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ABSTRACT BOOKLET

ORAL PRESENTATIONS

A LIGHT FIELD-DRIVEN LOGIC GATE IN A GRAPHENE HETEROJUNCTION

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Intense phase-controlled few-cycle laser pulses have enabled the control of electrons on sub-optical time scales in a great variety of systems ranging from atoms and molecules to solids. Over the last decade, steering electrons in solid-state materials and increasingly complex structures has emerged as a promising route towards electronics at unprecedented switching bandwidths, approaching 1 petahertz. For this aim we perform strong-field physics in graphene, an electrical conductor ideally suited to inject currents on sub-optical timescales, and with its single atomic layer nature a pure surface material. Here we show that we can distinguish and take advantage of two types of charge carriers in graphene: Real carriers, persisting after the light-matter interaction, and virtual carriers, existing during the interaction only.

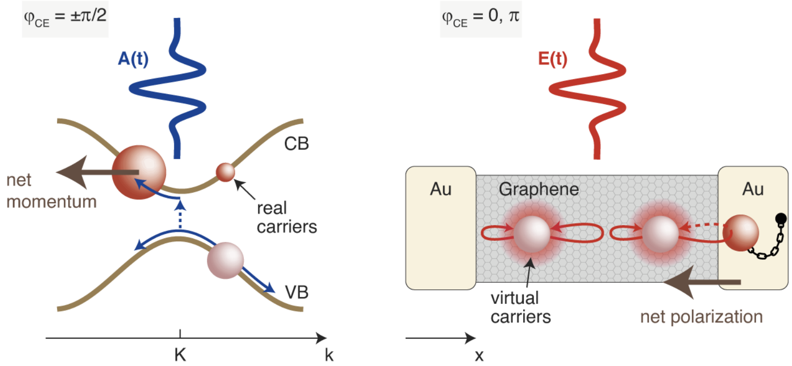

Figure 1 – Current generation by real and virtual charge carriers. Left, the temporal symmetry of the laser vector potential governs the generation of real carriers with net momentum, described in the momentum space representation of graphene. Right, in turn, in the real-space perspective, the symmetry of the laser electric field controls the net polarization of virtual carriers, which are collected by the gold electrodes.

We show that in a gold-graphene-gold heterostructure, the two types of carriers can be disentangled by their contribution to photo-generated currents, as they are susceptible to the carrier-envelope phase of the incident few-cycle laser pulses. The ultrafast dynamics are fully captured by atomistically detailed simulations on charge transport in the heterostructure. These insights now enable us to design and demonstrate a proof-of-concept experiment of an ultrafast logic gate with a potential bandwidth limited fundamentally by the frequency of light [1].

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AB-INITIO SIMULATION OF ULTRAFAST SPIN DYNAMICS: PHONOMAGNETISM AND THE GIANT OPTICAL SPIN HALL EFFECT.

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Real-time time-dependent density functional theory (TDDFT) is an ab-initio simulation method to study the quantum-mechanical electron dynamics on ultrafast time scales. In recent years, we have applied TDDFT to study ultrafast demagnetization[1], optically induced spin transfer between sub lattices[2], and transient EUV MCD spectra[3]. In our latest work, we show that coupling phonon excitations of the nuclei to spin and charge leads to femto-phono-magnetism, a powerful route to control magnetic order at ultrafast times. We identify strong non-adiabatic spin-phonon coupled modes that dominate early time spin dynamics. Activating these phonon modes we show leads to an additional (up to 25\%) loss of moment in FePt occurring within 40 femtoseconds of the pump laser pulse. Our finding demonstrates that the nuclear system, often assumed to play only the role of an energy sink aiding long time re-magnetisation of the spin system, can play a profound role in controlling femtosecond spin-dynamics in materials.

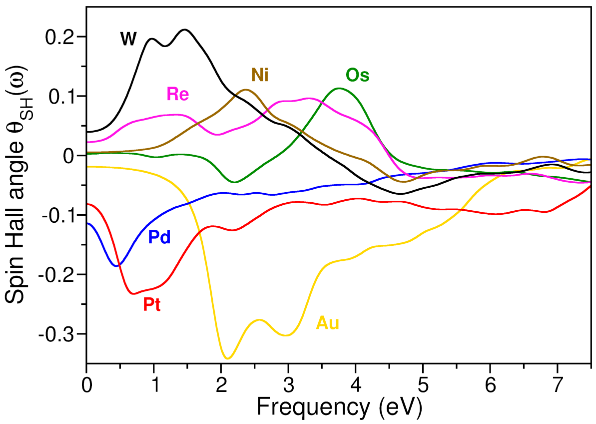

Figure 1: The frequency-dependent spin Hall angle, defined as the ratio of the transverse spin conductivity and the charge conductivity, for several transition metals.

We also study the spin Hall effect which plays a crucial role in spintronics, as both a generator (and detector) of spin current, which may be subsequently be used in spin-orbit-torque devices. We generalize the spin Hall angle to laser pulses of finite frequencies in the linear response regime and predict a giant optical spin Hall effect. Namely, for certain transition metal elements, at particular frequencies, the spin current can be a significant fraction of the charge current, and even exceed it for XUV frequencies. This can be seen in Fig.1, where giant spin Hall angles can be seen in Au and W at optical frequencies. Thus by tailoring the laser frequency, we can maximize the spin current while minimizing the charge current, and hence minimize a major source of heating in spintronic devices.

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Accessing the Real Time Flow of Energy Between Light, Carriers and Lattice in Solids with Attosecond Core-Level Spectroscopy

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Investigation of the emergence of a specific response of a molecular or solid-state system requires the ability to resolve the quantum multibody dynamics between light, carriers, and nuclei. Such investigation is still challenging due to the coupling between these various subsystems which occurs across vast temporal scales, ranging from attoseconds into picoseconds. Without discrimination of the various mechanisms, however, it is difficult to understand when and why an excitation manifests.

Here, we show that core-level x-ray absorption near edge structure (XANES) spectroscopy with attosecond soft x-ray (SXR) pulses meets the challenge in a single experimental method [1].

We study the dynamic flow of energy inside the layered semi-metal graphite and identify different regimes of carrier dynamics as we photoexcite the material with an ultrashort 11 ± 1 fs pump pulse at a photon energy of 0.7 eV, and a 15 ± 1 fs pulse at 1.6 eV, for pump fluences between 2.8 ± 0.2 mJ/cm2 and 81 ± 5 mJ/cm2. Attosecond-resolved measurements show the buildup of coherent charge oscillations which occur at occupied states below (holes) and unoccupied states (electrons) above the Fermi level, predominantly at the pump carrier frequency. An incoherent background, due to the dephasing of coherent charge oscillations, is rising within a few oscillations of the light field and signifies the ultrafast transfer of energy from the light field into the electron and hole excitation of the material. We find, that for a photo-doping with 1.6eV the ultrafast dephasing of the coherent carrier dynamics is governed by impact excitation (IE). For an excitation with 0.7eV photons, however, we only observe IE for electrons, while holes exhibit a switchover from IE to Auger heating (AH) already during the 11-fs duration of the infrared light field.

In contrast, a change in the XANES away from the Fermi level exhibits a markedly slower dynamic with a rise time for the incoherent contribution of $\tau^{0}_{\sigma^*} = 270 \pm 10$ fs and decay time of $\tau^{1}_{\sigma^*} = 4.3 \pm 1.3$ ps. This is in accord with the expected time evolution of phonon motion. A frequency analysis of the coherent signal contribution and a comparison with the phonon dispersion from two-temperature-model molecular dynamics simulations identifies the Raman-active $\Gamma - E_{2g}$ and the non-Raman-active $K – A'_1$ optical phonon modes at 46.4 ± 2.7 and 42.7 ± 1.1 THz, respectively

To elucidate the surprising early contribution from the (non-Raman-active) A'_1 mode, we calculated the equilibrium and the laser-excited potential energy surfaces along the A'_1 mode, and we found no displacement of the potential minimum. Thus, without the possibility for an immediate displacive excitation of the fully symmetric mode, we conclude that the observed coherent lattice motion must originate from the very strong electron-phonon coupling, thus acting almost impulsively.

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CHARGE CARRIER DYNAMICS IN EPITAXIAL METAL-HALIDE PEROVSKITES

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Metal-halide-based semiconductors with the perovskite structure (MHPs) and the empirical formula ABX_3 (A: stabilizing cation, B: metal, X: halide) are excellent candidates for next generation solar cells. They have high absorption coefficients, low exciton binding energies and large diffusion lengths of charge carriers. Their charge carrier dynamics is dominated by the interaction with polar phonons. Details of the underlying mechanisms are currently discussed [1]. In particular, the behavior of nascent high-energetic carriers is insufficiently understood [1,2].

For a systematic study, we prepared $CsPbBr_3$ and $CsSnBr_3$ by epitaxial growth in ultra-high vacuum. Our approach generates highly orientated thin films with well-defined (001) surfaces and very low defect densities. Angle-resolved photoelectron spectroscopy finds a valence band structure consistent with the one of single crystals [3]. This allows the detailed investigation of unoccupied states and electron dynamics of inorganic MHPs on the femto- and picosecond time scale with time-resolved two-photon photoelectron spectroscopy (2PPE). $CsPbBr_3$ and $CsSnBr_3$ exhibit each two decay channels of excited electrons. A fast decay τ_1 on the order of 100-200 fs can be assigned to the initial dynamics of high-energetic electrons. It is accompanied by the loss of excess energy with ~0.4 eVps^{-1} and might be related to the formation of large polarons or ultrafast transport [1,2].

The second time constant τ_2 of a few picoseconds corresponds to the diffusion of electrons at the conduction band minimum. It was used to determine the temperature-dependent electron mobility μ_e (T) in $CsPbBr_3$ from 2PPE-experiments (see Fig. 1).

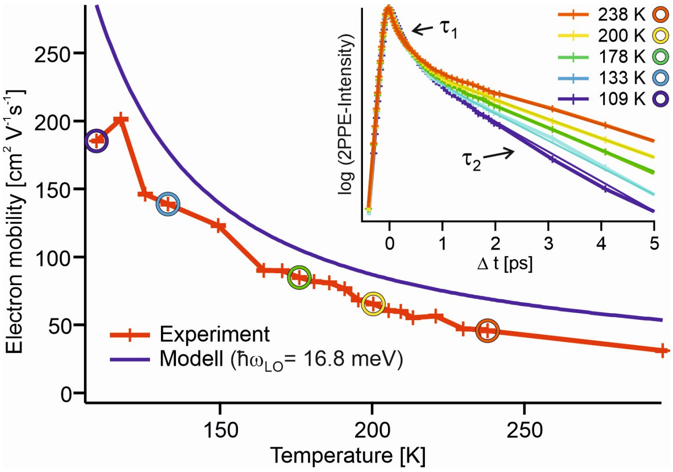

Figure 1 – μ_e (T) in epitaxial $CsPbBr_3$ together with underlying 2PPE-spectra at selected temperatures

The measured mobility is consistent with a model assuming a Fröhlich coupling of the electrons with one longitudinal-optical phonon ($\hbar\omega_{LO}$=16.8 meV) [4,5]. Corresponding electron-phonon scattering times are only 5 fs at 300 K. On this basis, an initial cooling rate of >3 eVps^{-1} would be expected. A comparison with the experimental rate points towards a screening of high-energetic carriers in MHPs.

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Density dependent mobility in photo-doped silicon

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Abstract: We have analyzed the interplay between photo-doping and mobility in SI-silicon by Time-Resolved Terahertz spectroscopy. Increasing a 400nm light pump photon flux, reveal the emergence of non-linear response in the recorded VIS pump-THz probe dynamics. The recorded change in dynamics is linked with an increase in the charge carrier scattering time as a function of pump-probe delay, as demonstrated from modelling the complex frequency resolved conductivity. A simple model for the diffusion of charge carriers from the surface towards the bulk explains the data rather well. Our results demonstrate a modulation of the charge carrier mobility as a function of photo-doping in the strong diffusive regime, which can be modelled by a Caughey-Thomas-like hyperbolic function vs charge carrier density.

1. Main Text

In this work we have studied the interplay between photo-doping and mobility in crystalline silicon by Time-Resolved Terahertz Spectroscopy (TRTS). By photo-exciting a SI silicon wafer above its band gap (pumping wavelength of 400 nm) we have time resolved the evolution of photogenerated charge-carriers optically (i.e., in a contactless way). As the Terahertz probe is primarily sensitive to free carriers, we can directly relate the changes in the absorption with the pump induced real photoconductivity[1] $\Delta T(t)/T \propto Re[\sigma(t)] = eN(t)\mu(t)$; where $Re[\sigma(t)]$ is the real part of the photoconductivity, $N(t)$ is the photo-induced carrier density, e is the electric charge, and $\mu(t)$ is the mobility. Figure 1a shows the optical pump-THz probe TRTS dynamics at a fluence of ~10,5 μJ/cm². The increase of the real conductivity as function of pump-probe delay is attributed to an increase of the charge carrier mobility. This is evident by retrieving the frequency-resolved complex conductivity at different pump-probe delay (see Figure 1b). To explain the data, we implement a simple diffusion model based on the Fick's law where the depth of the photogenerated carriers is a function of pump-probe delay. We can obtain the carrier density following $N(t) = N_s/d(t)$, where $d(t)$ is the effective thickness of the electron-hole gas as a function of time. A simple Caughey-Thomas-like hyperbolic function is able to model the scattering time (and hence mobility) dependence with photo-doping in the strongly diffusive regime (Figure 1c). These results are useful for theoretical modelling of charge carrier dynamics in silicon-based devices operating under strong injection and in the diffusive regime.

2. Tables and figures

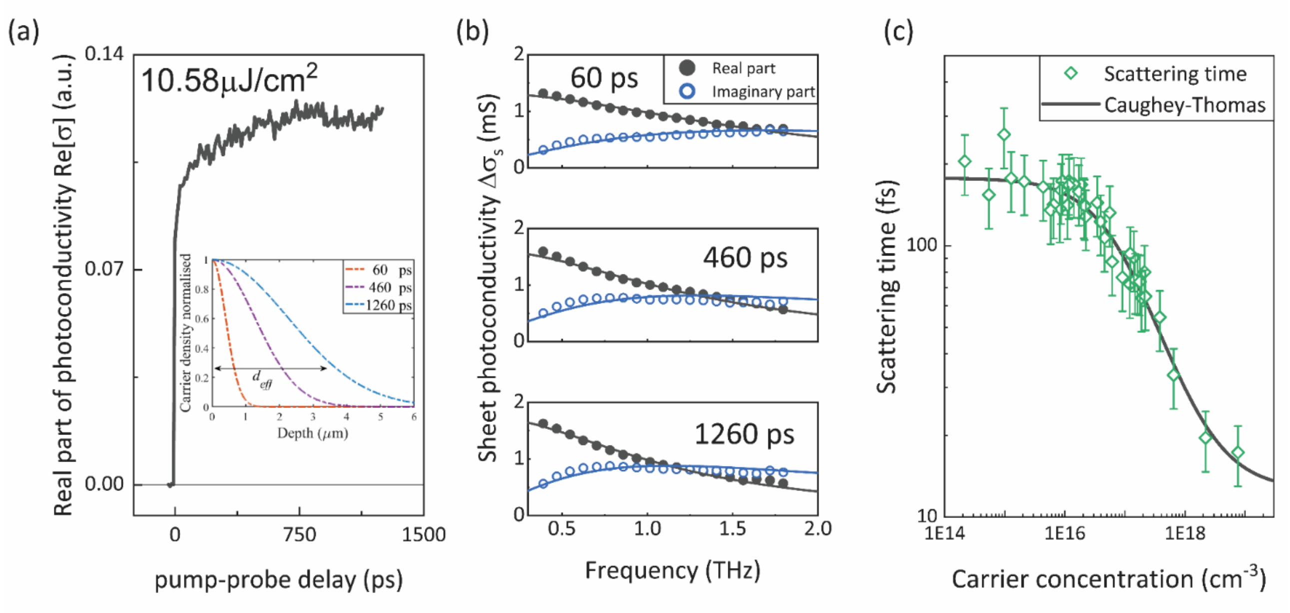

Figure 1. a) Real part of the photoconductivity as function of pump-probe delay for a fluence of 10,58 μJ/cm². Inset: numerical solution of the electron density profile with the diffusion model at 3 different pump-probe delays. b) Frequency-resolved complex conductivity measured at selected pump-probe delays at a fluence of ~10,5 μJ/cm². Black and open blue dots represents the real and imaginary part of photoconductivity, solid lines are best fits to Drude model. c) Scattering time as function of pump-induced charge carrier density. Solid line represents a Caughey-Thomas-like fit to the collected data.

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Disentangling Bloch states layer-by-layer in an antiferromagnetic topological insulator via layer-frequency lock-in

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Layer-by-layer material engineering has enabled exotic quantum phenomena. Meanwhile, deciphering electronic states layer-by-layer remains a significant experimental challenge. Here we introduce a new time-domain approach, Layer-Frequency Lock-in, to disentangle electronic states layer-by-layer in artificial heterostructures. Our approach is inspired by the depth resolution of magnetic resonance imaging, where there is a one-to-one mapping between the magnetic resonance frequency and depth. A similar layer-frequency correspondence can be utilized on digital superlattices enabled by time- and angle-resolved photoemission spectroscopy (trARPES): femtosecond pump pulses excite coherent lattice vibrations with layer-specific frequencies; photoemission spectroscopy tracks the dynamics of electronic band structures which are subsequently decomposed in the frequency domain. The frequency-domain ARPES (FD-ARPES) maps provide direct evidence for which electronic states couple to which phonon modes. Based on the layer-frequency correspondence, we deduce the layer origins of electronic states by identifying the substantial coupling with the corresponding phonon modes.

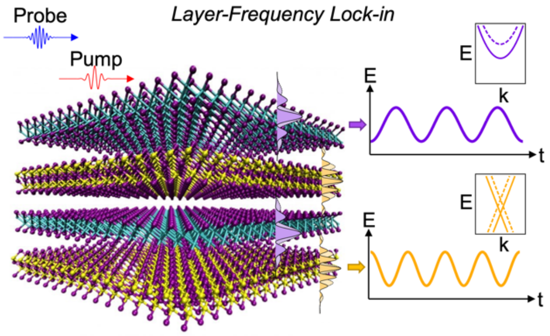

Figure 1 – Scheme of Layer-Frequency Lock-in Measurement. Using femtosecond infrared pulses to launch layer-specific coherent phonons, time- and angle-resolved photoemission spectroscopy allows us to decompose the electronic responses into different layers.

We demonstrate the first-time implementation of Layer-Frequency Lock-in to reveal the layer origins of key electronic states in a magnetic topological superlattice, $(MnBi_2Te_4)(Bi_2Te_3)$. Surprisingly, the Dirac state on the $MnBi_2Te_4$ termination exhibits predominant coupling with the Bi_2Te_3 A_{1g} distortion. Moreover, new quasi-2D states emerge on the $MnBi_2Te_4$ termination to couple with the $MnBi_2Te_4$ A_{1g} distortion. Our finding suggests that the Dirac state is buried underneath the top magnetic layer due to intrinsic structural changes. Our result potentially reconciles the controversy over the Dirac energy gap in $(MnBi_2Te_4)(Bi_2Te_3)_n$ materials. The layer-frequency correspondence can be harnessed to disentangle electronic states layer-by-layer in a broad class of van der Waals superlattices.

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DOES THE ORBITAL ANGULAR MOMENTUM OF LIGHT INFLUENCE ULTRAFAST DEMAGNETIZATION?

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Optical fields can carry an orbital angular momentum (OAM) of $L = l\hbar$ with the OAM quantum number $l \in \mathbb{Z}$. Since its discovery in 1992 [1], there has been a variety of applications of light with additional OAM, such as quantum entanglement, micromanipulation, communication, and microscopy [2].

Our research focuses on exploring the potential impact of the OAM of light on laser-induced ultrafast demagnetization of ferromagnetic materials. In this field, the question of how the angular momentum is conserved during the optically induced loss of magnetic order has not yet been fully answered. It has been shown that the spin angular momentum of light does not affect ultrafast demagnetization in ferromagnetic thin films such as Ni [3, 4]. However, pumping such a system with photons carrying OAM offers the potential to provide new insights, not only into the dissipation of angular momentum in the material but also into the interaction of optical OAM with matter in general. We investigate ultrafast demagnetization of ferromagnetic thin films induced by OAM light with time-resolved magneto-optic Kerr-effect measurements. We observed peculiar demagnetization dynamics that has so far not been observed for light without OAM.

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Dressing electrons with light

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Light interacts as a vector field with electrons in solid state matter, which I study by ultrafast photoemission spectroscopy and microscopy.[1-3] Applying an optical field introduces a time-periodic potential that transiently dresses the electron flow in momentum eigenstates of space periodic ionic potentials.[4,5] The Floquet engineering of electronic bands of metals appears in nonlinear multiphoton photoemission spectroscopy as displacement of electronic bands by n integer quanta of the photon energy, $\hbar\omega$.[6] At the onset of nonperturbative interactions, the photon field can also dress the momentum eigenstates causing field dependent energy shifts of Bloch bands in angle resolved multiphoton photoemission spectra. in also shift energies of the unperturbed eigenstates. I will discuss the attosecond time-scale Autler Townes dressing of noble metal Shockley surface and image potential states in resonant 1-3 photon excitation, and their Volkov replication in response to interferometrically defined optical fields.

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ELECTRONIC DYNAMICS IN SIDE-CHAIN-ENGINEERED THIOPHENE-BASED D-A ORGANIC POLYMERS FOR PHOTOVOLTAICS

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Thiophene-based polymers are promising candidates for solar cell, OLED or transistor (OFET) applications. An internal donor (D) – acceptor (A) system is formed by coupling thiophene polymers with pyrrole chains. Their charge transport behavior can be tuned by different electron acceptors, electron donors, and alkyl side chains. All three influence the electronic structure, HOMO and LUMO level positions, and interchain interactions. We employed diketopyrrolopyrrole [1] and fluorene as electron acceptors and various thiophene moieties as electron donors. Further, alkyl and urea modified side chains were studied.

We report results of temporally resolved photoemission studies on thiophene polymers on silicon substrates. Occupied electronic levels were analyzed via static UPS. The electron spectroscopy reveals LUMO-derived states, as well as excitonic and polaronic states in the bandgap. Dynamic 2PPE experiments were performed for the energetic position and dynamics of the unoccupied states. Details in the electron dynamics were resolved with tunable femtosecond UV radiation from a NOPA as pump pulse and the fourth harmonic (257 nm) of a compressed 500 kHz fiber laser as probe pulse. Always multiple exponential population decays were observed. Lifetimes, resolved according to the electron kinetic energy, can be grouped into three ranges, a fast one around a few 100 fs, an intermediate one of about 3 ps and 10 ps, and show also a slow component around 30 ps or longer. The fluorene acceptor yields significantly shorter lifetimes then the diketopyrrolopyrrole one.

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EXTENDING SUBCYCLE THZ-ARPES TO THE STRONG-FIELD REGIME

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Time- and angle-resolved photoemission spectroscopy (tr-ARPES) provides an indispensable tool to track electrons in solids within the band structure on ultrafast time scales. Recently, tr-ARPES has been combined with terahertz (THz) excitation, while the probe-pulse duration has been set shorter than the single optical cycle and allowed for studying subcycle electron dynamics in momentum space [1]. However, the field strength achieved with a center frequency of 1 THz was limited to a few kV/cm, and real strong-field phenomena were not accessible. Here, we introduce our latest subcycle THz-ARPES experiment in the mid-infrared (MIR) spectral region (center frequency of 25-40 THz), which enables us to reach a field strength of ~1 MV/cm even under surface screening. The technical development paves the way for visualizing various strong-field phenomena such as the emergence of Floquet-Bloch states [2] and high-harmonic generation (HHG) [3] directly in the band structure and on sub-optical-cycle time scales.

In our experiment, we excite electrons in topological surfaces state (TSS) of Bi_2Te_3 by using the MIR pump, which facilitates the generation of electric fields up to several tens of MV/cm. Combining the intense MIR pump with subcycle ARPES poses multiple technical challenges, including the requirement of a temporal resolution better than a single optical cycle, which unavoidably leads to an energy broadening of the ARPES spectra. We meet the former by utilizing a two-photon photoemission process with broadband 400-nm probe pulses compressed down to 17 fs. We also demonstrate that the visibility of the broadened ARPES maps is dramatically improved by applying a curvature-based image processing, an extension of a well-known second-derivative technique. Figures 1(a) and (b) show the photoelectron momentum streaking trace and the converted electric-field waveform on the surface. The peak field of 0.8 MV/cm is strong enough for HHG from TSS [3]. Curvature-filtered, streaking-compensated THz-ARPES images in Fig. 1(c) warrant the first observation of a number of key intrinsic material dynamics in the band structure: the formation of an asymmetric electronic distribution – a hallmark of lightwave-driven currents – and the formation of multiple sidebands are simultaneously resolved within a single optical cycle. Our experiments provide direct access to the transient dynamics of Floquet-Bloch states as well as to subcycle intra- and interband excitations underlying HHG.

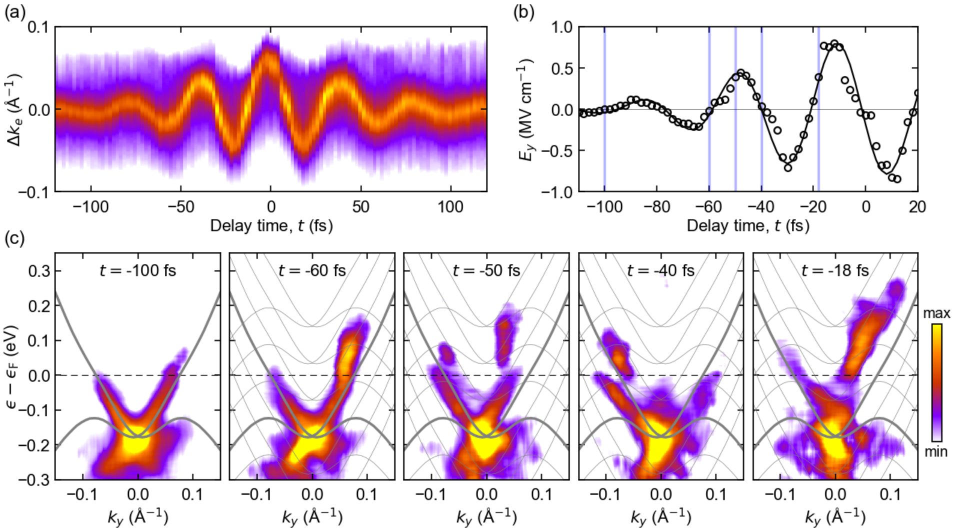

Figure 1 - (a) Photoelectron momentum streaking of a 25 THz driving field. (b) Electric-field waveform reconstructed from the streaking trace. The solid line shows an analytical fitting. (c) Curvature-filtered, streaking-compensated THz-ARPES maps measured at five temporal steps indicated in (b). The TSS band structures obtained by density functional theory calculations and their Floquet replicas are indicated by gray curves. (ϵ_F: Fermi level)

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FAR-FROM-EQUILIBRIUM ELECTRON-PHONON INTERACTIONS IN OPTICALLY-EXCITED GRAPHENE

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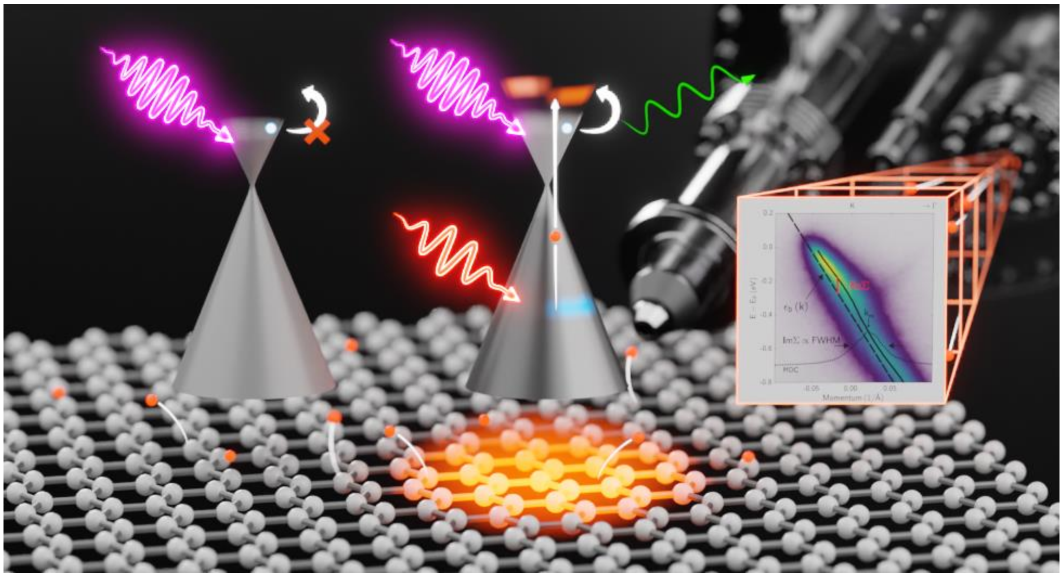
Comprehending far-from-equilibrium many-body interactions is one of the major goals of current ultrafast condensed matter physics research. Here, a particularly interesting but barely understood situation occurs during a strong optical excitation, where the electron and phonon systems are significantly perturbed from their equilibrium distributions.

In this work, we use time- and angle-resolved photoelectron spectroscopy (trARPES) to study such far-from-equilibrium many-body interactions for the prototypical material graphene. For this purpose, we optically excite the Dirac material into a strong NEQ regime, where the subsystems of electrons and phonons are in a non-equilibrium with respect to each other, and, in addition, cannot be described by Fermi-Dirac or Bose-Einstein distributions, respectively. We show that optically excited graphene exhibits a complex non-equilibrium many-body response by evaluating the linewidth of the Dirac state and thus the imaginary part of the quasiparticle self-energy Im(Σ) from spectrally deconvoluted trARPES data. We emphasize that this analysis is successful even though the energy resolution in our trARPES experiment is on the order of about 200 meV. By employing a first-principles theoretical modeling, we find that the observed complex experimental features in the time-dependent self-energy Σ are caused by ultrafast NEQ scatterings between optical phonons and photoexcited charge carriers, being active on timescales well below 100 fs. Our results advance the understanding of many-body physics in extreme conditions, which is important for any endeavor to optically manipulate or create emergent states of matter.



FEMTOSECOND ORBITAL TOMOGRAPHY OF EXCITON GENERATION AND RELAXATION DYNAMICS IN C_{60} THIN FILMS

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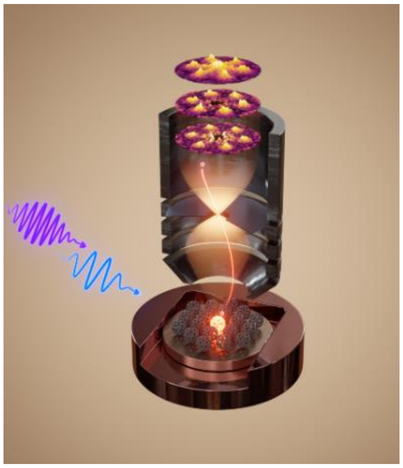

Figure 1 – Schematic overview of the experimental setup used for time-resolved photoemission orbital tomography of different excited states of crystalline C_{60}.

Photoemission orbital tomography is a powerful tool to probe the full electronic structure of oriented molecular thin films [1]. Recently, it has been demonstrated that in combination with ultrafast pump-probe spectroscopy it facilitates the access to the electronic excitation pathways subsequent to an optical excitation [2]. This approach also opens the door towards imaging excited state wavefunctions with nanometer spatial- and femtosecond time-resolution. In our work, we use ultrafast multidimensional photoemission spectroscopy to carry out femtosecond orbital tomography of exciton generation and relaxation dynamics in a C_{60} thin film. Here, it has been recently proposed that the exciton cascade involves the formation of delocalized charge-transfer excitons and subsequent two-step decay into a localized Frenkel excitonic state [3]. We gain access to real-space information by combining orbital tomography with calculations within the many-body perturbation theory framework of the Bethe-Salpeter equation, which enables us to gain insight into the transient localization dynamics of the electronic wavefunctions on femtosecond timescales. We also address the question of how the measured momentum fingerprints of two-particle correlated excitonic states, which are composed of both electrons and holes, can be interpreted in terms of single-particle orbitals, and to what extent many-body correlations and the final state of the remaining hole need to be considered in the process. Our findings demonstrate how photoemission orbital tomography can shed light on the real-space transient dynamics of out-of-equilibrium electronic wavefunctions, and in particular show how the delocalization of excitons in C_{60} evolves on the femtosecond timescale.

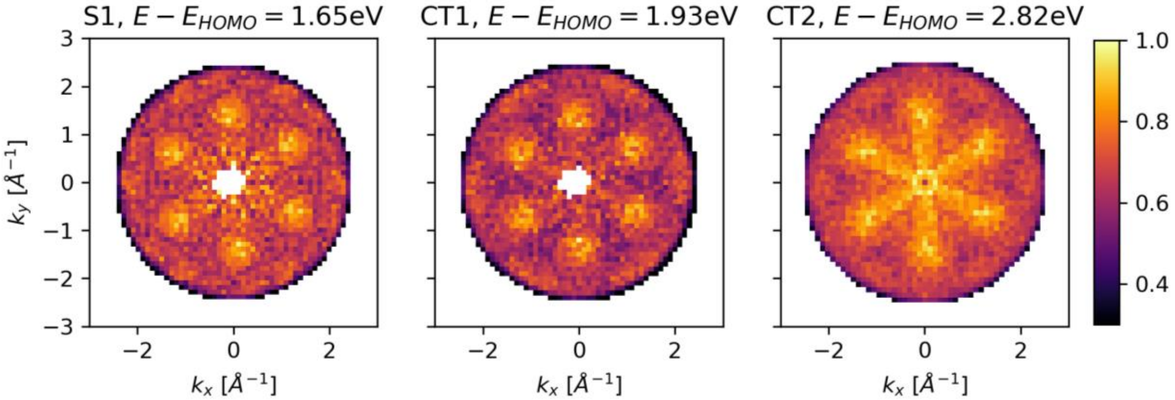

Figure 2 – Measured momentum fingerprints of the three excitonic states encountered in crystalline C_{60} after femtosecond optical excitation.

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INFLUENCE OF LOCAL SYMMETRY ON LATTICE DYNAMICS AND SPIN TEXTURE OF SURFACE STATES

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While global symmetries are decisive for determining bulk material properties, local symmetries can have a propound impact on confined electronic states, particularly those at surfaces. In this talk, we will examine the consequences on the lattice dynamics and spin-texture of topological surface states. Our experimental methodology spans time-resolved and spin-resolved photoemission spectroscopies.

First, we employ time- and angle- resolved photoemission spectroscopy (trARPES) to study coherent phonons in the topological semimetal Sb(111), and characterize their couplings to the bulk and topological surface states [1]. Despite the complexity of the response, we find that frozen-phonon density functional theory provides an adequate description of the binding-energy oscillations. This result benchmarks the connection between coherent phonons and the well-established equilibrium framework of electron-phonon coupling.

We then focus on coherent phonons in the topological insulator Bi_2Te_3. We find that the surface state couples to coherent modes that are forbidden in the bulk, including infrared-active phonons as well as those with non-zero wavevectors [2]. Our models show that these behaviors arise naturally due to the translational and inversion symmetries broken at the surface. By leveraging these locally broken symmetries, these effects expand the phase space for tailoring material properties on-demand, and they may provide a necessary ingredient for driving topological phase transitions via optical excitation.

Finally, we will present our first steps towards measuring the spin degree-of-freedom associated with these dynamics. We select the Rashba semiconductor BiTeCl as a model system to investigate strong spin-orbit coupling using spin-resolved ARPES. Our measurements reveal a strongly momentum-dependent spin texture in the surface states which can only be described by incorporating higher-order terms into the canonical Rashba Hamiltonian [3]. These terms should have a strong impact on the spin-dependent transport of charge carriers at the surface.

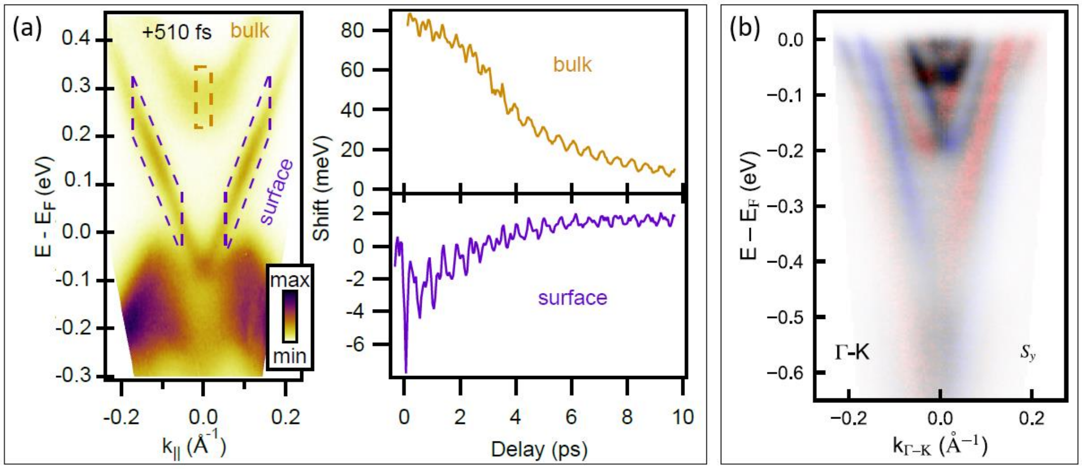

Figure 1

(a) Time-resolved ARPES measurements on the bulk and surface states of the topological insulator Bi_2Te_3.

(b) Spin-resolved ARPES measurements on the surface states of the Rashba semiconductor BiTeCl.

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LASER-DRIVEN NUCLEATION OF A TOPOLOGICAL PHASE & DEPTH-PROFILING OF MAGNETIZATION DYNAMICS

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Magnetic Skyrmions are spin textures which behave as quasi-particles and are characterized by a specific topology. Some types of skyrmions do exist in suitable thin magnetic multilayer film systems at room temperature, and they can be generated and efficiently moved laterally by spin-polarized current pulses. I will discuss how magnetic skyrmions can be generated via laser pulses instead of current pulses at unprecedented speed, with the topology change completed after 300 ps. Insight into the laser-induced formation mechanism comes from time-resolved scattering experiments at X-ray lasers in comparison with atomistic spin simulations. Local topology fluctuations in a transient high temperature phase are identified as the key element for this topological phase transition - a mechanism that may be applicable to phase transitions with a net change of topology in completely different material systems as well. [1]

So far, the experimental investigation of skyrmion nucleation mechanisms on the sub-picosecond timescale in real space has been hampered by the inability to reproducibly nucleate magnetic skyrmions at the same positions. Hence, one cannot build up statistics via pump-probe experiments at high repetition rates. I will report on recent progress in tackling this challenge. [2][3]

Thin magnetic films and stacked multilayers or heterostructures thereof are common sample systems to generate magnetic functionalities, such as all-optical magnetization switching or hosting structures like the skyrmions mentioned above. Even simple “single layers” typically contain growth-seed and cap layers. On a prototypical GdFe layer with a Ta cap layer and Pt seed layer, we demonstrate that time-resolved access to the magnetization depth profile is possible via pump-probe broadband TMOKE spectroscopy in the soft x-ray region around 150 eV photon energy [4], in conjunction with polarization-dependent magnetic scattering simulations [5], taking the layer structure into account. The depth-resolved temporal magnetization dynamics in this purely metallic system exhibits some unexpected features. For example, we observe a faster demagnetization of the GdFe layer at the interface to the seed (!) layer, while the IR pump laser is incident on the cap layer – illustrating the importance of depth-resolved information when trying to understand optically induced ultrafast dynamics in systems with heterogeneity on the few nanometer scale.

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MPSD Hamburg

Light-matter control of quantum materials: From light-induced superconductivity to cavity materials

In this talk I will discuss recent progress in controlling and inducing materials properties with light [1]. Specifically I will discuss recent experiments showing light-induced superconductivity through phonon driving in an organic kappa salt [2] and its possible theoretical explanation via dynamical Hubbard U [3]. I will then highlight some recent theoretical and experimental progress in cavity quantum materials [4], where the classical laser as a driving field of light-induced properties is replaced by quantum fluctuations of light in confined geometries. Ideas and open questions for future work will be outlined.

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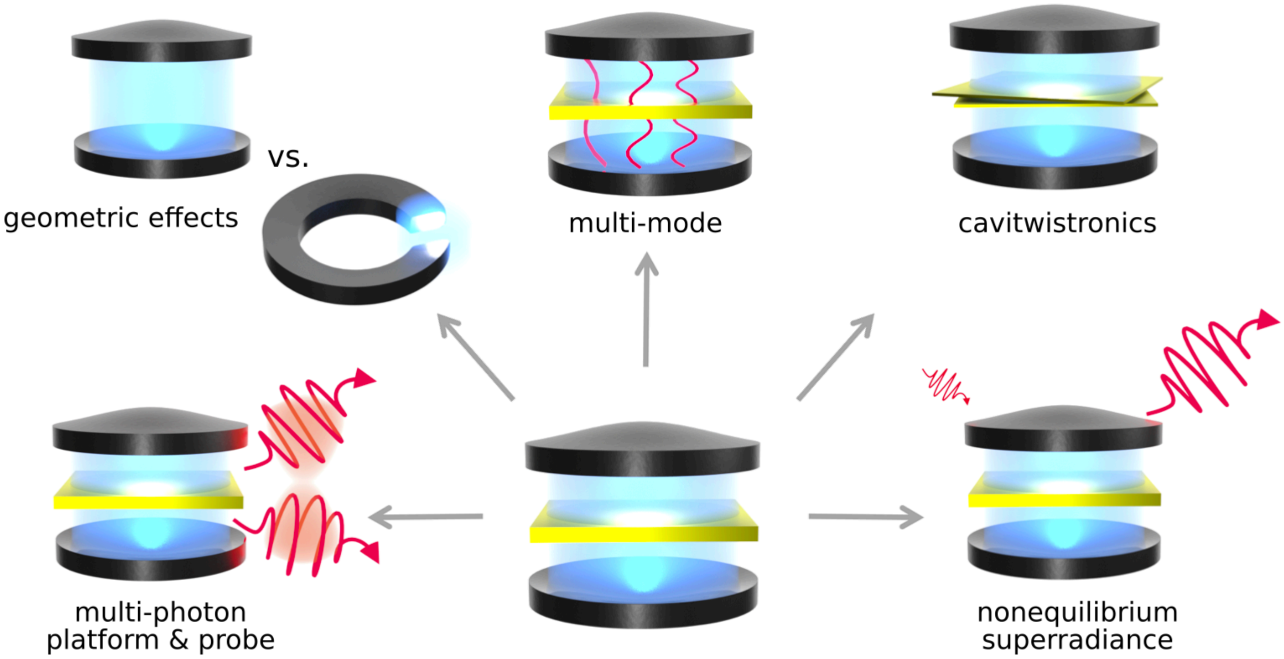

Machine Learning based Interatomic Potentials to Describe Laser-Excited Materials

Bernd Bauerhenne, Malwin Xibraku, Pascal Plettenberg, Martin E. Garcia

Femtosecond laser pulses create a transient state with hot electrons at temperature Te and cold ions, in which ultrafast nonthermal effects occur.
Later, the decay of this state due to electron-phonon interactions gives rise to ultrafast effects of more thermal character.
Quantum mechanical methods like Te-dependent density functional theory allow a precise description of the non-thermal state with hot electrons.
However, simulations of laser material interactions at surfaces on experimentally relevant sizes require to take a huge amount of atoms into account, which is only possible by the usage of interatomic potentials.
Most known interatomic potentials are determined assuming that electrons are in their ground state and cannot describe nonthermal effects initiated by bond-softening or hardening.
In order to take ultrafast thermanl and non-thermal effects on the same theoretical level, Te-dependent interatomic potentials are needed.
Here, we present two machine learning methods for developing highly accurate electronic temperature dependent interatomic potentials.
One is based on a polynomial functional form and the other one is represented by an artificial neural network.
We applied the methods to silicon and derived Te-dependent interatomic potentials that describe the phonon band structure, the cohesive energy curves for several crystal structures, the electronic specific heat, atomic forces and cohesive energies in excellent agreement with DFT calculations for a wide range of Tes.
Using our polynomial Te-dependent interatomic potential, we studied the influence of the non-thermal effects following a femtosecond laser excitation of a thin silicon film.
We also derived Te-dependent interatomic potentials for antimony and carbon on the same level of accuracy as for silicon.

Magnetization dynamics at FeRh surface: trigger of a sub-picosecond phase transition

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The meta-magnetic phase transition in FeRh attracted the interest of scientists for its appealing phenomenology and for the high degree of complexity. The stoichiometric FeRh metallic system is anti-ferromagnetic at room temperature and undergoes a first order phase transition (FOPT) as the temperature is risen above 420 K (150 C). Commensurate the magnetic transition, the lattice structure isotropic expands by about 1% in volume. The symmetry breaking induced by the surface also influence the magnetic state of the system: Rh-terminated surfaces stabilize a magnetic layer also at room temperature [1]. The FeRh free surface can sustain a graphene layer protecting the surface from contamination and oxidation [2]. We investigate the laser induced first order phase transition (FOPT) on a graphene covered FeRh surface by means of Time-Resolved Momentum Microscopy (TR-MM), focusing on the modifications of the electronic structure. We show that the presence of a spin polarized Fe-band close to the Fermi level is a clear fingerprint of the magnetic state of the system which can be used to follow the emergence of the ferromagnetic phase. The results show the appearance of a transient electronic phase characterized by strong modifications of the whole electronic structure as a precursor of the ferromagnetic phase, non-compatible with the common picture of photon-mediated electronic re-distribution [3]. The ensuing emergence of the ferromagnetic phase happens with an overall timescale of few hundreds of femtoseconds, which is, so far, the fastest recorded effect of the FOTP in FeRh. With the help of Time-Dependent Density Functional Theory (TD-DFT), we suggest that a crucial role in triggering the transition is played by the laser induced electronic redistribution with a net charge and spin transfer from the Rh to the Fe atoms.

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Microscopic dynamics of propagating and localized excitations at complex interfaces

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An excited electron propagates in condensed matter with its momentum **k** at an energy *E*(**k**) and experiences elastic and inelastic scattering processes, which lead to electronic relaxation, localization, and energy transfer to microscopic excitations of the nuclear and spin degrees of freedom. Experiments employing femtosecond time-resolved photoelectron spectroscopy exploited the surface sensitivity of the method and probed such scattering processes locally at or near the surface in the time domain [1]. Here, experimental results which analyze the non-local dynamics of excited electrons are reported and the sensitivity to buried media is demonstrated [2]. In these experiments one photon excites in Au/Fe/MgO(001) heterostructures electrons in Fe. Electron propagation through the layer stack to the Au surface is detected in linear and non-linear photoelectron spectroscopy in back side pump – front side probe experiments. We observe pronounced differences between front and back side pumping of the heterostructure which are attributed to (i) electron transport contributions through the layer stack and (ii) separate electron relaxation in the Fe and Au constituents.

Furthermore, work on alkali ions on Cu(111), onto which D_2O and Xe were coadsorbed as model systems for polar and non-polar solvents, will be reported. We find that the attraction between Cs^+ and Xe counterbalances the screened Coulomb repulsion between Cs^+ ions on Cu(111) and that the Cs 6s electron is repelled from Cu(111) due to xenon’s electron density [3]. This yields a dual, i.e., attractive or repulsive, response of Xe depending on the positive or negative charge of the respective counterparticle, which emphasizes the importance of the Coulomb interaction in these systems. In case of water coadsorption the solvent-solute arrangement on the surface is fundamentally different from the bulk. Ring like structures, in which the Cs^+ ions are located at the perimeter, form due to the competition between the Cs^+-water and the hydrogen bonds in combination with coupling to the substrate. The relaxation dynamics of the Cs 6s electron which is photo-excited by electron transfer from Cu(111), shows indications of a collective solute-solvent response. The relationship between coverage, geometric and electronic structure is essential for the chemical reactivity of such interfaces.

This work was done in close collaboration with the groups of D. Marx, A. Michaelides, K. Morgenstern, P. Saalfrank. It was funded by the Deutsche Forschungsgemeinschaft through the Collaborative Research Center CRC 1242 and the Cluster of Excellence RESOLV.

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MOIRÉ INTERLAYER EXCITONS IN SPACE AND TIME:
A TIME-RESOLVED MOMENTUM MICROSCOPY STUDY

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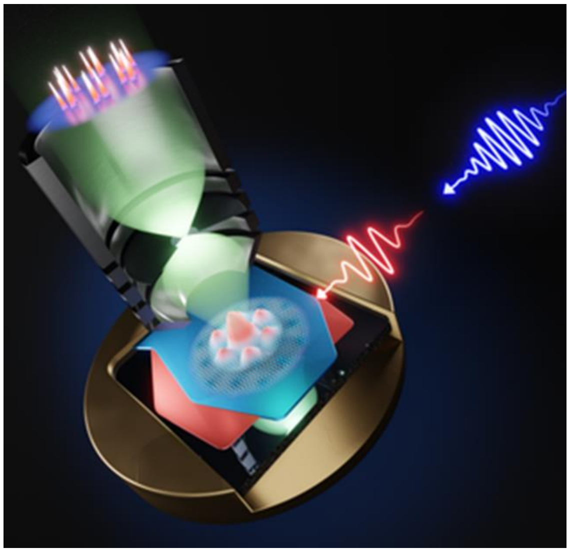

Transition metal dichalcogenides (TMDs) are an exciting model system to study ultrafast energy dissipation pathways, and to create and tailor new emergent quantum phases [1,2]. The versatility of TMDs results from the confinement of optical excitations in two-dimensions and the concomitant strong Coulomb interaction that leads to excitonic quasiparticles with binding energies in the range of several 100 meV. In TMD stacks consisting of at least two layers, the interlayer interaction can be precisely controlled by manipulating the twist angle: The misalignment of the crystallographic directions leads to a momentum mismatch between the high symmetry points of the hexagonal Brillouin zones. This strongly impacts the interlayer wavefunction hybridization, and, moreover, adds an additional moiré potential. Crucially, in this emergent energy landscape, dark intra- and interlayer excitons dominate the energy dissipation pathways. While these dark excitonic features are hard to access in all-optical experiments, time-resolved momentum microscopy [3] can provide unprecedented insight on these quasiparticles [4].

In my talk, I will present our recent results on the ultrafast formation dynamics of interlayer excitons in twisted WSe_2/MoS_2 heterostructures. First, I will report on the identification of a hallmark signature of the moiré superlattice that is imprinted onto the momentum-resolved interlayer exciton photoemission signal. With this data, we reconstruct the electronic part of the exciton wavefunction, and relate its extension to the moiré wavelength of the heterostructure. Second, I will show that interlayer excitons are effectively formed via exciton-phonon scattering, and subsequent interlayer tunneling at the interlayer hybridized Σ_W valleys on the sub-50 fs timescale. Finally, I will discuss our recent efforts to monitor the interlayer exciton formation dynamics with spatiotemporal resolution using femtosecond photoelectron dark-field microscopy.

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NON-EQUILIBRIUM DYNAMICS OF LASER-EXCITED ELECTRONS IN SOLIDS

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During femtosecond laser irradiation of solids, mainly the electrons in the material gain energy. Free electrons in the conduction band of a metal can directly absorb photons of visible light. In semiconductors and dielectrics, a band gap has to be overcome first, before further energy gain from the laser beam is possible. These absorption processes transfer the electronic system to a state of strong thermodynamic non-equilibrium, where the electronic energy distribution strongly differs from a Fermi distribution.

We simulate the dynamics of a large ensemble of excited electrons with help of complete Boltzmann collision integrals. For metals, we include the laser-excitation, electron-electron collisions as well as electron-phonon collisions [1]. For dielectrics, interband processes of energy- as well as particle transfer, such as secondary electron generation and Auger recombination, are included additionally [2]. The results show the athermal character of the non-equilibrium electron distribution, its thermalization to a Fermi distribution at elevated temperature and the energy loss caused by electron-phonon coupling.

We observe that electron-phonon interactions maintain a certain non-equilibrium of the electrons on a picosecond timescale. For metals with distinct features in the density of states, phonon emission results in energetically localized deviations of the electrons' distribution from a Fermi distribution [3]. For dielectrics, electron-phonon energy transfer continously drives a non-equilibrium between the energy content and the electron-hole density in the excited band system. This leads in turn to strong deviations of the electronic energy distribution from a thermal distribution on timescales up to the picosecond range.

In this presentation, we introduce our calculation method and the various relaxation processes, discuss their timescales and present examples of their mutual influence. We further show how such intertwined relaxation processes affect measurable or averaged quantities on ultrafast timescales, such as, e.g., the DC conductivity in noble metals, the magnetic polarization in itinerant ferromagnets, or the electron-phonon coupling strength in dielectrics.

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Observation of ultrafast interfacial Meitner-Auger energy transfer in a van der Waals heterostructure

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Interlayer charge- and energy transfer processes in atomically thin, layered van der Waals heterostructures are of fundamental importance for determining their properties in novel device concepts based on single active crystalline layers. In our work, we study the ultrafast excitation, relaxation and transfer processes in an epitaxial grown monolayer WSe_2/graphene heterostructure using time- and angle-resolved photoemission spectroscopy [1]. By measuring the non-equilibrium electronic structure, we identify a novel interfacial energy transfer mechanism: Meitner-Auger energy transfer, which describes the conversion of an exciton in the semiconductor to an intraband electron-hole pair in graphene, characterized by the excitation of deep-lying valence holes in graphene. Based on a systematic microscopic calculation of interlayer couplings, we identify Meitner-Auger energy transfer as the dominant transfer process surpassing the efficiency of the established Förster- and Dexter-type transfer.

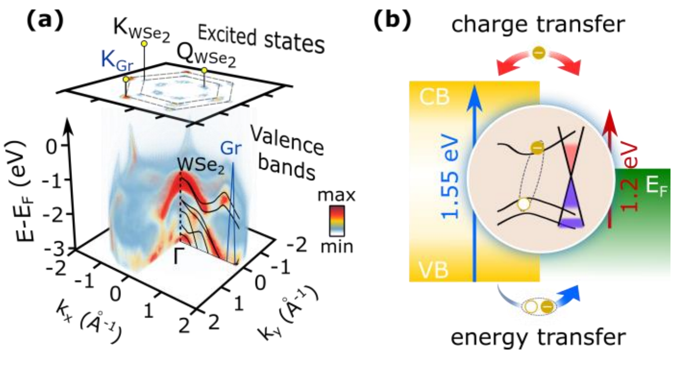

Figure 1 – (a) The 3D snapshot of the 4D data in ML-WSe_2/graphene heterostructure. (b) We selectively probe the intervalley charge and energy transfer by changing the pump wavelength.

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ORBITAL-RESOLVED OBSERVATION OF SINGLET FISSION

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Time- and angle-resolved photoemission spectroscopy (trARPES) provides a quantum-state-resolved picture of the ultrafast dynamics of quasi-particles in non-equilibrium states of matter. Utilizing a high-repetition-rate extreme ultraviolet laser source and a momentum microscope detector [1], we observe the formation and scattering of excitons resolved in energy, momentum, and time. We exemplify the signatures of excitonic states in trARPES for the layered semiconductor WSe_2. This spectroscopy reveals all key properties of the excitons like binding energy, exciton-phonon coupling, and the real-space distribution of the many-body wave functions through the Fourier transform of the photoelectrons' momentum distribution [2,3]. We then discuss exciton dynamics in a molecular crystal: ultrafast singlet fission in pentacene. This process converts a singlet exciton into two triplet excitons and thereby doubles the number of excited charge carriers. The primary step of singlet fission is the ultrafast creation of the correlated triplet pair. While several mechanisms have been proposed to explain this step, none has emerged as a consensus. The challenge lies in tracking the transient excitonic states. We track the primary step of singlet fission and obtain strong evidence for a charge-transfer mediated mechanism [4]. We gain intimate knowledge about the localization and the orbital character of the exciton wavefunctions recorded in momentum maps. This allows us to directly compare the localization of singlet and bitriplet excitons and decompose energetically overlapping states based on their orbital character. Orbital- and localization-resolved many-body dynamics promise deep insights into the mechanics governing molecular systems and topological materials.

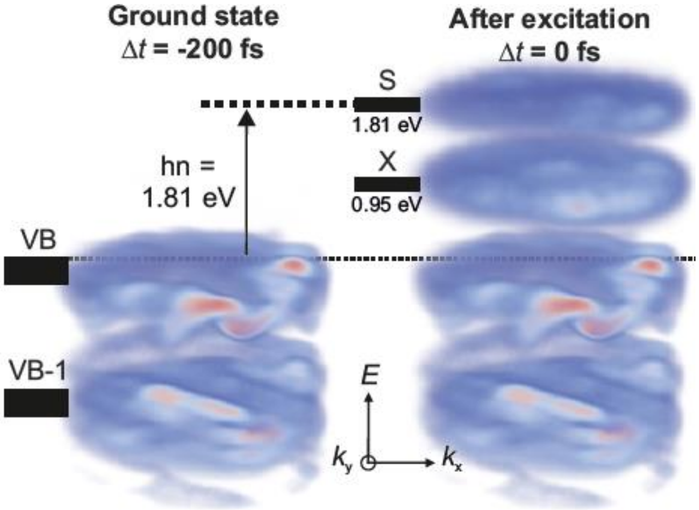

Figure 1 – Investigating singlet fission in pentacene with trARPES. Visualization of the energy-momentum distribution of the resonantly excited singlet at the moment of excitation (right). The singlet state features a pronounced charge-transfer character, indicated by a satellite signal X in addition to the Frenkel-like exciton signal S.

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PHOTON-INDUCED VERSUS PLASMON-INDUCED HOT ELECTRONS

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The mechanism of plasmonic enhancement in photochemistry, photocatalysis, and light-harvesting, and in particular the role of hot charge carriers, is still heavily debated. The question arises whether plasmon-induced and photon-induced hot charge carriers are fundamentally different or whether plasmonic enhancement is just an effect of field concentration that generates these charge carriers in larger numbers. A fundamental difference is known for bulk plasmon resonance [1-3], but this is far from clear for technologically important surface plasmons. Direct imaging of surface plasmon induced hot charge carriers could provide essential insights, but separating the influence of the driving laser, field enhancement, and fundamental plasmon decay has proven difficult.

Here we present an approach using a two-color femtosecond pump-probe scheme in time-resolved 2-photon photoemission (PEEM and momentum microscopy), supported by a theoretical analysis of the light and plasmon energy flow. We separate the energy and momentum distribution of plasmon-induced hot electrons from that of photoexcited electrons by following the spatial evolution of photoemitted electrons during the propagation of a surface plasmon polariton (SPP) pulse along a gold surface. We find plasmonic enhancement toward high excitation energies and small in-plane momenta, suggesting a fundamentally different mechanism of hot electron generation previously unknown for SPP [4].

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Probing (Dynamical) Orbital Properties of 2D Materials using Novel Dichroisms in (Time-Resolved) ARPES

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Angle-resolved photoemission spectroscopy - ARPES - is the most powerful technique to investigate the electronic eigenvalues (band structure) of crystalline solids. To completely characterize the electronic structure of (topological) materials, one needs to go beyond band structure mapping and probe the orbital texture, associated with Berry curvature and topological invariants. I will present new measurement methodologies in ARPES based on *i)* crystal rotation mimicking time-reversal symmetry operation [1,2], *ii)* continuous modulation of the ionizing radiation polarization axis [3], and *iii)* modulation of the light helicity [4]. I will show how such manipulation of the photoemission transition dipole matrix elements, complemented by minimal theory inputs, allows accessing the momentum-dependent orbital texture and Berry curvature. I will show how these novel dichroisms can be used to probe the emergence of topological Floquet-Bloch states in laser-driven transition metal dichalcogenides - TMDCs [5]. Last, I will briefly discuss the new beamline that we have developed at CELIA-Bordeaux, specifically designed for such novel dichroisms in trARPES (250kHz, 21.6eV, 120fs, and fully polarization tunable) [6].

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Probing and controlling phase transitions in low-dimensional materials using ultrafast low-energy electron diffraction

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Solid-state surface systems are particularly attractive because of their modified electronic, lattice and spin structures, resulting in strongly altered physical and chemical properties compared with the bulk [1]. We have recently developed ultrafast low-energy electron diffraction (ULEED) in a laser pump/electron-probe scheme to explore optically-induced structural dynamics at surfaces on their intrinsic time scales [2,3]. This talk will introduce the basic principles of ULEED and discuss our recent advances regarding the observation and control of surface-structural dynamics as well as phase transitions far from equilibrium. This involves, e.g., the tracking of nonequilibrium phonon populations via ultrafast diffuse scattering of low-energy electrons and the transfer of control tactics from femtochemistry to solids.

In this context, we report the coherent vibrational control over the phase transition in a quasi-one-dimensional Peierls insulator by manipulating the vibrational amplitudes of key lattice modes [4]. Specifically, we employ optical multi-pulse excitation schemes and monitor the structural transformation between the insulating (8×2) and the metallic (4×1) phase of atomic Indium wires [5,6] on Si(111) by ULEED. An analysis of the delay-dependent phase transition efficiency proves the distinct roles of shear and rotation phonons for the transition. Mode-selective excitation of these phonons enables us to explore the underlying potential energy surface (PES) and identify structural pathways across the transition state [7].

Furthermore, combining ULEED with *ab initio* molecular dynamics simulations, we prove the existence of a ballistic transformation mechanism, in which the concerted motion of the indium atoms can overcome the $(8\times2)\rightarrow(4\times1)$ barrier on the multidimensional PES, by harnessing the kinetic energy of the nuclei [7]. Our work illustrates that coherent excitation of collective modes evades entropic barriers and enables the dynamic control of materials functionality.

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PUSHING THE LIMITS OF LOCAL SPECTROSCOPY: RAMAN SCATTERING FROM A SINGLE ADATOM AND NANOCALE COHERENT PHONON SPECTROSCOPY

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Atomic-scale light confinement has recently been demonstrated in plasmonic cavities, where the ultimate confinement of the light field results from atomistic protrusions when a plasmonic “picocavity” is formed. Here we show that inelastic light scattering is dramatically enhanced by forming a quantum point contact (QPC) on a single adatom in a well-defined plasmonic picocavity that is controlled precisely using low-temperature scanning tunneling microscopy (STM). Raman spectra from a single silver atom on Ag(111) in the QPC regime exhibit salient spectral features originating from the interaction with both vibrations and electrons in the STM picocavity. The atomic-scale light confinement in the STM picocavity is reproduced by time-dependent density functional theory simulations including vibrational dynamics of the atomistic structures.

Furthermore, we demonstrate extreme spatial resolution using ultrafast laser-induced STM applied to local coherent phonon spectroscopy. Coherent phonons can provide microscopic insight into ultrafast lattice dynamics and coupling to other degrees of freedom. Excitation and relaxation of coherent phonons may be susceptible to the local nanoscale environment, calling for real-space observation of ultrafast lattice dynamics. We demonstrate nanoscale coherent phonon spectroscopy employing time-resolved STM in a plasmonic junction, and unveil spatial inhomogeneities of coherent phonon dynamics in ultrathin zinc oxide (ZnO) films with few nanometer spatial and femtosecond temporal resolution. The coherent phonons in ZnO are locally excited by the tightly-confined gap plasmon, and are probed via the photoinduced tunneling current through an electronic resonance of the ZnO film. In combination with tip-enhanced Raman spectroscopy and scanning tunneling spectroscopy, we reveal the involved phonon modes and a correlation of the local electronic structure with the coherent phonon dynamics.

Acknowledgements: The contributions by Shuyi Liu, Takashi Kumagai, Melanie Müller, Franco Bonafe, Heiko Appel, and Angel Rubio, are gratefully acknowledged.

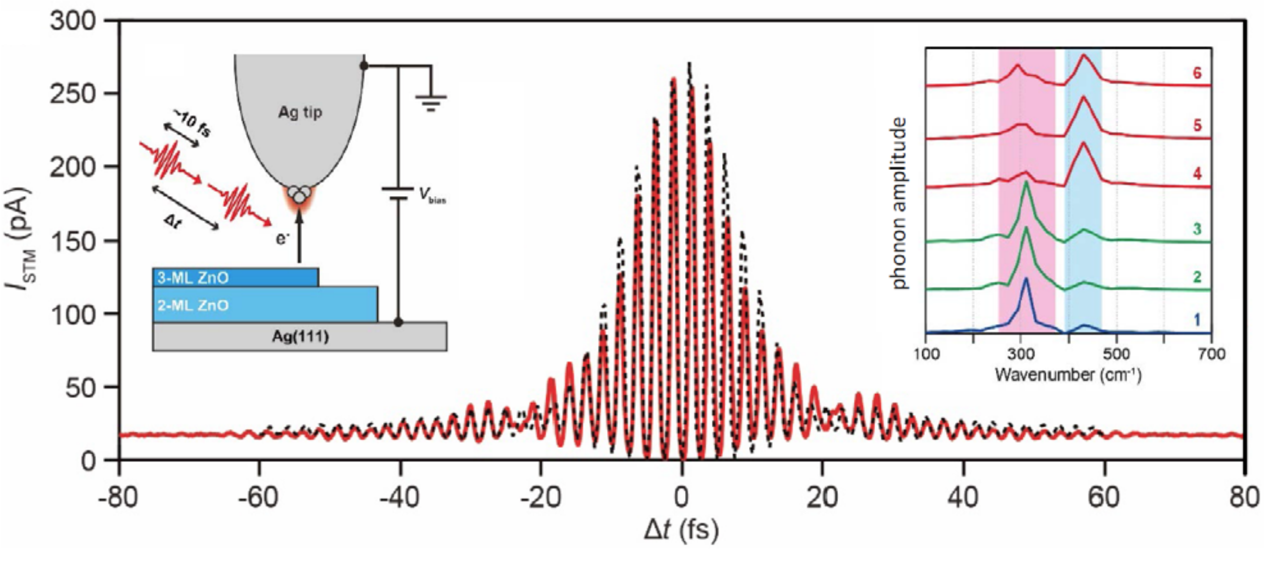

Figure 1: Ultrashort pulsed laser-induced STM in a plasmonic junction on Ag(111), 2-ML and 3-ML ZnO, respectively. Main panel: IAC trace of the photocurrent for a Ag tip–Ag(111) junction. The inset right shows FT spectra of the IAC traces at different sample positions on ZnO (with 2 nm steps).

Spin dynamics in Gd/Fe bilayers and its signature in the electronic structure

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Understanding ultrafast spin dynamics is not only a complex and fascinating challenge in fundamental physics, but carries the potential for magnetic recording based on all-optical switching (AOS) of the magnetic order [1]. AOS triggered by a single fs laser pulse was first observed in the ferrimagnetic alloy FeCoGd [2] and more recently in Pt/Co/Gd stacks [3]. The key to AOS is exchange of angular momentum between the oppositely aligned magnetic moments of the transition metal and rare earth sublattices [4]. Spin transport and exchange scattering are discussed as microscopic processes responsible for all-optical switching. However, triggered by the fs laser pulse, demagnetization occurs on a sub-ps timescale while reversal still requires >10 ps and reliable rewriting ~300 ps [5].

We will give microscopic insights to spin dynamics in Fe, Gd and Gd/Fe bilayers investigated by spin- and time-resolved photoemission as well as X-ray magnetic circular dichroism in reflection. Fe(2 nm)/Gd(5 nm) bilayers were grown on a W(110) single crystal. The in-plane magnetized bilayer shows a compensation point T_{cp} of 235 K, where $M_{Gd} = -M_{Fe}$ indicates that the whole 5-nm Gd film is ferromagnetic. Below T_{cp}, the magnetization of the Gd film M_{Gd} is aligned along the external field H. Above T_{cp}, M_{Fe} aligns parallel to H. At a starting temperature slightly above T_{cp}, we find ultrafast reversal of all Gd spins. Within 400 fs, the Gd XMCD contrast switches from +1 to -1. On the same timescale, the Fe film demagnetizes by only ~12%.

In a second experiment we used time- and spin-resolved photoemission spectroscopy to study the Gd surface state on the Gd/Fe bilayer. We show that spin transport between Gd and Fe layers leads to a 20% drop of the spin polarization at the Gd surface within 100 fs. The Fe layer acts as a spin filter so that mainly the majority spin population of the Gd surface state decreases. This is in clear contrasts to a pure Gd film on W(110), where spin transport into the tungsten substrate can be neglected and local spin-flip processes do not alter the spin polarization of the Gd surface state but reduce the exchange splitting by a spectral upshift. This significant difference is corroborated by the electron temperature. In the bilayer, hot electrons are efficiently removed from the Gd layer before they can heat the phonon system while in pure Gd/W(110) electron and phonon subsystems equilibrate at an elevated temperature after 1.5 ps. Moreover, a re-magnetization process characterized by an increased exchange splitting acts as negative feedback to the spin-transport-driven demagnetization. Our results evidence that ultrafast spin transport can be a main driver for the magnetization dynamics in ferrimagnetic Gd/Fe bilayers and is important for AOS in layered systems. We observe particular spectral signatures in the Gd electronic structure which reveal microscopic insights into ultrafast spin dynamics.

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This work contains collaborations of the projects A01 Weinelt, A03 Pontius/Schüßler-Langeheine (Helmholtz-Zentrum Berlin), and B02 Woltersdorf (Martin-Luther Universität Halle-Wittenberg) of the **CRC/TRR 227** Ultrafast Spin Dynamics and is funded by the Deutsche Forschungsgemeinschaft.

SUBCYCLE TIME-RESOLVED ARPES OF LIGHTWAVE-DRIVEN PROCESSES*

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Intense light fields offer spectacular opportunities to control and manipulate the properties of solids. Prominent examples are photo-induced insulator-to-metal transitions, light-induced superconductivity, the creation of Floquet-Bloch states and light-wave electronics. And there exist numerous other ideas for all-optical shaping of materials, to reveal hidden states of matter or to create novel quantum states and topological states [1]. Experiments along these lines can benefit tremendously from the capability of time- and angle-resolved photoelectron spectroscopy (tr-ARPES) to probe the electronic structure of the material. This holds in particular when methods beyond common pump-probe schemes are applied and the transient changes are detected with subcycle time resolution during the optical excitation.

As an example, I will discuss Floquet-Bloch sidebands that build up when the surface state of a topological insulator is driven with intense mid-infrared fields [2]. Starting with strong intraband currents, we observe how Floquet replicas emerge within a single optical cycle and how electrons in high-order sidebands scatter into bulk states and dissipation destroys the Floquet bands. A wealth of new applications of lightwave ARPES become available by employing laser high-harmonics for photoemission. In combination with orbital tomography [3], lightwave ARPES should make it possible to take slow-motion movies of molecular orbitals during charge transfer processes, during molecular vibrations, and during surface chemical reactions.

* Experiments performed in collaboration with the groups of Rupert Huber (Regensburg), Stefan Tautz (Jülich), and Robert Wallauer (Marburg).

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Tracking Electrons, Atoms and Quasiparticles in Real-Space and Real-Time

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Abstract

Chemical transformations in molecules are a consequence of valence electron motion and its eventual coupling to atomic motion, hence, tracking valence electron motion at the orbital level is the key to understanding and taming such transformations. Scanning tunnelling microscopy (STM) can passively and locally probe the valence electron density in molecules. Contemporary techniques in attosecond science[1,2], on the other hand, can generate and track the temporal evolution of a coherent superposition of quantum states of valence electrons by using strong laser fields, which can be probed only non-locally. In absence of the capability to trigger and probe electron dynamics at the single-orbital level, electron motion could only be inferred by reconstruction. In the talk, I will show you how dynamics of coherent superposition of valence electron states generated by < 6 femtosecond long carrier-envelope-phase (CEP) stable laser pulses, can be locally probed with picometer spatial resolution and 300 attosecond temporal resolution simultaneously, at the single orbital-level with the help of an STM, defying the previously established fundamental space-time limit[3,4].

Even though, by integrating ultrashort laser pulses with an STM we are now able to trace electron dynamics directly in molecules, the information about vibrational dynamics and its coupling with electronic motion is still limited. In order to overcome this barrier, we have recently integrated time-resolved tip-enhanced Raman spectroscopy[5] in conjunction with the experimental capability of tracking the electron dynamics. I will show the recent experimental results of tracking the phonon coherence and dephasing dynamics in a single graphene nanoribbon by the application of coherent anti-Stokes Raman spectroscopy (CARS) in an STM. We envisage that it will be soon possible to see a chemical bond formation/breaking dynamics through a transition state at the orbital level.

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Transient Optical Harmonics in Two-dimensional Layered Materials

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Recently, nonlinear optics in two-dimensional (2D) layered materials has attracted great attention [1-8]. Here, we will present our recent results on transient optical harmonics (second harmonics [6], third harmonics [7] and high-order harmonics [8]) in transition metal dichalcogenides. Our results indicate that transient harmonics is not only a promising characterization technique for fundamental science (e.g., carrier dynamics and excitonic states in monolayer semiconductors and their heterostructures, and fundamental nonlinear optics in 2D materials), but also a potential platform for disruptive photonic and optoelectronic applications, including all-optical modulation and computing.

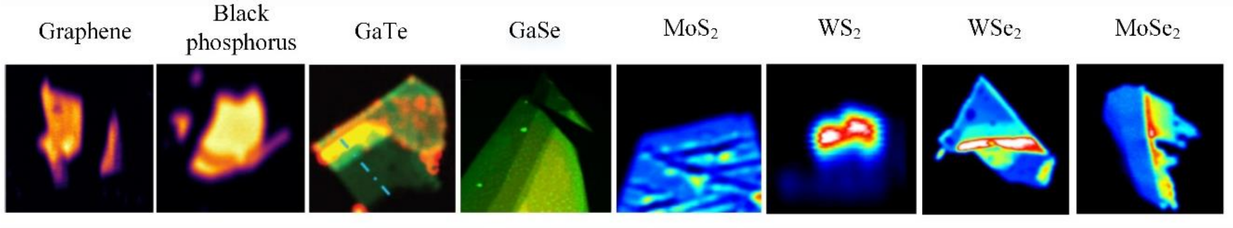

Figure 1. Nonlinear optical images of different two-dimensional layered materials[1].

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TUNABLE NONLINEAR PHONON RESPONSE OF MONOLAYER hBN

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Investigating nonlinear optical effects that occur during the interaction of light with materials at the atomic level is important for opto-electronic applications based on nanomaterials. Using first-principles calculations, we investigate the nonlinear optical response of monolayer hBN and demonstrate the plausibility of electrical tunability of vibrational modes with potential application in opto-electronics within the mid-infrared phonon-polariton region [1].

We proceed by first obtaining the potential energy surface and induced dipole density as a function of atomic positions. Then, solving the equation of motion either in the time domain and in a perturbative scheme, we obtain the nonlinear susceptibilities associated with second- and third-harmonic generation, as well as the Kerr effect. Besides the strong nonlinear response that situates this material on par with graphene as the most nonlinear materials in the mid-infrared region, we foresee that substantial frequency shifts exceeding the spectral widths of the phonon-polariton modes can be realized by applying in-plane DC fields that are attainable using existing lateral gating technology. We also predict quantum blockade at the few-quanta level in nanometer-sized hBN structures, with potential application in quantum technologies.

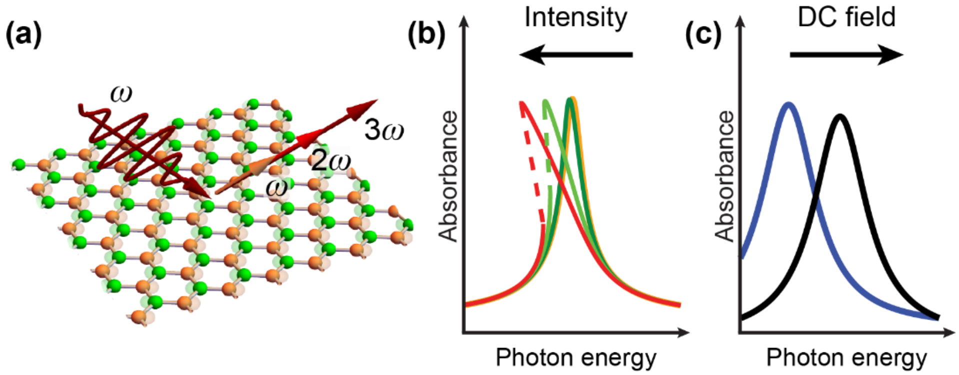

Figure 1 - (a) Illustration of monolayer hBN under normally impinging light irradiation with linear polarization along one of the B-N bond directions. (b,c) Spectral dependence of the absorbance for (b) increasing light intensity and (c) an applied DC field.

In summary, our work demonstrates that phonon polaritons in monolayer hBN with strong nonlinear response, long lifetime, and optical and electrical modulation configure a promising, robust platform that opens new possibilities in mid-infrared nonlinear optics with application in opto-electronics and quantum optics.

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Ultrafast charge transfer in heterostructures of two-dimensional materials

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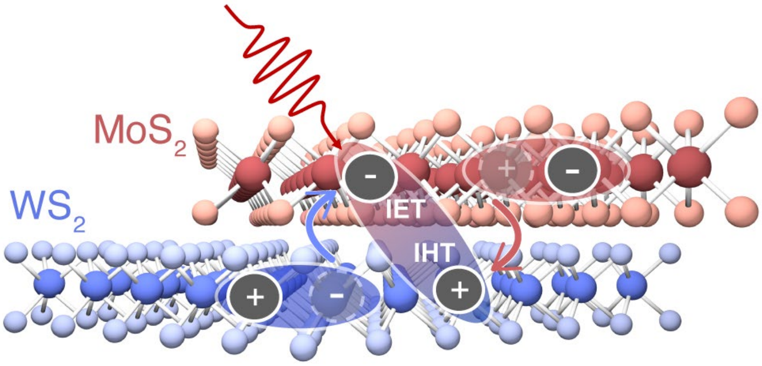

Heterostructures (HS) of two-dimensional materials offer unlimited possibilities to design new materials for applications to optoelectronics and photonics. In such HS the electronic structure of the individual layers is well retained because of the weak interlayer van der Waals coupling. Nevertheless, new physical properties and functionalities arise beyond those of their constituent blocks, depending on the type and the stacking sequence of layers. In this presentation we use high time resolution ultrafast transient absorption (TA) and two-dimensional electronic spectroscopy (2DES) to resolve the interlayer charge scattering processes in HS.

We first study a WSe_2/$MoSe_2$ HS, which displays type II band alignment with a staggered gap, where the valence band maximum and the conduction band minimum are in the same layer. By two-colour pump-probe spectroscopy, we selectively photogenerate intralayer excitons in $MoSe_2$ and observe hole injection in WSe_2 on the sub-picosecond timescale, leading to the formation of interlayer excitons (ILX). The temperature dependence of the build-up and decay of interlayer excitons provide insights into the layer coupling mechanisms [1]. By tuning into the ILX emission band, we observe a signal which grows in on a 400 fs timescale, significantly slower than the interlayer charge transfer process. This suggests that photoexcited carriers are not instantaneously converted into the ILX following interlayer scattering, but that rather an intermediate scattering processes take place We then perform 2DES, a method with both high frequency and temporal resolution, on a large-area W_S2/MoS_2 HS where we unambiguously time resolve both interlayer hole and electron transfer with 34 ± 14 and 69 ± 9 fs time constants, respectively [2]. We simultaneously resolve additional optoelectronic processes including band gap renormalization and intralayer exciton coupling.

Finally, we investigate a graphene/WS_2 HS where, for excitation well below the bandgap of WS_2, we observe the characteristic signal of the A and B excitons of WS_2, indicating ultrafast charge transfer from graphene to the semiconductor [3]. The nonlinear excitation fluence dependence of the TA signal reveals that the underlying mechanism is hot electron/hole transfer, whereby a tail the hot Fermi-Dirac carrier distribution in graphene tunnels through the Schottky barrier. Hot electron transfer is promising for the development of broadband and efficient low-dimensional photodetectors.

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Ultrafast exciton dynamics in atomically thin semiconductors

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Monolayer transition metal dichalcogenides (TMDs) and related van der Waals heterostructures exhibit a rich exciton physics including bright and a variety of dark states as well as spatially separated interlayer excitons. Solving 2D material Bloch equations for excitons, phonons and photons, we obtain a microscopic access to the interplay of optics, ultrafast dynamics and diffusion of excitons in these technologically promising materials. In joint theory-experiment studies we shed light on the importance of momentum-dark excitons in low-temperature photoluminescence spectra [1], temperature-resolved exciton-exciton annihilation [2], and exciton (anti-)funnelling [3] in strained TMD monolayers (Fig. 1a). Furthermore, we investigate twist-angle dependent trapping of excitons in moiré potentials [4] as well the ultrafast charge transfer dynamics in TMD heterostructures [5] (Fig. 1b).

The gained microscopic insights into the spatiotemporal exciton dynamics are crucial for understanding and controlling many-particle phenomena governing exciton optics, dynamics and transport in technologically promising 2D materials and related heterostructures.

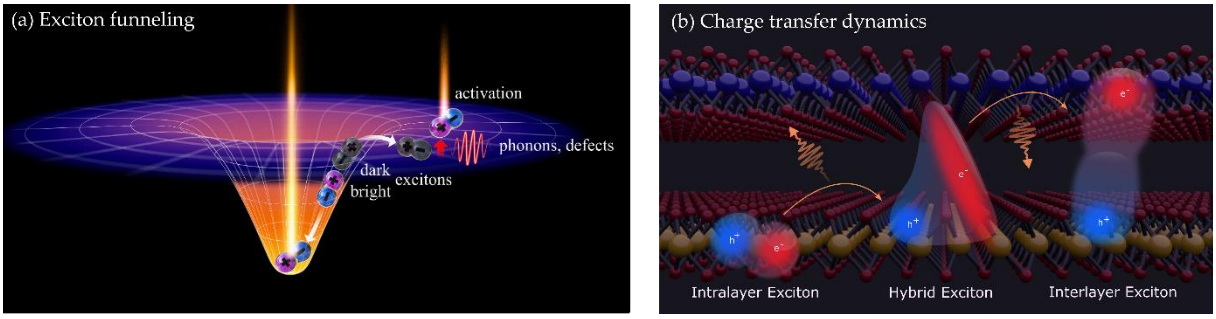

Figure 1: (a) Illustration of funneling of bright excitons and anti-funneling of dark excitons in strained TMDs. Dark excitons can be activated by phonons and/or defects making them visible in optical spectra. (b) Sketch of the charge transfer process in TMD heterostructures. Starting from an exciton localized in the bottom layer, phonon-mediated scattering to a hybrid exciton state allows for the charge transfer to the upper layer.

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Ultrafast Light to Steer Electroncs and Switch Magnetic Moments

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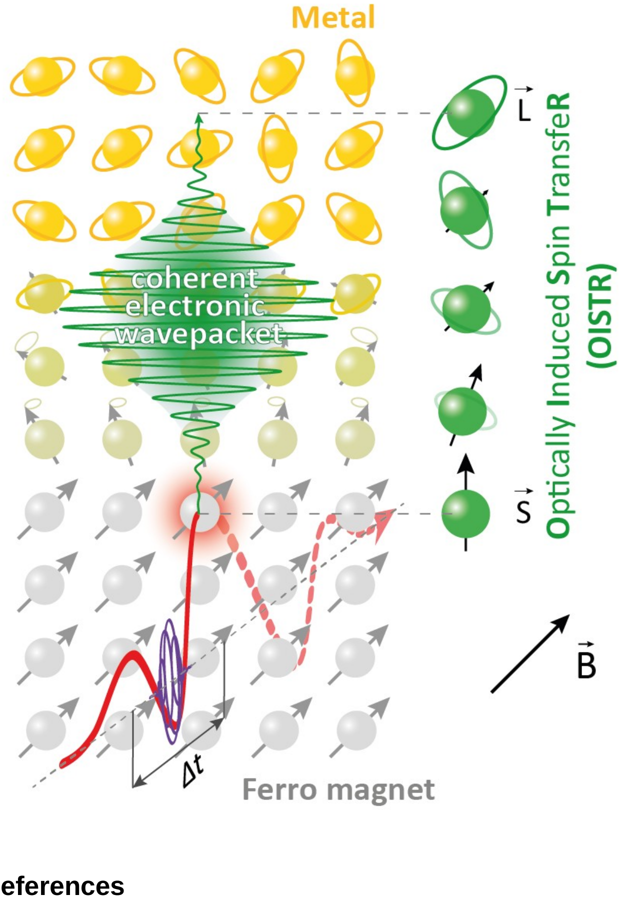
The enormous electric field strength of ultrafast laser waveforms allows to steer electronic motion and control electronic excitation so fast, that secondary processes disrupting coherence and striving for an equilibrium have hard time catching up – even in condensed phase systems.

We investigate the opportunities this temporal segregation offers to transfer coherent control ideas in atomic and molecular ensembles to solids. I will discuss two experiments demonstrating that single cycle optical fields allow manipulating electronic and spin degrees of freedom in solid state systems at optical clock rates faster than de-coherence. Sub-femtosecond carrier injection in the band-structure of dielectrics interfaced to electronic circuitry demonstrate the feasibility of ultrafast, coherent optoelectronic applications up to 1 Petahertz frequencies[1].



As a corollary of this ultrafast coherent modification of the electronic system, in suitably chosen hetero-structures also the spin system can be manipulated coherently.
Optically induced spin transfer is demonstrated as a route to the direct, all-optical manipulation of macroscopic magnetic moments on previously inaccessible attosecond timescales[2] and a path to arrange magnetic domain patterns with light.

Could this provide a pathway to turn something *more magnetic* via illumination? As an outlook of the talk I will discuss our recent activities targeted at inducing magnetic order via light in ferromagnetic/dielectric hetero-structures.

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Ultrafast modification of the spin-dependent energy level alignment of molecular/WSe$_2$ heterostructures

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One of the most crucial challenges in the field of 2D van der Waals materials is to devise new approaches to design their electronic and optical properties on the nanoscale. For transition metal dichalcogenides (TMDCs), this is mainly achieved by the formation of 2D heterostructures with other 2D materials. While this approach was successfully employed to tune the interfacial properties of such systems, it still reveals limitations in the tuneability of the interfacial energy level alignment as well as in the lateral dimensions of the heterostructures.

In this contribution, we introduce an alternative route to transiently functionalize the properties of TMDCs by the adsorption of molecular materials. As an exemplary case, we focus on a heterostructure consisting of the TMDC bulk crystal WSe$_2$ and the prototypical molecule C$_{60}$. Both were selected due to their intriguing optical and spin-dependent electronic properties. On the one hand, the WSe$_2$ valence band structure reveals an overall vanishing spin polarization despite the layer- and valley-dependent spin polarization of the individual tri-layers [1]. On the other hand, the excited state dynamics of fullerenes is dominated by charge transfer excitons that can transiently manipulate the energy level alignment of the surrounding material [2,3].

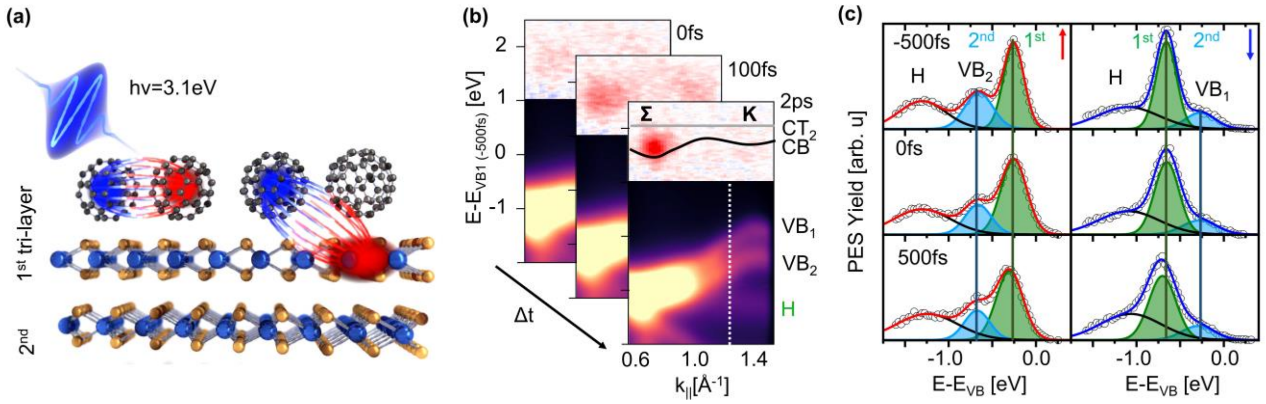

Fig.1 (a) Illustration of the C$_{60}$/WSe$_2$ heterostructure as well as the interfacial charge transfer after the optical excitation of the C$_{60}$ layer. (b) Snapshots of the time-resolved ARPES data set. (c) Spin-resolved spectral yield of the valence band structure at selected time delays illustrating the transient layer dependent WSe$_2$ valence band shift.

Using time-, spin-, and momentum-resolved photoemission, we monitor the ultrafast carrier dynamics as well as transient modifications of the valence band structure of the C$_{60}$/WSe$_2$ heterostructure. After the resonant optical excitation of the C$_{60}$ layer, we observe an ultrafast electron transfer from the C$_{60}$ into the WSe$_2$ layer as illustrated in Fig.1(a), and (b). The resulting charge separation is accompanied by an interfacial dipole that causes a transient shift of the C$_{60}$ and WSe$_2$ valence states. Crucially, the magnitude of the WSe$_2$ valence band shift is largest for the WSe$_2$ layer in direct contact with C$_{60}$ and decreases with increasing distance from the interface, see Fig. (c). This transiently lifts the spin-degeneracy of neighboring WSe$_2$ layers at the interface. Our findings hence provide a new approach for the ultrafast manipulation of spin functionalities of a molecular/TMDC heterostructures by optical excitation.

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ULTRAFAST NANO-IMAGING OF FAR-FROM-EQUILIBRIUM QUANTUM DYNAMICS

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Quantum phase transitions, polaronic carrier transport, coherent vibrations driving singlet fission, or electronic energy transfer in light-harvesting complexes emerge from the interplay of the elementary electronic, vibrational, and phononic quantum states. *Ultrafast nano-imaging* based on tip-enhanced femtosecond spectroscopy has demonstrated access to ultrafast carrier dynamics on the nanoscale in semiconductor, correlated-electron, or polaritonic materials. However, mostly limited to strong and short-lived carrier and collective polaritonic excitations, the contrast obtained to date has remained insufficient to probe with excited state specificity many-body interactions induced by strong perturbation among charge carriers, lattice phonons, or molecular vibrations.

Here, we demonstrate a generalizable approach to ultrafast nano-imaging based on non-degenerate *heterodyne* pump-probe infrared scattering scanning near-field optical microscopy (HPP IR *s*-SNOM) with low repetition-rate *modulated excitation*, which provides *simultaneous space, time, and frequency resolutions* with high sensitivity (Fig. 1A) [1]. A modulated femtosecond pump pulse drives the system into an excited state, followed by infrared heterodyne probing of the transient low-energy electronic and vibrational response. The induced *third-order nano-localized polarization* is isolated by sideband lock-in detection and directly detected interferometrically in the time-domain. Ultrafast HPP IR *s*-SNOM nano-imaging thus provides the time-resolved analogue of ground-state nano-FTIR spectroscopy, selectively resolving the transient and nano-localized *excited-state response*.

As representative applications to quantum materials, we resolving the transient domain dynamics in nano-imaging the electron dynamics associated with the ultrafast photoinduced insulator-to-metal transition (IMT) in vanadium dioxide (VO_2) (Fig. 1B) [1,2]. In another application, we directly resolve heterogeneity in polaron-cation coupling that controls the photovoltaic response in triple cation perovskite by imaging both the excited state vibrational dynamics as well as the coupled polaron dynamics (Fig. 1C-D) [3]. Further, imaging carrier dynamics and resolving the transient phonon softening as a local probe of substrate temperature, we establish ultrafast infrared nano-thermometry as a new tool to image thermal transport at semiconductor hetero-interfaces imaging the heterogeneity in ps thermalization at the example of the WSe_2/SiO_2 heterostructure [4].

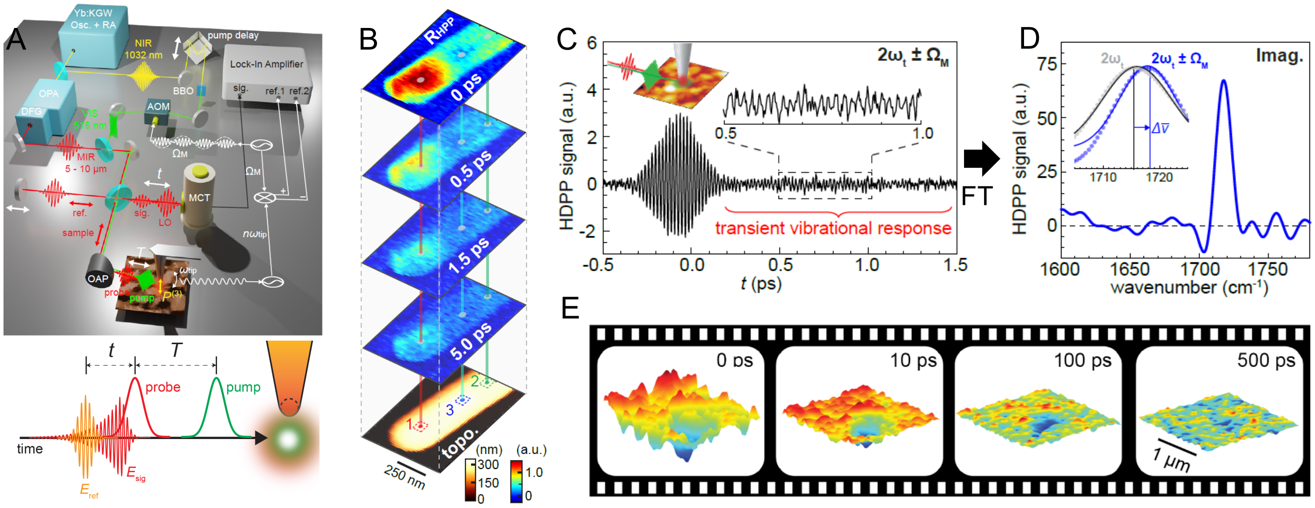

Fig. 1. *(A) Ultrafast nano-imaging with interferometric heterodyne detection (HPP IR s-SNOM) for quantitative excited state contrast. (B) Example of nano-imaging the ultrafast insulator-to-metal transition in VO_2. (C,D) Ultrafast local probe nano-spectroscopy of triple-cation perovskite excited state vibrational dynamics. (E) Ultrafast movie resolving associated transient heterogeneity of polaron-lattice dynamics.*

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Ultrafast polarization nanoscopy of charge carrier dynamics and correlations in 2D systems

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Advancements in electronics, quantum- and nanotechnology require nanoscale understanding of the internal processes in custom-tailored materials. Since these elementary building blocks of condensed matter are in constant motion, still images are not enough.

In this talk, I will show how femtosecond polarization nanoscopy unravels the dynamics of photoexcited charge carriers in custom van der Waals (vdW) heterostructures. I will introduce a novel terahertz nanoscopy technique to trace charge carrier dynamics in conducting and non-conducting materials [1]. We demonstrate ~40 nm spatial and sub-cycle temporal resolution and non-invasively probe the interlayer tunneling across an atomically sharp WSe_2/WS_2 interface. We see pronounced variations in the formation and annihilation of optically bright and dark excited states as a result of nanoscale strain and changes in atomic registry. Our results demonstrate that ultrafast nanoscopy is an indispensable tool to study intrinsically disordered materials like vdW heterostructures.

On a WSe_2 homobilayer, we precisely tune the density of excitons by photoinjection and observe the transition of a gas of strongly bound excitons into an electron-hole plasma. By revealing this excitonic Mott transition on the nanoscale, we circumvent averaging over unavoidable nanoscale inhomogeneities and extract the true nature of the process. We find a continuous transition with spatial variations which are not correlated with topography. Our results indicate that the exciton binding energy can be modulated on length scales inaccessible to other non-contact ultrafast probes. In the future, the technique could resolve the interplay between excitons and a broad variety of quantum phases in real space.

Furthermore, lightwave scanning tunneling microscopy was reported to induce local forces, which drive a coherent structural motion of a single-molecule switch in its electronic ground state [3]. The molecular switch even allows for quantitative sampling of atomic-scale waveforms in the tunnel junction [4]. I will discuss how these breakthroughs and their combination into ultrafast multi-messenger scanning probe microscopes could soon allow us to tailor (bio)chemical reactions or ultrafast phase transitions, on their intrinsic atomic-scale spatial and fs-temporal scales.

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ULTRAFAST STRUCTURAL DYNAMICS IN THE AU/SI(553) ATOMIC WIRE SYSTEM: FEMTOSECOND CHARGE TRANSFER AND DISORDER INDUCED METASTABILITY

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Deposition of 0.5 ML Au on the stepped Si(553) surface results in the formation of Au double-strand metallic atom wires with a two-fold periodicity along the terraces of the vicinal Si surface. The Si step edge atoms exhibit a three-fold periodicity, due to the presence of a chain of half occupied dangling bonds hosting an antiferro-magnetically coupled spin chain [1,2]. The system undergoes an order-disorder phase transition at Tc ≈ 100 K where the thermal creation of soliton-antisoliton pairs destroys the long-range ordering, accompanied by a 2D-1D dimensional crossover [3].

The ultrafast structural dynamics in this atomic wire system is studied by time-resolved ultra-fast reflection high energy electron diffraction. Upon impulsive excitation with an intense fs-optical pulse the streak-like intensity, indicative for the two-fold periodicity of the dimerized Au atoms, decreases on a sub-picosecond timescale. This reflects transient weakening of the dimerization within the Au double-strand which goes hand-in-hand with an in-surface charge transfer to the Si dangling bond chain.

The recovery of the ground state is facilitated by a two-stage process on picosecond to nanosecond time scales. Both processes exhibit a temperature dependence as well as a pronounced fluence dependence of the corresponding recovery time constants indicative for kinetic limitations during recovery: The greater the fluence of the excitation pulse, the faster the return to the ground state. Metastability of the excited state is facilitated by photo-doping induced disorder of the Si dangling bond chain where soliton-antisoliton pairs are excited. Weak energy barriers preventing the mobility of the solitons hinders the immediate recovery to the ground state which proceeds through mutual annihilation of the soliton and antisoliton.

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ULTRAFAST TRANSPORT AND ENERGY RELAXATION OF HOT ELECTRONS IN Au/Fe/MgO(001) INVESTIGATED BY LINEAR TIME-RESOLVED PHOTOELECTRON SPECTROSCOPY

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Optically excited electrons and holes are of particular interest in solid-state physics because analysis of their dynamics allows a microscopic understanding of the interactions in non-equilibrium states. Excited charge carriers in metals and semiconductors relax on a femto- to picosecond timescales due to electron-electron(e-e) and electron-phonon (e-ph) scattering. Here we want to discern the relaxation by such local inelastic processes and transport effects. To analyze the ultrafast dynamics of charge carriers, femtosecond time-resolved linear photoelectron spectroscopy was applied. This work is a continuation of previously performed two-photon photoelectron spectroscopy experiments [1], which provides data close to the Fermi energy E_F. Here, we report on first experimental results obtained by using 1.55 eV pump and 6 eV probe photons on an Au/Fe/MgO(001) epitaxial heterosystem. By comparing results obtained in our pump-probe experiments by Au side pumping as well as Fe side pump excitation, both with Au side probing, we can separate the contribution of transport from local effects, like electron-electron (e-e) and electron-phonon (e-ph) scattering. In case of the Fe side pump configuration, hot electrons are excited in the Fe layer, and subsequently injected into the Au layer. Then the electrons, that propagate to the surface and are probed by photoelectron emission spectroscopy with 6 eV photons. In the Fe side pumped data, we observe differences in relaxation compared to the Au pumped data. While in Au side pumping we see the maximum intensity for all observed energies at the temporal pump-probe overlap, Fe side pumping shows an increase of intensity in lower energies at later times. We attribute this difference to transport effects due to propagation of the hot electrons through the Au layer. Since the transport occurs on similar timescales as the e-e scattering, we conclude on a (super)-diffusive transport regime. We will present an energy E and Au thickness d_{Au} dependent analysis of the propagation and energy density $U(E,d_{Au})$ of electrons and holes above and below E_F. In Au side pumping we observe efficient transport away from the probed area into the Fe layer, which acts as an electron sink [2], because of its smaller scattering times. For the thinnest investigated Au layer of 5 nm Au side pumping and Fe side pumping cannot be discerned in the relaxation dynamics. We explain this observation by excitation of the Fe layer in Au side and Fe side pumping since 5nm Au transmit more pump light than it absorbs. Furthermore, a slow decaying intensity component after 500 fs in the vicinity of E_F, which persists at longer time delays up to 2 ps, is assigned to effects of e-ph coupling. Local relaxation and non-local transport mechanisms will be discussed.

This work was funded by the Deutsche Forschungsgemeinschaft through the Collaborative Research Center CRC 1242 (project number 278162697).

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Ultrafast Tunnel Magnetoresistance – A Photoemission Study

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Ultrafast light-field control of spin dynamics and magnetic moments paves the way for future coherent spintronic applications, spin transistors and data storage by establishing optical frequencies as their speed limit. Although the ultrafast manipulation of spins is restricted by the weak coupling between light and spin, a recent experimental and theoretical study [1] demonstrated the light-wave control of magnetic moments on a sub-fs time scale. Based on this work we set up an experimental program dedicated to the investigation of magnetization dynamics on surfaces. Magnetic tunnel junctions (MTJ), thereby, have been arousing ongoing interest due to their peculiar spin-transfer mechanism since the discovery of the large tunnel magnetoresistance. However, a complete microscopic understanding of the underlying physics is still lacking. We put the spotlight on the interface between a MgO-based tunnel barrier and a CoFeB wedge-like electrode in order to disentangle intertwined phenomena which come along with ultrafast demagnetization. Therefore, we developed a new visible photon energy microscopy scheme. By employing circularly-polarized visible light pulses and utilizing magnetic circular dichroism in a two-photon-photoemission (2PPE) experiment we establish a direct route to the buried interface, providing insight into the spatial arrangement and the electronic behaviour of the magnetic pattern near the Fermi level. Optical pumping, furthermore, reveals an unexpected response which subdivides the probed electronic system into two sections. Close to the Fermi level typical demagnetization curves appear with time-scales comparable with results from time-resolved magneto-optical Kerr effect measurements (TR-MOKE), whereas for specific energy/momentum intervals we observe a light-induced increase in magnetic moment. Together with DFT calculations we try to shed light on the microscopic processes involved in the ultrafast demagnetization and, especially, emphasize on the peculiarity of MTJs acting as an energy- and spin-filter.

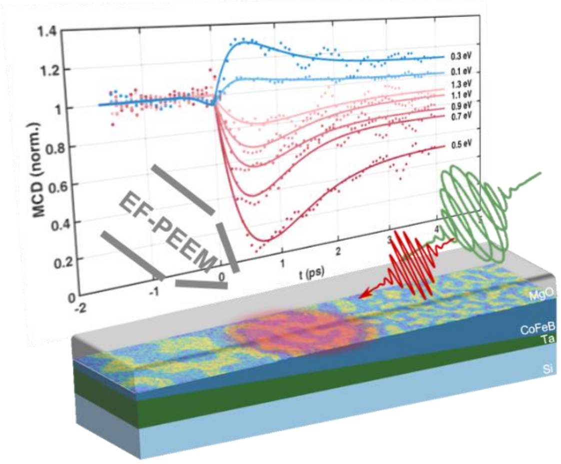

Figure 1 – A time-resolved and energy-filtered PEEM study on a half magnetic tunnel junction

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Unraveling Electron-Phonon and Phonon-Phonon Coupling in Momentum and Time with Ultrafast Electron Diffuse Scattering (UEDS)

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The nature of the couplings within and between lattice and charge degrees of freedom is central to condensed matter and materials physics. These interactions are essential to phenomena as diverse as superconductivity, charge density waves and carrier mobility in semiconductors and metals. Despite their fundamental role, detailed momentum-dependent information on the strength of electron-phonon coupling (EPC) and phonon-phonon coupling (PPC) across the entire Brillouin zone has proved to be very difficult to obtain.

In this talk I will describe an emerging pump-probe technique, ultrafast electron diffuse scattering (UEDS), that provides such information from the perspective of the phonon system directly [1,2]. Two recent examples of the application of UEDS to layered (2D) materials will be the focus. First, in the thermoelectric material SnSe – a strongly polar semiconductor – we directly observe the phonon dressing processes that yield carrier localization and polaron formation with UEDS [3]. In SnSe these phonon dressing dynamics are profoundly bimodal, with the fast (300 fs) process associated with the formation of a quasi-1D lattice distortion and a relatively large polaron and the slower (4 ps, an order of magnitude slower timescale) process associated with small polaron formation. The observations in SnSe are consistent with electron and hole polarons being different sizes, or the process of polaron formation being instrincially bimodal for both carriers in a manner reminiscent of Lars Onsager’s inverse snowball effect. Second, the extension of UEDS to a MoS_2 monolayer heterostructure will be demonstrated. These results reveal substrate dielectric screening of the electron-phonon interaction within the monolayer as well as the monomentum-dependent carrier-phonon equilibration. These results are compared directly against ab-initio simulations of these processes.

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POSTER
PRESENTATIONS

CARRIER RELAXATION IN UN-DOPED MONOLAYER MoS$_2$ THROUGH DEFECT STATES AND DEFECT ASSISTED AUGER RECOMBINATION

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Light-matter interaction in 2D materials is of great interest for a variety of photonic and optoelectronic applications. A strong confinement effect and electro-magnetic interaction among the charge carriers in these materials leads to a high exciton binding energy, strong exciton-exciton interactions and a large Auger capture rate. Due to the rising demand of 2D materials like Molybdenum Disulfide (MoS$_2$) monolayer in lasers, compact optical parametric amplifiers, and high-power detectors it is essential to study such material at highly excited conditions. Earlier experimental transient absorption measurement on *n*-doped MoS$_2$ with femtosecond time resolution shows that the inverse carrier lifetime increases linearly with the excitation fluence. Such dependence was explained using defect assisted Auger processes [1,2]. Experiments performed on an un-doped monolayer (ML) MoS$_2$ shows an initial fast fluence independent decay of the transient absorption signal. We show here that the dependence of the initial decay time on the fluence could be explained by just the capture of carriers at lower excitation fluences and decay to defect states and defect assisted Auger recombination at higher excitation fluences. Transient absorption ($\Delta\alpha$) measurements performed on a single un-doped ML MoS$_2$ by directly exciting and probing at the *A*-exciton shows an initial reduction in transmission within the duration of pump pulse followed by a fast recovery [3]. The fluence dependence of the inverse of the best fit time constant τ is shown in Fig 1 (a). At lower fluences (< 3 mJcm^{-2}) the $1/\tau$ remains nearly constant around 2 ps^{-1}. On the other hand, at higher fluences the measured $1/\tau$ increases linearly with pump fluence.

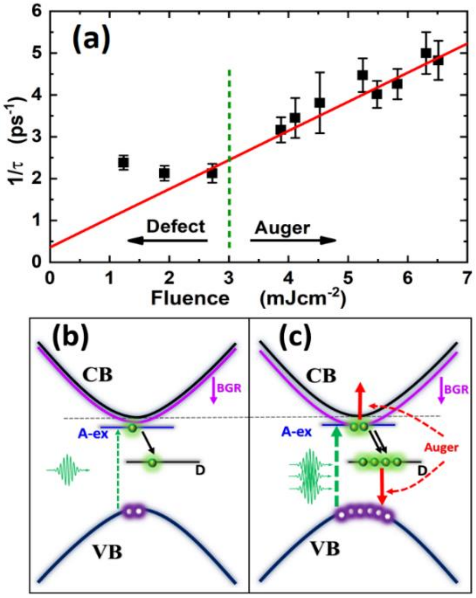

Figure 1: The fluence dependence of the measured relaxation rate $1/\tau$. Points are the experimental data. The line is the linear fit to data above 2.5 mJcm^{-2}; (b) and (c) are schematics for the physical processes responsible at the lower and higher fluence regime.

Earlier transient studies on *n*-doped MoS$_2$ by H. Wang, *et al.*, showed a continuous increase in $1/\tau$ without a constant $1/\tau$ at the lower fluence regime. It has been proposed that since the sample is *n*-doped, the defect states were completely filled before the arrival of the pump pulse and aids in the Auger process once excited by the pump pulse [1,2]. Defect states in the ML MoS$_2$ are known to trap carriers for few hundreds of picoseconds. Thus, in the undoped sample the observed decay of $\Delta\alpha$ at low fluence regime could be explained just by the decay of carriers to the defect states, see Fig. 1(b). At higher fluences it is expected that the number of defect states in MoS$_2$ get completely filled. Such filling should in fact increase the lifetime of carriers, which is opposite to the experimental observation. On the other hand, a scattering process like the Auger mechanism is expected to play a stronger role at higher excitation fluences. We show that the linear dependence of $1/\tau$ on excitation density observed at higher fluences (> 3 mJcm^{-2}) can be explained by the stronger contribution of defect assisted Auger relaxation in ML MoS$_2$.

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COHERENT RESPONSE OF THE ELECTRONIC SYSTEM DRIVEN BY NON-INTERFERING LASER PULSES

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The strength of light-matter interaction in condensed matter is fundamentally linked to the orientation and oscillation strength of the materials' optical transition dipoles. Structurally anisotropic materials, e.g. elongated molecules, exhibit optical transition dipoles with fixed orientations that govern the angular-dependent light-matter interaction. Contrary, free electron like metals should exhibit isotropic light-matter interaction with the light fields dictating the orientation of the optical transition dipoles. Here, we demonstrate that an anisotropic direction of the optical transition dipoles even exists in highly free electron like noble metal surfaces. Our time- and phase-resolved photoemission experiment reveals coherent interference effects on (110)-oriented silver surface after optical excitation with two non-interfering cross-polarized pulses. We explain this coherent material response within the density matrix formalism by an intrinsic coupling of the non-interfering light fields mediated by optical transition dipoles with fixed orientations in silver.

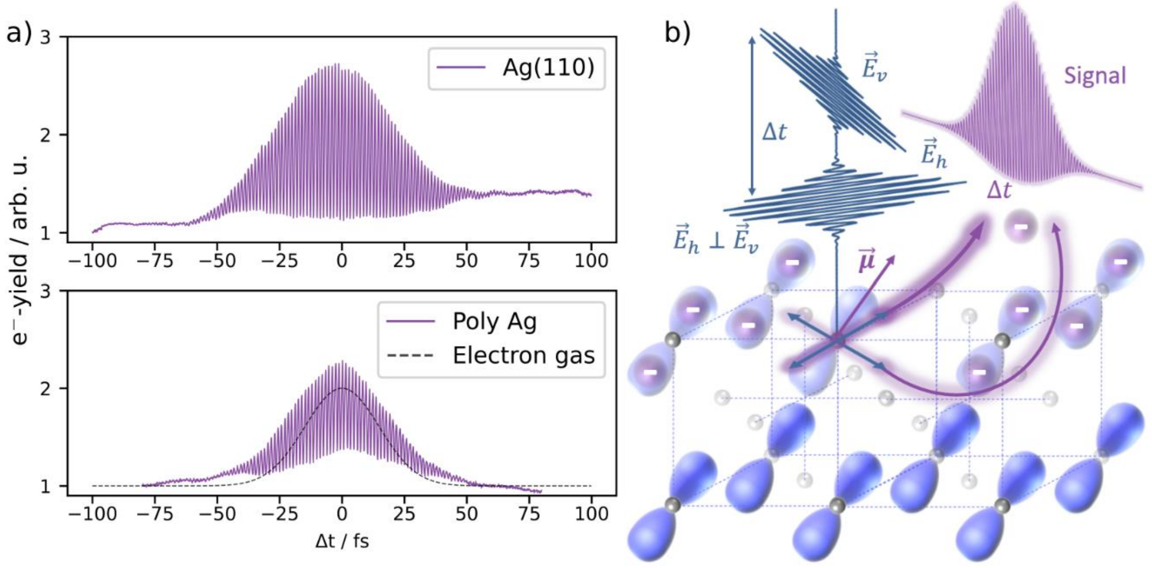

Figure 1 – Cross-polarized two-photon photoemission traces of a Ag(110) and a polycrystalline silver surface a) Energy- and momentum-integrated cross-correlation traces on Ag(110) (top) and polycrystalline silver (bottom) showing interference patterns despite the non-interfering pump-probe excitation scheme. The black dashed curve represents the expected behavior in the case of a free electron gas. b) Schematic of the coupling between the perpendicular electric fields and the optical transition dipole $\vec{\mu}$, which leads to coherent photoexcitation for the field components overlapping with the dipole.

COHERENT TIME-RESOLVED ABOVE-THRESHOLD PLASMOEMISSION FROM THE AU(111) SHOCKLEY SURFACE STATE

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Strong nonperturbative interactions of an intense driving light field with the electronic band structure in a solid can result in exotic material properties that do not exist under equilibrium conditions. For suitable driving conditions, Floquet theory predicts that the originally unperturbed electronic structure is modified by the formation of light-dressed electron states in strong fields. Observing such dressing requires electronic state resolution as well as precise control over the intense periodic driving field to overcome intrinsic dissipation and decoherence in the solid.

Here, we explore nano-focusing of femtosecond surface plasmon polariton (SPP) pulses on flat surfaces as a possible route towards strong-field control over electronic states within a solid using time- and angle-resolved photoemission spectromicroscopy. We observe above-threshold electron emission from the Au(111) Shockely surface state by the absorption of up to seven SPP quanta. Two-dimensional time-resolved photoelectron spectroscopy using a birefringent delay line provides us with direct access to the coherent and incoherent dynamics of the electron emission process with attosecond precision. The presented results clearly indicate the coherent nature of the interaction of the intense SPP nano-focus with the band structure of the material.

DIFFUSE SCATTERING AND NONTHERMAL PHONON POPULATIONS IN ULTRAFAST LEED

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Ultrafast diffraction and spectroscopy have significantly advanced our understanding of nonequilibrium dynamics in condensed matter, tracing excitation and relaxation pathways in and between electronic and lattice degrees of freedom. Recently, ultrafast electron diffuse scattering (UEDS) was introduced to characterize transient phonon populations from momentum-resolved maps of the diffuse background arising from inelastic scattering [1,2]. To date, these experiments were conducted in transmission through thin films, and at electron energies of tens of keV, primarily exhibiting bulk-sensitivity. Here, we transfer this scheme to surface-sensitive scattering at low electron energies and in a reflection geometry, which, in contrast to former experiments, is sensitive to phonon modes with out-of-plane polarization. The experiments are conducted with our ultrafast low-energy electron diffraction (ULEED) setup (see Fig. 1a), in which collimated electron pulses are generated at a nanoscopic needle emitter triggered by 400 nm laser pulses. This setup enables the investigation of ultrafast structural dynamics at surfaces in a laser-pump/electron-probe scheme with a temporal resolution down to 1.3 ps [3] at electron energies of 20-200 eV, combined with a high momentum resolution of $\Delta k_\mathrm{s} = 0.03\ \text{Å}^{-1}$.

In this contribution, by analyzing the delay-dependent diffuse scattering signal in ULEED, we track non-equilibrium phonon populations in the commensurate charge density wave phase of titanium diselenide, a prototypical 2D transition metal dichalcogenide. We observe transient momentum-dependent changes in the inelastic background following optical excitation, which we link to a change in population of acoustic phonons with out-of-plane polarizations. These are generated from high-energy phonons via phonon-phonon scattering in the later stages towards thermal equilibrium. A momentum-resolved map of background signal rise time (see Fig. 1b) evidences a decrease in rise time between the zone center and the boundary, which we attribute to the higher number of scattering pathways leading to the latter. Measurements taken at different pump fluences reveal accelerated dynamics for higher fluences across the whole Brillouin zone (see Fig. 1c). We explain this with the larger phonon population leading to increased scattering probabilities.

Overall, the evaluation of the diffuse scattering signal in ULEED represents an ideal complement to established UEDS methods due to the surface and out-of-plane sensitivity. It has the potential to provide fundamental insights to phonon dynamics at surfaces as well as relaxation pathways in 2D materials or heterostructures.

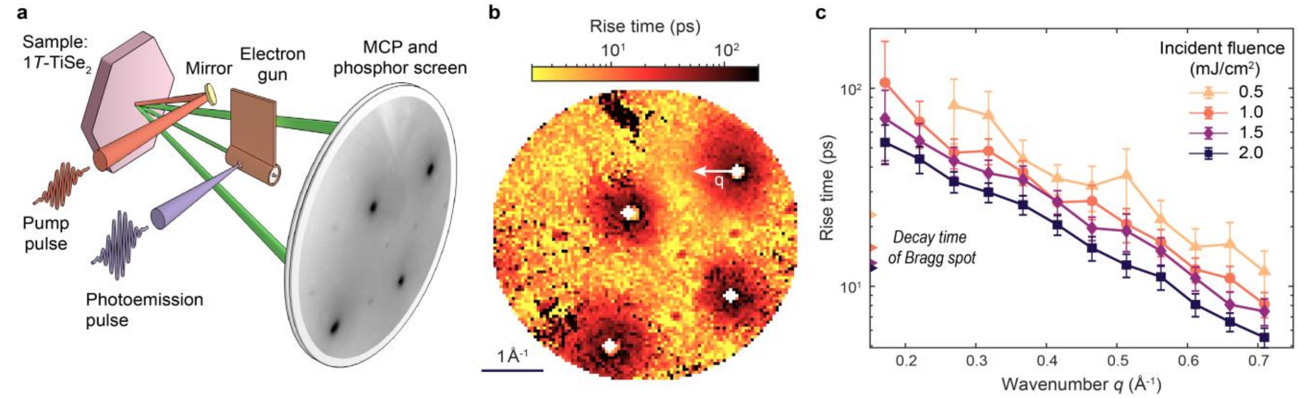

Fig. 1. a, Sketch of the ULEED setup. b, Momentum-resolved map of phonon rise times. c, Radial profile of phonon rise times for four different pump fluences.

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ELECTRON DYNAMICS AT A CONFINED COPPER - CUPROUS OXIDE INTERFACE

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Cuprous oxide (Cu_2O) is a promising photocatalytic material [1] but is unstable in contact with water. To reduce corrosion, we have prepared thin $Cu_{2-x}O$ films with a protective *h*-BN capping layer on Cu(111) by O_2 intercalation at partial pressures of O_2 from 0.01 to 1 mbar and temperatures up to 200 °C, a procedure similar to [2]. The atomic structure was unraveled with a combination of surface science techniques at the Swiss Light Source [3,4]. The *h*-BN layer remains intact and the $Cu_{2-x}O$ oxide film has a thickness of a few atomic layers (3-4 Å) with a structure displaying motifs similar to the Cu_2O(111) surface [5,6], comprising a chemisorbed O layer and a $Cu_{2-x}O$-like trilayer positioned in between the h-BN layer and substrate. A well-ordered *h*-BN/$Cu_{2-x}O$/Cu(111) sample was prepared for characterizing the electronic structure and electron dynamics with ultraviolet and pump-probe photoemission spectroscopy, and compared to *h*-BN/Cu(111) and Cu(111) reference samples. Angle-resolved photoelectron spectroscopy (ARPES) after oxidation shows the sharp σ and π bands of an intact *h*-BN monolayer and another non-dispersing state 1.2 eV below the Fermi level, attributed to the valence band of Cu_2O that is shifted downwards due to the contact to a metallic copper substrate. Time-resolved two-photon photoemission (tr-2PPE) spectra acquired with 3 eV pump and 6 eV probe photon energy show a short-lived state at 1.0 eV and a long-lived state at 0.1 eV above the Fermi level (Fig. 1). The higher energy state has a strong parabolic dispersion reminiscent of the conduction band of bulk Cu_2O, whereas the low energy state has no dispersion, pointing towards a localized defect state in the $Cu_{2-x}O$ layer. Such defect states were also observed on the reconstructed bulk Cu_2O(111) surface [7] and were attributed to charged oxygen vacancy states with very high capture coefficients for trapping conduction electrons [8]. We conclude that the electron dynamics in this confined layer are remarkably close to those in single-crystalline Cu_2O [6], and the visible conduction band shows the potential of this approach to create a stable photocathode.

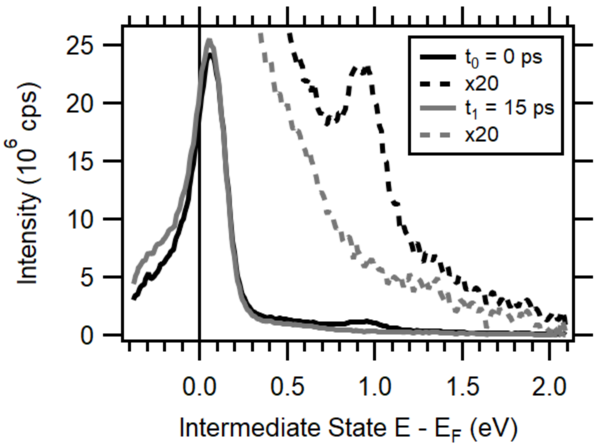

Figure 1 – tr-2PPE spectra at two delays after subtraction of the delay-independent background

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EXCITED STATE DYNAMICS IN PANCHROMATIC PORPHYRIN-NANOGRAPHENE CONJUGATES

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Graphene is a well-known material with an outstanding thermal and electrical conductivity, elasticity, hardness and mechanical strength [1]. It is currently being used as a transparent conductor, for organic light-emitting diodes, photodetectors or saturable absorbers for ultrafast lasers [2]. Nevertheless, the absence of an optical gap in the energy band structure jeopardizes many applications in optoelectronics.

Nanographenes (NGs) offer the possibility of introducing a bandgap, while preserving outstanding electric and mechanical properties. In this work, we use femtosecond optical transient absorption spectroscopy to systematically study, for the first time, photoexcited carrier relaxation in *β-,meso-,β-triplyfused porphyrin-nanographene* conjugates **1** and **2** (Fig.1) [3]. These compounds are panchromatic NGs with an onset of absorption matching the solar spectrum, making these compouns suitable candidate for high efficiency solar cells. Conjugate **2** performs a subpicosecond thermalisation process, followed by a band to band relaxation of the order of 100s ps. However, for conjugate **1** the last process is hastened, leading to a much faster photobleaching decay in the picosecond order.

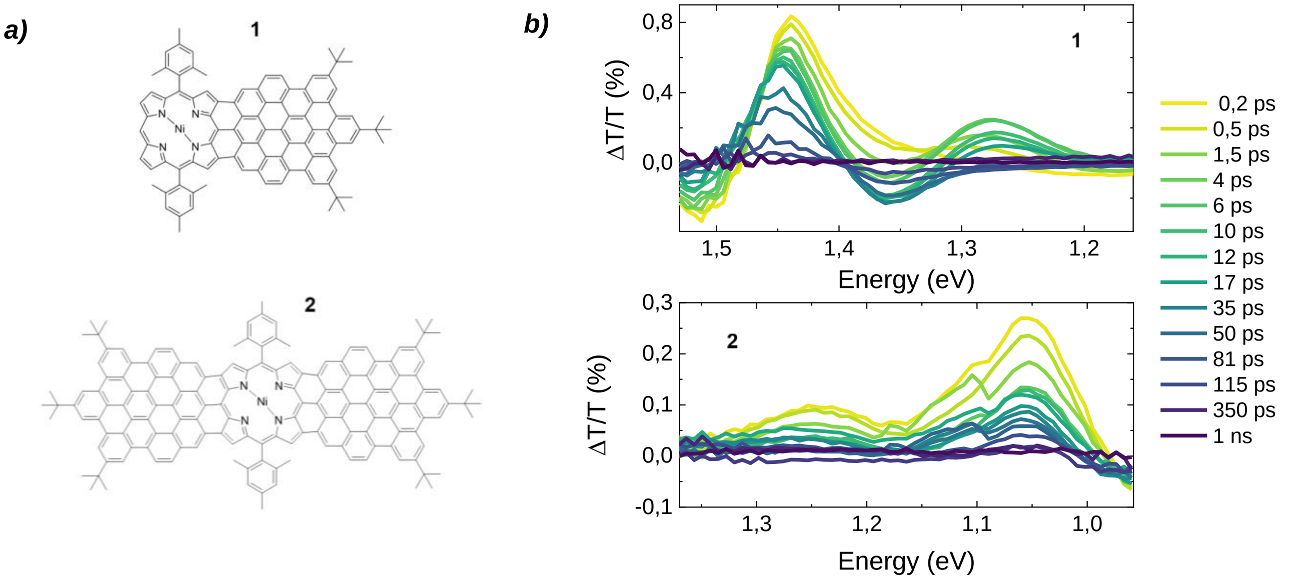

*Figure 1 – **a)** Structures of triply fused porphyrin-nanographene conjugates **1** and **2** [3], and **b)** differential transmission signal obtained for conjugates **1** and **2** during 1 ns after pumping with a 778 nm laser.*

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EXPLORING ELECTRON-PHONON INTERACTIONS USING FREQUENCY-DOMAIN ARPES

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Electron-phonon interaction (EPI) is one of the most fundamental quasiparticle interactions in solids governing for instance transport and thermodynamic properties of metals, but being also responsible for unconventional phenomena such as low-temperature superconductivity or the formation of charge-ordered phases.

Recent time- and angle-resolved photoemission spectroscopy (TRARPES) studies on coherent phonon-driven electronic processes illustrated that an advanced frequency domain analysis of such data can provide an intuitive and direct electronic band- and phonon mode-resolved view onto EPIs [1 - 3]. Even more, a theory work on this topic envisioned that such type of experimental data could in the future enable the quantitative evaluation of electron-phonon matrix elements directly from the experimental data without any additional input from theory [4].

In my presentation, I will present and discuss TRARPES data on the excitation of coherent phonons in the Weyl-semimetal *Td*-WTe_2. A Fourier-transform of the three-dimensional data set allows revealing electronic band-resolved information on EPI in this material. The phonon frequency-resolved data carry high-resolution information on band dispersions and depict the different strengths and types of couplings of the individual modes to the electronic bands. I particularly will discuss results on the transient modulation of a Dresselhaus-type spin splitting of electronic bands driven by the selective coupling of an interlayer shear mode of the layered compound. The latter results reveal real-time insights into electron-phonon coupled processes that are of vital importance for a light-driven topological phase transition in *Td*-WTe_2.

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GALLIUM-DEPENDENT PASSIVATION EFFICIENCY AT THE CdS/CIGS INTERFACE

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The bandgap of $CuIn_{1-x}Ga_xSe_2$ (CIGS) chalcopyrite semiconductors can be tuned between ~1.0 eV and ~1.7 eV for Ga contents ranging between x=0 and x=1. While an optimum bandgap of 1.34 eV is desirable for achieving maximum solar energy conversion in solar cells, state-of-the-art CIGS-based devices experience a drop in efficiency for Ga contents x>0.3 (i.e. for bandgaps >1.2 eV), an aspect that is limiting the full potential of these devices. The mechanism underlying the limited performance as a function of CIGS composition has remained elusive: both surface and bulk recombination effects have been proposed. Here, we use optical pump-THz probe spectroscopy (OPTP) to disentangle between surface and bulk effects in CIGS absorbers as a function of Ga content. The goal is achieved by comparing photo-generated charge carrier dynamics in air/CIGS and surface-passivated ZnO/CdS/CIGS samples. While surface passivation prevents surface recombination of charge carriers for low Ga content (x < 0.3; up to 1.2 eV bandgap), surface recombination is unaffected in higher bandgap alloys. Altogether, our results correlate well with the observed Ga-dependent efficiency trend in device performance and demonstrate that the drop of efficiency in cells made with larger bandgaps can be attributed, at least partially, to surface - rather than bulk - recombination at the CdS/CIGS interface. [1]

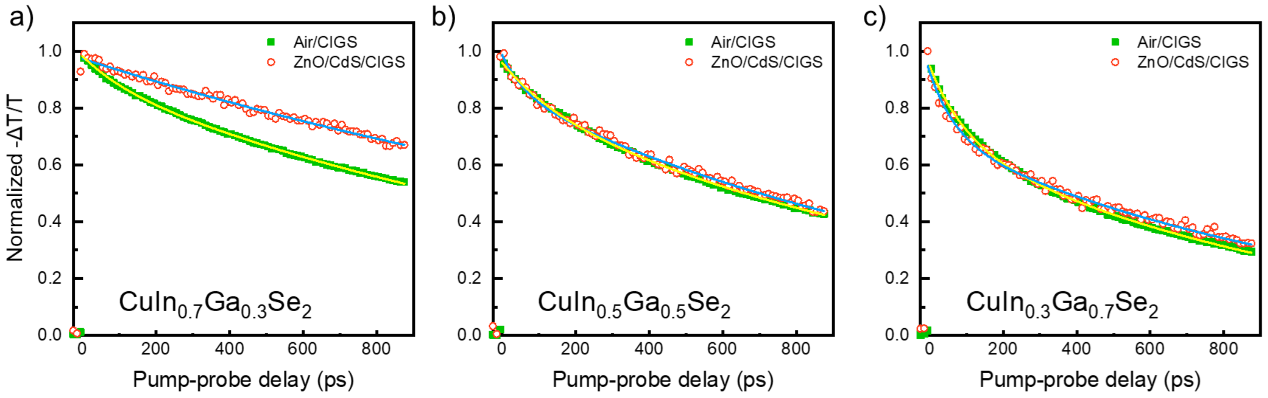

Figure 1 - Normalized optical Pump-THz Probe dynamics on CIGS samples with Ga-contents of x=0.3 (a), x=0.5 (b) and x=0.7 (c). Green squares represent the decay for the air/CIGS samples, while red open circles depict the transient signal for the ZnO/CdS/CIGS samples. Solid lines indicate the best bi-exponential fits to the data (single exponential for the low Ga ZnO/CdS/$CuIn_{0.7}Ga_{0.3}Se_2$ sample). In all cases, the photogenerated carrier density per pulse equals $1.7 \cdot 10^{18} cm^{-3}$.

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IMAGING OF VALLEY POPULATION IN MONOLAYER WS_2

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Two-dimensional transition metal dichalcogenides (TMDCs) host an exceptional variety of excitons. Upon optical excitation, bright excitons consisting of Coulomb-bound electrons and holes of same spin orientation are forming within the same valley. Due to band ordering and different binding energies other energetically favorable excitonic states exist with electron and hole located at different valleys. The formation of such excitons with non-zero center of mass momentum requires a momentum transfer and/or spin-flip. They are referred to as momentum- or spin-dark excitons as they are inaccessible by optical interband transitions and therefore difficult to study in all-optical experiments.

In our experiment, we combine a tunable pump and a high-harmonic probe laser source with a momentum microscope to map electrons after optical excitation from monolayer WS_2. Thereby, we get access to bright and dark states on an ultrafast timescale [1]. The high temporal resolution of our setup allows us to trace the early-stage excitons dynamics and observe the formation process of momentum-forbidden dark KΣ-excitons, which is mediated by strong electron-phonon coupling. To get access to all possible scattering pathways after excitation, we recently incorporated a new pumping scheme, that allows to excite the sample under an incident angle close to 0° with circularly polarized light. This results in an excitation, which is located purely within the K valley for one helicity and in the K’ valley for the other helicity (Fig. 1). In the case of an excitation at K we find that electrons scatter by spin-conserving processes towards K’ and Σ within a few tens of femtoseconds, while the formation of spin-forbidden excitons in the K valley or electron scattering towards Σ’ is significantly slower (50 – 100 fs).

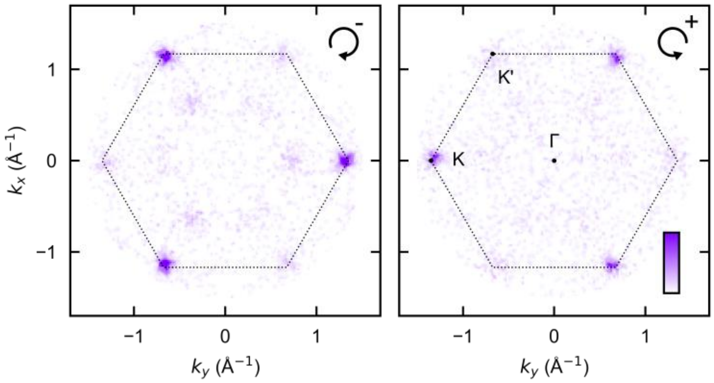

Fig 1: Momentum maps of the conduction band of monolayer WS_2 around temporal overlap. The system is excited with circularly polarized pump photons of opposite helicity as indicated in the upper right corner. Photon energy in both cases is resonant with the A 1s exciton (2.05 eV).

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INFLUENCE OF THE STATIC DIELECTRIC PERMITIVITY ON ULTRAFAST QUASIPARTICLE DYNAMICS IN WS_2C MONOLAYERS

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Unlike their bulk counterparts, transition metal dichalcogenite monolayers are ideal for the study of exciton physics due to their direct band gap. The Coulomb-bound electron-hole pairs, dominate the optical properties of TMDC monolayers and are stable at room temperature due to their high binding energy on the order of hundreds of meV [1]. Investigation of the influence of substrates on the exciton dynamics of TMDCs monolayers is crucial for their application in the field of electronics such as integrated circuits. Theoretically, any dielectric environment, such as of a substrate, should influence the Coulomb interaction between electrons and holes through screening and, thus, affect the excitonic properties of TMDCs monolayers.

This work is an extension of our recent fluence- and photon energy dependent studies of quasiparticle dynamics in WS_2 monolayers on fused silica (FS) and Si-SiO_2 substrates [2,3]. These showed that excitonic screening solely reduces the binding energy of the excitons, leading to a transient blue-shift of the exciton resonance. On the contrary, quasi free carrier (QFC) screening effectively causes a red shift, as the carrier-induced screening leads to a larger band gap renormalization than binding energy reduction [3]. Due to the very similar dielectric permittivities of the two substrates in these works, the quasiparticle dynamics were not affected by the change of template [2]. In this work, we investigate the fluence-dependent, time-resolved transmission contrast of WS_2 monolayers on sapphire and FS substrates and compare the exciton steady-state and dynamic response, as sapphire's dielectric permittivity is thrice the one of FS. Using the previously introduced formalism [2], we observed a blue shift of the exciton resonance followed by a redshift at later time delays in case of resonant excitation while an additional, initial red-shift is determined in case of QFC excitation. We characterize the underlying exciton and carrier population dynamics by the phenomenological model reported in [3] and compare the extracted parameters for the two different substrates. The effective band gap renormalization parameter is quite comparable for FS and Sapphire and they are in the order of 10^{-12} meV/cm^2 while the dynamic screening parameter due to exciton screenings leading to binding energy reduction is approximately 4 times higher in sapphire than for FS. The time constants for linear recombination channel of excitons and transfer of thermal energy to the sapphire substrate are in the order of tenths of ps while Auger recombination rate is around 0.07 cm^2/s. Similarly, the broadening parameters due to the scattering between quasiparticles are quite comparable in both substrates. Remarkably, neither scattering rates, relaxation time constants, nor band gap renormalization are influenced by the dielectric permittivity. Only the steady state exciton resonance and the photoinduced exciton binding energy reduction are modified by the change of the substrate's dielectric properties. This is likely related to the different Bohr radii and the resulting modified polarizability of the exciton population. These findings highlight and quantify the fragile interplay of the many elementary processes at play upon photoexcitation of TMDC monolayers that determine transient energy level alignment and subsequent non-equilibrium dynamics.

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PSEUDOHETERODYNE NEAR-FIELD IMAGING AT KHZ REPETITION RATES

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Scattering-type scanning near-field optical microscopy (SNOM) enables the measurement of the optical response of 2D materials and nano-structures beyond the diffraction limit. While it has been shown that time-resolved SNOM gives direct access to charge carrier dynamics on these functional surfaces [1], the use of pulsed laser sources in combination with a lock-in based signal detection forces compromises on AFM parameters and laser amplifier. The high-order demodulation technique used to extract the near-field from the total backscattered light requires the sampling rate to be significantly higher than the AFM tapping frequency of 70 – 350 kHz, thus requiring a laser repetition rate in the MHz regime. However, kHz class laser sources with their associated high peak powers are desirable in order to access the broad experimental toolkit based on nonlinear optics such as the use of optical parametric amplifiers, extending the available wavelengths into the visible and even near UV. Here we present quadrature-assisted discrete (quad) demodulation, circumventing these constraints by measuring the optical signals as well as the modulation phases for individual pulses [2]. This approach is based on the phase-domain sampling technique introduced by Wang *et al.* [3] and is fully compatible with established methods for background and noise suppression, e.g. pseudo-heterodyne (pshet) and balanced detection. The measurements, acquired with a fs laser source with a repetition rate of 200 kHz, provide a similar contrast and signal to noise ratio as results obtained by standard lock-in based methods. This is shown through a comparison of retraction curves and images on a SiO_2/Si test grating. SNOM measurements with a pulsed laser in the visible regime are illustrated by images of WS_2 layers on a SiO_2/Si substrate at different wavelengths, on and off resonance of the A-exciton in WS_2 at 629 nm. Establishing pshet SNOM at kHz repetition rates paves the way towards time-resolved SNOM and nano-spectroscopy beyond IR frequencies.

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SCANNING TUNNELING MICROSCOPE INDUCED-LUMINESCENCE OF AN ATOMICALLY-THIN SEMICONDUCTOR.

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Atomically thin semiconductors made from transition metal dichalcogenides (TMDs) are appealing systems for the investigation of efficient light-matter interactions. Their reduced dimensionality and reduced dielectric screening results in tightly bound excitons that dominate their optical response, even at room temperature [1]. However, the nanometer (nm) spatial extension of excitons makes them sensitive to the presence of atomic-scale inhomogeneities such as defects, adsorbates, dielectric disorder and local strain [2-3]. The nm-size character of these inhomogeneities lies at length scales below the spatial resolution accessible with standard optical spectroscopies. Furthermore, the proximity between the TMD with other 2D-materials can foster ultrafast energy and charge transfer phenomena happening in sub-picosecond (ps) timescales [4,5]. This underlines the necessity of experimental techniques able to address excitons with sub-nm spatial and sub-ps temporal resolution. Here, we present an approach using a scanning tunneling microscope (STM) to induce the luminescence of an $MoSe_2$/few layer graphene (FLG) heterostructure supported by a Au(111) substrate. This allows us to correlate the atomic-scale landscape with the locally induced optical response of the heterostructure. We demonstrated for the first time STM-induced luminescence (STML) from neutral, charged and localized excitons in TMDs at low temperatures (figure 1). We report sizeable variations in the emission of the heterostructure between different nm-sized areas. Hyperspectral STML mapping and time-resolved experiments are required for a complete understanding of the optical properties of the heterostructure. However, this study paves the way for novel investigations regarding the local properties of van der Waals heterostructures and highly localized excitons. We conclude by presenting an experimental setup capable of addressing exciton dynamics with femtosecond-temporal and nm-spatial resolution. This technique, known as THz-STM, relies on the free space coupling of broadband single cycle THz-pulses to an STM junction [6,7]. This setup has recently been developed by the research group of Dr. Melanie Müller at the department of physical chemistry headed by Prof. Martin Wolf at the Fritz Haber Institute, Berlin.

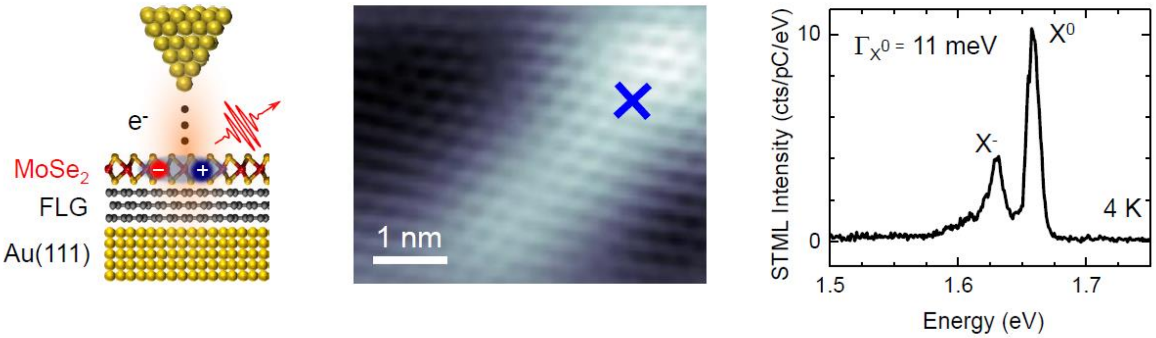

Figure 1 – Left: Artistic view of an STML experiment. An exciton is created by the tunneling electrons. The curly arrow represents a photon emitted after radiative recombination. Middle: Atomically-resolved STM image of the $MoSe_2$/FLG surface. Right: STML-spectrum acquired at the marked position in the middle image. The spectra is composed of two peaks corresponding to the neutral (X^0) and charged exciton (X^-).

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SIGNATURE OF MAGNON POLARONS IN ELECTRON RELAXATION ON TERBIUM REVEALED BY COMPARISON WITH GADOLINIUM

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Magnons and phonons are elementary excitations of spin and lattice subsystems that can form a hybrid quasiparticle in the presence of strong magnetoelastic coupling. Here we use spin-, angle-, and energy-resolved photoemission spectroscopy to show that magnon-phonon hybridization plays an important role for the relaxation of the electron surface-state in Tb, opening both majority and minority spin channels in electron-phonon scattering. This is attributed to the strong 4f spin-orbit coupling, as evidenced by a comparison of the lifetime broadening in the occupied surface states of Gd and Tb. Both ferromagnetic metals have a comparable valence electronic structure, but the magnetocrystalline anisotropy is much stronger in Tb as compared to Gd. Consequently, in Gd electron-phonon and electron-magnon scattering lead to spin-dependent photohole relaxation-rates. Unlike in Gd, the lifetime broadening of the occupied surface state in Tb is only weakly spin dependent and the mass enhancement parameter λ is twice the spin-averaged value of Gd. This difference in phase space is explained by the intimate coupling of phonons and magnons to form magnon polarons, which opens both minority and majority spin bands as decay channels [1].

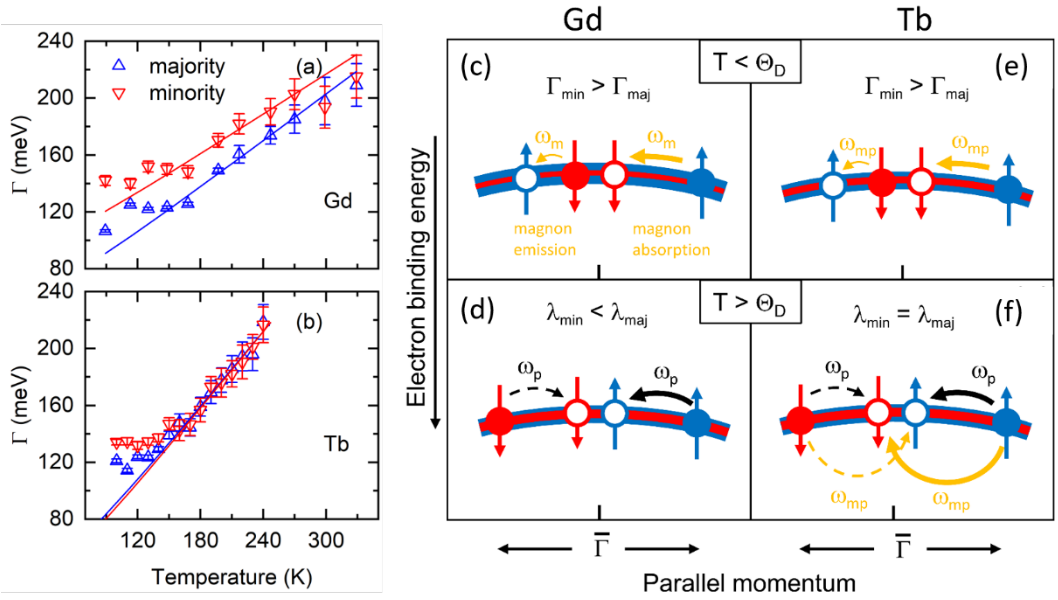

Figure 1 – Extracted linewidths $\boldsymbol{\Gamma}$ of the surface state on (a) ***Gd*** *and (b)* ***Tb*** *for both the spin-majority (blue, up-triangle) and spin-minority (red, down-triangle) components as a function of temperature. Sketch of intra-band scattering processes filling the photohole (open circle) in the occupied surface-state band of Gd (left column) and Tb (right column). Blue and red colors denote the spin majority and spin minority components, respectively. The degree of spin mixing of the surface-state band is indicated by the line thickness.* ***Gd*** *(c) In the temperature range $\boldsymbol{T} < \boldsymbol{\Theta_D}$ between 90 and 160 K electron-magnon scattering ($\boldsymbol{\omega_m}$) dominates. This leads in combination with the spin-dependent density of states to larger lifetime broadening in the spin minority channel $\boldsymbol{\Gamma_{min}} > \boldsymbol{\Gamma_{maj}}$. (d) For $\boldsymbol{T} > \boldsymbol{\Theta_D}$ spin conserving electron-phonon scattering dominates and leads to a spin-dependent mass-enhancement parameter $\boldsymbol{\lambda_{min}^{Gd}} < \boldsymbol{\lambda_{maj}^{Gd}}$.* ***Tb*** *(e) For $\boldsymbol{T} < \boldsymbol{\Theta_D}$ electron-(magnon-polaron) scattering ($\boldsymbol{\omega_{mp}}$) dominates and $\boldsymbol{\Gamma_{min}} > \boldsymbol{\Gamma_{maj}}$. (f) For $\boldsymbol{T} > \boldsymbol{\Theta_D}$ phonon scattering and magnon-polaron formation double the phase space for electron scattering and $\boldsymbol{\lambda_{min}^{Tb}} = \boldsymbol{\lambda_{maj}^{Tb}} = (\boldsymbol{\lambda_{min}^{Gd}} + \boldsymbol{\lambda_{maj}^{Gd}})$.*

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SPATIOTEMPORAL IMAGING OF BRIGHT AND DARK EXCITONIC QUASIPARTICLES IN TWISTED TMD HETEROSTRUCTURES

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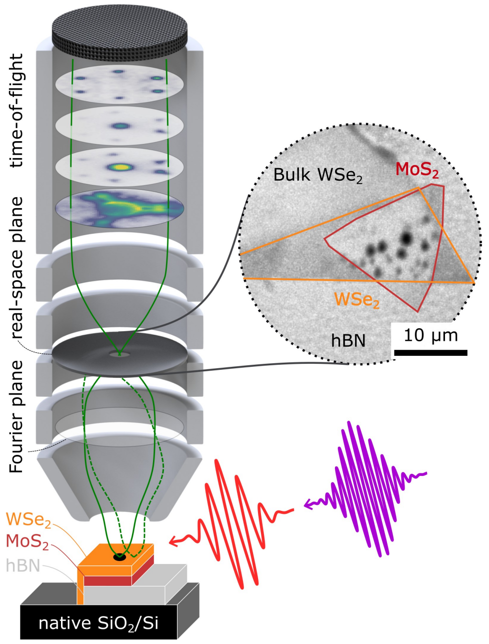
The weak Coulomb screening of charge carriers in van der Waals materials leads to exciting new possibilities to control material properties. For example, the optoelectronic response of Transition Metal Dichalcogenide (TMD) monolayers is defined by bright and dark exciton quasiparticles, which exhibit binding energies of several 100 meV. Dark excitons are momentum-indirect and thus not directly accessible with all-optical probe techniques. Despite this, understanding their formation and relaxation properties is crucial for understanding the material's overall light-matter interaction.

Furthermore, when creating a heterostructure out of two TMD semiconductor monolayers with a type II band alignment, interlayer excitons are formed [1]. The hole remains in the valence band maximum of one layer and the electron in the conduction band minimum of the other, resulting in a separation of charges – an atomically thin p-n junction.

In this complex interplay between intra- and interlayer excitons, the spatial and lateral dynamics on the fundamental nanometer length scale remain mainly unrevealed. How do specific exciton quasiparticles form, relax and diffuse in the presence of a heterojunction, stress-fields, inhomogeneities or edge states? Do dark excitons, hidden from direct probes by light, behave differently than bright quasiparticles? For this a technique with momentum, energy and high spatial resolution is needed.

Here, we tackle this challenge using time-resolved momentum microscopy [2] and femtosecond photoelectron dark-field microscopy to study the ultrafast formation dynamics of different excitonic species in twisted WSe_2/MoS_2 heterostructures in momentum and real space.



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Spectrally resolved spin dynamics of 3d-transition metals in EUV T-MOKE

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The use of light pulses in the extreme ultraviolet (EUV) and soft x-ray regions to probe ultrafast magnetization dynamics of a given material has found increasing application in the field of magneto-optical spectroscopy. It allows to get new insights into the light-matter interaction processes, because the technique does not only provide element-specificity but also the potential to investigate spectrally-resolved non-equilibrium spin dynamics in the band structure of the material. The latter has led to promising discoveries in the field of ultrafast magnetism, such as the optically-induced spin transfer process (OISTR) [1-3], and recently, to a number of publications on distinct spectral responses in transition metals [4-6].

Here, we investigate the spin dynamics of 3d-transition metals Co, Fe and Ni by use of EUV T-MOKE. The high-harmonic beamline is based on a fiber laser amplifier system with a repetition rate between 100 and 300 kHz, covering a photon energy range of 30-72 eV [7]. Being resonant to the M-absorption edges of the given elements, our high-harmonic spectrum allows us to resolve distinct spin dynamics at specific energies around the Fermi-level, as shown for the example for Ni in Fig 1. The pump energy can be tuned from 1.2 eV to the second harmonic of 2.4 eV, which allows the investigation of spin dynamics as a function of the pump energy. We discuss different processes leading to energy-dependent changes in the magnetic asymmetry at early times after the optical excitation, e.g. optically-induced charge/spin transfer and band renormalization in 3d-transition metals Co, Fe and Ni.

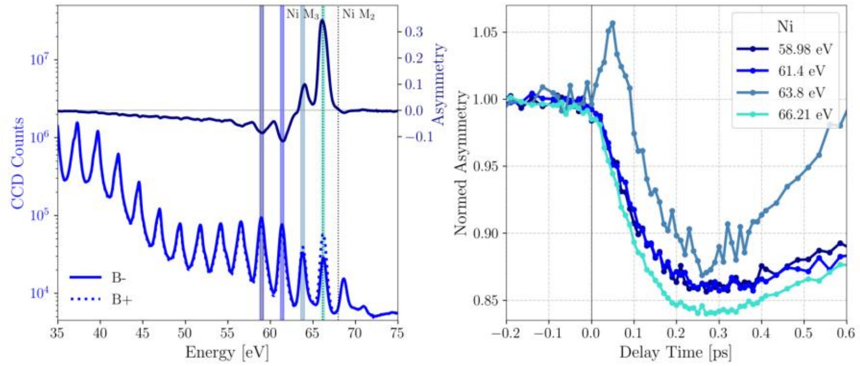

Figure 1 – Magnetic asymmetry together with typical HHG spectra for both magnetization directions (left). Spectral dynamics of the asymmetry for different energies around the $M_{2,3}$-edges of Ni after pumping with 1.2 eV (right).

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Spin-transport-driven ultrafast magnetization dynamics in a ferrimagnetic Gd/Fe bilayer

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Laser-induced spin transport has been proven to be a key ingredient in ultrafast spin dynamics, such as femtosecond demagnetization and all-optical switching (AOS). However, it is still debated to what extent ultrafast demagnetization generates spin currents or vice versa. We use time- and spin-resolved photoemission spectroscopy of the gadolinium surface state to study spin dynamics in a ferrimagnetic Gd/Fe bilayer. This prototype system for AOS was epitaxially grown on W(110). The magnetization M of the Gd and Fe film are in-plane and antiparallel aligned. First, we studied the temperature dependence of M_{Gd} via the spin polarization of the Gd surface state. The compensation temperature T_{CP}, where $M_{Gd} = -M_{Fe}$, depends on layer thicknesses. For a Gd(5 nm)/Fe(3 nm) bilayer, we observe reversal of the spin polarization of the Gd surface state at $T_{cp} \sim 230$ K. For Gd(2 nm)/Fe(3 nm) the compensation temperature is about 130 K. At higher temperature M_{Fe} aligns always parallel to the external field. For this case we have studied the ultrafast spin dynamics in a pump-probe experiment. Our dynamic findings suggest that spin transport between the antiferromagnetically coupled gadolinium and iron layers leads to an ultrafast drop of the spin polarization and an increased exchange splitting at the Gd surface. In the Gd/Fe bilayer, we find that after optical excitation the Gd surface state shows an ultrafast decrease of the spin polarization by 20 % within the first ~100 fs and a subsequent slower decrease by about 5% on the picosecond timescale. The increase of the exchange splitting acts like a bottleneck against the overall demagnetization of the Gd layer. In contrast, the pure Gd/W(110) film shows a constant spin polarization and a reduced exchange splitting of the surface state upon optical excitation [1]. This helps to exclude significant contributions of local spin-flips to spin-polarization dynamics. Therefore, any variation of spin polarization in the Gd/Fe bilayer must be attributed to spin transport. This is corroborated by the transient electron temperature that decays within 1 ps to the initial temperature before optically excited electrons can heat lattice. In pure Gd/W(110) ultrafast spin transport is absent and an elevated electron temperature of about 380 K is reached after a pump-probe delay of 1.5 ps via electron-phonon coupling. Our results provide clear evidence that magnetization dynamics in the Gd/Fe bilayer can be driven by spin transport. We see distinct signatures in the spin-dependent electronic structure that allow us to gain microscopic insights into ultrafast spin dynamics. These findings are of general importance for the understanding of magnetization switching in ferrimagnetic Gd-Fe compounds.

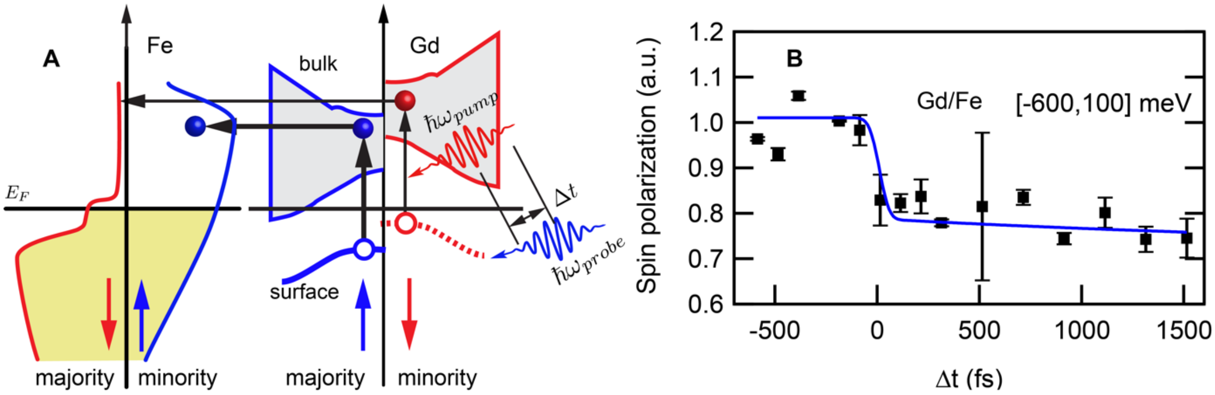

*Figure 1. (**A**) Schematic spin-resolved density of states of Fe and Gd. Due to the opposite in-plane magnetization of the Fe and Gd layers majority spin electrons in Gd (blue) become minority spin electrons in Fe and vice versa for spin minority electrons (red). The IR pump pulse excites electrons from the Gd surface state into Gd bulk states. While spin-majority electrons are transported into the Fe layer as indicated by the thick arrow, the transfer of spin-minority electrons is minor (thin arrow). The spin-dependent density of states of Fe acts as a spin filter. This leads to an ultrafast drop of the Gd spin polarization probed at the Gd surface. (**B**) Spin polarization of the Gd/Fe bilayer integrated from $E - E_F = -600$ to 100 meV. It shows an instantaneous decrease of about 20 % at 0 fs after photoexcitation, followed by a slower ps decrease.*

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STUDYING LONG-RANGE ORDER STRUCTURAL DYNAMICS OF ADSORBATES USING TIME-RESOLVED LOW ENERGY PHOTOELECTRON DIFFRACTION

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Time-resolved electron diffraction has evolved into a key instrument for the study of ultrafast structural dynamics in solids and at their surfaces. A high surface sensitivity, for instance required when probing ultrafast adsorbate-substrate interactions, can be achieved using trRHEED [1] and trLEED [2]. One of the main limitations of these types of experiments arises from the repulsive Coulomb interaction among the probe electrons resulting in a temporal spreading of the electron pulses and a deterioration of the time resolution.

Photoelectrons that are emitted from a substrate through an adsorbed layer of molecules can undergo diffraction and thus carry structural information of the overlayer. In low energy ARPES experiments one particularly addresses the long-range structural order of the adsorbates and, therefore, gains LEED-equivalent information [3]. This sensitivity may potentially be used in a trARPES experiment to probe the transient built-up and decay of disorder or ultrafast structural phase transitions in adsorbate overlayers in response to a pulsed photoexcitation. The time resolution of such an experiment is ultimately limited by the pulse width of pump and probe laser pulses, only.

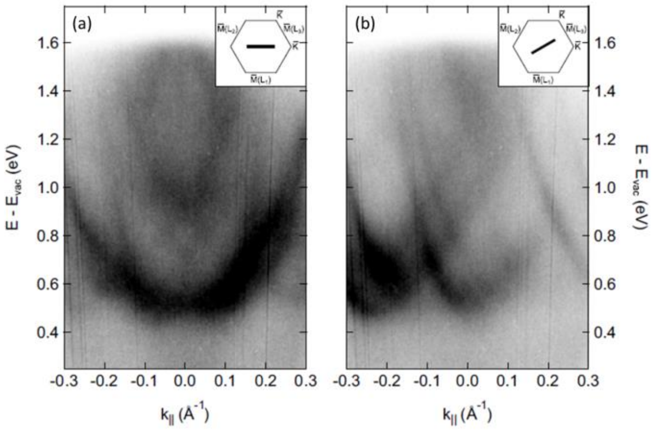

Figure 1 – Photoelectron diffraction pattern from an incommensurate adsorbate overlayer in a low-energy ARPES spectrum of 1 ML SnPc on Ag(111) measured (a) along the ΓK direction and (b) along the ΓM direction. Figure from Ref. [4]

In this poster we will present results of low-energy ARPES studies of SnPc adsorbed on Ag(111) and single crystalline graphite (SCG) which have been performed using 90 fs NUV pulses. We observe distinct spectral patterns in the ARPES spectra, which can be associated with the diffraction of the detected photoelectrons from the ordered SnPc overlayer [4]. Temperature dependent spectra in comparison with LEED data show that from the photoemission signal quantitative structural information for instance on the Debye-Waller factor or on a thermally induced structural phase transition of the adsorbate layer can be evaluated. First time-resolved experiment addressing the photoemission diffraction signal of SnPc/SCG will be presented and prove a time-resolution of ≈ 100 fs, essentially reflecting the laser pump-probe cross correlation signal.

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THEORETICAL INSIGHTS INTO TIME-RESOLVED PHOTOELECTRON ORBITAL IMAGING OF EXCITED-STATE DYNAMICS AT MOLECULE-SUBSTRATE INTERFACES

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Time-resolved momentum microscopy at free-electron laser (FEL) sources allows for gaining unprecedented insight into photo-induced dynamics at hybrid molecule-substrate interfaces, since it reveals details of electronic and structural transformations on atomic scale. We theoretically describe time- and momentum-resolved photoelectron spectroscopy of excited-state dynamics in molecules adsorbed on substrates. We reveal details of excited-state dynamics in a pentacene bilayer adsorbed on a silver substrate [1] and Copper phthalocyanine (CuPc) adsorbed on Titanium Diselenide (TiSe2) [2] from simulated photoelectron momentum maps compared to experimental data obtained at the Free-Electron Laser FLASH in Hamburg. We also demonstrate that time-resolved momentum microscopy can be used as a powerful tool to reveal photo-induced electron dynamics in molecules on attosecond to few-femtosecond time scales in the future.

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TIME-RESOLVED ARPES PROBING RABI-OSCILLATIONS AND LANDAU-ZENER-STÜCKELBERG INTERFERENCES IN GRAPHENE – A PROPOSAL

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At sufficiently high intensities, the interaction of few-cycle laser fields with solids gives rise to strong-field (non-perturbative) effects envisioning novel and exciting strategies for controlling optical and electronic properties via the electric field waveform on sub-femtosecond timescales, i.e., at petahertz frequencies. A striking example is the CEP-control of light-field-driven currents in graphene due to electronic quantum path interferences [Landau-Zener-Stückelberg (LZS) interferences] [1] resulting from the complex interplay of field-driven adiabatic intraband motion and non-adiabatic interband tunneling transitions [2]. Simulations show that the LZS-interferences give rise to characteristic asymmetries in the momentum-distribution of the residual conduction band population in the Dirac cone on top of a symmetric quasi-periodic pattern indicative for Rabi oscillations (see Fig. 1). Therefore, trARPES using few cycle femtosecond pump laser pulses seems to be in ideal tool for the investigation of these processes and there dependence on parameters such as pulse peak electric field strength E_0, laser polarization, and CEP phase.

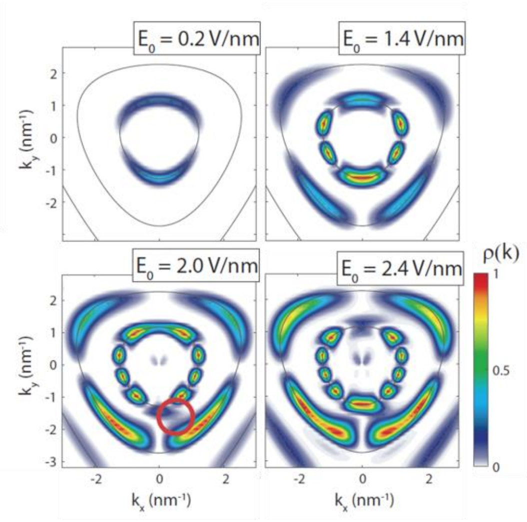

Figure 1 – Calculated residual conduction band population in graphene near the K-point for different values of E_0. The data show results for 800 nm, 5.5 fs laser pulse at $\varphi_{CEP}=\pi/2$ and a linear polarization along the x-direction. Figure from Ref. [1]

Based on preliminary results on graphite using 7 fs few-cycle NIR pump-pulses [3] we will discuss in this presentation the prospects, but also the challenges that arise in such type of trARPES experiment. We particularly will show that in spite of the critical problem of space charging at high pump peak intensities past experiments were already close or have even entered the relevant field-strength regime. Furthermore, simulations will illustrate, in which way we aim to probe the characteristic signatures of strong-field effects with the available experimental setup and how dephasing processes will affect the experimental data.

If successful, the experiments could establish an exciting strategy for probing in an direct manner light-matter interaction processes beyond the conventional perturbative single photon absorption regime. This includes for instance strong-field photoemission interferometry for the study of topological properties of two-dimensional materials [4], the reciprocal space exploration of coherently controlled electron trajectories [5], or energy- and momentum-resolved CEP-phase and two-pulse few cycle control experiments in novel 2D-materials.

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TIME-RESOLVED MOMENTUM MICROSCOPY OF MOIRÉ INTERLAYER EXCITONS

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Transition metal dichalcogenides (TMDs) are extensively studied because of their exceptional material properties such as the strong-light-matter coupling, spin-valley locking, and large exciton binding energies. The stacking of different TMDs can lead to even more intriguing electronic properties: In type-II band aligned TMD stacks, novel excitonic states can be created where the electron and the hole contribution to the exciton are separated between the van-der-Waals-coupled TMDs. The twist angle between these layers allows further manipulation of the interlayer coupling, and induces a moiré potential, which can further induce emergent material properties. While most of the studies on interlayer excitons were done with purely energy-resolved or optical methods, we make use of our setup for time-resolved momentum microscopy [1] that is perfectly suited to probe the femtosecond dynamics of excitonic features with energy- and in-plane momentum resolution.

On this poster, we discuss the ultrafast formation dynamics of the interlayer excitons, the distinct interlayer exciton momentum pattern and its moiré hallmark, and a real-space reconstruction of the different excitonic signatures on a twisted WSe_2/MoS_2 heterostructure [2].

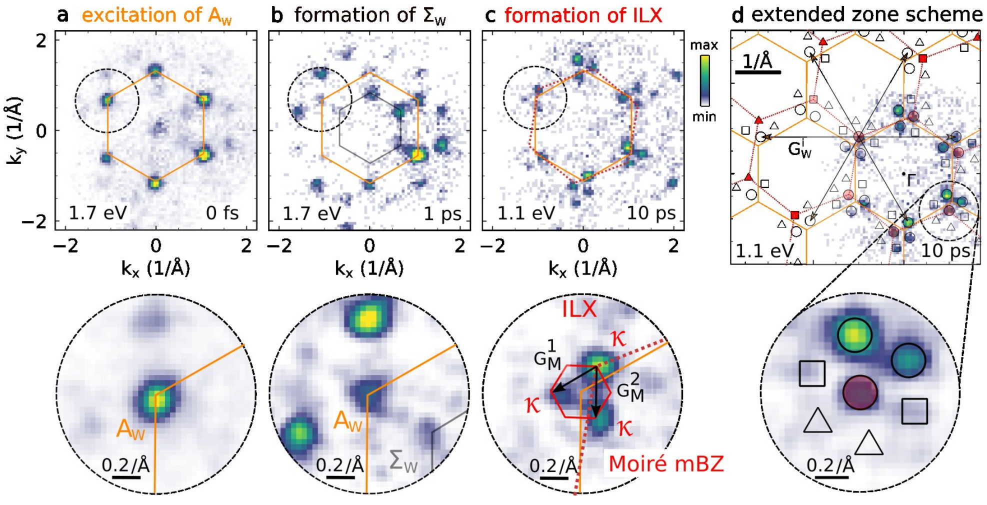

Figure 1 – Momentum-resolved fingerprints of the intralayer exciton, dark Σ exciton *and the interlayer exciton*

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Time-Resolved Photoemission Orbital Tomography of CuPc on Cu(001)-2O

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Photoemission orbital tomography is a powerful technique to image the electron distribution in occupied orbitals of molecules adsorbed at surfaces in momentum space by angle-resolved photoelectron spectroscopy. Recently, we have extended this technique by using high laser harmonics and a momentum microscope in a femtosecond pump-probe experiment to establish time-resolved photoemission orbital tomography (tr-POT) [1]. This allows to trace the population dynamics of initially unoccupied, optically excited molecular orbitals in order to study charge transfer across molecular interfaces on a femtosecond timescale.

In our first experiment, we were investigating a well-ordered 3,4,9,10-perylene-tetracarboxylic-dianhydride (PCTDA) layer on a Cu(001)-2O surface for which we were able to identify two distinct excitation pathways of the PTCDA molecules by changing the polarization of incident pump pulses [1]. The LUMO of PTCDA can be either populated from the HOMO by an intramolecular excitation or by electron transfer from the substrate and shows a surprisingly long lifetime of around 250 fs as shown in Fig. 1(c).

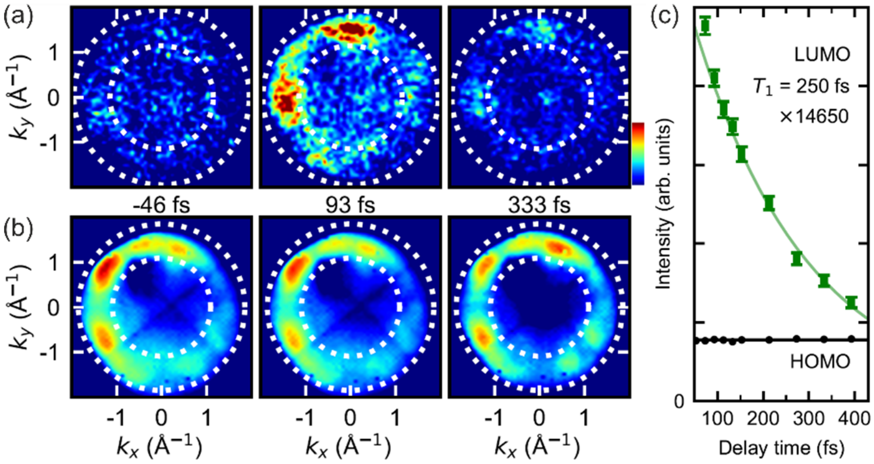

Figure 1*: (a,b): Momentum maps of the frontier orbitals LUMO and HOMO, respectively, of PTCDA on Cu(001)-2O at selected delay times between probe and pump pulse. (c) Corresponding PE intensity for both orbitals, showing an exponential decay for the LUMO.*

Here, we show for the example of a well-ordered monolayer of copper(II) phthalocyanine (CuPc) on the same Cu(001)-2O surface how the temporal evolution of the LUMO momentum distribution can be systematically disentangled from contributions of the projected HOMO which appears at the same final state energy in the case of resonant excitation from the HOMO into the LUMO. For this purpose, we tune the pump photon energy around the optical band gap of 1.8 eV for on- and off-resonant excitation, vary the pump polarization as well as the delay between pump und probe pulses and make use of the distinct momentum patterns of HOMO and LUMO. For p-polarized pump, we observe that the detected momentum distributions for CuPc are in fact dominated by the projected HOMO during temporal overlap between pump and probe pulses but gradually resemble the LUMO pattern at 0.5 eV above the Fermi edge for longer delays. The temporal evolution of the photoelectron intensity can be well described by a density matrix approach for a three level system which yields a LUMO population lifetime of about 50 fs.

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ULTRAFAST OPTICAL SPECTROSCOPY OF NICKEL OXIDE

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Strongly correlated materials form an integral part of modern opto-electronic studies and applications. In correlated transition-metal oxides (TMO), the strong electron-electron repulsion splits the *d*-bands into an occupied lower Hubbard band (LHB) and an unoccupied upper Hubbard band (UHB). The presence of additional oxygen (ligand) *p*-bands at the adjacent sites and their interaction with the Hubbard bands leads to complex properties governed by the Hubbard potential (U) and the charge transfer ($p \leftrightarrow$ LHB) gap Δ. In charge-transfer insulators Δ is smaller than U, and, thus, the *p*-band is located between the LHB and the UHB, as schematically depicted in Fig.1. Here we present excited state dynamics in nickel oxide (NiO), a charge transfer insulator with comparable U and Δ. Previous ultrafast two-photon photoemission studies by Gillmeister *et al.* have investigated the evolution of ultrafast photo-induced in-gap states and the coupling between antiferro-magnetically correlated spins and local Hund excitations. An UV-VIS absorption spectrum of NiO thin films on Si_3N_4 substrate shows three absorption peaks at 1.7 eV, 2.7 eV, and 4.5 eV. The 1.7 eV and 2.7 eV peaks are contributions due to multiple reflections from the 200 nm thick Si_3N_4 substrate on which NiO has been deposited. In time-resolved optical spectroscopy NiO is pumped above the band gap with a 4.8 eV photon energy and the ultrafast absorption spectrum is probed using time-delayed supercontinuum. The time-resolved absorption spectrum, see Fig. 2, shows within the first 500 fs positive pump-induced changes at 2.2 eV and 3.2 eV which are separated from substrate contributions. We also see a strong negative change around 4.3 eV. The relaxation dynamics of the positive and negative changes clearly differ. While the positive changes decay on time scales similar to the in-gap states reported by Gillmeister *et al*., the negative change represents the bleached ground state which recovers within 4 ps, potentially by interaction with phonons.

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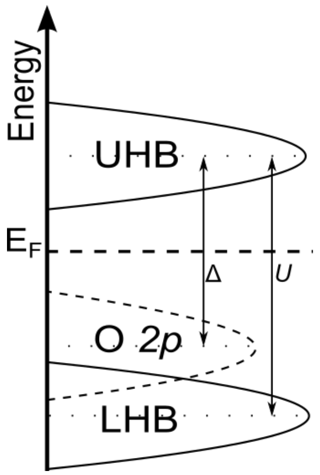

Figure 1: Band diagram of an Oxide Charge Transfer Insulator

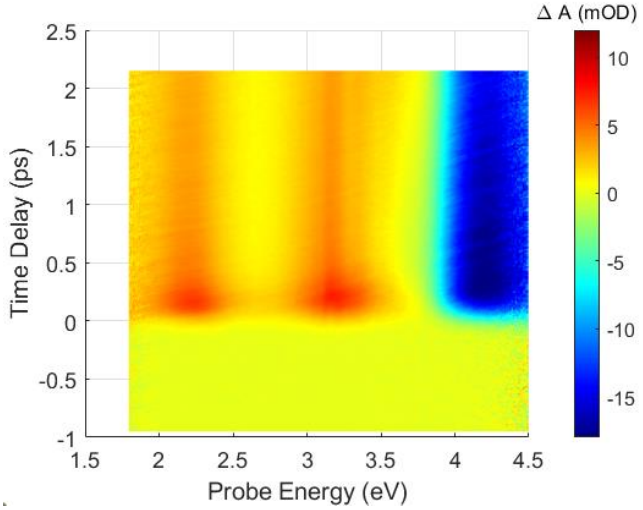

Figure 2: Experimental results: Transient absorption of a NiO thin film corrected for substrate contributions.

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