Ultrafast Dynamics in Solids, at Surfaces and Interfaces

Charge transfer, chemical reactions, photovoltaic processes or phase transitions are often triggered by an initial primary excitation that induces subsequent dynamics starting on an ultrafast timescale, i.e., attoseconds (10^{-18}) to femtoseconds (10^{-15}). In our research we apply such controlled primary excitations to study the impact of these excitations on dynamical mechanisms in materials, which are driven by non-equilibrium electron, spin, and lattice dynamics. A microscopic understanding of these ultrafast physics allows us to gain insight into the fundamental mechanisms that determine electronic, magnetic, and structural changes in materials. Our tools comprise time-resolved photoemission spectroscopy and magneto-optical techniques with pulsed laser sources, and in particular the application of photons in the extreme ultraviolet from high-harmonic generation table-top light sources.

Energy conversion in correlated electron materials

In materials with strong correlations, the interactions of spin, charge, and lattice determine the path of energy conversion after optical excitation. Depending on the dominant interaction, the deposited energy is directed into different forms of work inducing electronic, magnetic, and structural changes. Often, however, the main interaction that would be responsible for the pathway of energy flow after an excitation is hard to determine in thermal equilibrium. Here, ultrafast time-resolved spectroscopies are a powerful way to overcome this problem and to investigate non-equilibrium energy flow in correlated materials. In particular, time-resolved photoemission techniques are well suited, since they can follow in a direct manner the optically induced redistribution of charge carriers. Hence, ultrafast time-resolved mapping of the electrons' energies, spins, and momenta after a strong optical excitation sheds light on band-structure formation, its relaxation to equilibrium, and the pathways of energy flow that are determined by the material's spin-charge-lattice interactions.

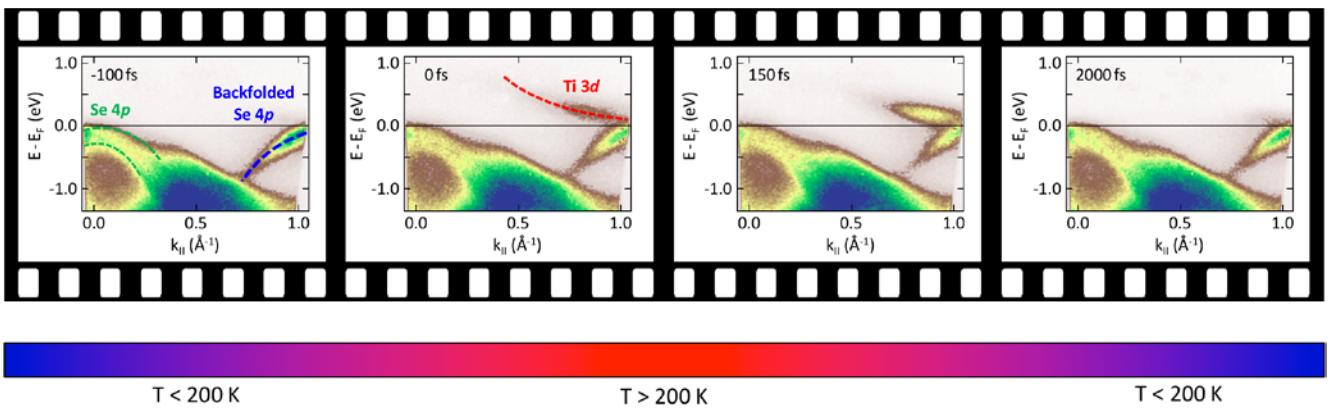
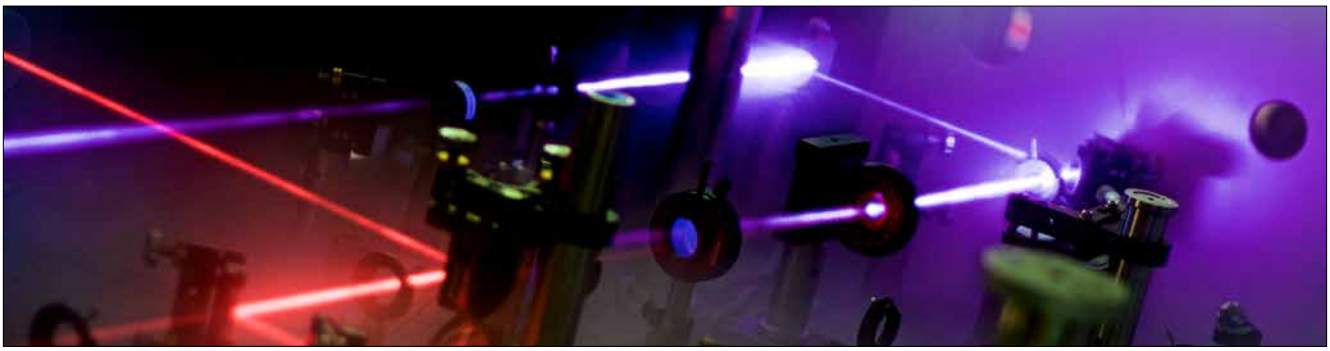


Fig. 1: Time-resolved snapshots of a photo-induced charge-density-wave to semi-metal phase transition, measured with time- and angle-resolved photoelectron spectroscopy, [1].



Ultrafast magnetization dynamics in nanostructures

The speed at which a magnetic state can be manipulated and, hence, data can be magnetically stored depends ultimately on the elementary spin-photon interaction, spin-scattering, and spin-transport processes. Until the mid-1990s, dynamics in magnetic systems were believed to occur on time scales of ~100 picoseconds or longer, determined by the interaction of the spins with the lattice. However, studies using femtosecond laser pulses starting from 1996 revealed the presence of other processes beyond this simple spin-lattice relaxation picture.

In this research field, we use novel methods based on the combination of coherent ultrafast X-ray pulses from laser-based high-harmonic generation with a variety of magneto-optical techniques. These combinations allow us to probe ultrafast spin dynamics with element-specificity and highest time-resolution. Highlights of our research are, e.g., elucidating the role of superdiffusive spin-currents in a femtosecond demagnetization process, and probing the timescale of the exchange interaction in a ferromagnetic alloy. Currently, we study cooperative effects of interacting magnetic subsystems in magnetic multilayers, alloys, and nanostructures.

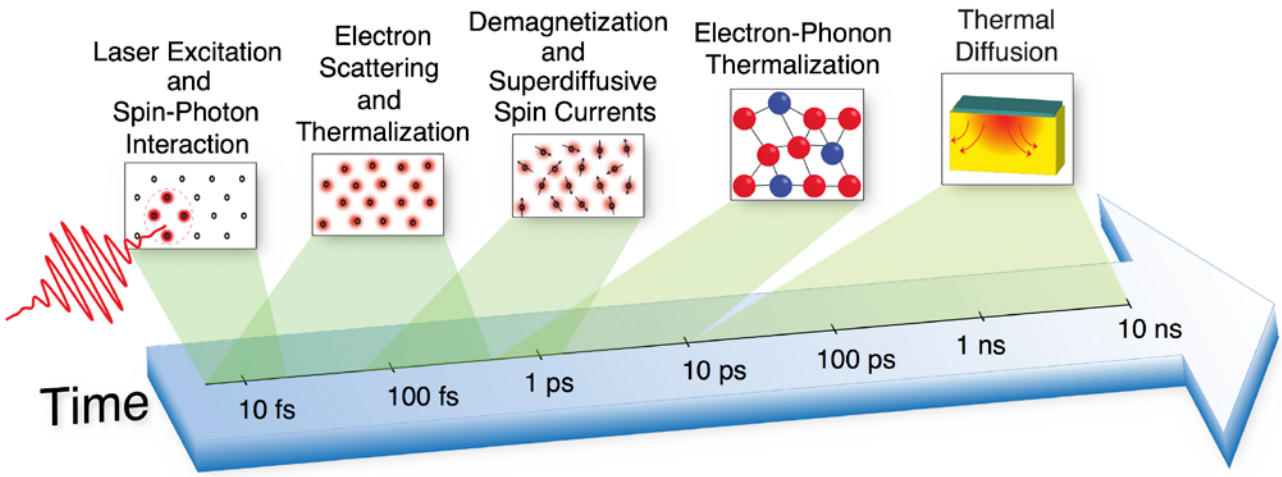
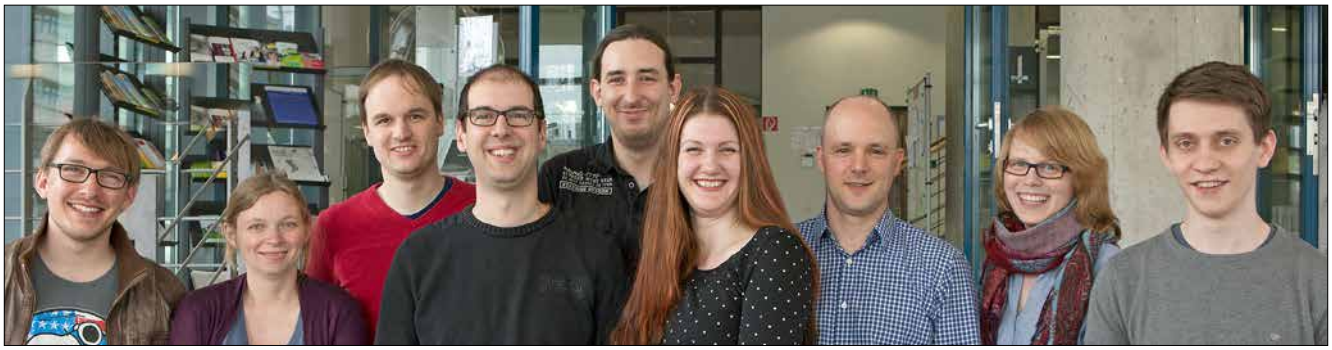


Fig. 2: Schematic timeline of ultrafast photon–electron–spin–lattice interactions after an ultrafast laser excitation. During the ultrafast excitation of the electron system by a femtosecond laser pulse, ultrafast spin-photon interaction can be a source of coherent magnetization dynamics. On a longer femtosecond timescale, various scattering processes between electrons, phonons, and magnons, as well as superdiffusive spin-currents determine the dynamic response of the material. The different contributions of the above-mentioned processes to the ultrafast magnetic dynamics are widely debated and a field of active research, [6].

The short life of electrons at interfaces

The idea in this research area is to study photo-stimulated electron dynamics after an optical excitation in real time. In particular, we are interested in the fate of excited electrons, i.e., their decay processes and their according ultrashort lifetimes. In general, the investigation of the dissipation of such "hot electrons" is of relevance, for instance, in femtochemistry, spin-dynamics, for spin-injection processes, and for energy conversion mechanisms. Since the lifetime of excited electrons plays a central role in all photo-stimulated processes, and also depends on a diverse range of physical parameters, our works extend from dynamics in quantum-well nanostructures to molecule/surface hybrid systems and topological materials with high spin-orbit coupling. As an experimental method to access the relevant ultrafast dynamical processes, we employ time-resolved two-photon photoemission spectroscopy. This real-time pump-probe technique is then combined with different photoemission methods which include angular- ("ARPES"), spin-, and/or real-space resolution.



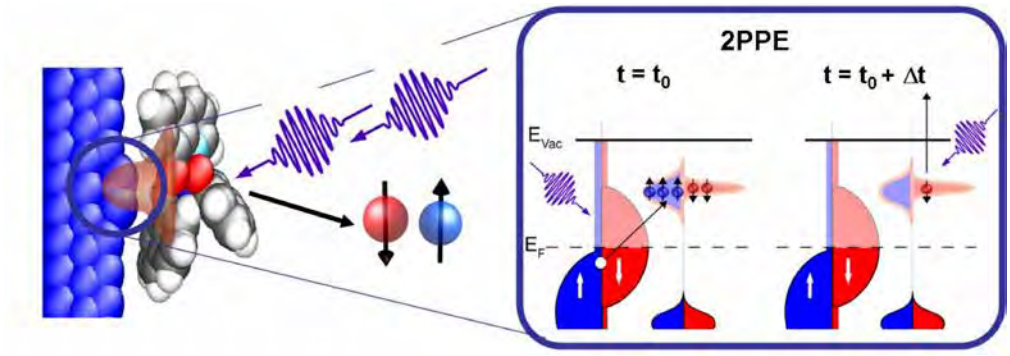


Fig. 3: Schematic of a time- and spin-resolved two-photon photoemission experiment from an organic molecule/metal hybrid system [5].

New tools to study ultrafast materials dynamics

Rapid progress in ultrafast X-ray science worldwide, both in high-harmonic generation (HHG) and X-ray free electron laser (FEL) sources, has paved the way for a new generation of light-matter experiments investigating ultrafast electronic, magnetic, and structural dynamics in materials. Here, we developed in recent years several ultrafast material science experiments that are based on the use of table-top HHG lightsources. By the virtue of the short wavelength pulses produced by high-harmonic generation lightsources, we could show that HHG is an ideal probe for even the fastest dynamics in matter. Using elemental absorption edges, site-specific magnetic, electronic, structural, and chemical dynamics can be captured, providing unique capabilities for the study of complex emerging materials.

In our activities, the development of such novel tools for the study of ultrafast dynamics in materials is an integral part of our research.

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Stefan Mathias, born in 1977 in St. Wendel, studied Physics at the University of Kaiserslautern and the University of Uppsala. From 2004 until 2008 he conducted his PhD in the group of Martin Aeschlimann on ultrafast dynamics on surfaces. In 2009, he was a visiting lecturer at the Kigali Institute of Science and Technology, Rwanda, and moved afterwards to JILA, University of Colorado and NIST, Boulder, USA as part of an EU Marie-Curie International Outgoing Fellowship. In 2012, he was appointed Junior-Professor for Laser Physics and Ultrafast Phenomena in Solids at the University of Kaiserslautern. He joined the University of Göttingen as a full professor in 2015, where he continues his research on ultrafast dynamics in materials.