Energy dissipation from a correlated system driven out of equilibrium

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In complex materials various interactions have important roles in determining electronic properties. Angle-resolved photoelectron spectroscopy (ARPES) is used to study these processes by resolving the complex single-particle self-energy and quantifying how quantum interactions modify bare electronic states. Here we employ a combined theoretical and experimental treatment of femtosecond time-resolved ARPES and show how population dynamics measured using time-resolved ARPES can be used to separate electron–boson interactions from electron–electron interactions. We demonstrate a quantitative analysis of a well-defined electron–boson interaction in the unoccupied spectrum of the cuprate $Bi_2Sr_2CaCu_2O_{8-x}$ characterized by an excited population decay time that maps directly to a discrete component of the equilibrium self-energy not readily isolated by static ARPES experiments [1].

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Theoretical modeling of non-equilibrium spectroscopy

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A powerful method to study the interactions between electrons and bosons in high-Tc superconductors is the measurement of the single-particle spectral function. The recent development of time-resolved ARPES (tr-ARPES) has allowed this measurement of be performed out of equilibrium, where the material is driven by an ultrafast laser pump pulse. We have developed a theoretical framework to complement to these experiments, and here we report on several aspects of electron-boson coupling out of equilibrium.

First, we will illustrate how time-resolved spectroscopy can be used to study the coupling between electrons and phonons observing the decay rate of the transient signals as a function of energy, momentum, and time. A sufficiently strongly coupled phonon will exhibit a signature in the tr-ARPES spectra as both a kink in the dispersion as well as a sharp change of the decay rates, and we will discuss how these effects appear out of equilibrium. [1][2] Second, we will focus on the return to equilibrium in systems with multiple interaction types, and show that there are two distinct types of scattering processes: those types of interactions that conserve the energy within a subsystem, and those that do not. While in equilibrium these two contribute equally to the linewidth, we will show that out of equilibrium they behave differently -- the first type are mainly responsible for thermalization within the electronic subsystem, whereas the second type drain the energy out. As a result, the scattering rates out of equilibrium can be vastly different from the linewidth, and the features of the second type of interactions can be clearly observed.[3][4]

In addition, I will present some aspects of non-equilibrium physics in BCS superconductors. We solve the Nambu-Gor'kov equations for superconductivity within the Migdal-Eliashberg approximation, obtaining a full dynamic description of non-equilibrium BCS superconductivity. The temporal behavior after a pump exhibits characteristic 2D oscillations, which we attribute to the Higgs, or amplitude mode[5]. Finally, motivated by recent experiments[6], I will illustrate how superconductivity can be enhanced or suppressed through non-linear phononics. By modifying the physical parameters, we can model the driving of a lattice distortion, leading to an enhanced Tc[7].

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Transient photo-induced melting of charge-density-wave in 1T-$TiSe_2$: a femtosecond time-resolved photoemission investigation

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The driving force leading to the charge-density-wave (CDW) phase transition in 1T-$TiSe_2$ is still highly debated. Among the proposed mechanisms, the excitonic insulators model, i.e. the Coulomb interaction between Γ holes and L electrons, has gained major attention thanks to recent time-resolved experiments [1,2]. However, lattice distortion and electron-lattice interaction should also play an important role, as suggested by theoretical calculations [3,4]. We employed femtosecond time- and angle-resolved photoemission spectroscopy to investigate the photo-induced melting of CDW in 1T-$TiSe_2$ single crystals. Our measurements reveal that spectral weight and energy position of the valence band undergo different dynamics after photo-excitation with ultrashort visible laser pulses. This discrepancy is analyzed in the contest of the excitonic model and the band-type Jahn-Teller mechanism.

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Electrons dynamics at the surface of CH3NH3PbI3

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The grain structure of heterogeneous CH3NH3PbI3 plays a major role in the conversion efficiency of solar cells based on hybrid perovskites. Nonetheless, the dynamics of electrons at the surface of crystallites facets is still poorly understood. I will discuss two photon photoemission spectroscopy experiments of CH3NH3PbI3. The proposed approach monitors the electronic distribution of photoexcited electrons, explicitly discriminating electronic thermalization from slower dynamical processes. The reported results establish the initial energy relaxation time of hot carriers, suggest the proximity of a photoinduced phase transition, sets a limit on the surface induced recombination and outline the effects of internal electric fields in proximity of the perovskite-vacuum interface. Controlling such fields in heterogeneous films may boost the macroscopic efficiency of solar cells to the theoretical limit.

Ultrafast evolution of the prototype out-of-equilibrium Mott-Hubbard material V_2O_3

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The study of photoexcited strongly correlated materials is attracting growing interest since their rich phase diagram often translates into an equally rich out-of-equilibrium behavior. With femtosecond optical pulses, electronic and lattice degrees of freedom can be transiently decoupled, giving the opportunity of stabilizing new states inaccessible by quasi-adiabatic pathways. The prototype Mott-Hubbard material V_2O_3 presents a transient non-thermal phase developing immediately after ultrafast photoexcitation and lasting few picoseconds. Recent results on this model system will be presented, combining different ultrafast techniques (time-resolved photoemission, reflectivity, and FEL-based X-ray diffraction) [1].

For both the insulating and the metallic phase, the formation of the transient configuration is triggered by the excitation of electrons into the bonding a_{1g} orbital, and is then stabilized by a lattice distortion characterized by a hardening of the A_{1g} coherent phonon, in stark contrast with the softening observed upon heating. These results show the importance of selective electron-lattice interplay for the ultrafast control of material parameters, and are relevant for the optical manipulation of strongly correlated materials.

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Controlling the mass of confined electrons with light in graphite

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Many-body interactions with the underlying bulk electrons determine the properties of confined electronic states at the surface of a metal. Using momentum resolved nonlinear photoelectron spectroscopy we show that one can tailor these many-body interactions in graphite, leading to a strong renormalization of the dispersion and linewidth of the image potential state. These observations are interpreted in terms of a basic self-energy model, and may be considered as exemplary for optically induced many-body interactions.

Electron Dynamics in Single-Layer Transition Metal Dichalcogenides

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

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

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

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Novel topological phases of matter: an ultrafast view

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We have recently proven the potentiality of time and angle resolved photoelectron spectroscopy (tr ARPES) to investigate the unoccupied electronic states of topological insulators (TIs) [1] and to track the dynamics of the topological protected Dirac particles after optical perturbation [2]. The coherent optical excitation of the laser can be also exploited to modify, at the ultra-short timescale, the material electronic transport properties. This approach has been successfully carried out on $ZrTe_5$ [3], a novel compound, which has recently puzzled the scientific community due to its poorly understood transport and topological properties [4, 5].

TIs represent the first exponents of a much wider class of compounds, characterized by non-trivial topological properties. Among all, 3D Dirac semimetals [4, 6], Weyl semimetals [7] and nodal line Dirac semimetal [8] have being subject of an increasing number of ARPES studies. We will examine the possibility of exploiting tr ARPES, based on novel HHG sources, to access the electron dynamics in these topological phases of matter.

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Excited-state band mapping with XUV-based trARPES

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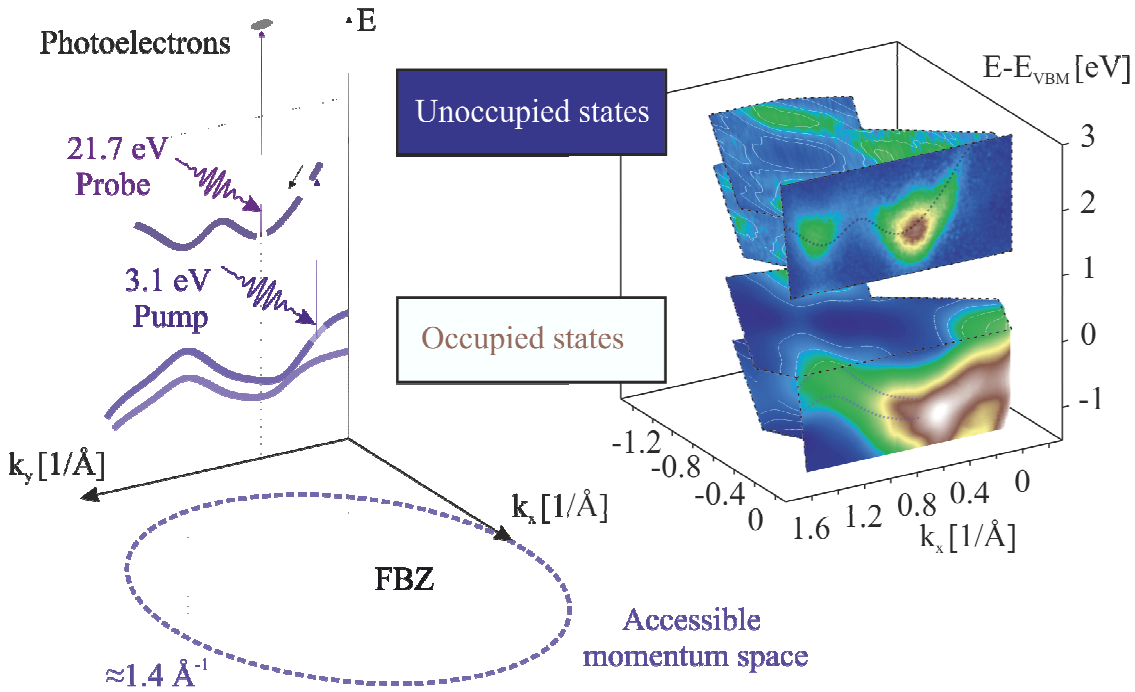
Time-resolved photoemission combined with scanning of the emission angle can extend established band-structure mapping to excited states which are only occupied out-of-equilibrium. The full potential of time- and angle-resolved photoemission spectroscopy (trARPES) is reached by performing the experiment at high repetition rates of hundreds of kHz, limiting space charge effects and data acquisition time. Extreme ultraviolet (XUV) photon energies grant access to the whole Brillouin zone: by performing high-harmonic generation at 0.5 MHz with a novel laser light source [1], we demonstrate trARPES with a photon energy of 21 eV, ≈100 meV resolution and sub-50 fs time resolution.

A 3.1 eV pump pulse populates the conduction band of the layered transition metal dichalcogenide WSe_2 followed by relaxation toward the band minimum within 1 picosecond. About 100 fs after excitation, most of the normally unoccupied conduction band states are populated due to scattering of the photo-excited carriers, allowing excited state band mapping throughout the whole Brillouin zone. This demonstrates how trARPES can measure the bandgap of a material in a single experiment with full time and momentum resolution.



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Ultrafast Excited State Dynamics and Insulator-to-Metal Transitions probed by trARPES

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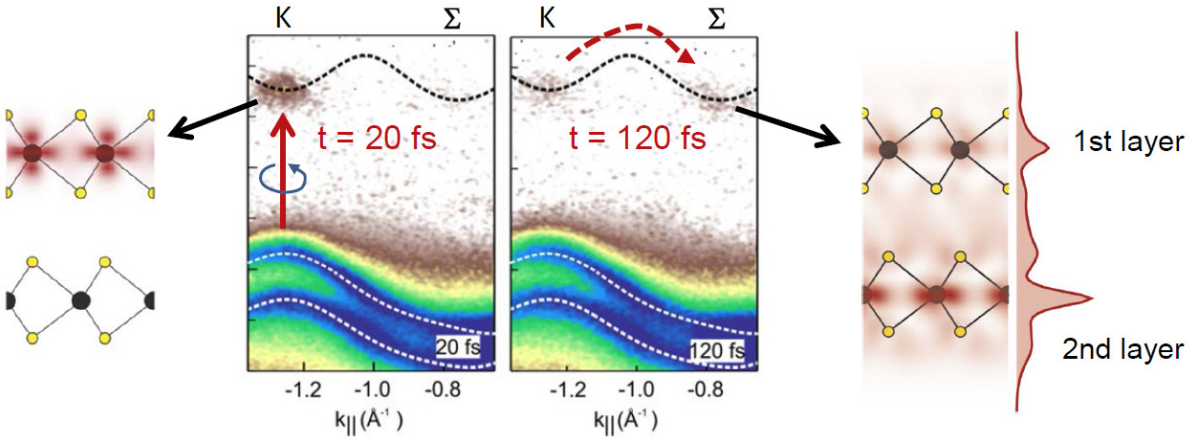
We employ high-harmonic-driven time- and angle-resolved photoemission spectroscopy (trARPES) to obtain a direct momentum resolved view of the ultrafast excited state dynamics and interlayer charge transfer in semiconducting transition metal dichalcogenides (TMDCs). While in bulk TMDCs the overall crystal structure is inversion symmetric, individual layers lack this property and in combination with strong spin-orbit coupling the energy degeneracy of electronic bands of opposite spin polarizations is lifted. Applying circularly polarized light leads to momentum- and spin-selective selective excitation of spin-polarized electrons in the K-valleys of the TMDC band structure. We show that, even in centrosymmetric samples of 2H-WSe_2 we can generate spin-, valley- and layer-polarized excited states in the conduction band which are localized within individual layers at the K points [1]. Subsequent ultrafast scattering populates states at the Σ-valley with a three-dimensional character facilitating optical control of inter-layer charge transfer (see figure).



Furthermore, we investigate the ultrafast photoinduced insulator-to-metal transition of quasi-one dimensional (1D) metal wires on In/Si(111) by trARPES. Starting from the insulating (8x2) phase we follow the gradual evolution of the electronic structure into the (4x1) phase on a 500 fs time scale. The gap at the Brillouin zone boundary is observed to close after 200 fs, while states at the zone center shift from above to below the Fermi level within 500 fs. A coherent phonon mode at 2.4 THz is observed in both phases. However, the absence of a coherent amplitude mode, as observed in Peierls systems such as the quasi-1D RTe_3 compounds [2], suggests a non-standard mechanism for the phase transition. We discuss this mechanism in the context of recent time-resolved electron diffraction results [3].

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Investigation of the intervalley scattering dynamics in MoS_2 by two-photon photoemission with a high-harmonic probe at 100 kHz repetition rate

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

In the first part of my talk I will focus on the development of the high-harmonic source that uses a Xe gas jet for efficient generation of 23.25 eV photons at 100 kHz repetition rate with only a few µJ pulse energy of 800 nm/40 fs fundamental laser pulses [1]. In the second part, I will discuss the application of this setup to the investigation of the electron dynamics in the conduction band of MoS_2 after optical excitation with different pump photon energies. Recently, we have shown that optical excitation above the A exciton resonance at ~1.8 eV with 2.05 eV pump pulses results in an immediate occupation of the conduction band at K followed by an ultrafast transfer to the conduction band minimum at Σ [2]. Subsequently, the occupation at both high-symmetry points decays slowly on a ps timescale. Here, we will present new data for pump photon energies in the range of 1.8–2.1 eV and show how the dynamics of this transfer depend on the excess energy above the exciton resonance.

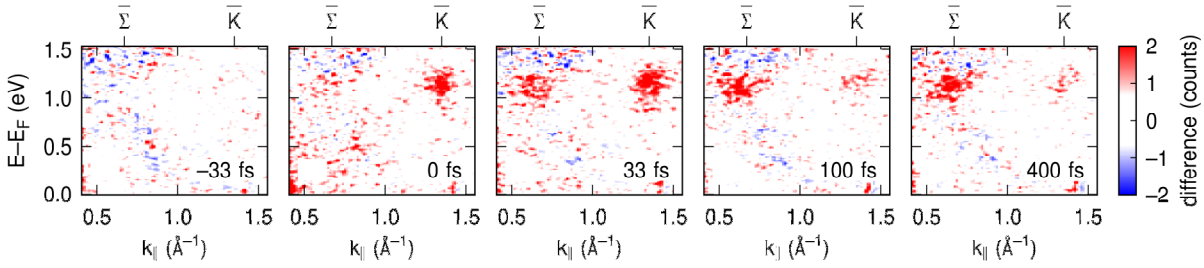

Fig. 1: Energy-momentum maps of the difference between the 2PPE intensity in the conduction band of MoS_2 after and before the optical excitation with 2.05 eV pump pulses for different pump-probe delays.

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High-repetition-rate XUV trARPES: A Sensitive Probe of Ultrafast Materials Dynamics

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Time- and angle-resolved photoemission spectroscopy (trARPES) represents a powerful approach to resolve the dynamics of the electronic structure, quasiparticles, and correlations in quantum materials. Since the accessible energy and momentum range both increase with photoelectron kinetic energy, incident photons in the extreme-ultraviolet (XUV) are highly desirable. Moreover, a high laser repetition rate is important for sensitive measurements with high photon flux due to space charge limitations. However, often trARPES is limited in repetition rate to maintain high peak intensities for XUV generation, or operates at high repetition rates yet with limited VUV probe photon energy. In this talk, I will discuss our recent development of a novel trARPES setup based on a 50-kHz source of bright and narrowband XUV harmonics, along with its application to ultrafast materials studies.

The experiments employ bright XUV pulses at 50-kHz repetition rate with average source flux exceeding 10^{13} ph/s at 22.3 eV. These are generated by driving high-harmonic generation with the UV second-harmonic of a Ti:sapphire laser amplifier, focused tightly into Kr gas. The cascaded scheme yields high conversion efficiency ($\approx 5 \times 10^{-5}$) corresponding to a two-orders-of-magnitude enhancement via dipole wavelength scaling and enhanced phase matching. Importantly, the spectral structure enables the direct, high-contrast isolation of a single harmonic with narrow bandwidth down to ≈60 meV. The XUV pulses are refocused with a toroidal mirror, delivering $\approx 10^{10}$-10^{11} ph/s flux to the sample. ARPES is performed in a customized UHV chamber equipped with a hemispherical electron analyzer, six-axis sample manipulator and cryostat, and side chambers for sample loading, storage and preparation. Part of the laser output is available for excitation up to mJ/cm^2 intensities.

Initial applications of the 50-kHz XUV trARPES setup to the study of electronic materials dynamics will be presented. The high flux enables rapid band mapping across the Brillouin zone of quantum materials, along with sensitive measurements that access e.g. the crossover from perturbative excitations to photo-induced phase transitions. We will discuss in detail experiments that track the fundamental dynamics of the semiconducting transition-metal dichalcogenide $MoSe_2$. At low temperatures, the 1.6 eV pump photon energy is tuned resonant to the absorption line of the strongly-bound A-excitons. After photo-excitation, a signal is immediately observed at the *K*-point, followed by rapid inter-valley scattering to the conduction band minimum at the Σ-point on a 70-fs timescale. Moreover, distinct excitation-dependent and dynamical energy shifts are observed in the transient photoemission spectra that indicate the observation of band-gap excitons via ARPES.

This work was carried out together with J. H. Buss, H. Wang, Y. Xu, F. Joucken, J. Maklar, S. Ulonska, P. Ranitovic, C. Ko, S. Tongay, J. Wu, Z. Hussain, A. Lanzara, and supported by the DOE Office of Science, Materials Sciences and Engineering Division under contract DE-AC02-05CH11231.

Tracking electron dynamics in Graphene and TMDCs

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Novel quantum materials such as graphene and transition metal dichalcogenides (TMDC) are attracting vast interest particularly for their application in spintronic and optoelectronic devices. Their properties are intrinsically governed by the electronic structure of the large momentum electrons (at the Brillouin zone K-point). In order to eject such electrons in vacuum and observe their dynamics, a high energy (>20 eV) photon source is required as well as ultrashort pulse duration. High Harmonic Generation source [1] combined to an Angle-Resolved PhotoEmission Spectroscopy (ARPES) end-station is a powerful tool to observe such electron dynamics. Here I will present an overview of our recent time-resolved ARPES studies [2-5] performed at the Artemis facility.

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Chronology of Photo-Excited Carriers at Surfaces Studied by Time-Resolved Photoemission Spectroscopy

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Photoemission spectroscopy using vacuum ultraviolet/soft X-rays has been the important experimental method to *probe* electronic structure of a matter *directly*. The time-resolved measurements can be carried out with synchrotron radiation (SR) or laser (FEL, HHG) beams to *chase* temporal variations of the non-equilibrium electrons or electronic states *in real time* [1]. Taking benefits of the ultra-short pulse-width of the lasers, elementary processes of dynamical phenomena are measured to seek for critical factors in phase transitions, for example. On the other hand, high brilliant SR has nowadays been used for the dynamical *operando* experiments to find the limiting processes in valuable chemical reactions. The academic and industrial demands in time-resolved photoemission spectroscopy have grown recently.

In the present talk, several topics of the time-resolved photoemission researches on surface science using two types of soft X-ray sources, SR and HHG, are presented.

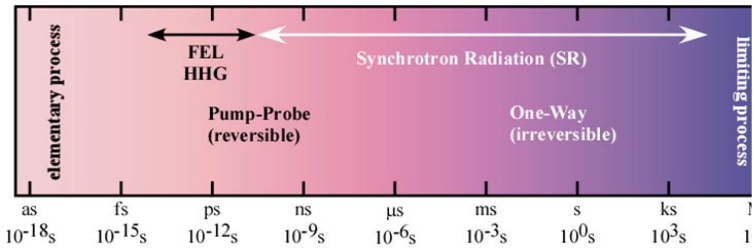

Figure Chronology-diagram of experiments using various types of vacuum ultraviolet/soft X-ray beams

At the soft X-ray high-brilliant beamline, SPring-8 BL07LSU[2], we observed unexpected intermediate molecules in the CO_2-to-alcohol reaction by time-resolved near-ambient pressure XPS [3] and tracked carrier transfer across interfaces of the regulated heterojunctions [4,5]. With a HHG laser, we studied ultrafast carrier dynamics of Dirac electrons in graphene layers on the designed substrates [6]. At the summary of the talk, chronology of the photo-excited carriers at surfaces is overviewed together with discussion of the concerting uses of various light beams as the experimental sources.

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Electron-Phonon Coupling and Unoccupied States in Quantum Materials

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Time- and angle- resolved photoemission spectroscopy (trARPES) offers a powerful toolset for studying dynamical phenomena in quantum material systems. Here we focus on two classes of experiments which highlight these unique capabilities. First, we study electron-phonon coupling by measuring the electronic response to coherently excited phonon modes. In the unconventional superconductor FeSe, we perform parallel trARPES and time-resolved x-ray diffraction experiments. By measuring the electronic and structural dynamics together, we directly quantify the electron-phonon coupling strength. Comparison with theory reveals the cooperative interplay of correlation effects in FeSe [1]. In the cuprate superconductor Bi2212, this approach offers a promising route toward quantifying the momentum-dependence of the coupling strength [2].

The second class of experiments employs two-photon photoemission (2PPE) to map the band structure of unoccupied electronic states. This approach has been used to discover new topologically-nontrivial states in Bi_2Se_3 [3]. We have recently extended this technique to study unoccupied states in Bi2212. These measurements establish a basis for comparison to Hubbard model calculations, which place constraints on the correlation strength U [4].

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Dynamic spin filtering at the Co-Alq_3 interface mediated by weakly coupled second layermolecules

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Spin injection and spin filtering at organic-metal interfaces aredominated by the specific interaction between the organic molecules and the inorganic magnets used as electrodes [1]. Here we demonstrate a spin-filtering mechanism based on the dynamical spin relaxation of the long-living interface states formed by the magnet and weakly physisorbed molecules. We investigate the case of Alq_3 on Co and, by combining spin- and time-resolved two-photon photoemission experiments with electronic structure theory, show that the observed long-time spin-dependent electron dynamics is driven by molecules in the second organic layer [2, 3]. The interface states formed by physisorbed molecules are not spin-split, but acquire a spin-dependent lifetime, that is the result of dynamical spin-relaxation driven by the interaction with the Co substrate. Such spin-filtering mechanism has an important role in the injection of spin-polarized carriers across the interface and their successive hopping diffusion into successive molecular layers of molecular spintronics devices.

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Influence of graphene on the interface states with selected transition metals (Ir, Cu, Ni)

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Graphene single layer, the most popularly mentioned and first discovered 2D material, has become the focus of worldwide research since the huge list of superlative properties. The presence of graphene overlayers on metallic surfaces affects the interface electronic properties including the surface states that, being characterized by a large spatial expansion into the vacuum of the associated wavefunctions, sensitively depend on the graphene-metal interaction [1].

Image states comparable to those of the clean metal surface were observed both in weakly (Gr/Ir) and strongly (Gr/Ru) interacting systems. In the former, the image state appears essentially unchanged by the presence of graphene [2]. Due to a strong corrugation of the graphene layer, the strongly interacting graphene/Ru interface shows two series of image potential states [3].

Herein, by using circularly polarized femtosecond laser pulses in non-linear photoemission measurements, we show that the unoccupied n = 1 image state in the Gr/Ir weakly interacting system exhibits a Rashba splitting as well as the occupied surface state. Due to the weak interaction, graphene grows on Ir(111) surface as an almost free-standing ordered sheet. As a consequence, the Rashba splitting on the surface state remains giant, while on the n=1 image state results fifty times smaller than that measured on the occupied one [4].

Moreover, by means of angle-resolved non-linear photoemission, we discuss two additional systems: the weakly interacting Gr/Cu(111) and the strongly interacting Gr/Ni(111). Comparing non-linear, angle-resolved photoemission data collected on graphene/Cu(111) and on a single layer graphene grown on polycrystalline copper, we provide evidence for the existence of an interface state at the Gr/Cu(111) which strongly hybridizes with the first image state. Moreover, as confirmed by calculations based on a one-dimensional model potential, this hybridization results in two states with a significant amplitude probability in the correspondence of Gr/metal well [5].

On Gr/Ni(111) interface, an occupied and two unoccupied surface states appear. One of the unoccupied state has been ascribed to the n=1 image potential state, the other to an interface state originating by the presence of graphene. In contrast with the Gr/Cu(111), in this system the bulk origin of the interface states can be clearly identified by ab initio calculation.

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Recent developments in the one-step description of time-resolved photoemission

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A brief introduction to the theory of angle-resolved photoemission spectroscopy (ARPES) of solids is given with an emphasis on the so-called one-step model of photoemission that describes excitation, transport to the surface and the escape to the vacuum in a coherent way [1].

Recently a theoretical frame for the description of pump-probe photoemission has been developed. The approach is based on a general formulation using the Keldysh formalism for the lesser Green function to describe the real-time evolution of the electronic degrees of freedom in the initial state after a strong pump pulse that drives the system out of equilibrium [2]. As a first application, the theoretical description of two-photon photoemission (2PPE) for Ag(100) within the SPR-KKR-approach is introduced [3]. Furthermore, this formalism and the corresponding numerical implementation has been generalized to the ferromagnetic case. First examples of fully spin-polarized 2PPE calculations on Fe(110) will be presented.

Finally, a new approach is sketched that allows to account also for dynamical correlations in the non-equilibrium state caused by the pump pulse by means of a corresponding two-time dependent self-energy $\Sigma(z,z')$. This central quantity of the scheme may be calculated, for example, up to second order in the framework of non-equilibrium DMFT in the so-called weak coupling approach [4].

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Signatures of ultrafast magnetization dynamics in the electronic band structure

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Femtomagnetism has become an established and active research field in magnetism. Still it is debated on which timescale the band structure and spin polarization of a ferromagnet changes after femtosecond laser excitation and how this affects the magnetization dynamics.

To approach these problems, we perform time- and spin-resolved two-photon photoemission experiments with laser pulses and time- and angle-resolved photoemission with high-order harmonic VUV source. We have studied ultrafast demagnetization for the local-moment ferromagnets Gadolinium and Terbium. In the lanthanides equilibration of the excited state involves more than one timescale, because the optical excitation occurs in the valence band but the magnetic moment is dominated by the localized 4*f* electrons.

Following the excitation by a femtosecond infrared pulse (0.95-eV and 1.55-eV photon energy @ Artemis and our Lab), we directly map the transient exchange splitting of the Gd and Tb valence bands near the center of the bulk Brillouin zone by time- and angle-resolved photoemission using VUV-pulses. Simultaneously we record the magnetic linear dichroism of the *4f* photoemission line. This allows us to compare the magnetization dynamics of *4f* core and *5d6s* valence electrons in one measurement.

We see striking differences between the Gd ($4f^7$, L=0) and Tb ($4f^8$, L=3), demagnetization dynamics, which we attribute to the difference in 4f spin – lattice coupling. The Gd magnetization dynamics in the first 100 fs hints at a coherent coupling of 5d and 4f spins.

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Bypassing the energy-time uncertainty in time-resolved photoemission

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The energy-time uncertainty is an intrinsic limit for time-resolved experiments, as it imposes a tradeoff between the duration of the light pulses used in experiments and their frequency content. In standard time-resolved photoemission, this limitation is reflected in a tradeoff between the temporal resolution and the resolution on the photoelectron kinetic energy. [1,2]
I will present a proposal for an experimental scheme that allows to disentangle the energy and time resolutions in time-resolved photoemission. I will show that dynamical information on all timescales can be retrieved from time-resolved photoemission experiments using suitably shaped light pulses of classical or quantum nature.
As a paradigmatic example, I will theoretically discuss the dynamical buildup of the Kondo peak, a narrow feature in the electronic response function arising from the screening of an impurity magnetic moment by conduction electrons. After a quench, the Kondo screening builds up on timescales shorter than the inverse width of the Kondo peak [3], and I will illustrate how the proposed experimental scheme could be used to measure such timescales.
The proposed approach provides an experimental framework to access the nonequilibrium response of collective electronic properties beyond the spectral uncertainty limit and will enable measurements which are now hindered by the energy-time uncertainty, e.g. the measurement of the melting of Mott gaps or of the dynamics of excited Higgs modes.

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Ultrafast Magnetization Relaxation Dynamics in $La_{0.66}Sr_{0.33}MnO_3$ Films

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Hole-doped rare-earth manganites, like $La_{0.66}Sr_{0.33}MnO_3$ (LSMO), display exotic phenomena such as concurrentcolossal magnetoresistance and half-metallicity which originate from the interplay of charge, spin, and orbitaldegrees of freedom [1]. The peculiar transport properties of LSMO thin films combined with the ferromagneticorder that persists up to about 350 K [2] render such system a most technologically attractive material for spininjection: the spin polarization at the Fermi level reaches about 100% for $T<T_{Curie}$[3].
The ultrafast manipulation of spin states in LSMO can be tested by state-of-the-art time-resolved pump-probetechniques. Previous studies by optical pump-probe spectroscopy have given evidence of photoinduced effects inferromagnetic manganites [4].
Photo-Electron Spectroscopy (PES) allows a direct measurement of the electronic structure; time-resolved PES isable to disentangle the delicate out-of-equilibrium interplay between electronic, spin and lattice degrees of freedom[5], an essential feature in the case of highly correlated materials. HArd X-ray PhotoElectron Spectroscopy(HAXPES) extends the probing depth of PES to the bulk of the solid (tens of nm), and therefore does not suffer ofthe modification induced by the surface.
We present here a pump-probe HAXPES study of the relaxation dynamics of LSMO thin films. We study thestructure of the Mn 2p core level and, in particular, the bulk-only screening channel proportional to the metallic andferromagnetic state in LSMO. We observe a large and ‘slow’ reduced lineshape change up to 200 picoseconds afterthe IR pumping. By comparison with all-optical techniques (Time-Resolved Magneto-Optical Kerr effect, TRMOKE)we are able to attribute the observed quenching to a collapse of magnetic order. The suddendemagnetization reduces the mobility of electrons in the solid, inducing a localization similar to a metal-insulatortransition.
Since LSMO is half-metallic, the direct exchange of energy between the optically excited electrons and the magneticorder is inhibited by the absence of final states for spin-flip scattering [3]. So we can follow the relaxation dynamicsas the energy is first dissipated in the lattice and then in a reduction of the magnetic order.

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Time resolved ARPES on n-doped and p-doped Topological Insulators

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Topological insulators (TIs) represent a new quantum state of matter that is currently attracting great attention thanks to their unique electrical transport properties. This growing interest is due, in particular, to their characteristic spin texture [1] and because these materials are ideal candidates for future spintronics devices [2,3].
The first observation of a persistent electron population in the photo-excited surface state (SSs) of TIs has focused the attention, in particular, on the out-of-equilibrium electronic properties of these materials [4,5]. In this perspective a detailed knowledge of the dynamical response of TIs after optical excitation is needed. The aim is to enhance the surface spin transport properties circumventing the limitation of the bulk transport. The most relevant mechanism, which was initially proposed as responsible for the relaxation dynamics of the photo-excited electrons, is the electron-phonon scattering. Hence, a deep understanding of the scattering mechanisms between the surface and bulk states of TIs is a key issue.
Recent experimental works have shown that the time-resolved version of the Angle Resolved Photoelectron Spectroscopy (tr-ARPES) may have a key role in the study of these systems.
We performed tr-ARPES experiments on a wide set of different doped TIs providing a complex picture. We display measurements performed on n-doped ($GeBi_2Te_4$, $GeBi_4Te_7$) intrinsic (Bi_2Te_3, $GeBi_2Te_4$) and p-doped (Sb_2Te_3, Sb_6Te_3). We observed different dynamics probing bulk and surface states. Experimental data clearly show that surface and bulk states play a crucial role in electron relaxation dynamics processes. Comparable behaviors' also indicates a strong coupling between the two.

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Multi-MHz time-of-flight electronic bandstructure imaging of graphene on Ir(111)

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In the quest for detailed spectroscopic insight into the electronic structure at solid surfaces in a large momentum range, we have developed an advanced experimental approach. It combines the 3D detection scheme of a time-of-flight momentum microscope with an optimized filling pattern of the BESSY II storage ring. Here, comprehensive data sets covering the full surface Brillouin zone have been used to study faint substrate-film hybridization effects in the electronic structure of graphene on Ir(111), revealed by a pronounced linear dichroism in angular distribution [1]. The method paves the way to 3D electronic bandmapping with unprecedented data recording efficiency.

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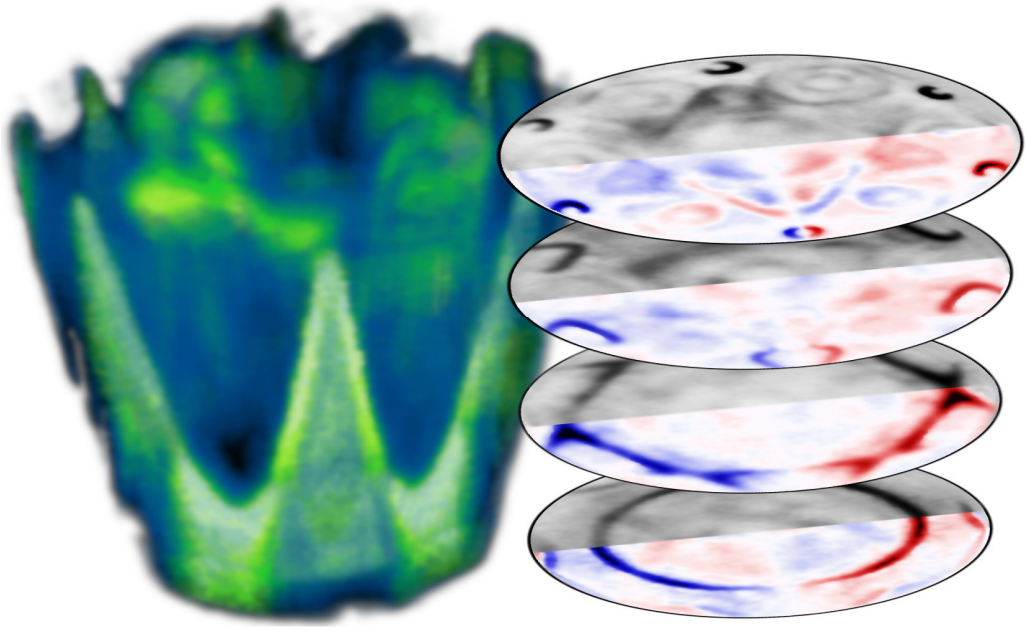

Fig1: *Three dimensional data set of the band dispersion of graphene on Ir(111) measured during 35min acquisition time with the ToF momentum microscope at BESSY-II (left), and linear dichroism in the angular distribution (LDAD) for $k_{||}$ momentum disks at selected binding energies (right). The red-blue color scale shows the LDAD asymmetry derived from a single acquisition due to the off-normal incidence of the light [1].*

Frontier research at FERMI

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The most recent light sources, extreme ultraviolet (EUV) and X-ray free electron lasers (FELs), have extended table top laser experiments to shorter wavelengths, adding element and chemical state specificity by exciting and probing electronic transitions from core levels. Through their unique properties, combining femtosecond X-ray pulses with coherence and enormous peak brightness, the FELs have enabled studies of a broad class of dynamic phenomena in matter that crosses many scientific disciplines and have led to major breakthroughs in the last few years. In this talk, we intend to discuss how the advances in the performance of the FELs, with respect to coherent control and multi-colour pulse production, may push the development of original experimental strategies to study non-equilibrium behaviour of matter at the attosecond–nanometer time–length scales. This would have a tremendous impact as an experimental tool to investigate a large array of phenomena ranging from nano dynamics in complex materials to phenomena that are at the heart of conversion of light into other forms of energy.

Femtosecond X-ray Photoemission Spectroscopy for Femtochemistry and Photovoltaics.

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The study of charge dynamics in chemical processes at surfaces by measuring in real time the changes of the electronic structure of the materials is nowadays possible thanks to the advent of free-electron lasers FELs. We studied by means of time resolved resonant x-ray emission (RXES) and time resolved x-ray absorption (XAS) photocatalytic reactions at surfaces by recording electronic structure changes in the femtosecond and picosecond timescale after an optical excitation. Besides photon in-photon out probing techniques like RXES, photon in-electron out techniques like PES can also be employed for the electronic structure study. I will discuss the advantages and limitations on the usage of time resolved photoemission (PES) for femtochemistry measurements at FELs. I will also present a preliminary time resolved PES experiment performed at a synchrotron facility to explore the charge dynamics induced by sunlight in donor/acceptor molecular system.

Magnetic and electronic properties during laser induced phase transitions.

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Electronic excitations induced by short laser pulses modify physical properties which can be studied using a wide number of time resolved experimental techniques. Although synchrotron radiation time resolution is poor to directly follow electronic excitations [1], synchrotron based experiments are precious to obtain a detailed characterization of the materials electronic properties including specific collective electronic excitations[2]. Spectroscopic results compared to "ab initio" theoretical description of initial states, electronic excitations and decay processes are necessary for a deep understanding of the observed phenomena[3]. Another advantage to include synchrotron based experiments for time resolved spectroscopic studies is the possibility to use soft X-rays circularly polarized photons which allows us to obtain on the same sample a direct measurement of electronic and magnetic properties[4].

I will present photoelectron spectroscopy experiments on core levels, valence band and resonant Auger electrons in pump/probe experiments with 800 nm short laser excitations to follow a laser induced demagnetization process and the Antiferromagnetic to Ferromagnetic phase transition in FeRh epitaxial layers.

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