O 38: Ultrafast electron dynamics at surface and interfaces II

Time: Tuesday 10:30–12:30 Location: TRE/MATH

O 38.1 Tue 10:30 TRE/MATH

Ultrafast momentum microscopy of exciton dynamics at ZnO surfaces — •Hashima Marukara, Paul Werner, Jan Philipp Bange, G S Matthijs Jansen, Wiebke Bennecke, Junde Liu, and Stefan Mathias — Georg-August-Universität Göttingen, I. Physikalisches Institut, Germany

ZnO is a prototypical wide-band-gap material with long-lived excitons, making it a central platform for modern optoelectronic and catalytic technologies, especially for investigating exciton-related phenomena [1]. Excitons*bound electron*hole pairs that can exist in various forms near the surface, significantly shape the material*s optical and electronic response and play an essential role in interfacial energy-conversion processes [2]. However, studies that directly characterize these excitons, particularly if they are defect-mediated, localized or mobile, or predominantly Frenkel- or Wannier-type, remain limited.

Here we address these questions using time-resolved momentum microscopy to map the fingerprints of excitons in energy, momentum and time [3]. We present first experimental momentum-space maps of the ZnO(10-10) surface exciton states and track their real-time evolution following photoexcitation. We observe clear signatures of surface exciton formation, including their emergence in the unoccupied band structure and their subsequent relaxation dynamics on ultrafast timescales.

[1] Gierster *et al.*, Nat Commun 12, 978 (2021) [2] Deinert *et al.*, Phys. Rev. Lett. 113 057602 (2014) [3] Bennecke *et al.*, Nature Phys. (2025), https://doi.org/10.1038/s41567-025-03075-5

O 38.2 Tue 10:45 TRE/MATH

Investigating the light-induced metallization of ZnO through time-resolved photoemission spectroscopy — Gian Marco Pierantozzi[1], Federico Motti[1], Matteo Gatti[2], •Monika Schied[1], Alice Crosato[3], Thomas Sartori[3], Riccardo Cucini[1], Giorgio Rossi[1,4], Fausto Sirotti[2], and Giancarlo Panaccione[1] — [1]CNR – Istituto Officina dei Materiali (IOM), Trieste, Italy — [2]École polytechnique, Paris, France — [3]Università Ca' Foscari Venezia, Italy — [4]Università degli Studi di Milano, Italy

With a direct bandgap of ∼3.4 eV and a high exciton binding energy (∼60 meV), ZnO is ideal for optoelectronics and excitonic studies. Under optical excitation, the system shows ultrafast metallization with several relaxation timescales [1,2]. The relevance of defects in stabilizing the photoinduced metallic state is known, but many aspects of the processes remain unclear. Here we investigate the ultrafast dynamics of the optically populated conduction band and the Zn 3d core level with time- and angle-resolved photoemission spectroscopy after resonantly pumping, in order to disentangle the excitations of defect levels. Our experiments cover the whole metallization process in ZnO. We observe a fast rise in the conduction band population within 100 fs, a slower temperature-dependent rise, probably due to carrier transport and redistribution, a decay on the ns timescale and a long-lasting metastable state. All these dynamics occur also pumping below resonance, albeit to a lower extent, highlighting the role of the defect levels.

[1] L. Gierster et al. Nat. Comm. 12:978 (2021);
[2] L. Gierster et al., Far. Disc. 237, 58 (2022)

O 38.3 Tue 11:00 TRE/MATH

Ultrafast electron dynamics in charge-transfer insulators $LaFeO_3$(001) and $BiFeO_3$(001) — •Friederike Wührl, Antonia Rieche, Anne Oelschläger, Kathrin Dörr, and Wolf Widdra — MLU Halle-Wittenberg

The electron dynamics in strongly correlated systems, where electron-electron correlation leads to band gap opening, are highly unexplored. One example is the prototypical charge transfer insulator NiO, which shows a remarkable short lifetime of <10 fs at the conduction band minimum, relaxing in a many-body in-gap state, which couples to the antiferromagnetic spin system [1]. Here we are extending our time-resolved studies to the unoccupied states of epitaxial antiferromagnetic $LaFeO_3$(001) and ferroelectric $BiFeO_3$(001) thin films grown on $SrRuO_3/DyScO_3$(001) substrates. We use two-photon photoemission in a UV-UV pump-probe experiment, exciting electrons across the band gap, which opens between strongly hybridized oxygen 2p/Fe 3d and Fe minority states for both perovskites. We find in both sytems a broad state at ∼ 1.3 eV above E_F with a short lifetime of 20 fs, whereas the conduction band minimum shows a multiexponential decay with timescales in the femtosecond to picosecond range. The different decay channels show an energy dependent increase of lifetimes towards the Fermi level. Comparing $BiFeO_3$ and $LaFeO_3$, the decay channels differ solely in their amplitudes, with a higher probability for the fast decay channel in $LaFeO_3$.

[1] Gillmeister *et al.* Nat. Commun. **11**, 4095 (2020).

O 38.4 Tue 11:15 TRE/MATH

Ultrafast Formation and Annihilation of Strongly Bound Anisotropic Excitons in CrSBr — •Lawson T. Lloyd[1], Tommaso Pincelli[1,2], Mohamed A. Wahada[1], Alessandro De Vita[1,2], Ferdinand Menzel[3], Ksenia Mosina[4], Túlio H. L. G. Castro[1], Alexander Neef[1,2], Andreas V. Stier[3], Nathan P. Wilson[3], Zdeněk Sofer[4], Jonathan J. Finley[3], Martin Wolf[1], Laurenz Rettig[1], and Ralph Ernstorfer[1,2] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany — [2]Technische Universität Berlin, Berlin, Germany — [3]Technische Universität München, Garching, Germany — [4]University of Chemistry and Technology Prague, Prague, Czech Republic

Excitons dominate the optical response and determine the functionality of atomically thin semiconductors. In the van der Waals magnetic semiconductor CrSBr, strongly bound, anisotropic excitons couple to the underlying spin order, which could be leveraged in next-generation opto-spintronics. However, mechanisms of exciton formation, dissociation, and interaction with free carriers remain largely unexplored. Employing time- and angle-resolved photoemission spectroscopy, we probe the nature of the exciton in CrSBr by directly imaging the nonequilibrium electronic states, extracting fundamental properties of the excitonic wavefunction and revealing the driving mechanisms of exciton relaxation on ultrafast timescales. Notably, we observe an excitation-density-dependent interconversion between bound excitons and quasi-free carriers, indicating that many-body effects govern the excited state dynamics during the initial stages of relaxation.

O 38.5 Tue 11:30 TRE/MATH

Large relative delays for photoemission final states less than 1 eV apart — •Tillmann Schabbehard, Andreas Gebauer, Walter Enns, Luis Maschmann, and Walter Pfeiffer — Universität Bielefeld, Universitätsstr. 25, 33615 Bielefeld

The reconstruction of attosecond beating by interference of two-photon transitions (RABBITT) allows probing photoemission dynamics in atoms, molecules and solids. However, the interpretation of retrieved relative delays between different emission channels in particular for emission from solids is controversial. Here we employ spin-orbit delays in GaSe by RABBITT, i.e., relative delays for emission from the different initial states of the spin-orbit doublet, to investigate the relative photoelectron kinetics in emission channels, which differ less than 1 eV in their final state energy (E_{final}). Despite the small spin-orbit splitting for both Ga3d and Se3d the extraction of relative delays for the two spin-orbit doublets yields 180±53 as and 94±45 as. These unexpectedly large delays can neither be attributed to intraatomic delay mechanisms nor to kinetic propagation effects in the final state continuum. This challenges common concepts about photoelectron emission dynamics, which predict much smaller delays for E_{final} differences below 1 eV. Notably, the Se3d spin-orbit delay in GaSe agrees well with those observed in Bi_2Se_3 and WSe_2 indicating a general high variability of emission dynamics as function of the E_{final}. Predictions based on the one step model, which take into account transport via propagating and evanescent states match observations much better, while modelling based on propagation effects in the final state alone fail.

O 38.6 Tue 11:45 TRE/MATH

Lightwave-driven flat bands in a van der Waals crystal — •Changhua Bao[1], Vincent Eggers[1], Manuel Meierhofer[1], Jakob Helml[1], Lasse Münster[1], Suguru Ito[2], Leon Machtl[1], Sarah Zajusch[2], Giacomo Inzani[1], Ludwig Wittmann[1], Marlene Liebich[1], Robert Wallauer[2], Ulrich Höfer[1,2], and Rupert Huber[1] — [1]Department of Physics, University of Regensburg, Regensburg, Germany — [2]Department of Physics, Philipps-University of Marburg, Marburg, Germany

Flat bands lead to an extremely high density of states and suppressed

kinetic energy, thereby increasing electronic correlations and leading to intriguing light-matter interaction. Here, we reveal the band structure of the van der Waals crystal $NbOCl_2$ using photoelectron momentum microscopy. We directly map out an electronic band that is flat throughout the entire Brillouin zone and well isolated from both the conduction and remote valence bands. By utilizing subcycle photoemission spectroscopy [1,2], the dynamics of the flat band under atomically strong mid-infrared light fields are explored. Intriguingly, the carrier field of light induces long-lived collective modes, which are resolved with subcycle precision. These collective modes not only accelerate the electrons but also renormalize the energy of the flat band. We investigate the microscopic mechanisms of these dynamics, highlighting novel possibilities of flat bands in strong-field physics.

[1] Reimann *et al.,* Nature **562**, 396 (2018); [2] Ito *et al.,* Nature **616**, 696 (2023)

O 38.7 Tue 12:00 TRE/MATH

Spin-orbit entangled exciton formation in a 2D correlated semiconductor — •Tania Mukherjee[1,2], Lawson T. Lloyd[1], Alessandro De Vita[1,2], Martin Wolf[1], Laurenz Rettig[1], Ralph Ernstorfer[1,2], and Tommaso Pincelli[1,2] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany — [2]Technische Universität Berlin, Berlin, Germany

Excitons govern the magneto-optical properties in spin-ordered 2D materials. $NiPS_3$, a layered antiferromagnetic semiconductor, has recently gained significant attention because of its unconventional excitonic behaviour. Previous literature suggests that excitons are composed of bound electron and holes pairs in a highly spin and orbitally entangled Zhang-Rice-singlet state, resulting in strong coupling with the underlying magnetic ordering.However, the microscopic mechanism that governs the exciton formation remains elusive and unexplored. Using time- and angle-resolved photoemission spectroscopy, we map the electronic structure and the excited state dynamics of $NiPS_3$ across the magnetic phase transition and, for the first time, observe directly the long-lived exciton footprint below the Néel temperature. This paves the way for exploring similarly complex excitonic behavior in other layered correlated materials.

O 38.8 Tue 12:15 TRE/MATH

Understanding resonantly enhanced light-induced superconductivity in K3C60 — •Michael A. Sentef[1,2], Juan I. Aranzadi[1], Paul Fadler[1], and Joseph Tindall[3] — [1]Institute for Theoretical Physics, University of Bremen — [2]Max Planck Institute for the Structure and Dynamics of Matter, Hamburg — [3]Center for Computational Quantum Physics, The Flatiron Institute, New York

Light-induced superconducting-like responses have been observed in K3C60 when driving with mid-infrared laser pulses [1], with a pronounced 10 THz resonance reported more recently [2]. Here we address the microscopic origin of this resonance. Using numerical calculations on finite clusters for a realistic multi-orbital model of K3C60 we simulate the effect of a periodic drive on the strongly correlated system. We find a sharp enhancement of pairing correlations when the drive frequency is tuned close to 10 THz, in agreement with experiment. The microscopic origin of this enhancement and its potential implications for a light-induced hidden phase [3] will be discussed. Our results thus provide a microscopic explanation for the resonantly enhanced light-induced superconducting-like state in K3C60 and strengthen its interpretation as being closely connected to superconducting pairing rather than to purely optical or nonthermal population effects.

[1] Mitrano et al., Nature 530, 461-464 (2016). [2] Rowe et al., Nature Physics 19, 1821-1826 (2023). [3] Budden et al., Nature Physics 17, 611-618 (2021).