MA 3: Ultrafast Magnetization Effects I

Time: Monday 9:30–12:45 Location: H 2013

Invited Talk MA 3.1 Mon 9:30 H 2013

Effects of Magnetization Inertia in Spin Dynamics — •Anna Semisalova — Faculty of Physics and CENIDE, University of Duisburg-Essen, Duisburg, Germany

Magnetization inertia has been shown to give rise to an additional motion of magnetization - a THz-frequency nutation, superimposed on the regular GHz precession [1-4], attractive for ultrafast magnonics [5]. Furthermore, in nanoparticles, magnetic nutation can be induced by a non-uniform spin configuration due to surface anisotropy [6]. In this talk, I discuss how to resolve effects of inertia on spin dynamics in anisotropic ferromagnets [7]. Within the inertial LLG equation, we find a reduction of the ferromagnetic resonance (FMR) frequency for both aligned and non-aligned modes due to inertia. We illustrate this phenomenon for model thin film systems with cubic and uniaxial magnetocrystalline anisotropy. Notably, for an out-of-plane magnetic field the FMR frequency dependence of a thin film becomes non-linear, in contrast to conventionally used Kittel formula. We also find that the nutation frequency increases with the magnetic anisotropy and the applied field. These higher-order corrections due to inertia are important for an accurate evaluation of magnetic anisotropy and g-factor, and for an interpretation of spin dynamics experiments at higher frequencies [7]. Support from DFG is gratefully acknowledged (SE 2853/1-1 | AL 618/37-1; CRC/TRR 270). [1] PRB 83, 020410(R) (2011); [2] PRB 102, 184432 (2020); [3] Nat. Phys. 17, 245 (2021); [4] JMMM 579, 170830 (2023); [5] PRB 103, 174435 (2021); [6] PRB 98, 165444 (2018); [7] PRB 106, 054428 (2022)

MA 3.2 Mon 10:00 H 2013

Ultrafast generation of nonthermal magnons in iron: *Ab initio* parameterized calculations — •Markus Weissenhofer[1,2] and Peter M. Oppeneer[1] — [1]Uppsala University, Uppsala, Sweden — [2]Freie Universität Berlin, Berlin, Germany

Ultrafast laser excitation of ferromagnetic metals gives rise to correlated, highly non-equilibrium dynamics of electrons, spins and lattice, which are, however, poorly described by the widely used three-temperature model (3TM). We develop a fully *ab initio* parameterized out-of-equilibrium theory based on a quantum kinetic approach – termed *(N+2) temperature model* – that describes magnon occupation dynamics due to electron-magnon scattering [1]. We apply this model to perform quantitative simulations on the ultrafast, laser-induced generation of magnons in iron and demonstrate that on these timescales the magnon distribution is non-thermal: predominantly high-energy magnons are created, while the magnon occupation close to the center of the Brillouin zone even decreases, due to a repopulation towards higher energy states. We show that the 3TM can be derived from our model and compare it with our microscopic calculations. In doing so, we demonstrate that the simple relation between magnetization and temperature computed at equilibrium does not hold in the ultrafast regime and that the 3TM greatly overestimates the demagnetization. Our calculations show that ultrafast generation of non-thermal magnons provides a sizable demagnetization within 200 fs and, thus, emphasize the importance of magnons for ultrafast demagnetization. [1] M.Weißenhofer and P.M. Oppeneer, arXiv:2309.14167

MA 3.3 Mon 10:15 H 2013

Spin nutation driven non-resonantly by ultrashort laser pulses — •A. De[1], J. Schlegel[2], A. Lentfert[1], L. Scheuer[1], B. Stadtmüller[1], P. Pirro[1], G. von Freymann[1,3], U. Nowak[2], and M. Aeschlimann[1] — [1]RPTU Kaiserslautern-Landau — [2]Universität Konstanz — [3]Fraunhofer ITWM, Kaiserslautern

The interaction of ultrashort laser pulses with ferromagnet can trigger a variety of new phenomena such as ultrafast demagnetization, all-optical switching, etc. In this work, we focus on the optically driven magnetization dynamics in the yet unexplored timescale between ultrafast demagnetization and the collective precession motion of the spin system. In this intermediate time window, the direction of the magnetic moment and angular momentum are transiently separated due to inertia. This results in additional oscillations, known as nutation, superimposed on the usual precession, with higher frequencies but smaller amplitudes and relaxation times. We experimentally observe nutation (with frequency around 100 GHz) in permalloy thin films by all-optical time-resolved magneto-optical Kerr effect (TR-MOKE) measurements. The nutation frequency shows a negligible dependence on magnetic field and film thickness. These results are confirmed by atomistic spin model simulations, providing insights into a deeper understanding of nutation at ultrafast timescales.

MA 3.4 Mon 10:30 H 2013

Sub-wavelength localised all-optical helicity-independent switching in GdTbCo using plasmonic gold nanodisks — Themistoklis Sidiropoulos, •Puloma Singh, Tino Noll, Michael Schneider, Dieter Engel, Felix Steinbach, Ingo Will, Denny Sommer, Clemens von Korff Schmising, and Stefan Eisebitt — Max Born Institut für Nichtlineare Optik und Kurzzeitspektroskopie, Berlin, Germany

All-optical helicity-independent switching (AO-HIS) is of interest for ultrafast and energy efficient magnetic switching in future magnetic data storage approaches. Yet, to achieve high bit density magnetic recording it is necessary to reduce the size of magnetic bits while controlling their shape and position. Metallic nanostructures that support localized surface plasmons enable electromagnetic confinement well below the diffraction limit and rare-earth transition metal alloys such as GdTbCo have demonstrated nanometer-sized stable domains. Here, we deposit plasmonic gold nanodisks on GdTbCo films and probe the magnetic state using magnetic force microscopy. We observe localised AO-HIS of the sample after resonant excitation of the gold nanodisks by a single 370 fs long laser pulse with a center wavelength of 1030 nm. We demonstrate that the strong localization of optical fields through plasmonic nanodisks enables nanoscale AOS-HIS at a sub-wavelength scale length which is comparable to structure sizes in commercial heat assisted magnetic recording. Moreover, we study the influence of the localized electromagnetic field enhancement by the plasmonic nanoparticles on the required fluence to switch the magnetization.

MA 3.5 Mon 10:45 H 2013

Influencing ultrafast demagnetization with the OAM of light — •Paul Herrgen, Eva Prinz, Benjamin Stadtmüller, and Martin Aeschlimann — Department of Physics and Research Center OPTIMAS, Rheinland-Pfälzische Technische Universität Kaiserslautern-Landau, 67663 Kaiserslautern, Germany

As bosons, photons always have a spin angular momentum, which is associated with left- or right-handed circular polarization. This angular momentum is limited to values of $\pm 1\hbar$. Additionally, the photons can also have an orbital angular momentum (OAM). With an angular momentum of $l\hbar$ with $l \in \mathbb{Z}$ it is possible to increase the total angular momentum of the photons drastically.

In contrast to the spin angular momentum, we found a significant influence of the orbital angular momentum of light on the ultrafast magnetization dynamics. Interestingly, the photonic OAM changes the magnetization only after the interaction of the light pulse with the material [1]. Furthermore, we find a clear relationship between the influence of photonic OAM and the angle between the wave vector of the OAM beam and the magnetization. These empirical results provide the basis for a microscopic model of the OAM-driven magnetization dynamics.

[1]: Prinz et al., arXiv:2206.07502

15 min. break

MA 3.6 Mon 11:15 H 2013

Ultrafast Spin Dynamics in non-collinear Antiferromagnetic Mn_3Sn — •Chongxiao Fan[1,2], Bingke Xiang[3], Dante Kennes[2,1], Angel Rubio[1,4], Yihua Wang[3], and Peizhe Tang[5,1] — [1]Max Planck Institute for the Structure and Dynamics of Matter, 22761 Hamburg, Germany. — [2]Institut für Theorie der Statistischen Physik, RWTH Aachen University, 52062 Aachen, Germany. — [3]State Key Laboratory of Surface Physics and Department of Physics, Fudan University, Shanghai 200433, China — [4]Center for Computational Quantum Physics (CCQ), The Flatiron Institute, 162 Fifth avenue, New York NY 10010, USA. — [5]School of Materials Science and Engineering, Beihang University, Beijing 100191, China

Ultrafast manipulation of magnetic moments is a captivating subject within contemporary magnetism and spintronics. In this work, we conduct a pump-probe experiment on the non-collinear antiferromagnet Mn_3Sn, which shows large anomalous Hall effect due to nontrivial topology of its electron band structure. The observed asynchronous alteration after the pump in both the magneto-optical Kerr signal and reflectivity implies changes linked not only to the electron occupation but also to a change of the spin configuration. The relatively low heat capacity of electrons will cause a rapid increase in electron temperature and thermoelectric current can generate polarized current. Spin dynamics calculation confirmed the possible formation of a net magnetization when the pump fluence is large. Our research uncovers novel phenomena within antiferromagnets, offering a fresh approach to comprehend ultrafast physics in magnetic systems.

MA 3.7 Mon 11:30 H 2013

Ab initio investigation of laser-induced ultrafast demagnetization of L10 FePt: Intensity dependence and importance of electron coherence — •Mrudul Muraleedharan S. and Peter M. Oppeneer — Department of Physics and Astronomy, Uppsala University, Uppsala, Sweden

We theoretically investigate the optically-induced demagnetization of ferromagnetic FePt using the time-dependent density functional theory (TDDFT). We compare the demagnetization mechanism in the perturbative and nonperturbative limits of light-matter interaction and show how the underlying mechanism of the ultrafast demagnetization depends on the driving laser intensity. Our calculations show that the femtosecond demagnetization results from a nonlinear optomagnetic effect akin to the inverse Faraday effect. The demagnetization scales quadratically with the electric field E in the perturbative limit, i.e., $\Delta M_z \propto E^2$. Moreover, the magnetization dynamics happens dominantly at even multiples $n\omega_0$, $(n = 0, 2, \cdots)$ of the pump-laser frequency ω_0. We further investigate the demagnetization in conjunction to the optically-induced change of electron occupations and electron correlations. Comparing the *ab initio* computed demagnetizations with those calculated from spin occupations, we show that electronic coherence plays a dominant role in the demagnetization process, whereas interpretations based on the time-dependent occupation numbers poorly describe the ultrafast demagnetization.

MA 3.8 Mon 11:45 H 2013

Ultrafast dynamics of different magnetic properties in a helical Heisenberg antiferromagnet — •Hyein Jung[1,2], Abeer Arora[2], Victoria Taylor[2], Túlio de Castro[2], Franziska Walther[3], Kristin Kliemt[3], Christian Schüssler-Langeheine[4], Niko Pontius[4], Urs Staub[5], Cornelius Krellner[3], Laurenz Rettig[2], Ralph Ernstorfer[1,2], and Yoav William Windsor[1,2] — [1]Technische Universität Berlin, Berlin, Germany — [2]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany — [3]Goethe-Universität Frankfurt, Frankfurt, Germany — [4]Helmholtz-Zentrum Berlin für Materialien und Energie, Berlin, Germany — [5]Paul Scherrer Institut, Villigen, Switzerland

Ultrafast control of spin order holds great promise for future devices. In particular, Eu-based magnetism is appealing due Eu's extremely high moment size. However, the spatially localized 4f magnetic states interact indirectly, mediated by the conduction electrons via Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction. Control of RKKY presents a new avenue for manipulation of spin order, which is of great need particularly for manipulation of antiferromagnets, which are largely considered the basis for next-generation spintronics. Here, we study ultrafast spin dynamics in the chiral antiferromagnet (AF) $EuCo_2P_2$ using resonant X-ray diffraction. We probe two magnetic observables: the size of the AF-ordered spins and their periodicity. We demonstrate that following ultrafast photoexcitation, they depart from the expected thermal behavior, revealing distinct nonthermal dynamics in magnon population and strength of the RKKY coupling between Eu 4f states.

MA 3.9 Mon 12:00 H 2013

Femtosecond spin-state switching in Fe(II) spin-crossover thin films — •Lea Kämmerer[1], G. Kämmerer[1], M. Gruber[1], J. Grunwald[2], T. Lojewski[1], L. Mercadier[3], L. Le Guyader[3], R. Carley[3], C. Carinan[3], N. Gerasimova[3], D. Hickin[3], B. E. Van Kuiken[3], G. Mercurio[3], M. Teichmann[3], S. K. Kuppusamy[4], A. Scherz[3], M. Ruben[4,5], K. Sokolowski-Tinten[1], A. Eschenlohr[1], K. Ollefs[1], C. Schmitz-Antoniak[6], F. Tuczek[2], P. Kratzer[1], U. Bovensiepen[1], and H. Wende[1] — [1]University of Duisburg-Essen and CENIDE — [2]Christian-Albrechts-University Kiel — [3]European XFEL — [4]Karlsruhe Institute for Technology — [5]Institut de Science et d*Ingénierie Supramoléculaires (ISIS) Strasbourg Cedex — [6]Technical University of Applied Science Wildau

Spin-crossover molecules have gained popularity in recent years due to their potential applications, including molecular switches. Understanding their switching dynamics is crucial for optimizing their properties, which involve an abrupt, broad thermal hysteresis as a result of cooperative switching at room temperature. X-ray absorption spectroscopy is a sensitive tool for studying the spin-state. Combining X-ray absorption spectroscopy with a femtosecond time-resolution at the European XFEL allowed observation of the underlying femtosecond switching dynamics in Fe(II) spin-crossover thin films. Optical laser pumping triggered the sub-picosecond light-induced low-spin to high-spin transition in this experiment. A detailed analysis of the dynamics reveals the transient population of an intermediate state.

MA 3.10 Mon 12:15 H 2013

Coupled spin-lattice dynamics from electronic structure — Ramon Cardias[1], Simon Streib[2], Zhiwei Lu[3], Manuel Pereiro[2], Anders Bergman[2], Erik Sjöqvist[2], Cyrille Barreteau[4], Anna Delin[3,2], Olle Eriksson[2], and •Danny Thonig[5,2] — [1]Universidade Federal Fluminense, Brazil — [2]University Uppsala, Uppsala — [3]KTH Royal Institute of Technology, Sweden — [4]Université Paris-Saclay, France — [5]Örebro University, Sweden

The interplay between spin and lattice degrees of freedom is important for a wide range of applications in, e.g., sustainable materials research or in ultrafast dynamics [1]. The here often used low-order parameterised energy description, however, is recently discussed to fail in particular at finite temperature [2].

We developed [3] a method that performs the coupled adiabatic spin and lattice dynamics based on the tight-binding electronic structure model, where the intrinsic magnetic field and ionic forces are calculated from the converged self-consistent electronic structure at every time step. By doing so, this method, implemented in *Cahmd* [4], allows us to explore limits of a given spin-lattice Hamiltonian.

We demonstrate how the dynamics of spin and lattice is strongly influenced by each other on the application to low-dimensional systems. For instance, we observed that a disordered magnetic configuration is able the induce significant lattice distortions.

[1] Phys. Rev. Lett. 76, 4250 (1996); Phys. Rev. B 95, 014431 (2017). [2] Sci. Rep. 10, 20339 (2020); Comp. Mat. Sci. 44, 888 (2009) [3] arXiv:2311.00765 [4] available at https://cahmd.gitlab.io/cahmdweb/.

MA 3.11 Mon 12:30 H 2013

Ultrafast dynamics of phase transitions in hematite — •Maik Kerstingskötter[1], Tobias Dannegger[1], András Deák[2], Levente Rózsa[3], László Szunyogh[2], and Ulrich Nowak[1] — [1]Department of Physics, University of Konstanz — [2]Department of Theoretical Physics, Budapest University of Technology and Economics — [3]HUN-REN Wigner Research Centre for Physics, Budapest

The iron oxide hematite is well suited for many spintronic applications and long-distance spin transport due to its insulating and antiferromagnetic properties and its low damping constant. In addition, hematite undergoes a low-temperature first-order phase transition called the Morin transition, leading to an abrupt 90° spin reorientation associated with a transition from a fully antiferromagnetic to a canted state with weak net magnetization. We simulate the nonequilibrium spin dynamics of this phase transition in response to sudden temperature and field variations with femtosecond resolution using atomistic spin dynamics simulations based on ab initio parameters [1].

[1] T. Dannegger et al., "Magnetic properties of hematite revealed by an ab initio parameterized spin model", *Phys. Rev. B*, **107**, 184426 (2023).