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Book of Abstracts

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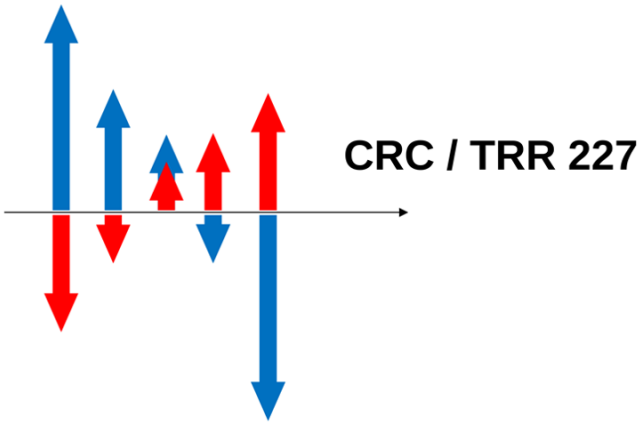

Contents

Sunday, March 4, evening / Opening Talk

A time domain perspective on electron-boson coupling in superconducting materials

Uwe Bovensiepen

Universität Duisburg-Essen

Experiments in the time domain allow to determine the electron-boson coupling by determining the second moment of the Eliashberg function $\alpha^2F(\omega)$ from the relaxation time constant of thermalized, hot electrons after optical excitation due to energy dissipation into further degrees of freedom. While this approach works well for conventional superconducting materials, it is under discussion for unconventional superconductors. In this talk, time- and angle-resolved photoemission on cuprate and Fe-based superconductors will be presented and the question of thermalized / non-thermalized distributions will be discussed. We show that coupling to specific bosonic excitations inhibits thermalization of the excited electron distribution. Analysis of the actual non-equilibrium distribution provides opportunities to analyze the electron-boson coupling without the assumption of a thermalized electron distribution.

Monday, March 5, morning / Invited Talk

Relaxation dynamics in strongly correlated materials - from models to materials

Denis Golez[1], Lewin Boehnke[1], Nikolaj Bittner[1], Martin Eckstein[2], Philipp Werner[1]

[1] *University of Fribourg*
[2] *Friedrich-Alexander-Universität Erlangen-Nürnberg*

Strong correlations between spin, charge and orbital degrees of freedom play an important role in materials and a recent development of ultrafast spectroscopies enabled to disentangle these relevant degrees of freedom by their temporal evolution. Due to the complexity of non-equilibrium dynamics it has become necessary to exchange ideas between experimental and theoretical community. I will present recent development of theoretical tools based on dynamical mean field theory (DMFT) that enable us to describe the non-equilibrium dynamics in strongly correlated materials and show how the theoretical development is approaching a realistic description of solids, which is crucial to provide a proper feedback to the experimental community.

I will start with a summary of the charge carrier relaxation after the photo-excitation in Mott insulators described within DMFT and continue how this formalism can be extended to more realistic description including the role of dynamical screening and non-local fluctuations (GW+EDFMT) [1,2]. First I will describe relevant relaxation channels deep in the Mott phase and close to the metal-insulator transition within a Hubbard model. The relevance of this description will be exemplified by the relaxation dynamics of the photo-emission spectra in 1T-TaS_2 [3]. Then I will introduce an advanced description including the effects of dynamical screening in the charge channel and open the question how to use the laser pulse to manipulate screening in Mott insulators. As an extreme example I will present a self-trapping of the system in the negative temperature state by a proper manipulation of the screening environment, which leads to the enhanced subgap response in the charge susceptibility. This population inversion leads to the low-energy anti-screening and I will comment on its experimental relevance. In the last part I will shed a light on the role of spin fluctuations in the relaxation dynamics, which can be analysed by an extension of DMFT [4], and exemplify how optical pump probe techniques can be used to detect some basic theoretical ideas in higher dimensional doped anti-ferromagnets, like string states, Trugman paths and the lack of spin-charge separation. At the end I will provide an outlook how to extend these tools to an ab-initio description of strongly correlated materials out of equilibrium.

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[4] N. Bittner, D. Golež, M. Eckstein, P. Werner, in preparation.

Monday, March 5, morning

Coupling between antiferromagnetism and an electronic surface state observed by time-resolved photoemission and x-ray diffraction

Yoav William Windsor[1], Chris W. Nicholson[1], A. Fedorov[2], Michele Puppin[1], K. Kummer[3], K. Kliemt[4], C. Krellner[4], Ralph Ernstorfer[1], M. Wolf[1], D. V. Vyalikh[5], Laurenz Rettig[1]

[1] *Fritz-Haber-Institut*
[2] *Leibniz-Institut für Festkörper- und Werkstoffforschung*
[3] *European Synchrotron Radiation Facility*
[4] *Goethe-Universität Frankfurt*
[5] *Donostia International Physics Center*

Intermetallics of type $R\,Rh_2Si_2$ (R is a rare earth) are antiferromagnets (AFM) with a Shockley surface state that becomes strongly spin-split in the AFM phase (up to 160 meV split). We study the coupling between the bulk AFM order and the surface spin polarization by probing the ultrafast dynamics of both properties in two complementary techniques. We use resonant X-ray diffraction (trRXD) to study the dynamics of the AFM order at the R ions' M edges, and we observe the narrowing of the spin-splitting gap using XUV time- and angle-resolved photoemission spectroscopy (trARPES). Comparison of the ultrafast demagnetization dynamics of the two orders allows us to address the coupling between them. The AFM dynamics also exhibited a coherent rotation of the entire AFM spin structure. This was resolved by measuring dynamics at several azimuths, which allows us to describe the entire AFM spin structure as a function of time. Lastly, differences in demagnetization timescales between $SmRh_2Si_2$ and $GdRh_2Si_2$ (300 fs vs 11 ps) suggest a strong variation in spin-lattice coupling, in analogy to 4f magnetization dynamics in Lanthanide metals.

Monday, March 5, afternoon

Terahertz dynamics of complex oxide interfaces

Wolf Widdra, Florian O. Schumann

Martin-Luther-Universität Halle-Wittenberg

In this presentation, the linear dynamics of ultrathin oxide films and oxide surfaces in the range from 1 to 50 THz upon electromagnetic excitation will be discussed. Surface vibrational spectroscopy based on high-resolution electron energy loss spectroscopy (HREELS) reveals the full dielectric characterization of the surface response of $BaTiO_3$, $SrRuO_3$, and $SrTiO_3$ with different intentional doping levels. The extracted surface dielectric function will be quantitatively compared with available bulk infrared data and allows the experimental determination of the surface-near doping level in oxides. HREELS data allow for the first time the determination of the complex dielectric functions of ultrathin oxide films down to single unit cell thicknesses. These data are discussed with respect of possible two-dimensional electron gases at the surface, electron-phonon coupling, as well as strain-driven phonon softening.

Support by the Sonderforschungsbereiche SFB-762 "Functional Oxide Interfaces" (projects A3 and B8) and SFB/TRR-227 "Ultrafast Spin Dynamics" (project A6) is gratefully acknowledged.

Monday, March 5, afternoon

Terahertz spin waves induced by femtosecond spin-current pulses in metallic multilayers: optimizing the optical excitation and tuning the frequencies

Liane Brandt[1], Mirko Ribow[1], Niklas Liebing[1], Ilya Razdolski[2], Georg Woltersdorf[1], Alexey Melnikov[1,2]

[1] *Martin Luther Universität Halle-Wittenberg*
[2] *Fritz-Haber-Institut*

The key challenge in a rapidly emerging field of modern spintronics consists in the push from the GHz to THz domain. In particular, it requires the development of techniques for the generation of femtosecond spin current (SC) pulses. With the help of magneto-induced second harmonic generation (mSHG) in the back pump-front probe scheme, we have demonstrated long-range spin transport on a femtosecond timescale upon laser excitation of the Fe film in epitaxial Fe/Au bi-layers [1]. Later on, in Fe/Au/Fe tri-layers, this technique allowed us to demonstrate 250 fs-short SC pulses and attribute their origin to the non-thermal spin-dependent Seebeck effect at ferromagnet/normal metal interfaces where the optically excited Fe film serves as a SC emitter [2]. With the time-resolved magneto-optical Kerr effect, we have demonstrated the excitation of sub-THz perpendicular standing spin waves (PSSW) in the opposite, 14 nm-thick Fe layer (collector) by interface-confined spin transfer torque (STT) exerted by these SC pulses [3].

Here, using both mSHG and magneto-optical Kerr effect detection in similar Fe/Au and Fe/Au/Fe structures with continuously varying thickness of the Fe emitter, we optimize the excitation of THz PSSWs and obtain new insights into the underlying electron and spin dynamics. In particular, from the SC generation efficiency dependence on the emitter thickness peaking at 4 nm, we estimate the escape depth of hot majority electrons in Fe of about 2 nm. To tune the PSSW frequencies, we fabricated an epitaxial Fe/Au/Fe structure with continuously varying Fe collector thickness from 1 to 17 nm. We demonstrate efficient STT-driven excitation of PSSWs with frequencies up to 2 THz corresponding to spin-wavelengths of 2 nm. Signatures of the higher-order PSSWs with frequencies up to 5 THz are also observed. We analyze the PSSW dispersion, damping, and amplitudes in the time domain as well as their dependence on the collector thickness and discuss new insights into the STT-induced THz spin dynamics.

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Monday, March 5, afternoon

Surface electron dynamics and oxygen reactivity of Dimethylsulfoxide films

Angelika Demling, Sarah B. King, Katharina Broch, Julia Stähler

Fritz-Haber-Institut

Dimethylsulfoxide (DMSO) is a widespread aprotic solvent proposed for use in lithium-air batteries [1]. However, the individual steps of electron transfer from the cathode into the solvated oxygen remain unknown. Using a model system of DMSO films on Cu(111) we investigate the fundamental steps in electron transfer using time- and angle-resolved two-photon photoelectron spectroscopy. On an ultrafast timescale we observe the formation and localization of a small polaron located at the surface of the second monolayer within 200 fs. Coverages with $>$ 2 monolayer equivalents additionally exhibit two surface-bound electronic states at 2.41 ± 0.05 eV and 2.30 ± 0.05 eV above the Fermi level whose lifetimes are on the order of several seconds. Detailed analysis reveals that both are formed from the same precursor state, most probably the small polaron. Upon exposure to O_2, the long-lived surface-bound electronic states are immediately quenched and a new state 2.53 ± 0.05 eV above the Fermi level appears with laser illumination. Moreover, the small polaron dynamics are shifted down in energy, approaching the energy of the oxygen-related state at times $>$ 200 fs implying that the small polaron forms the latter. Comparison to the gas phase O_2 [2] suggests that we are observing the formation of O_2^- at this model electrode/electrolyte interface.

[1] K. M. Abraham, J. Electrochem. Soc. **162**, A3021 (2015).
[2] K. M. Ervin *et al.*, J. Phys. Chem. A **107**, 8521 (2003).

Monday, March 5, afternoon

Surface photovoltage at the SiO_2/Si(100) interface studied by pump-probe photoemission with MHz high-order harmonics

Robin Kamrla[1,2], Cheng-Tien Chiang[1,2], Frank O. Schumann[2], Andreas Trützschler[1,2], Michael Huth[2], Wolf Widdra[1,2]

[1] *Martin-Luther-Universität Halle-Wittenberg*
[2] *Max-Planck-Institut für Mikrostrukturphysik*

The dynamics of the surface photovoltage (SPV) at the interface of SiO_2 and Si has been a subject of interest over the last decades [1-4]. In this contribution, we present time-resolved photoelectron spectroscopy (tr-PES) measurements on SiO_2/Si interfaces using a high-order harmonic generation (HHG) light source at megahertz repetition rates [5]. The observed dynamics of SPV as well as its doping and fluence dependence will be discussed in terms of the band structures and multiphoton transitions.

Charge carriers in the space charge region of *n*- and *p*-doped Si(100) (doping level $n = 10^{15} cm^{-3}$ each) are excited by femtosecond laser pulses with photon energies of $h\nu_{pump}$=1.2 or 2.4 eV. These excitations reduce the band bending at the SiO_2/Si interface, which can be monitored as energy shifts in the valence band photoelectron spectra using high-order harmonics of $h\nu_{probe}$=22.6 eV. On SiO_2/*p*-Si we determined a SPV of +250 meV towards higher energies upon excitation with $h\nu_{pump}$= 1.2 eV, whereas on SiO_2/*n*-Si the SPV reverses its sign and has a smaller magnitude of 140 meV [4]. Moreover, the SPV on the SiO_2/*p*-Si(100) interface depends logarithmically on the fluence for ϕ<2 nJ/mm^2 and ϕ >100 nJ/mm^2 [1-3], whereas it stays nearly constant in the intermediate fluence range. In strong contrast, for excitation with higher photon energy of $h\nu_{pump}$=2.4 eV, we observed only a logarithmic fluence dependence before its saturation at 350 meV at ϕ=20 nJ/mm^2.

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[2] W. Widdra, D. Bröcker, T. Gießel, I. V. Hertel, W. Krüger, A. Liero, F. Noack, V. Petrov, D. Pop, P. M. Schmidt, R. Weber, I. Will, B. Winter, Surf. Sci. **543**, 87 (2003).
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[5] C.-T. Chiang, M. Huth, A. Trützschler, M. Kiel, F. O. Schumann, J. Kirschner, W. Widdra, New J. Phys. **17**, 013035 (2015).

Tuesday, March 6, morning / Invited Talk

Ultrafast surface science with XUV pulses from high-harmonic generation

Stefan Mathias

Georg-August-Universität Göttingen

Ultrashort extreme-ultraviolet (XUV) pulses from high-harmonic generation (HHG) are a powerful tool for novel experiments in the area of ultrafast surface science. The short-wavelength nature of these sources provide important information related to all electronic, magnetic, structural, and chemical properties of a solid. In the beginning of my talk, I will exemplarily introduce a set of surface science experiments, where the usage of XUV light from HHG did considerably help to answer some of the key questions in the respective research field [e.g. 1,2]. I will further discuss current developments in HHG-based photoelectron spectroscopy, and show that it is now possible to map spin-dependent band structure dynamics using this technique. Here, we did study the response in Co to an ultrafast photo-induced spin current [3]. Via mapping of the spin-resolved band-structure dynamics, we find that optically driven femtosecond spin currents induce collective spin excitations on extremely fast timescales.

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[2] O. Kfir *et al.*, Science Adv. **3**, eaao4641 (2017)
[3] S. Eich *et al.*, Science Adv. **3**, e1602094 (2017)

Tuesday, March 6, morning

Elementary processes of laser induced ultrafast spin dynamics at Co/Cu(001) interfaces

Jinghao Chen[1], Andrea Eschenlohr[1], Uwe Bovensiepen[1], Tristan Müller[2], Peter Elliott[2], Eberhard K. U. Gross[2], John K. Dewhurst[2], Sangeeta Sharma[2]

[1] *Universität Duisburg-Essen*
[2] *Max-Planck-Institut für Mikrostrukturphysik*

The studies of spin injection across ferromagnetic/metallic interfaces are currently areas of interest, as the potential of layered structures being the basic building blocks of spintronic devices [1]. In technological applications one would like to have spin injection without spin-flip, therefore it is important to understand the microscopic nature of spin polarized transport and spin-flip at the interfaces.

In this work we report the experimental (time-resolved magnetization induced second harmonic generation [2]) and theoretical studies (time-dependent density functional theory [3]) at Co/Cu(001) interfaces. With the agreement between theory and experiment we identify a spin polarized transport from Co majority to Cu during the first 30 fs after optical excitation. Moreover the theory implies a simultaneous spin transfer from Cu minority to Co due to 1.55 eV pumping energy. Both experimental and theoretical data show that magnetization dynamics reaches its maximum change at 100 fs, in which the spin-orbit coupling changes the magnetization at time delays later than the saturation of the dynamics induced by charge transfer across the interface (30 fs). In conclusion we show that spin-flip processes can be induced by spin-orbit coupling without involving other scattering processes [4], which opens new possibilities for ultrafast control of spin dynamics.

We acknowledge funding from the DFG through SPP 1840.

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Tuesday, March 6, afternoon

Ultrafast electronic decay of above band gap excitations in NiO ultrathin films

Konrad Gillmeister[1], Cheng-Tien Chiang[1,2], Yaroslav Pavlyukh[1], Jamal Berakdar[1], Wolf Widdra[1,2]

[1] *Martin-Luther-Universität Halle-Wittenberg*
[2] *Max-Planck-Institut für Mikrostrukturphysik*

Nickel oxide, a type-II antiferromagnet with $T_{N\acute{e}el} = 523.6$ K, is a model system not only for a charge-transfer insulator but also a strongly correlated oxide. Despite of long-standing research its electronic structure is still object of great experimental as well as theoretical interest. The strong electronic correlation in NiO causes the appearance of lower and upper Hubbard bands with an energetic separation U which are formed by Ni $3d$ electrons. On the other hand, the presence of oxygen ligands leads to the formation of a charge-transfer gap (CTG) of the energy size $\Delta < U$.

In this contribution we focus on the electron dynamics after exciting electrons across the CTG of NiO ultrathin films. These films of thicknesses from 1 to 20 monolayers (ML) are grown on Ag(001) via molecular beam epitaxy. Using two-photon photoemission spectroscopy (2PPE) we find that the CTG ($\Delta \simeq 3.8$ eV) is formed in NiO films of 4 ML and beyond. Time-resolved measurements reveal an unexpectedly short electronic decay within less than 15 fs upon electron excitation across the CTG. This decay is coupled to the appearance of a coherent low-energy excitation which is long-living, thickness-, and temperature-dependent: periodic intensity modulations in the time-resolved 2PPE signal just above the vacuum cut-off are found for NiO film thicknesses ≥ 6 ML. These intensity modulations with a frequency of about 4.1 THz (17 meV) are visible for temperatures below 470 K for 10 ML (560 K for 20 ML). Their dephasing times are as long as 300 to 450 fs at room temperature and decrease with increasing temperatures.

The observed intensity oscillations are discussed in the framework of fundamental excitations in solids such as excitons, phonons, or magnons, and lead to coherent population of multi-electron excitations (*dd* excitations) within the NiO band gap that are subsequently probed.

Tuesday, March 6, afternoon

Development of a high power tabletop femtosecond pulsed X-Ray source

Manuel Bridger, Oscar Naranjo, Alexander Tarasevitch, Uwe Bovensiepen

Universität Duisburg-Essen

For a number of femtosecond time resolved soft X-Ray experiments, it would be a great improvement to have a femtosecond laser driven X-Ray source as a tabletop setup. In my doctorate thesis, I want to use High Harmonic Generation (HHG) to shift mid infrared laser pulses to the soft X-Ray range. To have enough power in the X-Ray range to perform absorption spectroscopy experiments, I use an infrared laser source with a power of about 30 W. This intense infrared laser light is generated by using an Optical Parametric Chirped Pulse Amplification (OPCPA) setup, which efficiently amplifies the initial laser pulses from a few nanojoule to a millijoule level. At a pulse repetition frequency of 20 kHz, the system is estimated to produce the mentioned power of 30 W of infrared laser light at the wavelength of 3500 nm. The goal is then, to use this setup to perform soft X-Ray pump-probe absorption spectroscopy experiments to analyze complex nanostructures in the time domain.

Tuesday, March 6, afternoon

High-repetition rate extreme ultraviolet HHG light source for femtosecond photoemission experiments

Christina Nolte[1], Amelie Schulte[1], Marco Merboldt[1], Germaine Arend[1], Steffen Hädrich[2], Tino Eidam[2], Jens Limpert[2], Sabine Steil[1], Daniel Steil[1], Stefan Mathias[1]

[1] *Georg-August-Universität Göttingen*
[2] *Active Fiber Systems GmbH, Jena*

Table-top coherent ultrashort extreme ultraviolet light sources from high-harmonic generation have been shown to enable a wealth of novel experiments in the field of ultrafast surface science [1,2]. However, the full potential of this approach has not yet been achieved because, to date, high harmonics generated by low-repetition rate Ti:sapphire lasers required a trade-off between photon flux, repetition rate, energy and time resolution [3].

Here, we present a HHG light source driven by a nonlinearly compressed 0.5 MHz fiber laser providing 10^{10} photons/s in single harmonics between $22-73$ eV. Two parallel beamlines allow for high harmonic generation in a tight focusing gas jet configuration with different fundamental drivers (1030 nm and 515 nm). In addition, our setup enables the direct generation of harmonics with bandwidths of the order of about 50 meV due to window-type Fano resonances in Argon. This makes the light source ideally suited for next generation femtosecond angle-resolved photoemission experiments.

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[2] S. Mathias *et al.*, J. Electron. Spectrosc. Relat. Phenom. **189**, 164 (2013).
[3] S. Eich *et al.*, J. Electron. Spectrosc. Relat. Phenom. **195**, 231 (2014).

Tuesday, March 6, afternoon

Beyond the molecular movie: ultrafast dynamics of bands and bonds during a photo-induced phase transition

Ralph Ernstorfer[1], Chris W. Nicholson[1], Aandreas Lücke[2], W. Gero Schmidt[2], Michele Puppin[1], Laurenz Rettig[1], Martin Wolf[1]

[1] *Fritz-Haber-Institut*
[2] *University of Paderborn*

In the Born-Oppenheimer picture, nuclear dynamics evolve across a potential energy surface determined by the time-dependent electronic structure and its transient occupation. In the case of photo-induced reactions, ab initio molecular dynamics (AIMD) simulations can provide the electronic and atomic structure of a system subsequent to impulsive excitation. Experimental access to the non-equilibrium distribution of electronic states of a material provides benchmarks which directly relate to the forces that govern the trajectory along the reaction coordinate during an ultrafast structural transition. Such an approach goes beyond the recording of a "molecular movie" [1], i.e., the determination of atomic structure as function of time. We demonstrate this concept by investigating a model phase transition system – indium nanowires at the (111) surface of silicon – which undergoes an order-order structural transition accompanied by an electronic metal-to-insulator transition [2]. Utilizing femtosecond time- and angle-resolved photoemission spectroscopy with a 0.5 MHz XUV laser source, we obtain direct access to the transient k-resolved electronic structure during the photo-induced phase transition (PIPT) in In/Si(111). By observing the dynamically changing band structure, a detailed reaction pathway is resolved, including temporal separation of the insulator-to-metal (200 fs) and structural (700 fs) phase transitions; the latter in extremely good agreement with recent time-resolved electron diffraction measurements [3]. The reaction pathway is reproduced by AIMD simulations, which reveal the crucial role played by localized photo-holes in shaping the potential energy landscape of the structural transition. This furthermore allows us to extend the description of ultrafast PIPTs to real space, and chart the ultrafast formation of chemical bonds during the phase transition.

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[3] Frigge, T. *et al.*, Optically excited structural transition in atomic wires on surfaces at the quantum limit. Nature **544**, 207 (2017).

Wednesday, March 7, morning / Invited Talk

Theoretical description of excitons signature in time resolved ARPES

Enrico Perfetto[1,4], Davide Sangalli[2,4], Andrea Marini[2,4], Gianluca Stefanucci[3,4]

[1] *Università di Roma Tor Vergata*
[2] *Istituto di Struttura della Materia of the National Research Council*
[3] *Università di Roma Tor Vergata, and INFN, Sezione di Roma Tor Vergata*
[4] *European Theoretical Spectroscopy Facility*

Time-resolved (TR) and angle-resolved photoemission (ARPES) in extended systems is often described in terms of electron dynamics into the conduction band. However the sample is probed into an excited state which is characterized by the same number of particle of the ground states. Such states are well described in the excitonic picture.

Here I illustrate an accurate diagrammatic approach we recently proposed [1] to calculate the TR-ARPES spectrum of systems with excitons. The diagrammatic approximation applies to the relaxed regime characterized by the presence of quasi-stationary excitons and vanishing polarization. The non-equilibrium self-energy diagrams are evaluated using excited Green's functions. The final result is an expression for the lesser Green's function in terms of quantities that can all be calculated in a first-principles manner. The validity of the proposed theory is illustrated in a one-dimensional model system with a direct gap. We discuss possible scenarios and highlight some universal features of the exciton peaks.

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Wednesday, March 7, morning

Two simple models for electronic states at surfaces and interfaces

Ulrich Höfer

Philipps-Universität Marburg

Ab-initio electronic structure calculations have become increasingly reliable and successful in describing relatively complex surfaces or interfaces consisting of hundreds of atoms. However, it is often difficult to extract general trends or to obtain a more global picture. In this talk, I will first present an ab-initio-based model potential which is able to predict the energy position and the wave function overlap of interface states between metals and a wide class of organic semiconductors [1]. As a second example for a simple physical model, I will discuss a scattering approach for two or more electronic resonances at metal surfaces and apply it to image-potential states [2]. Both models have not only provided fundamental insights into the origin and electronic properties of interface states and resonances. They also turned out to be very valuable in interpreting the results of time-resolved 2PPE experiments concerning the ultrafast dynamics of electron transfer processes.

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Wednesday, March 7, afternoon

Control of ultrafast photocurrents in the Dirac cone of the topological insulator Sb_2Te_3

Johannes Reimann[1], Kenta Kuroda[2], Jens Güdde[1], Ulrich Höfer[1]

[1] *Philipps-Universität Marburg*
[2] *University of Tokyo*

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 linear and circular polarized 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 magnitude and direction of this current both depend on the azimuthal orientation of the sample and reflect the threefold symmetry of the surface. For orientations in which no photocurrent is generated by linear polarized light, we are instead able to generate a photocurrent by circular polarized light and fully control its direction and magnitude by varying the light helicity.

[1] K. Kuroda *et al.*, Phys. Rev. Lett. **116**, 076801 (2016).

Wednesday, March 7, afternoon

Robustness of the charge-ordered phases in $IrTe_2$ against photoexcitation

Claude Monney[1], Adrian Schuler[2], Thomas Jaouen[1], Marie-Laure Mottas[1], Thomas Wolf[3], Michael Merz[3], Matthias Muntwiler[4], Luca Castiglioni[2], Philipp Aebi[1], Frank Weber[3], Matthias Hengsberger[2]

[1] *University of Fribourg*
[2] *University of Zurich*
[3] *Karlsruhe Institute of Technology*
[4] *Paul Scherrer Institute*

The transition metal dichalcogenide $IrTe_2$ undergoes two first-order structural and charge-ordered phase transitions on cooling below 270 K and below 180 K. It has been put forward that a Mott phase involving spin-orbit-coupled states [1] may be at the origin of the phase transitions in $IrTe_2$.

Here we present a time-resolved angle-resolved photoelectron spectroscopy study of $IrTe_2$ [2]. The possibility of inducing a phase transition by photoexcitation with near-infrared femtosecond pulses is investigated in the charge-ordered phases. We observe changes of the spectral function occurring within a few hundreds of femtoseconds and persisting up to several picoseconds, which we interpret as a partial photoinduced phase transition (PIPT). The necessary time for photoinducing these spectral changes increases with increasing photoexcitation density and reaches time scales longer than the rise time of the transient electronic temperature. We conclude that the PIPT is driven by a transient increase of the lattice temperature following the energy transfer from the electrons. However, the photoinduced changes of the spectral function are small, which indicates that the low-temperature phase is particularly robust against photoexcitation. We suggest that the system might be trapped in an out-of-equilibrium state, for which only a partial structural transition is achieved.

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Wednesday, March 7, afternoon

On the origin of photocurrents in the topological insulator Bi_2Se_3

A. S. Ketterl[1], S. Otto[2], M. Bastian[1], C. Gahl[1], B. Andres[1], J. Minar[3], H. Ebert[4], O. E. Tereshchenko[5], K. A. Kokh[6], J. Braun[4], Th. Fauster[2], M. Weinelt[1]

[1] *Freie Universität Berlin*
[2] *Friedrich-Alexander-Universität Erlangen-Nürnberg*
[3] *University of West Bohemia*
[4] *Ludwig-Maximilians-Universität München*
[5] *Novosibirsk State University*
[6] *Tomsk State University*

Topological insulators (TIs) 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 (DC) dispersion is occupied. Two-photon photoemission experiments have shown that Bi_2Se_3 exhibits a second unoccupied DC in the band gap between the second and third conduction band [1].

It has been disputed, whether photocurrents, induced in Bi_2Se_3 with circularly polarized light stem from the occupied DC [2]. We studied the momentum distribution of electrons excited with circularly polarized light from the first into the second DC 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. We find dichroic photoemission for circularly polarized pump pulses, both at resonant and off-resonant excitation. While a two-dimensional (E, k_y)-cut perpendicular to the plane of incidence would support the commonly assumed coupling of the photon angular momentum to the electron spin [3,4], it is inconsistent with the observed azimuthal (k_x, k_y) – patterns, which reflect the threefold crystal symmetry. Only for excitation out of the occupied bulk conduction band, we observe an asymmetric electron population, which would be a prerequisite for a photocurrent [5]. Thus our results question the topological origin of photocurrents in Bi_2Se_3 at near-IR excitation energies [3,4].

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Wednesday, March 7, afternoon

Intervalley scattering dynamics in MoS_2 imaged by 2PPE with high-harmonic probe

Robert Wallauer, Jens Güdde, Johannes Reimann, Ulrich Höfer

Philipps-Universität Marburg

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 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. 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 to 2.3 eV and show how the dynamics of this transfer depend on the excess energy above the exciton resonance.

Thursday, March 8, morning / Invited Talk

Dark exciton dynamics in atomically thin 2D nanomaterials

Ermin Malic

Chalmers University of Technology

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 regular bright excitonic states, there is also a variety of dark states that cannot be accessed by light due to the required momentum-transfer or spin-flip.

To model the exciton physics in TMD monolayers, we apply a microscopic approach combining semiconductor Bloch equations with the Wannier equation providing access to time- and momentum-resolved optical response and the non-equilibrium dynamics in TMDCs. In this talk, we review our recent work focusing on:

(i) Uncovering the full exciton landscape including bright as well as momentum- and/or spin-forbidden dark excitonic states [1].

(ii) Revealing the microscopic origin of the excitonic lifetime including radiative and phonon-assisted non-radiative relaxation channels [2,3].

(iii) Providing a fully quantum mechanical description of formation, thermalization, and decay of bright and dark excitons and their impact on photoluminescence and pump-probe spectroscopy [4,5].

(iv) Demonstrating signatures of dark excitons, when probing the intra-excitonic 1s-2p transition shortly after excitation and after exciton thermalization [6].

(v) Activation of momentum-forbidden dark excitonic states via efficient coupling with molecules and suggesting a novel dark-exciton-based sensing mechanism for molecules [7].

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Thursday, March 8, morning

Lightwave valleytronics in a monolayer of tungsten diselenide

Stefan Schlauderer[1], Fabian Langer[1], Christoph P. Schmid[1], Martin Gmitra[1], Jaroslav Fabian[1], Philipp Nagler [1], Christian Schüller[1], Tobias Korn[1], Peter G. Hawkins[2], Johannes T. Steiner[2], Ulrich Huttner[2], Stefan W. Koch[2], Mackillo Kira[3], Rupert Huber[1]

[1] *Universität Regensburg*
[2] *Philipps-Universität Marburg*
[3] *University of Michigan*

Steering the quantum motion of electrons in atoms and molecules by the carrier wave of an intense lightwave lies at the heart of high-harmonic generation and attosecond science [1]. Only recently, lightwave electronics has been extended to solids leading to the observation of dynamical Bloch oscillations [2], high-harmonic generation [3,4], quantum interference [5], and electron–hole collisions [6,7]. So far, however, these studies have mainly focused on controlling the translational motion of the electron's charge faster than a single cycle of light. Despite their potential as quantum information carriers, the internal degrees of freedom of crystal electrons, such as spin or valley pseudospin [8], have not been switchable on a subcycle scale yet.

Here, we demonstrate a novel control scheme for the valley pseudospin by lightwave-driven intraband transport. To this end, we resonantly prepare coherent electron–hole pairs in a monolayer of WSe_2 using a 100-fs near-infrared pulse. Simultaneously, an intense multi-terahertz waveform ionizes the electron–hole pairs, accelerates the constituent electrons and holes, and finally recollides them, resulting in the emission of a sequence of high-order sidebands accompanying the excitation spectrum. In contrast to bulk WSe_2 [7], our first-ever observation of high-order sideband generation in a monolayer crystal features a qualitatively new signature: odd-order sidebands. Their appearance under linearly polarized excitation can only be understood if the valley pseudospin is taken into account. The effectively symmetric excitation of both valleys [8] with linearly polarized near-infrared pulses compensates for the broken inversion symmetry. Yet, each valley emits circularly polarized light with opposite helicity. Hence, the contributions from the K and the K' valley are disentangled by their orthogonal polarizations. These pseudospin dynamics lead to cross-polarized even and odd sideband orders, which are directly confirmed by polarization-resolved experiments.

In order to directly trace how the valley pseudospin changes during lightwave acceleration, we prepare coherent excitons selectively in the K valley by a right-circularly polarized excitation pulse. If the electron–hole pairs recombine within the same valley, the sideband emission is expected to be polarized with the same helicity as the excitation pulse. Surprisingly, the emitted sidebands are polarized strongly elliptically with a predominant orientation perpendicular to the driving field. The overall elliptical polarization stems from counter-circularly polarized contributions of the non-excited K' valley due to lightwave-driven intervalley

transport. These dynamics are corroborated by quantum mechanical simulations, which predict transfer fidelities as high as 96% for realistic experimental parameters. Such observations mark the first lightwave-induced switching of the valley pseudospin, opening a completely new toolbox for optical-cycle-scale quantum information technologies.

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Thursday, March 8, afternoon

Ultrafast nanoscopy of photo-activated interface polaritons in black phosphorus heterostructures

Fabian Mooshammer[1], Markus A. Huber[1], Markus Plankl[1], Leonardo Viti[2], Fabian Sandner[1], Lukas Z. Kastner[1], Tobias Frank[1], Jaroslav Fabian[1], Miriam S. Vitiello[2], Tyler L. Cocker[1], Rupert Huber[1]

[1] *Universität Regensburg*
[2] *NEST, CNR Istituto Nanoscienze and Scuola Normale Superiore*

Surface polaritons in the mid infrared have emerged as a versatile platform for extreme light confinement and tailored nanophotonics [1,2]. These light-matter coupled modes can be hosted on two-dimensional materials such as graphene or hexagonal boron nitride. Whereas surface plasmons have been successfully modulated in gapless graphene, semiconducting layered materials and heterostructures are promising candidates for high-contrast, ultrafast control of polaritons.

Here, we report on the first ultrafast snapshots of a switchable surface polariton mode [3]. In a custom-tailored heterostructure, we sandwich the direct-gap semiconductor black phosphorus [4] (BP) between two SiO_2 layers. Upon ultrafast optical interband excitation, surface plasmon polaritons on BP couple to surface phonon polaritons on the adjacent SiO_2 layers to form hybrid interface polaritons. Using pump-probe scanning near-field optical microscopy (SNOM) in the mid infrared [5], we resolve the photo-activated polariton in real space as a standing wave pattern. We find that the hybrid polariton appears within 50 fs after photo-excitation, as electron-hole pairs are created within the BP layer. The subsequent recombination of the photogenerated carriers results in a decay of the hybrid mode within 5 ps. Remarkably, the hybrid polariton's wavelength is found to be almost constant throughout its entire lifetime. This decoupling of the hybrid polariton's momentum from the BP carrier density stands in stark contrast to the behavior of conventional plasmon polaritons [1,2].

Moreover, employing nano-spectroscopy, we trace the evolution of the hybrid polariton's energy and observe that it is strongly confined at a frequency of 33.8 THz. Furthermore, this instantaneous mode frequency is almost independent of the pump-probe delay time, which coincides with our theoretical modelling of the polariton hybridization. This switchable polariton mode with constant energy and momentum holds great promise for future ultrafast polaritonic devices because efficient incoupling throughout its entire lifetime can, for example, be provided by a monochromatic laser and a single grating.

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Thursday, March 8, afternoon

Occupied and unoccupied electronic structure of two-dimensional oxide quasicrystal

Cheng-Tien Chiang[1,2], Martin Ellguth[2], Florian O. Schumann[1], Christain Tusche[2,3], Stefan Förster[1], Richard Kraska[1], Wolf Widdra[1,2]

[1] *Martin-Luther-Universität Halle-Wittenberg*
[2] *Max-Planck-Institut für Mikrostrukturphysik*
[3] *Peter Grünberg Institut (PGI-6), Forschungszentrum Jülich*

In solid state physics, the electronic structure is often explained by examples of periodic systems. In reality, quasicrystals of metallic alloys with aperiodic atomic structure are well-known, whereas their valence electronic structure is still under debate [1]. Moreover, only very recently their unoccupied electronic structure has been studied [2]. In this contribution, we present angle-resolved photoelectron spectroscopy on the occupied and unoccupied electronic structure of a two-dimensional oxide quasicrystal (OQC), which is formed by the rewetting process of barium titanate on Pt(111) [3,4].

The occupied valence bands are investigated by the momentum microscope with He I excitation, and the photoelectron distribution over the whole hemisphere above the surface is mapped [5]. We decompose the energy-dependent photoelectron patterns according to symmetry considerations. As a results, clear dispersion of the oxygen 2p bands with a bandwidth of more than 0.5 eV at 5.2 eV below the Fermi level can be identified. Moreover, localized oxygen 2p states are observed at around 4 and 6 eV below the Fermi level, which are comparable to the O non-bonding and the Ti-O dpσ bands in bulk, respectively [6].

As preliminary studies on the unoccupied electronic states and their femtosecond dynamics, two-photon photoemission spectroscopy using a megahertz fiber laser system is performed. With a pump and probe photon energy of around 3.9 and 1.7 eV, the n=1 and 2 image potential states on OQC are identified at around 3.5 and 3.6 eV above the Fermi level.

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Thursday, March 8, afternoon

Excitation dynamics of the CuPc / PTCDA heterosystem on Ag(111)

Klaus Stallberg, Jonas Zimmermann, Alexander Lerch, Andreas Namgalies, Ulrich Höfer

Philipps-Universität Marburg

Stacked layers of copper-phthalocyanine (CuPc) and PTCDA on Ag(111) represent a structurally well-defined model for an organic donor-acceptor interface above a metal substrate. We investigate the excitation dynamics of this system by means of two-photon photoemission (2PPE). Besides direct 2PPE from the CuPc HOMO and photoemission involving the n=1 and n=2 image potential states (IPS), we observe an unoccupied Shockley-derived interface state (IS) at the PTCDA / Ag(111) interface. Photoemission from the IS for varying pump photon energies ranging from 1.6 eV to 2.5 eV mainly reflects the optical absorption spectrum of CuPc. This indicates efficient transfer of photoexcited electrons from the CuPc layer to the IS at the PTCDA / silver interface. Using time-resolved 2PPE we are able to follow the dynamics of this charge transfer process which takes place on a femtosecond timescale. Moreover, we discuss the observation of excitonic bands in the 2PPE spectra and investigate the relaxation dynamics of excitonic states in the CuPc / PTCDA donor-acceptor type heterosystem.

Thursday, March 8, afternoon

Different electron and hole dynamics in the Rashba material BiTeI

Sophia Ketterl[1], Marco Polverigiani[1], Vladimir Voroshnin[2], Beatrice Andres[1], Alexander Shikin[2], Martin Weinelt[1]

[1] *Freie Universität Berlin*
[2] *St. Petersburg State University*

BiTeI is a layered semiconductor without inversion symmetry. Strong spin-orbit coupling leads to a giant Rashba-splitting of the BiTeI surface and bulk bands. In general, semiconductors exhibit longer excited carrier lifetimes than metals and are thus favorable for spintronic applications. Crystalline BiTeI is known to cleave along the van-der-Waals-bonded layers of Te and I. This results in either Te- or I-terminated surfaces depending on the orientation of the crystal. For freshly cleaved samples we find a mixture of both terminations, which host surface states of either donor or acceptor character. Due to the concomitant down- and upward band bending the photoemission signal of both surface states unfavorably overlap. After one-day storage at a pressure of 10^{-9} mbar we clearly observe, as a bugaboo for all surface scientist, the surface band-structure of the Te-termination. This is attributed to the higher reactivity of the I-terminated surface, lifting its band bending upon rest-gas absorption [1].

In time-resolved photoelectron spectroscopy using an angle-resolving time-of-flight spectrometer, we measured the population dynamics of excited carriers in pure and Mn-doped BiTeI. Applying a 6.2-eV probe pulse, we monitor the Rashba-split surface state and conduction band on the Te-terminated surface. Its surface state has a nearly isotropic free-electron-like dispersion with positive effective mass. Excitation with a 1.5-eV pump pulse generates hot electrons with picosecond lifetimes τ, which show a Fermi-liquid-like behavior $\tau \sim (E - E_F)^{-2}$. Moreover, for higher laser fluence we observe an increase of the electron lifetime which scales with the laser fluence and thus density n of excited electrons as $\sim n^{5/6}$. The lifetimes of electrons do not depend on the sample temperature (50 *vs* 300 K).

In contrast, we find a very different behavior of the photohole. Their lifetimes do not follow Fermi-liquid theory, are rather constant, overall smaller than corresponding electron lifetimes, and show a clear temperature dependence. We attribute this behavior to field currents and/or the opening of an additional plasmon-emission channel as proposed in Ref. [2].

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Notes

	Sunday	Monday	Tuesday	Wednesday	Thursday	Friday
7:30		**Breakfast**				
8:30		**Denis Golez** Relaxation dynamics in strongly correlated materials – from models to materials	**Stefan Mathias** Ultrafast surface science with XUV pulses from HHG	**Davide Sangalli** Theoretical description of excitons signature in time resolved ARPES	**Ermin Malic** Dark exciton dynamics in atomically thin 2D nanomaterials	**Departure**
9:30		**Yoav William Windsor** Coupling between anti-ferromagnetism and an electronic surface state observed by time-resolved photoemission and x-ray diffraction	**Jinghao Chen** Elementary processes of laser induced ultra-fast spin dynamics at Co/Cu(001) interfaces	**Robert Wallauer** Intervalley scattering dynamics in MoS_2 imaged by 2PPE with high-harmonic probe	**Stefan Schlauderer** Lightwave valleytronics in a monolayer of tungsten diselenide	
10:05		**Breakout session & lunch break**				
16:00	**Arrival**	**Wolf Widdra** Terahertz dynamics of complex oxide interfaces	**Konrad Gillmeister** Ultrafast electronic decay of above band gap excitations in NiO ultrathin films	**Jens Güdde** Control of ultrafast photocurrents in the Dirac cone of the topological insulator Sb_2Te_3	**Fabian Mooshammer** Ultrafast nanoscopy of photo-activated interface polaritons in black phosphorus heterostructures	
16:35		**Alexey Melnikov** Terahertz spin waves induced by femtosecond spin-current pulses in metallic multilayers	**Manuel Bridger** Development of a high power tabletop femtosecond pulsed X-Ray source	**Claude Monney** Robustness of the charge-ordered phases in $IrTe_2$ against photoexcitation	**Cheng-Tien Chiang** Occupied and unoccupied electronic structure of two-dimensional oxide quasicrystal	
17:10		**Coffee break**				
17:40		**Angelika Demling** Surface electron dynamics and oxygen reactivity of Dimethylsulfoxide films	**Christina Nolte** High-repetition rate extreme ultraviolet HHG light source for femtosecond photoemission experiments	**Sophia Ketterl** On the origin of photocurrents in the topological insulator Bi_2Se_3	**Klaus Stallberg** Excitation dynamics of the CuPc / PTCDA heterosystem on Ag(111)	
18:15		**Robin Kamrla** Surface photovoltage at the SiO_2/Si(100) interface studied by pump-probe photoemission with MHz high-order harmonics	**Ralph Ernstorfer** Beyond the molecular movie: ultrafast dynamics of bands and bonds during a photo-induced phase transition	**Ulrich Höfer** Two simple models for electronic states at surfaces and interfaces	**Martin Weinelt** Different electron and hole dynamics in the Rashba material BiTeI	
19:15		**Dinner**				
20:30	**Uwe Bovensiepen** A time domain perspective on electron-boson coupling in superconducting materials					