WUPCOM'15

Winter School on Ultrafast Processes in Condensed Matter

1 – 6 March 2015,

Winklmoosalm, Reit im Winkl

Book of Abstracts

WUPCOM 15 Organizer

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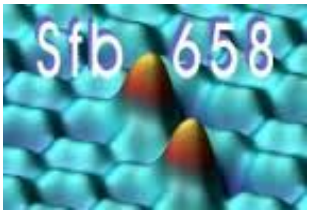

Scientific program

Sunday, March 1

16:00 – 18:00 Arrival and rental of skiing equipment

18:30 – 19:00 Registration

19:00 – 20:00 Dinner

Monday, March 2

7:30 – 8:30	Breakfast
	Morning session (Chair: Martin Weinelt)
8:30 – 9:15	**Jure Demsar** (Johannes Gutenberg-Universität Mainz) *Cooperative atomic motion probed by femtosecond electron diffraction*
9:15 – 9:30	Discussion
9:30 – 10:00	**Selene Mor** (Fritz-Haber-Institut der MPG, Berlin) *Ultrafast coherent phonon dynamics during the phase transition of the quasi one-dimensional Ta_2NiSe_5*
10:00 – 15:30	Outdoor workshop
	Afternoon session (Chair: Cheng-Tien Chiang)
16:00 – 16:30	**Abdul Samad Syed** (Universität Duisburg-Essen) *Ultrafast dynamics of a Pb overlayer structure grown on flat and vicinal Si(111)*
16:30 – 17:00	**Markus A. Huber (**Universität Regensburg) *Ultrafast sub-cycle terahertz nano-spectroscopy*
17:00 – 17:30	Coffee break
17:30 – 18:00	**Alexander Lerch** (Philipps-Universität Marburg) *Ultrafast electronic excitations at the buried GaP/Si interface*
18:00 – 18:30	**Alex Paarmann** (Fritz-Haber-Institut der MPG, Berlin) *Second Harmonic Spectroscopy in the Reststrahlen Band of 6H-SiC*
19:00 – 20:00	Dinner
20:00 –	More discussions...

Tuesday, March 3

7:30 – 8:30 Breakfast

Morning session (Chair: Jure Demsar)

8:30 – 9:15 **Sascha Schäfer** (Georg-August-Universität, Göttingen)
Ultrafast electron imaging and diffraction with nanoscale photoemitters

9:15 – 9:30 Discussion

9:30 – 10:00 **Petra Hein** (Christian-Albrechts-Universität zu Kiel)
Hot electron dynamics at 2H-MoS_2 surfaces: Time- and angle-resolved photoelectron spectroscopy results

10:00 – 15:30 Outdoor workshop

Afternoon session (Chair: Jens Güdde)

16:00 – 16:30 **Matthias Hohenleutner** (Universität Regensburg)
High-order harmonic generation by dynamical Bloch oscillations in a bulk solid

16:30 – 17:00 **Gerald Rohde** (Christian-Albrechts-Universität zu Kiel)
Time- and angle-resolved XUV ARPES at sub-15 fs temporal resolution

17:00 – 17:30 Coffee break

17:30 – 18:00 **Cheng-Tien Chiang** (MPI für Mikrostrukturphysik und Martin-Luther-Universität Halle-Wittenberg)
Double photoemission spectroscopy on correlated d electrons in solids using a megahertz high-order harmonic light source

18:00 – 18:30 **Sebastian Emmerich** (Universität Kaiserslautern)
trARPES with bright narrowband HHG pulses using frequency-doubled Ti:Sapphire lasers

19:00 – 20:00 Dinner

20:00 – More discussions...

Wednesday, March 4

7:30 – 8:30 Breakfast

Morning session (Chair: Uwe Bovensiepen)

8:30 – 9:15 **Michael Horn-von Hoegen** (Universität Duisburg-Essen)
Electron Diffraction at Surfaces: by now Ultrafast!

9:15 – 9:30 Discussion

9:30 – 10:00 **Thomas Vasileiadis** (Fritz-Haber-Institut der MPG, Berlin)
The effect of confinement in the electron-phonon coupling studied with ultrafast electron diffraction

10:00 – 15:30 Outdoor workshop

Afternoon session (Chair: Ralph Ernstorfer)

16:00 – 16:30 **Uwe Bovensiepen** (Universität Duisburg-Essen)
Competition of spin currents and spin-flip scattering in ultrafast magnetization dynamics of Co/Cu(001) analyzed by the complex time-resolved MOKE

16:30 – 17:00 **Alexey Melnikov** (Fritz-Haber-Institut der MPG, Berlin)
Direct monitoring of hot carrier-formed spin currents in metallic multi-layers by time-resolved magneto-induced second harmonic generation

17:00 – 17:30 Coffee break

17:30 – 18:00 **Moritz Barkowski** (Universiät Kaiserslautern)
Depth-dependent study of femtosecond spin-currents in Ni/Au

18:00 – 18:30 **Kamil Bobowski** (Freie Universität Berlin)
Ultrafast magnetization dynamics of Gadolinium measured by x-ray magnetic circular dichroism in reflection geometry

18:30 – 19:00 **Beatrice Andres** (Freie Universität Berlin)
Ultrafast magnetization dynamics in Gadolinium – Analyzing the transient spin polarization

19:00 – 20:00 Dinner

20:00 – More discussions...

Thursday, March 5

7:30 – 8:30 Breakfast

Morning session (Chair: Michael Horn-von Hoegen)

8:30 – 9:15 **Ralph Ernstorfer** (Fritz-Haber-Institut der MPG, Berlin)
Femtosecond electrons probing structural dynamics and ultrafast currents

9:15 – 9:30 Discussion

9:30 – 10:00 **Konrad Gillmeister** (Martin-Luther-Universität Halle-Wittenberg)
Electron dynamics in NiO ultrathin films

10:00 – 15:30 Outdoor workshop

Afternoon session (Chair: Alex Paarmann)

16:00 – 16:30 **Ishita Agarwal** (Universität Duisburg-Essen)
Femtosecond time-resolved two-photon photoemission spectroscopy of solavted electrons in $C_6H_5F/D_2O/Cu(111)$

16:30 – 17:00 **Harald Kirsch** (Fritz-Haber-Institut der MPG, Berlin)
Probing Water Dissociation and Structure at the α-Al_2O_3(0001) Surface Using Interface Specific Vibrational Dynamics

17:00 – 17:30 Coffee break

17:30 – 18:00 **Wibke Bronsch** (Freie Universität Berlin)
Ultrafast Exciton Dynamics in Thin Sexithiophene Films

18:00 – 18:30 **Jens Güdde** (Phillips-Universität Marburg)
Electron dynamics in the topological insulator Sb_2Te_3

18:00 – 18:30 **Martin Weinelt** (Freie Universität Berlin)
Electronic structure and dynamics of the topological insulator Sb_2Te_2S

19:00 – 20:00 Dinner

20:00 – More discussions...

Friday, March 6

8:00 – 9:00 Breakfast

9:00 – 10:30 Departure and return of skiing equipment

Monday, March 2

7:30 – 8:30	Breakfast
	Morning session (Chair: Martin Weinelt)
8:30 – 9:15	**Jure Demsar** (Johannes Gutenberg-Universität Mainz) *Cooperative atomic motion probed by femtosecond electron diffraction*
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18:00 – 18:30	**Alex Paarmann** (Fritz-Haber-Institut der MPG, Berlin) *Second Harmonic Spectroscopy in the Reststrahlen Band of 6H-SiC*
19:00 – 20:00	Dinner
20:00 –	More discussions...

Cooperative atomic motion probed by femtosecond electron diffraction

Maximilian Eichberger[1,2] and Jure Demsar[2]

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In numerous solids exhibiting broken symmetry ground states, changes in electronic (spin) structure are accompanied by structural changes. Femtosecond time-resolved techniques recently contributed many important insights into the origin of their ground states by tracking dynamics of the electronic subsystem with femtosecond light pulses [1,2]. Moreover, several studies of structural dynamics in systems with periodic lattice modulation (PLD) were performed [3,4,5]. Since intensities of the super-lattice diffraction peaks are in the first approximation proportional to the square of the PLD amplitude, their temporal dynamics provides access to cooperative atomic motion. This process takes place on a fraction of a period of the corresponding lattice vibration (typically 100 fs timescale). However, since energy transfer from the excited electronic system to the lattice takes place on a comparable timescale [1,3], contribution of the incoherent lattice motion on diffraction intensities has to be taken into account. We demonstrate an ultrafast transmission electron diffraction set-up, where relative changes in individual diffraction peaks of less than 1% can be studied. Taking a prototype two-dimensional charge density wave system 1T-TaS_2 as an example, we show, that by simultaneously tracking the dynamics of intensities in super-lattice peaks, lattice peaks and in the incoherent background over multiple diffraction orders the two processes can be effectively disentangled [6]. This approach provides direct access to the dynamics of the order parameter.

[1] H. Schaefer et al., Phys. Rev. Lett. 105, 066402 (2010); H. Schaefer et al., Phys. Rev. B 89, 045106 (2014).
[2] M. Porer et al., Nature Mat. (2014); K.W. Kim, et al., Nature Mat. 11, 497 (2012).
[3] M. Eichberger, et al., Nature 468, 799 (2010).
[4] N. Erasmus, et al., Phys. Rev. Lett. 109, 167402 (2012).
[5] T. Huber et al., Phys. Rev. Lett. 113, 026401 (2014).
[6] M. Eichberger, et al., in preparation

Ultrafast coherent phonon dynamics during the phase transition of the quasi one-dimensional Ta_2NiSe_5

Selene Mor[1], Marc Herzog[1], Claude Monney[1,2], Martin Wolf[1] and Julia Stähler[1]

[1]Fritz Haber Institut der Max Planck Gesellschaft, Faradayweg 4-6, 14195, Berlin, Germany
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Ta_2NiSe_5 is a layered compound in which atomic chains are aligned in layers, forming a quasi one-dimensional crystal structure. At 328 K, the system shows a structural change accompanied by an electronic phase transition (PT) from a low temperature (LT) (potentially excitonic) insulator to a semiconductor. Our aim is to unveil the microscopic mechanism underlying this PT by investigating the excited non-equilibrium dynamics after optical excitation. The system is excited with a femtosecond laser pulse at 800 nm and the mid-infrared (MIR) transient optical response is monitored by ultrafast optical spectroscopy. We observed a fast rise of transient reflectivity, which decays exponentially within 2-3 ps. This incoherent response is superimposed by several coherent phonon oscillations. In particular, we observe a 4 Thz phonon that is specific of the LT phase, which is confirmed by temperature dependent Raman spectroscopy. The time evolution of this LT phase phonon reveals a finite lifetime of few ps. Furthermore, a non linear behaviour is observed for both the coherent and incoherent response as a function of excitation fluence. The fluence and sample temperature dependence suggest that the PT can be driven by photoexcitation on an ultrafast timescale.

Ultrafast dynamics of a Pb overlayer structure grown on flat and vicinal Si(111)

Abdul Samad Syed[1], Vesna Mikšić Trontl[1], Manuel Ligges[1], Mathias Sandhofer[1], Ishita Agarwal[1], Isabella Avigo[1], Daniel Lückermann[2], Christoph Tegenkamp[2], Herbert Pfnür[2], and Uwe Bovensiepen[1]

[1]Fakultät für Physik und Zentrum für Nanointegration (Cenide), Universität Duisburg-Essen, 47048 Duisburg, Germany
[2]Institut für Festkörperphysik, Leibniz Universität Hannover, 30167 Hannover, Germany

Due to real space anisotropy of vicinal surfaces, hot electrons can be expected to exhibit different dynamics along and perpendicular to the steps [1] as compared to a flat surface. We made a comparative study of Pb overlayer structures grown on vicinal Si (557) and flat Si (111) using femtosecond time- and angle-resolved two-photon photoemission. We mapped the unoccupied electronic band structure near Γ (~ ±0.1Å^{-1}) and find two unoccupied states at $E-E_F$ =3.3 and 3.5 eV for the vicinal surface. We find unoccupied states in the similar binding energy region for Pb/ Si (111). The electronic structure of these bands is different, as compare to the Pb/Si (557) case potentially due to the different Pb coverage of 1/3 ML for Si (111) and 4/3 ML for Si (557). In pump-probe experiments combined with a position sensitive electron time of flight spectrometer [2] we analyze the ultrafast momentum dependent electron dynamics along two in plane directions. This allows to investigate the electronic relaxation time averaged over all accessible electron momentum which is found to be 24±5 and 35±5 fs for the states at 3.3 and 3.5 eV of Pb/Si (557) and very similar values for Pb/Si (111). Our observation is consistent with electronic structure density functional theory calculations of Pb/Si (111) [3] which find Pb derived states within an orientational band gap of Si (111) near $E-E_F$ = 4 eV. In addition on vicinal surfaces we observe a specific, momentum dependent population dynamics which is absent on the flat surface. This signature shows a delay in population buildup of about 5 fs as a function of angle with respect to the terrace direction. We assign this behavior to step-induced scattering which is absent along the step direction.

We gratefully acknowledge funding by the DFG through FOR1700. V. M.-T. acknowledges support by the Heinrich-Hertz Foundation.

[1] Roth et al., Phys. Rev. Lett. **88**, 096802 (2002).
[2] Kirchmann et al., Appl. Phys. A **91**, 211 (2008).
[3] S. Sakang, P. Kratzer, private communication.

Ultrafast sub-cycle terahertz nano-spectroscopy

M. A. Huber[1], M. Eisele[1], T. L. Cocker[1], M. Plankl[1], L. Viti[2], D. Ercolani[2], L. Sorba[2], M. S. Vitiello[2], and R. Huber [1]

[1]Universität Regensburg, Institut für Experimentelle und Angewandte Physik, Universitätsstraße 31, D-93053, Regensburg

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Many electronic properties of solids are governed by low-energy elementary excitations like plasmons, phonons and inter-level transitions in excitons. Ultrafast terahertz pulses have provided valuable insights into the dynamics of these processes [1], revealing, for example, the ultrafast build-up of screening following the injection of an electron-hole plasma by an ultrashort optical pulse [2]. However, terahertz spectroscopy is limited to a probe diameter of several micrometers or larger by diffraction. Scattering near-field scanning optical microscopy (s-NSOM) in the infrared can be used to gain further insights into the structure of low-energy excitations on their characteristic, nanometer length scales. Their behaviour in nanomaterials is of particular interest [3], as confinement may alter their properties.

Here, we demonstrate a new approach that combines the ultrafast, sub-cycle temporal resolution of time-resolved terahertz spectroscopy with the nanometer spatial resolution of s-NSOM. The novel setup allows us to trace the local ultrafast electron dynamics in indium arsenide nanowires with simultaneous 10 nm spatial and 10 fs temporal resolution [4]. After photoexcitation we observe the formation of a bulk plasma resonance in our probing spectrum. Tracing the resonance as function of pump-probe delay time allows us to extract the spatio-temporal carrier density at the surface of the nanowire. The dynamics feature a reduction of the carrier density on the picosecond scale, which agrees well with typical carrier trapping times at defect sites. More surprisingly, though, we also observe a very fast initial decay (<50 fs). To reveal the origin of this ultrafast decay we employed a novel technique we call femtosecond tomography. This approach allows us to trace the electron dynamics at different probing depths inside the material. The sub-picosecond decay was found to only happen close to the surface of the wire and can be explained by the ultrafast build-up of a surface depletion layer in the nanowire.

Our novel microscope should find further applications in a broad range of subjects: from carrier transport in nanostructures to transient local dynamics in chemical composites.

[1] R. Ulbricht *et al.*, Rev. Mod. Phys. **83**, 543 (2011).
[2] R. Huber *et al.*, Nature **414**, 286 (2001).
[3] J. M. Stiegler *et al.*, Nano Lett. **10**, 1387 (2010)
[4] M. Eisele *et al.*, Nature Photon. **8**, 841 (2014).

Ultrafast electronic excitations at the buried GaP/Si interface

Alexander Lerch, Kristina Brixius, Andreas Beyer
Kerstin Volz, Wolfgang Stolz and Ulrich Höfer

Philipps Universität Marburg, Fachbereich Physik und Zentrum für Materialwissenschaften, Renthof 5, 35032 Marburg, Germany

We investigated ultrafast electronic excitations and charge transfer processes at the epitaxially grown GaP on Si(001) interface by means of optical second-harmonic generation (SHG). 1.55-eV 45-fs pump pulses were used to excite the GaP/Si heterostructure. The SHG was detected in a non-collinear geometry. We observed pronounced SHG transients which are absent in the corresponding bulk materials. A fast initial increase of the SHG which decays in approximately 500 fs is followed by a slow rise on the time scale of several picoseconds. From the linear dependence of the transients on the pump fluence we exclude direct excitation of both GaP and Si bulk with direct band gaps of 2.8 eV and 3.4 eV, respectively. Instead we propose a direct excitation of electronic states at the interface by the 1.55-eV pump pulses which is consistent with an initial rise-time of the SHG signal faster than 30 fs.
A charge transfer of the excited carriers across the interface could build up an electric field on a picosecond time scale which is in agreement with the approximately 3-ps rise-time of the second transient component. The influence of the lattice temperature on the dynamics via electron-phonon scattering is discussed and confirms this assignment. Additionally, the interference between the transients and the static GaP bulk contribution was utilized to discriminate different interface components phase sensitively.

Second Harmonic Spectroscopy in the Reststrahlen Band of 6H-SiC

Alex Paarmann, Alexey Melnikov, Ilya Razdolski, Sandy Gewinner,
Wieland Schöllkopf, and Martin Wolf

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We experimentally study the spectral dependence of efficiency for optical second harmonic generation in the highly reflective Reststrahlen spectral region of the wide-band gap semiconductor 6H-SiC. Employing tunable picosecond mid-infrared laser pulses generated from the FHI free electron laser [1], we have developed a novel approach for spectrally resolved, mid-infrared second harmonic spectroscopy.
The Reststrahlen region [2], located between longitudinal and transversal optical phonon resonances, provides a unique window to study the nonlinear response of optical phonons in a crystal. We observe sharp resonances of the second harmonic signal at both transverse and longitudinal optical phonon fundamental frequencies, the latter being unexpected since no resonance in the linear and nonlinear optical response is expected for longitudinal optical modes. We discuss the underlying mechanism of Fresnel transmission resonances, which is tightly linked to the largely dispersing linear optical properties in the Reststrahlen band.
The negative real part of the dielectric function results in rapidly attenuated evanescent waves in this region, making these experiments highly sensitive to a thin near-surface layer. The forthcoming application of our approach to study field enhancements from localized surface phonon polariton resonances [3] is discussed, possibly opening up a new route to strongly drive optical phonon motion.

[1] W. Schöllkopf, S. Gewinner, W. Erlebach, H. Junkes, A. Liedke, G. Meijer, A. Paarmann, G. Von Helden, H. Bluem, D. Dowell, R. Lange, J. Rathke, A. M. M. Todd, L. M. Young, U. Lehnert, P. Michel, W. Seidel, R. Wünsch, and S. C. Gottschalk, Proc. FEL WEB04 (2014).

[2] S. Adachi, in *Opt. Prop. Cryst. Amorph. Semicond.* (Springer US, Boston, MA, 1999).

[3] J. J. D. Caldwell, O. O. J. Glembocki, Y. Francescato, N. Sharac, V. Giannini, F. J. Bezares, J. P. Long, C. Owrutsky, I. Vurgaftman, J. G. Tischler, V. D. Wheeler, N. D. Bassim, L. M. Shirey, R. Kasica, and S. A. Maier, Nano Lett. **13**, 3690 (2013).

Tuesday, March 3

7:30 – 8:30 Breakfast

Morning session (Chair: Jure Demsar)

8:30 – 9:15 **Sascha Schäfer** (Georg-August-Universität, Göttingen)
Ultrafast electron imaging and diffraction with nanoscale photoemitters

9:15 – 9:30 Discussion

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Hot electron dynamics at 2H-MoS_2 surfaces: Time- and angle-resolved photoelectron spectroscopy results

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trARPES with bright narrowband HHG pulses using frequency-doubled Ti:Sapphire lasers

19:00 – 20:00 Dinner

20:00 – More discussions...

Ultrafast electron imaging and diffraction with nanoscale photoemitters

Armin Feist, Max Gulde, Reiner Bormann, Simon Schweda, Gero Storeck,
Katharina Echternkamp, Sascha Schäfer, Claus Ropers

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The utilization of ultrashort electron pulses in laser-pump/electron-probe experiments allows for the investigation of structural dynamics on ultrafast timescales [1,2]. A crucial element in these studies, and often a limiting factor, is the performance of the pulsed electron source, both, in terms of temporal pulse width and electron beam brightness and emittance.
We will present our current experimental developments regarding the application of nanometric photocathodes for ultrafast electron imaging and diffraction experiments. Specifically, we employ sharp laser-triggered tungsten needle-emitters for ultrafast low-energy electron diffraction (ULEED) and ultrafast transmission electron microscopy (UTEM). In both cases, the enhanced optical field at the tip apex confines nonlinear photoemission to small emitter areas [3,4], yielding spatially well defined pulsed electron beams.
Electron pulses at low energies are ideally suited for the study of structural dynamics of surfaces and ultrathin films, and we demonstrate a first application of ULEED for the investigation of the laser-induced melting of a polymer superstructure on single-layer graphene [5].
In UTEM, the small effective source size of needle photocathodes enables electron focal spot sizes on the sample of down to 3 nm with electron pulse durations of about 350 fs.
With such tightly focused electron probes, we study in detail inelastic photo-induced scattering of electrons in an optically driven near-field [6]. Employing the nanoscale probing capabilities of our instrument, we unambiguously demonstrate the quantum coherent nature of this interaction, suggesting future applications in coherent control of electron pulses.
Finally, based on the present electron beam properties, we discuss future research directions of the Göttingen UTEM and ULEED for time-resolved electron imaging, diffraction and spectroscopy.

[1] R. J. D. Miller *et al.* `Making the molecular movie': first frames. *Acta Crystallogr., Sect. A* **66,** 137–156 (2010).
[2] A. H. Zewail, Four-dimensional electron microscopy. *Science* **328,** 187–93 (2010).
[3] C. Ropers, D. R. Solli, C. P. Schulz, C. Lienau, T. Elsaesser, Localized Multiphoton Emission of Femtosecond Electron Pulses from Metal Nanotips, Phys. Rev. Lett. 98, 043907 (2007).
[4] P. Hommelhoff, C. Kealhofer, M. A. Kasevich, Ultrafast Electron Pulses from a Tungsten Tip Triggered by Low-Power Femtosecond Laser Pulses, Phys. Rev. Lett. 97, 247402 (2006).
[5] M. Gulde *et al.* Ultrafast low-energy electron diffraction in transmission resolves polymer/graphene superstructure dynamics. *Science.* **345,** 200–204 (2014).
[6] A. Feist *et al.* Quantum coherent optical phase modulation in an ultrafast transmission electron microscope, submitted.

Hot electron dynamics at 2*H*-MoS_2 surfaces: Time- and angle-resolved photoelectron spectroscopy results

Petra Hein, Ankatrin Stange, Kerstin Hanff, Gerald Rohde,
Michael Bauer, and Kai Rossnagel

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Recent research on the layered semiconductor 2*H*-MoS_2 has mainly been concentrated on MoS_2 monolayers: In contrast to the bulk crystal, MoS_2 monolayers are direct bandgap semiconductors without an inversion center [1], making them promising candidates for valleytronic devices [2-4] with hot carrier lifetimes in the picosecond regime [5]. However, 2*H*-MoS_2 surfaces could be equally exciting: Due to a possible decoupling of the topmost sandwich, similarities to monolayers seem likely. For an investigation of this system, time- and angle-resolved photoelectron spectroscopy is the method of choice, as it provides high surface sensitivity and momentum resolution, while at the same time enabling us to map electronic processes on their fundamental time scales.
Here, we present a momentum-resolved study of the transient photoelectron intensity in the conduction band after excitation of 2*H*-MoS_2 surfaces with 3.2 eV pump pulses. Momentum-dependent intensity rise times between 30 fs and 150 fs and decay times in the order of several 100 fs allow us to identify direct electronic excitations as well as to track the electrons' relaxation pathways into the lowest energy states. These conduction band minima are finally depopulated within a few picoseconds. Possible explanations for this latter observation – such as diffusion processes, optical electron-hole recombination and trapping in defect states – are discussed.

[1] Kin Fai Mak *et al*., Physical Review Letters **105**, 136805 (2010)
[2] Ting Cao *et al*., Nature Communications **3**, 887 (2012)
[3] Hualing Zeng *et al*., Nature Nanotechnology **7**, 490 (2012)
[4] Kin Fai Mak *et al*., Nature Nanotechnology **7**, 494 (2012)
[5] Hongyan Shi *et al*., ACS Nano, Vol. **7**, No. 2, 1072 (2013)

High-order harmonic generation by dynamical Bloch oscillations in a bulk solid

Matthias Hohenleutner[1], Fabian Langer[1], Olaf Schubert[1], Christoph Lange[1], Ulrich Huttner[2], Stephan W. Koch[2], Mackillo Kira[2] and Rupert Huber[1]

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Bloch oscillations are one of the most fascinating quantum manifestations of electrons in crystalline solids. When a d.c. electric bias accelerates an electron, its de Broglie wavelength shortens. Once the latter equals twice the lattice constant, the wave should undergo Bragg reflection, causing electrons to oscillate in reciprocal and real space [1]. The experimental observation of this long-standing prediction has been challenging due to ultrafast scattering and dielectric breakdown under d.c. bias conditions. Recently, high-order harmonic (HH) generation in solids has been attributed to a dynamical version of Bloch oscillations [2]. Precise field control, however, has been out of reach due to the fluctuating carrier-envelope phase (CEP) of the laser pulses. Latest developments in field resolved multi-terahertz optics provide CEP-stable waveforms, which serve as a sub-cycle bias for high-field experiments [3]. Here, we employ phase-locked electromagnetic waveforms with peak fields as high as 72 MV/cm and adjustable carrier-envelope phase to control all-coherent charge transport in gallium selenide (GaSe) on femtosecond timescales [4]. The dynamics encompass the generation of phase-stable high-harmonics, spanning the entire terahertz-to-visible spectral domain between 0.1 and 675 THz in a single waveform. While HH generation in atoms is well explained by the semiclassical three-step model including ionization, acceleration and recollision [5], the quantum-mechanical wave nature of electrons dominates the physics of high-field transport in solids: As corroborated by our quantum mechanical five-band model, an intriguing interplay of coherent interband polarization and intraband transport in the regime of dynamical Bloch oscillations underlies the HH generation mechanism. A novel quantum interference of different ionization pathways allows for sensitive control of the emitted harmonics via the CEP of the driving waveform.

[1] C. Zener, Proceedings of the Royal Society A **145**, 523–529 (1934).
[2] S. Ghimire et al., Nature Physics **7**, 138–141 (2010).
[3] A. Sell et al., Optics Letters **33**, 2767–2769 (2008).
[4] O. Schubert et al., Nature Photonics **8**, 119-123 (2014).
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Time-resolved XUV ARPES at sub-15 fs temporal resolution

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Time- and angle-resolved photoelectron spectroscopy (trARPES) provides a most direct access to ultrafast electronic processes in condensed matter systems. Application of XUV photon pulses generated in High-Harmonic Generation (HHG) light sources allows probing the transient electronic structure within the entire Brillouin zone or even beyond [1]. The effective time-resolution of these types of experiments is given by the cross-correlation signal between IR or visible pump and XUV probe pulse. The pump pulse typically limits the time-resolution to values of ≥ 30 fs so that extremely fast phenomena such as characteristic melting times of electronic phases [2] or the thermalization of a nascent electron distribution [3] may remain unresolved.

Here we present a HHG-based trARPES setup exhibiting a substantially improved time-resolution. The optical pump-line of the experiment is operated using sub-6 fs pulses at a center wavelength of about 800 nm as generated in an amplifier-driven hollow-fiber chirped-mirror compressor ("white light source"). In the contribution, details of the source optimization procedure and its integration into the existing trARPES setup are described. First trARPES measurements on 1*T*-$TiSe_2$ and on HOPG are shown providing direct experimental evidence for a sub-15 fs time-resolution. Furthermore, the results from HOPG indicate that a nonthermal distribution is monitored on its way towards a Fermi-Dirac distribution.

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Double photoemission spectroscopy on correlated *d* electrons in solids using a megahertz high-order harmonic light source

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Photoelectron spectroscopy (PES) has provided immense insight into the electronic structure of materials. In PES, the energy dispersion of individual photoelectrons as a function of their momentum and spin orientation represents the ultimate information that could be unravelled. Under such experimental conditions, signatures of the correlation between electrons can only be indirectly retrieved. In strong contrast, double photoemission (DPE) experiments show promising potential to provide direct spectroscopic information on the electron correlation [1]. In DPE spectroscopy, a pair of photoelectrons is emitted after the excitation by one single photon, and the energy as well as the momentum of these two photoelectrons can be analyzed [2].

In this contribution we present laboratory DPE experiments on Ag(001) and NiO films using a high-order harmonic light source at megahertz repetition rates [3]. The light source is driven by a compact Yb-fiber laser system at 0.7 MHz and delivers a photon energy ranging from 14 to 40 eV [4]. From the two-dimensional energy distribution of the photoelectron pairs from a Ag(001) surface excited by a photon energy of 25 eV, we obtain the DPE spectrum as a function of the sum energy of the pairs (E_{sum}). At E_{sum}=7.5 eV, we identify features that can be attributed to emission of two *d* electrons. At a higher energy of E_{sum}=12.1 eV, a feature is assigned to emission of one electron from the *d* and the other from the *sp* band. For comparison, DPE from NiO films shows mainly features due to emission of two *d* electrons from the valence band. Our observation indicates the dominant role of *d* electrons in the DPE process and implies their general importance for electron correlation in solids ranging from metals to oxides.

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*tr*ARPES with bright narrowband HHG pulses using frequency-doubled Ti:Sapphire lasers

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Time- and angle-resolved photoemission spectroscopy (*tr*ARPES) using femtosecond extreme ultraviolet high harmonics has recently emerged as a powerful tool for investigating ultrafast quasiparticle dynamics in correlated-electron materials. However, the full potential of this approach has not yet been achieved because, to date, high harmonics generated by 800 nm wavelength Ti:Sapphire lasers required a trade-off between photon flux, energy and time resolution. Photoemission spectroscopy requires a quasi-monochromatic output, but dispersive optical elements that select a single harmonic can significantly reduce the photon flux and time resolution. In our work, we show that 400 nm driven high harmonic extreme-ultraviolet *tr*ARPES is superior to using 800 nm laser drivers since it eliminates the need for any spectral selection, thereby increasing photon flux and energy resolution to <150 meV while preserving excellent time resolution of about 30 fs [1].
Using this new capability, I will discuss recent results in the non-equilibrium dynamics of the charge-density wave material $TiSe_2$.

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Wednesday, March 4

7:30 – 8:30 Breakfast

Morning session (Chair: Uwe Bovensiepen)

8:30 – 9:15 **Michael Horn-von Hoegen** (Universität Duisburg-Essen)
Electron Diffraction at Surfaces: by now Ultrafast!

9:15 – 9:30 Discussion

9:30 – 10:00 **Thomas Vasileiadis** (Fritz-Haber-Institut der MPG, Berlin)
The effect of confinement in the electron-phonon coupling studied with ultrafast electron diffraction

10:00 – 15:30 Outdoor workshop

Afternoon session (Chair: Ralph Ernstorfer)

16:00 – 16:30 **Uwe Bovensiepen** (Universität Duisburg-Essen)
Competition of spin currents and spin-flip scattering in ultrafast magnetization dynamics of Co/Cu(001) analyzed by the complex time-resolved MOKE

16:30 – 17:00 **Alexey Melnikov** (Fritz-Haber-Institut der MPG, Berlin)
Direct monitoring of hot carrier-formed spin currents in metallic multi-layers by time-resolved magneto-induced second harmonic generation

17:00 – 17:30 Coffee break

17:30 – 18:00 **Moritz Barkowski** (Universiät Kaiserslautern)
Depth-dependent study of femtosecond spin-currents in Ni/Au

18:00 – 18:30 **Kamil Bobowski** (Freie Universität Berlin)
Ultrafast magnetization dynamics of Gadolinium measured by x-ray magnetic circular dichroism in reflection geometry

18:30 – 19:00 **Beatrice Andres** (Freie Universität Berlin)
Ultrafast magnetization dynamics in Gadolinium – Analyzing the transient spin polarization

19:00 – 20:00 Dinner

20:00 – More discussions...

Electron Diffraction at Surfaces: by now Ultrafast!

Michael Horn-von Hoegen

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Ultrafast structural dynamics at surfaces can be studied by electron diffraction using a combination of a pulsed electron gun with a fs laser system in a pump probe scheme under ultra-high vacuum conditions. The grazing incidence of electrons of 7 – 30 keV in reflection high energy electron diffraction (RHEED) geometry ensures high surface sensitivity. The sample is excited with 800 nm photons with pulses of 80 fs length, an energy of 1 mJ at 5 kHz repetition rate and a fluence of 1 – 8 mJ/cm2. Applying a tilted pulse front scheme for the laser pulses the temporal resolution of the entire setup has recently been improved to less than 400 femtoseconds [1-3]!
The huge potential of this technique is demonstrated with the non-equilibrium dynamics of the In induced (8x2) reconstruction on Si(111). This surface exhibits a Peierls-like phase transition at 130 K from a (8x2) ground state, which is accompanied by the formation of a charge density wave (CDW), to a (4x1) excited state. Upon excitation by the fs-laser pulse the (8x2) groundstate is driven into the excited (4x1) state at a sample temperature of 30 K. The surface is only excited electronically, the structural transition occurs in 350 fs, but the surface remains for almost one nanosecond in a super-cooled excited (4x1) high temperature state. An activation barrier of ~40 meV for the collective motion of the In atoms hinders the immediate recovery of the (8x2) groundstate. Such metastable situation – a hidden state of matter far away from equilibrium – is only accessible through the ultra-fast excitation by the fs-laser pulse. Relaxation to the (8x2) ground state is delayed on a timescale of ~ 500 picoseconds and is triggered by (8x2) remnants pinned at adsorbates that act as nucleation seeds – the same way that super-cooled water in a bottle freezes upon the insertion of seeds. The surface unit cells fall back into their ground state, one at a time, like a row of falling dominoes [4]. From a transient spot profile analysis of the width of the (8x2) spots we can determine the speed of the phase front which propagates at about 100 m/s.
The initial structural transition at a time scale of 350 fs is followed by a heating of the surface atoms on a time scale of 3.5 ps. The thermal motion of the indium and silicon atoms is observed by means of the Debye-Waller effect which causes a reduction of intensity of the diffraction spots. On a time scale of 20 ps this thermal motion has dissipated into the Si substrate as evident from the recovery of the intensity.

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The effect of confinement in the electron-phonon coupling studied with ultrafast electron diffraction

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In this talk we will present preliminary results regarding the effect of confinement in the electron-phonon coupling constant. Our approach consists of studying size-selected Au nanoclusters, the so-called ´magic clusters´, with ultrafast electron diffraction. Size-selection is based on the increased stability of these structures from both geometrical and electronic considerations [1]. Ultrafast diffraction provides the most direct way to observe the response of the lattice to electronic excitations caused by femtosecond laser pulses. Especially electron diffraction is an ideal tool for nanostructures because of the much higher scattering cross-section in comparison with X-rays. The lattice temperature is probed by the decrease of intensity of the diffraction peaks due to the Debye-Waller effect. Our first measurements are focused on Au clusters with 923 atoms that are located at the borders of bulk and nano regimes based on ultrafast optical measurements [2]. A strong enhancement of the kinetics of heat transfer from the electrons to the lattice is observed which indicates an enhancement of electron-phonon interaction in confined systems. Furthermore, from the position of diffraction peaks ultrafast expansion of the nanoparticles is probed with subpicometer resolution. As expected, coupling of the nanoclusters with the substrate has important effects but only in longer timescales. In conclusion our present findings suggest more rapid heat-transfer from the electronic to the lattice subsystem and demonstrate the ability of ultrafast diffraction to study photo-induced changes in nanostructured systems with subpicometer sensitivity in structural changes.

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Competition of spin currents and spin-flip scattering in ultrafast magnetization dynamics of Co/Cu(001) analyzed by the complex time-resolved MOKE

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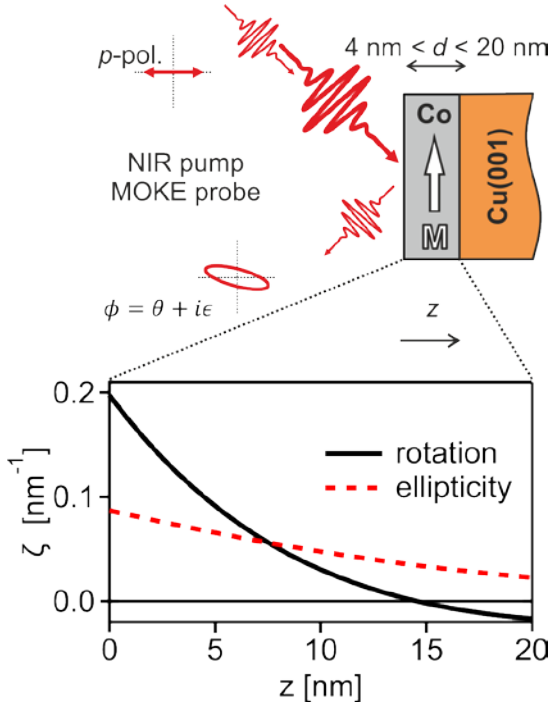
The ultrafast response of transition metal ferromagnets to femtosecond laser excitation is governed by spin-dependent electronic excitations, their propagation dynamics, as well as coupling to magnons and to the lattice [1-4]. To understand these individual elementary processes with characteristic time scales in the femtosecond regime, an experimental tool which is capable to disentangle the respective contributions is highly desired.



Here we demonstrate one step in this direction by analyzing *in situ* under ultra-high vacuum the spatio-temporal correlation of ultrafast time-dependent magnetization profiles $M(z,t)$ for epitaxial Co/Cu(001) films. Optical excitation of the Co-Cu interface leads to spin transport due to the large gradient in spin polarization across the interface. Thereby a spin current is generated which continues while electrons remain excited [2,4]. In the bulk part of the film and near the Co-vacuum interface the magnetization dynamics is dominated by the optical excitation density following the microscopic three temperature model [1] and its extensions considering gradients in the transient electron temperature.

In this study we analyze the ultrafast changes of the polarization rotation and ellipticity in the magneto-optical Kerr effect as a function of pump-probe delay t. By exploiting the effective depth sensitivity of the method, see the figure for sensitivity $\zeta(z)$, we monitor $M(z,t)$ as a function of film thickness d from 4 to 20 nm, which facilitates to disentangle transport and local effects. We find that before hot electron thermalization for $t < 100$ fs $M(z,t)$ is governed by spin-dependent transport effects, while at later delays, after hot electron thermalization ($t > 200$ fs), local spin-flip processes dominate.

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Direct monitoring of hot carrier-formed spin currents in metallic multi-layers by time-resolved magneto-induced second harmonic generation

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Within last years, superdiffusive transport of laser-induced hot carriers (HC) occurring in highly excited states of matter (which is especially important in the case of metals where a large amount of highly mobile HC can be excited in a few nm-thick layer), is extensively studied both experimentally and theoretically. Great interest to this type of transport is stimulated in particular by its potential use in ultrafast spintronics: being excited in the exchange-split band structure of a ferromagnet, HC may become spin-polarized and provide an ultrafast spin transport owing to different inelastic mean free paths of majority and minority HC. This superdiffusive spin transport [1] is also considered as an important contribution to the ultrafast magnetization dynamics, which defines the laser-induced demagnetization of ferromagnetic metals at the initial (first couple of hundreds femtoseconds) stage of the process. Utilizing the surface sensitivity of magneto-induced second harmonic generation (mSHG), we have demonstrated the HC spin transport in Au/Fe/MgO(001) bi-layers [2]. The SH field consists of two components, $\boldsymbol{E_{even}}$ and $\boldsymbol{E_{odd}}$, which are even and odd with respect to the magnetization reversal, and our interpretation of nonlinear-optical data was based on the assumption that the variations of $\boldsymbol{E_{even}}$ and $\boldsymbol{E_{odd}}$ are proportional to the variation of the HC density and transient magnetization at the surface of Au, respectively. Here we perform more detailed investigations and show that $\boldsymbol{E_{even}}$ and $\boldsymbol{E_{odd}}$ also contain sizable terms proportional respectively to superdiffusive electric and spin currents in the bulk of metallic layers. Analyzing the response of Au/Fe/MgO(001) and Fe/Au/Fe/MgO(001) structures, we observe contributions from both charge/particle and spin components of HC currents within 500 fs after the excitation, which can be experimentally separated from interface contributions of transient HC density and magnetization, respectively. Thus, we obtain a direct access to laser-induced superdiffusive spin currents which we can monitor with 20 fs time resolution. The underlying mechanisms will be discussed along with related details and perspectives of mSHG diagnostics. DFG (ME 3570/1, Sfb 616) and EU 7-th framework program (CRONOS) are acknowledged.

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Depth-dependent study of femtosecond spin-currents in Ni/Au

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We use depth-resolved femtosecond magneto-optical Kerr effect [1,2] to study ultrafast magnetization dynamics in Ni/Au. After impulsive photo-excitation, we see the wellknown rapid demagnetization in the Ni-layer (<100 fs), which is accompanied by the appearance of Kerr signal in the underlying Au layer [3].
A detailed analysis of the very different time-resolved rotation and ellipticity signals shows that these consist of weighted sums of the femtosecond magnetic response in the individual layers. Therefore, different transient signals (demagnetization times) of rotation vs. ellipticity do most often *not* indicate so-called optical artifacts [4], but must be expected and are fingerprints of complex magnetization dynamics by spin-currents and spin-scattering mechanisms in multilayer structures [5].

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Ultrafast magnetization dynamics of Gadolinium measured by x-ray magnetic circular dichroism in reflection geometry

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The ferromagnetic character of the lanthanide metal Gadolinium is determined by the magnetic moments of the localized 4*f* electrons. The exchange coupling between neighboring 4*f* magnetic moments is mediated by polarizing the 5*d*6*s* valence electrons, which is described with the RKKY interaction. Optical excitation of the valence electrons leads to a demagnetization of gadolinium, whose dynamics on the ultrafast timescale give rise to the question whether the demagnetization times for the 5*d*6*s* valence electrons and the localized 4*f* electrons are coupled.
Former studies combining time-resolved X-ray circular magnetic dichroism (XMCD) sensitive to the unoccupied 4*f* moments [1] and magneto-optical Kerr effect (MOKE) which probes the 5*d*6*s* valence electrons [2] show identical demagnetization times for both subsystems. This suggests a strong intra-atomic coupling that keeps the 4*f* and 5*d*6*s* moments aligned.
However, photoemission studies using high order harmonic generation (HHG) at 36 eV show a clear difference in the transient behavior of the 5*d*6*s* exchange splitting and the magnetic linear dichroism in the occupied 4*f* states upon laser excitation [3, 4].
In my talk I will present recent results of our experiments at the FEMTOSPEX beamline at BESSY II, which resolve possible explanations for the discrepancy of the XMCD and HHG data. The measurements were performed on a single-crystalline Gadolinium thin film using XMCD in reflection geometry to achieve a better comparability to the HHG measurements.

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Ultrafast magnetization dynamics in Gadolinium – Analyzing the transient spin polarization

Beatrice Andres[1], Marko Wietstruk[1], Cornelius Gahl[1], Jürgen Kirschner[2], and Martin Weinelt[1]

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As shown in the talk of Kamil Bobowski, many experiments have been carried out on the laser-induced ultrafast demagnetization of Gadolinium. All of them are by some means spin-dependent but so far no experiment has been performed with actual spin resolution. That is the gap we aim to fill.
We set up a spin- and time-resolved photoemission experiment capable of working with an amplified Ti:Sa laser system that usually suffers from repetition rates too low for spin resolution. Using 4 mJ/cm² of the fundamental (1.5 eV) to demagnetize Gd/W(110) and the 4th harmonic (6 eV) for direct photoemission, we measure the spin polarization and binding energy of the majority surface-state.
In my talk, I show that the spin polarization of the surface state remains constant for several tens of picoseconds. This is surprising because preliminary works lead to the conclusion that the surface state's spin polarization breaks down immediately within the duration of the pump pulse [1]. Unlike the stable spin polarization, the binding energy of the surface state shifts in accordance with the exchange splitting of the 5*d* bands in Ref [2]. This is in clear contrast to our results on thermal heating where the surface state's spin polarization decreases, while the exchange splitting remains finite even above the Curie temperature.

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Thursday, March 5

7:30 – 8:30 Breakfast

Morning session (Chair: Michael Horn-von Hoegen)

8:30 – 9:15 **Ralph Ernstorfer** (Fritz-Haber-Institut der MPG, Berlin)
Femtosecond electrons probing structural dynamics and ultrafast currents

9:15 – 9:30 Discussion

9:30 – 10:00 **Konrad Gillmeister** (Martin-Luther-Universität Halle-Wittenberg)
Electron dynamics in NiO ultrathin films

10:00 – 15:30 Outdoor workshop

Afternoon session (Chair: Alex Paarmann)

16:00 – 16:30 **Ishita Agarwal** (Universität Duisburg-Essen)
Femtosecond time-resolved two-photon photoemission spectroscopy of solavted electrons in $C_6H_5F/D_2O/Cu(111)$

16:30 – 17:00 **Harald Kirsch** (Fritz-Haber-Institut der MPG, Berlin)
Probing Water Dissociation and Structure at the α-Al_2O_3(0001) Surface Using Interface Specific Vibrational Dynamics

17:00 – 17:30 Coffee break

17:30 – 18:00 **Wibke Bronsch** (Freie Universität Berlin)
Ultrafast Exciton Dynamics in Thin Sexithiophene Films

18:00 – 18:30 **Jens Güdde** (Phillips-Universität Marburg)
Electron dynamics in the topological insulator Sb_2Te_3

18:00 – 18:30 **Martin Weinelt** (Freie Universität Berlin)
Electronic structure and dynamics of the topological insulator Sb_2Te_2S

19:00 – 20:00 Dinner

20:00 – More discussions...

Femtosecond electrons probing structural dynamics and ultrafast currents

Lutz Waldecker, Melanie Müller, Roman Bertoni, Thomas Vasileiadis, Alex Paarmann, and Ralph Ernstorfer

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The recent development of femtosecond electron and x-ray techniques for diffraction and imaging allows for the direct observation of structural dynamics in the course of photo-induced chemical or physical processes with atomic spatial and femtosecond temporal resolution. We investigate ultrafast structural as well as electronic dynamics in bulk materials, two-dimensional semiconductors, one-dimensional nanowires, and nanoparticles. Such studies require femtosecond probes strongly interacting with small-volume samples. We employ two experimental approaches employing sub-keV single-electron pulses and 100 keV electron bunches, respectively.

Electrons with energies ranging from 50 to 1000 eV exhibit extremely large scattering cross sections and high sensitivity to electric fields, but their pronounced dispersion during propagation in vacuum [1] so far prevented their use as femtosecond probe pulses in time-resolved experiments. Employing a laser-triggered point-like source of either divergent or collimated electron wave packets, we developed a hybrid approach for femtosecond point projection microscopy (fsPPM) and femtosecond low-energy electron diffraction (fsLEED) [2]. We investigate ultrafast electric currents in nanowires with sub-100 femtosecond temporal and few 10 nm spatial resolutions and demonstrate the potential of our approach for studying structural dynamics in crystalline single-layer materials.

The investigation of structural dynamics in thin film samples requires higher energy electrons. We developed a highly compact femtosecond electron diffractometer which allows for delivering 100 fs long pulses containing up to 5000 electrons at 100 keV to the sample [3]. We investigate the photo-induced structural dynamics in $Ge_2Sb_2Te_5$ (GST), a popular phase change material exhibiting two metastable crystalline states which can be switched by light or current pulses. We observe distinct differences between the dynamics of optical properties and lattice which we explain in terms of the resonant bonding present in these phase change materials [4]. Finally, we will briefly discuss recent results on the electron-phonon coupling in the semiconducting transition metal dichalcogenide material WSe_2.

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Electron dynamics in NiO ultrathin films

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The transition metal oxide NiO is not only a model system for a strongly correlated electron system but also known as a charge-transfer insulator. Although the electronic structure of NiO has been extensively studied over the last decades not much is known about the unoccupied oxide states and the electron dynamics. In this talk we report on the unoccupied Ni 3*d* states near the charge-transfer gap. For the investigation, ultrathin well-ordered NiO films from 1–20 monolayers (ML) thickness have been prepared on Ag(001) via molecular beam epitaxy.
With the use of angle-resolved two-photon photoemission spectroscopy (2PPE) it was possible to characterize all occuring unoccupied states. While interface effects dominate the electronic structure for films up to 3 ML, films of 4 ML and beyond converge to a bulk-like behavior. Time-resolved 2PPE yields lifetimes of less than 15 fs for all unoccupied Ni 3*d* states. This surprisingly ultrafast relaxation of conduction band electrons in the presence of a substancial charge-transfer band gap of about 3.8 eV can be explained by an efficient electronic decay into different many-electron states. The ultrafast dynamics seems to be a manifestation of the strong electron-electron correlation of NiO.

Femtosecond time-resolved two-photon photoemission spectroscopy of solvated electrons in $C_6H_5F/D_2O/Cu(111)$

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Solvated electrons are known to be responsible for many chemical reactions but their transfer dynamics to the reaction site are poorly known. In order to study these charge transfer dynamics (eg. Fig.1a), we performed time-resolved two-photon photoemission experiments on $C_6H_5X/D_2O/Cu(111)$, X= F and Cl. We report here the observed energies, workfunction changes and lifetimes of excited electronic states in amorphous $D_2O/Cu(111)$ and coadsorbed C_6H_5F systems after optical excitation with 3.8 eV pump pulse and 1.9 eV probe pulse. We observe a decrease in lifetime of the solvated electron state from 90±5 fs for bare D_2O to 60±5 fs for coadsorbed C_6H_5F. Further, after a total estimated exposure of $0.5x10^6$ photons per C_6H_5F molecule, a workfunction change of 200±50 meV was observed, as shown in Fig. 1b. Due to the absence of photo-induced workfunction changes in the single stack $D_2O/Cu(111)$ and $C_6H_5F/Cu(111)$, this change is concluded to be mediated by solvated electrons. Additional experiments on C_6H_5Cl were performed and an even larger photo induced workfunction change was observed. The increase in workfunction is explained by the formation of F^-/Cl^- as a photo-chemical dissociation product [1,2]. We conclude to have identified the charge transfer to C_6H_5F/Cl and a fingerprint of its photoinduced dissociation. Future experiments aim to systematically change the UV photon energy and the molecule acceptor level.

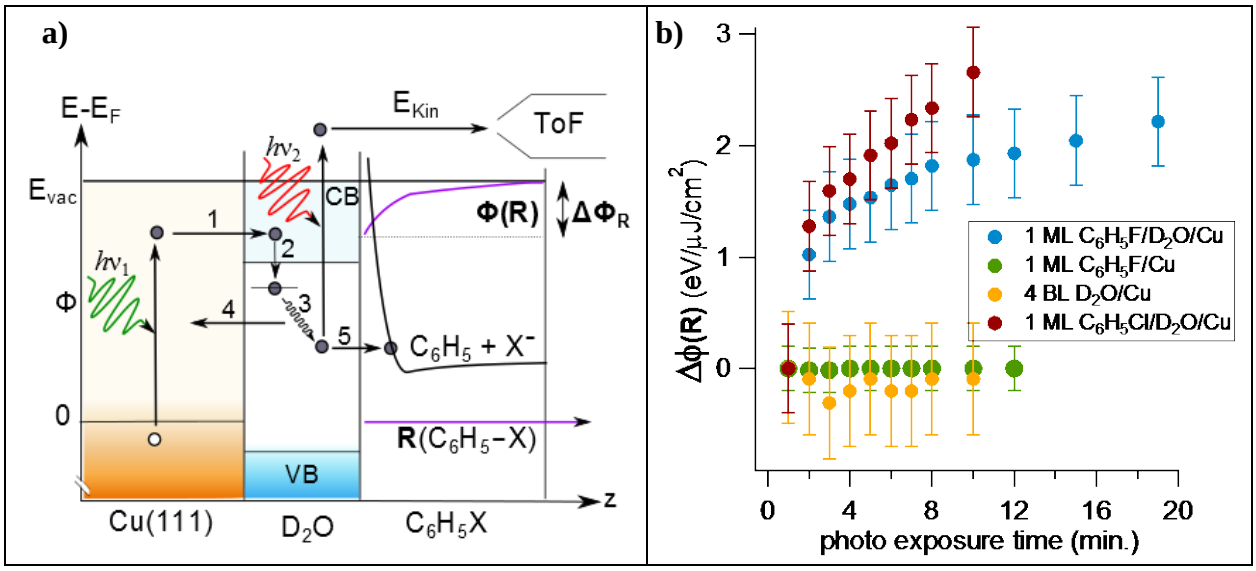

Fig. 1a) Charge transfer scheme in $C_6H_5X/D_2O/Cu(111)$, steps 1-4 depict the solvation process and step 5 shows the induced dissociation of the C_6H_5X molecule. The dissociated X^- is seen as an increase in the workfunction $\phi(R)$;
b) Temporal change in workfunction on photo illumination for different adsorbates on Cu(111).

[1] S. Ryu, et al., J. Am. Chem. Soc. **128**, 3500 (2006)
[2] M. Bertin, et al., Faraday Discuss. **141**, 293, (2009)

Probing Water Dissociation and Structure at the α-Al2O3(0001) Surface Using Interface Specific Vibrational Dynamics

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α-Al_2O_3(0001) surfaces exposed to water are ubiquitous in engineered applications and a useful model for more complicated alumino-silicate surface/water interaction omnipresent in the environment. Despite this ubiquity, gaining a molecular level understanding of water α- Al_2O_3(0001) interaction has proven challenging. It has recently been proposed, based partly on simulation, that the first layer of water on this surface is essentially hydrophobic: while hydrogen bonding occurs within this first layer, neither surface aluminols nor the first layer of molecular water donate a hydrogen bond to overlying bulk liquid. Direct experimental evidence for such a non-wetted water layer has thus far proven elusive. Here we probe the existence of this layer using interface specific vibrational relaxation as a probe of surface water structure. We find, using an ultrafast time resolved infrared pump - vibrational sum frequency probe scheme, that the T_1 of OH groups at the α-Al_2O_3(0001) surface is $\approx$ 5 ps: intermediate between that of surface aluminols in contact with excess water (< 1 ps) and that of isolated surface OH groups on oxide surfaces (> 100 ps). This intermediate T_1 is the first direct experimental insight consistent with a non-wetted interfacial water layer on the α-Al_2O_3(0001) surface.

Ultrafast Exciton Dynamics in Thin Sexithiophene Films

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Sexithiophene (6T) is one of the model systems for organic semiconductors. Therefore we are interested in the details of exciton formation and decay in thin 6T films [1]. As it is known the S1 absorption band of sexithiophene strongly depends on the mutual orientation of the molecules [2]. This allowed us to study the coverage-dependent orientation of 6T on the Au(111) surface by means of UV-vis spectroscopy. Following up these results we investigated the influence of the excitation energy on the exciton dynamics in this system by means of two-photon-photoemission spectroscopy.

[1] Varene et al. , Phys. Rev. Lett. **109**, 207106 (2012).
[2] Egelhaaf et al., Synthetic Metals **61**, 143 (1993).

Electron dynamics in the topological insulator Sb_2Te_3

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We present experimental results on the electron dynamics in the p-doped topological insulator Sb_2Te_3 using time- and angle-resolved two-photon photoemission (2PPE).
We have investigated the excitation and decay dynamics of electrons in the initially unoccupied topological surface state (TSS) as well as in the bulk conduction band for different pump photon energies in the range between 0.8 and 2.5 eV, for sample temperatures between 40 and 300 K as well as for different pump fluences between 0.5 and 2.5 mJ/cm^2.
For all investigated photon energies we find that the population of the TSS is dominated by an indirect excitation through the bulk conduction band. Experiments for different sample temperatures reveal that the decay out of the TSS is independent on temperature for energies below the conduction band minimum (CBM) whereas it becomes faster with lower temperature for energies above the CBM as we have already reported for Sb_2Te_2Se [1]. An increase of the pump fluence results in a slower decay which is less pronounced at low sample temperatures.
We will discuss the observed dynamics under particular consideration of electron transport into the bulk as an important decay channel resulting from the small optical penetration depth of Sb_2Te_3.

[1] J. Reimann, J. Güdde, K. Kuroda, E. V. Chilkov, and U. Höfer, Phys. Rev. B **90**, 081106(R) (2014).

Electronic structure and electron dynamics of the topological insulator Sb_2Te_2S

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The strength of the electron-phonon coupling in topological insulators is debated. Both strong and weak coupling constants have been reported [1,2].

We use time- and angle-resolved two-photon photoemission (2PPE) to study the electronic structure and ultrafast electron dynamics of the p-doped topological insulator Sb_2Te_2S [3]. Our experiments reveal a Dirac-cone like energy dispersion of the topological surface states above the Fermi energy. In the 2PPE experiment, the Dirac cone is populated by an infrared pump-pulse via direct absorption and interband scattering from the conduction band. In the Dirac cone we observe electron dynamics on the picosecond timescale. The spectra hint at a stepwise relaxation via small-energy transfer processes. Cooling of the system leads to faster electron dynamics. Simulations by rate equations suggest that these unexpected dynamics are governed by electron-phonon and defect scattering. Our measurements corroborate the supercollision model which was proposed for graphene [4].

[1] R. C. Hatch, M. Bianchi, D. Guan, S. Bao, J. Mi, B. Brummerstedt Iversen, L.Nilsson, L. Hornekær, and P. Hofmann, Phys. Rev. B **83** (2011) 241303.
[2] Z.-H. Pan, A.V. Fedorov, D. Gardner, Y. S. Lee, S. Chu, and T. Valla, Phys. Rev. Lett. **108** (2012) 187001.
[3] J. Reimann, J. Güdde, K. Kuroda, E. V. Chulkov, and U. Höfer, Phys. Rev. B **90** (2014) 081106(R); Erratum Phys. Rev. B **91**, 039903 (2015).
[4] M. W. Graham, S.-F. Shi, D. C. Ralph, J. Park, and P. L. McEuen, Nature Physics **9** (2013) 103.

	Monday	Tuesday	Wednesday	Thursday
8:30	**Jure Demsar** *Cooperative atomic motion probed by femtosecond electron diffraction*	**Sascha Schäfer** *Ultrafast electron imaging and diffraction with nanoscale photoemitters*	**Michael Horn-von Hoegen** *Electron Diffraction at Surfaces: by now Ultrafast!*	**Ralph Ernstorfer** *Femtosecond electrons probing structural dynamics and ultrafast currents*
9:30	**Selene Mor** *Ultrafast coherent phonon dynamics during the phase transition of the quasi one-dimensional Ta_2NiSe_5*	**Petra Hein** *Hot electron dynamics at 2H MoS_2 surfaces: Time- and angle-resolved photoelectron spectroscopy results*	**Thomas Vasileiadis** *The effect of confinement in the electron-phonon coupling studied with ultrafast electron diffraction*	**Konrad Gillmeister** *Electron dynamics in NiO ultrathin films*
16:00	**Abdul Samad Syed** *Ultrafast dynamics of a Pb overlayer structure grown on flat and vicinal Si(111)*	**Matthias Hohenleutner** *High-order harmonic generation by dynamical Bloch oscillations in a bulk solid*	**Uwe Bovensiepen** *Competition of spin currents and spin-flip scattering in ultrafast magnetization dynamics of Co/Cu(001) analyzed by the complex time-resolved MOKE*	**Ishita Agarwal** *Femtosecond time-resolved 2PPE spectroscopy of solvated electrons in $C_6H_5F/D_2O/Cu(111)$*
16:30	**Markus A. Huber** *Ultrafast sub-cycle terahertz nano-spectroscopy*	**Gerald Rohde** *Time- and angle-resolved XUV ARPES at sub-15 fs temporal resolution*	**Alexey Melnikov** *Direct monitoring of hot carrier-formed spin currents in metallic multi-layers by time-resolved magneto-induced second harmonic generation*	**Harald Kirsch** *Probing Water Dissociation and Structure at the α-Al_2O_3(0001) Surface Using Interface Specific Vibrational Dynamics*
17:30	**Alexander Lerch** *Ultrafast electronic excitations at the buried GaP/Si interface*	**Cheng-Tien Chiang** *Double photoemission spectroscopy on correlated d electrons in solids using a MHz HHG light source*	**Moritz Barkowski** *Depth-dependent study of femtosecond spin-currents in Ni/Au*	**Wibke Bronsch** *Ultrafast Exciton Dynamics in Thin Sexithiophene Films*
18:00	**Alex Paarmann** *Second Harmonic Spectroscopy in the Reststrahlen Band of 6H-SiC*	**Sebastian Emmerich** *trARPES with bright narrowband HHG pulses using frequency-doubled Ti:Sapphire laser*	**Kamil Bobowski** *Ultrafast magnetization dynamics of Gadolinium measured by XMCD in reflection geometry*	**Jens Güdde** *Electron dynamics in the topological insulator Sb_2Te_3*
18:30			**Beatrice Andres** *Ultrafast magnetization dynamics in Gadolinium – Analyzing the transient spin polarization*	**Martin Weinelt** *Electronic structure and electron dynamics of the topological insulator Sb_2Te_2S*