HL 54: Ultra-fast Phenomena II

Time: Thursday 15:00–17:15 Location: H17

HL 54.1 Thu 15:00 H17

Effect of the acceptor strength on intermolecular conical intersection dynamics in aggregates of quadrupolar dyes — •Katrin Winte[1], Somayeh Souri[1], Daniel Lünemann[1], Teresa Kraus[2], Elena Mena-Osteritz[2], Peter Bäuerle[2], Sergei Tretiak[3], Antonietta De Sio[1], and Christoph Lienau[1] — [1]Oldenburg University, Germany — [2]Ulm University, Germany — [3]Los Alamos National Laboratory, USA

Aggregated films of quadrupolar acceptor-donor-acceptor (A-D-A) molecules have emerged as promising materials for organic photovoltaics. The optoelectronic properties in the molecule are governed by an interplay between electronic and vibronic couplings in the molecule. In aggregated films, we have uncovered the existence of intermolecular conical intersections (CoIn) occurring on timescales of less than 50 fs, which funnel energy from the photoexcited bright state into the lower-lying dark electronic state of the aggregate [1]. This raises the question whether the quantum dynamics of the aggregates can be altered by modifying the intramolecular charge transfer character of the A-D-A monomers. Chemical intuition suggests that increasing the acceptor strength might accelerate charge transfer and with it the passage through the CoIn. To explore this hypothesis, we synthesize thin films of A-D-A molecules with varying acceptor strengths and study their ultrafast dynamics by femtosecond time-resolved spectroscopy. Our results show only minimal effects on the CoIn dynamics, suggesting that intermolecular vibronic couplings play the dominant role in the quantum dynamics. [1] A. De Sio et al., Nature Nano 16, 63 (2021).

HL 54.2 Thu 15:15 H17

Ultrafast decay of spin-polarization of semiconductor holes measured by attosecond transient absorption spectroscopy — •Lauren Drescher[1,2], Kylie Gannan[1], and Stephen Leone[1] — [1]Department of Chemistry, University of California, Berkeley, California 94720, USA — [2]Max-Born-Institut, Max-Born-Str. 2A, 12489 Berlin, Germany

Carrier excitation by light can lead to spin-polarization in semiconductor conduction and valence bands. While the dynamics of the spin-polarization of conduction band electrons are often well characterized, comparably little is known about the counterpart polarization of valence band holes, due to its much shorter lifetime.

Here we introduce circular dichroic attosecond transient absorption spectroscopy which allows to follow the spin-polarization of materials on attosecond timescales through coupling to the angular momentum of light. Corroborated by real-time time-dependent density functional theory calculations, our experimental results reveal the few femtosecond decay of semiconductor hole spin-polarization in germanium. Our method opens the door to measure spin dynamics in non-magnetic materials with extreme temporal resolution, high spectral resolution and core-level specificity.

HL 54.3 Thu 15:30 H17

Picosecond Femtojoule Resistive Switching in Nanoscale VO2 Memristors — •Sebastian Schmid[1,2], Laszlo Posa[1,3], Timea Török[1,3], Botond Santa[1,4], Zsigmond Pollner[1], Györgi Molnar[3], Yannik Horst[5], Janos Volk[3], Juerg Leuthold[5], Andras Halbritter[1,4], and Miklos Csontos[5] — [1]Department of Physics, Institute of Physics, Budapest University of Technology and Economics, H-1111 Budapest, Hungary — [2]Experimental Physics V, Center for Electronic Correlations and Magnetism, University of Augsburg, Augsburg 86159, Germany — [3]Institute of Technical Physics and Materials Science, HUN-REN Centre for Energy Research, 1121 Budapest, Hungary — [4]HUN-REN-BME Condensed Matter Research Group, H-1111 Budapest, Hungary — [5]Institute of Electromagnetic Fields, ETH Zurich, 8092 Zurich, Switzerland

The dynamics of the Mott transition in correlated electron oxides could provide a sustainable alternative to the von Neumann computation by exploiting device-level functional complexity at low energy consumption. We fabricated nanoscale VO2 devices and tested them in our picosecond timeresolution, real-time resistive switching experiments, using 20 ps short and <1.7 V amplitude voltage pulses. There we observed tunable resistance states from insulator-metal transitions with down to 15 ps incubation times and switching energies starting from a few femtojoule. These orders of magnitude improvements from other memristive devices open up new possibilities for neuromorphic computing applications, outperforming the human brain at size and speed, with competitive energy consumption.

HL 54.4 Thu 15:45 H17

Agreement between Theoretically Predicted and Measured Bragg Peak Decay in Bismuth Following Femtosecond Laser Excitation — •Bernd Bauerhenne[1], Jimiben Patel[1], Sahar Bakhshi[1], Sascha Epp[2], and Martin Garcia[1] — [1]Institute of Physics, University of Kassel, Heinrich-Plett-Straße 40, D34132 Kassel, Germany — [2]Max-Planck-Institut für Struktur und Dynamik der Materie, 3 Luruper Chaussee 149, 22761 Hamburg, Germany

We investigated the time-resolved Bragg peak decay in bismuth films, 30 nm and 50 nm thick, following excitation with femtosecond (fs) laser pulses. These measurements were conducted using x-ray pulses sourced from a free-electron laser. To explain the observed Bragg peak decay, we developed an electronic temperature (Te) dependent interatomic potential for bismuth, generated using forces and energies from ab initio molecular dynamics (MD) simulations at elevated Te levels. Additionally, we computed the optical properties and the Te-dependent electron-phonon coupling constant for bismuth using ab initio methods. Employing these calculated quantities, we conducted MD simulations on similarly laser-excited antimony films, 30 nm and 50 nm thick. The comparison between our theoretical predictions and experimental measurements of Bragg peak decay exhibited an agreement, affirming the accuracy of our model. This model effectively incorporates the fs-laser induced modifications of the potential energy surface and the dynamic influences of electron-phonon coupling, providing a robust framework for understanding laser-material interactions in ultrafast processes.

15 min. break

HL 54.5 Thu 16:15 H17

Ultrafast Time-Domain Spectroscopy Reveals Coherent Vibronic Couplings Upon Electronic Excitation in Crystalline Organic Thin Films — •Naby Hadilou[1], Somayeh Souri[1], Daniel Timmer[1], Daniel C. Lünemann[1], Katrin Winte[1], Antonietta De Sio[1], Martin Esmann[1], Sebastian Anhäuser[2], Michele Guerrini[1], Ana M. Valencia[1], Caterina Cocchi[1], Gregor Witte[2], and Christoph Lienau[1] — [1]Oldenburg university, Germany — [2]Philipps-Universität Marburg, Germany

Coherent coupling between electronic excitations and molecular vibrations significantly influences the optical and charge transport properties of organic semiconductors and may have profound effect on technologically relevant processes such as excitons fission. Highly ordered crystalline films are ideal for probing such couplings since they enable studies of individual domain. Here, we report first polarization-resolved pump-probe experiments probing the ultrafast dynamics of crystalline perfluoropentacene thin films grown on different substrates with 10-fs time resolution. Coherent oscillations in the spectra reveal vibronic couplings to a high-frequency, 25-fs, in-plane deformation mode that is insensitive to the optical polarization, while the coupling to a lower-frequency, 85-fs, out-of-plane ring bending mode depends significantly on the crystalline and molecular orientation. Raman spectra confirm this interpretation and highlight the dominance of solid-state effects on vibronic couplings. Our results represent a first step toward uncovering the role of anisotropic vibronic couplings for singlet fission processes in crystalline molecular thin films.

HL 54.6 Thu 16:30 H17

Attosecond light-driven charge injection in germanium — •Giacomo Inzani[1,9], Lyudmyla Adamska[2,3], Amir Eskandarias[4], Nicola Di Palo[1], Gian Luca Dolso[1], Bruno Moio[1], Luciano Jacopo D'Onofrio[4,5], Alessio Lamperti[6], Alessandro Molle[6], Rocio Borrego-Varillas[7], Mauro Nisoli[1,7], Stefano Pittalis[2], Carlo Andrea Rozzi[2], Adolfo Avella[4,5,8], and Matteo Lucchini[1,7] — [1]Dept. of Physics, Politecnico di Milano, Italy — [2]CNR - Istituto Nanoscienze, Modena, Italy — [3]Dept. of Physics, Mathematics and Informatics, University of Modena & Reggio Emilia, Italy — [4]Dip. di Fisica, Università degli Studi di Salerno, Italy — [5]CNR - SPIN, UoS di Salerno, Italy — [6]CNR - IMM, Unit of Agrate Brianza, Italy — [7]IFN - CNR, Milano, Italy — [8]Unità CNISM di

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The injection of charges from the valence to the conduction band of a semiconductor induced by an ultrashort pulse can tailor its electro-optical properties. This process typically occurs on time scales shorter than the laser period - for visible light, in the order of one femtosecond. Despite its relevance, few experiments studied the charge excitation process with attosecond temporal resolution. In this work, we combine attosecond transient reflection spectroscopy measurements to a dual cutting-edge theoretical approach, demonstrating that photoexcitation in Ge cannot be ascribed to a single physical mechanism. The interplay of multi-photon absorption, light-induced band dressing, and intra-band motion is crucial for determining the overall charge injection.

HL 54.7 Thu 16:45 H17

Dephasing Effects in High-Harmonic Generation from Solids — •Francisco Navarrete and Dieter Bauer — Institut für Physik, Universität Rostock, 18051 Rostock, Deutschland

In the calculation of high-order harmonic generation (HHG) in solids, introducing a dephasing time is often crucial for accurately reproducing the distinct spectral peaks observed experimentally in the first plateau region [1,2]. In this contribution, we present analytical and numerical studies on the non-integer contributions to the interband HHG spectrum and investigate how dephasing affects not only the spectral structure but also the amplitude and wavelength dependence of the harmonics. Using a simplified two-band model, we numerically solve and also approximate the semiconductor Bloch equations via a saddle-point method [3] to elucidate these effects. Our findings provide closed analytical expressions for these contributions, offering insights into the interplay between dephasing and electron dynamics in solids and the mechanisms shaping the HHG spectrum.

References:
[1] Vampa et al., Phys. Rev. Lett. 113, 073901 (2014)
[2] Cavaletto et al., Nat. Rev. Phys. (2024) (accepted)
[3] Navarrete et al., Phys. Rev. A 100, 033405 (2019)

HL 54.8 Thu 17:00 H17

Ultrafast Dynamics of Vanadium Dioxide Phase Transformation — •Liuyue Yang, Daniel Sandner, and Hristo Igle — Laser and X-Ray Physics E11, TUM School of Natural Sciences, TUM, James-Franck-Str. 1 85748 Garching, Germany

Vanadium dioxide has been discovered to have a metal-to-insulator transformation (MIT) at 68 °C. To investigate the dynamics of MIT, we use the pump-probe experiment to obtain the reflective transient mid-infared spectrum of VO2. In the photoinduced VO2 MIT, the phase transformation can occur at high pump power. The reflectivity of VO2 increases significantly at the metal state. At high temperature, the phase transformation occurs at lower pump power. Moreover, we observed a reflectivity oscillation in wavelength around the zeropoint of pump delay. The oscillation frequency decreases rapidly with the delay time