

## TT 74: Focus Session: Ultra-Fast Magnetism under Electronic Nonequilibrium Conditions (organized by MA)

Organizers: U. Bovensiepen (U. Duisburg-Essen), Martin Eckstein (U. Hamburg), M. I. Katsnelson (U. Nijmegen)

Rapid control of magnetism is of high technological relevance for ultra-fast magnetic storage [Kirilyuk et al., RMP 82, 2731 (2010)]. Because the magnetic order emerges from electronic correlations, magnetism control can be pushed to the extreme time limit with ultra-short laser pulses that bring the electronic state out of equilibrium. For example, recent photoemission studies resolve the dynamics of the exchange splitting in ferromagnetic Gadolinium [Carley et al., PRL 109, 057401 (2012)], and photo-induced phase transitions are possible in materials with coupled orbital and spin order [Wall et al., PRL 103, 097402 (2009)]. When electrons are driven out of equilibrium, the magnetic exchange interaction itself can be manipulated by ultrafast carrier (photo) doping or by dressing the electronic states with light. On the theory side, recent developments like nonequilibrium dynamical mean-field theory [Aoki et al., RMP 86, 779 (2014)] allow to investigate electronic correlations in magnetically ordered systems out of equilibrium. This session will highlight recent experimental and theoretical developments which provide an understanding for the dynamics of magnetism in a regime where separation of slow spin and fast electron dynamics is no longer valid.

Time: Wednesday 15:00–17:45

Location: H 1012

### Invited Talk TT 74.1 Wed 15:00 H 1012

**Ultrafast optical tuning of ferromagnetism in EuO via the carrier density** — ●MANFRED FIEBIG — ETH Zürich, Department of Materials, Vladimir-Prelog-Weg 4, 8093 Zurich, Switzerland

The interest in manipulating magnetic order by ultrashort laser pulses has thrived since it was observed that such pulses can be used to alter magnetization on a sub-picosecond timescale. In many cases demagnetization by laser heating dominates the dynamics; this is well described by the classic three-temperature model — assuming energy exchange between thermalized reservoirs of electrons, spins, and the lattice. Here we demonstrate a mechanism that allows the magnetic order of a material to be enhanced or attenuated at will. This is possible in systems simultaneously possessing a low, tunable density of conduction band carriers and a high density of magnetic moments. In such systems the thermalization time can be set such that adiabatic processes dominate the photoinduced change of the magnetic order — the three-temperature model is bypassed. In ferromagnetic  $\text{Eu}_{1-x}\text{Gd}_x\text{O}$  we thereby demonstrate strengthening as well as weakening of the magnetic order by  $\sim 10\%$  and within  $\leq 3$  ps by optically controlling the magnetic exchange interaction. A theory backing up and expanding our experimental results will be presented.

### Invited Talk TT 74.2 Wed 15:30 H 1012

**Intra-atomic exchange in ultrafast magnetism** — ●MARTIN WEINELT — Freie Universität Berlin, Fachbereich Physik, Arnimallee 14, 14195 Berlin, Germany

The exchange interaction is the defining element in the formation of magnetic order in atoms and solids. Therefore the role of intra- and interatomic exchange during ultrafast magnetization dynamics needs to be explored. In the atomic magnetism of lanthanide metals localized  $4f$  and itinerant  $5d$  orbitals contribute to the overall magnetic moment. In general it is assumed that the intra-atomic exchange coupling is fast enough to be treated as an instantaneous process. We studied the magnetization dynamics of the lanthanide metals gadolinium [1] and terbium by time-resolved photoemission employing femtosecond higher-order harmonic vacuum-ultraviolet pulses. Recording in parallel  $4f$  magnetic linear dichroism and  $5d$  exchange splitting we observe distinct spin dynamics in Gd, which show the breakdown of the intra-atomic exchange upon femtosecond laser excitation. An orbital-resolved Heisenberg model [2] explains well the state-dependent two timescales of magnetization dynamics in Gd metal, which differ by one order of magnitude. Due to its much stronger spin-lattice coupling, Tb shows a distinctly different magnetization dynamics.

[1] Robert Carley *et al.*, Phys. Rev. Lett. **109** (2012) 057401.

[2] Soenke Wienholdt *et al.*, Phys. Rev. B **88** (2013) 020406(R).

15 min. break

### Invited Talk TT 74.3 Wed 16:15 H 1012

**Laser induced ultrafast demagnetization in solids: a time-dependent density functional theory perspective** — ●SANGEETA

SHARMA, J. K. DEWHURST, K. KRIEGER, P. ELLIOTT, and E. K. U. GROSS — Max-Planck-Institut für Mikrostrukturphysik, Weinberg 2, D-06120 Halle, Germany.

Ultrafast manipulation of spins in a controlled manner is a milestone of solid state physics. The motivation for this is to use electronic spin for storing binary data, which can then be optically manipulated using lasers. Recent experiments have demonstrated that demagnetization or spin-reorientation processes can be induced by femtosecond laser pulses. However, we are still far from achieving optimally controlled manipulation of spins required for production of devices. One of the reasons behind this is the lack of full understanding of the phenomena leading to demagnetization.

Time-dependent density functional theory (TDDFT) is a formally exact method for describing the real-time dynamics of electrons under the influence of an external field – for example vector potential of the intense laser pulse. We use spin-resolved TDDFT to study of the process of optical demagnetization. The advantage of such a technique is clear from the fact that it is fully ab-initio in nature. Our analysis shows that the demagnetization occurs as a two step process where first the electrons make transitions to excited states, followed by spin-orbit-mediated spin-flip transitions which lead to a loss of moment. Non-collinearity of the spins does not play significant role in the demagnetization process.

### Invited Talk TT 74.4 Wed 16:45 H 1012

**Ultrafast control of the exchange interaction with electric fields** — ●JOHAN H. MENTINK — Max Planck Research Department for Structural Dynamics, University of Hamburg-CFEL, 22761 Hamburg, Germany — Radboud University Nijmegen, Institute for Molecules and Materials, Heyendaalseweg 135, 6525 AJ Nijmegen, The Netherlands

The strongest interaction between microscopic spins in magnetic materials is the exchange interaction  $J_{\text{ex}}$ . Therefore, ultrafast control of  $J_{\text{ex}}$  keeps the promise to control spins on ultimately fast timescales, potentially bypassing fundamental speed limits for the control of magnetism with magnetic fields. In this talk we provide theoretical evidence that the exchange interaction can be manipulated on ultrashort timescales by strong electric field pulses. Focusing on the prototype Mott-Hubbard insulator, we find very different effects for resonant and off-resonant photo-excitation. In the former case, the electron distribution is changed and the subsequent relaxation of photo-excited carriers causes an ultrafast reduction of  $J_{\text{ex}}$ . Conversely, off-resonant driving allows for an ultrafast and reversible control of  $J_{\text{ex}}$ , *i.e.*, it is active while the field is on, but leaves the electronic state unexcited after the pulse is switched off. In the regime of weak-driving strength the modification of  $J_{\text{ex}}$  is proportional to the intensity of the electric field and we find that  $J_{\text{ex}}$  can be enhanced and reduced for frequencies below and above gap, respectively. Furthermore, for strong driving even the sign of the exchange interaction can be reversed and we show that this causes time reversal of the associated quantum spin dynamics.

## Invited Talk

TT 74.5 Wed 17:15 H 1012

**Controlling, probing and harnessing the strongest force in magnetism** — ●ALEXEY KIMEL — Radboud University Nijmegen, Institute for Molecules and Materials, 6525 AJ Nijmegen, The Netherlands

The idea to change magnetic properties of media with the help of light has long intrigued people in physics and chemistry. Naturally, this raises the question about the speed limit of the optical control of magnetism. Fundamentally, magnetic order is a macroscopic manifestation of a quantum mechanical exchange coupling between spins. This exchange coupling represents the largest interaction in magnetism. It can be associated with an effective magnetic field of 100-1000 T. The

strength can be appreciated from the fact that magnetic order in condensed matter survives well above room temperature. Obviously, harnessing the exchange interaction is the way to achieve the ultimately fastest magnetic switching. How can we control, probe and harness the exchange interaction for ultrafast magnetic switching? Here we demonstrate that the exchange interaction can be manipulated through ultrafast laser excitation in a large class of transition metal oxides. We show that using ultrashort laser pulses one can monitor laser-induced dynamics of the energy of the exchange interaction with subpicosecond temporal resolution. Finally, we suggest a scenario in which the strength of the exchange interaction is employed to achieve the fastest possible magnetic switching.