O 34: Ultrafast Electron Dynamics at Surface and Interfaces III

Time: Tuesday 10:30–13:15 Location: TRE Phy

O 34.1 Tue 10:30 TRE Phy

Resolving Intervalley Exciton Couplings in Atomically Thin Semiconductors with Multidimensional Spectroscopy — •Lawson Lloyd[1,2], Ryan Wood[2], Fauzia Mujid[2], Siddhartha Sohoni[2], Karen Ji[2], Po-Chieh Ting[2], Jacob Higgins[2], Jiwoong Park[2], and Gregory Engel[2] — [1]Fritz-Haber-Institut, Berlin, Germany — [2]The University of Chicago, Chicago, IL, USA

Atomically thin transition metal dichalcogenides (TMDs) have emerged as promising candidates for next-generation optoelectronic applications. In particular, an optically addressable "valley" degree of freedom at the K/K' points can be used to store and readout information by exploiting the valley-dependent chiral optical selection rules. However, intervalley coupling after optical excitation leads to a loss of the valley polarization on ultrafast timescales. Identifying the microscopic mechanisms driving valley depolarization is therefore critical to advancing useful applications.

Here, leveraging multidimensional electronic spectroscopy, we track the exciton dynamics and couplings in large-area monolayer MoS_2 with femtosecond and valley resolution. We show that intervalley coupling between exciton states occurs both on the timescale of excitation (∼10 fs) and with minimal dependence on the excitation fluence, temperature, or sample grain size. These results shed light on the strong many-body interactions governing the femtosecond exciton dynamics in TMDs and the factors limiting the realization of novel technologies built around the valley degree of freedom.

O 34.2 Tue 10:45 TRE Phy

Electron dynamics after a spin- and valley-polarized electronic excitation in WS_2 — •Lasse Münster[1], Sarah Zajusch[1], Raul Perea-Causin[1], Samuel Brem[1], Katsumi Tanimura[1], Jens Güdde[1], Yaroslav Gerasimenko[2], Rupert Huber[2], Ermin Malic[1], Ulrich Höfer[1], and Robert Wallauer[1] — [1]Fachbereich Physik, Philipps-Universität Marburg, Germany — [2]Fachbereich Physik, Universität Regensburg, Germany

The excitonic landscape of monolayer transition metal dichalcogenides (TMDCs) comprises optically accessible bright excitons as well as spin- and momentum-forbidden dark excitonic states. We image the formation process of these states after optical excitation on an ultrashort timescale with time-resolved momentum microscopy.

We excite monolayer WS_2 resonantly to the A 1s exciton with circular polarized light. This excitation leads to a population, which is located purely within the K valley for one helicity and in the K' valley for the other helicity. In the case of an excitation at K, electron scatter to K' and Σ by spin-conserving processes within a few tens of femtoseconds. Furthermore, we observe the formation of spin-forbidden excitons in the K valley and electron scattering towards Σ'. Both of these processes involve a spin-flip and are significantly slower (50 - 100 fs).

O 34.3 Tue 11:00 TRE Phy

Influence of the substrate dielectric permittivity on ultrafast quasiparticles dynamics in WS_2 monolayers — •Subhadra Mohapatra[1,2], Lukas Gierster[1,2], Stefano Calati[1,2], Nicholas Michael Olsen[3], Quiyang Li[3], Xiaoyang Zhu[3], and Julia Stähler[1,2] — [1]Humboldt-Universität zu Berlin, Institut für Chemie — [2]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Abt. Physikalische Chemie, Berlin, Germany — [3]Columbia University

In our recent fluence-and photon energy-dependent studies [1,2] of quasiparticle dynamics in WS_2 monolayers on fused silica (FS) and Si-SiO_2 substrates, we observed that excitonic screening reduces the binding energy of the excitons, however the role of screening due to substrates and the quantitative nature of dynamic screening due to excitons remained inconclusive. On further investigation of such fluence-dependent quasiparticle dynamics studies using a higher dielectric permittivity of a sapphire substrate, we found that scattering rates, relaxation time constants, and band gap renormalization are not influenced by the dielectric permittivity. On the contrary, the dynamic screening parameter of the excitons is approximately two times higher in FS than Sapphire and Bohr radius is approximately 2.5 % higher in sapphire than for FS, which must be a direct consequence of the increased dielectric permittivity likely leading to more delocalized excitons.

References:

[1] Calati et al. PCCP 23(39) (2021).
[2] Calati et al. arXiv:2204.02125 (2022).

O 34.4 Tue 11:15 TRE Phy

Probing the ultrafast hole-transfer mechanism in a twisted TMD heterostructure — •Marcel Reutzel[1], Jan Philipp Bange[1], David Schmitt[1], Wiebke Bennecke[1], Giuseppe Meneghini[2], AbdulAziz AlMutairi[3], Daniel Steil[1], Sabine Steil[1], R. Thomas Weitz[1], Samuel Brem[2], G. S. Mathijs Jansen[1], Stephan Hofmann[3], Ermin Malic[2], and Stefan Mathias[1] — [1]I. Physikalisches Institut, Georg-August-Universität Göttingen, Göttingen, Germany — [2]Fachbereich Physik, Philipps-Universität, 35032 Marburg, Germany — [3]Department of Engineering, University of Cambridge, Cambridge CB3 0FA, U.K.

In type-II band aligned TMD heterostructures, long-lived interlayer excitons (ILX) can be formed via two different processes: Optically excited intralayer excitons either transfer the exciton's electron or hole component across the interface. Using momentum microscopy [Rev. Sci. Ins. 91, 063905 (2020)], we have shown that the transfer of the exciton's electron proceeds via exciton-phonon scattering and layer hybridized Σ-excitons [Schmitt *et al.*, Nature 608, 499 (2022)].

In this contribution, we show that the Coulomb correlation between the exciton's hole and electron can be used to probe the hole-transfer dynamics: The intralayer exciton's hole transfers from the VBM of MoS_2 into the VBM of WSe_2, which is imprinted onto the photoemission signal as an apparant upshift of the mean photoelectron energy. Our analysis provides new insights on the ultrafast hole-transfer mechanism in the WSe_2/MoS_2 heterostructure, and, more generally, on the photoemission signature of Coulomb correlated electron-hole pairs.

O 34.5 Tue 11:30 TRE Phy

Resolving momentum-dependent phonon buildup at a 1T-TiSe2 surface using diffuse scattering in ultrafast LEED — •Felix Kurtz[1], Tim Dauwe[1], Sergey Yalunin[1], Gero Storeck[2], Jan Gerrit Horstmann[1], Hannes Böckmann-Clemens[1], and Claus Ropers[1,2] — [1]Max Planck Institute for Multidisciplinary Sciences, Am Fassberg 11, D-37077 Göttingen — [2]4th Physical Institute, University of Göttingen, Friedrich-Hund-Platz 1, D-37077 Göttingen

Electron-phonon and phonon-phonon couplings govern the energy flow into and within the lattice following femtosecond laser excitation. The resulting transient phonon population can be directly deduced from momentum-resolved maps in ultrafast electron diffuse scattering [1]. Here, we employ ultrafast low-energy electron diffraction (ULEED) [2] to investigate structural dynamics and pathways for energy relaxation in the optically-excited charge-density-wave phase of 1T-$TiSe_2$. By analyzing the diffuse scattering background, we track the nonequilibrium evolution of the phonon population and observe strongly momentum-dependent rise times. Specifically, we find a rapid buildup of zone-boundary phonons within few picoseconds, followed by a substantially slower generation of low-energy zone-center acoustic phonons. Our findings are corroborated by *ab-initio* DFT calculations of phonon-phonon scattering rates. We believe that the presented approach is generally applicable to a variety of systems and provides fundamental insights into surface phonon dynamics and relaxation pathways.

[1] L. P. René de Cotret, *et al.*, Phys. Rev. B 100, 214115 (2019)
[2] G. Storeck, *et al.*, Structural Dynamics 7, 034304 (2020)

Topical Talk O 34.6 Tue 11:45 TRE Phy

Photoemission orbital tomography for excitons — •Peter Puschnig, Andreas Windischbacher, Melvin Hodžić, and Christian S. Kern — Institute of Physics, NAWI Graz, University of Graz, Austria

In photoemission orbital tomography (POT), the photoemission angular distribution (PAD) of oriented molecular layers is interpreted in terms of the Fourier transform of the initial molecular orbital from which the electron is emitted. Recently, it has been demonstrated that POT can in principle also be applied to optically excited states using a femtosecond pump-probe setup, which images the excited electron-hole pairs of a system, the excitons, on ultrafast time scales [1]. However, a rigorous and generally applicable connection between the measured PAD and the spatial structure of the excitons is lacking. By considering the expansion of the exciton wave function in the product basis of

valence and conduction state orbitals, as typically done when solving the electron-hole Bethe-Salpeter equation or Casida's equations in the framework of TDDFT, we demonstrate in this contribution that the PAD is given by the Fourier transform of a coherent sum of the electronic part of the exciton wave function. This relation, which is based on a plane wave final state, as well as the unexpected consequences of the hole for the measured kinetic energy spectrum an exciton is illustrated for a series of organic molecules in the gas phase for which the PAD is also simulated explicitly, and without resorting to a plane-wave final state, by means of a real-time, real-space TDDFT approach.

[1] Wallauer et al., Science 371, 1056-1059 (2021).

O 34.7 Tue 12:15 TRE Phy

Ultrafast lattice dynamics of Niobium diselenide ($2H$-$NbSe_2$) — •Victoria C. A. Taylor[1], Yoav William Windsor[1,2], Hyein Jung[1], and Ralph Ernstorfer[1,2] — [1]Fritz Haber Institute — [2]TU Berlin

Within the transition metal dichalcogenide family, $2H$-$NbSe_2$ is unusual as it exhibits metallic, charge density wave (CDW), and superconducting phases, evidencing strong electron-lattice interactions. Ultrafast techniques enable investigation of such interactions on femtosecond timescales, and several studies have investigated the ultrafast electronic response of $2H$-$NbSe_2$. However, none have directly probed the ultrafast response of the lattice. We present a femtosecond electron diffraction (FED) study on $2H$-$NbSe_2$ in its metallic phase, probing both coherent and incoherent phononic responses that result from electron-phonon and phonon-phonon scattering. From these data we explore these processes and the associated timescales.

O 34.8 Tue 12:30 TRE Phy

Non-equilibrium dynamics of bulk VSe2 — •Wibke Bronsch[1], Manuel Tuniz[2], Denny Puntel[2], Alessandro Giammarino[2], Fulvio Parmigiani[1,2], and Federico Cilento[1] — [1]Elettra-Sincrotrone Trieste, Trieste, Italy — [2]Università degli Studi di Trieste, Trieste, Italy

By means of time- and angle-resolved photoemission spectroscopy (tr-ARPES), we investigate the effect of optical excitation on the electronic properties of the transition metal dichalcogenide VSe2. The electronic band structure of VSe2 has recently been subject of investigation ranging from the bulk to the monolayer regime, in search for the manifestation of the opening of a band gap in its CDW phase [1,2]. However, at present, only a few studies on the effect of an ultrafast optical excitation are available [1,3]. In our contribution we present a study on the bulk material. By selecting the polarization of the probe pulses, tr-ARPES allows us to disentangle states with different orbital character, originating from the V and Se valence bands. Our tr-ARPES data show indication for a novel photoinduced state near the Fermi level, lasting for several picoseconds after photoexcitation.

[1] Biswas et al., Nano Letters 21, 1968-1975 (2021).
[2] Umemoto et al., Nano Research 12, 165-169 (2019).
[3] P. Majchrzak et al., Phys. Rev. B 103, L241108 (2021).

O 34.9 Tue 12:45 TRE Phy

Ultrafast charge separation and charge density wave suppression in monolayer $TiSe_2$ on graphite — •Sebastian Buchberger[1,2], Charlotte Sanders[3], Yu Zhang[3], Emma Springate[3], Paulina Majchrzak[4], Jill Miwa[4], Philip Hofmann[4], Søren Ulstrup[4], and Phil D. C. King[1] — [1]University of St Andrews, UK — [2]MPI for Chemical Physics of Solids, Dresden, Germany — [3]Central Laser Facility, UK — [4]Aarhus University, Denmark

The properties of two-dimensional materials and their heterostructures are often governed by the interplay of several coexisting interactions which can be difficult to disentangle in equilibrium. Time and angle resolved photoelectron spectroscopy (trARPES) is emerging as a powerful method to investigate such systems, allowing the observation of ultrafast processes such as the buildup of charge screening[1], optically induced bandgap renormalisation[2] and interlayer charge separation[3]. Here we present a trARPES study on monolayer $TiSe_2$ grown epitaxially on graphite. Monolayer $TiSe_2$ is a small indirect bandgap semiconductor that exhibits an unconventional charge density wave (CDW) in its ground state, whose origin and nature are still not fully established[4]. We study how the electronic structure evolves upon photoexcitation, tracking band shifts and spectral weight variations to disentangle charge separation, screening, and dynamical suppression of the CDW.

[1]T. Rohwer et al., Nature 471, 490-493 (2011), [2]S. Ulstrup et al., ACS Nano 10, 6315-6322 (2016), [3]S. Aeschlimann, Sci. Adv. 6, eaay0761 (2020), [4]P. Chen et al., Nat. Commun. 6:8943 (2015)

O 34.10 Tue 13:00 TRE Phy

Manipulation of the charge-density-wave in VTe2 by femtosecond light pulses — •Manuel Tuniz[1], Wibke Bronsch[2], Denny Puntel[1], Giovanni Di Santo[2], Luca Petaccia[2], Davide Soranzio[3], Davide Bidoggia[1], Maria Peressi[1], Fulvio Parmigiani[1,2], and Federico Cilento[2] — [1]Dipartimento di Fisica, Università degli Studi di Trieste, Italy — [2]Elettra - Sincrotrone Trieste S.C.p.A., Italy — [3]Institute for Quantum Electronics, ETH Zurich, 8093 Zurich, Switzerland

Comparing equilibrium and out-of-equilibrium angle-resolved photoemission spectroscopy (ARPES and tr-ARPES), we investigate the effect of an optical excitation on the electronic properties of the charge-density wave (CDW) system VTe2. Recently, the modification of the material's electronic structure triggered by CDW formation has been discussed because the strongly orbital-dependent changes may give rise to a topological change in specific bands [1]. In our contribution we show clear modifications to the electronic band structure of VTe2 induced by the emergence of the CDW phase. Moreover, our tr-ARPES experiments open the possibility to perturb the CDW phase and study the relaxation dynamics of this non-equilibrium state to the ground state of the system.

[1] Mitsuishi, N. et al. Nat Commun 11, 2466 (2020)