

Symposium Electron-driven processes: Atomic-scale insights from theory and experiment (SYED)

jointly organized by
the Surface Science Division (O),
the Chemical and Polymer Physics Division (CPP),
the Thin Films Division (DS),
the Semiconductor Physics Division (HL),
the Magnetism Division (MA), and
the Metal and Material Physics Division (MM)

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As traditional approaches for the conversion of light, electrical, and chemical energy are reaching their natural performance boundaries, future advancement demands fundamental understanding of the intricate interplay between light, electronic, and atomic degrees of freedom in materials. Recent progress in (nonlinear) spectroscopic experimental characterisation of ultrafast quasiparticle dynamics, in particular electron-phonon interactions, together with a drastic boost in our ability to theoretically describe and simulate electron-nuclear coupling at the femtosecond-nanometer scale have given rise to new applications of ultrafast science in plasmonics, organic and hybrid electronics, nanooptics, and electron-driven processes in so-called quantum materials. This symposium aims to provide a cross-disciplinary forum, bringing together experimentalists and theoreticians to assess the progress and future challenges in ultrafast electron and nuclear dynamics in materials.

Overview of Invited Talks and Sessions

(Lecture hall HSZ 01)

Invited Talks

SYED 1.1	Thu	9:30–10:00	HSZ 01	Ultrafast electron dynamics at laser-irradiated surfaces — ●BAERBEL RETHFELD
SYED 1.2	Thu	10:00–10:30	HSZ 01	Unraveling Momentum-Dependent Electron-Phonon Coupling and its Role in the Origin of Charge Density Wave Phases — ●BRADLEY SIWICK, MARTIN OTTO, JAN-HENDRIK POHLS, LAURENT RENE DE COTRET, MARK SUTTON
SYED 1.3	Thu	10:30–11:00	HSZ 01	Light MATTERs!!! — ●HRVOJE PETEK, ANDI LI, ZEHUA WANG, MARCEL REUTZEL
SYED 1.4	Thu	11:15–11:45	HSZ 01	Quantum localization and delocalization of charge carriers in molecular organic crystals — ●JOCHEN BLUMBERGER
SYED 1.5	Thu	11:45–12:15	HSZ 01	Single-Atom Catalysis (SAC): How Structure Influences Reactivity — ●GARETH PARKINSON

Sessions

SYED 1.1–1.5	Thu	9:30–12:15	HSZ 01	Symposium Electron-driven processes
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SYED 1: Symposium Electron-driven processes

Time: Thursday 9:30–12:15

Location: HSZ 01

Invited Talk

SYED 1.1 Thu 9:30 HSZ 01

Ultrafast electron dynamics at laser-irradiated surfaces — ●BAERBEL RETHFELD — Department of Physics and Research Center OPTIMAS, Technische Universität Kaiserslautern, Erwin-Schroedinger-Strasse 46, 67663 Kaiserslautern, Germany

When a femtosecond laser pulse of moderate intensity irradiates a solid surface, the energy is mainly absorbed by the electrons within the crystal. We calculate numerically the transient distribution function of laser-excited electrons with help of full Boltzmann collision integrals. Based on their density of states, we can trace different materials' response to an ultrafast stimulus of photonic excitation, as well as the interplay of electron-electron and electron-phonon interaction. Our results show that the initial characteristic signatures of the energy absorption are thermalized on a timescale of a few femtoseconds up to tens of femtoseconds. However, the cooling of the heated electron system by the cold phonons induces a further nonequilibrium, persisting on picosecond timescales [1].

We present possible consequences of the electron-phonon interaction on the energy distribution of hot electrons in dependence on material properties like band gap or particular features of the density of states.

[1] S.T. Weber and B. Rethfeld, Phys. Rev. B **99**, 174314 (2019)

Invited Talk

SYED 1.2 Thu 10:00 HSZ 01

Unraveling Momentum-Dependent Electron-Phonon Coupling and its Role in the Origin of Charge Density Wave Phases — ●BRADLEY SIWICK^{1,2}, MARTIN OTTO¹, JAN-HENDRIK POHLS¹, LAURENT RENE DE COTRET¹, and MARK SUTTON¹ — ¹McGill University, Department of Physics, Center for the Physics of Materials, Montreal, Canada — ²McGill University, Department of Chemistry, Montreal, Canada

The nature of the couplings within and between lattice and charge degrees of freedom is central to condensed matter and materials physics. These interactions are essential to phenomena as diverse as superconductivity, charge density waves and carrier mobility in semiconductors and metals. Despite their fundamental role, detailed momentum-dependent information on the strength of electron-phonon coupling (EPC) and phonon-phonon coupling (PPC) across the entire Brillouin zone has proved elusive. This talk will describe a new technique, ultrafast electron diffuse scattering (UEDS), which provides such information. Specific applications of UEDS to 2D materials including graphite, TiSe₂ and TaSe₂ will be presented. The data demonstrate that UEDS patterns can separate the influence of the electronic susceptibility from the inelastic exchange of energy between the electron and phonon systems due to the technique's profound sensitivity to photoinduced changes to the phonon system. In TiSe₂, this confirms key role of excitonic correlations to the phonon softening and CDW mechanism.

Invited Talk

SYED 1.3 Thu 10:30 HSZ 01

Light MATTERS!!! — ●HRVOJE PETEK¹, ANDI LI¹, ZEHUA WANG¹, and MARCEL REUTZEL^{1,2} — ¹Department of Physics and Astronomy, University of Pittsburgh, Pittsburgh, Pennsylvania 15260, USA — ²Present address: I. Physikalisches Institut, Georg-August-Universität Göttingen, Göttingen, Germany

Light interacting with solid-state matter under perturbative conditions excites primarily electric dipole transitions between k-vector dependent eigenstates of the periodic lattice potentials. A time-periodic light potential, however, can modify the electronic band structure of a solid through nonperturbative interactions entangling the light-matter interaction, and opening a route to tailor material properties with light at will. I will describe two examples: 1) It will happen at zero field strength when the complex dielectric response function $\text{Re}[\epsilon(\omega)] \sim 0$ and $\text{Im}[\epsilon(\omega)]$ is small. This happens for ionic solids above the longitudinal optical phonon frequency and for metals where the $-\epsilon_\infty(\omega) \sim \epsilon_{\text{Drude}}(\omega)$, or near the interband absorption threshold. This epsilon near-zero (ENZ) response coincides with the excitation of collective ion or electron responses, i.e., in case of metals, the bulk plasmon excitation. I will describe the ENZ response of single crystalline, low-index surfaces

of Ag, as measured by ultrafast multiphoton photoemission (mPP) spectroscopy. This bulk plasmonic response of Ag is fundamentally responsible for all plasmonic responses of silver, and yet the observed mPP spectra fundamentally contradict significant aspects that we expect from theory. 2) I will also report the high field response of Cu(111) surface where light is sufficiently strong to dress the electronic bands through Floquet engineering and Stark shift effects. The optical dressing opens the way to modify quasiparticles in solids at will, where, for example, electrons can be transformed into holes on subfemtosecond time scale. Our studies inform how light, electronic, and atomic degrees of freedom in solids can overcome their natural performance boundaries.

15 min. break**Invited Talk**

SYED 1.4 Thu 11:15 HSZ 01

Quantum localization and delocalization of charge carriers in molecular organic crystals — ●JOCHEN BLUMBERGER — University College London, Gower Street, London, WC1E 6BT, UK

Charge carrier transport (CT) in organic semiconducting materials is at the heart of many exciting and revolutionising technologies. Yet, our fundamental understanding of CT in the active layers of these devices that could rationalise experimental observations and guide further advances in the field is still rather limited. Charge carriers in organic semiconductors (OSs) are often described by one of two extremes: by a travelling wave propagating through the material or by a particle hopping from one molecular fragment to the next. Here we show that neither of these simplified pictures applies. Solving the time-dependent electronic Schrödinger equation coupled to nuclear motion for eight different organic molecular crystals, we find that the excess charge carrier forms a polaron delocalized over 10-20 molecules in the most conductive crystals.[1] The polaron propagates through the material by diffusive jumps over several lattice spacings (nanometers) at a time during which it expands to about twice its size. Computed localization lengths and charge mobility are in excellent agreement with experimental estimates, where available, and exhibit strong positive correlation in agreement with the recently proposed transient localization theory.

[1] S. Giannini, A. Carof, M. Ellis, H. Yang, O. G. Zigos, S. Ghosh, and J. Blumberger, Nature Comm., vol. 10, p. 3843, 2019.

Invited Talk

SYED 1.5 Thu 11:45 HSZ 01

Single-Atom Catalysis (SAC): How Structure Influences Reactivity — ●GARETH PARKINSON — TU Wien, Vienna, Austria

The field of "single-atom" catalysis emerged as the ultimate limit of attempts to minimize the amount of precious metal used in heterogeneous catalysis. There are many reports of active catalysts for important heterogeneous, electrochemical and photocatalytic reactions, but it has become clear that metal adatoms behave very differently to supported nano-particles. This is because they form chemical bonds with the support and are often charged. Thus, SAC systems resemble coordination complexes used in homogeneous catalysis, and there is growing excitement that SAC could achieve similar levels of selectivity, and even "hetero-genize" problematic reactions currently performed in solution. Homogeneous catalysts are designed for purpose based on well-understood structure-function relationships, while in SAC, the complexity of real systems means that the structure of the active site is difficult to determine, never mind design. I will describe how we are using Fe₃O₄(001) as a precisely-defined model support to unravel fundamental mechanisms in SAC. First, I will describe systematic studies of CO adsorption across several metals (Cu, Ag, Au, Ni, Pt, Ir, Rh) and show that adsorbate-induced structural changes affect the stability of the catalyst and the desorption energy. Then I will describe how the local structure of Ir₁/ and Rh₁/Fe₃O₄(001) SACs change with preparation, and how CO adsorption stabilizes the system through square-planar and octahedral binding environments reminiscent of Ir₁ and Rh₁ based metalorganic complexes.