O 5: Ultrafast Electron Dynamics at Surfaces and Interfaces I

Time: Monday 10:30–12:30 Location: MA 041

O 5.1 Mon 10:30 MA 041

Electronic Structure and Carrier Dynamics of the Bulk Semiconductor Magnet CrSBr — •Lawson Lloyd[1], Tommaso Pincelli[1,2], Túlio de Castro[1], Ferdinand Menzel[3], Andreas Stier[3], Nathan Wilson[3], Martin Wolf[1], Laurenz Rettig[1], and Ralph Ernstorfer[1,2] — [1]Fritz Haber Institute, Berlin, Germany — [2]TU Berlin, Berlin, Germany — [3]TU Munich, Garching, Germany

Van der Waals (vdW) layered magnets have the potential to enable novel optoelectronic and spintronic applications. Among these, CrSBr is a direct band gap semiconductor that hosts interlayer antiferromagnetic order, a highly anisotropic, quasi-1D electronic structure, and strongly bound excitons. However, understanding how the carrier and exciton dynamics couple to the underlying spin order is currently lacking. Here, we employ time- and angle-resolved photoemission spectroscopy to map the temperature-dependent band structure and carrier dynamics in bulk CrSBr. We observe an electronic band splitting that emerges below the magnetic transition temperature, which we interpret as arising from a super-exchange mechanism giving rise to intralayer ferromagnetism. Time-resolved measurements reveal a rapid band renormalization of the lower conduction band that occurs during photoexcitation, pointing towards complex many-body effects governing the excited state dynamics and optical properties. These results provide important experimental observations of the low-temperature electronic band structure and shed further light on the microscopic interactions driving carrier dynamics and spin order in this vdW magnet.

O 5.2 Mon 10:45 MA 041

The wandering spins of Fe_3GeTe_2: itinerant vs localized behaviour in the electronic structure — •Tommaso Pincelli[1,2], Tania Mukherjee[1,2], Lawson Lloyd[2], Jyoti Krishna[3], Túlio de Castro[2], Shuo Dong[4], Victoria Taylor[2], Yoav William Windsor[1,2], Martin Wolf[2], Laurenz Rettig[2], and Ralph Ernstorfer[1,2] — [1]Institute of Optics and Atomic Physics, Technische Universität Berlin, Berlin, Germany — [2]Fritz Haber Institute of the Max Planck Society, Berlin, Germany — [3]Max-Born-Institut (MBI), Berlin — [4]Institute of Physics, Beijing National Laboratory for Condensed Matter Physics, Beijing, China

Iron Germanium Telluride (FGT) is a van der Waals metallic ferromagnet that has shown a wealth of exotic phenomena, ranging from 2D ferromagnetism with gate-tunable Curie temperature to heavy fermion behaviour. Despite its interest, its electronic structure and the microscopic interactions giving rise to magnetic order are still poorly understood: evidence exists of both itinerant and localized magnetic behaviour, an anomaly in a purely 3d electronic system. By employing time-resolved angle-resolved photoemission spectroscopy (trARPES), we observe clear evidence of the closure of the Stoner exchange gap, a clear indication of an itinerant character of the electronic excitations. This is in contrast with previous results of quasi-equilibrium studies, which ascribed FGT demagnetization to localized excitations. We also observe, by employing frequency domain ARPES, the impact of phononic excitations, resulting in an A_{1g} phonon strongly coupled with the bandstructure at the Brillouin zone centre.

O 5.3 Mon 11:00 MA 041

Electron Dynamics in the Conduction Band of the van-der-Waals Magnet $Cr_2Ge_2Te_6$ — •Stephan Schmutzler, Jan Böhnke, Martin Weinelt, and Cornelius Gahl — Fachbereich Physik, Freie Universität Berlin, Arnimallee 14, 14195 Berlin, Germany

The electron dynamics in the central region of the Brillouin zone of $Cr_2Ge_2Te_2$ has been investigated by time-resolved ARPES. Optical excitation with a photon energy of 1.55 eV leads to a transient broadening of the valence band and simultaneously excites an electron population located at 0.7-1.3 eV above the Fermi level. Within 1 ps, this population continuously relaxes towards states at 0.5 eV. Considering their long lifetime of >3 ps, we attribute these states to the conduction band minimum. Our findings are in contrast to theoretical predictions that locate the minimum close to the $\overline{K}$-point of the surface Brillouin zone [1,2]. The broad momentum distribution in the conduction band suggests enhanced scattering in the excited material.

[1] D.-Y. Wang et al. Phys. Rev. B 107, 125148 (2023)

[2] Y. F. Li et al., Phys. Rev. B 98, 125127 (2018)

O 5.4 Mon 11:15 MA 041

Ultrafast charge and spin response to linear and circular polarized laser excitations in naturally layered delafossites $PdCoO_2$ and $PtCoO_2$ from time-dependent DFT — •Mike Bruckhoff, Markus E. Gruner, and Rossitza Pentcheva — Faculty of Physics and Center of Nanointegration, CENIDE, University of Duisburg-Essen

In the framework of real-time time-dependent density functional theory, we investigate the layer-resolved, ultrafast electronic dynamics after laser excitation of the non-magnetic, metallic delafossites $ACoO_2$ A = Pd, Pt. We simulate the responses to linear and circular polarized laser pulses with multiple laser frequencies and laser fluences. We observe a marked difference in the redistribution of atomic charges regarding in-plane vs. out-of-plane linear polarized laser pulses, which results in a charge transfer from the A-layer into the CoO_2-layer and vice versa depending on the laser frequency and polarization direction. In addition we encounter for $PdCoO_2$ low-frequency charge oscillations within the CoO_2-layer which occur after the decay of the laser pulse. These oscillations are accompanied by significant transient spin magnetic moments reaching values up to $0.08\mu_B$. Circular polarized laser pulses excite these low-frequency charge and spin oscillations in both delafossites using less intense laser pulses.
Funding by DFG within CRC1242 is gratefully acknowledged.

O 5.5 Mon 11:30 MA 041

Ultrafast lattice dynamics of $MoSe_2$ monolayers — •Victoria C. A. Taylor[1], Yoav W. Windsor[1,2], Elizaveta Patutkina[2], Hyein Jung[1,2], Jannik Malter[1], Mustafa Hemaid[3], Tobias Korn[3], and Ralph Ernstorfer[1,2] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany — [2]Technische Universität Berlin, Berlin, Germany — [3]Institut für Physik, Universität Rostock, Rostock, Germany

Transition metal dichalcogenides (TMDCs) are a promising family of 2D materials, which exhibit rich electronic phenomena. In particular, several notable TMDCs, such as $MoSe_2$, exhibit an indirect to direct band gap transition when reduced to monolayer thicknesses. Although extensive studies have investigated ultrafast electronic properties of TMDC monolayers, comparatively few have directly addressed the lattice response to photoexcitation.

Femtosecond electron diffraction (FED) is an ideal tool to study photoexcited lattice dynamics, as it provides quantitative information on coherent and incoherent atomic vibrations. Such information enables a direct understanding of energy flow from the electronic subsystem to the lattice via electron-phonon coupling, and within the lattice via phonon-phonon relaxation. We present FED measurements of $MoSe_2$ monolayers under different excitation conditions, in particular at the A-exciton resonance. As such, the results enable direct observation of the behaviour of phonons under ultrafast "nonthermal" conditions, i.e. those in which Bose Einstein statistics are not maintained.

O 5.6 Mon 11:45 MA 041

GHz manipulation of low-energy electron pulses — •Dennis Epp[1,2], Benjamin Schröder[1,2], Marcel Möller[1,2], and Claus Ropers[1,2] — [1]Department of Ultrafast Dynamics, Max Planck Institute for Multidisciplinary Sciences, 37077 Göttingen, Germany — [2]4th Physical Institute - Solids and Nanostructures, University of Göttingen, 37077 Göttingen, Germany

Electron pulses are a powerful probe in time-resolved diffraction and microscopy. One of the main challenges in such experiments is the dispersion-induced pulse broadening, both due to Coulomb interactions and the initial photoelectron energy distribution [1]. One technique to overcome this challenge is radio-frequency (RF) pulse compression, which is routinely used to control high-energy electron pulses [2]. However, corresponding schemes have not been developed for energies below a few kilo-electronvolts.

In this contribution, we demonstrate longitudinal phase-space manipulation of low-energy electron pulses using RF fields. Specifically,

we combine a millimetre-sized photoelectron gun [3] with a synchronized compression cavity driven at a frequency of 2.5GHz. The change in pulse duration induced by the cavity is measured by the deflection of transient space-charge cloud at a metal grid, resulting in a two- to four-fold compression of 80-,100- and 120-eV electron pulses [4].

[1] Dwyer, et al., Phil. Trans. R. Soc. 364,741-778 (2006). [2] Kassier, et al., Applied Physics B 109, 249-257 (2012). [3] Vogelgesang, et al., Nature Physics 14,184-190 (2018). [4] Epp et al., under review.

O 5.7 Mon 12:00 MA 041

Ultrafast Low-Energy Photoelectron Diffraction for the Study of Adsorbate-Surface Interactions with 100 fs Temporal Resolution — •Hermann Erk, Carl Jensen, Stephan Jauernik, Petra Hein, and Michael Bauer — Christian-Albrechts-Universität Kiel, Germany

Ultrafast Low-Energy Electron Diffraction (ULEED) is a promising tool for studying the structural dynamics of ordered adsorbate layers following photoexcitation with femtosecond light pulses. However, the temporal spread of the probing electron pulse due to Coulomb interaction while it propagates toward the sample surface substantially limits the time resolution of this technique so far [1].

In this contribution, we present an alternative experimental scheme based on time- and angle-resolved ARPES that uses the photoelectrons generated in the substrate and diffracted by the adsorbate overlayer on the way to the electron detector to monitor ultrafast adsorbate structural dynamics. For tin-phthalocyanine adsorbed on single-crystalline graphite, we show that the intrinsic time resolution of the photoemission experiment of 100 fs is preserved in the diffraction process by the adsorbate overlayer. On a few ps timescale, we identify a transient and distinct decrease in the photoelectron diffraction intensity that we attribute to an increase in the adsorbate superlattice temperature due to phononic coupling with the substrate. The interpretation is substantiated by a comparison with phonon-temperature transients for graphite simulated based on a three-temperature model.

[1] S. Vogelgesang, et al. Nature Physics 14, 184 (2018)

O 5.8 Mon 12:15 MA 041

Development and characterization of an ultrafast scanning electron microscope — •Paul H. Bittorf[1] and Nahid Talebi[1,2] — [1]Institute for Experimental and Applied Physics, Kiel University, Leibnizstraße 19, D-24118 Kiel, Germany — [2]Kiel Nano, Surface and Interface Science KiNSIS, Kiel University, Christian-Albrechts-Platz 4, D-24118 Kiel, Germany

A variety of processes are involved within the interaction of free-electron wave packets with light or matter, such as coherent and incoherent excitation of the investigated material. Understanding these interaction processes of pulsed electron beams with shaped light fields or nanostructured matter is crucial to achieve a favored shaping of electron wave packets, as well as exploring the dynamics of material excitations. Therefore, we developed an ultrafast scanning electron microscope (USEM) for exploring the dynamics at the nanoscale and at the femtosecond time regime. Our setup is based on the combination of a commercial SEM with an ultrafast laser system, where a laser-driven pulsed electron source is achieved via the photoemission process. In addition to the electron beam excitation, a time-delayed laser pulse is focused onto the sample to induce an optical near-field and implement a time-resolved pump-probe measurement. The interaction properties of electrons with matter are analyzed through the emitted light, either cathodoluminescence (CL) or photoluminescence. Here, we report on the technical aspects of the development and characterization of our USEM setup and present measurements of the photoemitted pulsed electron beams and a novel fiber-based CL detector.