USD 10

Ultrafast Surface Dynamics

June 11-16, 2017
Inzell, Germany

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WELCOME

to the 10th International Symposium on *Ultrafast Surface Dynamics*. This meeting takes place for the tenth time at a remote location in order to facilitate discussion and informal interaction among all participants. We wish you a pleasant stay at the foothills of the Alps in Inzell and hope that you enjoy the scientific program of the USD 10. We are looking forward to stimulating talks and discussions.

Uwe Bovensiepen, Nora Dörmann, and Michael Bauer

HISTORY

Ultrafast Surface Dynamics (USD) is a biannual international conference and aims at informal, scientific exchange in the field of ultrafast dynamics at surfaces and interfaces. Previous meetings took place

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

SCOPE

The conference focuses on questions about fundamental electronic, atomic and spin-dependent processes at interfaces, which evolve on time scales from a few picoseconds to attoseconds. The program of USD 10 will cover, e.g., experiments on dynamics of electron transfer processes on the atomic scale, the generation and detection of transient photocurrents at topological surface states, and attosecond relaxation processes during the photoemission of electrons. A further focus will be the current development in theoretical physics regarding questions of 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 functional theory. The main objectives of the event are the exchange of such current scientific results, the initiation and deepening of bilateral and multilateral cooperation among the research groups, as well as the discussion and development of general visions on future research in or around the particular research area.

USD ANNIVERSARY

This year's edition of the international symposium on *Ultrafast Surface Dynamics* in 2017 represents two anniversaries. It is the tenth meeting which takes place exactly twenty years after the first USD symposium. We commemorate this anniversary in a special session which will highlight major achievements in the past and future opportunities and challenges.

PROGRAM

At the meeting, the recent progress in the field of *Ultrafast Surface Dynamics* will be presented. We have grouped the presentations into the following topics which are represented by the following distinguished international invited speakers.

- Electron Dynamics and Electronic Energy Transfer at Surfaces and Interfaces
  A. Nilsson, K. Watanabe, J. Zhao, M. Shibuta
- Surface and Interface Magnetisation Dynamics
  S. Sharma, M. Cinchetti, A. Eschenlohr
- Ultrafast Plasmonics and Nanooptics
  C. Ropers, M. Raschke, P. Hommelhoff
- Dynamics in Low-dimensional Materials
  I. Gierz, Ph. Hofmann, R. Ernstorfer, E. Malic, R. Huber
- Novel Phenomena / Materials
  M. Horn-von Hoegen, B. Moritz, K. Kuroda
- Attosecond Surface Science
  L. Castiglioni, M. Murnane, E. Krasovskii

FINANCIAL SUPPORT

We gratefully acknowledge the financial support provided by the *Deutsche Forschungsgemeinschaft* as the main funding source of the meeting. We also thank the *Wilhelm und Else Heraeus-Stiftung* for travel grants for students who are members of the *German Physical Society*.

We thank the collaborative research centers *SFB 1083* (Marburg) and *SFB/TRR 173* (Kaiserslautern / Mainz) for their financial support for the USD anniversary session.

Poster sessions and the conference outing are cosponsored by the companies *Amplitude Systems, Clark-MXR, Coherent, KMLabs, Laser Quantum, Light Conversion, NKT Photonics, ScientaOmicron, SPECS, Spectra-Physics*, *Thorlabs, TOPAG*, which is highly appreciated.

INTERNATIONAL STEERING COMMITTEE

Uwe Bovensiepen
Pedro M. Echenique
Tony F. Heinz
Ulrich Höfer
Yoshiyasu Matsumoto
Toshiaki Munakata
Hrvoje Petek
Martin Weinelt
Martin Wolf
Xiaoyang Zhu

Sunday, 11 June 2017

Arrival

18:00 Welcome & Introduction

Chair: Ulrich Höfer

18:20 **Anders Nilsson**
Probing Surface Catalysis in Real Time

19:00 **Margaret Murnane**
Capturing the Fastest Charge and Spin Dynamics in Nanosystems using High Harmonic Beams

19:40 Reception

20:00 Dinner

Monday, 12 June 2017

7:45 **Breakfast**

Chair: Margaret Murnane

9:00 **Masahiro Shibuta**
Two-Photon Photoemission Spectroscopy and Microscopy of Photoexcited Electron in Organic Molecular Films

9:40 **Petra Tegeder**
Ultrafast Exciton Dynamics in Organic Semiconductors

10:00 **Benjamin Stadtmüller**
Optically induced Quasi-Particle Dynamics and Transient Polarization of a C_{60} *Film*

10:20 **Coffee break**

11:00 **Jin Zhao**
Phonon Assisted Ultrafast Charge Transfer at van der Waals Heterostructure Interface

11:40 **Kazuya Watanabe**
Non-Equilibrium Adsorbate Dynamics Induced by Electronic Friction

12:30 **Lunch**

14:00 **Outdoor Workshop**

Chair: Martin Wolf

15:40 **Sangeeta Sharma**
Laser Induced Ultrafast Spin Dynamics:
A Time-Dependent Density Functional Theory Perspective

16:20 **Stefan Mathias**
Spin-Resolved Band-Structure Evolution during Ultrafast Demagnetization in Cobalt

16:40 **Beatrice Andres**
Don’t Judge a Lanthanide by its Surface State?
Magnetization Dynamics on Gd and Tb(0001)

17:00 **Coffee break**

17:40 **Mirko Cinchetti**
Dynamic Spin Filtering at Hybrid Ferromagnetic Metal-organic Interfaces

18:20 **Andrea Eschenlohr**
Ultrafast Inhomogeneous Magnetization Dynamics Analyzed
by Linear and Interface-sensitive Nonlinear Magneto-Optics

19:00 **Dinner**

20:20 **Poster Session** (posters with odd numbers)

Tuesday, 13 June 2017

7:45 Breakfast

Chair: Hrvoje Petek

9:00 Claus Ropers
Ultrafast Electron Diffraction and Microscopy Enabled by Nanoscale Photoemitters

9:40 Markus Raschke
Ultrafast Nanoscopy: Imaging Structure, Function, and
Dynamics of Matter on its Natural Length and Times Scales

10:20 Coffee break

11:00 Peter Hommelhoff
Quantum Path Interference in Two-Color Coherently Controlled Electron Emission off Tungsten Tips and in Attosecond Electron Dynamics in Graphene

11:40 Michael Horn-von Hoegen
Ultrafast non-Equilibrium Structural Dynamics in the Si(111)-In (8x2)<->(4x1) System at the Quantum Limit

12:30 Lunch

14:00 Outdoor Workshop

Chair: Toshiaki Munakata

15:40 Brian Moritz
Optical Materials Design of TMDCs and Frustrated Mott Insulators

16:20 Frank Oliver Schumann
Correlated Electron Dynamics at Surfaces Investigated via He Ion Neutralization

16:40 Alexander Paarmann
Mid-Infrared Nonlinear Nanophotonics Using Surface Phonon Polaritons

17:00 Coffee break

17:40 Kenta Kuroda
Optical Control over Spin-Polarized Surface States in Topological Insulators

18:20 Kazuki Sumida
Temperature-induced Prolonged Duration of non-Equilibrium Surface Dirac Fermions in Topological Insulators

18:40 Yoshiyasu Matsumoto
Enhancement of Hole Trapping at Curved Surface of Titania Nanoparticles with Water Adsorbate

19:00 Dinner

20:20 Poster Session (posters with even numbers)

Wednesday, 14 June 2017

7:45 Breakfast

Chair: Petra Tegeder

9:00 Philip Hofmann
Electron Dynamics in Single-Layer Transition Metal Dichalcogenides

9:40 Ralph Ernstorfer
Momentum-Resolved View on Electrons, Phonons and Their Coupling in WSe_2

10:20 Coffee break

11:00 Robert Wallauer
Intervalley Scattering Dynamics in MoS_2 *Imaged by 2PPE with High-harmonic Probe*

11:20 Ermin Malic
Excitonic Fingerprint of Atomically Thin 2D

12:00 Kunie Ishioka
Ultrashort Acoustic Pulses Generated at Buried GaP/Si Interface

12:20 Sebastian Otto
Dynamics of p-doped Topological Insulators $SnBi_xSb_{2-x}Te_4$ *and* $GeSb_2Te_4$

12:45 Lunch

14:00 Conference Outing

19:00 Conference Dinner

Thursday, 15 June 2017

7:45	**Breakfast**
	Chair: Jin Zhao
9:00	**Isabella Gierz** *Femtosecond Control of Quasiparticle Interactions in Graphene*
9:40	**Ryuichi Arafune** *Spin-Texture Inversion in Image Potential States of Ir(111) covered by Graphene*
10:00	**Sven Aeschlimann** *Ultrafast Momentum Imaging of Chiral Interband Excitations in Graphen*
10:20	**Coffee break**
11:00	**Guido Fratesi** *Substrate Induced Ultrafast Electron Injection Dynamics at Organic-Graphene Interfaces*
11:20	**Yoav William Windsor** *Ultrafast Electron Dynamics of the Surface States of Intermetallic* $GdRh_2Si_2$ *Probed by XUV trARPES*
11:40	**Rupert Huber** *Lightwave Electronics in Layered Semiconductors and single Molecules*
12:30	**Lunch**
	USD 10 Anniversary Session *Past and Future of USD 10*
15:00	**Chair: Thomas Fauster** *Introduction*
15:20	**Helmut Zacharias** *Femtosecond Laser Induced Associative Desorption of Molecular Hydrogen from Graphite*
16:00	**Hrvoje Petek** *Ultrafast Coherent Multidimensional Multiphoton Photoemission Spectroscopy*
16:40	**Coffee break**
17:20	**Tony F. Heinz** *Ultrafast Surface Dynamics - without the Bulk: Probing 2D Materials*
18:00	**Martin Aeschlimann, Ulrich Höfer**
19:00	**Dinner**
20:20	**Martin Wolf** *The Forefront of Time-resolved Photoemission: From the Past to the Future*

Friday, 16 June 2017

7:45 **Breakfast**

Chair: Michael Bauer

9:00 **Manuel Ligges**
Time-domain Identification of Electronic Correlations in 1T-TaS_2

9:20 **Selene Mor**
Ultrafast Electronic Band Gap Control in an Excitonic Insulator

9:40 **Eugene Krasovskii**
Kinematic and Dynamic Properties of a Bloch-Wave Packet Excited by an Ultrashort Light Pulse

10:20 **Coffee break**

11:00 **Walter Pfeiffer**
Intra-atomic Delays in Attosecond Time-resolved Solid State Photoemission

11:20 **Johann Riemensberger**
Energy Dependent Photoemission Delays from the (0001) Surface of Magnesium

11:40 **Luca Castiglioni**
Attosecond Dynamics in Photoemission from Noble Metal Surfaces

12:20 **Closing**

12:45 **Lunch**

14:00 **Outdoor Workshop**

ORAL CONTRIBUTIONS

Sunday, 11 June 2017

Overview Talks

18:20 **Anders Nilsson (invited)**
Probing Surface Catalysis in Real Time

19:00 **Margaret Murnane (invited)**
Capturing the Fastest Charge and Spin Dynamics in Nanosystems using High Harmonic Beams

Probing Surface Catalysis in Real Time

A. Nilsson

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In heterogeneous catalysis, reactants adsorbed on surfaces are converted to products, which eventually desorb via various intermediates. The transition state separates reactants and intermediates from products and the free energy required to reach it determines the kinetics of an elementary chemical reaction. Many surface reaction intermediates are, however, transient species with a short residence time and the population of species in the transition state region is near-zero making their observation a challenge during steady state conditions. Ultrafast pump-probe techniques have, however, opened up opportunities by promoting a sufficient population of molecules in transient states to allow detection on short time scales. Here recent results on probing chemical reactions on surfaces using X-ray free-electron lasers LCLS (Linac Coherent Light Source) at SLAC National Accelerator Laboratory) will be presented. Four examples will be shown CO desorption, Oxygen activation, CO oxidation and CO hydrogenation on Ru(0001). We demonstrate that both transient intermediates and the transition state region can be detected in surface chemical reactions. I will describe my vision of the future development of this field where the various FEL facilities can play an important role.

Capturing the Fastest Charge and Spin Dynamics in Nanosystems using High Harmonic Beams

M. Murnane

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High harmonic generation (HHG) is a unique quantum light source with fundamentally new capabilities – producing fully spatially and temporally coherent beams with linear or circular polarization throughout the extreme ultraviolet (EUV) and soft X-ray region, all on a tabletop. This talk will review recent developments in HHG sources, as well as exciting advances in imaging and spectroscopy of materials. [1-5] It is now possible to image below the wavelength limit in the EUV/x-ray region for the first time, by combining HHG sources with coherent diffractive imaging (CDI) techniques. In other recent work, we measured the shortest lifetime of any state to date, at 212±30 attoseconds, corresponding to an excited state in the band structure of a material. More recently, in particular, a new technique called attosecond-ARPES (angle resolved photoemission) harnesses HHG pulse trains to measure the fastest electron dynamics intrinsic to materials, making it possible to distinguish sub-femtosecond electron scattering and screening for the first time. Finally, bright, phase matched, circularly and linearly polarized HHG now span from the EUV to the soft X-ray region at kHz repetition rates, ideally suited for a host of applications in science and technology.

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[3] K.M. Hoogeboom-Pot, J.N. Hernandez-Charpak, T. Frazer, E.H. Anderson, W. Chao, R. Falcone, X. Gu, R. Yang, M. M. Murnane, H.C. Kapteyn, D. Nardi, PNAS **112**, 4846 (2015).

[4] E. Turgut, C La-o-Vorakiat, J. Shaw, H. Nembach, D. Rudolf, R. Adam, S. Mathias, M. Aeschlimann, C. Schneider, T. Silva, H. Kapteyn, M. Murnane, Phys. Rev. Lett. **110**, 197201 (2013).

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Monday, 12 June 2017

Electron and Energy Transfer Dynamics

9:00 **Masahiro Shibuta (invited)**
Two-Photon Photoemission Spectroscopy and Microscopy of Photoexcited Electron in Organic Molecular Films

9:40 **Petra Tegeder**
Ultrafast Exciton Dynamics in Organic Semiconductors

10:00 **Benjamin Stadtmüller**
Optically induced Quasi-Particle Dynamics and Transient Polarization of a C_{60} *Film*

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Phonon Assisted Ultrafast Charge Transfer at van der Waals Heterostructure Interface

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Magnetization Dynamics

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Magnetization Dynamics on Gd and Tb(0001)

17:40 **Mirko Cinchetti (invited)**
Dynamic spin filtering at hybrid ferromagnetic metal-organic interfaces

18:20 **Andrea Eschenlohr (invited)**
Ultrafast Inhomogeneous Magnetization Dynamics Analyzed
by Linear and Interface-sensitive Nonlinear Magneto-Optics

Two-Photon Photoemission Spectroscopy and Microscopy of Photoexcited Electron in Organic Molecular Films

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Ultrafast carrier dynamics in organic molecular films are required to be clarified spectroscopically and microscopically to develop well-designed organic devices. Here, we have investigated the time evolution of photoexcited electrons for organic thin films by time-resolved two-photon photoemission spectroscopy (2P-PES) and microscopy (2P-PEEM).

As for example, Fig. 1(a) shows the 2P-PES spectra for 1 monolayer (ML) C_{60} film on HOPG graphite taken with varying the pump-probe delay. The peaks at E_F + 0.7 eV and 1.8 eV originate from the photocarrier electrons in the lowest unoccupied molecular orbital (LUMO) and the next LUMO (LUMO+1) of C_{60}, respectively, in which the photocarriers are generated by a dissociation of hot-exciton with pump photon.[1] The photocarrier electrons can be imaged with 2P-PEEM in real space. Figs. 1(b) and (c) compare the one-photon (1P-) and 2P-PEEM images of 0.2 ML C_{60} film on HOPG. The contrasts in both images are totally reversed, and the bright area in the 2P-PEEM indeed corresponds to the C_{60} islands.[2] In other words, photocarrier electrons in the C_{60} nanostructured islands are successfully imaged.

We will also introduce the 2P-PES study on alkanethiolate self-assembled monolayers (SAMs) fabricated on Au(111). Long-lived image potential electrons ranging up to 100 ps depending on the thickness of alkyl layer have been resolved, indicating well-insulating property with the molecular monolayer.[3] Furthermore, a deposition and chemical substitution of functional molecules onto the SAM have revealed the characteristic excited states and dynamics of the molecules electronically decoupled surface system.[4] These spectroscopic and microscopic observations are essential to understand the charrier generation and transport in organic thin film devices.

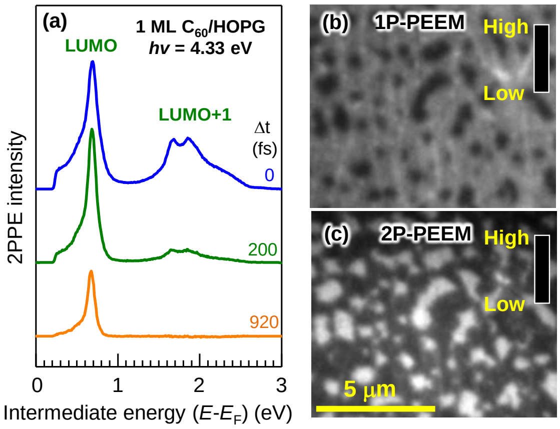

Fig. 1. (a) 2P-PES, (b) 1P-PEEM (hv = 5.63 eV), and (c) 2P-PEEM (hv = 4.23 eV) for C_{60} film on HOPG.

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[2] M. Shibuta et al., Appl. Phys. Lett. **109**, 203111 (2016).
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[4] M. Shibuta et al., J. Am. Chem. Soc. **136**, 1825 (2014).

Ultrafast Exciton Dynamics in Organic Semiconductors

M. Hänsel[1], L. Bogner[1], V. Belova[2], K. Broch[2], F. Schreiber[2], P. Tegeder[1]

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Understanding the dynamics of electronically excited molecular states after optical excitation in organic semiconductors is necessary for improvement and optimization of potential molecule-based devices such as organic photovoltaic cells or organic light emitting diodes. Using femtosecond time-resolved spectroscopies, namely two-photon photoemission (2PPE) and second harmonic generation (SHG), we studied the exciton dynamics at interfaces with thiophene and perylene derivatives, respectively.

For instance, SHG is applied to investigate the photoinduced ultrafast charge transfer (CT) dynamics across the donor/acceptor, P3HT/C_{60} interface. The de-excitation of hot singlet excitons in the conduction bands of the polymer into localized excitonic states is observed. In the presence of the electron acceptor, the ultrafast population of a CT state is identified as the dominating relaxation channel. Interestingly, the charge transfer yield correlates with the excitation wavelength and rises with increasing excess energy. In well-ordered diindenoperylene and perylene alkyldiimide films, respectively, we elucidated the exciton dynamics in the femtosecond regime using SHG.

In the case of dicyanovinyl (DCV)-substituted sexithiophene (DCV6T), the dynamics of excitonic states involving processes ranging from femtoseconds to several tens of picosseconds are resolved using 2PPE. The decay of the excitonic states is considerably slower than in DCV-dimethyl-pentathiophene (DCV5T-Me_2). We ascribe this behavior to weaker intermolecular couplings in the DCV6T film. Despite the faster exciton decay, DCV5T-Me_2 is known for a better solar cell efficiency compared to that of DCV6T. We suggest that this is due to the concomitant better exciton and charge carrier transport in well-coupled DCV5T-Me_2 molecular structure.

Optically Induced Quasi-particle Dynamics and Transient Polarization of a C_{60} Film

B. Stadtmüller[1,2], S. Emmerich[1,2], D. Jungkenn[1], N. Haag[1], C. Schott[1], S. Eich[1], M. Rollinger[1], M. Maniraj[1], M. Aeschlimann[1], M. Cinchetti[3], S. Mathias[4]

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The performance of organic based electronic devices is determined by the energy level alignment of the valence and conduction band as well as by the excitation dynamics of electrons in these materials. Although both aspects are intrinsically linked by the polarizability of organic materials, they have mainly been discussed separately so far.

In this work, we present new insight into the ultrafast band structure dynamics of thin C_{60} films on Ag(111). The combination of time- and angle resolved photoemission (tr-ARPES) with a fs-XUV light source allows us to simultaneously follow the transient evolution of the occupied as well as of the unoccupied band structure after fs-optical excitation with visible light. The unoccupied part of the C_{60} band structure reveals the well-known quasi-particle dynamics of the exciton formation and its decay in C_{60}. Most interestingly, we observe transient changes of the linewidth of all occupied molecular orbitals upon optical excitation which can be directly linked to the characteristic timescales of the exciton decay of C_{60}. These observations are attributed to a transient polarization of the molecular film caused by the exciton formation at distinct C_{60} sites.

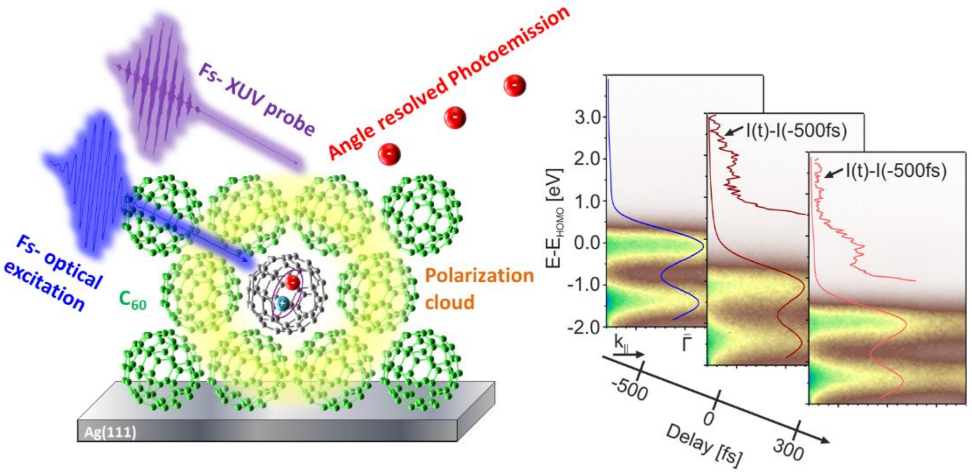

Fig. 1: The left side shows a schematic sketch of the experiment. A C60 multilayer film is optically excited by a fs-light pulse and the transient changes of the band structure are monitored by tr-ARPES. Typical tr-ARPES maps of the transient band structure of a C_{60} film are shown on the right side.

Phonon Assisted Ultrafast Charge Transfer at van der Waals Heterostructure Interface

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The ultrafast dynamics of photo-excited charge carriers plays an important in optoelectronics and solar energy conversion. Using nonadiabatic molecular dynamics simulation, we study the ultrafast charge dynamics at CH_3OH/TiO_2 and MoS_2/WS_2 interfaces. For CH_3OH, we study the forward and backward hole transfer between TiO_2 and CH_3OH as well as the whole hole relaxation process to valance band maximum (VBM). First, we found that the hole trapping ability of CH_3OH depends on the adsorption structure strongly. Second, we found the time scales of forward hole transfer process from TiO_2 to CH_3O (hole trapping process) and hole relaxation to VBM strongly depend on the temperature. The hole relaxation process to VBM is also slow down significantly. This can be interpreted by the reduction of the non-adiabatic coupling and the phonon occupation. For MoS_2/WS_2, we show that instead of direct tunneling, the ultrafast interlayer hole transfer is strongly promoted by an adiabatic mechanism through phonon excitation. At room temperature, the interlayer charge transfer in MoS_2/WS_2 is ultrafast with a timescale of 20 fs which is in good agreement with the experiment. This ultrafast hole transfer process can be suppressed by decreasing the temperature to 100K. The atomic level picture of phonon-assisted ultrafast mechanism revealed in our study is valuable both for the fundamental understanding of ultrafast charge carrier dynamics as well as for the design of novel quasi-2D devices.

Non-Equilibrium Adsorbate Dynamics Induced by Electronic Friction

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Hot electrons in metal substrate couple to vibrational degrees of freedom of adsorbates leading to non-thermal reactions. The non-adiabatic coupling emerges as frictional force on adsorbates inducing stochastic vibrational excitations [1]. The potential for steering the surface chemistry largely depends on how large degree of non-equilibrium can be attained by the ultrafast heating of adsorbates. Characterizing non-equilibrium statistics among multidimensional vibrational degrees of freedom is crucial to understand and develop hot carrier-driven chemistry. Despite the numerous efforts, the evolution of vibrational excitation in an ultrafast timescale still remains to be elucidated. Toward this goal, we have developed vibrational sum-frequency generation spectroscopy with phase-sensitive detection combined with UHV [2] and have demonstrated its potential to reveal structure [3] and dynamics of surface adsorbates [4].

In this talk, we focus on multidimensional vibrational dynamics of carbon monoxide (CO) on Cu(100) along hot-carrier induced desorption [5]. Instantaneous frequency and amplitude of the CO internal stretching mode are tracked with a sub-picosecond time resolution that is shorter than the vibrational dephasing time. These experimental results in combination with numerical analysis based on Langevin simulations enable us to extract non-equilibrium distributions of external vibrational modes of desorbing molecules. Superstatistical distributions are generated with mode-dependent frictional couplings in a few hundred femtoseconds after hot-electron excitation, and energy flow from hot-electrons and intermode anharmonic coupling play crucial roles in subsequent evolution of the non-Boltzman distributions.

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Phys. Rev. Lett. **117**, 186101 (2016).

Laser-induced Ultrafast Spin Dynamics: A Time-Dependent Density Functional Theory Perspective

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Ultrafast manipulation of spins in a controlled manner is a milestone of solid state physics. The motivation for this is to use electronic spin for storing binary data, which can then be optically manipulated using lasers. The advantage of such a technique would be reduction in the size and efficiency of the storage device by several orders of magnitude. Recent experiments have demonstrated that demagnetization or spin-reorientation processes can be induced by femtosecond laser pulses. However, we are still far from achieving optimally controlled manipulation of spins required for production of devices. One of the reasons behind this is the lack of full understanding of the phenomena leading to demagnetization.

Time-dependent density functional theory (TDDFT) is a formally exact method for describing the real-time dynamics of electrons under the influence of an external field- for example vector potential of the intense laser pulse. We use spin-resolved TDDFT to study of the process of optical demagnetization. The advantage of such a technique is clear from the fact that it is fully ab-initio in nature.

Our analysis shows that the demagnetization is a highly system dependent phenomenon; in bulk of simple metals like Fe, Co and Ni demagnetization occurs as a two-step process where first the electrons make transitions to excited states, followed by spin-orbit-mediated spin-flip transitions which lead to a loss of moment [1,2]. In the bulk of complex systems like Heuslers demagnetization can partially occur without spin-orbit interaction, just by transfer of moment between two different spin-lattices [3]. In metallic interfaces this process of demagnetization is, to a large degree, dominated by spin currents across the interface. These observations appear to preclude the development of a single simple model describing spin dynamics in materials on short time scales.

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Spin-Resolved Band-Structure Evolution During Ultrafast Demagnetization in Cobalt

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The evolution of the electronic band structure of the simple ferromagnets Fe, Co, and Ni during their well-known ferromagnetic-paramagnetic phase transition has been under debate for decades, with no clear and contradicting experimental observations so far. Here, we show that using time- and spin-resolved photoelectron spectroscopy, we can make a movie showing the evolution of the electronic properties in real time after excitation with an ultrashort laser pulse. This allows us to monitor large transient changes in the spin-resolved electronic band structure of Cobalt. We show that the loss of magnetization is not only found around the Fermi-level, where the states are affected by the laser excitation, but reaches much deeper into the electronic bands. We find that this demagnetization process cannot be explained by a loss of the exchange splitting of the spin-polarized bands, but instead shows rapid band mirroring after the excitation, which is a clear signature of ultrafast magnon generation. Our result helps to understand band-structure formation in these seemingly simple ferromagnetic systems, and gives first clear evidence of the transient processes relevant to femtosecond demagnetization. [1]

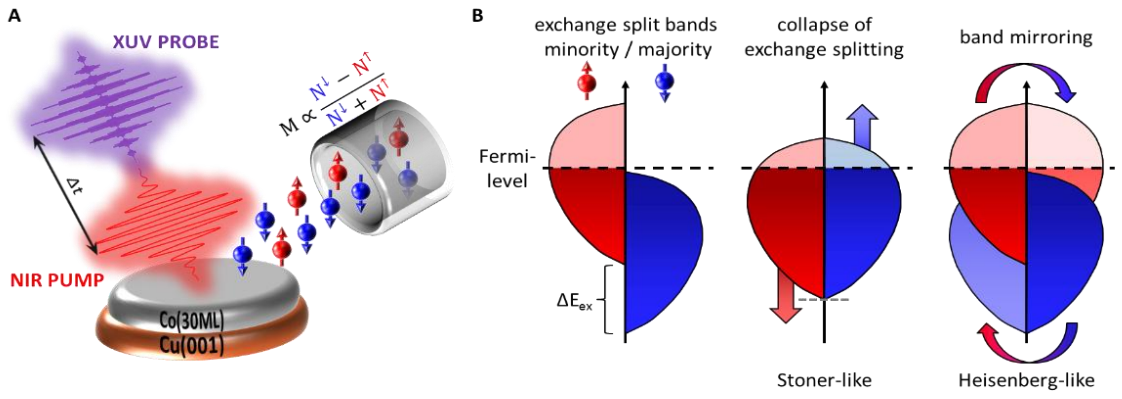

Fig. 1: Schematic of the time- and spin-resolved photoemission experiment and the potential response of the electronic and spin systems to laser-induced demagnetization. (A) The in-plane magnetized 30 ML Co film is excited with 74±1 fs, 1.6 eV near-infrared laser pulses that induce demagnetization. The evolution of the band structure is measured via spin- and time-resolved photoemission using extreme ultraviolet (XUV) 33±7 fs, 22 eV pulses from high-harmonic generation (HHG). (B) (left) Exchange-split density of states for a 3d ferromagnet. (middle) Reduced magnetization in the Stoner-like picture via a potential collapse of the exchange splitting and (right) in the localized spin picture via band mirroring.

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Don’t Judge a Lanthanide by its Surface State? Magnetization Dynamics on Gd and Tb(0001)

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Time-resolved photoemission of bulk bands is a formidable challenge on magnetic metals. If spin-resolution is added to measure the complete magnetization dynamics, it becomes currently almost impossible. Therefore, instead of measuring bulk states, we utilize the unique magnetic properties of the $5d_{z^2}$ surface states on Gd(0001) and Tb(0001), which mirror the magnetism of the bulk. This way, we can use the fundamental of a 300 kHz regenerative Ti:Sapphire-laser amplifier as pump pulse and its fourth harmonic for direct photoemission (probe), which leads to sizable spin-resolved count rates in an exchange-scattering detector.

In spin- and time-resolved photoemission experiments on the laser-induced demagnetization of Gd [1] and Tb, we find that the exchange splitting of the surface state follows that of the *(5d6s)* valence bands [2]. In contrast, the spin polarization of the surface state appears to reflect the magnetization of the *4f* core levels [3]. We can thus use the surface state to monitor the properties of the different bulk states contributing to magnetism in the lanthanides.

This way, we confirm, in a spin-resolved experiment, the enormous difference between the Gd- and Tb-demagnetization times previously found in XMCD [4]. Besides this difference observed in the surface states’ spin polarization, we still find similar response times for the exchange splitting of both materials.

We thus show that the laser-induced demagnetization of the lanthanides is driven by different processes. In Gd, the ultrafast reduction of the exchange splitting can be explained by Elliott-Yafet spin-flip scattering, while spin-orbit coupling leads to a much slower decrease of the spin polarization. In Tb both spin-flip scattering and spin-orbit coupling act on the ultrafast time scale reducing exchange splitting as well as spin polarization within the first picosecond. The significantly different response of the spin polarization in Gd (~ 15 ps) and Tb (~ 1 ps) follows the dynamics of the *4f* magnetic moments, which exhibit a strongly different spin-orbit coupling in both materials.

In summary, comparing the demagnetization times for bulk and surface states, it seems legitimate to judge a lanthanide by its surface state.

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Dynamic Spin Filtering at Hybrid Ferromagnetic Metal-Organic Interfaces

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Recent developments in molecular spintronics indicate that the deposition of organic molecules on the strongly reactive surfaces of ferromagnetic metals leads to a change in the local magnetic properties of the atoms hybridized with the molecule, such as exchange interaction, magnetic moments, and magnetocrystalline anisotropy. In organic spintronics devices, the specific interaction between the organic molecules and the inorganic magnets used as electrodes also dominates the spin injection and spin filtering across the hybrid ferromagnet-organic interface [1].

Here we demonstrate a spin-filtering mechanism based on the dynamical spin relaxation of the long-living interface states formed by the magnet and weakly physisorbed molecules. We investigate the case of Alq_3 on Co and, by combining spin- and time-resolved two-photon photoemission experiments with electronic structure theory, show that the observed long-time spin-dependent electron dynamics is driven by molecules in the second organic layer [2,3]. The interface states formed by physisorbed molecules are not spin-split, but acquire a spin-dependent lifetime, that is the result of dynamical spin-relaxation driven by the interaction with the Co substrate. Such spin-filtering mechanism has an important role in the injection of spin-polarized carriers across the interface and their successive hopping diffusion into successive molecular layers of molecular spintronics devices.

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Ultrafast Inhomogeneous Magnetization Dynamics Analyzed by Linear and Interface-sensitive Nonlinear Magneto-Optics

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The interaction of femtosecond optical pulses with magnetic materials presents new opportunities for manipulating magnetization on ultrafast timescales. In particular, laser-induced spin currents in metallic ferromagnets and heterostructures drive spin dynamics in a non-local manner, between different components of the sample structure. We analyze fs spin currents across interfaces, and competing processes such as phonon-mediated spin flips, employing epitaxial Co/Cu(001) films in a 0.4-20 nm thickness range as a well-defined model system. The bulk-sensitive magneto-optical Kerr effect (MOKE) and non-linear magneto-optics, i.e. time-resolved magnetization-induced second harmonic generation (mSHG), which is surface and interface sensitive in cubic crystals, serve as complementary time-resolved probes.

Exploiting the depth sensitivity of MOKE, we identify spin currents and spin-flip scattering via characteristic spatial profiles in the transient magnetization of Co after femtosecond laser excitation. We find that on time scales before hot electron thermalization (<100 fs) the Co demagnetization is governed by spin-dependent transport effects, while local spin-flip processes dominate subsequently [1]. The nearly destructive interference of the surface and buried interface contributions to mSHG in Co/Cu(001) makes the overall signal particularly sensitive to differences in the transient magnetization redistribution in Co. From a characteristic Co film thickness dependence of the magnetization dynamics within the first several hundred femtoseconds we conclude on a limited mean free path of the ultrafast spin current of about 3 nm [2].

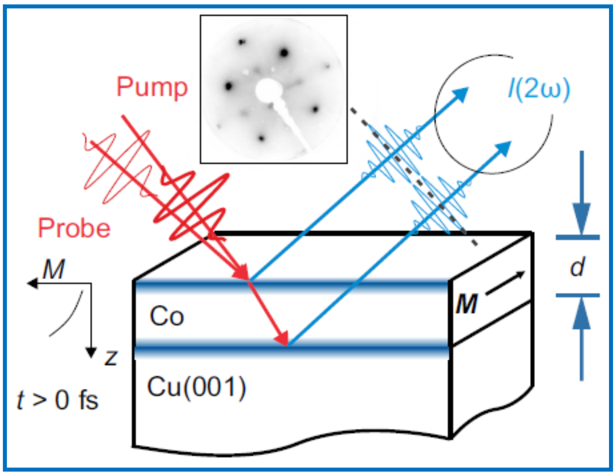

Fig. 1 Schematic experimental setup for analyzing the interface-sensitive mSHG yield in reflection from Co/Cu(001) for different Co film thicknesses d ranging from 0.4 nm to 10 nm in a pump-probe experiment. Ultrafast demagnetization due to the pump pulse will lead to a spatially inhomogeneous change in the magnetization M along the film depth z.

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Tuesday, 13 June 2017

Plasmonics Nanooptics

9:00 **Claus Ropers (invited)**
Ultrafast Electron Diffraction and Microscopy Enabled by Nanoscale Photoemitters

9:40 **Markus Raschke (invited)**
Ultrafast Nanoscopy: Imaging Structure, Function, and
Dynamics of Matter on its Natural Length and Times Scales

11:00 **Peter Hommelhoff (invited)**
Quantum Path Interference in Two-Color Coherently Controlled Electron Emission off Tungsten Tips and in Attosecond Electron Dynamics in Graphene

Novel Phenomena and Materials

11:40 **Michael Horn-von Hoegen (invited)**
Ultrafast non-Equilibrium Structural Dynamics in the Si(111)-In (8x2)<>(4x1) System at the Quantum Limit

15:40 **Brian Moritz (invited)**
Optical Materials Design of TMDCs and Frustrated Mott Insulators

16:20 **Frank Oliver Schumann**
Correlated Electron Dynamics at Surfaces Investigated via He Ion Neutralization

16:40 **Alexander Paarmann**
Mid-Infrared Nonlinear Nanophotonics Using Surface Phonon Polaritons

17:40 **Kenta Kuroda (invited)**
Optical Control over Spin-Polarized Surface States in Topological Insulators

18:20 **Kazuki Sumida**
Temperature-induced Prolonged Duration of non-Equilibrium Surface Dirac Fermions in Topological Insulators

18:40 **Yoshiyasu Matsumoto**
Enhancement of Hole Trapping at Curved Surface of Titania Nanoparticles
with Water Adsorbate

Ultrafast Electron Diffraction and Microscopy Enabled by Nanoscale Photoemitters

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Novel methods in ultrafast electron microscopy [1], diffraction and spectroscopy promise unprecedented insights into the dynamics of structural, electronic and magnetic processes on the nanoscale. A key to the realization of such technologies is the generation of high-quality beams of ultrashort electron pulses.

This talk will present the development and application of ultrafast imaging and spectroscopy based on localized electron sources, with a focus on Ultrafast Low-Energy Electron Diffraction (ULEED) [2,3]. Initially developed in a transmission geometry for monolayer and bilayer films [2], ULEED now allows for the study of structural dynamics from surfaces in backscattering diffraction [3]. Using this technique, we investigate the transformation between charge-density wave (CDW) phases at the surface of the transition metal dichalcogenide 1T-TaS_2. Harnessing the high transverse coherence of the low-energy electron beam, we resolve the coarsening of a nascent incommensurate CDW phase, and identify the phase-ordering kinetics with the annihilation of topological defects in the charge order.

In the future, based on a continuing miniaturization of ultrafast electron guns [4], ULEED will enable the study of structural dynamics in a wide range of surface systems, including surface reconstructions and molecular adsorbates.

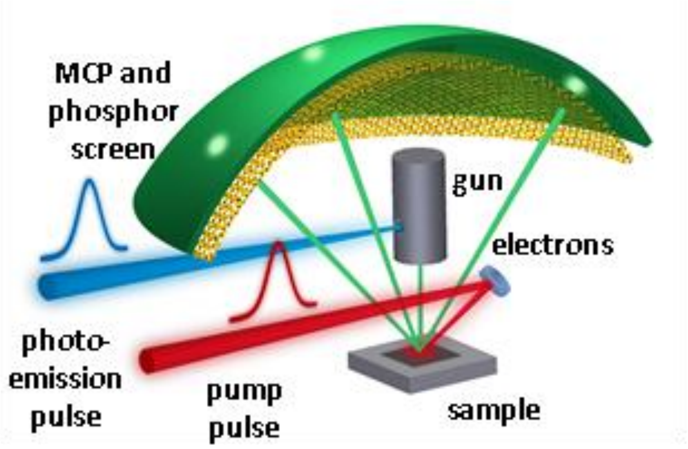

Fig. 1: Schematic of Ultrafast Low-Energy Electron Diffraction (ULEED)

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Ultrafast Nanoscopy: Imaging Structure, Function, and Dynamics of Matter on its Natural Length and Time Scales

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I will present the advances in multimodal linear, nonlinear, and spatio-temporal nano-imaging for the study of fundamental optical and plasmonic phenomena, coupled single molecule or quantum dynamics, with unprecedented nanometer spatial and femtosecond resolution, sensitivity and precision [1-6]. To gain the desired *simultaneous nanometer spatial resolution with spectroscopic specificity and femtosecond temporal resolution* we combine plasmonic and optical antenna concepts with ultrafast and shaped laser pulses to precisely control optical excitation on femtosecond time and nanometer length scales from the visible to THz spectral range. In the implementation with scattering scanning nearfield microscopy (*s*-SNOM) or other tip-enhanced microscopy modalities with nonlinear, ultrafast, and IR and Raman vibrational spectroscopies, the resulting enhanced and qualitatively new forms of light-matter interaction enable deep-subwavelength spatially resolved imaging of heterogeneities and nano-confinement as they define the properties of most functional materials. I will present several new concepts extending tip-enhanced spectroscopy into the nonlinear and ultrafast regime for nano-scale imaging and spectroscopy of surface molecules and nanosolids. Examples include the adiabatic nano-focusing for nm-resolved imaging of the few-fs plasmon coherence [1] (Fig. 1), ultrafast and nonlinear probing of structure and dynamics in quantum materials [2-4], vibrational nano-imaging of molecular structure, coupling, and dynamics down to the single molecule level [5,6] (Fig. 2), and the transition from classical to quantum plasmonic emitter coupling and femtosecond control for electron wavepackets for ultrafast electron imaging and STM.

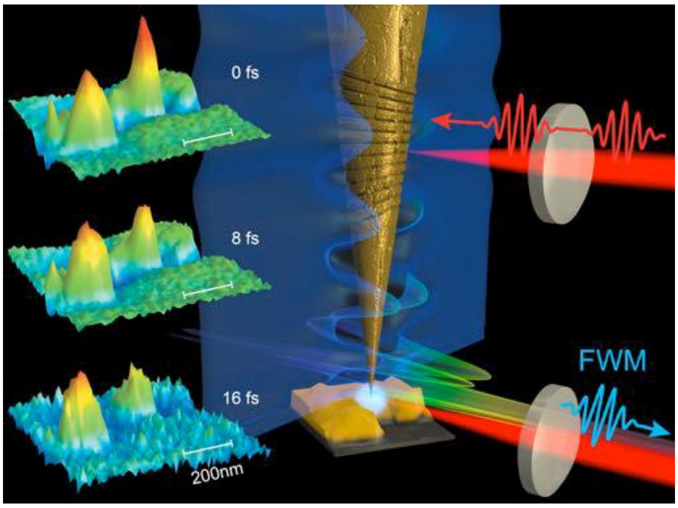

Fig. 1. Nonlinear four-wave mixing nano-focused imaging of coherent plasmon dynamics with few femtosecond and nanometer resolution [1].

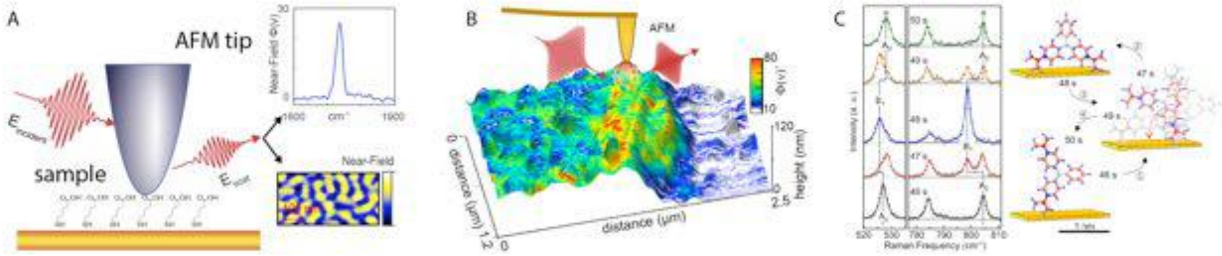

Fig. 2. Tip-enhanced/tip-scattering vibrational Raman and IR nano-spectroscopy and -imaging probing structure, order, and dynamics in self-assembled monolayer (A), molecular electronic materials (B), and intra- and intermolecular vibrational redistribution in single molecule spectral diffusion (C).

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Quantum Path Interference in Two-Color Coherently Controlled Electron Emission off Tungsten Tips and in Attosecond Electron Dynamics in Graphene

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Two topics around the interaction of surfaces with well-controlled femtosecond laser pulses will be discussed. In the first part, we will show that multi-photon photoemission from the apex of a tungsten tip can be coherently controlled with the optical phase between two light fields. With femtosecond laser pulses at around 1560 nm and their second harmonic focused on the tip, we observe an emission current modulation as large as 97.5 % as function of the relative phase between the pulses. Two different quantum emission channels in the material interfere with a high degree of coherence, which we ascribe to the confined nature of the emission site. Time-dependent density functional theory (TDDFT) simulations and ground-state density functional theory (DFT) support the observations.

In the second part of the talk, we will discuss the interaction of two-cycle carrier-envelope phase (CEP) controlled laser pulses at around 800 nm with a stripe of monolayer graphene on SiC. We observe a telltale change of the direction of the CEP-dependent current induced in the graphene stripe when we increase the optical peak field strength. We explain this and other observations with a transition from the weak-field into the strong-field regime. In the strong-field regime, coupled intraband-interband electron dynamics arise. The electron excitation can be interpreted as Landau-Zener transitions from the valence to the conduction band. For linear polarization, repeated Landau-Zener transitions events are possible within a single optical cycle, resulting in quantum path interference for reaching the same final state in the conduction band. These repeated coherent crossings of Landau-Zener transition regions give rise to Landau-Zener-Stückelberg interferometry, which we observe for the first time in a (quantum) system not engineered for the purpose.

Both experimental results bear promise for applications of light-field control of electrons in various settings, and on sub-optical, i.e. attosecond, time scales.

Ultrafast non-Equilibrium Structural Dynamics in the Si(111)-In (8x2)<->(4x1) System at the Quantum Limit

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The Indium induced (4×1) reconstruction on Si(111) is a famous prototype for 1D atomic wires at surfaces. Indium atoms form parallel zigzag chains with anisotropic metallic conductivity. At 130 K a metal-insulator transition to the (8×2) ground state takes place. A Peierls-like distortion causes periodicity doubling, opening of a bandgap, and formation of a CDW. A robust hysteresis of 10 K width during temperature cycling proves that this phase transition is first-order [1]. The non-equilibrium structural dynamics of the (8×2) is studied by ultra-fast electron diffraction [2,3]. We use a pulsed electron gun in a RHEED geometry with a femtosecond-laser system in a pump probe setup. A tilted pulse front scheme [4] improves the temporal resolution to less than 350 fs.

Upon photo excitation the (8×2) ground state is driven in 350 fs to the (4×1) excited state as observed through the transient RHEED spot intensity. The transition is described in an accelerated displacive excitation scenario which relies on transient changes in the potential energy surface. The population of specific long living electronic states facilitates the structural transition as they directly couple to the characteristic soft phonon modes. The strong coupling between substrate and adsorbate is responsible for the sub-picosecond structural response by dephasing and damping the characteristic phonons in 1/4th of their oscillation period.

Transient heating of the In atoms from 30 to 80 K occurs delayed on a time scale of 2.2 ps. Thus the phase transition is driven by electronic entropy and not thermally [4]. Cooling of the In layer to the substrate occurs on a time scale of 20 ps. An energy barrier for the atoms collective motion from the (4x1) state to the (8x2) state hinders the immediate recovery of the ground state: the In layer remains for nanoseconds in a super cooled metastable (4×1) state, which is not accessible under equilibrium conditions. The relaxation into the (8×2) ground state happens through the nucleation of the (8×2) at pre-existing adsorbates [5,6] which trigger a 1-dim. Recrystallization front propagating with 100 m/s as determined from a transient spot profile analysis of the (8×2) spots.

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Optical Materials Design of TMDCs and Frustrated Mott Insulators

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We discuss manipulation of the topology of a band insulator and induction of topological order in a Mott insulator, accessing transient Floquet steady states via broad pump pulses. First, a realistic material description of monolayer transition-metal dichalcogenides (TMDCs) leads to a novel mechanism to optically induce topologically-protected chiral edge modes, facilitating optically-switchable conduction channels, that are insensitive to disorder. Our strategy is to understand non-equilibrium Floquet-Bloch bands and topological transitions directly from *ab initio* calculations, using WS_2 to illustrate control of chiral edge modes not qualitatively sensitive to microscopic materials details [1]. We extend these ideas to strongly-correlated systems and show that pumping frustrated Mott insulators with circularly-polarized light can drive an effective spin system across a phase transition to a chiral spin liquid (CSL). The transient time evolution of a Kagome lattice Hubbard model is well-captured by an effective spin description, where circular polarization promotes a staggered scalar spin chirality directly to the Hamiltonian level. We fingerprint the phase diagram and find a stable photo-induced CSL in proximity to the equilibrium ground state [2]. These results suggest new avenues to marry dynamical symmetry breaking, strong interactions, and *ab initio* materials modelling, to access elusive phase transitions that are not readily accessible in equilibrium.

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Correlated Electron Dynamics at Surfaces Investigated via He Ion Neutralization

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The availability of intense and short light pulses has opened up a new and active research field. This allows to address fundamental questions on the time evolution of the electron dynamics leading to electron emission. For example, if two photons in close temporal proximity excite a sample, will the system react as if two independent energy quanta are donated, or does it recognize the two photons as one energy quantum? Theory answered this for two-photon double ionization of He via an intense photon beam with 70 eV photons. The time scale of interest lies in the range 300 as- 4.5fs. [1]

We describe an experiment on a solid surface using an alternative route. The neutralization of a single He^{2+} ion near a metal surface is usually described in two sequential steps. This makes two energy quanta available which are known to result in electron emission via an Auger type process. The neutralization of these two steps takes place on a time scale of about 2-20 fs. Hence, there is a finite probability for two energy quanta to become available in the interesting range of 300 as to 4.5 fs.

We show that the neutralization of one He^{2+} ion leads to the emission of an electron pair. Via coincidence spectroscopy we give evidence that a sizable amount of these electron pairs originate from a correlated single step neutralization of the ion involving a total of 4 electrons from the metal. These correlated electron pairs cannot be explained in the common picture of two consecutive and independent neutralization steps. We infer a characteristic time scale for the correlated electron dynamics in the metal of 40-400 as. This means within this time the surface recognizes the two energy quanta as one unit. [2]

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Mid-Infrared Nonlinear Nanophotonics Using Surface Phonon Polaritons

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Surface polaritons are the key building block of nanophotonics, since these excitations provide sub-diffractional light localization accompanied by significant optical field enhancements. Many previous studies have focused on surface plasmon polaritons (SPPs) at the surface of noble metals. Recently, an alternative approach was introduced using surface *phonon* polaritons[1] (SPhPs) which can be excited in the mid-infrared at the surface of polar dielectrics, where optical phonon resonances lead to the negative dielectric permittivity required for the surface polariton formation.

Here, we undertake a first step towards the largely unexplored domain of mid-infrared nonlinear nanophononics. We study the nonlinear-optical response of (i) localized SPhPs in sub-diffractional nanostructures[2] and (ii) propagating SPhPs using Otto-type prism coupling.[3] Employing an infrared free-electron laser as an intense, narrowband, and tunable excitation source for second harmonic generation (SHG) spectroscopy[4] in both approaches, we observe drastic enhancement of the SHG yield for resonant excitation of SPhPs.

We discuss the unique features of SPhPs as opposed to SPPs, including the large selection of non-centrosymmetric SPhP materials leading to a wide spectral tunability. In combination with the high quality of the SPhP resonances, several appealing scenarios emerge which facilitate SPhPs for nonlinear and ultrafast surfaces studies, for instance SPhP-enhanced surface vibrational spectroscopy.

[3] J. D. Caldwell et al.,Nanophotonics **4**, 1 (2015).
[4] I. Razdolski et al., Nano Letters **16**, 6954 (2016).
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[6] A. Paarmann et al., Applied Physics Letters **107**, 081101 (2015).

Optical Control over Spin-Polarized Surface States in Topological Insulators

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A number of challenging efforts have been recently made for a coherent control of highly spin-polarized topological surface states (TSS) in various topological insulators, suggesting ultrafast optospintronic devices based on the TSS. However, these attempts generally neither realized a selective photoexcitation of the TSS, since the optical response is typically governed by the bulk properties, nor do they exclusively probe the excitation of the TSS, because the applied techniques are not surface sensitive. In my talk, I will introduce two surface-sensitive photoemission experiments that directly visualize surface photoexcitation realizing coherently control over the spin-polarized TSS.

The first topic is a direct band mapping of a photoexcitation in the TSS of Sb_2Te_3 with time-resolved two-photon photoemission using ultrashort mid-infrared laser pulses. As the most remarkable finding, the mid-infrared excitation permits a direct population of the unoccupied TSS owing to a novel optical coupling across the Dirac point. In addition, the direct optical transition induces a pronounced asymmetry of the transient TSS population in *k*-space which indicates an excitation of a net spin-polarized photocurrent even with linear pump polarization [Phys. Rev. Lett. **116**, 076801 (2016)]. Moreover, the pump polarization can control the asymmetric population, *i.e.* the surface photocurrent, through the coherent optical transition of the TSS, [Phys. Rev. B **95**, 081103(R) (2017)]. By observing the decay of the asymmetric population, the ultrafast dynamics of the photocurrent in the surface Dirac-cone of the TSS is directly investigated.

Secondary, I will show coherent spin control of photoelectrons from Bi_2Se_3 by using spin- and angle-resolved photoemission spectroscopy combined with polarization variable 7-eV laser (Laser-SARPES). It is demonstrated that the direction of the photoelectron spin polarization can be easily controlled in three-dimension only by the direction of the laser field. A brief photoemission model with considerations of quantum interference processes reproduces the results. This result will increase the capabilities of SARPES to directly access the spin-dependent quantum interference [Phys. Rev. B **94**, 165162 (2016)].

This work was conducted in collaboration with J. Riemann, J. Güdde, U. Höfer, K. A. Kokh, O. E. Tereshchenko, A. Kimura, K. Yaji, F. Komori, T. Kondo, S. Shin. Funding by the Deutsche Forschungsgemeinschaft and JSPS Postdoctoral Fellowship for Research Abroad and SFB 1083 are greatly acknowledged.

Temperature-induced prolonged duration of non-equilibrium surface Dirac fermions in topological insulators

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Topological insulators (TIs) possess the spin-polarized massless Dirac fermions at the surface or the interface exhibiting much suppressed backscattering protected by time-reversal symmetry. Recently, the ternary TIs $(Sb_{1-x}Bi_x)_2Te_3$ have attracted a great deal of attention due to its tunable bulk carrier [1]. In addition, the quantum Hall effect (QHE) that stems from surface Dirac fermions was observed in $(Sb_{1-x}Bi_x)_2Te_3$ films under a magnetic field [2]. Furthermore, Cr- or V-doped films have been found to show the anomalous QHE [3,4]. Therefore, the ternary TIs $(Sb_{1-x}Bi_x)_2Te_3$ are the most promising candidate for the dissipationless device applications. In order to reveal the transport properties, here, we have performed time- and angle-resolved photoemission spectroscopy implementing pump-probe method [5] for $(Sb_{1-x}Bi_x)_2Te_3$.

Figure 1 shows the band dispersion and the temperature dependence of recovery time at the upper Dirac cone (UDC) of $(Sb_{0.73}Bi_{0.27})_2Te_3$. It clearly indicates that the recovery time drastically elongated upon increasing temperature. Concerning the mechanism of the recovery, we consider that the electron-phonon scatterings are playing a minor role. In the ~10-ps or ~100-ps time region, the electron-hole recombination thus may be the dominant channel for the slow recovery. Moreover, this temperature induced long duration is also due to the Pauli blocking effect at Dirac cone. In the presentation, we also show the prolonged duration of the intrinsic TIs.

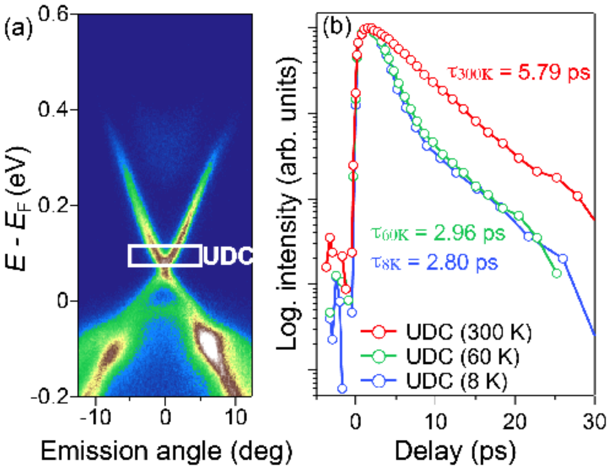

Fig. 1 (a) Photo-excited band dispersion E(k) of $(Sb_{0.73}Bi_{0.27})_2Te_3$ *recorded at pump-probe delay time t = 1.33 ps. (b) Temperature dependence of recovery time at the upper Dirac cone.*

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[5] Y. Ishida et al., Rev. Sci. Instrum. **85**, 123904 (2014).

Enhancement of Hole Trapping at Curved Surface of Titania Nanoparticles with Water Adsorbate

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Water splitting with heterogeneous photocatalyst has great potential for establishing a solid route for direct conversion of sunlight to chemical fuel, i.e., hydrogen. Despite intensive research in the past, the mechanism of water splitting still remains unclear. Photogenerated carriers created in a photocatalyst nanoparticle are rapidly trapped, and successive redox reactions take place at trap sites through trap-mediated charge transfer at the surface of a photocatalyst nanoparticle. Thus, it is of paramount importance to clarify carrier trap sites and the carrier trapping mechanism. We have tackled this problem experimentally and theoretically. The choice of photocatalyst in this study is anatase TiO_2, a benchmark photocatalyst, with two distinguishably different particle shapes: nanosphere (NS) and well faceted nanocrystalline (NC).

Water adsorption coverage was carefully controlled by changing water vapor pressure, and IR spectra and picosecond transient absorption in the mid IR were measured. In the case of NS, we have observed surface species with substantially red shifted OH stretching frequencies and an increase in hole trapping ability with increasing water coverage. In contrast, we have not observed such species and any overage dependence of hole trapping ability in the case of NC. IR spectra and hole trapping ability of surface species on NS were simulated with large-scale DFT calculations. The calculations clearly indicate that a strongly bound surface OH group at the curved surface of NS can trap a hole and this trapping ability is enhanced by hydrogen bonding with a neighboring water molecule.

Both the experimental and theoretical results provide a clear understanding of the mechanism of hole trapping that is an important step in the early stage of water oxidation on TiO_2. This finding not only leads to a molecular-level understanding of water oxidation with metal oxides, but also provides a new insight to the strategy of surface engineering of photocatalysts.

Wednesday, 14 June 2017

Low Dimensional Materials

9:00 **Philip Hofmann (invited)**
Electron Dynamics in Single-Layer Transition Metal Dichalcogenides

9:40 **Ralph Ernstorfer (invited)**
Momentum-Resolved View on Electrons, Phonons and Their Coupling in WSe_2

11:00 **Robert Wallauer**
Intervalley Scattering Dynamics in MoS_2 *Imaged by 2PPE with High-harmonic Probe*

11:20 **Ermin Malic (invited)**
Excitonic Fingerprint of Atomically Thin 2D

12:00 **Kunie Ishioka**
Ultrashort Acoustic Pulses Generated at Buried GaP/Si Interface

12:20 **Sebastian Otto**
Dynamics of p-doped Topological Insulators $SnBi_xSb_{2-x}Te_4$ *and* $GeSb_2Te_4$

Electron Dynamics in Single-Layer Transition Metal Dichalcogenides

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Changing the dimensionality of a material results in significant modifications of its electronic properties. This is even the case if the parent material already has a layered structure with little interaction between the layers, as in the case of graphene, bilayer graphene and single-layer transition metal chalcogenides.

While the static electronic properties of novel two-dimensional materials can be studied by standard angle-resolved photoemission spectroscopy (ARPES), investigations of the ultrafast carrier dynamics require both time- and angular resolution and thus time-resolved (TR)-ARPES. There is, moreover, the technical requirement of high photon energies since the interesting part of the aforementioned materials' electronic structure (i.e. the (gapped) Dirac cone) is placed at the two-dimensional Brillouin zone boundary. Recently, it has become possible to probe states at such high k by TR-ARPES, thanks to the arrival of ultrafast high harmonic laser sources.

Here we study single layer MoS_2 and WS_2 on different substrates [1,2] using TR-ARPES. For these materials, not only the decay of an excited carrier population is of interest but also the very size of the direct band gap. Due to strong excitonic effects and large exciton binding energies, the gap size cannot be determined by optical experiments. In TR-ARPES, it can be inferred from the simultaneous spectroscopy of the valence band and the partly populated conduction band. Depending on the dielectric properties of the substrate and the number of excited carriers, we observe a combination of static and dynamic band gap renormalization [3,4]. For WS_2 on Ag(111), we can address the valley degree of freedom by selective excitations using circularly polarized light [5].

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[3] G. Cabo et al., Nano Letters **15**, 5883 (2015).
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Momentum-Resolved View on Electrons, Phonons and Their Coupling in WSe_2

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The description of the coupling between the electronic and vibrational degrees of freedom is typically based on the assumption of thermal distribution functions. In nanoscale materials and heterostructures, however, a refined understanding of electron-phonon interaction is required. This is achievable with pump-probe techniques providing momentum-resolved information on ultrafast electron and phonon dynamics. We employ XUV-based time- and angle-resolved photoelectron spectroscopy (trARPES) and femtosecond electron diffraction (FED) to obtain a state-resolved understanding of microscopic coupling beyond ensemble-average descriptions, see figure 1. Specifically, I will discuss electron and phonon dynamics in the semiconducting transition metal dichalcogenide WSe_2. TrARPES reveals the distribution and evolution of excited states in the Brillouin zone and their potential spin- and pseudospin-polarization [1]. The complementary momentum-resolved view on phonon dynamics is obtained by FED [2]. By combining this information, a microscopic picture of electron-lattice coupling and energy flow emerges.

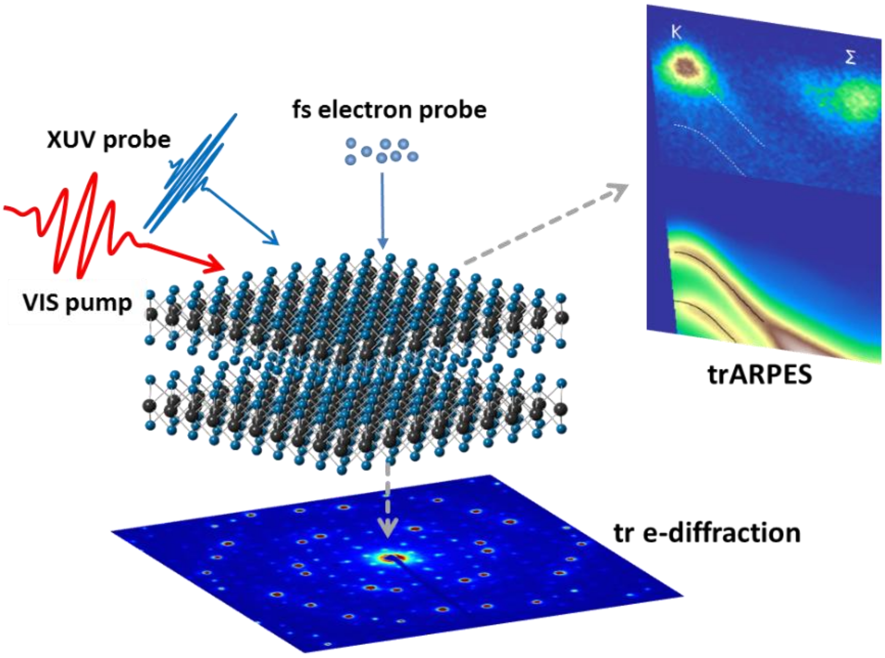

Fig. 1 Illustration of the experimental approaches used for obtaining momentum-resolved information on the ultrafast dynamics of electrons and phonons and for revealing electron-phonon coupling.

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[2] Waldecker et al., arXiv:1703.03496 (2017).

Intervalley Scattering Dynamics in MoS_2 Imaged by 2PPE with High-harmonic Probe

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We report on the application of time- and angle-resolved two-photon photoemission (2PPE) with a high-harmonic probe for the investigation of the electron dynamics in the topmost layer of bulk MoS_2 in momentum space. For this purpose, we have combined a high-repetition rate high-harmonic source [1] with tunable femtosecond pump pulses and a 3D (k_x, k_y, E) electrostatic electron spectrometer.

We discuss the application of this setup for the investigation of the electron dynamics in the conduction band of MoS_2 after optical excitation with different pump photon energies. Recently, we have shown that optical excitation above the A exciton resonance at 1.8 eV with 2.05 eV pump pulses results in an immediate occupation of the conduction band at K followed by an ultrafast transfer to the conduction band minimum at Σ [2]. Subsequently, the occupation at both high-symmetry points decays slowly on a ps timescale. We present new data for pump photon energies in the range of 1.8 - 2.3 eV and show how the dynamics of this transfer depend on the excess energy above the exciton resonance.

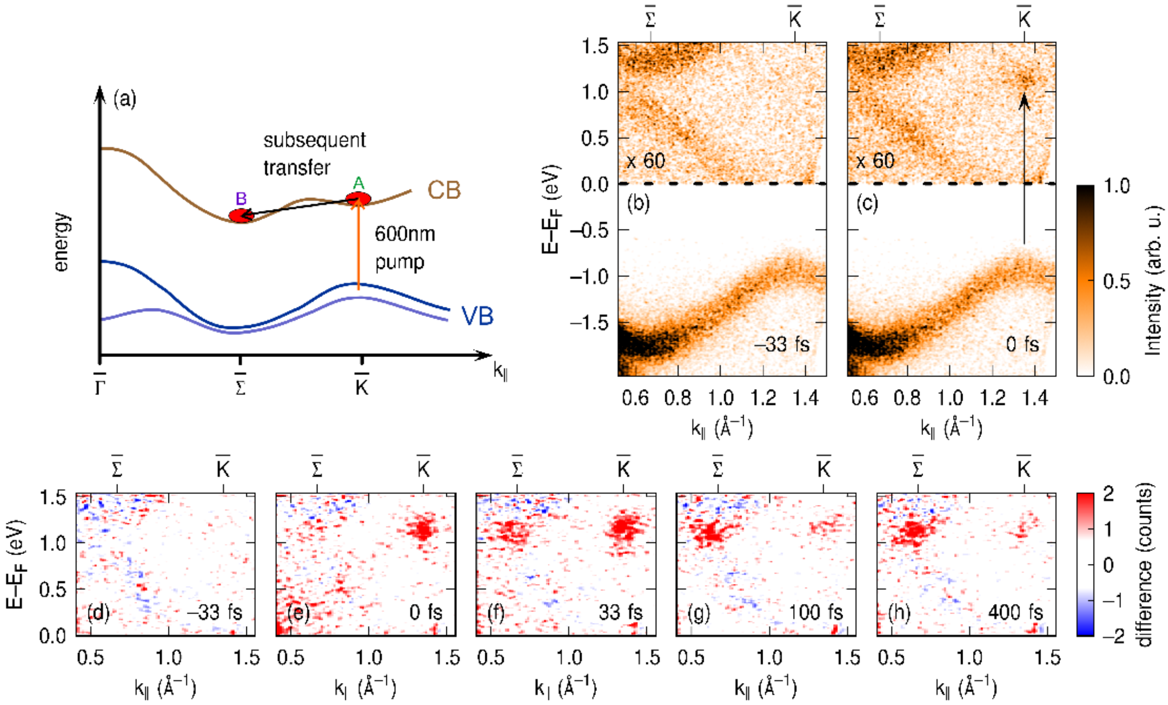

Fig 1: a) Excitation scheme of the experiment. b) Energy/momentum map before and c) at temporal overlap of pump and probe pulses. d-h) Difference spectra at various time delays.

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[2] R. Wallauer et al., Appl. Phys. Lett. **109**, 162102 (2016).

Excitonic Fingerprint of Atomically Thin 2D Materials

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Monolayers of semiconducting transition metal dichalcogenides (TMDs) build a new class of atomically thin two-dimensional materials. They exhibit a remarkably strong Coulomb interaction giving rise to the formation of tightly bound excitons. In addition to optically accessible bright excitonic states, there is also a variety of dark states that turn out to play a crucial role. To model these materials, we apply a microscopic approach combining the Wannier equation with TMD Bloch equations providing access to experimentally accessible excitonic absorption and photoluminescence spectra. In this talk, we review our recent work focusing on microscopic understanding of the excitonic finger print characterizing TMD materials:

(i) We present the excitonic absorption spectrum featuring a pronounced Rydberg-like series of excitonic transitions with binding energies in the range of 0.5 eV [1].

(ii) We investigate the microscopic origin of the homogeneous linewidth of excitonic transitions including radiative and phonon-assisted non-radiative relaxation channels as well as the appearance of exciton- phonon sidebands [2].

(iii) We show a significant disorder-induced coupling of bright and dark excitons offering a strategy to circumvent optical selection rules and make dark states visible in optical spectra [3].

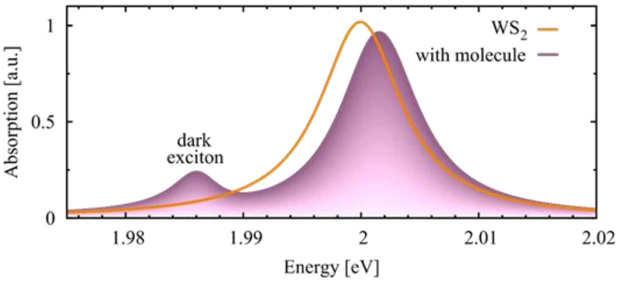

Fig. 1: Excitonic absorption spectrum of pristine and molecule-functionalized WS_2. Efficient exciton- molecule coupling gives rise to an additional peak that can be ascribed to a dark exciton [4].

(iv) We predict a novel sensor mechanism for molecules based on the appearance of an additional peak in absorption spectra (cf. Fig. 1). This peak can be ascribed to a dark exciton and only appears in the presence of attached molecules [4].

(v) We provide a microscopic view on the time- and energy-resolved formation and thermalization of bright and dark excitons as well as their impact on the photoluminescence in different TMD materials. In particular, we demonstrate the crucial role of intervalley dark excitons.

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Ultrashort Acoustic Pulses Generated at Buried GaP/Si Interface

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U. Höfer[3], H. Petek[4], C. J. Stanton[2]

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Absorption of an ultrashort laser pulse at a solid surface can lead to generation of coherent acoustic phonons in the form of an ultrasonic strain pulse. Coherent acoustic phonons can be excited efficiently via thermoelastic effect at a metal surface; in semiconductors, however, the generation is generally much less efficient and often requires a metallic thin film on top to act as an opto-acoustic transducer. In the first half of the present study we report on the intrinsic optical generation and detection of coherent acoustic phonons at (001)-oriented bulk Si and GaP without metallic phonon transducer structures [1]. We find that the generation of the acoustic pulse in these indirect semiconductors can be explained in terms of the deformation potential electron-phonon coupling. Comparison with our theoretical model reveals that the experimental strain pulses have finite build-up times of 1.2 and 0.4 ps for GaP and Si, which is comparable with the intervalley scattering time constant required for the photoexcited electrons to reach the conduction band minimum. In the second half, we report on the acoustic phonons generated at a buried interface of GaP/Si(001). GaP layers are grown by MOVPE on n-type Si(001) substrates and confirmed to have well-defined interfaces and surfaces [2]. Transient reflectivity traces exhibit pulse-like responses, which appear at later time delays for thicker GaP films. The arrival time τ of the pulse is a linear function of the GaP film thickness L. The slope, L/τ=5.83 nm/ps, is in a good agreement with the velocity of the LA phonon in GaP, confirming that a strain pulse generated at the GaP/Si interface gives rise to the pulse-like response when it arrives at the surface. The acoustic pulse shows a surprisingly short temporal width of < 1 ps, which would correspond to a spatial width of <5 nm or <20 atomic Ga-P bilayers, which suggests that it is generated exclusively at the abrupt GaP/Si interface.

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[2] A. Beyer, A. Stegmüller, J. O. Oelerich, K. Jandieri, K. Werner, G. Mette, W. Stolz, S. D. Baranovskii, R. Tonner, K. Volz, Chem. Mater. **28**, 3265 (2016).

Dynamics of p-doped Topological Insulators SnBi$_x$Sb$_{2\text{-}x}$Te$_4$ and GeSb$_2$Te

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The electronic structure of septuple-layered antimony telluride crystals with different p-doping is studied with time- and angle-resolved two-photon photoemission. All materials show a topological surface state (TSS) at the (111)-surface, where the Dirac point is found between E_F + 0.38 eV for GeSb$_2$Te$_4$ and E_F + 0.25 eV for SnBi$_{0.2}$Sb$_{1.8}$Te$_4$. After initial excitation (E_{photon} = 1.55 eV) the TSS is populated mainly from the conduction band minimum (CBM) according to the evolution of its temporal population (**Fig. 1 b**). Similar to the case of SnSb$_2$Te$_4$ [1] the electrons in the topological surface state decay rather fast into the partially unoccupied valence band maximum. With increasing amount of Bi in the SnSb$_2$Te$_4$ compound the group velocity of the TSS decreases, while a faster decay within the conduction band is observed.

The helical spin structure of the TSS is mapped with circular dichroism using circularly polarized probe pulses. Figure **1 a** shows the characteristic cross section of the asymmetry at the Dirac point. However, in none of the systems a time-dependent circular or linear dichroism was found within the TSS, using polarized pump pulses (**Figs. 1 c and d**).

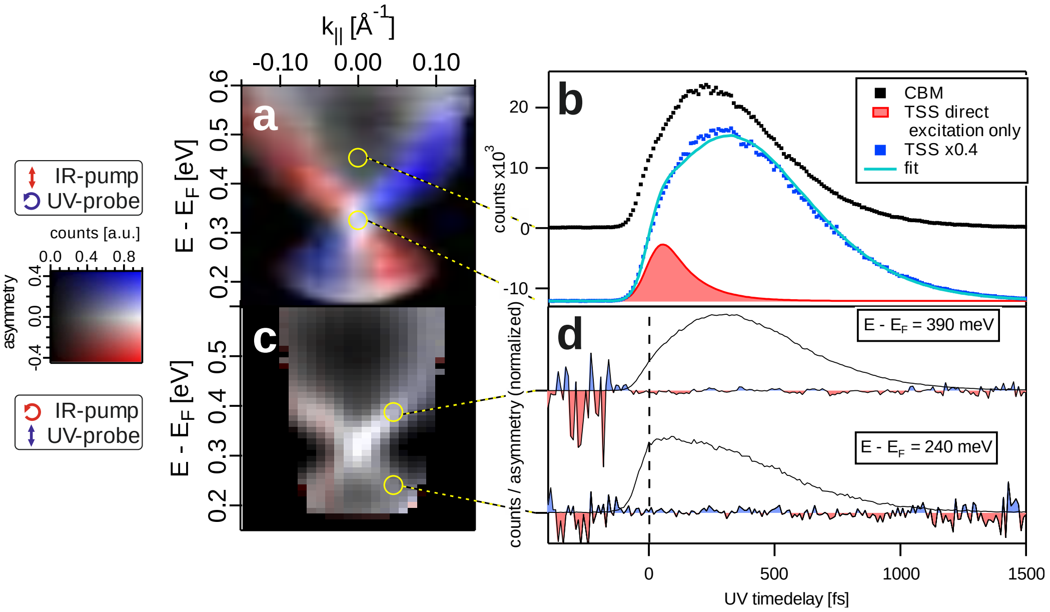

Fig. 1 a) Dirac cone of SnBi$_{0.2}$Sb$_{1.8}$Te$_4$ measured with linearly polarized pump and circularly polarized probe pulses and c) with circularly polarized pump and linearly polarized probe pulses. b) Time-resolved measurements of CBM and TSS with fit of a rate equation model [1]. d) Asymmetry and sum of counts from off normal time-resolved measurements with circularly polarized pump pulses of different helicity and linearly polarized probe pulses.

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Thursday, 15 June 2017

Low Dimensional Materials

9:00 **Isabella Gierz (invited)**
Femtosecond Control of Quasiparticle Interactions in Graphene

9:40 **Ryuichi Arafune**
Spin-Texture Inversion in Image Potential States of Ir(111) covered by Graphene

10:00 **Sven Aeschlimann**
Ultrafast Momentum Imaging of Chiral Interband Excitations in Graphen

11:00 **Guido Fratesi**
Substrate Induced Ultrafast Electron Injection Dynamics at Organic-Graphene Interfaces

11:20 **Yoav William Windsor**
Ultrafast Electron Dynamics of the Surface States of Intermetallic $GdRh_2Si_2$
Probed by XUV trARPES

11:40 **Rupert Huber (invited)**
Lightwave Electronics in Layered Semiconductors and single Molecules

Anniversary Session

15:00 **Thomas Fauster (chair)**
Introduction

15:20 **Helmut Zacharias**
Femtosecond Laser Induced Associative Desorption of Molecular Hydrogen from Graphite

16:00 **Hrvoje Petek**
Ultrafast Coherent Multidimensional Multiphoton Photoemission Spectroscopy

17:20 **Tony F. Heinz**
Ultrafast Surface Dynamics - without the Bulk: Probing 2D Materials

18:00 **Martin Aeschlimann, Ulrich Höfer**

20:20 **Martin Wolf**
The Forefront of Time-resolved Photoemission: From the Past to the Future

Femtosecond Control of Quasiparticle Interactions in Graphene

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Driven solids are different from matter at equilibrium because the interaction with a periodic driving field results in the formation of new effective Hamiltonians with new eigenstates. We exploit this idea for electronic structure control by shining tailored laser pulses onto various low-dimensional materials.

Recently, two different driving schemes have been developed: Resonant excitation of infrared-active lattice vibrations (“phonon pumping”) has been used to induce superconducting-like states at temperatures far above the equilibrium critical temperature in cuprates [1] and K_3C_{60} [2]. Further, strong circularly polarized mid-infrared fields have been used to generate photon-dressed states resulting in a topological phase transition in Bi_2Se_3 [3].

We combine these driving schemes with time- and angle-resolved photoemission spectroscopy at extreme ultraviolet wavelengths to investigate the electronic properties of driven solids over the whole first Brillouin zone and beyond.

I will show that resonant excitation of the in-plane bond-stretching phonon in bilayer graphene leads to a transient threefold enhancement of the electron-phonon coupling constant [4,5]. Although the microscopic origin of this effect is far from understood, our findings have important implications for a possible light-induced superconducting state in graphene and related materials [2]. I will end with a short outlook on other projects we are currently working on, such as one-dimensional Indium wires and C_{60} films.

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[5] E. Pomarico et al., Phys. Rev. B **95**, 024304 (2017).

Spin-texture Inversion in Image Potential States of Ir(111) Covered by Graphene

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Rashba effect, which is expressed by a Hamiltonian of HR= $\alpha_R\ \sigma\cdot(\hat{z}\times k)$, is one of the characteristic spin-orbit interactions in 2-dimensional electron gas (2DEG) at solid surfaces and interfaces. Fig. 1 shows the spin texture calculated from this Hamiltonian. Interestingly, spin-textures determined experimentally in literatures are opposite to that shown in Fig. 1; the Rashba parameter α_R determined by the experiment is negative. Strictly speaking, Rashba effect that is initially predicted by Rashba has not been detected yet at solid surfaces. While theoretical works on the sign of α_R are reported [1], a definitive conclusion has not been reached.

In this talk, we will present our recent results on angle-resolved two-photon photoemission spectra of the Ir(111) surface covered by graphene. The spin-polarized surface resonances in the occupied region [2] enable us to characterize the spin of the unoccupied states without a circular dichroism technique [3] nor a spin-resolved one [4]. We found the spin-splitting of the image potential states (IPSs) as well as the occupied surface resonance, and that α_R are positive for the high order (n ≥ 2) IPSs. This is the first result, in our knowledge, showing the spin-momentum locking that Rashba originally expected.

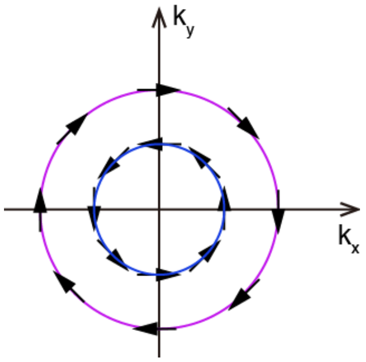

Fig. 1: Spin-texture for the Rashba model in 2DEG.

[1] H. Bentmann et al., Phys. Rev. B **84**, 115426 (2011).
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[2] Varykhalov et al., Phys. Rev. Lett. **108**, 066804 (2012).
[3] T. Nakazawa et al., Phys. Rev. B **94**, 115412 (2016).
[4] B. Schmidt et al., Phys. Rev. Lett. **95**, 107402 (2005).

Ultrafast Momentum Imaging of Chiral Interband Excitations in Graphene

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Due to the chiral nature of Dirac carriers, photo-excitation in graphene leads to a distinct anisotropic distribution of electron-hole pairs in momentum space. In fact, interband excitation is forbidden along the direction of the electric field and maximum perpendicular to it. We use time- and angle-resolved photoemission spectroscopy to track the generation and relaxation of these anisotropic photo-carrier distributions as a function of energy, momentum, and time (Fig. 1).

We show that the photo-excited electron distribution rapidly relaxes into a quasi-thermal state with an azimuth-dependent temperature, consistent with ultrafast collinear electron-electron scattering. Azimuthal relaxation is found to re-establish an isotropic electron temperature on longer timescales through optical phonon emission. We show that this process is strongly dependent on the type of static doping: In *n*-doped graphene, for which photo-excited carriers are generated close to the Fermi level, the anisotropy lives far longer than in *p*-doped graphene. We attribute this to the strong suppression of azimuthal relaxation due to a reduced phase space for optical phonon emission in the *n*-doped case.

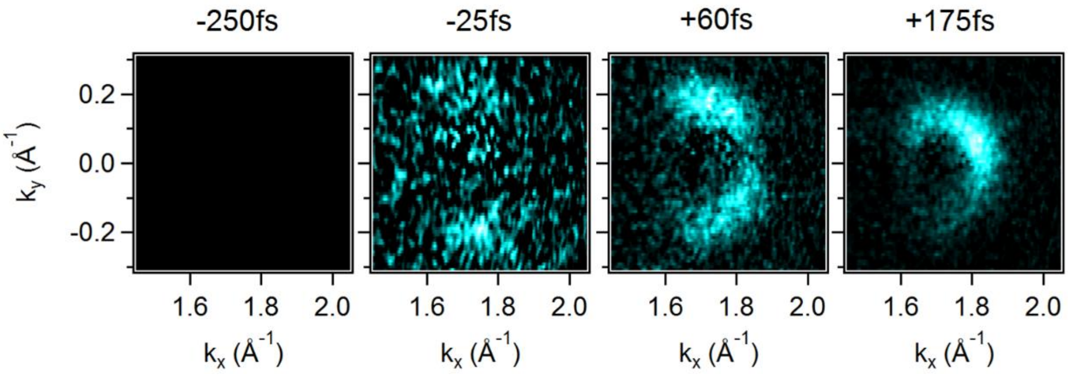

Fig. 1 Photoemission spectra of the photo-excited carriers as a function of k_x and k_y at four different time delays. t=-250fs: no excitation, t=-25 fs and t=60 fs: buildup of anisotropic carrier distribution, t=175fs: isotropic carrier distribution is obtained. The remaining azimuthal intensity modulation is due to photoemission matrix elements.

Substrate Induced Ultrafast Electron Injection Dynamics at Organic-Graphene Interfaces

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Electron core-level spectroscopies can effectively be used to investigate electron transfer rates at organic/inorganic interfaces occurring within few femtoseconds. The core-level excitation at an adsorbed molecule strongly perturbs the system and calls for a proper theoretical description. On the other hand, it induces novel phenomena such as backward electron transfer (substrate-to-molecule) as we measure by X-ray resonant photoemission and calculate by a theoretical framework based on density-functional theory (DFT) [1]. The rates can be controlled by varying molecular properties like the adsorption angle [2], as well as by tailoring the substrate like we show here for molecules on graphene.

N1s core excitation induces ultrafast electron transfer (τ=4fs) for bipyridine molecules on epitaxial graphene/Ni(111), which is characterized by a strong hybridization between C and metal states. We demonstrate that this interface can be decoupled by the addition of a second layer of graphene, so that the one in contact with the molecule is less hybridized with Ni underneath. In that case, transfer rates decrease by about one order of magnitude in the experiments and in the simulations, whereas no transfer is in principle expected for molecules on freestanding graphene within the current description.

[1] G. Fratesi, C. Motta, M. I. Trioni, G. P. Brivio, D. Sánchez-Portal, J. Phys. Chem. C **118,** 8775 (2014).

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Ultrafast Electron Dynamics of the Surface States of Intermetallic $GdRh_2Si_2$ Probed by XUV trARPES

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Rare-earth (“*RE*”) intermetallics of the form $RE\mathrm{Rh}_2\mathrm{Si}_2$ have attracted significant attention for many years due to the wide variety of exotic behaviors they exhibit, including complex magnetic phases, valence fluctuations, heavy-fermion and Kondo behavior or even superconductivity. For the specific cases of $EuRh_2Si_2$ and $GdRh_2Si_2$ it has recently been shown that their 4f antiferromagnetic (AFM) order leads to a large splitting of Si-derived Shockley surface states at the corner of the Brillouin zone (BZ) on Si-terminated surfaces [1,2]. This large exchange splitting, which is due to a ferromagnetic spin polarization of the surface, demonstrates a strong interaction of the non-magnetic Si surface states with the underlying *RE* magnetism.

To study this fascinating interplay between *RE* antiferromagnetism and the surface electronic structure in this compound, we performed time- and angle-resolved photoelectron spectroscopy (trARPES) on Si-terminated surfaces of $GdRh_2Si_2$. In order to access the relevant surface states at the M-points of the BZ, we employed our newly developed XUV trARPES setup operating at 21.8 eV photon energy, with a repetition rate of 500 kHz, and with sub-40 fs temporal resolution. Following photoexcitation by 800 nm pulses, we observe population of several unoccupied states both at the center and the border of the BZ, followed by a very fast relaxation towards the Fermi level and subsequent cooling through electron-phonon coupling within ~500 fs. Surprisingly, the exchange splitting of the surface states at the M-points reduces only on a much longer timescale of ~10 ps. Whereas this demagnetization timescale is much longer than the ones observed in most metallic ferromagnetic systems, it is comparable to the timescale of 4f demagnetization observed recently by trARPES in Gd metal [3]. This indicates a similar demagnetization process in this intermetallic compound despite the lack of net magnetization due to its AFM order.

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[2] M. Güttler et al, Sci. Rep. **6**, 24254 (2016).
[3] B. Frietsch et al., Nature. Commun. **6**, 8262 (2015).

Lightwave Electronics in Layered Semiconductors and Single Molecules

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The advent of atomically strong phase-locked laser pulses has opened the door to a new strong-field limit, where the light wave acts like a classical acceleration force [1]. Recently, intense terahertz (1 THz = 10^{12} Hz) fields have allowed us to drive dynamical Bloch oscillations and subcycle quantum interferences between multiple electron bands in bulk semiconductors, generating high-harmonic radiation over more than 13 optical octaves [2]. This mechanism can be exploited to shape the high-harmonic carrier wave [2] for solid-state attosecond sources. Strong THz fields can also accelerate and collide electrons and holes that form excitons in bulk and monolayer dichalcogenides, such as WSe_2. This novel concept of a quasiparticle collider provides key insights into the structure and dynamics of quasiparticles in a similar way to how conventional accelerators expose the nature of elementary particles [3].

Far-field spectroscopy is restricted by the diffraction limit. We explore two ways of tracing subcycle dynamics on the nanoscale: (1) Combining electro-optic sampling with near-field scanning optical microscopy we demonstrate simultaneous 10 fs and 10 nm resolution. The ultrafast nanoscope allows us to resolve ultrafast photo-switching of surface plasmon polaritons on black phosphorus [4]. (2) Lightwave electronics at the junction of a low-temperature scanning tunneling microscope allows us to control the quantum motion of single electrons into and out of a single orbital of one molecule. Utilizing this process, we take the first-ever femtosecond snapshot images and movies of a single vibrating molecule directly in the time domain and with sub-angstrom precision [5]. Our results offer a radically new way of accessing the microcosm at optical clock rates.

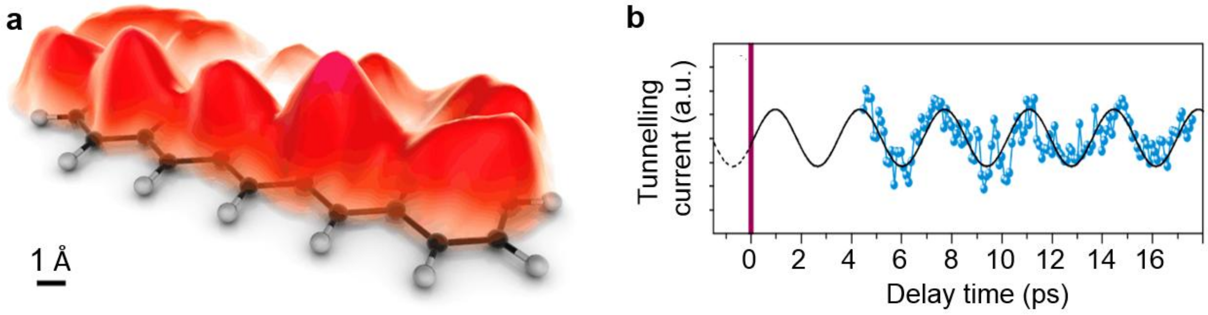

Fig. 1 a, Femtosecond snapshot of a single orbital of an individual pentacence molecule. b, Out-of-plane vibrations of this molecule are directly resolved in space and time.

[1] P. B. Corkum, F. Krausz, Nature Phys. **3**, 381 (2007).

[2] O. Schubert et al., Nature Photonics **8**, 119 (2014); M. Hohenleutner et al., Nature **523**, 572 (2015); F. Langer et al., Nature Photon., doi:10.1038/nphoton.2017.29 (2017).

[3] C. Poellmann et al., Nature Mater. **14**, 889 (2015); F. Langer et al., Nature **533**, 225 (2016).

[4] M. Eisele et al., Nature Photon. **8**, 841 (2014); M. A. Huber et al., Nature Nanotech. **12**, 207 (2017).

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Ultrafast Surface Processes - Early Ideas and Their Realization

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In the early 90ies it became evident – through discussions at various conferences and first publications – that hot electrons strongly influence the outcome of laser-induced surface reactions. Thus both the direct investigation of the electron dynamics in the time domain as well as hot electron driven actions on surfaces started to evolve. Topics like vibrational energy transfer, electronic excitations of adsorbates and substrates, and even plasmonics were discussed. A few examples will be given to sketch the development of the field since then. The talk will finish by addressing recent electronic interactions on non-metal systems, like strongly coupled optical phonons and excitons.

Ultrafast Coherent Multidimensional Multiphoton Photoemission Spectroscopy

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In the 20 year history of the symposium on Ultrafast Surface Dynamics our understanding of the interaction of light with solids and solid surface has greatly expanded, yet the role of coherence remains poorly understood. We usually take the comforting notion that optical transitions occur between black lines that are connected by upward arrows, but experience, which we often ignore, tells us that this is not always the case. I will introduce two examples where the Coherent Multidimensional Multiphoton Photoemission Spectroscopy [1-3] provides insights into the coherent response of metal surfaces. One example will address the d-band catastrophe in copper, where we find that the interband bulk transitions strongly affect a surface 3PP process even though they are otherwise spectroscopically silent. In another example, I will discuss how attosecond coherent multi-electron Coulomb scattering enables optical processes that seemingly violate energy conservation and energy-time uncertainty in a femtosecond 3PP experiment.

[1] X. Cui, C. Wang, A. Argondizzo, S. Garrett-Roe, B. Gumhalter, H. Petek, Nature Phys. **10**, 505 (2014).

[2] S. Zhang, C. Wang, X. Cui, Y. Wang, A. Argondizzo, J. Zhao, H. Petek, Phys. Rev. B **93**, 045401 (2016).

[3] S. Tan, L. Liu, Y. Dai, J. Ren, J. Zhao, H. Petek, H., J. Am.Chem. Soc. (ASAP) (2017).

Ultrafast Surface Dynamics - without the Bulk: Probing 2D Materials

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In this talk, we will ask the audience to indulge us in some reflections on the remarkable progress we have witnessed over the history of the USD meeting in our ability to probe important and fascinating dynamical phenomena occurring at surfaces and interfaces. In terms of measurement techniques, what were once very new methods like time resolved-photoemission and nonlinear optical spectroscopy have evolved from rather esoteric approaches to widely used probes. At the same time, new experimental methodologies, such as time-resolved x-ray spectroscopy, are emerging and offer great potential for further advances in the field.

On the materials side, we will also provide an update on the evolution of materials that solve the common problem of discriminating against the bulk response by the most unsubtle approach possible, i.e., by simply by not having any bulk. Here we refer to the 2D van der Waals materials, including graphene and the 2D semiconductors, such as transition metal dichalcogenide monolayers. The ultrafast dynamics of these systems and their heterostructures have been the subject intense interest and excitement. We will discuss some of these recent results, as well as possible avenues for future research, by way of welcoming the next decades of USD.

The Forefront of Time-resolved Photoemission: From the Past to the Future

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In this after dinner talk I will present a short history of milestones in time- and angle-resolved photoemission spectroscopy and will honor, in particular, the important contributions of Martin Aeschlimann and Ulrich Höfer to this field. Based on a critical look where we stand now I will discuss some challenges and directions for future developments.

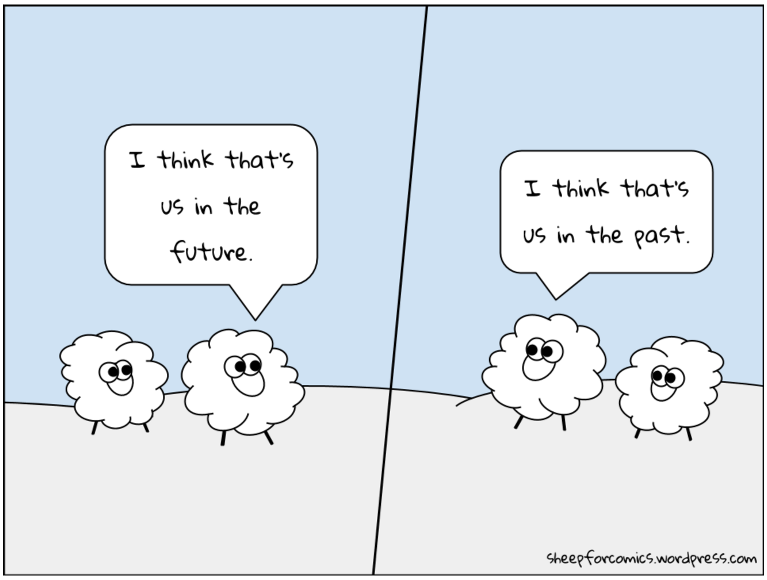

Fig. 1 …from the past to the future…

Friday, 16 June 2017

Low Dimensional Materials

9:00 **Manuel Ligges**
Time-domain Identification of Electronic Correlations in 1T-TaS_2

9:20 **Selene Mor**
Ultrafast Electronic Band Gap Control in an Excitonic Insulator

Attosecond Surface Science

9:40 **Eugene Krasovskii (invited)**
Kinematic and Dynamic Properties of a Bloch-Wave Packet Excited by an Ultrashort Light Pulse

11:00 **Walter Pfeiffer**
Intra-atomic Delays in Attosecond Time-resolved Solid State Photoemission

11:20 **Johann Riemensberger**
Energy Dependent Photoemission Delays from the (0001) Surface of Magnesium

11:40 **Luca Castiglioni (invited)**
Attosecond Dynamics in Photoemission from Noble Metal Surfaces

Time-domain Identification of Electronic Correlations in 1*T*-TaS$_2$

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Strongly correlated systems exhibit intriguing properties, caused by intertwined microscopic interactions that might be hard to disentangle under equilibrium conditions. Here, we used time-resolved photoemission spectroscopy to isolate and access fundamental Mott-physics in the (quasi) two-dimensional transition-metal dichalcogenide 1*T*-TaS$_2$. Doublon-holon recombination is studied by monitoring the upper Hubbard band that is observed in the commensurate and nearly commensurate phases of the system. The dynamics are found to occur on time scales as short as only a few electronic hopping cycles $\hbar/J$. Despite strong electron-phonon coupling in 1*T*-TaS$_2$ the dynamics can be reproduced within a single-band Hubbard model in the absence of additional bosonic coupling channels. We furthermore find that hole-doping governs the relaxation dynamics on ultrashort time scales, which opens the way for macroscopic control of ultrafast processes in such systems.

Ultrafast Electronic Band Gap Control in an Excitonic insulator

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Strongly correlated material systems often exhibit a rich phase diagram originating from the interaction between various degrees of freedom. Monitoring the electron and lattice dynamics after photoexcitation helps to disentangle the elementary processes associated with phase transitions and verify if a phase transition can be photoinduced. Here, we studied the ultrafast dynamics of Ta_2NiSe_5 by means of time- and angle-resolved photoelectron spectroscopy (trARPES) and transient reflectivity measurements. Ta_2NiSe_5 is proposed to support an excitonic insulator (EI) phase below $T_c \approx 328$ K, combined with a structural change. Such an EI phase is expected to occur in small gap semiconductors with strong electron-hole interaction as excitons can form spontaneously and condense into a ground state. Below T_c trARPES around Γ shows a strong fluence dependent valence band depopulation. This effect saturates at 50% of the equilibrium occupancy, above a critical fluence F_C = 0.2 mJ cm^{-2}, causing complete absorption bleaching. This is also reflected in a saturation of the transient optical response in the near-IR at F_C. A coherent phonon at 4 THz, characteristic of the EI/monoclinic phase, persists above F_C (Fig. 1), indicating that the photoinduced *structural* transition is hindered by pump absorption saturation. trARPES reveals that below F_C the band gap *shrinks* transiently due to photoenhanced screening of the Coulomb interaction (Fig. 2). However, above F_C the band gap at Γ transiently *widens* and recovers to its equilibrium value after ≈ 1.5 ps. Hartree-Fock calculations reveal that the band gap widening is due to photoenhancement of the exciton condensate, persisting until the system undergoes interband relaxation. Our results prove it is possible to manipulate exciton condensates with light and to gain ultrafast control of semiconductor band gaps.

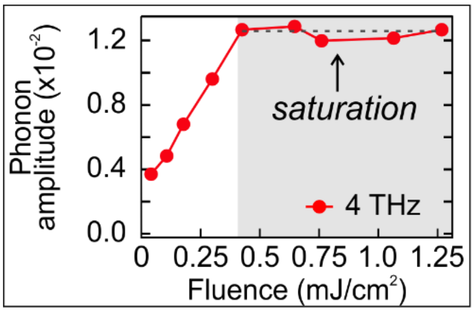

Fig. 1 Amplitude of the 4 THz phonon function of fluence.

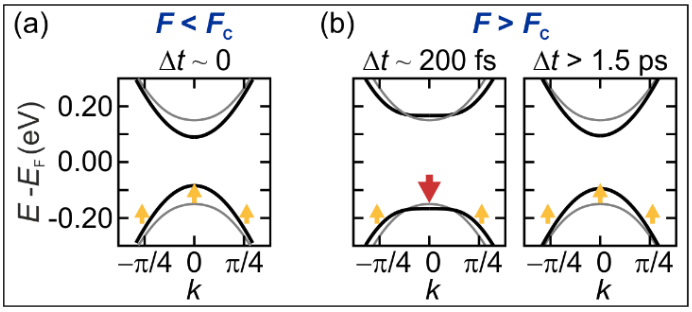

Fig. 2(a) Band gap narrowing for $F < F_C$. (b) At Γ band gap mode as a widening followed by quasi-thermal shrinking for $F > F_C$.

Kinematic and Dynamic Properties of a Bloch-Wave Packet Excited by an Ultrashort Light Pulse

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Electron excitation and transport through the surface in an attosecond streaking experiment [1] on a solid is discussed. We emphasize the crucial role of the exciting field in electron propagation and show that owing to a strong lattice scattering of the photoelectron during the optical pumping, the wave packet may move faster than with the group velocity and faster than the free electron [2]. The interaction of the packet with the crystal lattice slows down the temporal evolution of its energy spectrum, Fig. 1, and while the spectrum keeps changing the packet propagates faster than a packet with a stationary spectrum. At the same time, for a weak lattice scattering, the phenomenologi-cal result that the photoelectron moves with the group velocity $dE/d\hbar k$ and traverses on average the distance equal to the mean free path is proved to hold even at very short travelling times. The quantum nature of the outgoing electron is shown to become important when it is accelerated by a laser field inside the crystal at energies sufficiently close to a band gap [3]. The results are obtained by solving the non-stationary Schrödin-ger equation numerically exactly for a model of laser-assisted photoemission that realistically describes the lattice scattering [3]. Apart from the standard treatment of inelastic processes with an absorbing potential, a microscopic approach to electron damping is introduced. These findings offer a novel interpretation of the delay time in streaking experiment and shed new light on tunneling in opto-electronic devices.

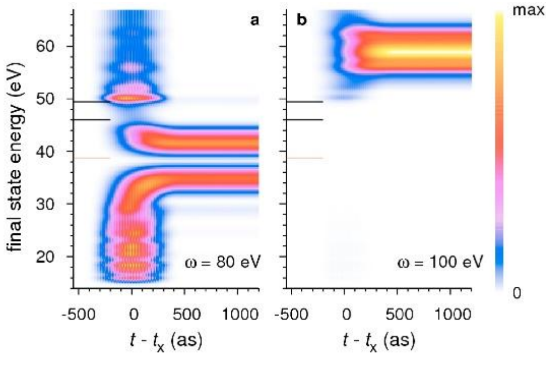

Fig. 1 Temporal evolution of the photoelectron spectrum from a localized state at E=-41 eV excited by a 1000 asec light pulse and $\hbar\omega$=80 eV (a) and 100 eV (b). The gaps in the spectrum (at 39 and at 48 eV) are caused by the interaction with the crystal lattice. Owing to the gaps, at 80 eV the evolution is much slower: at t=200 asec the spectrum is still spread over a range of 40 eV.

[1] L. Cavalieri et al., Nature **449**, 1029 (2007).
[2] E. E. Krasovskii, C. Friedrich, W. Schattke, P. M. Echenique, Phys. Rev. B **94**, 195434 (2016).
[3] E. E. Krasovskii, Phys. Rev. B **84**, 195106 (2011).

Intra-atomic Delays in Attosecond Time-resolved Solid State Photoemission

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Attosecond EUV pulses allow investigating electron dynamics with attosecond temporal resolution by recording shifts of the photoelectron streaking in a simultaneously present IR field [1]. The origin of the relative photoemission delays is not yet fully understood demonstrating our limited understanding of the photoemission process.

As demonstrated here attosecond time-resolved photoemission from the van der Waals crystal WSe_2 combined with theoretical modelling shows that intra-atomic effects such as the centrifugal barrier and other intra-atomic corrections significantly affect the relative delays between different emission channels. Identification of this effect is enabled both by the long-term stability of the inert WSe_2 surfaces and by the detection of four qualitatively different emission channels (VB, Se 4s, W 4f, and Se 3d core levels). The observed delays cannot be attributed to photoelectron propagation in the solid but intra-atomic corrections must be taken into account to match the experimental results. The latter effect is known for photoemission from free atoms [2] but, most interestingly, requires a revision of models for solid state photoemission from localized states.

State-of-the-art photoemission models emphasize the translational invariance in the solid and include scattering by the surface. In contrast, the effect of the significant intra-atomic delay is related to the fact that the photoelectron initially experiences a spherically symmetric environment. Only after some time, as the wave propagates to neighboring atoms, the photoelectron feels the structure of the crystal. This sheds new light on the fundamental mechanism involved in the photoemission process and shows that a refined model of photoemission accounting for these local effects is needed to correctly model the kinematics of the photoemission process.

[1] A.L. Cavalieri et al., Nature **449,** 1029 (2007).
[2] M. Ossiander et al., Nature Phys. published online (2016). doi:10.1038/nphys3941.

Energy Dependent Photoemission Delays from the (0001) Surface of Magnesium

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Soft X-Ray photoemission is a powerful tool for the investigation of electronic band structure of solids. Because of the small inelastic mean free path of the released electrons within the solid sample, it is limited to few atomic layers and sub-femtosecond timescales. The development of the attosecond streaking technique [1] provides experimentalists with a powerful new tool to measure the temporal dynamics of the photoelectric effect on attosecond time scales [2]. In recent literature, we find a contradiction between streaking and RABBIT experiments that respectively suggest ballistic electron transport [3] and final-state lifetime [4] to be the main contribution to observed photoemission delays.

In this study, we determine the relative photoemission time delay and chirp between the 2p-core state and valence-/conduction-band states close to the Γ-point of the magnesium (0001) surface at three photon energies between 112 and 135 eV. We find a distinct relation between the band-structure related photoemission resonance at 135 eV (c.f. Fig 1.a) and the temporal structure (c.f. Fig. 1.c) of the outgoing photoelectron wave packets even at final-state energies exceeding 100 eV.

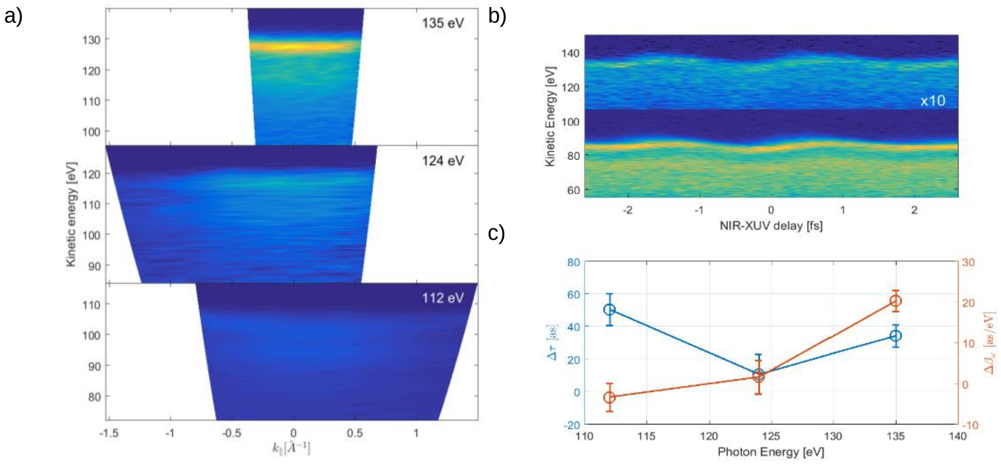

Fig. 1 a) Soft X-Ray ARPES of the Mg(0001) cb/vb measured with attosecond pulses. Spectra normalized to the respective 2p photoemission intensity. b) Attosecond streaking spectrogram of Mg(0001) at photon energy 135 eV c) Energy dependent relative photoemission delay(blue) and chirp(red) of 2p level w.r.t. cb/vb

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Attosecond Dynamics in Photoemission from Noble Metal Surfaces

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High-harmonic generation (HHG) has paved the way to time-resolved experiments with XUV probe pulses using table-top systems. Such XUV pulses are intrinsically ultra-short on the order of a few 100 attoseconds and have enabled the study of photoemission dynamics in atoms [1] and solid surfaces [2] in the time domain. Pioneering attosecond streaking [3] experiments revealed that electrons emitted from different initial states are released into vacuum at different times. We extended an interferometric two-photon technique named RABBITT [4] based on attosecond XUV pump pulses and a phase-matched infrared (IR) probe field from gas phase to condensed matter [5]. Experiments on Ag(111) and Au(111) surfaces revealed a strong energy dependence of the delays for photoemission from the valence *d*-band [6]. The origin of photoemission delays in solid surfaces has been subject of numerous theoretical investigations and effects such as initial state localization, resonances and electron transport were invoked. Our experiments were sensitive to final state effects and will be discussed in terms of final state band structure and electron propagation. Interpretation of the observed delays requires precise knowledge of the probe field distribution at the surface. The RABBITT technique was used in another experiment to sample the IR field distribution on a Cu(111) surface [7].

In a different, more conventional IR pump – XUV probe setup, we took advantage of the ultra-short nature of the photoemission process to investigate structural dynamics by means of photoelectron diffraction. Studying the displacive excitation of coherent phonons in Bi(111) we found a significant phase lag between the modulation of the electronic structure and the phonon, which could be attributed to the early evolution of the hot electron distribution [8]. Finally, I will show how the streaking method can be used to study the effective field of low-frequency THz pulses at metal surfaces.

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POSTER CONTRIBUTIONS

1 Femtosecond and Picosecond Spin Dynamics Separated by Spin and Time Resolved Photoelectron Spectroscopy

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The laser-driven ultrafast loss of the magnetization in a 3d ferromagnet has been observed two decades ago [1], yet is still an unresolved problem of solid state physics. The main questions surround the transfer of angular momentum away from the spin system. One of the most successful models describes the spin flips by the Elliot-Yafet scattering process, where phonons enhance the spin flip probability [2].

In a magneto-optical Kerr experiment, we implement a pump-pump-probe scheme in order to study the effect of excitations on the demagnetization. The first pump pulse excites the electron gas, which then transfers energy to the lattice and the spin system. We study the effect of the first pump pulse on the demagnetization caused by the second pump pulse. We find an ultrafast loss of the magnetization followed by picosecond spin dynamics commonly referred to as the “type II” behavior [2]. The femtosecond part is unaffected by previously created excitations, whereas the picosecond part is strongly affected by the first pump pulse [3].

We developed a spin and time resolved photoemission experiment based on a novel imaging SPLEED spin filter [4] and a higher harmonics source. By time, spin and energy resolved photoemission, we separate the ultrafast demagnetization from the slow, yet highly non-linear demagnetization effect observed in Kerr experiments [5].

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[5] R. Gort, et al., to be published.

2 Understanding the DEA Transfer Mechanism in $C_6H_5X/D_2O/Cu(111)$ System with 2PPE

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Despite the widespread interest in dissociative electron attachment on ice surfaces, its charge transfer dynamics and electron attachment mechanisms are not well understood. In this work, we study excess electron driven reactions via electron attachment as a function of the reactant's electronegativity and probe C_6H_5X /D_2O/Cu(111) with two-photon photoemission spectroscopy. An exponential increase in workfunction ($\Delta\Phi$) with fast (λ_{fast}) and slow (λ_{slow}) rates with photoexposure time is observed (Fig. 1 Right). This is shown to be due to the excess electrons in water which ionize or dissociate C_6H_5X to $C_6H_5X^-$ or $C_6H_5^{\cdot}$ + X^-. λ_{slow} shows inverse dependence on the halogen's electronegativity (and C-X bond strength). It is thus assigned to the ionization of C_6H_5X by electron attachment of excess electrons. Theory calculations of the ionizing π* states for C_6H_5X also corroborates with our finding. Additionally, theory shows that defect sites and vacancies provide favourable trapping centres for both the excess electrons and the adsorbed C_6H_5X. λ_{fast} can be assigned to electron attachment at the most favourable trapping sites. Fluence dependent measurements were also performed which showed rich dynamics for C_6H_5Br and C_6H_5Cl. Through comparative analysis and with the help of theoretical calculation, we were able to establish the role of defects and vacancies in ice in enhancing electron attachment processes. C_6H_5X /D_2O/Cu(111) proved to be an excellent system for studying such charge transfer dynamics.

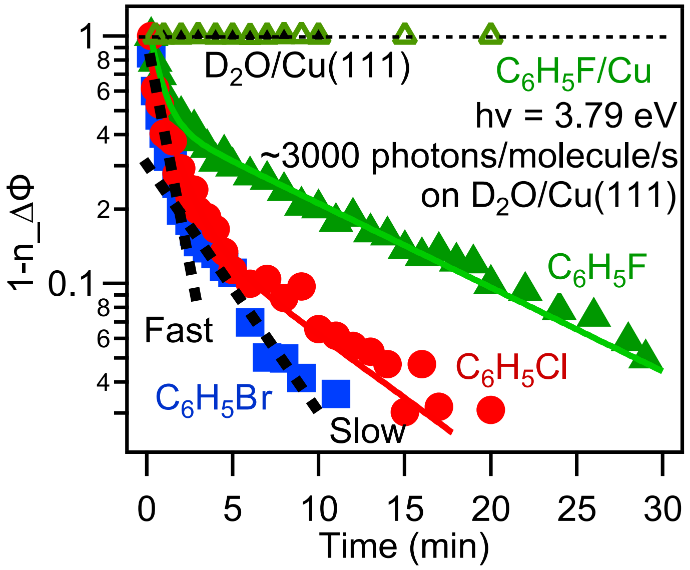

Fig. 1 Exponential change in workfunction change as a function of photoexposure time. Two rates are observed, and they are slowest for C_6H_5F and fastest for C_6H_5Br.

3 Momentum- and Thickness-dependent Lifetimes of Electrons in Ultrathin CoO and NiO Films

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Ultrathin antiferromagnetic cobalt oxide (CoO) and nickel oxide (NiO) films of 2 to 10 monolayers (ML) on Ag(001) are investigated by time- and angle-resolved two-photon photoelectron (2PPE) spectroscopy.

In the 2PPE spectra on 2 ML CoO films, unoccupied states at 3.6 to 4.0 eV above the Fermi level (E_F) are observed with pump-probe photon energies of 4.2 and 1.8 eV. These unoccupied states have a large effective mass up to 3 electron masses (m_e) and a momentum-dependent lifetime of around 9 to 23 fs. In addition, we resolve the lifetime at the conduction band minimum of CoO, which was first discovered by S. Otto *et al.* in their static 2PPE experiments [1]. These results together with the thickness dependent 2PPE spectra are discussed in view of the theoretical calculations by A. Schrön *et al.*, in which surface states of CoO have been predicted [2].

On 2 to 9 ML of NiO films, we observe at 3.7 to 4.2 eV above E_F image potential resonances. These resonances have a strongly thickness-dependent effective mass of 0.9 to 3 m_e, and a lifetime of less than 120 fs [3]. Moreover, we estimate an upper bound for the lifetime of the conduction band minimum at 2.5 eV above E_F as 10 fs.

Our results show that at both NiO and CoO surfaces the lifetime of unoccupied electronic states is much shorter than in conventional semiconductors, which is in the range of several picoseconds [4]. Our observation implies a significant influence to ultrafast electron dynamics due to electron correlation in transition metal oxides.

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4 Ultrafast CO Oxidation and Desorption Dynamics on Pd Nanoparticles

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While femtosecond pulse-driven heterogeneous surface chemical dynamics, such as photodesorption from the single crystal surface, have been studied and modeled successfully for several systems, nanoparticle (NP) surfaces, which offer new and unexpected reaction paths and outcomes, are not well understood, so far. Femtosecond two-pulse correlation spectroscopy, temperature program desorption spectroscopy, and density functional theory calculations are used to elucidate the mechanisms and the ultrafast dynamics of carbon monoxide (CO) oxidation and desorption reaction on rutile $TiO_2(110)$-supported Pd NP surfaces. Two different time-scales for the CO oxidation reaction on Pd NPs are observed: a fast channel (time constant ~ 7 picosecond) is found for the oxidation reaction when strongly bound CO is involved and a slow channel (time constant ~ 15 picosecond) is found for the oxidation reaction when weakly bound CO is involved. Temperature programmed desorption spectroscopy confirms that the fast and the slow channels are associated with CO oxidations mostly at (111) facets and at edges of the Pd NPs, respectively. Furthermore, we find that phonon mediated CO activation determines the ultrafast dynamics of CO oxidation reaction on Pd Nanoparticles. Along with CO oxidation measurements, we have also performed two-pulse correlation measurements of femtosecond pulse-driven desorption of carbon monoxide (CO) from rutile $TiO_2(110)$-supported Pd NP surfaces. One of the important findings includes that the CO desorption at Pd NP surfaces is purely phonon-mediated; while, the same at extended single crystal Pd(111) surface is electron-mediated. Furthermore, numerical simulation of the experimental results based on two temperature model coupled with phonon-mediated coupling to adsorbates directly indicates that electron-phonon coupling constant of Pd NPs is different from bulk value. We also find that initial temperature (in the 98 – 375 K temperature range) of Pd NPs controls the CO desorption yield and nature of the CO desorption dynamics.

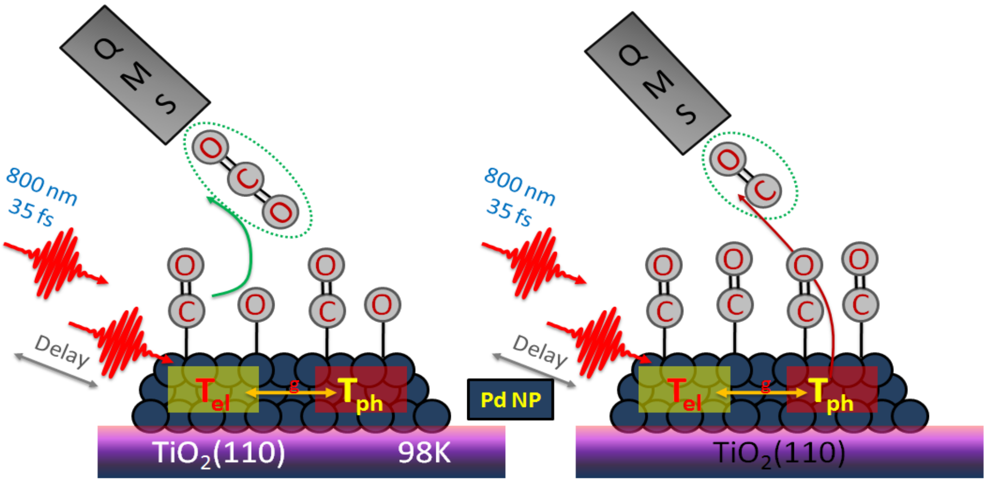

Fig. 1 Ultrafast dynamics of CO Oxidation and Desorption reactions on PdNP

5 Hot Carrier Dynamics in Semiconducting Transition-Metal Dichalcogenides - Valley-Polarized Excitation and Momentum-Dependent Relaxation

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Semiconducting transition-metal dichalcogenides (TMDC) such as WS_2 and MoS_2 have recently attracted considerable interest due to a transition from an indirect to a direct band gap semiconductor when thinned down to a single layer [1]. As these peculiarities are potentially useful for applications in electronics, spintronics, and photonics a basic knowledge of the relevant interaction channels governing carrier excitation and relaxation in these materials is of fundamental interest. We used time- and angle-resolved photoelectron spectroscopy (trARPES) to study different aspects of ultrafast carrier dynamics in the semiconducting TMDC $2H$-MoS_2. Momentum-dependent population and relaxation processes within the conduction-band energy landscape are monitored following photoexcitation with laser pulses in the UV-spectral regime. Our results provide a very direct view onto intervalley- and intravalley scattering processes and gives evidence that defect states play a key role in the final depopulation of the conduction band [2]. Furthermore, we are able to selectively address K and K' valleys in the trARPES experiment using circularly polarized 590 nm laser pulses owing to the lack of inversion symmetry in the top layer of $2H$-MoS_2 [3]. Similar to recent trARPES results reported for $2H$-WSe_2 [4], dichroism is observed in the excited state population as well as dynamics.

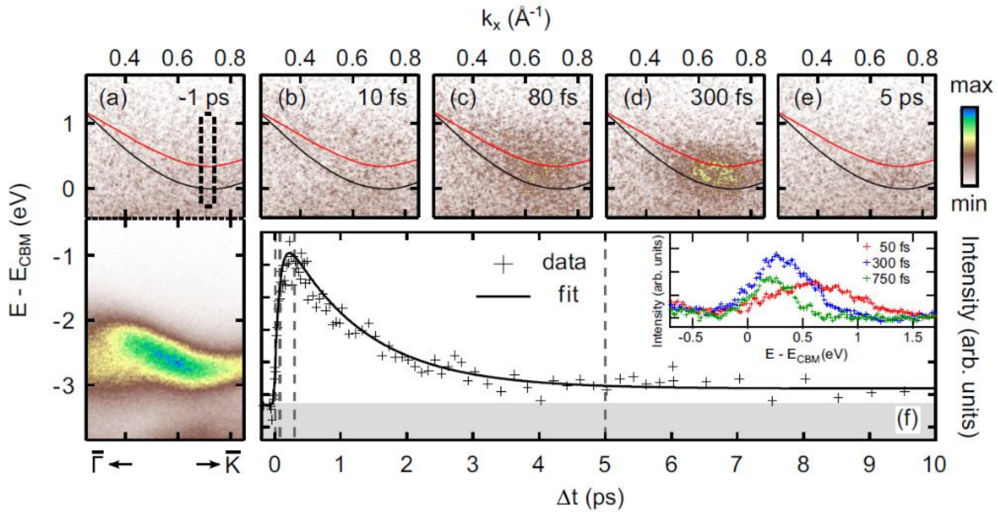

Fig. 1 TrARPES data on carrier relaxation in MoS2 probed along the ΓK-direction; (a) – (e) ARPES snapshots recorded at different time delays after photoexcitation; (f) photoemission intensity transient at 0.53 ΓK.

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6 Ultrafast Energy Relaxation in Thin Au and Mo Films

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The energy relaxation after short pulse laser heating in thin Au (27 nm) and Mo (10 nm) films on a glass substrate is investigated by ultrafast optical transient transmittance spectroscopy. A broadband probe range from 300 to 900 nm, a time resolution better than 50 fs, pump-probe delays up to 1 ns and a variable pump fluence between 40 and 2800 μJ/cm^2 is combined to study the nonequilibrium charge carrier dynamics. The pump pulse is centered at 475 nm (2.61 eV) in order to excite interband transitions in both materials. Despite intensive experimental and theoretical studies on the nonequilibrium charge carrier dynamics in thin gold films, a detailed experimental investigation of the pump fluence dependence with interband pumping is still missing to the best of the authors' knowledge.

In Fig. 1b) the time evolution of the pump induced transmittance change is shown. Red corresponds to increased transmission and blue to increased absorption. Besides the typical decay of the overall signal due to the cooling processes, an ultrafast spectral reshaping is observed which is connected to the evolution of the nonthermal and thermalized electron population. A first analysis immediately indicates that the description of the initial energetic relaxation (< 200 fs), and thus the spectral signatures, via the extended two-temperature model is insufficient after interband pumping. For the analysis a spectral fit model with empirical peak functions is employed as seen in Fig. 1a). The analysis of the spectral width of the functions allows us to follow the population and temperature changes in time (Fig. 1c)). This empirical approach is generally applicable, even for films and samples with unknown or unpredictable spectral changes.

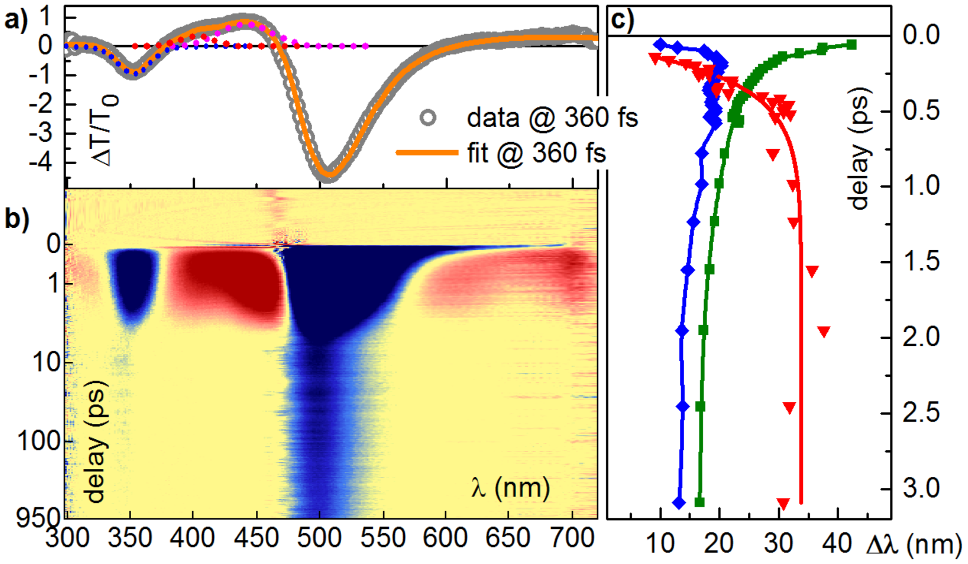

Fig. 1: a) Data and empirical fit with contributions at 360 fs pump-probe delay. b) Time evolution of the transmittance change after excitation with a 475 nm pump pulse and a pump fluence of 72 J/cm2. c) Time evolution of the spectral linewidth of the chosen peak functions up to 3 ps pump-probe delay. The solid lines serve as a guide to the eye.

7 Exciton Relaxation and Migration Dynamics in α-Sexithiophene/Au(111)

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Optical excitation of an organic semiconductor leads to exciton generation. The corresponding absorption bands are usually strongly broadened due to coupling to vibrational modes. In organic electronics, the dynamics of such excitonic states with respect to their energetic evolution as well as their migration through the material are decisive for device performance. We investigated the exciton dynamics in the model system of sexithiophene (6T) on a Au(111) surface by time-resolved two-photon photoemission (2PPE) spectroscopy. In crystalline 6T films, vibrational excess energy is dissipated on a time scale of <100 fs resulting in an excitonic state which is stable in energy for >100 ps. In amorphous 6T films, however, the energetic relaxation from a broad initial distribution of excited states is strongly slowed down and reaches an excitonic state with a binding energy comparable to that in the crystalline film on a time scale >10 ps. This slow process is attributed to the migration of excitons towards sites with higher binding energy. Thus the morphology of the organic layer strongly influences the excited state dynamics and energetics relevant for organic electronics.

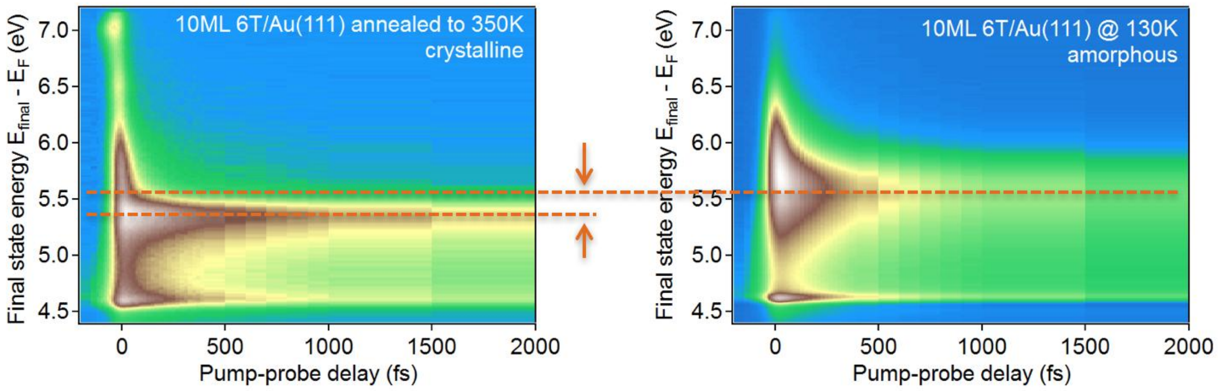

Fig. 1 Two-photon photoemissionm intensity as a function of final state energy versus pump-probe delay for a crystalline (left) and an amorphous (right) 6T film. Pump and probe photon energies are 3.14 and 4.25 eV, respectively.

8 Femtosecond Spin Resolved Photoelectron Spectroscopy

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The study of femtosecond spin dynamics using photoemission is challenging, as space charge effects limit the maximum number of electrons, which can be emitted per pulse [1]. Therefore, one typically uses high repetition rate laser systems, which works well for two-photon photoemission. The use of conventional single channel spin detectors in combination with higher photon energies from free electron lasers [1] as well as higher harmonic sources [2] is difficult. For example, the combination of spin and energy resolution with femtosecond time resolution was not feasible using a Mott detector [3].

Here, we present experiments with a novel imaging SPLEED based spin filter [4] in combination with a hemispherical energy analyzer. This way, one can detect a range of kinetic energies in parallel, while analyzing their spin polarization. This concept leads to a large improvement of the detection efficiency compared to a Mott spin polarimeter. For emitting the electrons, we designed a higher harmonic source, which is optimized for stability and emits radiation at 21eV. This way, we can detect spin dynamics at different energies of the valence band as well as the cascade [5,6].

We will present first experimental results from this setup and discuss future experimental opportunities using novel x-ray sources and spectrometer concepts.

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9 Fingerprint of Bulk Half-metallicity in the Surface Electron Spin Dynamics: Application to Fe_3O_4

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Half-metallic materials are very promising candidates for spintronic devices with a potential for generating fully spin polarized currents or high spin filtering effects. Nevertheless, the characterization of their bulk properties by photoemission remains very challenging due to surface reconstruction [1]. Here we propose a novel approach to show evidence of bulk half-metallicity in a material by monitoring the electron spin dynamics at the Fermi level after photoexcitation.

For the theoretical interpretation, the Boltzmann equation is numerically solved for the half-metallic bulk density of states of Fe_3O_4. On a short time scale, the electron thermalization leads to the same electronic temperature in both spin channels but with different chemical potentials. At the bottom of the majority spin conduction band, due to the presence of the gap, electrons have a long relaxation time and can slowly diffuse toward the metallic surface reducing the Fermi level spin polarization.

Experimentally we performed time- and spin- resolved photoemission measurements in the vicinity of the Fermi level using 4.65 eV photon energy. The static spin-resolved electron distribution curves are very well reproduced by fully relativistic photoemission calculations. In the time-resolved measurements, the Fermi level spin polarization is reduced over the first picosecond delay. We attribute this effect to the bulk – surface dynamics [2] as previously observed in different systems. Highly spin-polarized electrons from the bulk majority bands are injected in the metallic surface reducing the spin polarization observed in static experiments. This evolution of the spin polarization is qualitatively predicted by our bulk and surface thermalization calculations and is a direct consequence of the presence of the bulk bandgap in majority spin channel. So, the combination of time- and spin- resolved photoemission is a powerful approach to test the half-metallicity in solids.

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[2] Cacho et al., Phys. Rev. Lett. **114**, 097401, (2015).

10 Femtosecond Electron Dynamics in 1D Indium Wires

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The (4x1) reconstruction of In/Si(111) is a widely studied quasi one-dimensional system that undergoes a complex metal-to-insulator transition at T_{CDW}=110K. The mechanism that triggers this transition is a combination of a Peierls instability and a shear distortion between two neighboring Indium chains.

We study the photo-induced electron dynamics of In/Si(111)-(4x1) both in the metallic and the insulating phase with time- and angle-resolved photoemission spectroscopy at extreme ultraviolet wavelengths. In Fig. 1a) and b) we show the equilibrium band structure and the pump-induced changes of the photocurrent after excitation with femtosecond pulses at $\hbar\omega_{pump}$=1.5eV, respectively. Integration of the photocurrent over a region close to the Fermi level (black box in Fig. 1b) yields the pump-probe traces for temperatures above (panel c) and below T_{CDW} (panel d). On top of an exponentially decaying pump-probe signal that we attribute to the relaxation of photo-excited carriers, we observe oscillations at 1.8 THz (0.4 THz) for $T>T_{CDW}$ ($T<T_{CDW}$). We assign these modes to coherent phonons of the Indium chains and the amplitude mode of the charge density wave, respectively.

We will discuss the mechanism of charge density wave melting and recovery as well as the emerging coherent dynamics.

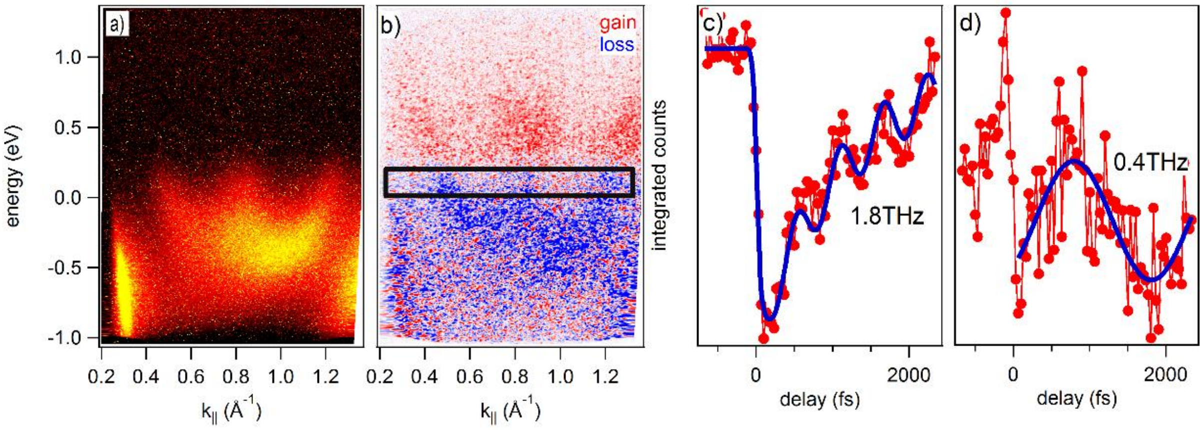

Fig. 1 (a) ARPES snapshot of In/Si(111)-(4x1) at negative time delays, (b) pump induced changes of the photocurrent, (c) pump-probe trace for the metallic phase and (d) for the insulating phase.

11 Nonadiabatic Molecular Dynamics Investigation on Charge Transfer Dynamics at CO_2/TiO_2 Interface

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CO_2 photo-reduction is one of the most important photocatalytic reactions on TiO_2 surface. Therefore, it is very important to understand the photo-generated charge carrier dynamics at CO_2/TiO_2 interfaces. Using nonadiabatic molecular dynamics combined with constrained DFT, we investigate the electron transfer dynamics at CO_2/TiO_2 interface. Our results show that bending vibration mode of CO_2 can be easily excited by electron injection to CO_2. If the life time of the excited electron on CO_2 is longer than 15 fs, the CO_2 dissociation is able to happen afterwards. Along with the bending vibration and dissociation of CO_2, the excited electron will first transfer from CO_2 to TiO_2 within 5 fs mediated by the adiabatic charge transfer mechanism. After that, it will transfer back to CO_2 molecule within 50 fs since the CO_2 LUMO is stabilized by the bending vibration significantly. Our results show that the excitation of bending vibration of CO_2 by electron injection is the key factor for CO_2 photo-reduction on TiO_2.

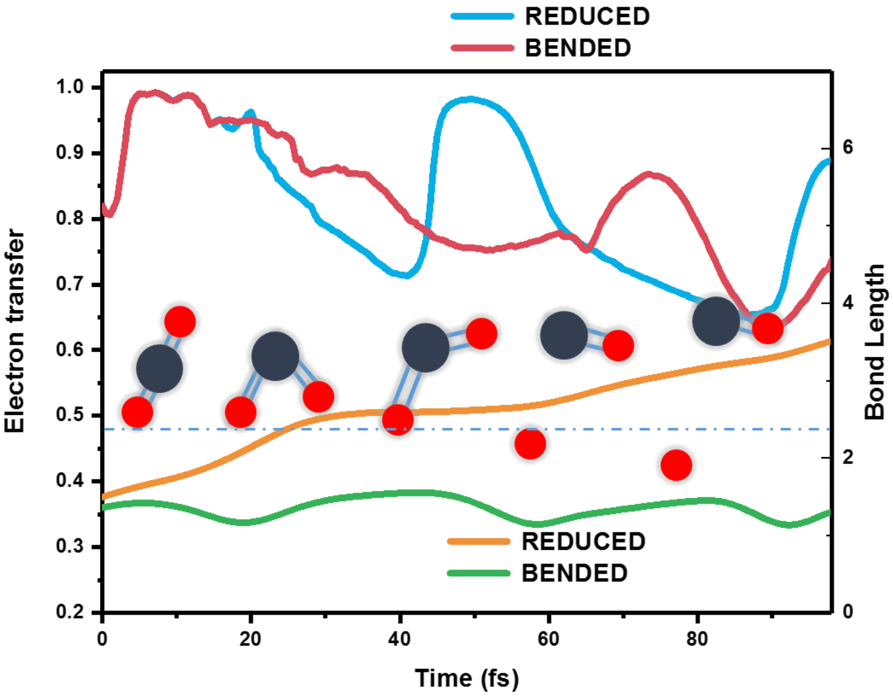

Fig. 1 Schematic diagram of hot electron dynamics and proton transfer process

12 Towards Probing THz-Driven Transient Electronic States with Time-Resolved ARPES

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In this contribution, we present the concept for a novel experimental setup at the THz facility TELBE at Helmholtz-Zentrum Dresden-Rossendorf [1], which combines THz excitation with intrinsically surface-sensitive time- and angle-resolved photoelectron spectroscopy (tr-ARPES). THz excitation provides resonant access to a multitude of fundamental modes, e.g., lattice vibrations, molecular rotations, spin precession and the motion of free electrons. Thereby it can be a handle to control highly relevant transient material properties, from metal-insulator-transitions [2] to superconductivity [3] and catalytic activity [4]. The obvious way of probing these THz-driven dynamics is tr-ARPES which enables direct access to the surface electronic states on a femtosecond timescale, thereby complementing current purely optical techniques. However, the implementation of THz pump – tr-ARPES probe experiments (cf. Fig. 1) has been impeded, because of the unmet requirement for high duty cycle THz sources (>> 10 kHz repetition rate quasi-cw) to provide sufficient statistics for tr-ARPES. This limitation is overcome at TELBE which offers tunable and CEP-stable THz pulses at a repetition rate of 100 kHz based on superradiant emission, and timing stability of < 30 fs due to a novel pulse-to-pulse diagnostics scheme [5]. Over the next three years, a tr-ARPES setup shall hence be implemented at the THz facility TELBE which aims at establishing feasibility and dynamic range despite obvious obstacles such as residual streaking of the photoelectrons by the high THz excitation fields. This contribution discusses the challenges and the opportunities of tr-ARPES in first THz control experiments and will outline the current design of the planned tr-ARPES endstation.

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13 Self-amplified Photo-induced Gap quenching in a Correlated Electron Material

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The formation of ordered phases in correlated electron materials is determined by a delicate balance between the interactions of electrons with other degrees of freedom such as the lattice or spin system. While these interactions are difficult to disentangle in thermal equilibrium, they show characteristic timescales during the relaxation process after strong optical excitation with intense fs-light pulses.[1] Here, we use femtosecond extreme-ultraviolet time- and angle-resolved photoelectron spectroscopy[2] to study the ultrafast photo-induced suppression of the charge-density wave (CDW) in 1*T*-TiSe$_2$. Monitoring the non-equilibrium electron dynamics occurring at femtosecond laser excitation, we identify hot-carrier multiplication as the primary driving force for the ultrafast CDW suppression. As soon as the optically excited carriers have relaxed to a quasi-equilibrium hot Fermi-distribution electron gas, the CDW suppression stops. Theoretical calculations of the hot carrier scattering processes and the screening properties further link the carrier multiplication to the observed CDW gap dynamics.[3]

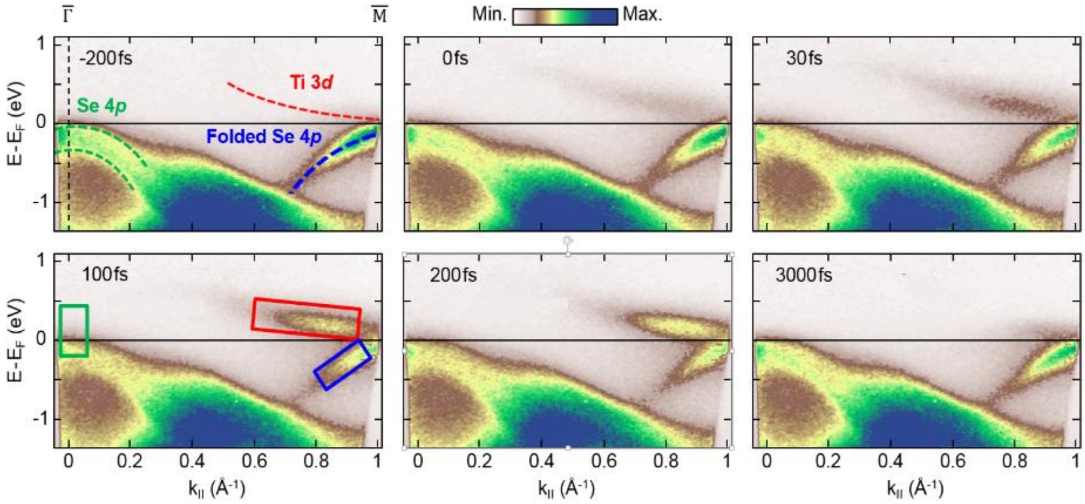

Fig. 1: Photoemission maps of the electronic response of the backfolded Se 4p states (blue area), and the hot-electron dynamics in the Ti 3d band (red area) at different times. The absorbed fluence of the 1.6 eV, 32 fs, p-polarized pump pulses was 0.47 mJ cm^{-2}. The trAPRES data were recorded with fs- XUV radiation (22 eV, p-polarization) obtained by higher harmonic generation.

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14 Band Structure Effect in Attosecond Photoemission Delays from Cu(111) Surface

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The study of photoemission delays using attosecond spectroscopy triggered many questions regarding electron propagation on the attosecond timescale and atomic length-scale [1]. One particular debate is related to the influence of the band structure on atomic scale propagation. Here we provide an answer and show that the band structure can still affect the electron transport over few atomic layers and within hundreds of attoseconds. We measured two-photon two-color absolute photoemission delays [2,3] of electrons excited from two initial bands within the 3*d*-valence band manifold of a copper sample, using the RABBITT attosecond metrology technique. We compared our experimental results with propagation times calculated using group velocities derived from the electronic band structure of copper, and propagation times using the free electron velocities. We observe that the electronic band structure information is able to reproduce the photoemission delays that the free-electron model fails to recover. This implies that the band structure can still affect electron transport within ångströms of propagation distance and on the attosecond timescale. We define an upper bound of 320 ± 40 as for an electron to be able to assume its effective mass.

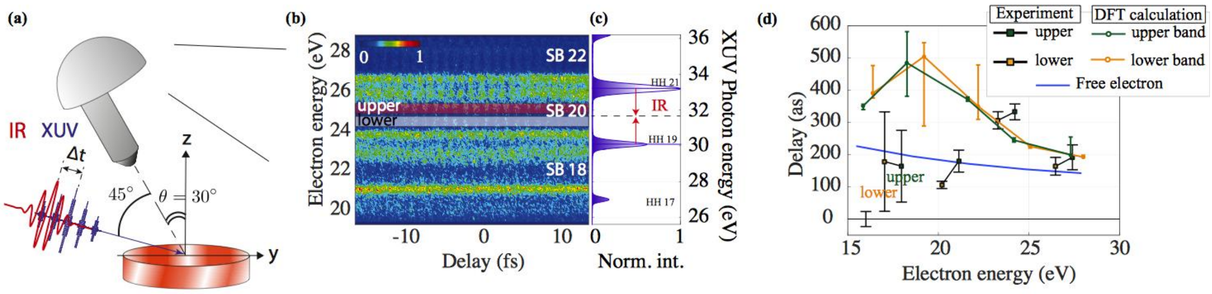

Fig. 1 (a) Schematic of the experiment. (b) RABBITT trace acquired from the copper sample showing a split signal in the sideband due to the split valence bands investigated. (c) XUV spectrum used for the experiment and schematic of the two-color two-photon interference leading to the sideband signal. (d) Absolute photoemission delays (black) extracted from the RABBITT traces, for the different parts of the split band: upper and lower, as a function of electron energy. Calculated propagation times for free-electron final state (in blue) and for Bloch state electrons excited from the upper (in green) and lower part (in yellow) of the valence band.

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15 Impact of Streaking Field Penetration, EUV Pulse Shirp, and Initial State Properties on Attosecond Time-resolved Photoemission from Solids

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The availability of single attosecond extreme ultra-violate (EUV) pulses paved the way to observe electron dynamics on their natural time scale. Ever since attosecond resolution in time-resolved photoemission experiments was first demonstrated [1] the impact of various effects and experimental parameters on the observed relative photoemission delays is not yet fully understood.

Here, attosecond time-resolved photoemission from solid surfaces is simulated based on solving the one-dimensional time-dependent Schrödinger equation (1D-TDSE) [2]. The so obtained photoelectron wave-packet evolution dynamics in a single-particle picture allows studying the impact of various parameters on the photoemission delays. From simulated streaking spectrograms photoemission time delays due to electron propagation in the one-dimensional potential landscape are extracted. The model calculation allows varying critical parameters such as the initial state wave function, the photoelectron inelastic mean free path, the electron-hole interaction, the penetration of the IR-field into the solid, and the chirp of the EUV pulse. By variation of these parameters, their impact on photoemission delays is systematically investigated and related to their corresponding relevance in actual attosecond streaking experiments.

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16 Turnkey High Repetition Rate XUV Source Driven by Yb-lasers

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Coherent extreme ultra violet (XUV) radiation generated by high harmonic generation (HHG) is a powerful tool to study ultrafast dynamics in atomic and molecular systems with a temporal resolution ranging from the femtosecond down to the attosecond scale. Ti:sapphire lasers are the most common lasers used for HHG, and typically deliver 30 fs pulses at a central wavelength $\lambda = 800$ nm, with pulse energies up to hundreds of mJ. The central wavelength can be converted further into the mid-infrared, by using optical parametric amplification (OPA). Due to the cutoff law of HHG, scaling as λ^2 for a given laser intensity, it allows extending the XUV spectral range into the soft-X rays. However, the average power of these laser systems cannot easily be scaled beyond 10 W, preventing HHG at repetition rates higher than 10 kHz. Increasing the repetition rate of HHG drivers comes with a number of advantages, such as increased photon flux [1,2], reduction of the acquisition time in coincidence experiments to study molecular dynamics such as COLTRIMS, and the possibility to study the electronic structure of matter via photoemission spectroscopy and microscopy, where low doses are needed to avoid space-charge effects [3]. The recent development of high power femtosecond lasers based on Yb, together with adaptation of the HHG process following scaling rules to allow phase matching in tight focusing configuration [1], has allowed HHG at high repetition rates and average powers [2].

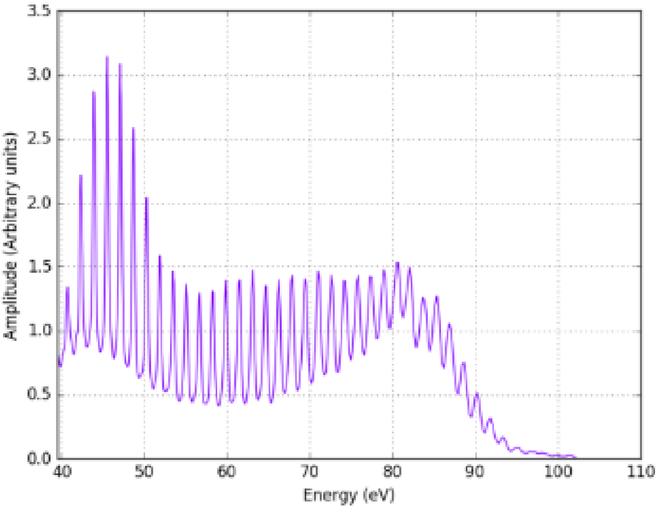

Fig. 1 XUV spectrum generated in a thin argon gas jet by an OPCPA laser at $\lambda = 1.55$ µm and a repetition rate of 125 kHz.

We drove HHG with high repetition rate few-cycle optical parametric chirped pulse amplifier at 1.55 µm described in [4]. The OPCPA laser is driven by a Tangerine, Amplitude Systems pump laser. The OPCPA source delivers 22 µJ 50 fs pulses at 125 kHz repetition rate. An XUV beamline has been optimized for this laser system, based on a tight focusing configuration with high atomic density at the gas jet target [1]. The laser pulses are focused onto an argon gas jet. As shown in fig. 1, the use of a long driving laser wavelength extends the spectral cut-off to 95 eV (Fig. 1). The XUV radiation generated is highly reproducible on day to day basis. This photon energy is suitable for several applications, for example attosecond time-resolved spectroscopy at surfaces [7], time resolved ARPES using a high harmonic generation source [8], atomic inner shell spectroscopy [5] or attosecond nanoscale near-field sampling [6]. We will discuss the experimental results such as XUV spectrum shape and cutoff and photon flux dependence on the experimental parameters, e.g. atomic gas jet density and focus position with respect to the gas jet.

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17 HELIOS – A Facility for Time and Angle Resolved Surface Studies

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For the investigation of the surface electron dynamics and electronic structures of crystalline solids, the most direct technique is that of angle resolved photoemission spectroscopy (ARPES). In this contribution, we present a newly installed set-up for both angle- and time-resolved photoemission spectroscopy (TR-PES). The High Energy Light Induced Overtone Source (HELIOS) is an in house source of Extreme Ultra Violet (XUV) radiation of energies between 15-70 eV. The source consists of a commercial Ti:Sapphire laser which operates at 10 kHz and pulse energy of 1 mJ. The output is split, where the majority (~800 µJ) produces high harmonics. These harmonics pass through a home built monochromator to produce an energy resolution of 100 meV. The remaining 200 µJ serves as pump whose energy can be changed by an Optical Parametric Amplifier which can tune the pump wavelength from 240 nm-20 µm. The pump probe cross correlation has been measured to be ~40 fs.

The source has two beam lines, one dedicated for the study of magnetic dynamics using Transverse Mangeto-Optical Kerr Effect (TMOKE) and the other has an Angle Resolved Time of Flight (ARTOF) spectrometer which is measures the time of flight of the emitted electron, thereby it's kinetic energy, and the angular distribution of photoelectrons which aids in determining k-space information. Thus, by combining the pump probe set up along with the ARTOF spectrometer, we can study time and angle resolved electron dynamics of the surface. The performance of the ARTOF station has been characterized by phase experiments, like side band measurement in Helium and the next scheduled experiments will include surface studies of solid samples. One prominent example is the study of electronic structure dynamics of photocatalytic materials, like tungsten trioxide (WO_3) and hematite iron oxide (Fe_2O_3) thin films.

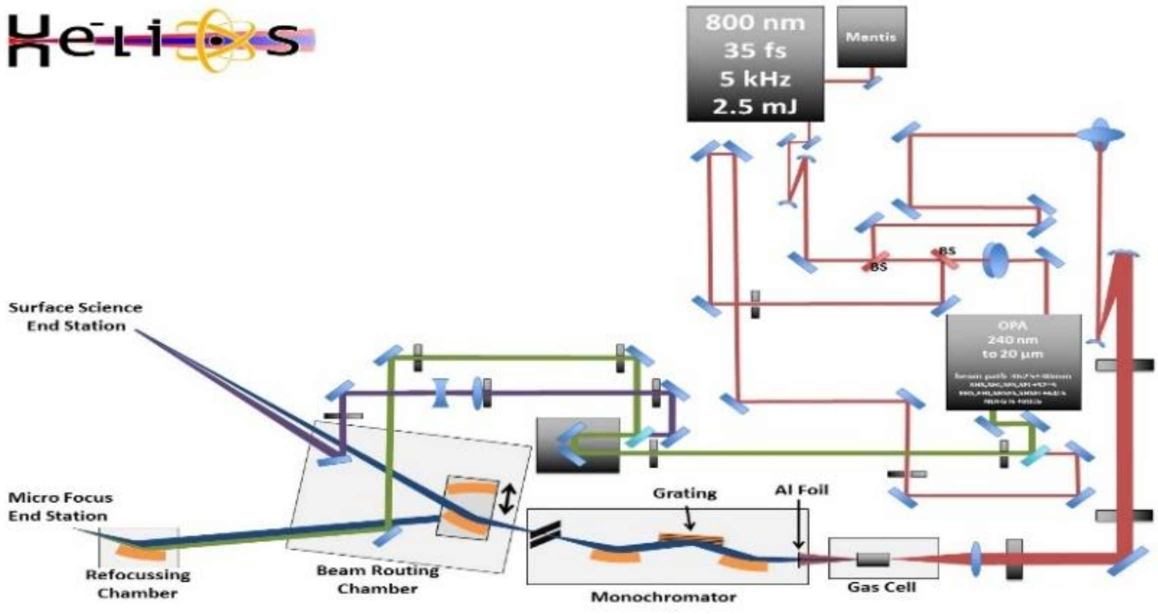

Fig. 1 Schematic drawing of beam path at HELIOS.

18 Attosecond XUV and VUV Photoelectron Wavepacket Broadening from Solid Surfaces

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Attosecond streaking on single crystal solids have previously been investigated, with relative photoemission time delays between core and valence levels measured. I will present results that show attosecond streaking’s more broad applicability to disordered solids, and measurements of photoelectron wavepacket broadening relative to gas phase streaking for extreme ultraviolet photon energies. Measured broadening was consistent with a ballistic electron transport model. I will also present an overview of ongoing experimental work on polycrystalline gold surfaces to investigate wavepacket broadening at lower photoelectron energies, where significantly more broadening is anticipated due to electron transport with greater inelastic mean free paths.

19 Optical Control of Ultrafast Currents in the Topological Surface State of Sb_2Te_3

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We present energy-momentum mapping of the photocurrent in the Dirac cone of the topological insulator Sb_2Te_3 by means of time- and angle-resolved two-photon photoemission (2PPE) after optical excitation with ultrashort mid-infrared laser pulses. Recently, we have demonstrated that linear polarized mid-infrared pulses permit the generation of photocurrents with ps-lifetime in the initially unoccupied part of the Dirac cone of Sb_2Te_3 by a direct optical excitation [1]. Here, we show that the magnitude and direction of this current depend on the azimuthal orientation of the sample as well as on the polarization of the pump light [2]. For azimuthal orientations for which no photocurrent is generated by linear polarized pump light, we are instead able to generate a photocurrent by elliptical polarized light and fully control its direction and magnitude by varying the light helicity of the mid-infrared light by a $\lambda/4$-plate as shown in Fig. 1.

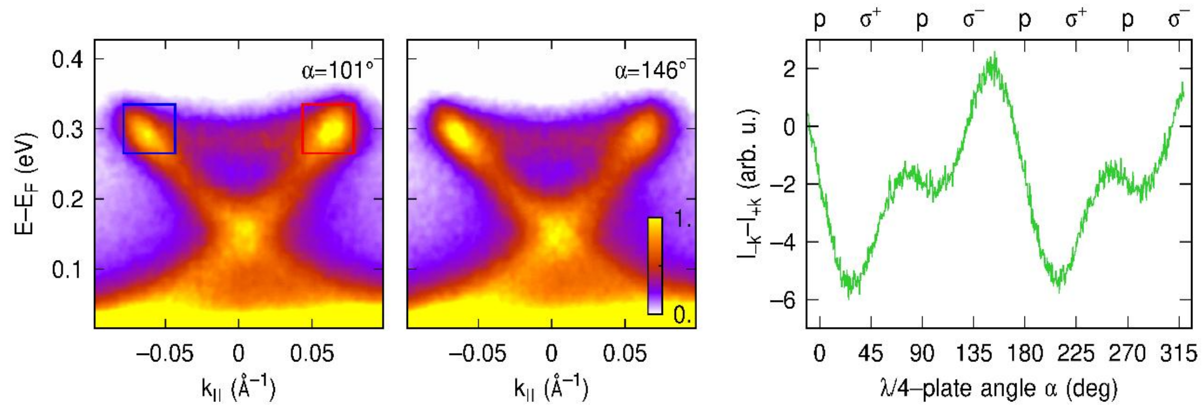

Fig. 1: Angle-resolved 2PPE spectra of Sb2Te3 for mid-IR pump (h□=0.33 eV) of different helicity. The intensity contrast between ±k|| at the energy of the direct optical excitation (E-EF=0.3 eV) can be controlled by the orientation of a1/4-plate.

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20 Studying the Type-II Weyl Semimetal *Td*-WTe$_2$ with Two-Photon Photoemission

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With their first experimental observation in 2015, topological Weyl semimetals have attracted enormous attention: In these materials, the long sought-after Weyl fermions are realized as quasi-particle excitations in condensed matter [1]. Weyl semimetals feature an unusual electronic structure with touching points in the bulk band structure – the so-called Weyl points – that are connected by unique topological surface Fermi arcs. The key technique to gain access to these signatures is angle-resolved photoelectron spectroscopy (ARPES) [2,3].

Here, we present a combined photoemission and two-photon photoemission (2PPE) study of the type-II Weyl semimetal *Td*-WTe$_2$. Using a widely tunable femtosecond laser system with photon energies between 1.3 eV and 5.0 eV, we are able to access a variety of both occupied and unoccupied electronic states. The static and time-resolved results are discussed under consideration of band structure calculations and current ARPES studies of *Td*-WTe$_2$.

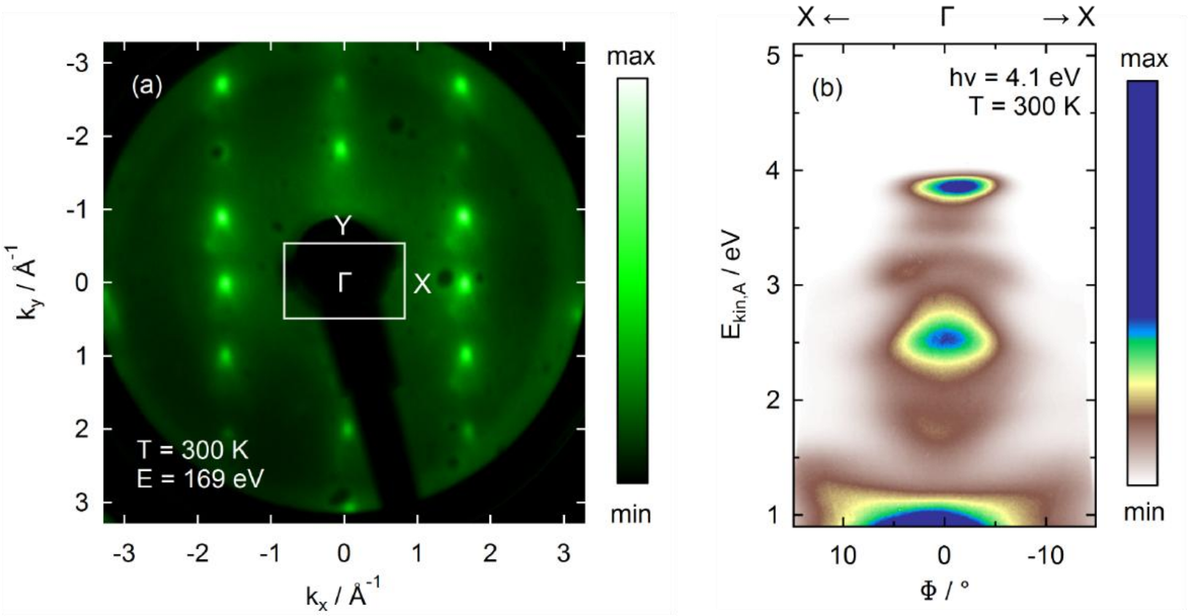

Fig. 1 (a) LEED image of Td-WTe2 demonstrating the sample quality and orientation of the Brillouin zone. (b) Example of a 2PPE intensity map along the high symmetry direction ΓX.

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21 Photo-induced Ultrafast Potential Shift Observed at a Metal-Semiconductor Interface of a Bi Thin Film

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A photo-induced potential shift at a surface and an interface is a central mechanism in optoelectronic applications such as solar cells and photodiodes. The simple picture of photo-induced electron-hole separations across a band-bending region has been widely accepted. The relaxation process in ns ~ µs time scales has also been studied with several time-resolved techniques and well interpreted considering thermal diffusion and tunneling processes of excited carriers [1]. However, the ultrafast (fs ~ ps) process, namely the very first stage in the formation and relaxation of a potential shift, has been investigated only recently [2]. Especially, studies at material interfaces are still quite limited. Here, we report a photo-induced ultrafast potential shift observed at a metal-semiconductor interface of a Bi thin film on a Ge substrate, by performing time- and angle-resolved photoemission spectroscopy (TrARPES), a unique direct probe for electronic dynamics.

The experiment was performed using a 1.5 eV pump pulse and a 5.9 eV probe pulse generated from a 250 kHz repetition Ti:Sapphire laser system. The total time and energy resolution were about 300 fs and 15 meV, respectively. Figure 1 (a) shows band structures of a Bi thin film on a Ge substrate observed by ARPES without pump excitation. Injection of a pump pulse caused both a dynamical variation of the band position and its occupation as shown in Fig. 1 (b)-(e). From a rigid-shift behavior of the peak positions, we can attribute the former to a photo-induced potential shift at the interface. We further discuss details of the temporal evolutions, the pump fluence dependence, and a possible origin of this ultafast potential dynamics.

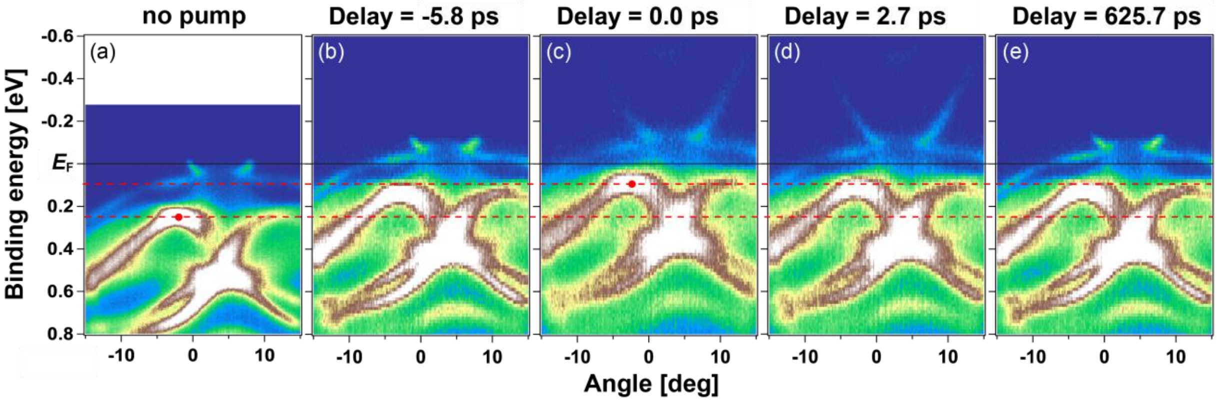

Figure 1 (a) The APRES spectra of a Bi thin film on a Ge substrate measured without pump excitation. (b)-(e) Evolution of the ARPES spectra after excitation according to the temporal delay between pump and probe pulses. The pump fluence was set to 0.1mJ/cm2. Red dashed lines are guides for highlighting the potential dynamics.

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22 Mahan Cone Formation in the Adsorbate System Tin-Phthalocyanine on Ag(111)

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The adsorption of tin-phthalocyanine (SnPc) on Ag(111) has attracted considerable interest due to peculiarities in the adsorbate-substrate interaction and switching capabilities reported for this model-type organic/inorganic system [1,2]. In this work Low Energy Electron Diffraction (LEED) and Photoemission/Two-Photon Photoemission (PES/2PPE) is used to study how structural and electronic properties of this model system are correlated in the sub-monolayer coverage regime. Photoemission data are recorded using a high-repetitive and tunable noncollinear optical parametric amplifier offering the possibility to address various resonant transitions of the electronic structure at the metal/organic interface. The study focusses on the incommensurate adsorption phase of SnPc which is formed at room temperature in the coverage regime between 0.90 ML and 1.0 ML. PES and 2PPE data of SnPc/Ag(111) (Fig. 1(a) and Fig. 1(b)) reveal the formation of Mahan Cones [3] associated with direct optical transitions within the Ag sp-band [4] which are strongly modified in the presence of the incommensurate adsorbate layer. Mahan cone simulations under consideration of the lattice structure of the SnPc overlayer along different high symmetry directions show a very good agreement with the experimental data (Fig. 1(c)).

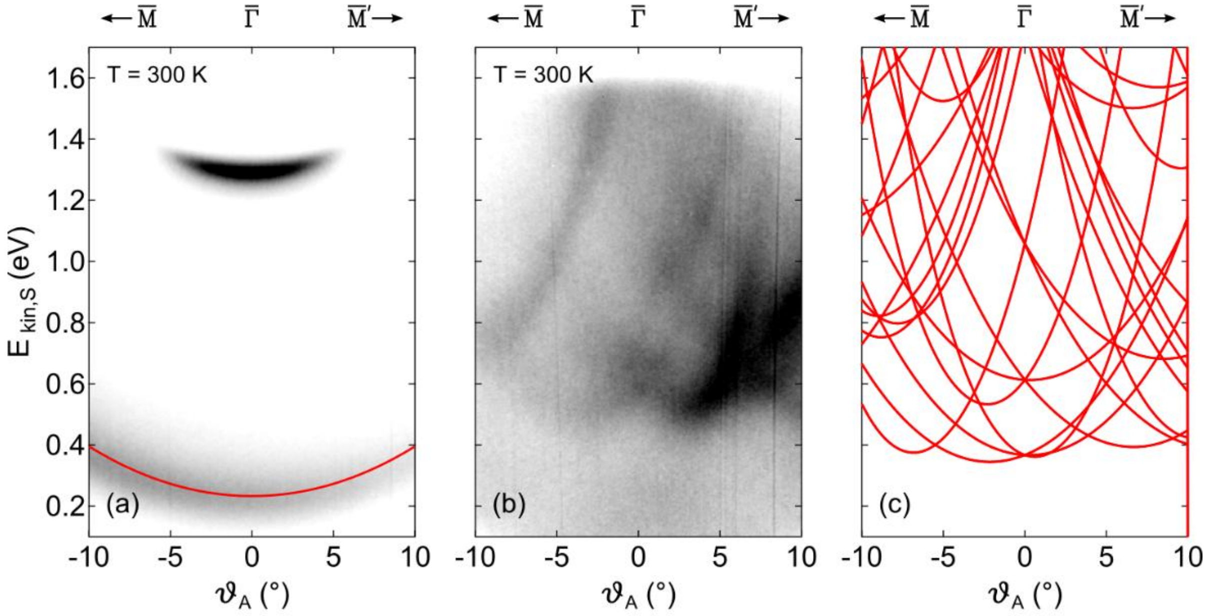

Fig. 1 PES intensity maps of (a) pristine Ag(111) and (b) 1ML SnPc/Ag(111) measured along the high symmetry direction M; (c) Calculated Mahan cone cuts for 1 ML SnPc/Ag(111) under consideration of the lattice structure of the incommensurate SnPc overlayer.

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23 Pump-probe Photoelectron Spectroscopy with MHz High-Order Harmonics: Surface Photovoltage Shifts at the SiO_2/Si(001) Interface

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Time- and fluence dependent surface photovoltage shifts at the SiO_2/Si(001) interface are studied with pump-probe photoelectron spectroscopy using MHz high-order harmonics in combination with a time-of-flight spectrometer. The high-order harmonics from 13 to 42 eV are generated by up to 14 µJ pulses at 1.2 eV with a repetition rate from 0.2 MHz to 1.0 MHz. At photon energies of 25 and 30 eV, the bandwidth of a monochromatized single harmonics is about 80 meV.

High harmonics at 23eV are used to record the angle-resolved valence band spectrum upon IR excitation at 1.2eV. A pulse energy of abput 0.25 µJ is used to optically excite charge carriers at the interface between a 15 Å thick SiO_2 film and a *p*-doped Si(001) substrate. As a consequence of the IR excitation at, an energy shift up 320 meV is observed in the photoelectron spectrum from the valence band maximum of SiO_2. This shift is known as surface photovoltage shift due to time-dependent band bending upon optical charge carrier generation [1].

On the poster we will present the setup [2,3] and discuss the observed fluence- and time-dependent surface photovoltage. In total, our results demonstrate an efficient design of pump-probe photoelectron spectroscopy with high-order harmonics at MHz repetition rates with high energy resolution.

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New J. Phys. **17**, 013035 (2015).

24 On the Origin of Photocurrents in the Topological Insulator Bi2Se3

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Topological insulators host metallic topological surface states with helical spin structure. This makes them promising materials for the generation of spin-polarized currents. The TI Bi_2Se_3 is intrinsically n-doped and thus the topological surface states at the Γ point with a Dirac cone dispersion is occupied. Two-photon photoemission experiments have shown that Bi_2Se_3 exhibits a second Dirac cone in the band gap between the second and third conduction band [1].

We studied the excitation of electrons with circularly polarized light in the second Dirac cone of Bi_2Se_3 with two-photon photoemission and one-step photoemission calculations. To map the momentum distribution, we use an angle-resolved time-of-flight spectrometer (Themis, SPECS), which detects all photoelectrons emitted in a cone of 30° opening angle. The second Dirac cone can be populated from the first Dirac cone. We find dichroic photoemission for circularly polarized pump pulses, both at resonant (cf. Fig. 1) and off-resonant excitation. Only for excitation out of the occupied bulk conduction band, we observe an asymmetric electron population, which would be a prerequisite for a spin-polarized photocurrent. Thus our results question the topological origin of photocurrents in Bi_2Se_3 at near-IR excitation energies [2].

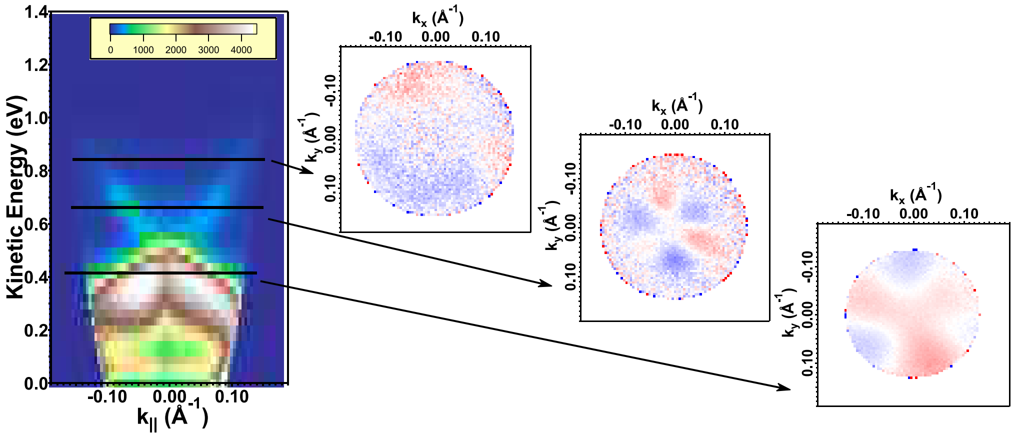

Fig. 1: 2PPE signal and circular dichroism upon resonant excitation from the first to the second Dirac cone.

[1] D. Niesner et al., Phys. Rev. B **86**, 205403 (2012).
[2] C. Kastl et al., Nature Commun. **6**, 6617 (2015).

25 Ultrafast Electron Dynamics and Small Polaron Formation in DMSO Films on Cu(111)

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Dimethylsulfoxide (DMSO) is an ubiquitous chemical in a wide range of applications from organic chemistry synthesis to electrochemistry and battery technology. It has been identified an electrolyte in lithium-oxygen batteries, which could offer high energy storage density. Despite DMSO's wide-spread use, the electronic states of excess electrons in DMSO have not been fully investigated. In our current work, the ultrafast electron solvation dynamics in DMSO have been investigated using time- and angle-resolved two-photon photoemission spectroscopy of condensed DMSO films on a Cu(111) single crystal. At DMSO coverages higher than 1 ML, initially a delocalized electronic state is formed in DMSO at 2.83 ± 0.05 eV relative to the Fermi level, with an effective mass of approximately 1.8 times the mass of a free electron. As shown in Figure 1, this state simultaneously undergoes localization and energetic stabilization where the peak energy shifts to lower energies at a rate of approximately 1 eV/ps as the peak dispersion flattens. By 200 fs after formation, this state exhibits negative dispersion, indicating the formation of a localized electronic state, and ten-fold slower peak shifting before decaying completely within 1 ps. We attribute peak energy and electron localization changes to small polaron formation in DMSO. At coverages >2 monolayers, the decay of the small polaron state is concomitant with the rise of a surface state at 2.30 ± 0.05 eV at the DMSO/vacuum interface. This surface state has an extremely long lifetime, on the order of seconds, which increases with increasing average DMSO coverage and exhibits possible reactivity with oxygen. As DMSO is a common electrolyte, our results on non-equilibrium electronic states in DMSO at surfaces are important for understanding electrochemistry.

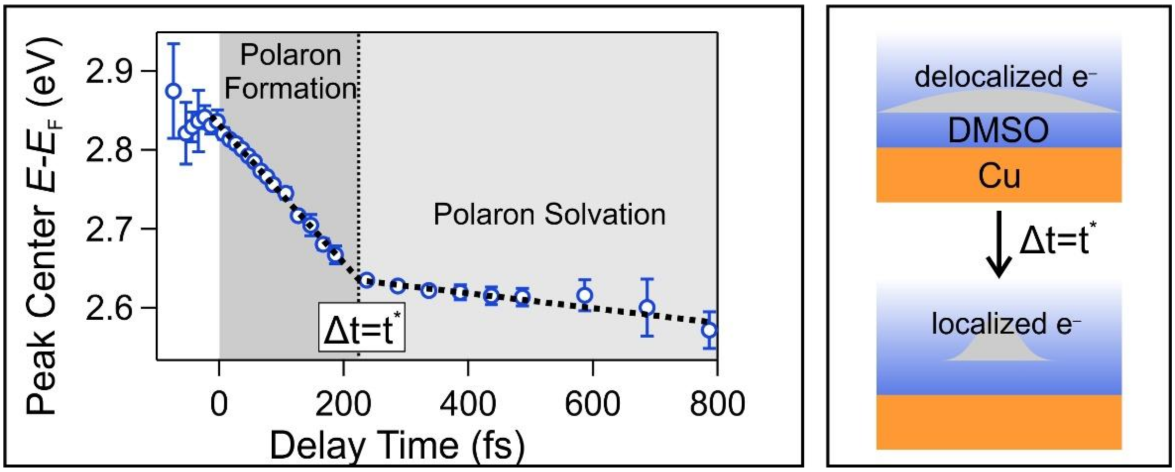

Fig. 1 Two regions of peak center shifting due to small polaron formation and subsequent solvation.

26 Time Resolved Electron Diffraction: Dynamics of Surface Structures on the Femtosecond Timescale

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For the determination of surface structures, a variety of techniques are established, one of which is electron diffraction. This technique has been successfully expanded with temporal resolution on basis of the pump and probe principle: pump fs-IR-laser- and probe electron-pulses [1]. In our group, we increased the temporal resolution from several tens of picoseconds to below 350 femtoseconds within the last decade.

In this presentation the principles of time resolved electron diffraction for high energy electrons (TR-RHEED) are discussed and illustrated by two examples: thin Pb-films on a Si(111) substrate and one monolayer of adsorbed Indium atoms on Si(111) (see figure). With time resolved electron diffraction not only the dynamics of transient structural changes can be observed but in addition the transient surface temperature can be extracted from the data. As electron diffraction is sensitive to the atomic motions, it thus yields complementary information on energy transfer processes between the electron and phonon subsystems and energy dissipation by phonon transport effects. Thin epitaxial Pb-films on Si(111) serve as an example to demonstrate how the surface and bulk is heated on time scales of 3 ps and 5 ps, respectively. This is at least a factor of three slower than observed in time resolved photoemission experiments [2]. The cooling of the thin film is determined by the reduced energy transport across the Pb/Si-Interface. For films of 4 nm thickness, the cooling time constant is 300 ps. In contrast, the In/Si(111)-system shows a much faster response of 350 fs which is ascribed to the non-thermal melting of the groundstate charge density wave that is formed in the Indium atomic chains at low temperatures.

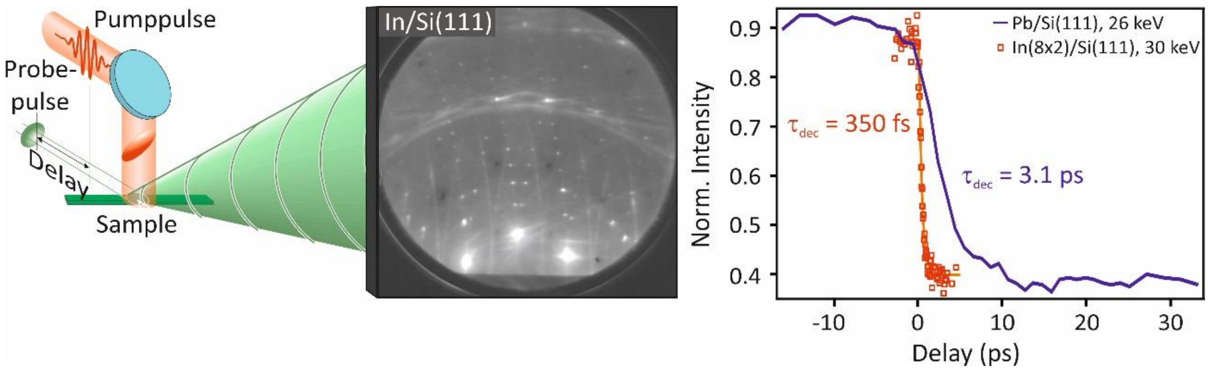

Fig. TR-RHEED: Principle of the setup and transient diffraction spot intensity of Pb and In on Si(111)

[1] A. Janzen, B. Krenzer, O. Heinz, P. Zhou, D. Thien, A. Hanisch, F.-J. Meyer zu Heringdorf, D. von der Linde, M. Horn-von Hoegen, Rev. Sci. Inst. **78**, 013906 (2007).

[2] L. Rettig, P.S. Kirchmann, U. Bovensiepen, New J. Phys. **14**, 023047 (2012).

27 Dynamics of Interface-state-mediated Charge Transfer Processes at Metal/organic Heterolayer Systems

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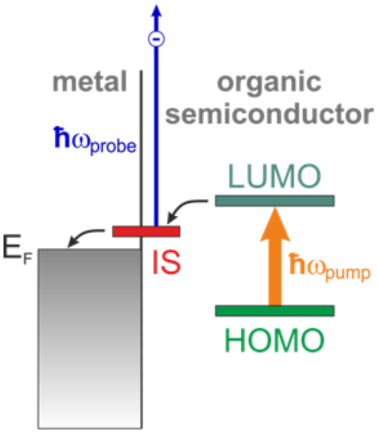
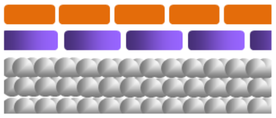
Time resolved two-photon photoemission (2PPE) studies can be a powerful experimental technique to investigate the microscopic mechanisms of charge carrier injection and extraction at metal-molecule contacts. At many well-defined interfaces between ordered organic molecular layers and single-crystal metal surfaces, interface-specific electronic states, located energetically between the Fermi level of the metal and the LUMO of the organic semiconductor, have been identified. Here, we explore their role in the charge transfer by conducting experiments at several heterolayer systems of controlled thickness grown on Ag(111).

Tuneable pump pulses ranging from 1.6 to 2.5 eV allowed us to selectively excite specific organic layers. Time-delayed UV pulses probe the transient population of the interface state (IS). A strong resonant enhancement of the IS intensity at pump photon energies corresponding to the optical gap of the molecular layers is observed. The buildup of the IS population is delayed with respect to the pump pulse by 60 fs for a bilayer of PTCDA/Ag(111) and by 160 fs for PTCDA/TiOPc/Ag/111). Comparison with CuPc/ PTCDA/ Ag(111) and a bilayer of TiOPc/Ag(111) clearly show that these resonances are the result of a HOMO-LUMO excitation in the second or third organic layer followed by a fast electron transfer to the IS. From the IS, the electrons decay into the metal on time scales between 25 and 100 fs depending on the exact energy position of the IS relative to the Fermi level.





Our results show that metal/organic interface states, located between Fermi level and LUMO, mediate the transfer between metals and organic semiconductors. Moreover, they highlight how individual steps to the charge transfer processes can be identified and resolved in time by means of 2PPE.

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28 Delayed Triplet Formation via Hybrid Charge Transfer Exciton (HCTE) State at CuPc/GaAs Interface

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Combining organic and inorganic semiconducting material is a promising way to meet the demands of high power conversion efficiency, low cost, and environmental compatibility by incorporating the merits of either mateirals. Interestingly, it has been reported that significantly different photo-generation mechanism between these two materials results in unique physical behavior such as the presence of the hybrid "Frenkel-Wannier exciton" at organic/inorganic hybrid interface [1] although its dynamical behavior is not clear yet. Thus, in the organic/inorganic hybrid system, ultrafast dynamics of charged carriers (exciton) are an important puzzle to be solved out for device efficiency improvement along with each intrinsic material issue. In this research, we focus on gallium arsenide (GaAs) as inorganic materials in the hybrid system owing to its outstanding properties such as high electron mobility and high efficiency for photovoltaic applications. Also, C_{60} and CuPc molecules, which are generally used as accepter and donor in organic photovoltaics (OPVs), are selected as organic counterpart to design the organic/inorganic hybrid systems. For four hybrid systems, C_{60}/*p*- or *n*-GaAs and CuPc/*p*- or *n*-GaAs, we first compare each interfacial electronic structure using ultraviolet photoelectron spectroscopy and inverse photoemission spectroscopy. Based on each interfacial electronic structure, ultrafast charge transport and separation dynamics are studied by using time-resolved two-photon photoemission spectroscopy, in detail. Doping type of GaAs and corresponding space charge field control specific charge separation behaviors upon light absorption. While C_{60}/*p*-GaAs shows electron injection or separation to organic layer, CuPc/*p*-GaAs hole injection behavior depending on light intensity. In addition, the photo-generated carriers within GaAs exhibit difference time evolution in sub-picosecond range depending on each interfacial electronic structure. Especially, in the case of CuPc/*p*-GaAs hybrid system, we observe the interesting phenomenon of the delayed triplet formation in CuPc. Since direct electron excitation within CuPc is neglected, the delayed triplet is probably generated via “hybrid charge transfer exciton (HCTE) states”, [2] which is formed by hole injection from GaAs to CuPc. However, the HCTE is a dark state in 2PPE when CuPc covers the top surface. Here we could obtain the lifetimes of HCTE and triplet states, respectively, when carriers in inorganic material part are initially generated. Finally, we discuss about hybrid type material design for enhanced charge separation.

[1] A. E. Jailaubekov, A. P. Willard, J. R. Tritsch, W.-L. Chan, N. Sai, R. Gearba, L. G. Kaake, K. J. Williams, K. Leung, P. J. Rossky, X. Y. Zhu, Nature Mater. **12**, 66 (2013).

[2] C. K. Renshaw and S. R. Forrest, Phys. Rev. B **90**, 045302 (2014).

29 New Instrumentation for spin-integrated and spin-resolved momentum microscopy – METIS and KREIOS

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Two new momentum microscopes are presented by SPECS: our newly developed time-of-flight momentum microscope METIS and the energy dispersive and filtered momentum microscope KREIOS. Both are using an optimized lens design which provides simultaneously highest energy, angular and lateral resolution. The lens provides a full 2π solid acceptance angle with highest angular resolution. In contrast to standard ARPES measurements with conventional hemispherical analyzers, electronic structure data from and beyond the 1st Brillouin zone is recorded without any sample movement. In addition the lens of such an instrument can work in a lateral imaging mode for microscopy as well. This enables navigation on the sample and reduces the size of the area under investigation in ARPES down to a few micrometers in diameter. This combination of large acceptance angle, high angular resolution and small acceptance area, makes this instrument the ideal tool for electronic structure studies on small samples or sample areas. The design is compact with a straight optical axis. Operation modes are (kx,ky,Ek) data acquisition by operation in energy filtered k-space imaging, (ToF-) PEEM mode, energy-filtered real space imaging and micro-spectroscopy mode.

The 3D (kx, ky, Ek) data recording is done with a 2-dimensional delayline detector, with a time resolution of 150 ps and count rates up to 8 Mcps. It uses channelplates with 40 µm spatial resolution. While the x,y position of an incoming electron is converted into kx,ky wave vector, the kinetic energy Ek is determined from the flight time t in METIS or obtained directly by the energy filter in KREIOS. Spin-resolved imaging is achieved by electron reflection at a W(100) spin-filter crystal prior to the 2-dimensional delayline detector. Electrons are reflected in the [010] azimuth at 45° reflection angle. Varying the scattering energy one can choose positive, negative, or vanishing reflection asymmetry.

Besides a description on how the instruments work data from both instruments on different single crystalline materials will be presented.

30 Ultrafast Imaging of Nanoscale Carrier Dynamics by Femtosecond Point-projection Microscopy

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Nowadays, semiconductor technology has reached the nanoscale, at which device functionality becomes strongly dependent on dimensionality. The physical properties of low-dimensional systems are influenced by quantum confinement and surface effects, and carrier dwell times become comparable to the time scale of fundamental relaxation processes. Semiconductor nanowires (NWs), in particular, are envisioned as building blocks for future nanoelectronic and nanophotonic devices and are ideal model systems to study the physics of quasi 1D-structures. Device performance is usually dictated by carrier lifetime, and in NWs the relaxation and recombination processes are crucially influenced by the surface and spatially vary across heterojunctions and grain boundaries. Understanding the spatially resolved dynamics of charge carriers in NWs is thus of particular importance. Most time-resolved techniques, however, spatially average over the entire nanostructure.

We developed femtosecond point-projection microscopy (fsPPM) as a new approach to study ultrafast carrier dynamics in nanostructures in real space and time [1]. In fsPPM, low-energy electron pulses photoemitted from a metal nanotip are transiently deflected by time-varying nanoscale fields in the vicinity of nanoobjects positioned several µm in front of the tip. Employing the optical pump - electron probe scheme, fsPPM reveals nanometer spatially-resolved information about ultrafast currents upon photoexcitation, as demonstrated by mapping the ultrafast inhomogeneous response of oxidized axially doped InP-NWs to above-bandgap photoexcitation. Depending on doping, photoexcited charge carriers are radially separated on the 100 fs time-scale by surface space charge fields. The accumulated charge at the surface is neutralized on the few-picosecond time scale, indicating ultrafast surface recobination via oxide-related surface states.

The sensitivity as well as the spatio-temporal resolution of fsPPM can be significantly increased by decreasing the tip-sample distance. At sub-µm distances fsPPM merges into femtosecond low-energy electron holography, promising real-space imaging of ultrafast charge movement with single-charge sensitivity [2] and few-femtosecond temporal resolution. As a first step in this direction, we recently demonstrated that broadband nanofocused surface plasmon polaritons can efficiently trigger electron emission from the apex of a gold nanotip within less than 10 fs [3]. This provides the foundation for ultrafast holography with low-energy electrons.

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[2] Latychevskaia et al., Nano Lett. **16**, 5469 (2016).
[3] Müller et al., ACS Photonics **3**, 611 (2016).

31 Optical Materials Design of TMDCs and Frustrated Mott Insulators

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We discuss manipulation of the topology of a band insulator and induction of topological order in a Mott insulator, accessing transient Floquet steady states via broad pump pulses. First, a realistic material description of monolayer transition-metal dichalcogenides (TMDCs) leads to a novel mechanism to optically induce topologically-protected chiral edge modes, facilitating optically-switchable conduction channels, that are insensitive to disorder. Our strategy is to understand non-equilibrium Floquet-Bloch bands and topological transitions directly from *ab initio* calculations, using WS_2 to illustrate control of chiral edge modes not qualitatively sensitive to microscopic materials details [1]. We extend these ideas to strongly-correlated systems and show that pumping frustrated Mott insulators with circularly-polarized light can drive an effective spin system across a phase transition to a chiral spin liquid (CSL). The transient time evolution of a Kagome lattice Hubbard model is well-captured by an effective spin description, where circular polarization promotes a staggered scalar spin chirality directly to the Hamiltonian level. We fingerprint the phase diagram and find a stable photo-induced CSL in proximity to the equilibrium ground state [2]. These results suggest new avenues to marry dynamical symmetry breaking, strong interactions, and *ab initio* materials modelling, to access elusive phase transitions that are not readily accessible in equilibrium.

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32 Attosecond Delays in the Photoemission from the Layered, Centrosymmetric Bi2Te3 and non-Centrosymmetric BiTeCl Crystals

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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 with unprecedented resolution [1]. The physical origin of these delays is not yet fully understood and various theoretical models coexist demonstrating our still limited understanding of the fundamentals of the photoemission process.

Here we report attosecond time-resolved photoemission from the layered crystals Bi_2Te_3 and non-centrosymmetric BiTeCl. For the latter, the lack of inversion symmetry allows studying relative photoemission delays for differently terminated but well-defined and inert surfaces. In addition, photoelectron propagation effects such as the inelastic mean free path can be determined experimentally because of the reversed layer stacking for differently terminated surfaces. This reduces the ambiguities for classical electron trajectory calculations performed to model the observed relative photoemission delays. However, electron propagation alone cannot explain the measured relative delays. Taking into account local atomic effects and many body corrections reduce the discrepancy between experimental observations and theoretical predictions but still yield no satisfactory explanation. This shows that additional mechanism in the photoemission process significantly influence the photoelectron dynamic and hence a refined model of photoemission is needed.

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33 Ultrafast Peierls Transition in In/Si(111) Nanowires

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Quasi-1D In/Si(111) nanowires undergo a structural transition from a 4x1 to an 8x2 unit cell at around 120 K, concomitant with a metal-to-insulator transition. A recent combined DFT and Raman study [1] points strongly to a Peierls-like scenario, whereby a combination of shear and rotary distortions lead to the opening of electronic band gaps.

Here we investigate the dynamics of the electronic structure during the ultrafast photoinduced 8x2 to 4x1 transition using XUV time- and angle-resolved photoemission spectroscopy (trARPES) at 500 kHz with sub-50 fs temporal resolution. Starting from the 8x2 phase we follow the gradual evolution of the electronic structure into the 4x1 phase on a femtosecond time scale. The gap at the 8x2 Brillouin zone boundary is observed to close already after 200 fs, while states at the zone centre shift from above to below the Fermi level within 500 fs. Moreover, the structural transition into the 4x1 phase, as manifested by the splitting of the m_2-m_3 bands, is completed after 700 fs. The distinct time-scales of the insulator-to-metal and structural transitions, and the lack of a coherent modulation by an amplitude mode as observed in the RTe_3 compounds [2], clearly differ from expectations for a "standard" Peierls-like scenario. A complete description of the transition thus requires understanding of the subtle interplay between transient electronic and structural changes following optical excitation.

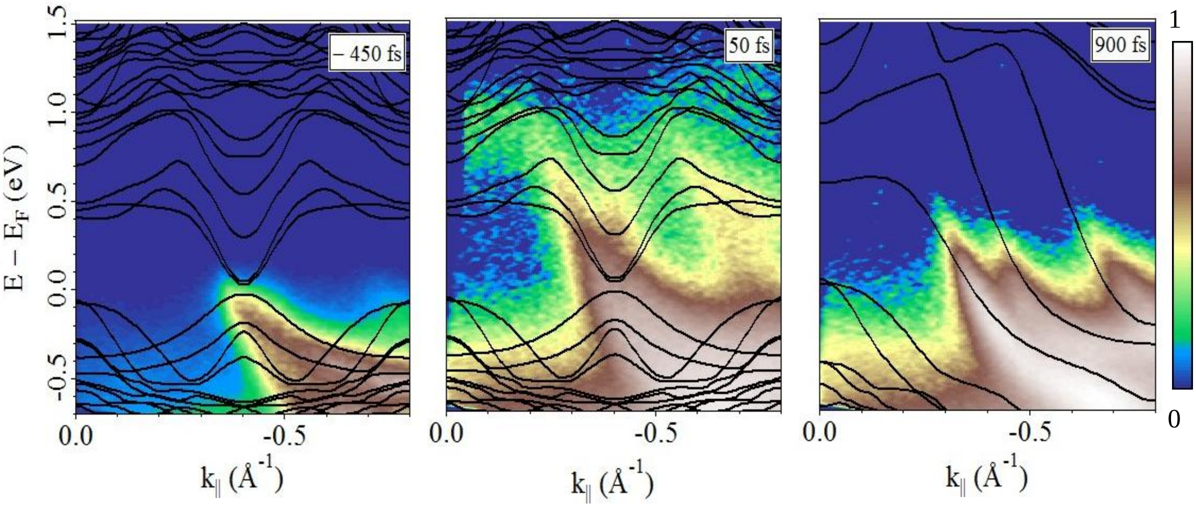

Fig. 1 Band structure of In/Si(111) nanowires at different delays shown in linear (left) and log (centre & right) colour scales revealing the 8x2 to 4x1 phase transition. The DFT band structure of the 8x2 and 4x1 phases are overlaid on the respective images.

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34 Ultrafast Magnetic Imaging with Circularly Polarized High Harmonics

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The recently demonstrated generation of circularly polarized extreme ultraviolet (XUV) radiation from high-harmonic light sources [1,2] in combination with lensless imaging techniques opens a new and powerful route for the spatially-resolved study of ultrafast magnetization dynamics [3].

Integrating the in-line production of a bi-circular field [4] in a recent implementation of high-harmonic-based coherent diffractive imaging [5] allows us to generate bright circularly polarized harmonics in the 25-70 eV photon energy range. Using the circularly polarized high harmonic pulses, magnetic imaging via x-ray magnetic circular dichroism (XMCD) at the M-absorption edges of Co, Ni, Mn and Fe becomes possible.

Here, we demonstrate the first nanoscale magnetic imaging with high harmonic radiation employing Fourier transform holography [6] at the cobalt M edge. Our experiment opens the path for imaging of ultrafast magnetization dynamics and chiral phenomena with nanometer spatial resolution, high temporal resolution down to the attosecond time scale combined with element-specificity.

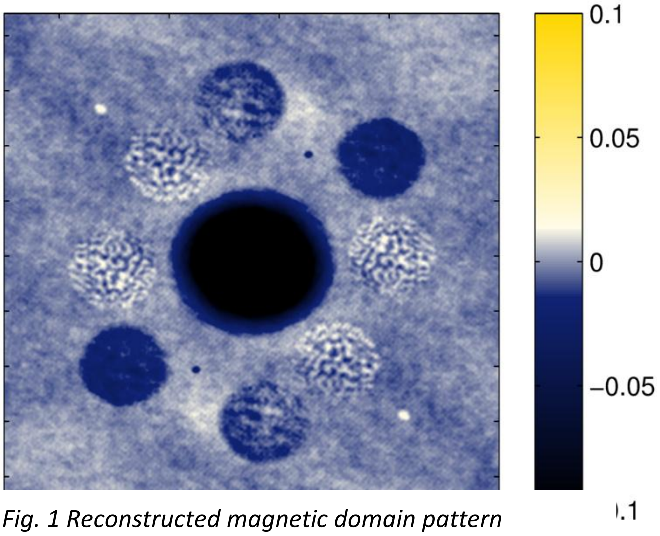

Fig. 1 Reconstructed magnetic domain pattern of a Co/Pd multilayer structure.

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35 High Energy Few-cycle OPCPA System with Variable Repetition Rate from 0.1 – 4 MHz

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The generation of intense few-cycle laser pulses by Optical Parametric Chirped Pulse Amplification (OPCPA) is a novel, innovative technology to obtain high peak intensities [1], as well as ultrashort, µJ-level pulses at high repetition rate in the MHz range with the advantage of improved statistics and higher photon fluxes [2].

We present experimental results from a compact Ti:sapphire oscillator seeded OPCPA system (venteon OPCPA, Laser Quantum GmbH) with high pulse energy > 25µJ delivering few-cycle pulse durations < 8 fs (Fig. 1(b)) at 200 kHz with a long-time active stabilization and operation over several hours with low rms noise < 1 %. The repetition rate can be easily changed via a push button between 0.1 and 4 MHz (Fig. 1(a)). Alternatively to few-cycle pulse generation, the same setup can work in a tuning mode providing wavelength tunability from 680 to 1050 nm and more than 15 µJ (at 100 kHz) pulse energy nearly over the whole tuning range (Fig. 1 (c)). The tuning speed is only limited to the speed of mechanical movement and we demonstrated a switching speed of more than 250 nm/ms for changing the central wavelength from 740 to 910 nm in less than 680 µs.

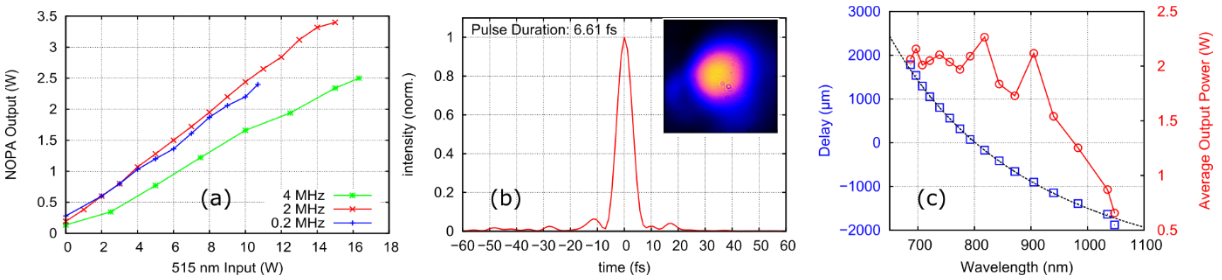

Fig. 1 (a) Measured pulse duration with SPIDER of 25 µJ NOPA output at 200 kHz. Inset: Corresponding output beam profile. (b) Slope of the compressed parametric output energy at 500 kHz in dependence on the 80 MHz background energy and the associated pulse suppression. (c) Central wavelength tuning of the OPCPA (measured central wavelength (blue dots), material dispersion (blue) and the corresponding output power (red)).

Maintaining a high contrast between the amplified pulse and the 80 MHz seed pulse background becomes challenging when the repetition rate of the OPCPA is increased towards a MHz regime. We demonstrate a simple and efficient scheme to improve this contrast by 36 dBc whereas the NOPA output power only decreases by 3.3 dBc. Therefore, our OPCPA system is ideally suited for a wide range of applications due to the versatile output specs without alteration of the setup.

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36 Two-color Coherent Control of Above-threshold Photoemission from Single Tungsten Nanotips

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We demonstrate that multi-photon photoemission including above-threshold orders from a nanotip can be coherently controlled with the optical phase between two light fields [1]. By focusing 74 fs drive pulses at 1560 nm and their second harmonic at 780 nm onto the tip and changing the optical phase between the two colors, we observe an emission current modulation of up to 97.5 %. Hence the electron current can be strongly increased (by a factor of 3.7) or almost fully turned off due to interference between two different quantum channels in the material. We argue that the extremely high degree of coherence evidenced by this near-unity current modulation depth is due to the confining nature of local field enhancement at the nanotip. The observed quantum interference between different emission pathways is supported by time-dependent density functional theory (TDDFT) simulations and ground-state density functional theory (DFT). Because of the pointy solid-state nature of the nanotip, we can apply large DC fields, offering an additional degree of freedom to investigate the modulation contrast of the photoemitted electron yield. By doing so we show that the non-cooperative distribution of electron emission leads to a decrease in modulation contrast [2].

Furthermore, ongoing research results on femtosecond laser-triggered electron emission in the multiphoton and strong-field regime from other emitters will be discussed.

[1] M. Förster, T. Paschen, M. Krüger, C. Lemell, G. Wachter, F. Libisch, T. Madlener, J. Burgdörfer, P. Hommelhoff, Phys. Rev. Lett. **117**, 217601 (2016).

[2] T. Paschen, M. Förster, M. Krüger, C. Lemell, G. Wachter, F. Libisch, T. Madlener, J. Burgdörfer, P. Hommelhoff, Journal of Modern Optics, in press.

37 Itinerant and Localized Magnetization Dynamics in Antiferromagnetic Ho

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The exchange interaction between different magnetic moments is the dominating force determining spin ordering in magnetic materials. In particular, the interplay of itinerant d-electron and localized f-electron magnetic moments in 4f metals and their alloys opens up new routes to control their magnetic behavior on ultrafast timescales. Here we use surface-sensitive time-resolved resonant magnetic x-ray diffraction at the Ho L_3 absorption edge in grazing-incidence geometry to investigate the demagnetization dynamics in antiferromagnetically ordered metallic Ho after femtosecond optical excitation [1]. By tuning the x-ray energy to the electric dipole (E1) or quadrupole (E2) transition we investigated selectively and independently the spin dynamics of the itinerant 5d and localized 4f electronic subsystems. The simultaneous demagnetization of both subsystems demonstrates strong intra-atomic 4f-5d exchange coupling. The demagnetization time scales which are very similar to ferromagnetic 4f systems, suggest that the loss of magnetic order occurs via a similar spin-flip process in both cases.

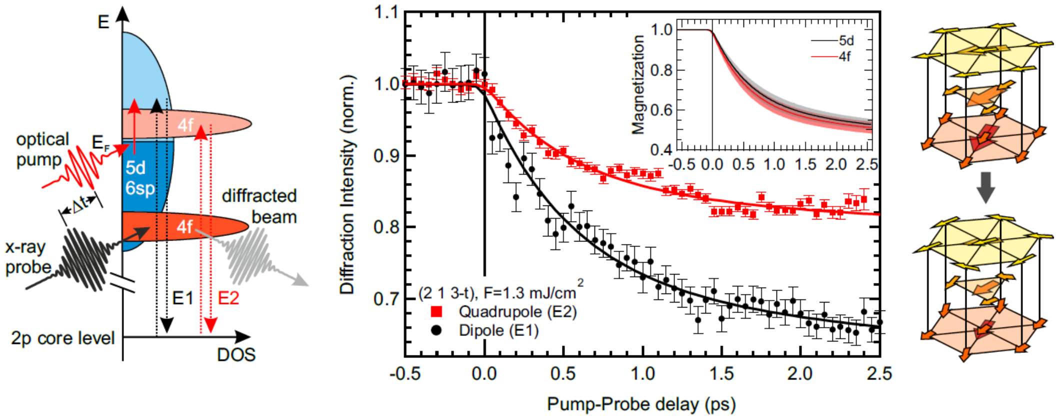

Fig. 1: Tuning of the x-ray probe energy across the Ho L3 absorption edge to the dipole (E1) and quadrupole (E2) transition allows selective probing of the ordered itinerant 5d and localized 4f moments (left). The equivalent demagnetization dynamics of 5d and 4f electrons demonstrate very strong intra-atomic exchange coupling (middle). The observed demagnetization timescales indicate spin-flip scattering in the ferromagnetically ordered layers as dominant demagnetization channel (right).

[1] L. Rettig et al., Phys. Rev. Lett. **116**, 257202 (2016).

38 Resolving Initial Stages of Carrier Thermalization in Graphite Using Time-resolved ARPES with Sub-15 fs Temporal and Near Fourier-limited Spectral Resolution

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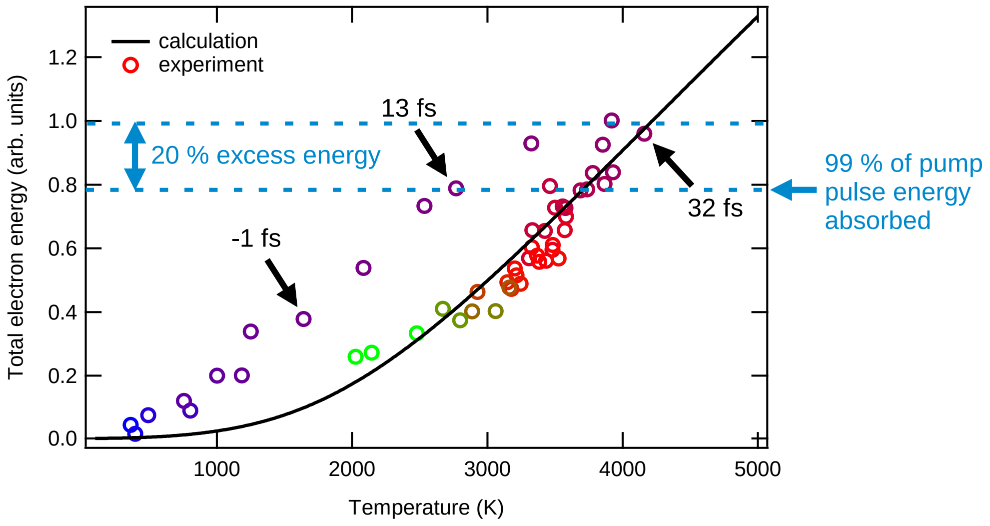
Carrier dynamics in graphitic materials have been subject of theoretical and experimental investigations in recent years. State-of-the-art experimental approaches address the time scales of excitation and relaxation of hot carrier distributions, aiming to break their evolutions down into the underlying fundamental scattering processes as ultimately governed by electron-electron and electron-phonon interaction.

Using time- and angle-resolved photoelectron spectroscopy with sub-15 fs temporal and near Fourier-limited spectral resolution, we report here on the build-up of an out-of-equilibrium electron distribution in graphite within 13 fs and its further evolution during the thermalization towards a Fermi-Dirac distribution.

We discuss potential processes causing this transition and compare the results to corresponding data and calculations reported for graphene [1,2]. Moreover, indications for a momentum redistribution of the hot carriers on ultrafast time scales are presented following a momentum-selective photoexcitation.



Total energy of conduction band electrons as function of equilibrium electron temperature for different pump-probe time delays (color-coded open circles) for graphite; the black curve represents the calculated values for a thermalized electron gas. The comparison between experiment and calculations indicate that thermal equilibrium is reached about 30 fs after time zero.

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[2] Malic et al., Phys. Rev. B. **84**, 205406 (2011).

39 Two-Photon Photoemission from the Giant Rashba System Bi/Ag(111)

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We study the electronic structure of the (√3 x √3)*R*30° substitutional surface alloy Bi/Ag(111) by time- and angle-resolved, mono- and bichromatic two-photon photoemission (2PPE) with different photon energies *hν* ranging from 1.51 eV to 4.89 eV. The obtained photoemission patterns are particularly rich in structure and feature direct transitions between surface and bulk states. Most notably, we find an intermediate state with a lifetime of ~ 34 fs. Due to its energetic position and characteristic Rashba-type downwards dispersion (Rashba parameter ~ 4 eVÅ), it might well be identified as the unoccupied spin-split p_xp_y-derived surface state ($m_j = ½$) close to the Fermi level [1-3]. However, our time-resolved and photon-energy-dependent measurements rather point towards a location close to the vacuum level, where neither theory nor previous experiments feature any spin-split states. A prominent parabolic feature ~ 4.3 eV above the Fermi level is ascribed to the $n = 2$ image-potential state (IPS) with a corresponding binding energy of ~ 0.17 eV and a lifetime of ~ 27 fs, while the $n = 1$ IPS cannot be clearly identified. We attribute this to a resonant transition from the occupied Rashba-split sp_z-derived surface state [4] into the $n = 2$ IPS. The former is observed as an initial state in 2PPE, confirmed by one-photon photoemission at $h\nu = 6.2$ eV. In addition, circular dichroism provides information on the unconventional spin texture [2, 5] of both, the occupied and unoccupied electronic structure of this giant Rashba system.

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[2] S. N. P. Wissing et al., Phys. Rev. Lett. **113**, 116402 (2014).
[3] L. El-Kareh et al., Phys. Rev. Lett. **110**, 176803 (2013).
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[5] R. Noguchi et al., Phys. Rev. B **95**, 041111(R) (2017).

40 Intrinsic Carrier Cooling in High Mobility Graphene on SiC(000-1)

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Graphene, a two-dimensional carbon crystal with a gas of massless Dirac fermions, has promise as a material that is useful in photonic and electronic devices. A comprehensive understanding of carrier cooling in photo-excited graphene is necessary for their applications, however, as competing cooling processes, electron–optical-phonon scattering, and disorder-assisted electron–acoustic-phonon scattering, so-called "supercollision", complicate the problem. It has been challenging to decouple these scattering processes and thus, intrinsic carrier dynamics in graphene is not understood.

In our study, we succeeded in fabricating high mobility graphene grown on a SiC(000-1) C-terminated surface for which the carrier mobility exceeded 100,000 cm^2 V^{-1} s^{-1} and performed time- and angle-resolved photoemission spectroscopy (TRARPES) to understand intrinsic carrier cooling in graphene (without supercollisions). As an example, Figure 1 shows the TRARPES spectra of the C-face graphene taken at several delay times. In the poster session, we will show the observed carrier dynamics and discuss cooling mechanism of photo-excited carriers in high mobility graphene.

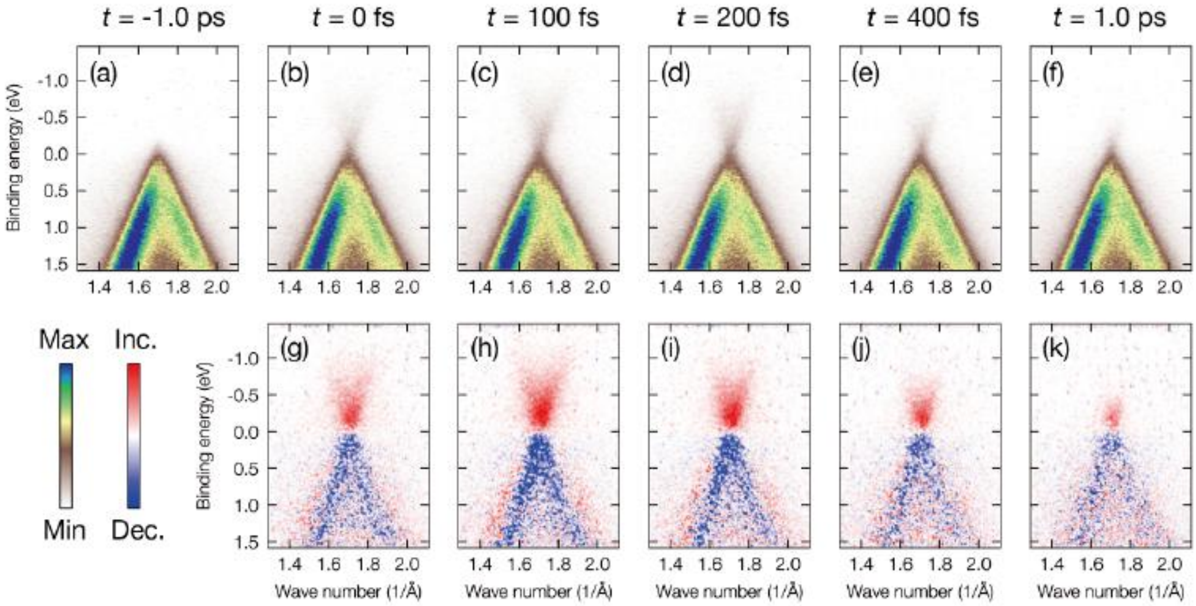

Fig. 1. (a-f) TRARPES spectra and (g-k) its difference spectra taken at specified delay times. All spectra were measured along Γ-K-M direction of the two-dimensional Brillouin zone. Probe light : 27.9 eV, Pump light : 1.55 eV (650 μJ/cm2) and Temperature : 300 K.

41 Ultrafast Spin Dynamics in the Half-metallic Multisublattice Ferromagnet NiMnSb

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NiMnSb is the prototypical half-metallic ferromagnet, first predicted by de Groot in 1983[1]. As such it has been strongly investigated using both theoretical and experimental methods mostly concerning its structural and static magnetic properties (see, e.g., Ref. 2). Compared to other half metals[3,4], however, little interest has been paid to its dynamical properties after optical excitation, especially concerning the fact that it is a multisublattice ferromagnet. A recent theoretical work using TD-DFT predicts quite intriguing spin dynamics in this system[5] with a strong transient increase of the Ni moment.

Here, we present first investigations of thin films of NiMnSb using both the time-resolved magneto-optical Kerr effect (MOKE) and transient reflectivity. In visible MOKE we find a rather unusual evolution of the dynamical magnetization on femto- to picosecond timescales with increasing laser fluence, starting from an apparent initial increase in magnetization followed by a “normal” demagnetization as in the 3d-ferromagnets for low fluences, to an apparent sign change of magnetization on short timescales for high fluences. We discuss this behavior with regard to Ref. 5 and present first element-resolved MOKE data on NiMnSb using a high-harmonic tabletop transverse MOKE setup.

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[5] P. Elliott, T. Müller, J. K. Dewhurst, S. Sharma, E. K. U. Gross, Sci. Reports **6**, 38911 (2016).

42 Ultrafast Response of 1*T*-$TaS_{2-x}Se_x$ Single Crystals

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We present optical femtosecond relaxation dynamics in single crystals of 1*T*-$TaS_{2-x}Se_x$ with 6%, 7.5%, 17% and 19% selenium doping in the strong pump excitation regime. We observe a transition to a new hidden state (H-state) which is inaccessible under normal equilibrium conditions and can only be reached after a quench with a single femtosecond laser pulse at 1.6-4.8 mJ/cm^2 fluence. Similarly as in the pioneering report [1,2], a notable change in the coherent phonon spectra followed by the change in the reflectivity, plays the role of the fingerprint of the successful switching to a new state via optical path. The major output is observation of the significantly higher temperature stability of the H-state in comparison with the parent undoped 1*T*-TaS_2 material.

Complementary to these optical experiments, intense experimental efforts are currently undergoing in order to explore the ultrafast response on femto– to picosecond time scales by means of femtosecond time– and angle–resolved photoemission spectroscopy (trARPES). Tracking the time–, frequency– and momentum–dependent influence of the photoexcitation on the electronic structure as the system undergoes a transition to hidden state might greatly enhance the understanding of the switching mechanism and the nature of the switched state.

The authors acknowledge financial support by the Deutsche Forschungsgemeinschaft through SFB 1242 and Alexander von Humboldt Foundation.

[1] L. Stojchevska et al., Science. **344**, 177 (2014).
[2] I. Avigo et al., Proc. SPIE **9931**, Spintronics IX, 99313V (2016).

43 Ultrafast Optical Response of Gr/Alkali Atoms/Ir(111)

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Graphene (Gr) plasmon, which is the collective in-plane oscillation of electrons in Gr, has been gathering much attention owing to its strong field confinement, long propagation length and tunability. Although Gr plasmon has only been observed in the mid-IR and THz region, the excitation with visible light is strongly desirable. In this work, we study the optical response of alkali-intercalated Gr with steady-state and time-resolved reflectivity measurements using visible light.

Gr on Ir(111) was prepared by chemical vapor deposition under an ultrahigh vacuum condition ($p < 4 \times 10^{-8}$ Pa) [1]. Steady-state reflectivity changes of *p*-polarized light induced by Cs or Li deposition were measured by a halogen lamp and polychromater with a CCD detector. Time-resolved reflectivity measurements were conducted by using femtosecond laser pulses (2.0 - 2.3 eV, 40 fs, 1 kHz).

Fig. 1a shows the reflectivity changes caused by Cs intercalation. The large absorption bands are attributed to the excitation of Gr plasmons activated by both high-level doping from alkali metals and an intrinsic moiré superstructure [1]. Fig. 1b shows the time-resolved reflectivity changes induced by pumping the absorption band, in which clear oscillatory features were observed. The oscillations are due to coherent surface phonons, indicating that the plasmon resonance energy is modulated by the coherent phonons.

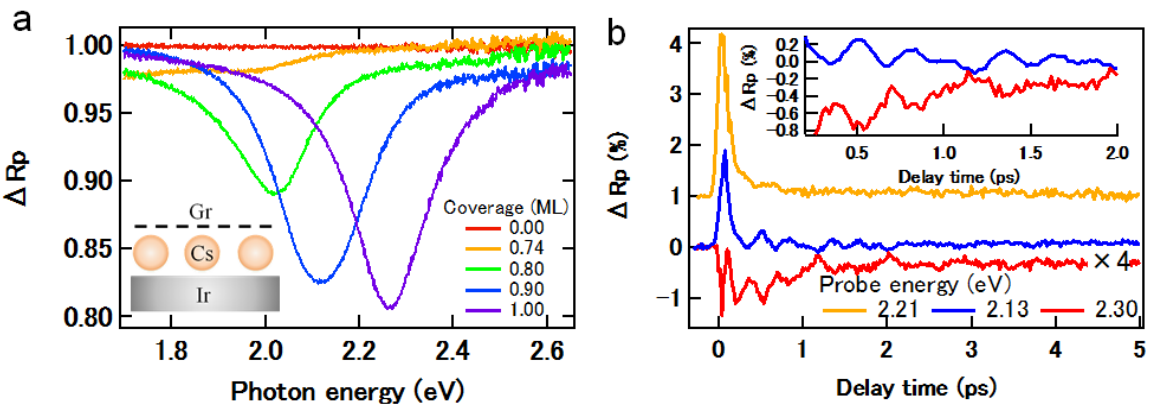

Fig. 1(a) Steady-state and (b) time-resolved reflectivity changes of Cs-intercalated Gr/Ir(111).

[1] M. Petrović et al., Nature Commun., **4**, 2772 (2013).

44 A Bi-directional Surface Photovoltage of Topological Insulator

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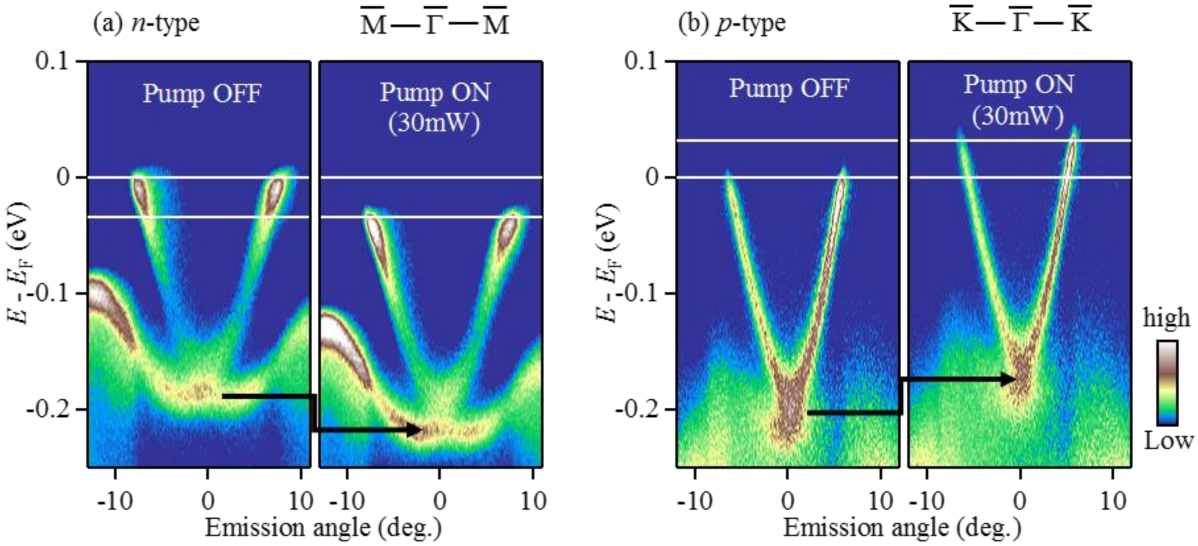
Topological insulators (TIs) possess spin-polarized metallic states on surface as a result of the non-trivial topology of the bulk band structure. The surface of TIs is thus considered as a promising platform for novel spin-electronic functions. Generating spin-polarized surface currents out of the spin-polarized metallic states would be a milestone function, and optical means provides several pathways to this end. One way is to utilize the surface photovoltage (SPV) effect, as proposed recently [1, 2]. To date, SPV effect was observed for the bulk-insulating TI, Bi_2Te_2Se [2]. However, due to the limited controllability of bulk carrier concentrations, only a uni-directional shift of the surface potential has been demonstrated [1, 2]. An ambipolar SPV is crucial for taking full control of the surface current by light.

Here, we have performed time- and angle- resolved photoelectron spectroscopy (TARPES) on bulk-insulating samples of *n*-type and *p*-type Bi_2Te_3 implementing pump-probe method. The experiment was carried out with linearly polarized pump (*hv*=1.48 eV) and probe (5.92 eV) pulses generated by Ti:sapphire laser system operating at a repetition rate of 250 kHz [3]. Under a non-zero pump pulse, a downward (an upward) photovoltaic shift of the bands with respect to that without pumping (Pump OFF) has been observed as shown for *n*-type (*p*-type) sample [Fig.1(a)(Fig.1(b))]. We have also found that the amount of the shifts at upper band edge is larger than that at the Dirac point, which signifies a pump induced filling. Our finding open a way to ambipolar optical control of spin-polarized current generation on the surface of TIs in the next generation opto-spinelectronic devices.



ig. 1 TARPES images of (a) p-type (b) n-type Bi_2Te_3 at t=4 μs acquired with pump OFF and ON.

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45 Phonon Assisted Ultrafast Charge Transfer at van der Waals Heterostructure Interface

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The van der Waals (vdW) interfaces of two-dimensional (2D) semiconductor are central to new device concepts and emerging technologies in light-electricity transduction where the efficient charge separation is a key factor. Contrary to general expectation, efficient electron-hole separation can occur in vertically stacked transition-metal dichalcogenide (TMD) heterostructure bilayers through ultrafast charge transfer between the neighboring layers despite their weak vdW bonding. In this report, we show using ab initio non-adiabatic molecular dynamics (NAMD), that instead of direct tunneling, the ultrafast interlayer hole transfer is strongly promoted by an adiabatic (AD) mechanism through phonon excitation. At room temperature the interlayer hole transfer in MoS2/WS2 is ultrafast occurring on 20 fs, which is in good agreement with the experiment. This process can be suppressed by decreasing the sample temperature to 100K, which reduces the phonon occupation; the charge transfer is then dominated by direct tunneling, which happens on a longer time scale than 300 fs. The atomic level picture of phonon-assisted ultrafast mechanism revealed in our study is valuable both for the fundamental understanding of ultrafast charge carrier dynamics at vdW hetero-interfaces as well as for the design of novel quasi-2D devices for optoelectronic and photovoltaic applications.

46 Photoinduced coherent electron transfer at a heterojunction interface

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Charge transfer in transduction of light to electrical or chemical energy at heterojunctions combining metallic, semimetallic, or semiconducting materials is commonly assumed to occur by incoherent process whereby hot electrons generated in the absorbing material are transported across the heterojunction interface under the influence of a potential gradient (Fig. 1a). Charge transfer can also occur instantaneously by coherent coupling of electronic bands by light at the heterojunction interface (Fig. 1b). Physical insights into how photoinduced interfacial charge transfer occurs can be clarified by methods that pump and probe both the donor and acceptor states on the time scale of electronic relaxation in solids. Applying multidimensional interferometric time-resolved multiphoton photoemission spectroscopy (mPP), we investigate the coherent electron transfer from Ag nanoclusters to their supporting graphite substrate. mPP spectra reveal a resonant coherent two-photon transition from an Ag chemisorption-induced interface state (IFS) to the σ symmetry free-electron-like interlayer band (σ-ILB) of graphite (Fig. 1c). Coherent photoemission spectroscopy shows that the resonant electron transfer is completed within 10 fs by dephasing of the coherently coupled states.

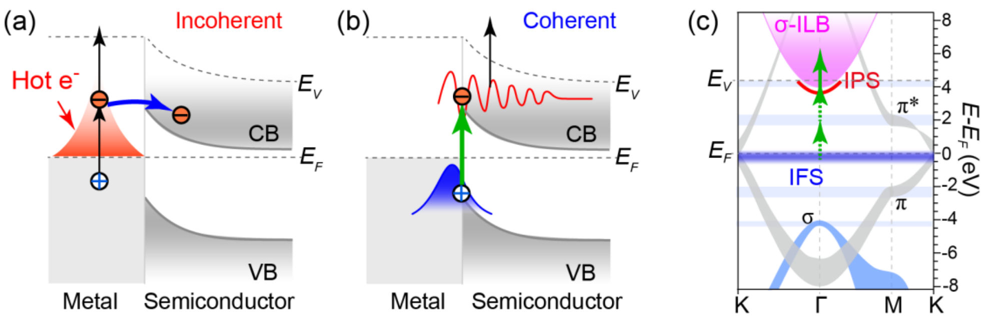

Fig. 1. (a) The incoherent and (b) coherent charge transfer schemes at the metal/semiconductor interface. (c) The k|| band structure of Ag/graphite in the Γ-M-K direction. IFS: Ag chemisorption induced occupied interface state; IPS, image potential state of graphite; σ-ILB: the interlayer band of graphite. The green arrows indicate the resonant coherent two-photon transition from IFS to σ-ILB.

47 Spin-dependent elastic charge-transfer times of core-excited Argon on magnetic surfaces

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Ultrafast charge transfer from photo-excited Argon on magnetic surfaces was found to depend strongly on the electron spin. Core-hole-clock spectroscopy experiments have shown that electrons excited to the Ar 4s state with majority spin are significantly longer-lived on Fe(110) and Co(0001) than those with minority spin [1]. Here we model adsorbed core-excited Ar on the corresponding semi-infinite metal substrates using density functional theory and Green's function techniques [2,3]. To describe the excited adsorbate, we include a core-hole on the Argon pseudopotential and add the removed electron back to the electronic density by constraining it to occupy the Ar 4s level. The extractions of the elastic linewidths of the Ar 4s resonances fully support the experimental findings and rationalize them in terms of the relative energetic position of the resonance and the projected band gap of the substrate's electronic band structure. The role of the adsorption height is also analyzed in our calculations.

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[2] M. Brandbyge, et al., Phys. Rev. B **65**, 165401 (2002)
[3] D. Sánchez-Portal, Prog. Surf. Sci. **82**, 313-335 (2007)

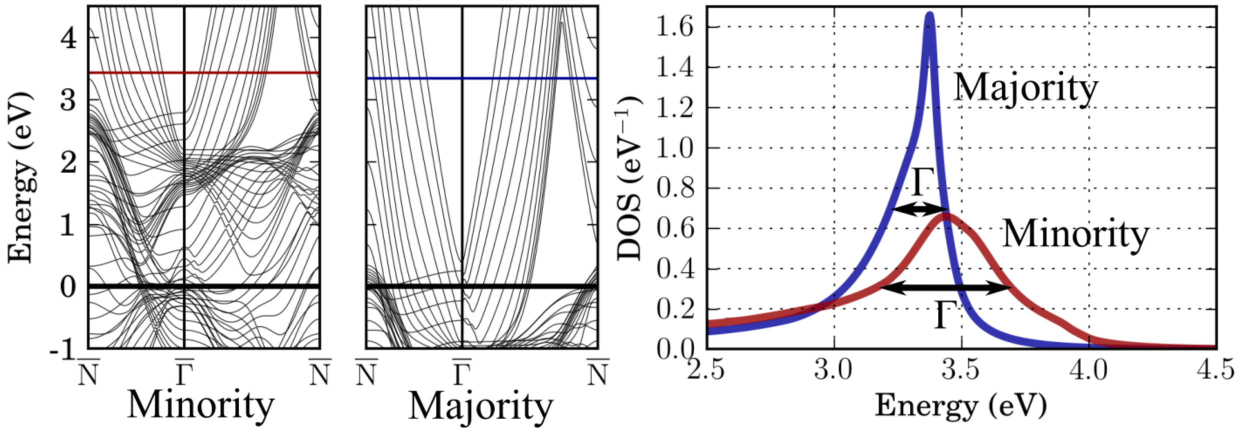

Fig. 1 Left panels: Band structure of Fe(110) for both spin projections and energetic positions of the resonance peaks (thin horizontal lines). The resonances coincide with a band gap at the Γ-point. Right panel: Projected density of states and corresponding linewidths Γ for both spins of the Ar 4s-state

48 Femtosecond laser induced associative desorption of molecular hydrogen from graphite

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The formation of molecular hydrogen via radiation-induced associative desorption from dust particles may play an important role in photon dominated regions of the interstellar medium and the interstellar cycle of matter. We investigate the desorption of molecular hydrogen and deuterium from HOPG following surface excitation with fs-laser pulses at $\lambda = 400$ nm. Desorbed neutral species are analyzed by internal state selective two-photon ionization ((1+1) REMPI) via the $C\ ^1\Pi \leftarrow X\ ^1\Sigma$ XUV transition.

The desorbing hydrogen molecules show a strong non-equilibrium behavior. The recorded rotational populations present a non-thermal distribution that can be fitted by two rotational temperatures for low and high J”. The average rotational energy is determined to $E_{rot} = 390$ cm^{-1} and 480 cm^{-1} for v” = 0 and v” = 1, respectively. The vibrationally excited state is stronger populated than the ground state. State specific kinetic energy measurements show a significantly lower energy for v” = 1: $E_{kin} = 170$ meV for v” = 0 and $E_{kin} = 100$ meV for v” = 1. A nonlinear fluence dependence of the desorption yield allows two-pulse correlation measurements, which give an insight into the relevant desorption mechanisms. The FWHM is about 450 fs which suggest an electron induced desorption mechanism. Measurements for molecular deuterium reveal similar results.

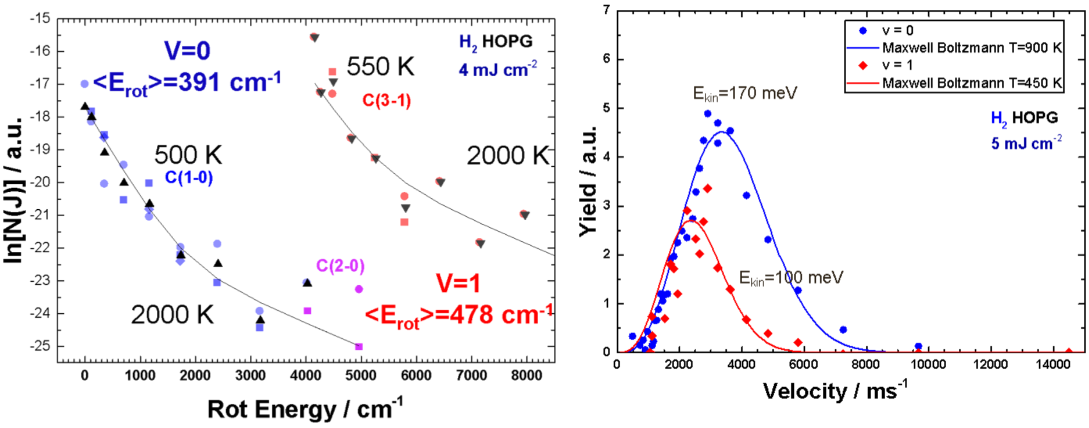

Fig. 1 Molecular hydrogen desorption from graphite. The data indicate a highly populated vibrationally excited state and a non-thermal desorption mechanism

Development and Application of Ultrafast Transmission Electron Microscopy (UTEM)

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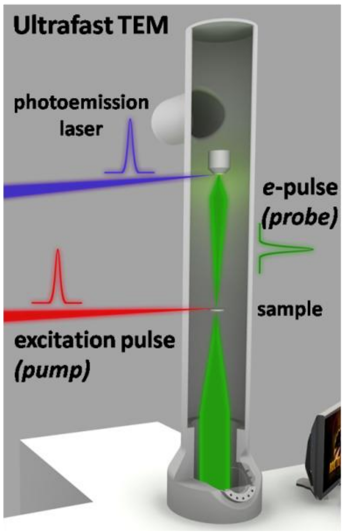
Electron microscopy is tremendously successful in unravelling material structures and compositions on the atomic scale, with a temporal resolution governed by detector response times. Utilizing a stroboscopic approach, ultrafast electron diffraction [1] and microscopy [2,3] techniques provide for a unique access to processes on ultrashort time scales. In ultrafast transmission electron microscopy (UTEM), combining nanoscale spatial resolution of electron microscopy with femtosecond temporal resolution of optical spectroscopy, a pulsed electron beam of sub-picosecond duration is able to probe rapid processes [2]. To further extend the UTEM methodology and to make full use of the imaging capabilities of state-of-the-art TEMs, electron pulses with enhanced coherence properties are required.



In this contribution, the implementation of an advanced UTEM instrument utilizing laser-triggered field emitters will be presented, including first applications harnessing the superior spatial electron beam properties. The Göttingen UTEM instrument is based on a JEOL JEM 2100F Schottky-field-emission TEM, which we modified to allow for optical sample excitation and the generation of ultrashort electron pulses [4,5]. The nanoscopic photoelectron source [4-6] employs localized single-photon photoemission from the front facet of a tip-shaped ZrO/W(100) Schottky emitter. Highly coherent ultrashort electron pulses with a normalized emittance of a few nm·mrad are achieved. Electron focal spot sizes down to 0.8 nm, an electron pulse width of 200 fs (full-width-at-half-maximum) and a spectral bandwidth of 0.6 eV are demonstrated [5]. We give an overview of current experiments in ultrafast phase-contrast imaging and local probing. Forming a well-collimated electron probe, the large transverse coherence length (up to 10% of the beam diameter) allows for the recording of high quality diffraction data from small sample areas. Phase-contrast imaging is shown by imaging magnetic nanostructures with Lorentz microscopy. Furthermore, ultrafast convergent beam electron diffraction near the edge of an inhomogeneously excited single crystalline graphite thin film, providing for spatio-temporal analysis of the local strain evolution with nanometer spatial and femtosecond temporal resolution [7].

In a further line of applications, we utilize the interaction of fast electrons with intense optical near-fields [4,8,9] to establish quantum coherent control of free electron pulses, tailoring the longitudinal and transverse electron beam properties by light [4,10]. As an example, we describe the optically-induced preparation of attosecond electron pulse trains, potentially pushing the limits of the temporal resolution in UTEM into the sub-optical-cycle regime [4,11].

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Program Ultrafast Surface Dynamics 10

time	Electron / Energy Transfer Dynamics	Magnetisation Dynamics	Plasmonics Nanooptics	Novel Phenomena and Materials	Low Dimensional Materials	Attosecond Surface Science
	Sun, 11.6.	Mon, 12.6.	Tue, 13.6.	Wed, 14.6.	Thu, 15.6.	Fri, 16.6.
9:00	ARRIVAL Registration	M. Shibuta	C. Ropers	Ph. Hofmann	I. Gierz	M. Ligges
9:20						S. Mor
9:40		P. Tegeder	M. Raschke	R. Ernstorfer	R. Arafune	E. Krasovskii
10:00		B. Stadtmüller			S. Aeschlimann	
10:20		Coffee break	Coffee break	Coffee break	Coffee break	Coffee break
10:40						
11:00		J. Zhao	P. Hommelhoff	R. Wallauer	G. Fratesi	W. Pfeiffer
11:20				E. Malic	W. Windsor	J. Riemensberger
11:40		K. Watanabe	M. Horn-von Hoegen		R. Huber	L. Castiglioni
12:00				K. Ishioka		
12:20				S. Otto		Closing
13:00		Lunch	Lunch	Lunch	Lunch	Lunch
14:00		Outdoor Workshop	Outdoor Workshop	Conference Outing		Outdoor Workshop
14:20						
14:40						
15:00					Th. Fauster	
15:20					H. Zacharias	
15:40		S. Sharma	B. Moritz			
16:00					H. Petek	DEPARTURE
16:20		S. Mathias	F. O. Schumann			
16:40		B. Andres	A. Paarmann		Coffee break	
17:00		Coffee break	Coffee break			
17:20					T. F. Heinz	
17:40		M. Cinchetti	K. Kuroda			
18:00	Welcome				M. Aeschlimann	
18:20	A. Nilsson	A. Eschenlohr	K. Sumida		U. Höfer	
18:40			Y. Matsumoto			
19:00	M. Murnane	Dinner	Dinner		Dinner	
19:20				Conference Dinner		
19:40	Reception					
20:00	Dinner					
20:20		Poster Session	Poster Session		After Dinner Colloquium	
20:40						
21:00						
21:20						