MA 9: Ultrafast Magnetization Effects II

Time: Monday 15:00–18:30 Location: H 2013

Invited Talk MA 9.1 Mon 15:00 H 2013

Optical control of 4f orbital state in rare-earth metals — •N. Thielemann-Kühn[1], T. Amrhein[1], W. Bronsch[2], S. Jana[3], N. Pontius[3], R. Y. Engel[4], P. S. Miedema[4], D. Legut[5], K. Carva[6], U. Atxitia[1], B. E. van Kuiken[7], M. Teichmann[7], R. E. Carley[7], L. Mercadier[7], A. Yaroslavtsev[7], G. Mercurio[7], L. Le Guyader[7], N. Agarwal[7], R. Gort[7], A. Scherz[7], S. Dziarzhytski[4], G. Brenner[4], F. Pressacco[4], R.-P. Wang[4,10], J. O. Schunck[4,10], M. Sinha[4], M. Beye[4], G. S. Chiuzbăian[9], P.. Oppeneer[8], M. Weinelt[1], and C. Schüssler-Langeheine[3] — [1]Freie Universität Berlin — [2]Elettra-Sincrotrone Trieste — [3]Helmholtz-Zentrum Berlin — [4]Deutsches Elektronen-Synchrotron — [5]IT4Innovations & Nanotechnology Centre — [6]Charles University — [7]European XFEL — [8]Uppsala University — [9]Sorbonne Université — [10]Universität Hamburg

Rare-earth 4f metals play a key role in materials which allow all-optical switching and their large spin-orbit coupling can result in strong magneto-crystalline anisotropy (MCA) - a property highly relevant for stable magnetic information storage. In time-resolved X-ray absorption (XAS) and Resonant inelastic X-ray scattering (RIXS) studies on different 4f-metals, we found that 5d-4f inelastic electron-electron scattering leads to selective excitation in the 4f shell, which change the angular momentum J. As a consequence, the MCA and thus coupling of the 4f-spin system to its environment is manipulated on fs-timescales. In the case of Dy we observed ferro- and antiferromagnetic dynamics to be strongly affected by the altered electronic state.

MA 9.2 Mon 15:30 H 2013

Ultrafast demagnetization dynamics in Fe studied by tr-ARPES — •Xinwei Zheng, Christian Strüber, and Martin Weinelt — Freie Universität Berlin, Fachbereich Physik, Arnimallee 14, 14195 Berlin

We investigate the ultrafast magnetization dynamics of Fe(110) on W(110) by time- and angle-resolved photoemission spectroscopy (tr-ARPES) with an IR pump pulse and a high-order harmonic XUV probe pulse. After laser excitation, transient changes to the band structure with tr-ARPES are observed. In contrast to a small reduction of the exchange splitting by only 3%, the magnetic linear dichroism signal calculated from the asymmetry between spectra for two opposite in-plane magnetization directions drops by 60% within 150 fs. This result proves that spin-mixing dominates the ultrafast demagnetization dynamics in Fe [1]. Moreover, we measure a stronger reduction of the asymmetry in the minority spin band close below the Fermi level compared to that in the majority spin band at a binding energy of 0.3 eV. This indicates that the spin system in Fe is out of equilibrium for about 1-2 ps after laser excitation [2]. The pump-fluence dependence of the MLD signal and the electronic temperature implies that a significant amount of energy is transferred into the spin system within 150 fs, leading to spin excitations particularly within the minority spin band.

[1] E. Carpene *et al.*, Phys. Rev. B. **78**, 174422 (2008)
[2] R. Gort *et al.*, Phys. Rev. Lett. **121**, 087206 (2018)

MA 9.3 Mon 15:45 H 2013

Unraveling light-driven spin transfer and hot carrier dynamics by EUV magneto-optical spectroscopy — •G. S. Matthijs Jansen[1], Christina Möller[1], Henrike Probst[1], John Kay Dewhurst[2], Marcel Reutzel[1], Sangeeta Sharma[2], Daniel Steil[1], and Stefan Mathias[1] — [1]I. Physikalisches Institut, Georg-August-Universität Göttingen — [2]Max-Born-Institute for Non-linear Optics and Short Pulse Spectroscopy, Max-Born Strasse 2A, 12489 Berlin, Germany

In recent years, extreme ultraviolet magneto-optical spectroscopy has helped to identify a number of unique ultrafast magnetization processes, including optical inter-site spin transfer (OISTR) and delayed demagnetization in Fe-Ni alloys. However, the interpretation in terms of the element-resolved magnetic moment in non-equilibrium is not always straightforward, as we have recently shown for transverse magneto-optical Kerr spectroscopy of Ni and Fe-Ni thin films [1, 2]. To overcome this issue, we demonstrate that an angle-resolved analysis enables complete access to the magneto-optical response function, and moreover enables a quantitative comparison to ab-initio theoretical calculations. Through such an analysis, we are able to verify the existence of OISTR in Fe-Ni alloys and furthermore shine light on the intimate connection between OISTR and delayed demagnetization [2].

[1] H. Probst et al., arXiv preprint arXiv:2306.02783 (2023)
[2] C. Möller et al., arXiv preprint arXiv:2306.02793 (2023)

MA 9.4 Mon 16:00 H 2013

Phonon mediated spin-spin interaction — •Daniel Schick[1], Markus Weissenhofer[2,3], Akashdeep Kamra[4], and Ulrich Nowak[1] — [1]Fachbereich Physik, Universität Konstanz, Konstanz, Germany — [2]Department of Physics and Astronomy, Uppsala University, Uppsala, Sweden — [3]Department of Physics, Freie Universität Berlin, Berlin, Germany — [4]IFIMAC - Condensed Matter Physics Center, Universidad Autónoma de Madrid, Madrid, Spain

The transfer and control of angular momentum is a key aspect for spintronic applications. Recently, it has been realized, that even the lattice can contribute to the exchange of angular momentum via chiral phonons. To simulate spin lattice coupling, we developed a numerical method, which is able to conserve the total angular momentum, allowing us to precisely track changes thereof in transfers between the spin and lattice system [1, 2]. In our simulations, magnetic spins can excite chiral phonons, which themselves are able to elicit a response of other spins, creating an effective spin-spin coupling via phonons. We discuss this interaction by driving spins to investigate the processes involved and determine the importance of the chirality of the phonons. In finite-temperature simulations, we investigate the thermodynamic implications of this interaction.

[1] S. Mankovsky et. al, Phys. Rev. Lett. **129**, 067202, (2022)
[2] M. Weißenhofer et. al, Phys. Rev. B **108**, L060404, (2023)

MA 9.5 Mon 16:15 H 2013

Non-Hermitian phase transition in optically-driven ferromagnetic semiconductors — •J. Li[1], M. Matsubara[2], K. Kliemt[3], N. Kaya[3], I. Reiser[3], M. Turaev[4], C. Krellner[3], S. Pal[5], J. Kroha[4], and M. Fiebig[1] — [1]ETH Zurich, Switzerland — [2]Tohoku Uni., Japan — [3]Goethe Uni. Frankfurt, Germany — [4]Bonn Uni., Germany — [5]NISER Bhubaneshwar, India

Phase transition in thermodynamic equilibrium is a heavily studied topic. Recently, however, non-equilibrium phase transitions have been drawing attention. Such transitions occur typically in open quantum systems dissipatively coupled to the environment and/or driven by external fields. As a result, the Hamiltonian eigenvalues can be complex-valued, which is why these transitions are referred to as non-Hermitian phase transition (NHPT). While most of the previous works studied bosonic quantum systems, we report our observation of a NHPT in a fermionic quantum system, i.e., the ferromagnetic semiconductor EuO. EuO has a Curie temperature (T_C) of 69 K. In our study, it is driven out of equilibrium by a 120 fs optical pump pulse at 1.55 eV. We probe the relaxation dynamics by measuring the pump-induced reflectivity change using another time-delayed 120 fs optical probe pulse at 1.31 eV. Below T_C, we find a clear signature of bi-exponential decay of the pump-induced reflectivity change, which becomes a damped, oscillatory behavior above T_C, indicating the appearance of complex eigenvalues of the optical response function. Our results reveal that the non-equilibrium situation triggers a NHPT in EuO that is marked by an exceptional point where two eigenvalues coalesce.

MA 9.6 Mon 16:30 H 2013

Nucleation and domain growth during the laser-induced metamagnetic phase transition in FeRh identified by UXRD — •Maximilian Mattern[1], Steffen Zeuschner[1], Jon Ander Arregi[2], Vojtěch Uhlíř[2], and Matias Bargheer[1,3] — [1]Institut für Physik und Astronomie, Universität Potsdam, Germany — [2]CEITEC BUT, Brno University of Technology, Czech Republic — [3]Helmholtz-Zentrum Berlin, Germany

We use time-resolved x-ray diffraction (UXRD) to study the laser-induced first-order antiferromagnetic (AFM) to ferromagnetic (FM) phase transition in FeRh that is accompanied by a gigantic expansion ($\approx 0.6\%$). The laser-induced rise of this expansion accesses the transient FM volume fraction independent of the orientation of the magnetic moment. By comparing two samples with thicknesses below and above the optical penetration depth, we disentangle optically induced nucleation on an intrinsic 8 ps timescale in the near-surface

region from the growth of the nucleated FM domains into the depth of the thick inhomogeneously excited FeRh film by slow heat diffusion.

We observe this 8 ps nucleation timescale even in the absence of a transient hot Fermi-distribution of the electrons, when we increase the pump pulse duration up to 10.5 ps. This indicates that hot electrons are not required for driving the phase transition on its intrinsic timescale. Instead, we identify a non-linearity in the laser-induced expansion due to the treshold behaviour of the first-order phase transition, that only starts when the energy successively deposited by the ps-pump pulse overcomes the local critical treshold.

15 min. break

MA 9.7 Mon 17:00 H 2013

Ultrafast electron scattering in a ferromagnetic two-sublattice system — •Ariyan Tavakoli, Kai Leckron, and Hans Christian Schneider — Physics Department and Research Center OPTIMAS, RPTU Kaiserslautern-Landau, Germany

We investigate an exchange coupled two-sublattice system as a model for a ferromagnetic alloy, as introduced in [1]. In addition to the exchange coupling between an itinerant and a localized spin system, and the electron-phonon scattering in the itinerant system we also include the electron-electron Coulomb interaction in the itinerant system. We study numerically the heat-induced ultrafast magnetization dynamics due to electron-electron, electron-phonon, Elliott-Yafet spin-flip and exchange scattering and discuss different scenarios for the demagnetization and relaxation dynamics of the sublattice. Our results show the impact of electron-electron scattering for the spin dynamics on ultrashort timescales.

[1] K. Leckron, A. Baral and H. C. Schneider, "Exchange scattering on ultrafast timescales in a ferromagnetic two-sublattice system", Appl. Phys. Lett. 120, 102407 (2022).

MA 9.8 Mon 17:15 H 2013

Time-resolved resonant magnetic small-angle scattering with a laser-driven soft-X-ray plasma source — •Leonid Lunin[1], Niklas Schneider[1], Konstanze Korell[1], Martin Borchert[1], Michael Schneider[1], Johannes Tümmler[1], Stefan Eisebitt[1,2], Bastian Pfau[1], and Daniel Schick[1] — [1]Max-Born-Institut für Nichtlineare Optik und Kurzzeitspektroskopie, Berlin, Germany — [2]Institut für Optik & Atomare Physik, TU Berlin, Germany

Resonant soft-X-ray scattering methods provide unique possibilities to study nanometer-scale magnetization dynamics with element selectivity and on ultrafast timescales when employing ultrashort pulsed X-ray sources. Based on a laser-driven plasma X-ray source, we have developed a novel instrument to carry out time-resolved magnetic small-angle X-ray scattering (SAXS) experiments in the soft-X-ray range between 500 and 1500 eV with sub-10 ps temporal resolution. In this contribution, we show time and element-resolved magnetic SAXS results of a photoexcited GdFe multilayer sample. The 2D scattering patterns collected contain information on the local magnetization, periodicity, and distribution of the magnetic maze domains in GdFe. The development of an online processing software for a hybrid-pixel detector enables us to drastically enhance the signal-to-noise ratio of the time-resolved data and reduce overall dead-times of the setup. Due to the flexibility of our laboratory-scale setup, we can further vary the sample environment, e.g., by applying external magnetic fields as well as cryogenic temperatures, and observe significant differences in the ground-state-dependent dynamics of the magnetic domains.

MA 9.9 Mon 17:30 H 2013

Ballistic or Diffusive transport? Signature in Complex Kerr response of magnetized films — •Sanjay Ashok, Jonas Hoefer, Martin Stiehl, Martin Aeschlimann, Hans-Christian Schneider, Baerbel Rethfeld, and Benjamin Stadtmueller — Department of Physics and Research Center OPTIMAS, RPTU Kaiserslautern-Landau

Transport mechanisms play an important role in ultrafast demagnetization. We calculate the influence of diffusive and ballistic transport on ultrafast magnetization dynamics in metallic magnets with varying film thicknesses [1]. We then simulate the probe-angle dependent Kerr-rotation and -ellipticity (CMOKE) dynamics [2, 3].

Our simulations reveal probe-angle dependence in the Kerr-ellipticity dynamics for the diffusive transport case. In contrast, no angle dependent signatures were found for the case when ballistic transport is predominant. No probe-angle dependence are found in the Kerr-rotation dynamics with either of the transport mechanisms.

Our theoretical predictions are compared with probe angle dependent CMOKE measurements on a 40 nm thick Nickel film. The angle dependence of the measured Kerr signals closely matches the simulated response with diffusive transport. Thus, we demonstrate the utility of probe-angle dependent CMOKE technique and, importantly, that the ballistic transport can be neglected in a 40nm thick Nickel film.

[1] Ashok et al. APL, 120 142402 (2022)
[2] Traeger et al. PSS, 131, 201 (1992)
[3] Kuch et al. Springer Surf. Sci. 57, (2015)

MA 9.10 Mon 17:45 H 2013

Influence of thermal disorder on ultrafast demagnetization — •Franziska Ziolkowski, Oliver Busch, Ingrid Mertig, and Jürgen Henk — Martin Luther University Halle-Wittenberg, Halle, Germany

Ultrafast demagnetization induced by ultrashort laser pulses in magnetic materials remains a topic of intense investigation. To explain the rapid quenching of magnetization various spin flip and spin transfer mechanisms are discussed in literature. With the theoretical framework evolve [1] we investigate such ultrafast magnetization dynamics.

Our approach is based on a real-space tight-binding model and includes optical excitation as well as coupling to an external heat bath. The occupation matrix yields the system's observables and is evolved in time by the Lindblad equation. This approach allows to reveal and to tune the relevant underlying processes.

Our study examines the influence of thermal disorder within the magnetic configuration on ultrafast demagnetization. We determine the regime of influence, which is the pertinent parameter space for this effect. Furthermore a comparison is made between spin current generation and ultrafast demagnetization in homogeneous systems versus ferromagnetic-nonmagnetic heterostructures.

[1] Töpler *et al* 2021 *New J. Phys.* **23** 033042

MA 9.11 Mon 18:00 H 2013

Real-space imaging of ultrafast dynamics in nanoscale magnetic domains — •Hung-Tzu Chang[1], Sergey Zayko[1], Timo Schmidt[2], Murat Sivis[1,3], Manfred Albrecht[2], and Claus Ropers[1,3] — [1]Max Planck Institute for Multidisciplinary Sciences, 37077 Goettingen, Germany — [2]Experimental Physics IV, Institute for Physics, University of Augsburg, 86159 Augsburg, Germany — [3]IV Physical Institute, University of Goettingen, 37077 Goettingen, Germany

Direct visualization of photoinduced dynamics of nanoscale spin textures is crucial for understanding the mechanisms of ultrafast demagnetization and all-optical magnetic switching. Here, we image the real-space response of magnetic domains in optically excited Co-based magnetic thin films with circularly polarized femtosecond extreme ultraviolet pulses [1]. Our experiment provides direct observations of domain wall dynamics at picosecond timescales, as well as the photoinduced disappearance of small domains, which should further advance the understanding of spin dynamics in nanomaterials.

[1] Zayko et al. Nat. Commun. 12, 6337 (2021)

MA 9.12 Mon 18:15 H 2013

Exciton-driven phonon dynamics in a magnetic layered semiconductor CrI_3 — •Martin Pavelka, Vishal Shokeen, Ruslan Chulkov, Alexander Yaroslavtsev, Johanna Rogvall, David Muradas, Ulrich Noumbe, Mahmoud Abdel-Hafiez, Venkata Kamalakar Mutta, Oscar Grånäs, and Hermann Dürr — Uppsala University, Uppsala, Sweden

CrI_3 is a prototype of a new class of magnetic 2D materials displaying ferromagnetic order down to the monolayer limit [1]. Its optical and magneto-optical spectra feature several excitonic dd-transitions characterized by spin-polarized electron and hole pairs with evidence of strong coupling to optical phonons [2].

In this talk, we will describe the ultrafast optical and magneto-optical responses of CrI_3. Our two-colour femtosecond spectroscopy allows us to investigate the interplay of exchange interaction and electron-phonon coupling. Following the optical generation of excitons, we observe coherent oscillations in the time-domain. Fourier transformation of the spectra shows two coherent phonon modes, the A^1_{1g} and A^2_{1g} symmetries, at frequencies of 2.4 and 3.9 THz, respectively. Theoretical calculations show that both modes couple to the magnetic order, in agreement with our observations. This finding is contrary to the previous report [2]. The starting phase of these two impulsively excited modes indicates a magnetic contribution to their

generation. We also explore the fluence dependence of these phenomena to assess the possible influence of neighbouring excitons.

[1] Nature 546, 270-273 (2017). [2] Nat Commun 13, 4473 (2022).