excitation. This is achieved by time- and momentum- resolved photoemission (tr-ARPES) with a fs-XUV light source which allows us to simultaneously follow the transient evolution of the excited and occupied band structure after fs-optical excitation with visible light. Optical excitation of the fullerene  $C_{60}$  leads to transient changes of the linewidth of all molecular valence states which can be directly linked to the characteristic timescales of the exciton decay in  $C_{60}$ . Using model simulations, we demonstrate that the transient broadening of all molecular levels is mediated by the dielectric screening of the molecular film due to the formation of non-interacting excitons with different local charge distributions, i.e. to excitons with charge transfer (CT) character. These transient modifications of the energy level alignment are not limited to  $C_{60}$ , but can also occur for other endohedral fullerenes such as  $Sc_3N@C_{80}$  and reveal characteristic changes for different sample temperatures. In this way, our study shows that excitons of dominant CT character act as charge defects in organic thin films that severely influence the transient energy positions of the transport levels of the entire molecular film.

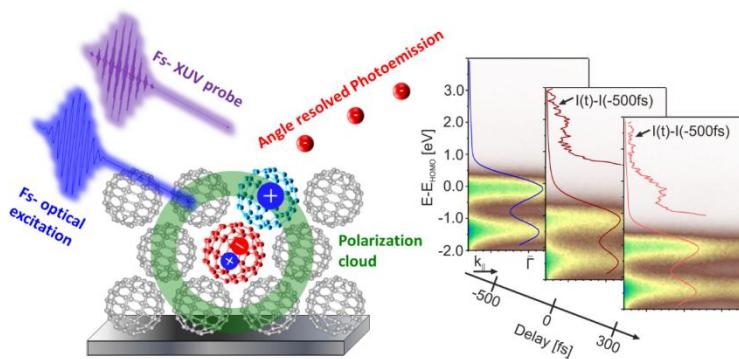


Fig. 1. The left side shows a schematic sketch of the experiment. A  $C_{60}$  multilayer film is optically excited by a fs-light pulse and the transient changes of the band structure are monitored by tr-ARPES. Typical tr-ARPES maps of the transient band structure of a  $C_{60}$  film are shown on the right side.

# 100-kHz Beamline with High Harmonic Generation for Photoemission at U.K. Artemis Facility

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At the U.K.'s Central Laser Facility (CLF) Artemis Facility, we provide a user facility for pump-probe angle-resolved photoemission spectroscopy (ARPES) studies, for interrogation of ultrafast dynamics in solid-state systems based on high-harmonic generation. Our system allows for the study of dynamics ranging from the fs to the ps regimes, with tunable polarisation in both the pump and probe.

We are presently in the process of upgrading our beamline, with the development of a new 100-kHz laser system. Our aim is to be able to offer time and energy resolution of around 50 fs and 100 meV, respectively, for probe energies of  $h\nu = 25 - 40$  eV; further, we plan to offer higher probe energies up to approximately  $h\nu = 100$  eV (for example, for pump-probe studies of shallow-lying core level states).

I will present here some of the details of our upgrade plans, and their consequences for the scientific scope of our facility. The new laser system will generate opportunities for time-resolved ARPES measurements with excellent time and energy resolution, and favourable statistics and space charge conditions relative to lower-repetition-rate systems.

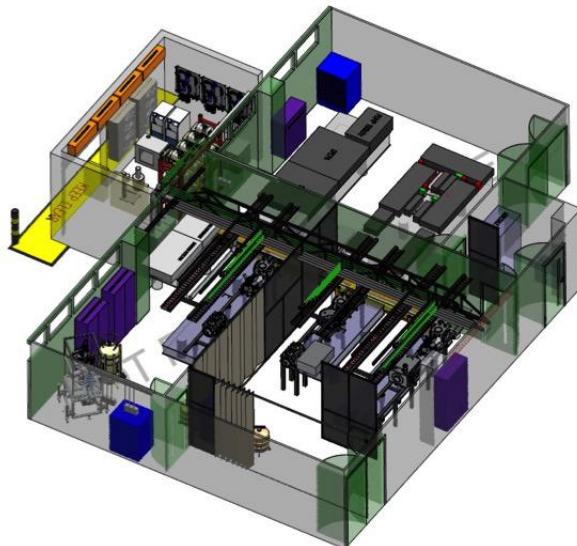


Fig. 1: Schematic layout of new lab space and beamlines.

# Femtosecond EUV Beamline for Ultrafast Materials Research and Development

Zhonghui Nie<sup>1</sup>, Ion Cristian Edmond Turcu<sup>1,2</sup>, Yao Li<sup>1</sup>, Xiaoqian Zhang<sup>1</sup>, Liang He<sup>1</sup>, Jian Tu<sup>1</sup>, Zhiqiang Ni<sup>1</sup>, Huangfeng Xu<sup>1</sup>, Yequan Chen<sup>1</sup>, Xuezhong Ruan<sup>1</sup>, Fabio Frassetto<sup>3</sup>, Paolo Miotti<sup>3</sup>, Nicola Fabris<sup>3</sup>, Luca Poletto<sup>3</sup>, Jing Wu<sup>4</sup>, Qiangsheng Lu<sup>5</sup>, Chang Liu<sup>5</sup>, Thorsten Kampen<sup>6</sup>, Ya Zhai<sup>7</sup>, Wenqing Liu<sup>8</sup>, Cephise Cacho<sup>9</sup>, Xuefeng Wang<sup>1</sup>, Fengqiu Wang<sup>1</sup>, Yi Shi<sup>1</sup>, Rong Zhang<sup>1</sup> and Yongbing Xu<sup>1,10</sup>

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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  $2.3 \times 10^{11}$  photons/second ( $2.3 \times 10^8$  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 1kHz, 4W to 1 MHz, kW repetition rate and average power respectively.

# Valley-polarized excitation in singly-oriented monolayer WS<sub>2</sub>/Au(111)

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Time- and angle-resolved photoelectron spectroscopy (trARPES) is employed to study the valley-selective excitation and near-surface dynamics of carriers in singly-oriented monolayer WS<sub>2</sub>/Au(111). Upon photoexcitation with circularly polarized light ( $\lambda \approx 590$  nm), we observe a selective population of both valence and conduction band at K and K', respectively, cp. Fig. 1. The quantitative analysis of the data indicates a valley polarization in the valence band of  $\approx 90\%$  which is consistent with the fraction of domains with mirror orientation as determined in diffraction experiments [1]. On the contrary, the valley polarization in the conduction band only reaches values of  $\approx 70\%$ . We explain the difference in the polarization by the spin-orbit splitting of the conduction band being much smaller in comparison to the splitting of the valence band and promoting intervalley scattering processes from K to K'. Different carrier dynamics of valence and conduction band population further hint to intervalley scattering being of relevance for the depolarization of the conduction band.

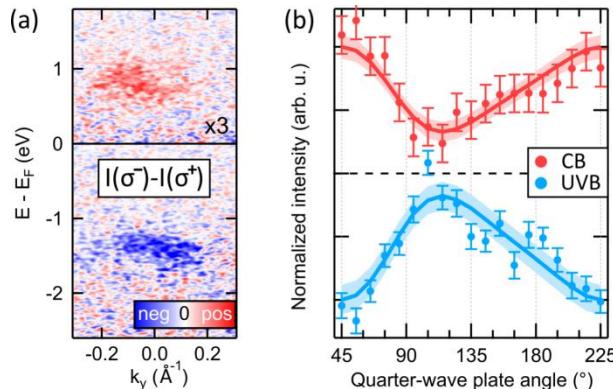


Fig. 1: TrARPES data of WS<sub>2</sub>/Au(111) at the K point at  $\Delta t = 50$  fs. (a) Difference intensity map generated from trARPES datasets recorded at excitation with left ( $\sigma^-$ ) and right ( $\sigma^+$ ) circularly polarized light, respectively, as set by a quarter wave plate. (b) Conduction band (CB) and upper valence band (UVB) intensities as a function of the quarter wave plate angle.

[1] L. Bignardi et al., Phys. Rev. Mat. **3**, 014003 (2019)

# Ultrafast Broadband Charge Collection from Clean Graphene/CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> Interface

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Photocarrier generation in a material, transportation to the material surface, and collection at the electrode interface are of paramount importance in any optoelectronic and photovoltaic device. In the last collection process, ideal performance comprises ultrafast charge collection to enhance current conversion efficiency and broadband collection to enhance energy conversion efficiency. Here, for the first time, we demonstrate ultrafast broadband charge collection achieved simultaneously at the clean graphene/organic-inorganic halide perovskite interface<sup>1</sup>. The clean interface is realized by directly growing perovskite on graphene surface without polymer contamination. The tunable two-color pump-probe spectroscopy, time-resolved photoluminescence spectroscopy and time-dependent density functional theory all reveal that the clean-interfacial graphene collects band-edge photocarriers of perovskite in an ultrashort time of ~100 fs, with a current collection efficiency close to 99%. In addition, graphene can extract deep-band hot carriers of perovskite within only ~50 fs, several orders faster than hot carrier relaxation and cooling in perovskite itself, due to the unique Dirac linear band structure of graphene, indicating a potential high energy conversion efficiency exceeding Shockley–Queisser limit. Adding other graphene superiority of good transparency, high carrier mobility and extreme flexibility, clean-interfacial graphene provides an ideal charge collection layer and electrode candidate for future optoelectronic and photovoltaic applications in two dimensions.

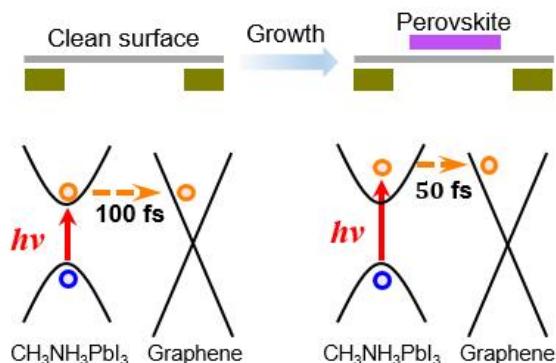


Fig. 1 Ultrafast broadband charge collection.

1. Hong H, et al. Ultrafast Broadband Charge Collection from Clean Graphene/CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> Interface. Journal of the American Chemical Society, 2018, 140, 14952.

# Lattice Disorder-engineered Energy Splitting between Bright and Dark Excitons in CsPbBr<sub>3</sub> Quantum Wires

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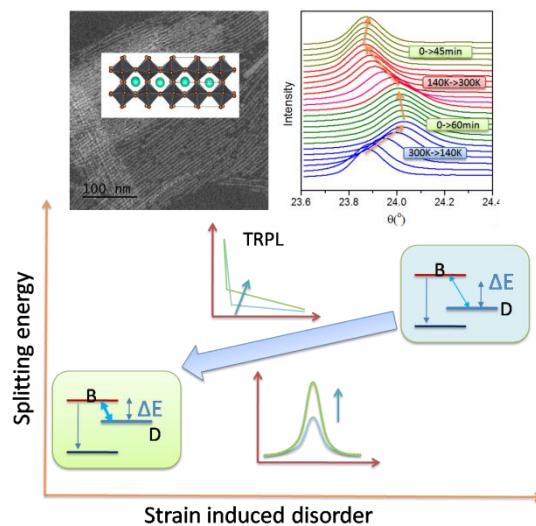
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Excitons in nanostructured semiconductors often undergo strong electron-hole exchange interaction, resulting in bright-dark exciton splitting with the dark exciton usually being the lower energy state. This unfavorable state arrangement has become the major bottleneck for achieving high photoluminescence quantum yield (PLQY). However, the arrangement of dark and bright exciton states in lead halide perovskites is under intense debate due to the involvement of many complicated factors. We present here the first experimental evidence to demonstrate that the strain is a crucial factor in tuning the energy splitting of the bright and dark excitons, resulting in different PL properties.



# Energy and momentum streaking in subcycle THz-ARPES

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C. Lange<sup>2</sup>, R. Huber<sup>2</sup>, and U. Höfer<sup>1</sup>**

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Recently, we have demonstrated a novel concept of subcycle THz-ARPES, which permits a direct access to carrier transport in non-trivial band structures with high time resolution [1]. In this experiment, we have employed time- and Angle-Resolved Photoelectron Spectroscopy (ARPES) to observe how the carrier wave of a THz pulse accelerates Dirac fermions in the topological surface state of  $\text{Bi}_2\text{Te}_3$ . In addition to this acceleration within the sample, the THz electric field leads also to an energy and momentum streaking of the photo-emitted electrons. For UV photoemission pulses that are much shorter than the period of the THz driving field, this streaking can be used for an in-situ sampling of the electric field at the sample surface, but also needs to be taken into account for a correct interpretation of the photoelectron spectra.

In this contribution, we will discuss how the acceleration within the sample and the streaking in front of the surface both affect energy and momentum of the detected photoelectrons in a different manner, and how these contributions can be disentangled. We will show that for samples with high reflectivity in the THz regime, the electric field of p-polarized THz pulses predominantly leads to energy streaking while the strongly suppressed streaking by s-polarized pulses is dominated by small momentum variations. We will further discuss the influence of electron scattering and the energy-momentum dispersion in the sample on the detected time-integrated energy and momentum distribution of the photoelectrons.

- [1] J. Reimann, , S. Schlauderer, C. P. Schmid, F. Langer, S. Baierl, K. A. Kokh, O. E. Tereshchenko, A. Kimura, C. Lange, J. Gütde, U. Höfer, and R. Huber, “Subcycle observation of lightwave-driven Dirac currents in a topological surface band”, *Nature* 562, 396 (2018).

# Anomalous photoluminescence enhancement in antimony telluride-molybdenum disulfide ( $\text{Sb}_2\text{Te}_3/\text{MoS}_2$ ) heterostructure

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Antimony telluride ( $\text{Sb}_2\text{Te}_3$ ) is known as a typical topological insulator (TI) with energy gap in bulk and gapless topological states at surface. Besides the novel TI-related properties,  $\text{Sb}_2\text{Te}_3$  also has van der Waals (vdW) layered structures, and its surface states can be relatively robust at interfaces between  $\text{Sb}_2\text{Te}_3$  and other semiconductors. Molybdenum disulfide ( $\text{MoS}_2$ ), as a most widely studied vdW layered TMDs material in recent years, is an excellent candidate to combine with  $\text{Sb}_2\text{Te}_3$  to explore interfacial interaction between 2D material and TI and its potential applications for optoelectronic devices.

In this study, we report the observation of an anomalous photoluminescence (PL) enhancement in CVD-grown heterostructures composed of multilayer  $\text{Sb}_2\text{Te}_3$  and monolayer  $\text{MoS}_2$ . Interestingly, the PL intensity of  $\text{MoS}_2$  excitons in the heterostructure area is enhanced (by 30-40%) compared with that of the bare monolayer  $\text{MoS}_2$  under the excitation of 532 nm laser source. To identify the origin of this PL enhancement, PL spectra and transient differential reflection are measured under different temperatures. It is found that the enhanced  $\text{MoS}_2$  excitonic radiation in the heterostructures is contributed by the hot photocarriers generated within the  $\text{Sb}_2\text{Te}_3$  by optical excitation, which then transfer to the  $\text{MoS}_2$  layer facilitated by their excess energy. Our findings provide important complementary insights into the charge transfer processes in vdW heterostructures, especially under the situation of non-resonant excitation.

# Laser Induced Phase Transition of MoS<sub>2</sub>

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Molybdenum disulfide (MoS<sub>2</sub>) is a typical transition metal disulfide (TMD) as a two-dimensional material. It has potential applications in many fields such as electronic transistors, optoelectronic devices and catalysis since its special band structure. There are many different phases of MoS<sub>2</sub>, in which the 2H phase is the semiconductor and the 1T phase is the metal, due to different stacked patterns of S-Mo-S layers. These phases could switch each other under special conditions.

Here we use the Helium lamp and laser angle-resolved photoemission spectroscopy to study the band structure of bulk MoS<sub>2</sub>, we found that it only shows the characteristics of 2H phase with Helium lamp. However, after visible light illumination, both the band information of 2H phase and the image potential state of the 1T metal phase can be observed, and the lifetime of the metal phase is longer than the repetition rate of the laser pulses. We have also found that the electrons in valence band of 2H phase can go through interband transition into the image potential state of 1T metal phase. These results show that the visible light can induce a local phase transition of 2H to 1T in the bulk MoS<sub>2</sub>, and this phase transition is accompanied by charge transfer between the semiconductor phase and the metal phase of MoS<sub>2</sub>. Our results may provide insightful understanding for the applications of MoS<sub>2</sub> in future optoelectronic devices.

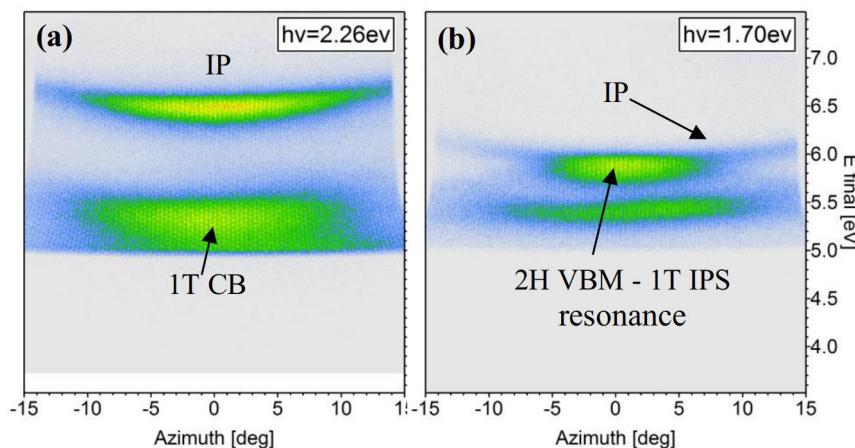


Fig. 1 Laser angle-resolved photoemission spectroscopy. (a)The image potential state of the 1T metal phase and conduction band of 1T phase. (b) Resonance of 2H VBM and 1T IPS.

# Non-Adiabatic Molecular Dynamics of Spin-Resolved Exciton

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Quantum dynamics simulation is crucial but still a challenge especially in condensed matter physics and other many-body systems. From the single particle (electron or hole) to multi-particle (exciton, trion, etc.) dynamics, the calculation will show more accurate results and reveal more essential physics. Here we introduce a method to calculate the dynamics of a spin-resolved exciton, in which the spin of bound electron and hole could be involved. In our approach, the Bethe-Salpeter equation (BSE) describes the binding of an exciton<sup>[1]</sup> and the spin-orbit coupling gives the channel for the spin flip. On the other hand, based on J.C. Tully's fewest switches surface hopping<sup>[2]</sup> (FSSH) and O.V. Prezhdo's classical path approximation<sup>[3]</sup> (CPA), an algorithm is developed to describe a (quasi-)particle's movement. By considering the excited state dynamics, the electron-phonon interaction, the excitonic effect and the spin degree of freedom, our approach gives us a powerful instrument to study plenty of time-dependent problems, and it could provide us with a more comprehensive physics picture.

[1] Rohlfing, M.; Louie, S. G., *Physical Review B* **2000**, 62 (8), 4927-4944.

[2] Tully, J. C., *J. Chem. Phys.* **1990**, 93 (2), 1061-1071.

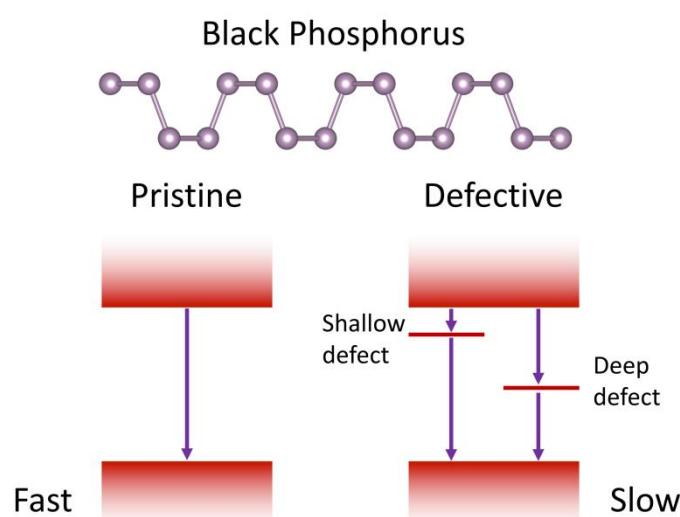
[3] Akimov, A. V.; Prezhdo, O. V., *J Chem Theory Comput* **2013**, 9 (11), 4959-72.

# Suppression of Electron-Hole Recombination by Intrinsic Defects in 2D Mono-Element Material

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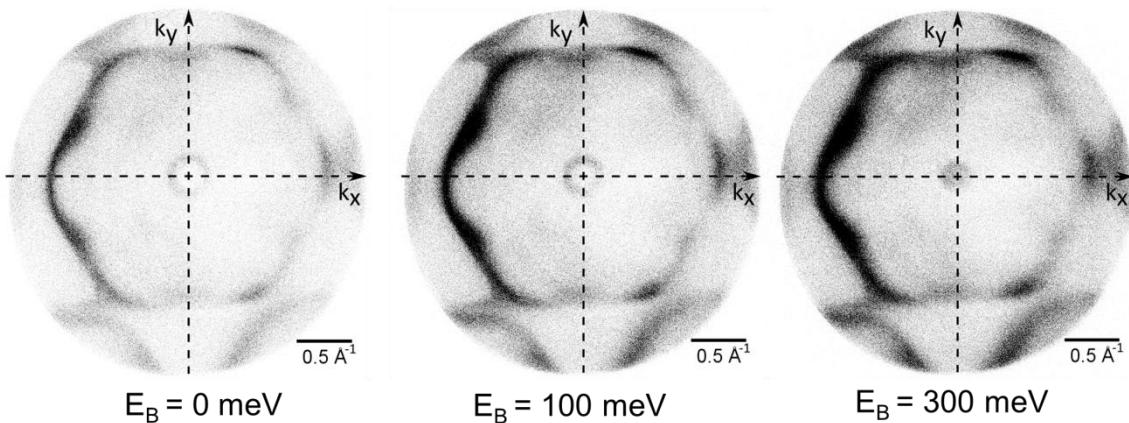
Nonradiative electron-hole ( $e-h$ ) recombination through the intrinsic defects in semiconductor has been known as one of the elemental problems in the solar energy conversion. The simple Shockley-Read-Hall (SRH) model, in which the deep trap defect states in the band gap are proposed as nonradiative  $e-h$  recombination centers, has been widely used for decades. In this report, by using time-dependent DFT based *ab initio* nonadiabatic molecular dynamics (NAMD) method, we find that the SRH model fails to describe the  $e-h$  recombination behavior for defects in two-dimensional (2D) mono-elemental material such as monolayer black phosphorous (BP). Through the investigation of three intrinsic defects with shallow and deep defect states in monolayer BP, it is found surprisingly that none of these defects accelerate the  $e-h$  recombination significantly despite of their distinct electronic structures. The deep trap defect state can even suppress the  $e-h$  recombination. Further analysis shows that because monolayer BP is a mono-elemental material, the distinct impurity phonon, which often induces fast  $e-h$  recombination, is not formed. Moreover, due to the flexibility of 2D material, the defects scatter the phonons present in pristine BP, generating multiple modes with lower frequencies. Through electron-phonon coupling with these low-frequency phonons, the nonadiabatic coupling and the coherence time between the electronic states are reduced, which suppresses the  $e-h$  recombination. We propose that the conclusion can be extended to other mono-elemental 2D materials, which can be an important guidance for the future design of functional semiconductors. In addition, the NAMD method, in which the electron-phonon coupling is included at *ab initio* level, paves a new way to understand the nonradiative  $e-h$  recombination in semiconductors.



# Time-resolved momentum microscopy with a 1 MHz high-harmonic generation lightsource

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Recent developments in laser technology and electron spectrometers allow for the build-up of a new generation of time-resolved photoelectron emission experiments. With regard to lightsource development, coherent extreme-ultraviolet femtosecond pulses can today routinely be produced via high-harmonic generation (HHG) at repetition rates up to 1 MHz, which is essential in time-resolved photoemission spectroscopy/ microscopy to avoid space charge effects. In addition, new and highly-efficient electron spectrometers are available that map the full parallel momentum space of the electronic structure (ARPES), or can be used as photoemission electron microscopes (PEEM) with several tenths of nanometers spatial resolution. On our poster, we will present first time-resolved measurements with such a high-repetition rate HHG photoemission setup. We will discuss detection efficiency, space charge effects and energy-, time-, momentum- and spatial resolution that can be achieved.



# Structural relaxation along with S<sub>1</sub>-T<sub>1</sub> conversion in carbazol-benzonitrile derivatives

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Thermally activated delayed fluorescence (TADF) molecules are gathering attention due to their potential to boost the efficiency of organic light emitting devices without precious metals.<sup>1</sup> The key is to optimize the balance between efficient emission and reverse intersystem crossing (RISC) which converts dark triplet excited states to bright singlet excited states, but microscopic understanding of the process is limited. Here, we focus on four carbazol-benzonitrile derivatives that possess identical energy gap between S<sub>1</sub> and T<sub>1</sub> but show distinct TADF activities.<sup>2</sup> We systematically studied the excited-state structures of typical TADF molecules using time-resolved infrared vibrational (TR-IR) spectroscopy in conjunction with quantum chemical calculations.

Fig. 1 shows TR-IR spectra in the region of 1250-1450 cm<sup>-1</sup>. TADF active molecules such as 4CzBN remains similar TR-IR spectra along with intersystem crossing (Fig.1a), implying the similarity in the molecular geometry between S<sub>1</sub> and T<sub>1</sub>. In contrast, TADF inactive molecules such as p-3CzBN show significant changes in TR-IR spectra along intersystem crossing (Fig.1b), suggesting drastic change in the geometry. This implies that restricting structural relaxation is important to keep the activation energy barrier for reverse intersystem crossing minimum to design efficient TADF molecules. The structural dynamics in solid environments will also be discussed.

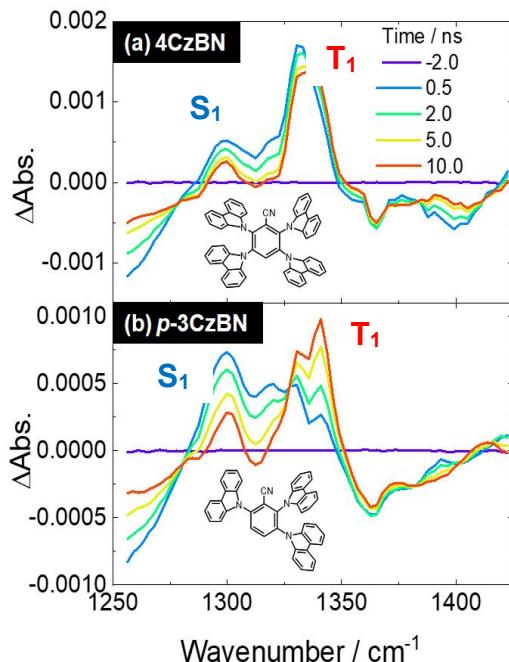


Fig. 1. Temporal evolutions of TR-IR spectra of (a) 4CzBN and (b) p-3CzBN in THF solution after photoexcitation. (inset) Molecular structures of each molecule.

[1] Uoyama, Adachi, et al., *Nature*, **492**, 234-238 (2012). [2] Hosokai, Adachi, et al., *Sci. Adv.*, **3**, e1603282 (2017).

# Spin Excitation and Magnetic Anisotropy of Mn Clusters on Graphene Moiré Patterns

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The magnetic structures comprising few-atom clusters could be exploited as bits in ultrahigh density data storage or spin-valves in nanoelectronics devices. Robust control and manipulation of the magnetization dynamics of these nanoclusters is a vital issue because their magnetic properties are highly susceptible to the environment. Here, we employ low temperature scanning tunneling microscopy and spectroscopy (STM/STS) to study the spin excitation of Mn clusters formed on single-layer graphene grown on Ru(0001) surface. The spatially resolved STS reveals spin states are highly confined within the nanoclusters. However, the site-dependent lineshapes and excitation energies indicate spin anisotropic distributions that might be modulated by the underlying graphene moiré patterns. Using noncontact atomic force microscopy (nc-AFM) and kelvin probe force microscopy (KPFM), we determine the configuration of Mn clusters and their adsorbed sites, as well as charge transfer from underlying graphene. Furthermore, we find that the spin properties of these Mn clusters can be markedly influenced by bonding with molecular ligands.

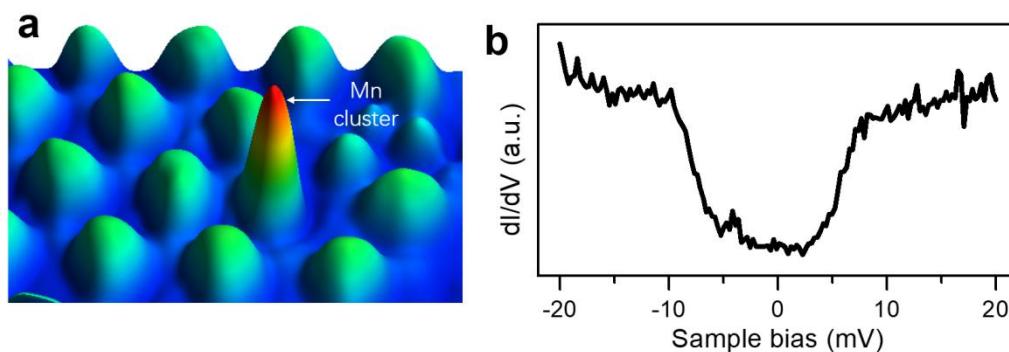


Fig. 1 a, STM image of a Mn cluster formed on graphene moiré patterns/Ru(0001), and a spin excitation spectroscopy acquired over the Mn cluster is shown in b.

# Cooperative evolution of intraband and interband excitations for high harmonic generation in strained MoS<sub>2</sub>

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Modulating electronic structure of two-dimensional (2D) materials represents an exciting avenue for tailoring their optoelectronic properties. Here, we identify a strain-induced, cooperative effect of intraband and interband excitations contributing to high harmonic generation (HHG) in prototype dichalcogenide MoS<sub>2</sub> monolayer. We find that besides the dominant intraband contributions, interband current is also indispensable in modulating HHG. The HHG yields increase linearly with the compressive strain since flatter band dispersion and Berry curvature enhance both interband and intraband dynamics. Band structure can be retrieved with high reliability by monitoring the strain-induced evolution of HHG spectra, suggesting that strain not only provides an additional knob to control HHG in solids, but also marks a way towards a complete understanding of underlying microscopic mechanisms.

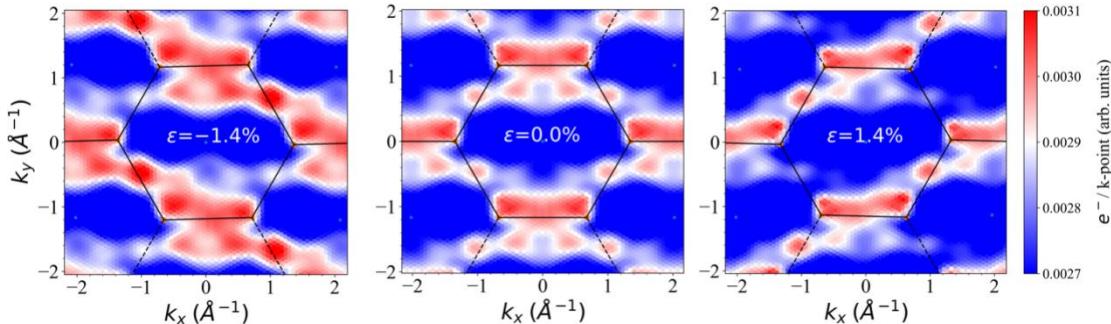


FIG. False color representation of the momentum space resolved distribution of the excited electrons under different strain conditions.

# Visualization of surface plasmon polaritons propagating at dielectric/metal interfaces by 2P-PEEM

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Visualization of surface plasmon polaritons (SPPs) propagating at dielectric/metal interfaces is indispensable in opening up opportunities for a new photonics utilizing the plasmonic phenomena. Two-photon photoelectron emission microscopy (2P-PEEM) is one powerful method to directly observe the SPP propagation induced by a pump photon. With this model, a beating of surface polarization between SPPs and pump photon itself is probed as a lateral distribution of photoemission (Figure 1) [1].

Here, we demonstrate the visualization of SPPs propagating at the dielectric/metal interfaces of fullerene ( $C_{60}$ )/Au(111) and alkanethiolate self-assembled monolayers (SAMs)/Au(111), through a dual-color 2P-PEEM using near-infrared (NIR) pump and ultraviolet (UV) probe photons.

In both systems, SPPs propagating in sub-mm scale are clearly imaged. Furthermore, the beating patterns quantitatively reflect the physical properties of SPPs at the interfaces (e.g. SPP dispersion, phase and group velocities). In the case of  $C_{60}$ /Au(111), the SPP properties are modulated sensitively depending on the  $C_{60}$  coverage, where the analytical results nicely fit to a simple Drude model. On the other hand, the SPP dispersion for SAM/Au(111) obviously deviates from the model assuming simply *n*-alkane layers on Au(111). The results imply that a strong chemical interaction at the gold-sulfur interface largely affects the SPP properties [2]. Precise evaluation of SPP properties for nanoscale functional films will be of high importance for a fine control of future plasmonic nanodevices.

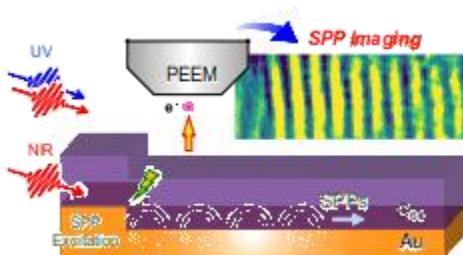


Figure 1. Schematic of SPP visualization at  $C_{60}$ /Au(111) interface by dual-color 2P-PEEM.

[1] M. Shibuta, T. Eguchi, and A. Nakajima, *Plasmonics* **8**, 1411 (2013).

[2] K. Yamagiwa, M. Shibuta, and A. Nakajima, *Phys. Chem. Chem. Phys.* **19**, 13455 (2017).

# Quantum Dynamics of Photo-induced Water Splitting on Graphitic Carbon Nitride

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Hydrogen production from photocatalytic water splitting provides an ideal way to harvest solar energy. One of the most promising photocatalyst is graphitic carbon nitride ( $\text{g-C}_3\text{N}_4$ ) thanks to its outstanding thermal and chemical stability, low cost and nontoxic nature. Despite many experimental studies, the microscopic quantum motion of photocatalysis remains elusive. Here we use newly-developed time-dependent density functional theory (TDDFT) molecular dynamics to explore the nonadiabatic process of photo-induced water splitting on graphitic carbon nitride. Different from traditional scheme, hole transfer plays a key role in water photosplitting, as described by a three-step model. First, the  $\text{g-C}_3\text{N}_4$  is photoexcited with electron transfer from nitrogen to carbon atoms. Then, water is attracted to nitrogen atom with a hole transfer from  $\text{g-C}_3\text{N}_4$  to water molecule, leading to a weaker O-H bond. Last, the O-H bond is activated and breaks with a proton transferred from water to  $\text{g-C}_3\text{N}_4$ . This work presents a first real time quantum simulation of photo-induced water splitting by first principles, which helps designing more effective photocatalysts.

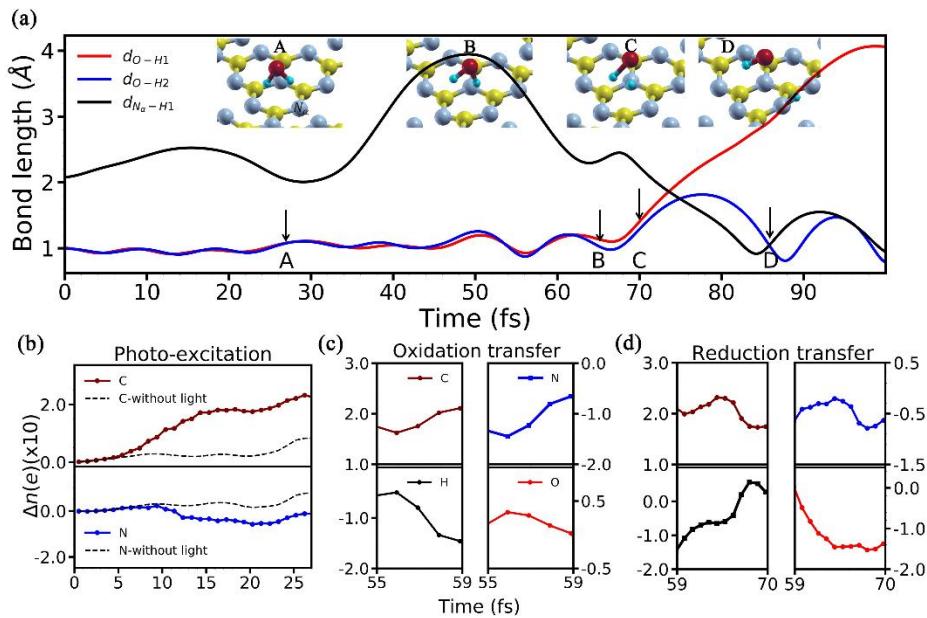


Figure 1. The dynamics of the photocatalytic water splitting process on  $\text{g-C}_3\text{N}_4$  upon photoexcitation

# Laser induced demagnetization in two dimension material $\text{NiCl}_2$

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The overarching goal of the field of femtomagnetism is to control the magnetic structure of matter on a femtosecond time scale. The temporal limits to the light-magnetism interaction are governed by the fact that the electron spin interacts indirectly with light, with current studies showing a laser induced global loss in the magnetic moment on a time scale of the order of a few 100 s of femtoseconds. In this work, we report *ab initio* simulations of the quantum dynamics of electronic charge and spins when subjected to intense laser pulses. Performing these purely electron-dynamics calculations for two dimension  $\text{NiCl}_2$  causes a significant change in the moments on the order of 30 fs. We show that the underlying physics of demagnetization for  $\text{NiCl}_2$  is dominated by a combination of the flow of spin-currents and spin-flips.

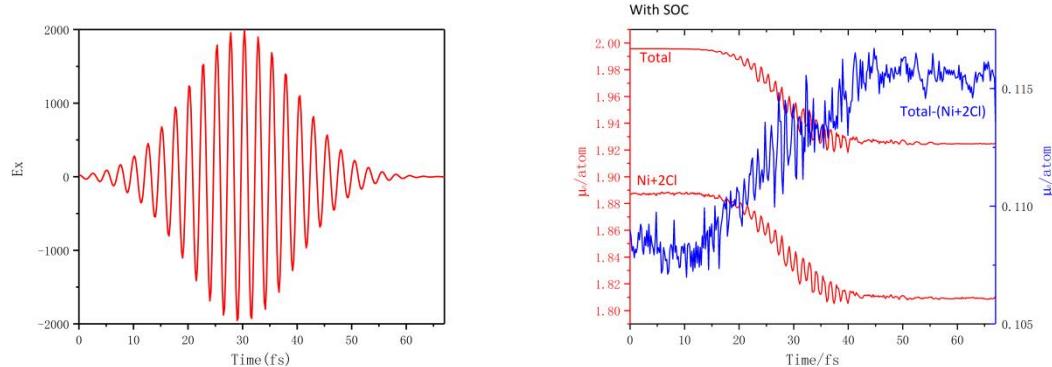


Fig. 1 Left: The shape of laser field. Right: Change of magnetic moment as a function of time.

- (1) Huang, B. et al., Nature 546, 270–273 (2017)
- (2) H. Liu et al., J. Phys. Chem. Lett. 2018, 9, 6709–6715
- (3) P. Tengdin et al., Sci. Adv. 2018;4: eaap9744
- (4) W. You et al., Phys. Rev. Lett. 121, 077204 (2018)

# Tracking energy transfer dynamics in luminescent Eu<sup>3+</sup> complex

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Trivalent europium (Eu<sup>3+</sup>) complexes are attractive materials for luminescence applications because of their narrow line-width emission due to the transition between shielded 4f-4f transitions. To overcome a small extinction coefficient ( $\varepsilon < 10 \text{ M}^{-1}\text{cm}^{-1}$ ) of Eu<sup>3+</sup> itself, the luminescent Eu<sup>3+</sup> complex needs to contain organic ligands with a large extinction coefficient. However, design strategy for efficient energy transfer from the ligands to the metal center is not yet established. Here, we have studied energy transfer dynamics in luminescent Eu(hfa)<sub>3</sub>(DPPTO)<sub>2</sub> (Fig. 1a) using time-resolved photoluminescence as a function of sample temperature.

The complex was pumped at the  $\pi-\pi^*$  transition of the ligands (267 nm). Time-resolved emission was recorded using either a streak camera or photomultiplier tube coupled with a monochromator. The solid sample was supported on a He cryostat to tune its temperature in the range of 20–300 K.

Fig. 1b shows the emission spectra at different delay time at room temperature. We observed three distinct emissions from Eu(hfa)<sub>3</sub>(DPPTO)<sub>2</sub>: (1) Broad emission in 300–450 nm which decays in a few ns (fluorescence from triphenylene ligands), (2) Narrow emission at 586 nm decaying in 1.5 s (4f-4f transition from the <sup>5</sup>D<sub>1</sub> state of Eu<sup>3+</sup>), and (3) Narrow emission bands at 595 and 620 nm that shows rise in 1.5 s and decays in >100 s (4f-4f transition from the <sup>5</sup>D<sub>0</sub> state). The rise and decay time scales correspond to the energy cascade from initially excited ligands to Eu<sup>3+</sup> metal center excited states (Fig. 1c). Interestingly, the emission dynamics from the <sup>5</sup>D<sub>1</sub> state showed additional emission rising in 0.5 s at 20 K (Fig. 1d), which might indicate additional energy transfer channel activated at low temperature due to suppression of non-radiative decay.

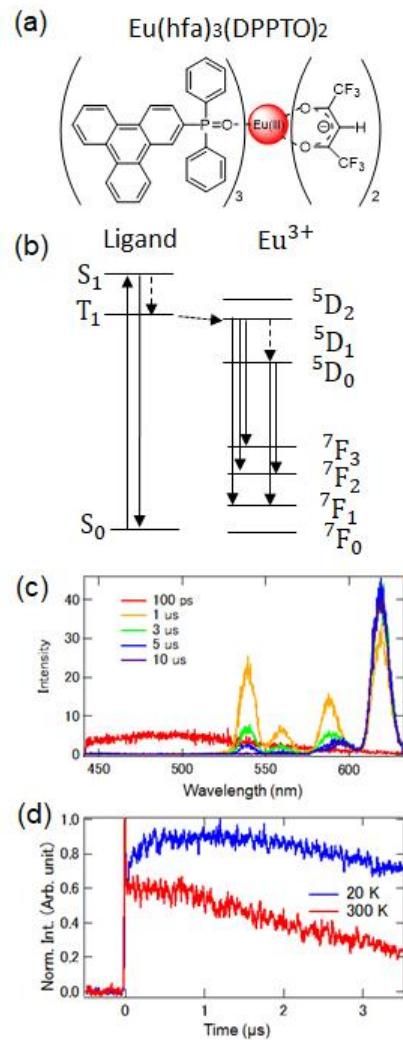


Fig.1. (a) Structure of Eu(hfa)<sub>3</sub>(DPPTO)<sub>2</sub>. (b) Emission spectra at different delay times. (c) The energy diagram of the Eu(hfa)<sub>3</sub>(DPPTO)<sub>2</sub>. (d) Temporal profiles of emission from <sup>5</sup>D<sub>1</sub> state (580–590 nm) at 300 K (red) and at 20 K (blue).

# Enhanced two photon photoemission at the interface of metal phthalocyanines and HOPG

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Metal phthalocyanines(MPcs) are widely used in organic semiconductor industry because of their outstanding photoelectric properties and thermal stability. We investigate the interactions of MPcs and HOPG surfaces using photoemission spectroscopy (PES) and relaxation process of the excited states with two-photon photoemission spectroscopy (2PPE). We observed greatly enhanced photoemission process at the interfaces of MPcs/inorganic substrates(HOPG, BP) compared to the pristine substrates and this enhancement is ascribed to the selection rules as well as the increased density of states introduced by MPcs which can be excited by the ~400nm light. The lifetime of the excited states are measured to be around 60 fs with time-resolved 2PPE. Our results provide valuable reference to the photoelectric and photovoltaic research fields.

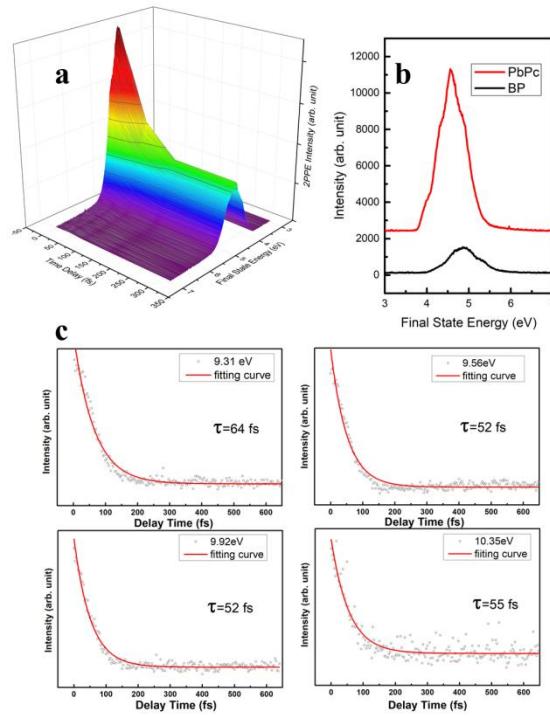


Fig. 1 (a) 2PPE spectra for 1 ML PbPc on BP as a function of delay time. (b) The enhanced 2PPE of PbPc/BP compared to the pristine BP. (c) The two-photon correlation signals for different energy levels.

# Temperature Dependent Ultrafast Dynamics of Heterogeneous O<sub>adsorbed</sub> + O<sub>adsorbed</sub> = O<sub>2,gas</sub> Reaction on Single Crystal Pd(100) Surface

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The interaction of atomic oxygen with transition-metal is of pivotal importance for a variety of industrially and environmentally relevant processes, including CO oxidation in automobile exhaust catalytic converters. As the recombination of two atoms forming a molecule, which then leaves the surface, represents one of the simplest surface chemical reactions, the recombinative desorption of molecular Oxygen from Pd single crystal surface attracted early attention of femtosecond surface chemistry community. Misewich and coworkers<sup>1</sup> followed the desorption of oxygen molecules from a molecular oxygen-covered Pd(111) surface. This molecular oxygen-covered Pd(111) surface can be prepared only at temperatures lower than 200 K (for an example, Misewich's work was performed at 140 K). At the low temperature (below 200 K), molecular oxygen can be molecularly adsorbed on the palladium surface. At temperatures above 300 K, molecular oxygen completely dissociates to oxygen atoms on the palladium surfaces, rendering atomic oxygen adlayer surface. So far, no recombinative desorption of (O+O=O<sub>2</sub>) was observed from the experiments performed with adsorbed atomic oxygen on the Pd(111) surface and it was concluded by Heinz and coworkers that femtosecond laser excitation in the range of 2 to 5 mJ/cm<sup>2</sup> does not initiate recombinative desorption of oxygen.<sup>2</sup> On the contrary, our work<sup>3</sup> is the first report on femtosecond laser-induced recombinative surface chemistry of adsorbed oxygen atoms on the Pd single crystal surface. The process of recombinative desorption of molecular oxygen (O<sub>adsorbed</sub> + O<sub>adsorbed</sub> = O<sub>2,gas</sub>) from Pd(100) single crystal surface, under femtosecond laser irradiation, has been investigated with the help of pre- and post-radiation temperature programmed desorption (TPD) measurements. This surface photochemistry is found to depend strongly on initial surface temperature. 400 K temperature was found to be threshold temperature above which this reaction is active for absorbed fluence of 2.86 mJ/cm<sup>2</sup>. The desorption-yield is found to be linear with respect to the absorbed fluence. Furthermore, the dynamics for the recombinative O+O=O<sub>2</sub> desorption has been explored with the help of two pulse correlation spectroscopy(2PC). The dynamics is found to be strongly dependent on the initial temperature, which leads to form different adsorbate coverage and surface structures.

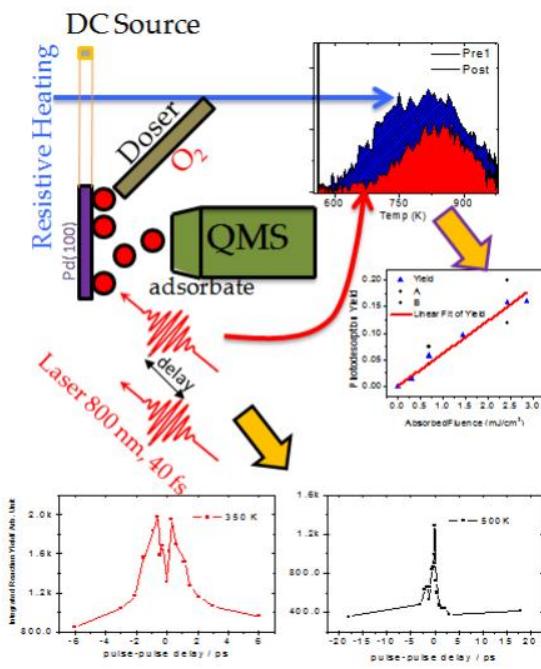


Figure 1: Summary for femtosecond laser-induced recombinative  $O+O=O_2$  reaction on Pd(100) and temperature dependent dynamics

1. Misewich, A. et al *J. Chern. Phys.*, **1994**, *100*, 736.
2. Heinz, T.F. et al *Proc. SPIE*, **1994**, *276*, 2125.
3. Banerjee, S. et al *J. Phys. Chem. C*, **2018**, *122*, 26039.

# Ultrafast carrier redistribution and photovoltage generation at a metal-semiconductor interface

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A photo-induced potential shift occurring at a surface and interface, namely photovoltage, is an essential phenomenon for optoelectronic applications and enhanced reactivity in photocatalysts. The dynamical behavior was intensively studied using time-resolved photoelectron spectroscopy (TrPES) at synchrotron facilities in ns  $\sim$   $\mu$ s time scales [1]. Thanks to recent developments of TrPES techniques [2], the ultrafast (fs  $\sim$  ps) processes have also been reported [3], however, whose underlying mechanism is not yet fully understood. We recently discovered an ultrafast shift of PES spectra measured on a metal-semiconductor interface of a Bi thin film on a Ge substrate. As shown in Fig. 1(a), the temporal evolution already started before time zero, which is a clear signature of a dynamical change in the net potential. We also performed numerical simulations of redistribution of hot carriers across the space charge region. The results demonstrate that photovoltage can generate instantly after photoexcitation and can decay in such an ultrafast time scale. The present study takes first step in understanding the ultrafast photovoltage and towards an attracting dynamics study utilizing unique properties of ultrathin Bi films [4].

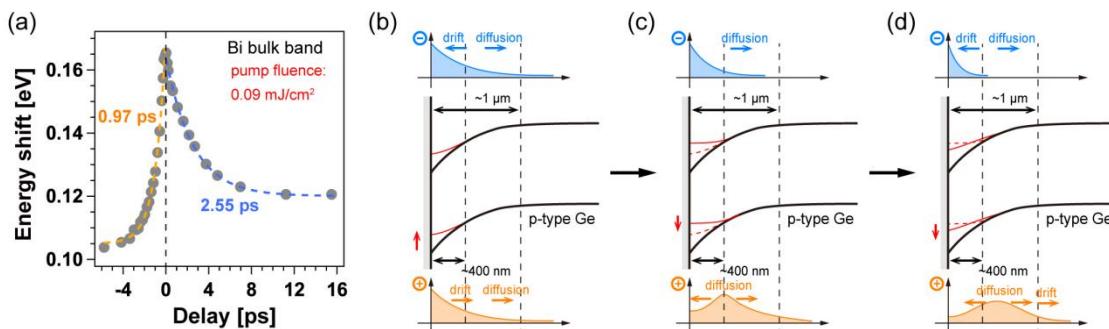


Fig. 1 (a) Temporal evolution of peak positions in TrPES spectra of a 40 bilayer Bi film grown on a p-type Ge(111) substrate. The pump and probe energies are 1.5 eV and 5.9 eV, respectively. (b)-(d) Schematics of ultrafast carrier redistribution and concomitant modulation of the interface band bending.

- [1] S. Yamamoto and I. Matsuda, J. Phys. Soc. Jpn. **82**, 021003 (2013).
- [2] Y. Ishida *et al.*, Rev. Sci. Instrum. **85**, 123904 (2014).
- [3] Y. Ishida *et al.*, Scientific Reports **5**, 8160 (2015), M. Neupane *et al.*, Phys. Rev. Lett. **115**, 116801 (2015), Y. Hazama *et al.*, Phys. Rev. Applied **10**, 034056 (2018).
- [4] S. Ito *et al.*, Phys. Rev. Lett. **117**, 236402 (2016), Phys. Rev. B **97**, 155423 (2018), to be submitted.

# Momentum dependent quasi-particle lifetime in metals investigated by time-resolved momentum microscopy

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Electron dynamics in solid state systems and at interfaces play a crucial role for the performance of nanoscale electronic and spintronic devices. Therefore, it is essential to investigate the electron dynamics and the corresponding energy and momentum dissipation mechanisms in prototypical materials used for such devices.

So far, most investigations focused on the inelastic lifetimes of hot electrons, which can be directly obtained with time-resolved two-photon photoemission (tr-2PPE), using a pump-probe setup. Here we will go beyond this conventional approach and investigate the momentum dependent lifetime of electrons and the corresponding (quasi-)elastic scattering phenomena. This is achieved by combining the well-established tr-2PPE technique with momentum spectroscopy in a ToF-PEEM operated in k-space mode. This enables us to analyze auto- and cross-correlation traces for different intermediate state energies at each point of the accessible momentum space, which can be directly translated into momentum dependent lifetime map representations (see Fig1b).

As first model systems, we focused on bulk states of simple noble metal surfaces such as Ag(110) and Ag(111) using different angles of incidence of laser light. Here we already observe lifetime differences of several femtoseconds, which can be correlated to the occupied as well as the unoccupied part of the band structure. Our first results already point to a complex momentum dependent lifetime of electrons even for simple material systems.

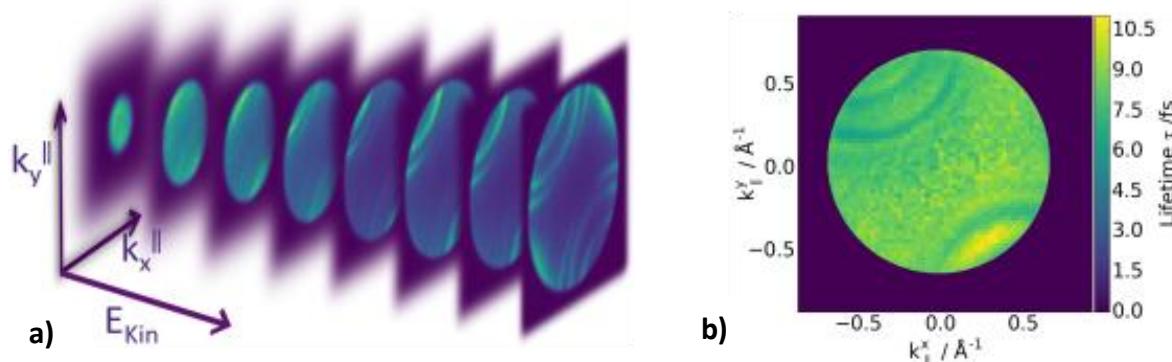


Figure 1. a) Time of flight data obtained in our momentum microscope from illuminating a Ag(110) crystal under normal incidence with a photon energy of 3.1eV. b) By introducing a delay between two identical pulses, we perform cross-correlation measurements and can directly translate the photoemission yield into electron lifetimes.

# New long-lived metastable state mediated by mode-selective electron-phonon coupling in 1T-TaSe<sub>2</sub>

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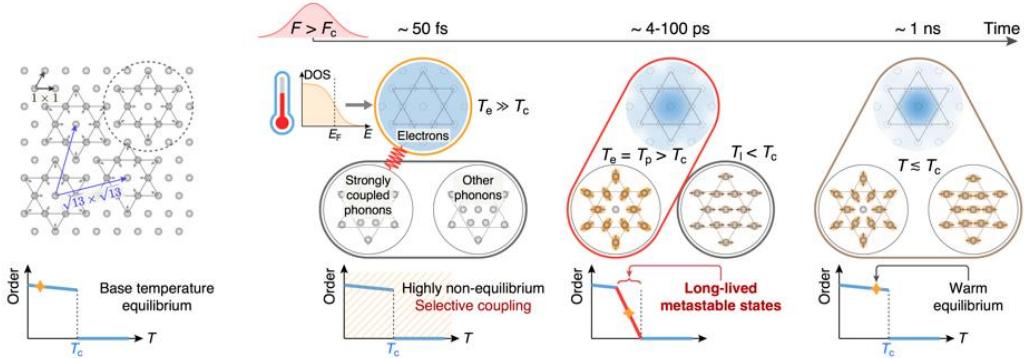
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Quantum materials represent one of the most promising frontiers in the quest for faster, lightweight, energy efficient technologies. However, their inherent complexity and rich phase landscape make them challenging to understand or manipulate in useful ways. Ultrafast excitation with a strong laser pulse enables the controllability over strongly correlated electronic states through non-equilibrium pathway. Here we use time- and angle-resolved photoemission spectroscopy, combined with femtosecond lasers, to measure the dynamic electron temperature, band structure and heat capacity of strongly correlated materials [1]. We then show that this is a very sensitive probe of phase changes in materials, because electrons react very quickly, and moreover generally are the smallest component of the total heat capacity. This allows us to uncover a new long-lived metastable state in the charge density wave material 1T-TaSe<sub>2</sub>, that is distinct from all of the known equilibrium phases (Fig. 1): it is characterized by a significantly reduced effective heat capacity that is only 30% of the normal value, due to selective electron-phonon coupling to a subset of phonon modes. As a result, significantly less energy is required to melt the charge order and transform the state of the material than under thermal equilibrium conditions. This metastable CDW phase, with a continuously tunable order parameter depending on the laser fluence, is stable up to hundreds of picoseconds, thus markedly enriching the landscape of metastable ordered phases of TaSe<sub>2</sub>. Moreover, this charge ordered metastable phase can be verified by measuring the band structure and breathing mode to reveal its nature and microscopic interactions [2]. The creation and optical manipulation of the long lived metastable phase with ultrashort lasers pave the way for future studies to inform new theories, as well as to map the full functional phase space of quantum materials for application.



**Fig. 1 New long-lived metastable state mediated by mode-selective electron-phonon coupling.** The upper left panel shows the top view of the Ta plane in 1T-TaSe<sub>2</sub>. In the CDW state, displacement of the Ta atoms leads to a  $\sqrt{13} \times \sqrt{13}$  superstructure consisting of 13-atom star-of-David clusters. After laser excitation, the evolution of the sample is determined first by the electron temperature and then by electron-phonon coupling, which depend on the fluence. For strong laser excitation, the electron-phonon coupling switches from nearly homogeneous to mode-selective. The resulting inhomogeneity within the phonon bath drives the material into a new long-lived metastable CDW state. The blue shading represents the electron density in the real-space, the grey circles represent Ta atoms, both amplitudes are exaggerated for better visualization. T<sub>e</sub>, T<sub>p</sub> and T<sub>i</sub> refer to the temperatures of the electron, strongly-coupled phonons, and the rest of the phonon bath, respectively

[1] X. Shi, W. You, Y. Zhang, Z. Tao, P. M. Oppeneer, X. Wu, R. Thomale, K. Rossnagel, M. Bauer, H. Kapteyn, M. Murnane, *Science Advances* **5**, eaav4449 (2019).

[2] In preparation.

# Electron-phonon interactions in transient states after ultrafast excitation

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Electron-phonon interactions are ubiquitous in condensed matter physics and play an important role in superconductivity, charge density waves in transition metal diselenides, relaxation of hot electrons, the carrier mobility in semiconductors. We investigate the linewidth and electron phonon coupling in several 2D materials under different Fermi-Dirac distribution with density functional perturbation theory and Wannier interpolation method. Our results provide information about electron-phonon coupling of the relaxation process of transient states after ultrafast excitation.

# Oscillating kinetic energy of the photoemitted electrons in tr-PES

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Cephise Cacho<sup>3</sup>, Richard Chapman<sup>3</sup>, Emma Springate<sup>3</sup>, Serguei Molodtsov<sup>2</sup>, and Martin  
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We measured ultrafast time-resolved photoemission spectra on Gd (0001) and W (110) with an IR pump, XUV probe set up at the Artemis facility at CLF. We observed an oscillating kinetic energy of the photoemitted electrons before time zero. Such oscillations have been observed on the surface state of Gd by Bovensiepen *et al.* [1]. They described the oscillations with a ponderomotive acceleration of the emitted photoelectrons in a transient grating formed by the interference between the incident and reflected parts of the pump pulse. We performed a further investigation on these oscillations by varying the wavelengths of the pump pulse and found that the amplitude and the phase of the oscillations are pump wavelength-dependent. They show different behavior than predicted by the ponderomotive acceleration model. By comparing the amplitude of the oscillations on different bands, we discovered that the oscillating kinetic energy depends on the initial state of photoemission.

[1] Bovensiepen *et al.*, Phys. Rev. B, 79, 045415 (2009)

# Photogenerated Carrier Dynamics at Anatase/Rutile TiO<sub>2</sub> Interface

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TiO<sub>2</sub> is an intensively studied photocatalytic material owing to its low cost and high activity. The anatase/rutile (A/R) mixed-phase TiO<sub>2</sub> is recognized as an effective strategy to achieve high photocatalytic efficiency by the type-II band alignment favorable to spatial charge separation. However, the atomic structure, as well as the exact band alignment of the A/R mixed-phase TiO<sub>2</sub>, is very difficult to identify either in experimental measurements or theoretical simulations. Moreover, the time-dependent photogenerated carrier dynamics, which can determine the photocatalytic efficiency, has not been studied at the atomic scale. In this report, we use adaptive genetic algorithm to search the stable interface structures. We find that the band alignment is determined by the interfacial atomic structures. Especially, with oxygen vacancy (O<sub>v</sub>) at the interface, band alignment can be reversed as compared to that of the stoichiometric interface. Then, we select one stoichiometric and one defective structure to study the photogenerated carrier dynamics using the time-dependent *ab initio* nonadiabatic molecule dynamics. We find that in the stoichiometric system, for both the electron and the hole, the charge transfer happens within 400 fs, which is much shorter than the electron-hole recombination timescale at ns to  $\mu$ s magnitude, which suggests that the charge transfer can occur efficiently at the interface before they recombine. For the defective A/R system with O<sub>v</sub>, we find that the electron will be trapped by the defect state within 1 ps, while the hole dynamics is not affected. Our study provides atomic insights into the understanding of the band alignment and photogenerated carrier dynamics at the mixed A/R TiO<sub>2</sub> interface, which provides valuable guidance for functional material design for solar energy conversion.

# Electrically driven single-photon superradiance from molecular chains in a plasmonic nanocavity

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When N two-level emitters undergo dipole-dipole coupling, new collective states are formed as a result of quantum superposition of product states with well-defined phases. Such coherent intermolecular dipole-dipole interactions can lead to various phenomena such as coherent energy transfer, photosynthesis, and new quantum engineered systems [1,2]. One of the interesting features of quantum coherent collective states is single-photon superradiance (SPS), namely the generation of a singly excited collective state through symmetric quantum superposition of all possible states [3,4]. However, quantitative studies of SPS in molecular systems confront challenges concerning the precise control and characterization of intermolecular dipole-dipole coupling, including the number of coupled monomers (N), intermolecular distances, dipole arrangements and orientations as well as their phase relations. Furthermore, it would be highly desirable to have optical access to each individual constituent of a molecular system such as a chain.

By exploiting scanning tunneling microscope induced luminescence technique [5], here we demonstrate electrically driven SPS from artificially constructed molecular chains of up to 12 molecules. Sub-nanometer resolved spectroscopic imaging for molecular chains reveals the local optical response of superradiant states and the associated quantum coherence nature. Second-order photon correlation measurements for these superradiant peaks show evident photon antibunching effects with single-photon purities  $g^{(2)}(0) < 0.5$ , indicating the occurrence of SPS and the formation of single-exciton superradiant state. The combination of spatially resolved spectral measurements with theoretical considerations reveals that nanocavity plasmons dramatically modify the linewidth and intensity of emission from the molecular chains, but they do not dictate the intrinsic coherence of the superradiant states. Our results open up new routes to study energy transfer and quantum many-body physics at individual molecule level.

- [1] Scholes, G. D. et al., *Nature*, **2017**, 543, 647
- [2] Hestand, N. J. and Spano, F. C., *Chem. Rev.*, **2018**, 118, 7069.
- [3] Dicke, R. H. , *Phys. Rev.*, **1954**, 93, 99.
- [4] Scully, M. O. and Svidzinsky, A. A. *Science*, **2009**, 325, 1510.
- [5] Kuhnke, K., Große, C., Merino, P. and Kern, K., *Chem. Rev.*, **2017**, 117, 5174

# Ultrafast photoinduced phase transition of MoTe<sub>2</sub>

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Among transition metal dichalcogenides (TMDs), MoTe<sub>2</sub> is of particular interest in the field of phase engineering, due to the small free energy difference between its semiconducting 2H phase and metallic 1T' phase, as well as the novel quantum phenomena such as topological states and Weyl states discovered in its 1T' and T<sub>d</sub> phases. In this study, we perform time-resolved reflection spectroscopy with varying pump fluences on a CVD-grown MoTe<sub>2</sub> film, composed of single crystalline grains of 2H phase and polycrystalline 1T' phase.[1] Under excitation fluences above  $\sim 10 \text{ mJ/cm}^2$ , the 2H-MoTe<sub>2</sub> grains demonstrate transient features very distinct from those under the lower excitation but similar to the photoresponse of the metallic 1T'-MoTe<sub>2</sub> area, suggesting photoinduced (partial) phase transformation. Temperature-dependence of transition characteristics in the transient differential reflection signals are also examined to obtain a comprehensive understanding on this optically induced phase transition. Our findings open up new opportunities for ultrafast phase control of the novel TMDs materials by optical methods, and also provide enlightening insights into the physics underlying their polymorphism.

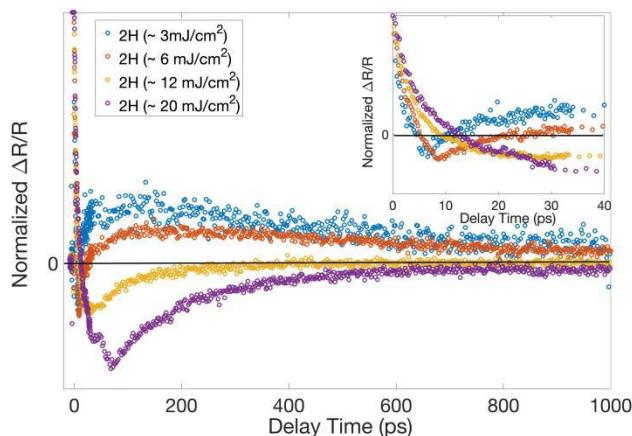


Fig. 1 Transient differential reflection of 2H-MoTe<sub>2</sub> measured under different pump fluences. The pump and probe wavelengths are 800 nm and 1050 nm, respectively.

[1] X. Xu, S. Chen, et al., *J. Am. Chem. Soc.* **141**, 2128 (2019).

# State resolved transient charge transfer on h-TiO<sub>2</sub>(110) by fs time-resolved photoemission

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Time-resolved pump-probe photoemission spectroscopy has been used to study the dynamics of charge carrier recombination and trapping on hydroxylated rutile TiO<sub>2</sub>(110). An XUV (20 - 30 eV) probe was used to monitor the population of electrons and holes in the conduction and valence bands. Two types of pump excitation were employed, one in the infrared (0.95 eV) and the other in the UV (3.5 eV) region. With IR excitation, electrons associated with defects are excited into the bottom of the conduction band from the polaronic states within the band gap, which are retrapped within 45±10 fs. Under UV excitation, the electrons in these band gap states as well as valence band electrons are excited into the conduction band. In addition to the fast polaron trapping observed with IR excitation, we also observe a long lifetime (about 1 ps) component to both the depletion of hot electrons at the bottom of the conduction band and the refilling of the band gap states. This points to a band gap state mediated recombination process with a ps lifetime.

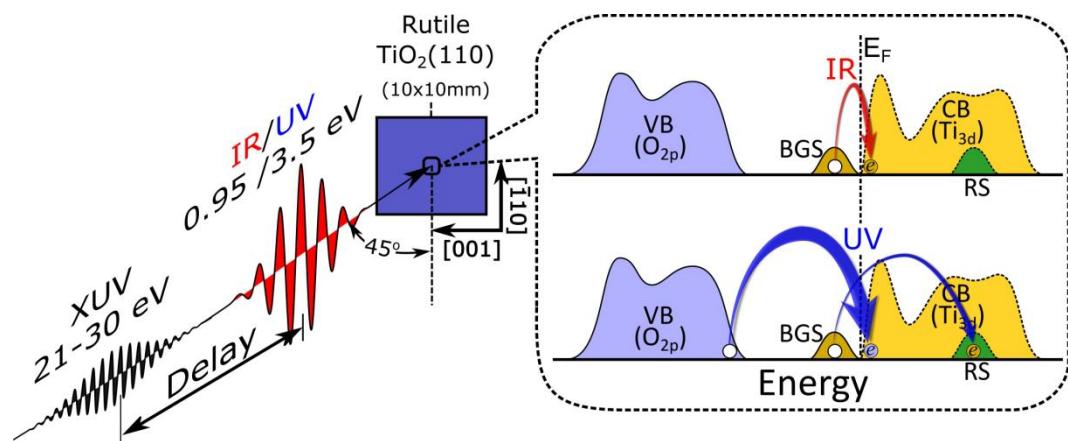


Fig. 1 Time-resolved pump-probe photoemission spectroscopy has been used to study the dynamics of charge carrier recombination and trapping on hydroxylated rutile TiO<sub>2</sub>(110). Two types of pump excitation were employed, one in the infrared (0.95 eV) and the other in the UV (3.5 eV) region

# Ultrafast Photocarrier Dynamics of Atomically Thin Bismuth Oxselenide Crystals

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Identifying novel two-dimensional (2D) materials with high mobility and suitable bandgap is a key issue of modern electronics. Besides the recently investigated 2D materials such as graphene, transition metal dichalcogenides (TMDs) and black phosphorus, the air-stable bismuth oxselenide crystals ( $\text{Bi}_2\text{O}_2\text{Se}$ ) has also attracted tremendous attention as a newly emerging candidate for optoelectronic devices, due to its high mobility and large electronic bandgap. However, the fundamental photocarrier behaviors of  $\text{Bi}_2\text{O}_2\text{Se}$ , determining the performance of related optoelectronic applications, still remain elusive. In this study, we research the ultrafast photocarrier dynamics in CVD-grown single crystalline  $\text{Bi}_2\text{O}_2\text{Se}$  nanoflakes through two-color pump-probe technique. Our experimental results reveal a  $\sim$ ns photocarrier lifetime in  $\text{Bi}_2\text{O}_2\text{Se}$  samples, much longer than those of other 2D materials. Photocarrier dynamics under high photocarrier density is dominated by bimolecular recombination, and a bimolecular recombination constant of  $3.3 \times 10^{-6} \text{ cm}^2 \text{s}^{-1}$  is extracted. Such low value of the bimolecular recombination constant is much smaller than those of bulk  $\text{Bi}_2\text{O}_2\text{Se}$  and TMDs, and also indicates that the 2D  $\text{Bi}_2\text{O}_2\text{Se}$  should be a promising material for optoelectronic devices. Meanwhile the photocarrier diffusion has been also measured, where the diffusion coefficient is  $\sim 31 \text{ cm}^2/\text{s}$ . Our findings could promote the understanding of photo-physics in  $\text{Bi}_2\text{O}_2\text{Se}$ , and provide valuable guidelines for device design based on  $\text{Bi}_2\text{O}_2\text{Se}$ .

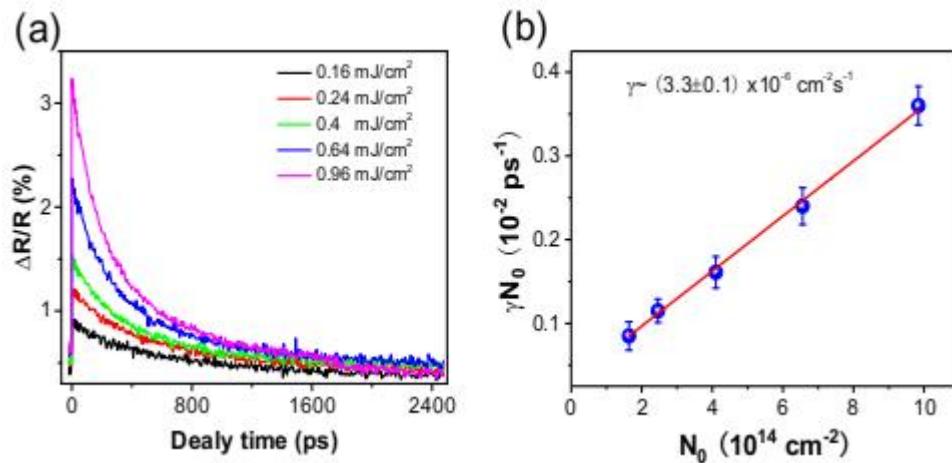


Fig. 1 (a) Dynamical signals of  $\text{Bi}_2\text{O}_2\text{Se}$  under different pump fluences; (b) the fitting of bimolecular recombination constant  $\gamma$  and the blue dots are extracted from (a).

# Spin Dynamics of Ultrafast Demagnetization in Ni

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We employ time-dependent *ab initio* non-adiabatic molecular dynamics (NAMD) to study the ultrafast demagnetization in Ni. Under the interaction of spin-orbital coupling, the spin flip of laser-excited electrons occurs in a time scale of hundreds of seconds. However, the spin minority electrons trend to conserve their spin because nonadiabatic coupling is stronger than spin-orbital coupling. This is a key factor of the demagnetization in Ni.

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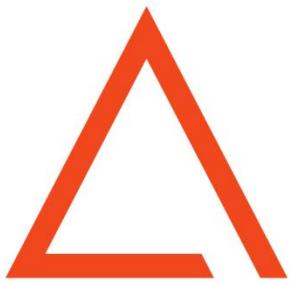
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