O 67: Ultrafast Electron Dynamics II

Time: Wednesday 15:00–17:45 Location: H11

O 67.1 Wed 15:00 H11

Polaron formation in NiO analysed by transient absorption spectroscopy — •Mahendra Kabbinahithlu[1], Björn Sothmann[1], Fred Hucht[1], Sergey Kovalenko[2], Tobias Lojewski[1], Nico Rothenbach[1], Katharina Ollefs[1], Heiko Wende[1], Uwe Bovensiepen[1], Julia Stähler[2], and Andrea Eschenlohr[1] — [1]Universität Duisburg-Essen, Fakultät für Physik und Center for Nanointegration (CENIDE), Lotharstraße 1, 47057 Duisburg, Germany — [2]Humboldt-Universität zu Berlin, Institut für Chemie, Brook-Taylor-Straße 2, 12489 Berlin, Germany

Polaron formation is the process by which free electrons in a material find a lower energy localized state by distorting their surrounding lattice. The polaron formation timescales and its fluence dependent dynamics in nickel oxide (NiO) from time-resolved optical absorption studies is discussed here.

NiO is pumped above the band gap with 3.98 eV energy photons, and is probed using a time-delayed supercontinuum. The time-resolved absorption spectrum shows negative ground state bleach and positive excited state absorption and in addition, a time-delayed and energy separated appearance of a positive feature at 3.35 eV that is discussed as the signature of polaron formation. With increasing fluence,the build-up time of this 3.35 eV signature decreases from 4 ps down to 1.1 ps, indicating the transition from isolated polarons to the formation of a polaron band. The spectrally distinct transition between low fluence and high fluence regimes is used to estimate the polaron size and the dynamics is modelled by coupled rate equations.

O 67.2 Wed 15:15 H11

Sharp Exciton Mott Transition in WS_2 and its Ultrafast Decay — •Subhadra Mohapatra[1], Lukas Gierster[1], Samuel Palato[1], Nicholas Michael Olsen[2], Xiaoyang Zhu[2], and Julia Stähler[1] — [1]Humboldt-Universität zu Berlin, Institut für Chemie — [2]Columbia University

The excitonic Mott transition (EMT) in transition metal dichalcogenides is reported to be either discontinuous [1] or continuous [2]. To resolve the ambiguity, here we study the optical response of WS_2 across the Mott density. Using a complex lineshape analysis, we separate the optical response in the photo-excited part of the sample from the unexcited regimes. In agreement with a continuous EMT, a gradual increase in the bleach of the sample-averaged absorption is observed. However, the lineshape analysis unveils a sharp discontinuity in the transient dynamics of the A and B exciton resonances above a critical photoexcitation density. This is attributed to plasma formation followed by band gap renormalization. The plasma decays with a laser fluence-independent time constant of 0.65 ps which is attributed to phase separation into excitonic and plasma regions, in agreement with the literature [3,4]. This work not only provides the first detailed experimental investigation of the EMT close to the critical limit but also highlights the role of spatial inhomogeneous charge carrier distribution for the widely used transient optical spectroscopies of 2D materials. References: [1] Bataller et al. Nano Lett 19(2) (2019). [2] Chernikov et al. Nat Photonics 9(7) (2015). [3] Steinhoff et al. Nat Commun 8 (1) (2017). [4] Koch et al. Physica status solidi (b) 238(3) (2003).

O 67.3 Wed 15:30 H11

Ultrafast formation of electron polarons in rutile TiO_2(110) — •xiang zhang, lukas gierster, and julia stähler — Humboldt-Universität zu Berlin, Institut für Chemie

The electron dynamics at TiO_2 surfaces are widely studied, mainly focusing on the defect-induced electronic states [1,2]. Additionally, recent calculations suggest that upon across-bandgap excitation, electron polarons form within 25 fs by trapping of photoexcited electrons inside the deformed lattice [3]. Using time-resolved photoelectron spectroscopy, we observe the instrument response-limited (<30 fs) rise of a photoinduced electron population at 0.3 eV below E_F. To elucidate whether this feature originates from polaron formation, experiments are conducted with different laser fluences, photon en- ergies and under varying surface conditions. Based on this, competing mechanism as exciton formation, surface photovoltage effects and band gap renormalization can be excluded. Our results suggest that photoexcitation of the rutile TiO_2(110) surface creates small sub-surface polarons within only 30 fs.

Reference:
[1] A. Argondizzo et al. J. Phys. Chem. C 120, 12959-12966(2016).
[2] Y. Zhang et al. J. Phys. Chem. Lett. 10, 5265-5270(2019).
[3] C. Gao et al. J. Phys. Chem. C 125, 27275-27282(2021).

O 67.4 Wed 15:45 H11

Time-resolved two-photon photoemission of antiferromagnetic $LaFeO_3$ — •Friederike Wühl, Antonia Rieche, Anne Oelschläger, Kathrin Dörr, and Wolf Widdra — MLU Halle-Wittenberg

Basic quantities as band gaps or the lifetime of electrons at the conduction band minimum are of strong interest for optoelectronic devives. In this context, strongly correlated oxides are rarely studied. One such example is 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].

We present time-resolved two-photon photoemission (2PPE) data on above band gap excitation of electrons in $LaFeO_3$, a charge-transfer insulator with antiferromagnetic order, which exhibits weak ferromagnetism through spin canting. The band gap opens between hybridized O 2p – Fe $3d_{\uparrow}$ and minority Fe 3d $t_{2g\downarrow}$ states. Thin films of 9 nm $LaFeO_3$ were prepared on $SrRuO_3/DyScO_3$ substrates using pulsed laser deposition. Low-energy electron diffraction confirmed the formation of single crystalline layers with a c(2x2) superstructure, indicative for the long-range magnetic ordering. In one-colour and two-colour UV-UV pump-probe experiments ($h\nu$ = 3.4 eV and 4.2 eV) we find three unoccupied states at 0.3, 1.3, 2.1 eV above E_F, the middle one being resonantly pumped from Fe $e_{g,\uparrow}$ states and exhibiting a short lifetime of 22 fs. At the conduction band minimum we observe a biexponential decay with lifetimes of 40 fs and 1.1 ps.

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

O 67.5 Wed 16:00 H11

Photo-induced charge-transfer renormalization in NiO — •T. Lojewski[1], D. Golez[2,3], K. Ollefs[1], L. Le Guyader[4], L. Kämmerer[1], N. Rothenbach[1], R. Y. Engel[5], P. S. Miedema[5], M. Beye[5,6], G. S. Chiuzbaian[7], R. Carley[4], R. Gort[4], B. E. Van Kuiken[4], G. Mercurio[4], J. Schlappa[4], A. Yaroslavtsev[4,8], A. Scherz[4], F. Döring[9], C. David[9], H. Wende[1], U. Bovensiepen[1,10], M. Eckstein[11], P. Werner[12], and A. Eschenlohr[1] — [1]Univ. Duisburg-Essen & CENIDE — [2]Jozef Stefan Inst. — [3]Univ. of Ljubljana — [4]European XFEL — [5]DESY — [6]Stockholm Univ. — [7]Sorbonne Univ. — [8]MAX IV Lab. — [9]PSI — [10]Univ. of Tokyo — [11]Univ. of Hamburg — [12]Univ. of Fribourg

For strongly correlated materials, like the charge transfer insulator NiO, the interplay between the interaction-based localization and the itinerant behaviour of electrons is essential in determining the electronic properties. In these materials, resonant photoexcitations result in convoluted dynamics involving dynamical screening induced band shifts, charge redistributions and *d-d* type excitations. By combining fs time-resolved X-ray absorption spectroscopy and dynamical mean-field theory, we disentangle the intertwined dynamics. We find long-lived redshifts of the Ni *L* and O *K* edges ($>$ 10 ps), arising from a combination of Hartree shifts and renormalization of local interactions. We also identify a short-lived Ni L_3 pre-edge feature ($<$ 1 ps) related to photo-induced *d-d* transitions [1]. Financial support by DFG through SFB 1242 is acknowledged. - [1] T. Lojewski et al., Phys. Rev. B, in press (ArXiv:2305.10145)

O 67.6 Wed 16:15 H11

Non-equilibrium carrier dynamics and band structure of graphene on 2D silicon — •Maria-Elisabeth Federl[1], Theresa Glaser[1], Niclas Tilgner[2], Thomas Seyller[2], and Isabella Gierz[1] — [1]University of Regensburg — [2]Technical University Chemnitz

Confinement heteroepitaxy, where novel 2D structures are stabilized at the interface between epitaxial graphene and SiC substrate, provides a pathway to engineer proximity-coupling between the massless carriers in graphene and the carriers in the underlying layer. If the latter is a Mott insulator, exotic electronic properties might emerge due to hybridization between itinerant and localized electrons. The Si-rich (3x3) structure on the surface of SiC(0001) was proposed to be Mott insu-

lating with a bandgap of 1eV [1]. We used confinement heteroepitaxy to prepare a graphene monolayer on top of this putative 2D Mott insulator [2] and searched for indications of interlayer hybridization using time- and angle-resolved photoemission spectroscopy (trARPES). Our findings are consistent with the occurrence of ultrafast charge transfer between graphene and the Si-rich surface structure that we attribute to interlayer hybridization in agreement with predictions from density functional theory [2].

[1] Surf. Sci. 445, 109 (2000)
[2] Phys. Rev. B 94, 245421 (2016)

O 67.7 Wed 16:30 H11

Direct view on ultrafast charge transfer between C60 molecules and graphene — •Michael Herb, Maria-Elisabeth Federl, and Isabella Gierz — University of Regensburg

Interfacing 0D molecules with 2D materials holds great potential for various applications in materials science, electronics, and nanotechnology. For example, C60/graphene hybrids have been proposed to serve as supercapacitors or sensitive UV-visible photodetectors. We use time- and angle-resolved photoemission spectroscopy (trARPES) to investigate the interfacial non-equilibrium charge carrier dynamics of a sub-monolayer C60 film deposited on single-layer graphene. Upon excitation below the HOMO-LUMO gap of C60, we observe a shift of the graphene Dirac cone towards higher binding energies and a concurrent shift of the C60 molecular levels towards lower binding energies. These shifts indicate a photoinduced hole doping of graphene that persists for several picoseconds. Based on these findings we propose a possible microscopic pathway for ultrafast charge transfer across the C60-graphene interface.

O 67.8 Wed 16:45 H11

Light-induced hidden state studied by ultrafast angle-resolved photoemission spectroscopy — •Junde Liu[1], Pei Liu[3], Liu Yang[3], Sung-Hoon Lee[4], Mojun Pan[2], Famin Chen[2], Jierui Huang[2], Bei Jiang[2], Mingzhe Hu[2], Yuchong Zhang[2], Zhaoyang Xie[3], Gang Wang[3], Mengxue Guan[3], Wei Jiang[3], Huaixin Yang[2], Jianqi Li[2], Chenxia Yun[2], Zhiwei Wang[3], Sheng Meng[2], Yugui Yao[3], Tian Qian[2], and Xun Shi[3] — [1]University of Göttingen, I. Physikalisches Institut, Germany — [2]Institute of Physics, Chinese Academy of Sciences, Beijing, China — [3]Beijing Institute of Technology, Beijing, China — [4]Kyung Hee University, Yongin, Republic of Korea

The non-volatile and ultrafast optical manipulation of material properties offers profound insights into light-matter interactions and holds great potential for optoelectronic applications. However, the discovery of such transitions is often serendipitous, and their practical implementation remains limited, underscoring the need for systematic investigation. In this talk, I will focus on laser-induced nonvolatile phase transitions in transition metal dichalcogenides (TMDs), highlighting the critical role of interlayer order in the formation of hidden states. By employing ultrafast laser excitations (single-pulse writing, pulse-train erasing and pulse-pair control), systematic angle-resolved photoemission spectroscopy (ARPES) characterizations, and comparative density functional theory (DFT) calculations, I aim to unravel the mechanisms that form and stabilize these hidden states, paving the way for novel methods to optically control low-dimensional materials.

O 67.9 Wed 17:00 H11

A machine-learning approach to understanding ultrafast carrier dynamics in the 3D Brillouin zone of $PtBi_2$ — Paulina Majchrzak[1], Charlotte Sanders[2], Yu Zhang[2], Andrii Kuibarov[3], Oleksandr Suvorov[3], Tami Meyer[1], Gesa Siemann[1], Emma Springate[2], Iryna Kovalchuk[3,4], Saicharan Aswartham[3], Grigory Shipunov[3], Bernd Büchner[3], Sergey Borisenko[3], and •Philip Hofmann[1] — [1]Department of Physics and Astronomy, Aarhus University, DK — [2]Central Laser Facility, Harwell, UK — [3]IFW Dresden, Germany — [4]Kyiv Academic University, UA

We examine the electron dynamics of the type-I Weyl semimetal $PtBi_2$ by time- and angle-resolved photoemission spectroscopy. By varying the probe photon energy over a wide range, we are able to explore differences throughout the three-dimensional Brillouin zone. For these experiments, the photoemission intensity is measured as a function of emission angle, electron kinetic energy, time delay and probe photon energy. In order to discover trends in this multi-dimensional data set, we apply k-means, an unsupervised machine learning technique. This reveals k_z-dependent differences in dynamics–in particular, we observe dynamics that are faster in the parts of the Brillouin zone that host most of the bulk Fermi surface than in parts close to the Weyl points.

O 67.10 Wed 17:15 H11

Tracing thermal and athermal electrons in laser-excited metals — •Markus Uehlein[1], Henry Snowden[2], Christopher Seibel[1], Tobias Held[1], Sebastian T Weber[1], Reinhard J Maurer[2,3], and Baerbel Rethfeld[1] — [1]Department of Physics and Research Center OPTIMAS, RPTU Kaiserslautern-Landau, Germany — [2]Department of Chemistry, University of Warwick, United Kingdom — [3]Department of Physics, University of Warwick, United Kingdom

Understanding the energy- and time-resolved electronic properties in metals at the initial athermal stage after femtosecond laser irradiation is of fundamental importance for many applications in surface science. In the time domain, where no electron temperature is defined, the non-equilibrium dynamics are usually described with microscopic methods, such as the Boltzmann equation. However, especially when investigating ballistic transport processes, a description with full Boltzmann collision integrals [1] is challenging due to the numerical effort.

We present a model that describes the athermal carriers efficiently. To that end, we consider thermal and athermal electrons separately. We use a temperature-based approach for the thermal electrons and the phonons, while we trace the distribution of the athermal electrons explicitly. Such a separation allows to conserve particles and energy even when a relaxation time approach is applied. We show the energy-resolved dynamics and find good agreement with time-resolved two-photon photoemission spectroscopy measurements [2].

[1] B. Y. Mueller and B. Rethfeld; Phys. Rev. B **87**, 035139 (2013)
[2] F. Kühne *et al.*; Phys. Rev. Res. **4**, 033239 (2022)

O 67.11 Wed 17:30 H11

Influence of carbon buffer layer on non-equilibrium carrier dynamics of epitaxial graphene on SiC(0001) — •Johannes Gradl[1], Leornard Weigl[1], Neeraj Mishra[2,3], Stiven Forti[2], Camilla Coletti[2,3], and Isabella Gierz[1] — [1]University of Regensburg, Germany — [2]Istituto Italiano di Tecnologia, Pisa, Italy — [3]Istituto Italiano di Tecnologia, Genova, Italy

The carbon buffer layer resting at the interface between epitaxial graphene and SiC(0001) substrate is believed to be electronically dead with two non-dispersing bands located well below the Fermi level [1]. We use time- and angle-resolved photoemission spectroscopy (trARPES) to show that this picture breaks down away from thermal equilibrium and on ultrafast time scales. We find that photo-doping of the graphene - buffer layer heterostructure increases the carrier concentration inside the Dirac cone. Supported by pump fluence and pump wavelength dependent measurements we attribute this transient charging to direct electronic transitions between the buffer layer and the graphene layer and subsequent relaxation of the non-equilibrium charge carrier distribution. [1] Phys. Rev. B 77, 155303 (2008)