6th Ultrafast Magnetism Conference

UMC 2024

2 – 6 September ⇅ Berlin, Germany

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Ultrafast Magnetism Conference – UMC 2024

UMC 2024 - the 6th in the conference series - is dedicated to the field of ultrafast spin and magnetization dynamics and in particular to ultrafast dynamic processes in magnetic materials on picosecond, femtosecond and attosecond time scales. Previous UMC conferences took place in Strasbourg (2013), Nijmegen (2015), Kaiserslautern (2017), York (2019), and Nancy (2022).

The scientific topics of the UMC 2024 conference include the following:

- Spin dynamics and ultrafast switching
- Spin phononics
- Angular momentum beyond spin
- Spin transport and devices
- 2D and new materials

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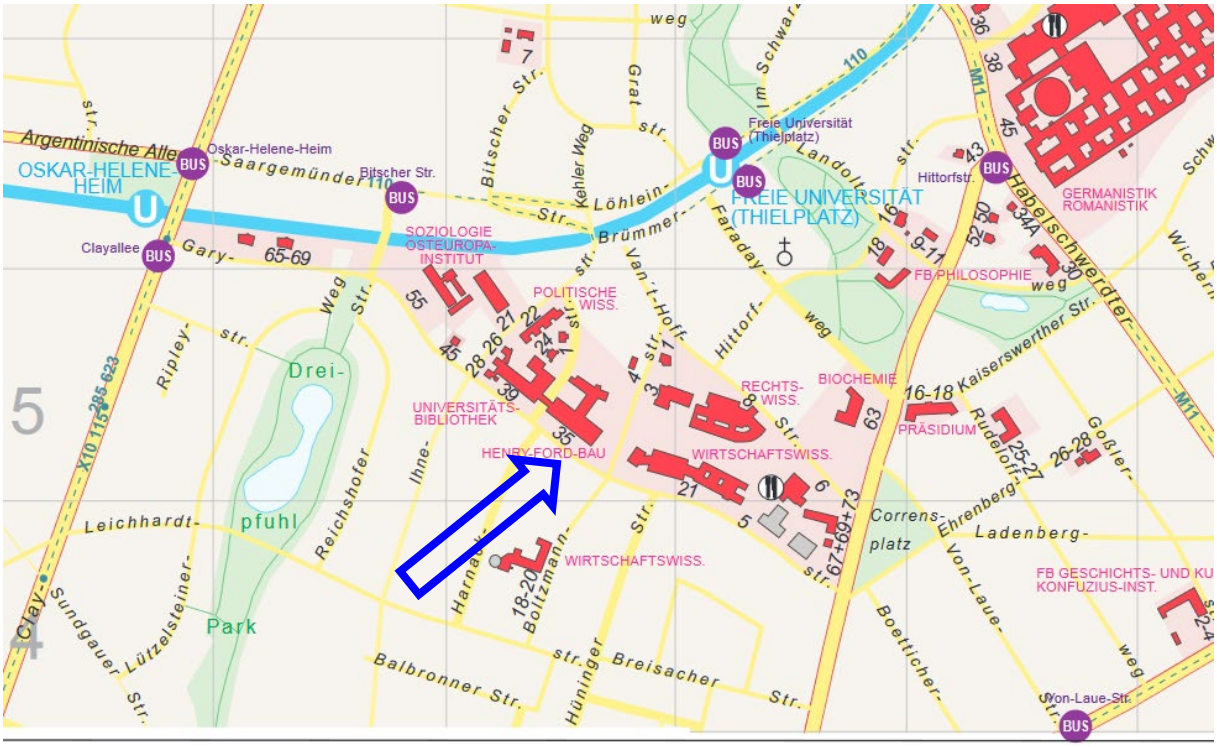

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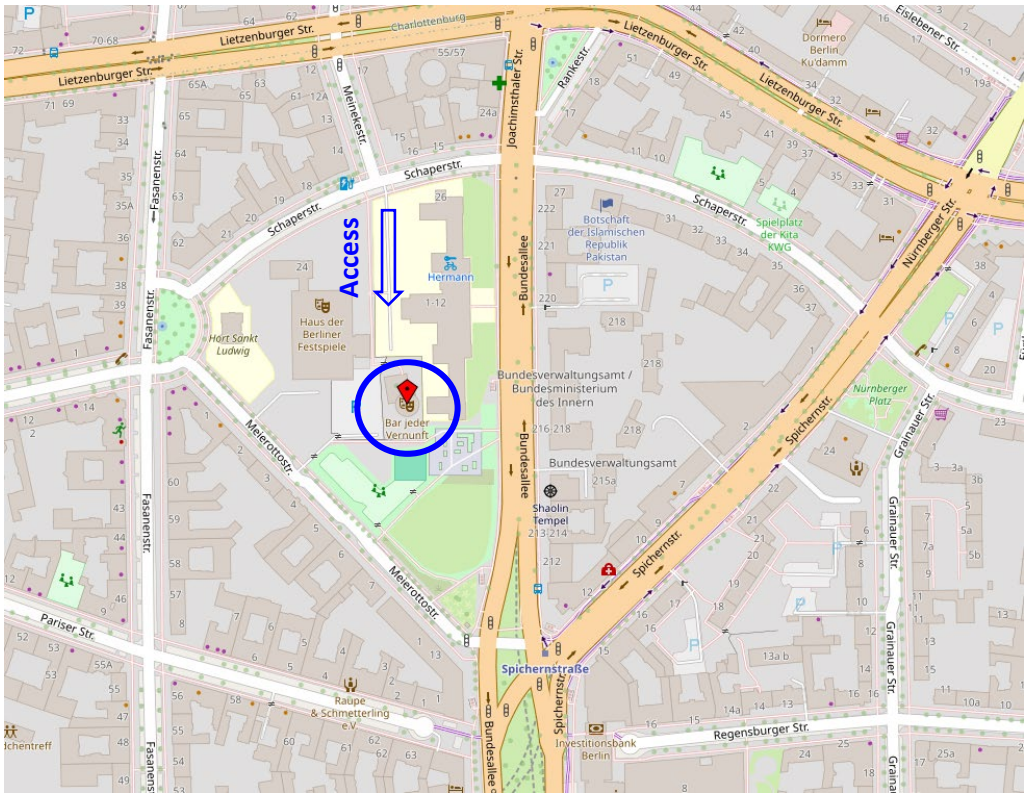
* For the non-German speakers: Note the pun in the German language – *bar jeder Vernunft* could also mean “devoid of all reason” 😉



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Program Overview

time	**Sept 2, 2024 Monday**	**Sept 3, 2024 Tuesday**	**Sept 4, 2024 Wednesday**	**Sept 5, 2024 Thursday**	**Sept 6, 2024 Friday**
08:30	*Registration*	*Registration*			
09:00	Stéphane Mangin (Tutorial)	Dmytro Afanasiev (Tutorial)	Martin Aeschlimann (Tutorial)	Bert Koopmans (Tutorial)	Sangeeta Sharma (Tutorial)
10:00	Eva Díaz	Jasmin Jarecki	Yongshan Liu	Gyung-Min Choi	MengXing Na
10:20	Martin Hennecke	Oksana Chubykalo-Fesenko	Mrudul Muraleedharan S		Tommaso Pincelli
10:40	Coffee break	Coffee break	Coffee break	Coffee break	Coffee break
11:20	Johan Mentink	Carl Davies	Urs Staub	Roopali Kukreja	Maciej Dąbrowski
12:00	Boyu Zhang	Karel Carva	Ulrich Nowak	Peter Kubaščík	Stephan Wust
12:20	Petr Nemec	Yuichi Saito		Deeksha Gupta	Luca Felipe Haag
12:40	Lunch	Lunch	Lunch	Lunch	Mirko Cinchetti
14:00	Kihiro Yamada	Andrea Eschenlohr	Tom Seifert	Georg Woltersdorf	13:20 *Closing*
14:40	José M. Lendínez	Dinar Khusyainov	Viktoriia Radovskaia	Jean-Yves Chauleau	
15:00	Chen Xiao	Lukas Körber	Hans Christian Schneider	Reza Rouzegar	
15:20	Coffee break	Coffee break	Coffee break	Coffee break	
16:00	Sergiu Ruta	Matthias Riepp	Nele Thielemann-Kühn	Anya Grafov	
16:20	Jakob Walowski	Daniel Schick		Matthijs Jansen	
16:40	Tomasz Zalewski	Dominik Juraschek	Laurenz Rettig	Chiara Ciccarelli	
17:00	Break		Alberto Anadón		
17:20	Thom Janssen	Start – 17:30		Start – 17:30	
17:40	Clemens von Korff Schmising	Poster session I		Poster session II	
18:20		Food & Drinks	Start – 18:30	Food & Drinks	
			Conference dinner		

Daily schedule

MONDAY Sept. 2, 2024

	08:55	Martin Weinelt	*Freie Univ. Berlin, Germany*	Conference Opening
Chair: *Martin Weinelt*	09:00	Stéphane Mangin	*Université de Lorraine, France*	Ultrafast all optical switching in spintronic devices – *(Invited Tutorial)*
	10:00	Eva Díaz	*Université de Lorraine, France*	Energy-efficiency of spin-orbit torque switching in the picosecond regime
	10:20	Martin Hennecke	*Max-Born-Inst. Berlin, Germany*	Depth-resolved dynamics of ultrafast all-optical switching
	10:40 – 11:20		*Coffee break*	
Chair: *Piet Brouwer*	11:20	Johan Mentink	*Radboud Univ., Netherlands*	Understanding ultrafast nanomagnetism localized in space, fluctuating in time – *(Invited)*
	12:00	Boyu Zhang	*Beihang Univ., Beijing, China*	Single-shot laser-induced switching of an exchange biased antiferromagnet
	12:20	Petr Nemec	*Charles Univ., Czech Republic*	Ultrafast modification of spin ordering in non-collinear antiferromagnetic metal Mn_3NiN by laser-polarization-dependent torque
	12:40 – 14:00		*Lunch break*	
Chair: *Andrea Eschenlohr*	14:00	Kihiro Yamada	*Tokyo Inst. of Technology, Japan*	Magnetization switching in Pt/Co/Pt multilayers by circularly polarized ultrashort optical pulses – *(Invited)*
	14:40	José M. Lendínez	*Inst. de Ciencia de Materiales de Madrid, Spain*	On the conditions for femtosecond laser-induced ultrafast toggle switching in 2D ferrimagnetic alloys
	15:00	Chen Xiao	*Beihang Univ., Beijing, China*	Ultrafast toggle switching of a ferromagnet using single-shot laser pulse
	15:20 – 16:00		*Coffee break*	
Chair: *Matias Bargheer*	16:00	Sergiu Ruta	*Sheffield Hallam Univ., UK*	Ultrafast heat-induced flows of magnons induced by femtosecond laser pulses
	16:20	Jakob Walowski	*Univ. Greifswald, Germany*	Exploring the impact of the inverse Faraday-effect on all-optical helicity-dependent magnetization switching
	16:40	Tomasz Zalewski	*Univ. Bialystok, Poland & Radboud Univ., Netherlands*	Ultrafast all-optical toggle switching of magnetization in garnets without relying on heat
	17:00 – 17:20		*Break*	

Chair: Andrei Kirilyuk	17:20	Thom Janssen	*HFML-FELIX, Radboud Univ., Netherlands*	Phononic switching in antiferromagnetic iron borate: statics and dynamics
	17:40	Clemens von Korff-Schmising	*Max-Born-Inst. Berlin, Germany*	The fundamental spatial limit of ultrafast all-optical switching probed by transient grating spectroscopy – *(Invited)*

TUESDAY Sept. 3, 2024

Chair: *Peter Oppeneer*	09:00	Dmytro Afanasiev	*Radboud Univ., Netherlands*	Light-driven ultrafast phonomagnetism – *(Invited Tutorial)*
	10:00	Jasmin Jarecki	*Univ. Potsdam & Max-Born-Inst., Berlin, Germany*	Controlling magnetization precession by heterostructure design
	10:20	Oksana Chubykalo-Fesenko	*Inst. de Ciencia de Materiales de Madrid, CSIC, Spain*	Route to minimally dissipative switching in magnets via terahertz phonon pumping
	10:40 – 11:20		*Coffee break*	
Chair: *Christine Boeglin*	11:20	Carl Davies	*HFML-FELIX, Radboud Univ., Netherlands*	Ultrafast switching of magnetization by polarized phonons – *(Invited)*
	12:00	Karel Carva	*Charles Univ., Czech Republic*	Microscopic picture of phonon induced magnetization changes
	12:20	Yuichi Saito	*Lancaster Univ., UK*	Antiferromagnetic spin dynamics induced by optical phonons in $FeBO_3$
	12:40 – 14:00		*Lunch break*	
Chair: *Georg Schmidt*	14:00	Andrea Eschenlohr	*Univ. Duisburg-Essen, Germany*	Insight into non-equilibrium states of materials with strong electronic correlations or spin-phonon coupling through femtosecond time-resolved x-ray absorption spectroscopy – *(Invited)*
	14:40	Dinar Khusyainov	*Radboud Univ., Netherlands*	Evolution of laser-induced nanoscale stochastic domains in Pt/Co/Pt trilayers
	15:00	Lukas Körber	*Helmholtz-Zentr. Dresden, Germany & Radboud Univ. Netherlands*	Self-induced Floquet states generated by nonlinear magnon interactions in a magnetic soliton
	15:20 – 16:00		*Coffee break*	

Chair: *Wolfgang Kuch*	16:00	Matthias Riepp	*Sorbonne Université & Université de Strasbourg, France; DESY, Germany*	Terahertz-driven coherent spin dynamics in labyrinth-type domain networks
	16:20	Daniel Schick	*Max-Born-Inst. Berlin, Germany*	Picosecond dynamics of magnetic domains in FeGd multilayers probed by laser-driven resonant soft-X-ray scattering
	16:40	Dominik Juraschek	*Tel Aviv Univ., Israel*	Chiral phononics: Controlling magnetism with a twist – *(Invited)*
	17:30 – 21:00		Poster Session 1	

WEDNESDAY Sept. 4, 2024

Chair: *Wolfgang Hübner*	09:00	Martin Aeschlimann	*RPTU Kaisers-lautern-Landau, Germany*	Expanding the horizons of angular momentum: Beyond spin – *(Invited Tutorial)*
	10:00	Yongshan Liu	*Beihang Uni., Beijing, China*	Efficient CoPt-based orbitronic terahertz emitters via orbital-to-charge conversion
	10:20	Mrudul Muraleedharan S	*Uppsala Univ., Sweden*	Ultrafast laser-induced magnetization in a two-dimensional semiconductor
	10:40 – 11:20		*Coffee break*	
Chair: *Stefan Eisebitt*	11:20	Urs Staub	*Paul Scherrer Inst., Switzerland*	Coupling phonons, magnons and orbitals: an x-ray view – (*Invited)*
	12:00	Ulrich Nowak	*Univ. Konstanz, Germany*	The role of angular momentum in ultrafast spintronics – *(Invited)*
	12:40 – 14:00		*Lunch break*	
Chair: *Carl Davies*	14:00	Tom Seifert	*Freie Univ. Berlin, Germany*	Driving and probing terahertz spin and orbital currents – *(Invited)*
	14:40	Viktoriia Radovskaia	*Radboud Univ., Netherlands*	Dynamic control of antiferromagnetic spin-wave spectrum via light-driven electronic excitation
	15:00	Hans Christian Schneider	*RPTU Kaisers-lautern-Landau, Germany*	Interplay of electron-magnon scattering and spin-orbit induced electronic spin-flip scattering in a two-band Stoner model
	15:20 – 16:00		*Coffee break*	

Chair: *Bert Koopmanns*	16:00	Nele Thielemann-Kühn	*Freie Univ. Berlin, Germany*	Optical control of the 4f electronic state in rare-earth metals – *(Invited)*
	16:20	Laurenz Rettig	*Fritz-Haber-Inst. Berlin, Germany*	Ultrafast angular momentum transfer in RKKY-coupled 4f antiferromagnets
	16:40	Alberto Anadón	*Université de Lorraine, IJL, France*	Disentangling the ultrafast demagnetization and spin accumulation in magneto-optical detection
	18:30 –		Conference Dinner at the "Bar jeder Vernunft"	

THURSDAY Sept. 5, 2024

Chair: *Stephane Mangin*	09:00	Bert Koopmans	*Eindhoven Univ. of Technology, Netherlands*	Ultrafast spintronics – *(Invited Tutorial)*
	10:00	Gyung-Min Choi	*Sungkyunkwan Univ., Seoul, South Korea*	Spin-torque-driven ultrafast dynamics of noncollinear antiferromagnet Mn_3Sn – *(Invited)*
	10:40 – 11:20		*Coffee break*	
Chair: *Ralph Ernstorfer*	11:20	Roopali Kukreja	*UC Davis, USA*	Unraveling optically induced ultrafast modification of nanoscale magnetic textures – *(Invited)*
	12:00	Peter Kubaščík	*Charles Univ., Czech Republic*	THz spectroscopy of spin-Hall magnetoresistance in fully metallic systems from 0.3 up to 30 THz
	12:20	Deeksha Gupta	*CNRS et Université de Strasbourg, France & Helmholtz-Zentrum Berlin, Germany*	Tuning of the ultrafast demagnetization by ultrashort spin polarized currents in multi-sublattice ferrimagnets
	12:40 – 14:00		*Lunch break*	
Chair: *Tobias Kampfrath*	14:00	Georg Woltersdorf	*Martin-Luther-Univ. Halle-Wittenberg, Germany*	Ultrafast spin transport in layer structures with MgO tunnel barriers – *(Invited)*
	14:40	Jean-Yves Chauleau	*SPEC CEA/ Saclay, France*	Terahertz antiferromagnetic dynamics induced by ultrafast spin currents
	15:00	Reza Rouzegar	*Freie Univ. Berlin, Germany*	Terahertz spin-conductance spectroscopy: probing coherent and incoherent ultrafast spin tunneling
	15:20 – 16:00		*Coffee break*	

Chair: *Clemens von Korff Schmising*	16:00	Anya Grafov	*JILA & Univ. of Colorado, USA*	Probing ultrafast spin transfer effects in Heusler alloys with extreme ultraviolet high harmonics
	16:20	Matthijs Jansen	*Univ. Göttingen, Germany*	Unraveling light-driven spin transfer and hot carrier dynamics by EUV magneto-optical spectroscopy
	16:40	Chiara Ciccarelli	*Univ. of Cambridge, UK*	Extracting spin from an antiferromagnet at picosecond timescales
	17:30 – 21:00		Poster Session 2	

FRIDAY Sept. 6, 2024

Chair: *Ulrich Nowak*	09:00	Sangeeta Sharma	*Max-Born-Inst. & Freie Univ. Berlin, Germany*	All about two dimensional materials – *(Invited Tutorial)*
	10:00	MengXing Na	*Radboud Univ., Netherlands*	Ultrafast dynamics in the ferroelectric and antiferromagnetic phase of a van der Waals multiferroic NiI_2
	10:20	Tommaso Pincelli	*Technische Univ. Berlin & Fritz-Haber-Institut Berlin, Germany*	Insights in the magnetic excitation spectrum of Fe_3GeTe_2 from femtosecond band structure dynamics
	10:40 – 11:20		*Coffee break*	
Chair: *Sangeeta Sharma*	11:20	Maciej Dąbrowski	*Univ. of Exeter, UK*	All-optical control of spins in van der Waals magnets – (Invited)
	12:00	Stephan Wust	*RPTU Kaisers-lautern-Landau, Germany*	Optical excitation of spin polarization in the altermagnet RuO_2
	12:20	Luca Felipe Haag	*RPTU Kaisers-lautern-Landau, Germany*	Optical excitation effects in altermagnetic RuO_2
	12:40	Mirko Cinchetti	*Technische Univ. Dortmund, Germany*	THz lattice and spin dynamics in a 2D antiferromagnet driven by d-d transitions – (Invited)
	13:20	Stefan Eisebitt	*Max-Born-Inst. Berlin, Germany*	Closing Remarks

Abstracts

- **Monday, 02 Sept. 2024**
- **Tuesday, 03 Sept. 2024**
- **Wednesday, 04 Sept. 2024**
- **Thursday, 05 Sept. 2024**
- **Friday, 06 Sept. 2024**

Ultra-fast all optical switching in spintronic devices

Jon Gorchon, Thomas Hauet, Michel Hehn, Julius Hohlfeld, Gregory Malinowski, Stéphane Mangin

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In 1996, Bigot et al. made a significant discovery that established the new field of ultra-fast magnetism. They demonstrated that excitation with a femtosecond laser pulse could induce sub-picosecond demagnetization of a thin Ni film [1]. However, it was not until ten years later that Theo Rasing's group in Nijmegen demonstrated complete deterministic all-optical switching (AOS) using circularly polarized laser pulses on a GdFeCo ferrimagnetic alloy [2]. This phenomenon was subsequently termed all-optical helicity-dependent switching (AO-HDS) [3], observable in a wide range of samples [4,5]. A major drawback of AO-HDS in most magnetic films is its requirement for a large number of pulses, rendering the process relatively slow [6]. Conversely, single-pulse all-optical helicity-independent switching (AO-HIS) emerges as a more promising approach for practical applications, owing to its need for just one laser pulse and significantly quicker response [7]. However, in the case of metallic samples, AO-HIS appears to be restricted to Gd-based materials (alloys and multilayers) [8-11] and MnRuGa [12]. Recently, another type of single-pulse all-optical switching has been observed, attributed to magnetization precession during anisotropy reorientation [13,14]. Despite its novelty, this all-optical precessional switching process is comparatively slow, occurring over several hundred picoseconds.

During the seminar we will examine magnetization reversal resulting from the direct interaction between the ultra-short laser pulse and the magnetization. Additionally, we will discuss how light can produce heat pulses or spin-polarized femtosecond current pulses which can then be exploited to reverse the magnetization of thin ferromagnetic films and magnetic heterostructures [15,16,17].

Part of the ultra-fast magnetism community is now focusing on deterministic magnetization switching induced by single femtosecond or picosecond laser pulses in spintronic devices, such as spin valves [18-24] and tunnel junctions [25,26]. During the talk we will show recent results on ultra-fast magnetization reversal (starting within less than one picosecond) in various ferrimagnetic and ferromagnetic spin-valve structures as shown in Figure 1, both perpendicularly magnetized [18-23] and in-plane magnetized [24]. Concerning perpendicularly magnetized systems, we have recently demonstrated optically induced ultrafast magnetization reversal occurring in less than a picosecond in rare-earth-free archetypal spin valves ([Pt/Co]/Cu/[Co/Pt]) commonly utilized for current-induced spin-transfer torque (STT) switching [23]. We discovered that the magnetization of the free layer can be switched from parallel to antiparallel alignment, akin to STT switching, revealing the presence of an unexpected, intense, and ultrafast source of opposite angular momentum in our structures.

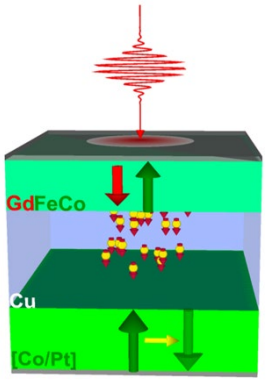

Fig.1: Sketch of a spin-valve structure used to demonstrate femto-second single pulse switching of each magnetic layer independently. The generation of femto-second spin current is shown to play a major role [17-23].

Additionnaly, we will also show results on ultrafast optical control of exchanged biased structures. Indeed, we have shown that for an exchanged biased IrMn/GdCo bilayer, not only can a single laser pulse switch the ferrimagnetic GdCo layer, but it can also change the sign of the exchange bias, demonstrating that the magnetic configuration in the antiferromagnetic IrMn layer is affected by the laser pulse [27]. These findings pave the way for ultrafast magnetization control by combining concepts from spintronics and ultrafast magnetism.

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Energy-efficiency of spin-orbit torque switching in the picosecond regime

Eva Díaz (1), Alberto Anadón (1), Pablo Olleros-Rodríguez (2), Harjinder Singh (1), Héloïse Damas (1), Paolo Perna (2), Martina Morassi (3), Aristide Lemaître (3), Michel Hehn (1), Jon Gorchon (1)

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Spin-orbit torques (SOTs) have been used to effectively control the magnetization down to the sub nanosecond (ns) regime [1]. Recently, SOT have been demonstrated in the picosecond (ps) regime with current pulses of a few ps, generated using a photoconductive switch [2, 3]. Nevertheless, many aspects about SOT at such an ultrafast scale, such as reversal mechanisms and energy requirements, remain to this date unexplored. In this work, we report the characterization of SOT switching over 7 orders of magnitude in pulse duration, from the μs down to the ps scale, in both ferromagnetic and ferrimagnetic devices of micrometric size. To achieve this feat, we have developed a technique to stretch the pulses generated with a photoconductive-switch, allowing us to bridge the pulse duration gap left between commercial pulse generators (for pulses >100 ps) and the photoconductive switches reported so-far (~6-9 ps).

We report that, for all samples, the energy consumption of SOT switching decreases by over an order of magnitude when pulse duration enters the ps regime. Moreover, we project a record-low energy cost of 9 fJ for a 100 x 100 nm2 ferrimagnetic device, lower even than the cost of switching the same sample with a femtosecond laser pulse. We have performed micromagnetic and macrospin simulations, where we correctly predict the decrease in energy consumption into the ps regime and, furthermore, reveal a transition from non-coherent domain nucleation and propagation---as observed down to the ns regime [4, 5]---into coherent magnetization reversal entering the ps regime. We speculate that coherent reversal is made energetically favorable by the ultrafast injection of spin and heat, which minimizes the losses to the lattice and substrate.

These results demonstrate that both fast and energy-efficient electrical control of magnetization is possible, opening great possibilities for application in SOT-based magnetic memories.

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Depth-resolved dynamics of ultrafast all-optical switching

Martin Hennecke (1), Daniel Schick (1), Themistoklis Sidiropoulos (1), Jun-Xiao Lin (2), Zongxia Guo (2), Grégory Malinowski (2), Michel Hehn (2), Lutz Ehrentraut (1), Matthias Schnuerer (1), Clemens von Korff Schmising (1), Stéphane Mangin (2), Stefan Eisebitt (1,3)

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The use of ultrashort light pulses in the femtosecond range allows the manipulation of magnetic order on fundamentally limiting time and length scales, inducing intriguing phenomena like ultrafast demagnetization, optically induced spin transport and all-optical magnetization switching (AOS) [1]. The ability to deterministically reverse the magnetization of a nanolayer on extremely short time and length scales makes AOS a promising approach to be exploited for ultrafast information processing and macroscopic functionality in today's opto-spintronics applications, relying on a localized optical control over the magnetic order in different layers and across the interfaces of complex magnetic heterostructures [2]. While AOS has so far been observed in a range of nanoscale transition metals and rare-earth-based alloys and multilayers, the underlying mechanisms are still under investigation [3,4].

Here, we combine temporal and spatial resolution to reveal the ultrafast evolution of AOS along the depth of a magnetic heterostructure consisting of a 10-nm-thin, in-plane magnetized, ferrimagnetic GdCo layer [5]. To this end, we perform broadband transverse magneto-optical Kerr effect (TMOKE) spectroscopy at the Gd N5,4 resonance (≈150 eV), using femtosecond soft x-ray pulses provided by a laboratory-scale light source based on high-harmonic generation [6]. Analyzing the time-resolved TMOKE data via magnetic scattering simulations [7] enables a quantitative determination of the transient magnetization depth profiles evolving within the 10 nm GdCo layer after infrared photoexcitation, with 40 fs temporal and <1 nm depth resolution. The data obtained for different excitation fluences below and above the switching threshold shows that the magnetization reversal is mediated by the transient formation and growth of a domain-like region with reversed spin direction, which starts to propagate from the surface of the magnetic layer towards its bottom. Within a few picoseconds and depending on the fluence, this region either expands over the entire depth of the magnetic thin film or relaxes back to the initial, unswitched state.

The achieved time- and depth-resolved view on the switching process provides direct insight into the microscopic mechanisms and spatial propagation of the ultrafast magnetization reversal within a magnetic thin film system, which is key to a better understanding of such phenomena in different material classes and sample geometries exhibiting AOS.

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Understanding ultrafast nanomagnetism localized in space, fluctuating in time

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Despite decades of downscaling of magnetic bits, magnetism at length scales below ~10 nanometer and time scales below ~1 nanosecond is a poorly understood regime. This regime of "ultrafast nanomagnetism" is only recently becoming accessible experimentally thanks to new magnetic imaging methods based on femtosecond x-ray and electron pulses. Interestingly, results obtained with such experimental techniques disclose counter-intuitive results for which established theories do not provide an adequate explanation.

In this talk, we focus on reviewing recent progress in ultrafast nanomagnetism from the perspective of theory and simulations. We showcase examples where atomistic spin models greatly assisted in corraborating key experimental observations concerning the ultrafast nucleation of skyrmions in ferromagnetic multilayers. For example, it was found that the nucleation proceeds by transiently passing through a topological fluctuations state, in which the dynamics is governed by exchange interactions, the strongest interaction in magnetism. This leads to picosecond nucleation times, being much faster than accessible with external fields [1].

Furthermore, to gain understanding of the mechanisms underlying ultrafast nanoscale fluctuations, we support the simulations with phenomenological modeling [2]. Specifically, we propose a model which describes topological fluctuations as a competition between nucleation and decay of toplogical textures over an energy barrier. A simple rate-equation model provides an intuitive picture of nonequilibrium topological fluctuation states and features excellent qualitative agreement with the much more advanced atomistic spin dynamics simulations. Furthermore, we show that this model can be generalized to gain understanding in helicity-dependent control of stochastic domain structures that have recently been observed using magnetic-force microscopy studies [3].

The systematic approach combining simulations and phenomenological modelling provides a new path to the development of a general theory of ultrafast nanomagnetism. This has not only great relevance to stimulate and support experiments, but is also capable of identifying conceptually new pathways to challenge the limits for the fastest, smallest and most energy-efficient writing of magnetic bits.

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Single-shot laser-induced switching of an exchange biased antiferromagnet

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Ultrafast manipulation of magnetic order has challenged the understanding of the fundamental and dynamic properties of magnetic materials. So far single-shot magnetic switching has been limited to ferrimagnetic alloys, multilayers, and designed ferromagnetic (FM) heterostructures. In FM/antiferromagnetic (AFM) bilayers, exchange bias (He) arises from the interfacial exchange coupling between the two layers and reflects the microscopic orientation of the antiferromagnet. Here the possibility of single-shot switching of the antiferromagnet (change of the sign and amplitude of He) with a single femtosecond laser pulse in IrMn/CoGd bilayers is demonstrated. The manipulation is demonstrated in a wide range of fluences for different layer thicknesses and compositions. Atomistic simulations predict ultrafast switching and recovery of the AFM magnetization on a timescale of 2 ps. The results provide the fastest and the most energy-efficient method to set the exchange bias and pave the way to potential applications for ultrafast spintronic devices.

Ultrafast modification of spin ordering in non-collinear antiferromagnetic metal Mn_3NiN by laser-polarization-dependent torque

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Recent breakthroughs in electrical detection and manipulation of antiferromagnets have opened a new avenue in the research of non-volatile spintronic devices. Antiferromagnetic (AFM) ordering leads to a reduced sensitivity to magnetic field perturbations, multi-level stability and ultra-fast spin dynamics. However, these features also make the characterization of AFM materials, in particular of thin metallic films suitable for spintronics, a major challenge [1]. One from a few available experimental approaches is to use magneto-optical (MO) effect which depends quadratically on the sublatice magnetization, the Voigt effect [1]. The applicability of pump-probe experimental technique based on Voigt effect for the investigation of magnetic anisotropy was demonstrated in compensated AFM metal CuMnAs, where antiparallel ordering of two spin sublattices is present [2]. Recently, Voigt effect was used to study quenching of AFM order also in non-collinear antiferromagnetic metal Mn_3Sn [3].

In this contribution, we report on an ultrafast modification of spin ordering in non-collinear cubic antiperovskite AFM metal Mn3NiN [4] by laser-induced torque. We show that linearly-polarized femtosecond pump pulses generate a torque on spins, with a torque direction controlled by the pump linear polarization orientation. We interpret the torque as a result of selective excitations of Mn electronic spin states, with the excitation strength (calculated by first-principles methods) dependent on the angle between the pump polarization orientation and the direction of atom-specific vectors, reflecting the local uniaxial anisotropy. For all three spins, the torque is directed within the triangular spin domain plane. Nevertheless, the probe-detected MO Voigt effect is dominated by the out-of-domain-plane spin tilt due to the resulting spin dynamics, which we modelled by LLG equation. Moreover, the experimentally measured MO signal is a result of the signal averaging from eight possible magnetic domain arrangements, corresponding to the Γ4g spin orientation being located in one of eight (111) planes.

Magnetization switching in Pt/Co/Pt multilayers by circularly polarized ultrashort optical pulses

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For thin magnetic metals, the magnetization is switchable by circularly polarized ultrashort laser pulses, referred to as all-optical helicity-dependent switching (AO-HDS). AO-HDS was first demonstrated in ferrimagnetic GdFeCo1 and later in ferromagnetic metals, including multilayered Co/Pt and Co/Ni stacks and granular FePt medium2. Because state-of-the-art magnetic hard-disc drives use local heating by laser illuminations3, the additional use of the optical helicity should be beneficial for further decreasing the energy consumption of writing a magnetic bit. However, the path toward the application is steep for difficulties, such as the slow dynamics, the necessity of multiple optical pulses, etc. Furthermore, it remains an outstanding issue that the dominant mechanism is unclear, i.e., which thermal or nonthermal effects dominate the AO-HDS.

In this presentation, I would like to present our recent progress in studies of the AO-HDS in ferromagnetic Pt/Co/Pt multilayers that are most typical for AO-HDS. The main driving force of the AO-HDS by multiple fs pulses was explored in the near-infrared to the visible spectral range. The helicity-dependent laser absorption by the magnetic circular dichroism predominantly accounts for the monotonic increase of the switching efficiency with increasing the wavelength from 0.5 μm to 1.2 μm4. We also found that a single pair of a fs linearly polarized pulse and a ps circularly polarized pulse with a time separation of a few picoseconds enables us to deterministically switch the spins in a helicity-dependent way on a picosecond time scale5. In the middle of these experiments, we were inspired to switch the magnetization by “x-ray” magnetic circular dichroism resulting from the core-to-valence electric-dipole transitions. Using the cutting-edge Japanese x-ray free electron laser SACLA, we discovered fs circularly polarized x-ray pulses control the magnetization when the photon energy is set at the absorption edge of Pt.

These projects were conducted in collaboration with Th. Rasing group (Radboud University), A. V. Kimel group (Radboud University), A. Kirilyuk group (Radboud University), T. Ono group (Kyoto University). M. Suzuki (Kwansei Gakuin University), Y. Tanaka group (University of Hyogo), I. Matsuda group (University of Tokyo), R. W. Chantrell group (University of York), and Advanced Light Source and Optics Research Group, XFEL Utilization Division (SACLA).

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On the conditions for femtosecond laser-induced ultrafast toggle switching in 2D ferrimagnetic alloys

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The Mermin-Wagner theorem establishes that long-range magnetic order is prohibited in two-dimensional (2D) isotropic magnets with short-range interactions due to the effect of thermal fluctuations. Nevertheless, magnetically stable 2D van der Waals (vdW) materials have been recently discovered, where magnetic anisotropy is crucial to stabilize magnetic order [1]. These materials offer the opportunity to explore fundamental questions so far unexplored. A prominent example is the field-free, all-optical ultrafast magnetization switching, which has been experimentally demonstrated in ferrimagnetic GdFe alloys. Despite recent efforts to understand the mechanisms underlying this effect in bulk ferrimagnetic alloys [2], the implications of the dimensional effects in 2D systems are still undetermined.

In this study, we explore the impact of reduced dimensionality on the ultrafast magnetization switching effect in 2D ferrimagnets by using atomistic spin dynamics (ASD) simulations. We note that ASD techniques account for thermal fluctuations, which considerably influence the magnetic behaviour of 2D materials [3]. With this approach, we aim to understand how the magnetic anisotropy affects the switching phenomenon in 2D structures compared to bulk systems.

Firstly, our results show that while for 3D systems the critical temperature linearly depends on the anisotropy value, for 2D structures this dependence is logarithmic. Secondly, we find that the temperature dependence of the magnetic order relaxation time upon a sudden temperature increase for a 3D system slightly depends on the anisotropy value. In contrast, for a 2D system, the behaviour as a function of the anisotropy value is much richer. Finally, we demonstrate that femtosecond laser pulse-induced toggle switching in 2D ferrimagnets is possible. However, unlike for 3D systems, for which the anisotropy value barely influences the switching process, for 2D systems the switching is only possible for relatively high anisotropy values.

Our findings open the door to the understanding the influence of dimensionality on the fundamental mechanisms of laser-induced ultrafast toggle switching. Furthermore, our results encourage the search for novel 2D ferrimagnetic materials with increased magnetic anisotropy, making them potential candidates to exhibit this effect.

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Ultrafast toggle switching of a ferromagnet using single-shot laser pulse

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Laser pulses are proved to be the fastest-ever way to manipulate magnetization, enabling the instantaneous transition of magnetic states on femtosecond to picosecond timescales[1]. The single-shot optical writing of spintronic devices holds great promise for advancing high-speed spin memory and logic applications. Over the past decade, the mechanism of all-optical switching (AOS) has been extensively studied. To date, the phenomenon of single-pulse switching has been observed exclusively in ferrimagnets based on rare-earth transition metals, with Gd being particularly prominent [2]. The substantially different dynamics of two sublattices in these ferrimagnets have been identified for achieving single-shot AOS [3,4], while ferromagnets, with only one sublattice, present a formidable challenge for single-pulse switching. In this study, we introduce for the first time the ultrafast toggled switching of ferromagnets using a femtosecond laser pulse. Here, we demonstrate efficient optical control and toggle switching of X/CoFeB/MgO heterostructures, which exhibit strong perpendicular magnetic anisotropy. The switching behaviors are quantitively investigated through a combination of optical writing and electrical reading techniques. The resulting phase diagrams reveal a competition between the external in-plane magnetic field and laser fluence. The energy fluence required for this process is calculated to be as low as 1.1 mJ/cm², with a high switching ratio exceeding 95%. Notably, single-shot all-optical switching has also been achieved in Ti/CoFeB/MgO heterostructure by replacing the in-plane field with tilted anisotropy. Further analysis indicates that the switching mechanism is associated with the ultrafast loss of anisotropy in the CoFeB/MgO layer, similar to the thermal anisotropy torque predicted previously [5]. Above all, we have studied femtosecond single-shot optical magnetization switching in a widely used ferromagnet, uncovering a regime distinct from conventional ferrimagnets. This work paves the way for breaking the speed limits of magnetic tunneling junctions and accelerates the application of ultrafast opto-magnetism.

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Ultrafast heat-induced flows of magnons induced by femtosecond laser pulses

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Understanding the fundamental physics of energy transfer in spin textures via magnon flows and domain wall motion are important for technological application. In the research community controlling magnetisation dynamics using femtosecond laser pulse excitation (all-optical switching) has been intensely investigated as an energy-efficient method. The ferrimagnet, GdFeCo, is a material known to undergo ultrafast switching within 1-2ps after excitation with femtosecond laser pulses.

In this work, we present via numerical methods the presence of ultrafast pure magnonic currents at sub-picosecond time scale due to local spin temperature imbalances. The magnons propagate ballistically and have acritical role in deterministic switching of the bilayer systems and ferrimagnetic alloys, but on a localised level. During the ultrafast laser pulse, the electrons heat up rapidly and cool down in a few ps, leading to a change in the spin system configuration, which can be described by the spin temperature.

To quantity the magnon propagation we constructed a bilayer system with different spin temperatures. The high-energy magnons from the hot regions will cross the interface and propagate into the cold region. The propagation is visualised where the high energy magnons are formed at the interface and then propagate into the cold region, with a speed of propagation in the order of km/s. In addition, we will show how spin wave calculation can be used to indicate the potential of magnons modes available in each of the layers and the impact on magnons transport across the interface.

The very localised nature of the mechanism is promising for utilization in nanoscale devices. This could lead to a new design of systems that undergo all-optical switching. In particular, combining structures with different functionalities will lead to improved overall performance. The results indicate that using chemical inhomogeneities can expand the rare-earth concentration range for deterministic all-optical switching by using the interface magnon currents to compensate the energy required for bridging the FM-AFM states.

Exploring the impact of the inverse Faraday-effect on all-optical helicity-dependent magnetization switching

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The fastest deterministic data recording technique solely using ultrashort laser pulses is all-optical helicity-dependent magnetization switching (AO-HDS). Granular high data density magnetic storage media developed for heat-assisted magnetic recording (HAMR) provide an ideal playground for investigating the interplay of effects conducting magnetization switching. In the latest perception, we identify two effects, the magnetic circular dichroism (MCD) and the inverse Faraday effect (IFE), as the forces driving the switching process. During photon absorption, which leads to a rapid temperature rise and thus to magnetization quenching, the MCD ensures two distinct electron temperatures due to helicity-dependent absorption. This effect already holds a nonvanishing probability for magnetization switching by generating the right amount of spin noise for each spin channel. At the same time, the IFE induces a magnetic moment within the material, enhancing the switching probability. We present AO-HDS experiments using ultrashort laser pulses (< 200 fs) in the near-infrared range from 800 nm to 1500 nm.

The investigations demonstrate a strong dependence of the switching efficiency on the absorbed energy density elevating the electron temperature close to the Curie point and allowing for the IFE to take full effect by inducing a magnetic moment for deterministic switching while in the quenched magnetization state. On the one hand, we do not observe enhanced switching from an increased MCD. On the other hand, a higher induced magnetization usually improves the switching rate if the electron temperatures reach at least the transition temperature vicinity. From this, we conclude that electron temperatures around the Curie point are crucial, making a necessary condition for switching if a slight temperature difference between spin states exists. However, the magnetic moment generated by the IFE is required within the switching process to provide a distinct deterministic character and improve the switching efficiency over a wide temperature range. In practical terms, a stronger absorption leads to a higher induced magnetic moment and magnetization switching at lower applied laser fluences.

Ultrafast all-optical toggle switching of magnetization in garnets without relying on heat

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The search for new mechanisms and possibilities for ultrafast all-optical switching of magnetization is a widely researched topic, motivated both by its application potential and its value for fundamental research on light-matter interactions. To date, ultrafast switching has been demonstrated in many materials, allowing for either deterministic control, where switching can be governed by light polarization, or in a nondeterministic manner, where toggle switching based on laser-induced heating mechanisms in metals has been demonstrated [1].

One of the most intriguing examples of all-optical switching is the least-dissipative mechanism of coherent ultrafast photo-magnetic switching demonstrated in iron-cobalt garnet dielectrics (YIG:Co) [2]. In this case, the YIG:Co was tilted from the surface normal to its substrate (GGG) by a 4° miscut, introducing a non-equilibrium of easy magnetization axes and degeneracy between magnetic states. Therefore, a single linearly polarized pulse was capable of switching magnetization and the subsequent laser pulse with orthogonal linear polarization could then switch the magnetization back.

Here, we demonstrate that the regime of ultrafast toggle switching can also be realized via a mechanism without relying on any heat. By using a YIG:Co sample with a similar composition but without miscut and with pure cubic symmetry, we show the ability to toggle magnetization between two stable magnetic states. Every incident femtosecond pump pulse with a wavelength of 1300 nm and set liner polarization toggles the magnetization state. Therefore, the final magnetization state depends on the initial magnetization state and the number of incident linearly polarized pulses. Thus, an even number of incident pulses leaves the same magnetic state, and an odd number of pulses reverses it. In contrast to laser-heat-induced magnetization reversal, this new regime of ‘cold’ toggle switching can be observed in ferrimagnets without the compensation point and in an exceptionally broad temperature range (200-450 K) [3].

Moreover, by employing the double pumping scheme, we show that forward and backward magnetization switching is achievable in two distinctive scenarios: using identical linearly polarized laser pulses and with pulses having orthogonal polarization planes [4].

Conducting measurements as a function of temperature allowed us to examine the influence of the magnetocrystalline contribution to the total magnetic anisotropy, which defines the energy landscape and the trajectories of magnetization movement leading to switching. Through the control of magnetic anisotropy, the toggle switching can be accompanied by lower dissipation compared to the mechanism of laser-induced heating, but the dissipation and the switching time are shown to be competing parameters.

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Phononic switching in antiferromagnetic iron borate: statics and dynamics

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Using the crystalline lattice to manipulate magnetic ordering represents an intriguing direction of future research, particularly when attempting to modify the rather robust magnetization of an antiferromagnet (AFM). The impervious nature of AFMs, however, makes it very difficult if not impossible to directly manipulate their magnetization with stray magnetic fields. Alternatively, all-optical mechanisms - based on the use of ultrashort laser pulses - can interface directly with antiferromagnetic ordering while arguably offering the fastest possible speeds [1]. While full-scale all-optical switching of order within an AFM has not yet been discovered [2], the direct excitation of THz optical phonon modes at resonance has been clearly shown to be capable of temporarily modifying order in a variety of other systems [3,4]. In particular, it was recently demonstrated that the magnetization of iron-garnet films can even be permanently switched by illuminating the ferrimagnet with infrared (IR) optical pulses tuned in frequency to match those of longitudinal optical phonon modes [5]. The central role of strain in the process was unambiguously confirmed by the highly non-trivial spatial distribution of the switched magnetization. From this, an intriguing question arises: how does this mechanism operate and manifest in an AFM?

Here, we study how magnetization can be all-optically rotated in the canted antiferromagnet iron borate, utilizing mid- to far-infrared ultrashort light pulses. We discover that this excitation gives rise to a peculiar pattern of metastable magnetic domains with a tunable symmetry, which can be qualitatively understood by considering how light-induced radial strain interacts with the Néel vector [6]. Further, we investigate the dynamics of reversal process in details. We apply single-shot time-resolved pump-probe microscopy technique, using intense IR pulses from a free-electron laser for excitation. We observe the formation of the pump-induced magnetic switching as a function of pump wavelength, intensity, and duration, as well as strength and direction of the applied magnetic field. On a nanosecond time scale, we observe a rapid motion of the reversed domains in absence of any magnetic field, but with velocities of several km/s. We finally discuss the specifics brought by the AFM order into the reversal process.

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The fundamental spatial limit of ultrafast all-optical switching probed by transient grating spectroscopy

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In order to establish all-optical techniques in future computing, it is crucial to understand the processes that define the fundamental spatial limits of ultrafast all-optical switching (AOS). According to the current understanding of AOS, the key to achieving magnetization reversal upon laser excitation is to confine sufficient energy within the electronic system for a characteristic time scale before it is locally transferred to the lattice or, in the case of nanoscale excitation gradients, lost by ultrafast lateral energy transport. Therefore, we need experimental studies that provide direct access to lateral energy transport and its effects on AOS with sub-10-nanometer spatial and sub-picosecond temporal resolution. An elegant approach is the Fraunhofer diffraction of transient gratings induced by interference of nanometer wavelength radiation.

In this contribution, we present transient grating spectroscopy performed at the free-electron laser FERMI in Trieste, Italy. We have imprinted excitation gratings in a GdFe alloy with periodicities between 87 and 17 nm by interference of two coherent femtosecond light pulses with photon energies between 30 and 150 eV. The subsequent ultrafast evolution of the magnetization pattern was probed by diffraction of a third, time-delayed pulse tuned to the Gd N-edge at a wavelength of 8.3 nm (150 eV). In a first experiment we studied the simultaneously recorded first- and second-order diffraction and, by performing real-space reference measurements with a wide-field magneto-optical microscope, conclusively demonstrated the ultrafast emergence of all-optical switching on an 87 nanometer length scale [1].

In a follow-up experiment, we were able to follow the ultrafast dynamics of magnetization gratings with systematically decreasing periodicities down to only 17 nm, i.e. with excitation gradients approaching the mean free path of the excited electrons. Combining our experimental data with atomistic spin dynamics and microscopic temperature model simulations, we extracted information about the spatio-temporal evolution of the electronic temperature and predicted a phase diagram for AOS for nanometer-scale periodic excitations. Our results suggest that the minimum size for AOS in GdFe alloys induced by nanoscale periodic excitation is around 25 nm, and that this limit is governed by ultrafast lateral electron diffusion and the threshold for optical damage.

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Light-driven ultrafast phonomagnetism

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Light in the form of ultrashort pulses has enabled control of magnetism at the ultimate time and length scales, where traditional magnetic field excitation fails. This approach has particularly advanced the control of antiferromagnets — materials without net magnetization and typically insensitive to magnetic fields.

In this tutorial, I will review recent breakthroughs in the optical control of magnetism, focusing on the resonant driving of elementary lattice excitations, known as phonons. Although phonons are typically associated with heating, which destroys magnetism, recent experiments have demonstrated that light-driven phonons can control and even induce magnetic order.

I will demonstrate how light-driven phonons can cause net distortions in the crystalline lattice, generating effective phonomagnetic fields and enabling controllable switching between different anti- and ferromagnetic phases.

Controlling magnetization precession by heterostructure design

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Magnetization precession is another effect in magnetic materials that holds technological potential, as it could enable energy-efficient magnetization switching with minimal heat deposition. Excitation mechanisms of magnetization precession have been studied individually in many different systems but rarely examined all together and compared regarding their efficiency in driving the precessional motion. In typical ultrafast experiments heat and strain is excited simultaneously and a separate evaluation of these effects is rather difficult. Nanoscale heterostructure design can control laser-induced heat dissipation and strain propagation, and therefore also their contribution and efficiency for exciting magnetization precession.

Here, we incorporate MgO layers at different positions into the experimental platform of metallic Pt(5 nm)-Cu(100 nm)-Ni(20 nm) heterostructures to block the propagation of hot electrons and thus vary the thermal load and strain profile in the buried Ni layer in three variations of the platform. We use a combination of time-resolved magneto-optical Kerr effect (MOKE) measurements in polar MOKE geometry to observe the out-of-plane magnetization as well as complementary ultrafast x-ray diffraction (UXRD) experiments, that show the capability of our platform to tailor the spatio-temporal shape of the transient heat and strain. This experimental approach provides a complete characterization of the time-dependent quantities determining the effective field changes inducing the magnetization precession. We model the observed magnetization dynamics based on the Landau-Lifshitz-Gilbert equation and use the calibrated temperature and strain as inputs to fully describe the response of the magnetic Ni layer, which allows us to disentangle the individual contributions of the driving effects to the excitation of the magnetization precession.

Our results uniquely highlight the role of quasi-static strain as a driver of precession, when the magnetic material is rapidly heated via electrons. The effective magnetic field change originating from demagnetization partially compensates the change induced by quasi-static strain. The strain pulses can be shaped via the nanoscale heterostructure design to efficiently drive the precession, paving the way for opto-magneto-acoustic devices with low heat energy deposited in the magnetic layer.

Route to minimally dissipative switching in magnets via terahertz phonon pumping

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One of the fundamental questions in ultrafast magnetism is understanding the angular momentum transfer between spins and lattice, with recent results demonstrating possibilities to excite spin dynamics via phonon excitation [1]. In this work we will present our recent modeling of atomistic spin-lattice dynamics simulations [2]. which produces a reliable angular momentum transfer from spins to lattice and vice versus.

Within our model we demonstrate that efficient coherent magnetisation switching of an insulating magnet can be triggered by a THz pulse by excitation of THz phonons [3]. We find that switching is driven by excitation near the P-point of the phonon spectrum in conditions where spins typically cannot be excited and when manifold k phonon modes are accessible at the same frequency. The mechanism of switching is via local magneto-elastic fields created by atom's displacements. In the conditions of the absence of spinwave excitations, practically all phonon angular momentum is transferred to a precessional magnetisation switching. The spin temperature calculated during the switching process shows a minimum increase (in the order of mK), Our model determines the necessary ingredients for low-dissipative switching and provides new insight into THz-excited spin dynamics with a route to energy efficient ultrafast devices.

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Ultrafast switching of magnetization by polarized phonons

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Over the last few decades, breakthrough experiments have demonstrated that ultrashort laser pulses have the capacity to both temporarily manipulate and permanently switch magnetic ordering [1]. A variety of different mechanisms can facilitate all-optical switching of magnetization, relying for example on ultrafast exchange-driven demagnetization [2] or the excitation of electronic transitions at resonance in dielectric iron-garnet films [3].

Spin-lattice coupling plays a central role in the research fields of magnetism and spintronics. Very recently, it has been demonstrated that ultrafast demagnetization involves the transfer of angular momentum from the spins to the lattice [4]. Reciprocally, it is also feasible for vibrations of the lattice (phonons) to modulate magnetic interactions such as exchange [5]. Going further, it is even possible for one to drive at resonance circularly-polarized optical phonons – the angular momentum of the latter can give rise to emergent magnetization [6] and large effective magnetic fields [7].

Here, I will discuss our experimental measurements studying how we can create and manipulate magnetization by selectively driving optical phonons at resonance. To achieve this, we use ultra-narrowband optical pulses in the infrared spectral range as delivered by the free-electron lasers at the FELIX facility. Upon, for example, irradiating magnetic heterostructures mounted atop paramagnetic substrates with circularly-polarized infrared pulses, we find that the magnetic ordering can be switched only when the pulse drives at resonance doubly-degenerate optical phonons within the substrate [8]. The direction of switching can be controlled by adjusting the helicity of the incident pulses. So far, we have identified this mechanism in structures grown on glass-ceramic, c-cut sapphire and zinc oxide substrates. The spectral dependence of the helicity-dependent switching follows the population of optical phonons found in the different substrates.

In summary, our results demonstrate that optical phonons can serve as an efficient and potentially universal lever for controlling magnetic ordering across ultrafast timescales.

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Microscopic picture of phonon induced magnetization changes

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Strong photon pumping can drive matter to unique transient states otherwise inaccessible in equilibrium conditions and even change its magnetic properties. Especially interesting is the case of soft polar phonons coherently excited by the photon pulse into a nonlinear regime. If these phonons are strongly coupled to the spin system, this may lead to a change of magnetic order. Phonons excited in a controlled way have indeed been shown to modify magnetic interactions so that the system turns from a collinear antiferromagnet to a canted weak ferromagnet [1]. We investigate microscopic aspects of nonlinear spin-phonon interactions leading to magnetic order modifications. We have evaluated magnetic exchange interactions under the influence of specific non-equilibrium phonon populations for prospective systems, employing first principles calculation methods together with the frozen phonon method.

Circularly polarized phonons are capable of modifying or reorienting magnetic moments while the resulting magnetization direction can be controlled by the pulse helicity. These phonons have recently been shown to create transient magnetization in paramagnetic CeF3 [2], and also reorient magnetization of a ferrimagnet in a heterostrucure [3]. The magnetic field due to chiral phonons has been ascribed to an effective coupling between the phonon angular momentum and the electron magnetic momentum [4]. The magnetization change caused by the interaction with circularly polarized phonons exhibits some similarity to that originating from circularly polarized light via the inverse Faraday effect [5]. We evaluate the spin and orbital contribution to magnetization induced in the presence of circular ionic motion, and extract the associated coupling described above on the level of first-principles description. This allows us to predict the induced magnetization values for different materials, and study which properties affect it.

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Antiferromagnetic spin dynamics induced by optical phonons in FeBO3

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Phono-magnetism is a new approach to manipulating magnetic order via direct lattice excitation, based on the strong spin-phonon coupling. Conventionally, a femtosecond laser pulse in the visible or near-infrared spectral region has been used for ultrafast demagnetization and driving coherent spin precession. In this case, most energy is dissipated into the lattice because the laser photon energy (over 1 eV) is three orders of magnitude larger than the intrinsic magnetic energy scale defined by exchange interaction (10 meV). The energy transfer from photons to spins is extremely inefficient.

In contrast, an ultrashort pulse in the mid-infrared region allows us to directly drive ionic lattice vibrations, avoiding hot carrier excitation. Therefore, the lower photon energy of mid-infrared (1-100 meV) is a promising excitation source to control the magnetic state more efficiently.

Here, we report the coherent spin dynamics assisted by phonons in a canted-antiferromagnet: FeBO3. This iron oxide has a relatively simpler unit cell with the number of phonons smaller than in the materials studied before. Thus, each optical phonon is well-isolated in the spectrum, which is an ideal property for exciting only single phonon mode selectively using the tunable mid-infrared light source with a 100 fs pulse duration.

We demonstrate that by pumping FeBO3 with a linearly polarized pulse around 8 μm, which is in resonance with the antisymmetric stretching phonon mode, the large amplitude coherent antiferromagnetic resonance mode is excited. We attribute the excitation mechanism to the phononic inverse Cotton-Mouton effect, eliminating other possibilities of thermal origin like hot carrier generation, or thermal expansion. Then, by tuning the pump wavelength, we show that the maximum amplitude is reached when pumping at the wavelength of 10 μm, which is shifted from the central wavelength of the target ionic vibration mode. Our theoretical modelling indicates that this red shift is an intrinsic property of the phono-magnetic effect. Besides this important conclusion, we also achieved the first demonstration of optically phonon-induced coherent spin dynamics at room temperature, making the case for practical applications of phono-magnetism much stronger.

Insight into non-equilibrium states of materials with strong electronic correlations or spin-phonon coupling through femtosecond time-resolved x-ray absorption spectroscopy

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The excitation of solid materials with femtosecond laser pulses generates non-equilibrium states that are interesting for both fundamental and application-related reasons, as their properties can be quite different from those generated by e.g. thermal excitation or chemical doping. However, once several degrees of freedom couple strongly and/or electronic correlations are prominent, simple descriptions via e.g. rate models become insufficient to describe ultrafast dynamics. I will discuss for three recent examples how x-ray absorption spectroscopy with femtosecond time resolution, in conjunction with theoretical calculations, is able to reveal and characterize non-equilibrium states by analyzing the transient electronic structure and spin state through characteristic, time-dependent spectral features: The interplay of local electronic correlations with ultrafast demagnetization in ferromagnetic Ni [1], photodoping of the charge transfer insulator NiO [2] and ultrafast spin-state switching at room temperature of spin-crossover molecules condensed in thin films [3].

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Evolution of laser-induced nanoscale stochastic domains in Pt/Co/Pt trilayers

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The switching of magnetization with the help of ultrashort laser pulses, also known as all-optical switching or AOS, is an intriguing subject with potential implications for future data storage and information processing technology. Ferromagnets are vital for writing magnetic bits, but understanding AOS in ferromagnets is challenging [1–4]. After the discovery of helicity-dependent AOS in ferromagnetic multilayers [5], it was realized that, unlike in ferrimagnets, where switching can be achieved with a single laser pulse [6], switching ferromagnets requires a sequence of pulses. The consequence of the cumulative nature of AOS in ferromagnets is the occurrence of processes manifesting themselves across different length and time scales, ranging from nano- to millimeters and pico- to milliseconds. These processes may involve magnetic circular dichroism, the spin-Seebeck and inverse Faraday effects, magneto-dipolar interactions, etc. Moreover, femtosecond experiments have shown that laser irradiation can lead to the formation of nanoscale topological spin textures [7]. Understanding the interplay of these mechanisms requires novel techniques that are able to resolve nanoscale magnetization dynamics beyond conventional magneto-optical imaging.

In this work, we employ in-situ magnetic force microscopy (MFM) to study the AOS of Pt/Co/Pt trilayers. Our measurements reveal that the first few laser pulses trigger the emergence of a stochastic magnetic domain network (SDN), of which the smallest characteristic size is ~200 nm. By varying the number of pulses, laser fluence, and light helicity, we observe a change in the nucleation behavior from a stochastic process to a deterministic one. Interestingly, we observe that the stochastic domain networks do not grow via the temperature gradient created with circularly polarized laser pulses, as was suggested before1. Still, instead, they either grow in number to homogeneously fill the irradiated space or shrink and annihilate. To explain our experimental results, we create a phenomenological macrospin model that includes local exchange as well as MCD in the calculation of switching probability (contribution by R. Liefferink et al.). We find that the complexity of the domain wall in the SDN regime is key to enhancing MCD-derived domain growth and eventual transition from SDN formation to deterministic switching.

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Self-induced Floquet states generated by nonlinear magnon interactions in a magnetic soliton

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Floquet engineering has emerged as an effective tool to tune the quantum properties of various systems in condensed matter physics. In particular, periodic driving of electronic systems has been proven to significantly alter band structures and spin interactions. In this study, we show how Floquet physics can be enriched by the intrinsic nonlinearities of magnetic systems, allowing for self-induced Floquet states without resonant low-frequency drive. In a combined experimental and theoretical study, we investigate the emergent Floquet physics in a magnetic soliton, particularly in magnetic disks in the vortex state. Such a vortex exhibits a rich magnon spectrum composed of several high-frequency magnons localized to the outer region of the vortex, as well as a low-frequency gyration mode corresponding to a translation of the vortex core. This gyration mode makes a magnetic vortex very susceptible to periodic driving at frequencies far below the rest of the magnon spectrum. Due to this separation of time scales, resonantly driving the gyration mode corresponds to the periodic modulation of the equilibrium seen by the high-frequency modes, resulting in additional Floquet bands in the magnon spectrum. In the first order of approximation, these bands are equidistantly spaced by the gyration frequency. Nonlinear interactions between high-frequency magnons and the gyration mode finally renormalize the Floquet bands at small wave numbers. Remarkably, this nonlinear coupling between the low-frequency gyration mode and the rest of the magnon spectrum allows for parametric excitation of the gyration at high powers by resonantly driving a single high-frequency magnon mode. This, in return, results in a self-induced Floquet spectrum that can be observed as a spontaneous magnon frequency comb and does not require resonant driving of the gyration mode.

Terahertz-driven coherent spin dynamics in labyrinth-type domain networks

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The controlled manipulation of spins on ultrashort timescales is among the most promising solutions for novel high-speed and low-power-consumption spintronic and magnetic recording applications. To do so, ultrashort terahertz (THz) light pulses can be used to drive coherent spin dynamics on femtosecond timescales. The existence of such THz-driven coherent spin dynamics could be demonstrated in various materials including single-crystalline antiferromagnets and polycrystalline ferromagnets. In polychrystalline ferromagnets, mainly in-plane magnetized thin films with negligible magnetic anisotropy energy (MAE) have been studied so far finding a quasi-instantaneous response of the spin system to the THz magnetic field. Here, we present THz-driven coherent spin dynamics in polycrystalline ferromagnetic Co/Pt multilayers that exhibit a perpendicular magnetic anisotropy (PMA) and a labyrinth-type domain network with nanometer-sized domains. For the first time, we could resolve the coherent spin dynamics on the nanoscale employing THz-pump–X-ray resonant magnetic scattering (XRMS) at the free-electron laser FLASH. Our results revealed THz-driven ultrafast demagnetization as well as coherent spin oscillations that are governed by a time-dependent damping. Instead of the quasi-instantaneous response observed in materials with negligible MAE, we find a temporal lag between the ultrafast demagnetization and the start of the coherent spin oszillations in Co/Pt multilayers with PMA. We could understand this behavior by the interplay of lattice heating and PMA reduction with the consequence of an upper speed limit for THz-induced magnetization switching in PMA media. Finally, we found highly correlated dynamics between domain and domain-wall contributions. As decorrelation effects, e. g., by ultrafast spin-dependet electron transport, seem to be absent, our results suggest the applicability of a THz-driven magnetization control in magnetic nanostructures.

Picosecond dynamics of magnetic domains in FeGd multilayers probed by laser-driven resonant soft-X-ray scattering

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Magnetic domains are fingerprints of the complex interactions within magnetic materials. In addition to a local magnetic order, these emergent textures exhibit lateral periodicities on the nanoscale with a specific orientation and distribution. Statically, the relevant magnetic interactions can be tailored by growing heterostructures of magnetic nanolayers and by applying external stimuli such as magnetic fields and temperature changes. However, the laser-driven dynamics of magnetic domains result from an intricate interplay of local and non-local processes in the depth and in the plane of the sample.

Here, we investigate the ultrafast dynamics of magnetic maze domains in a ferrimagnetic [Fe(0.4nm)/Gd(0.5nm)]116 multilayer sample by time-resolved resonant magnetic small-angle-X-ray scattering (SAXS). This technique is an ideal tool to probe the local and lateral magnetic order element selectively on the relevant femto- to picosecond time and nanometer length scale - but so far the use of this approach has been exclusive to installations at X-ray free-electron lasers. We utilize a novel, laboratory-based setup for transient SAXS experiments at the Fe L (707 eV) and Gd M (1189 eV) absorption edges with 9-ps-temporal resolution [1-4] to benefit from the strong magnetic contrast and large penetration depths at such high photon energies.

Upon photoexcitation, we observe distinctively different time scales for the quenching and recovery of the local magnetization compared to the changes in the domain periodicity. In contrast to previous work [5-7], we find both a transient decrease and increase in the domain periodicity for different pump-probe delays. Based on a detailed analysis of the time-resolved SAXS signal in reciprocal space and heat diffusion simulations [8], we understand these results as indicators for a strongly inhomogeneous magnetic order along the depth of the 100-nm-thick sample. Temperature-dependent experiments reveal a clear dependence of this type of ultrafast domain reorganization on the magnetic ground state of the sample before laser excitation. Our findings highlight the necessity to dissect the magnetization dynamics jointly on both the relevant time and length scales as an essential feature for the knowledge-based development of future functional materials.

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Chiral phononics: Controlling magnetism with a twist

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Chiral phononics is an emerging field that utilizes the angular momentum of circularly polarized lattice vibrations to manipulate the properties of quantum materials. When phonons are driven resonantly with an ultrashort circularly polarized laser pulse, light makes the ions in the material behave like electromagnetic coils, producing circular motions of the atoms around their equilibrium positions in the crystal. This induces real and effective magnetic fields that have been predicted and measured on the tesla-scale, unlocking an unprecedented means for the control of magnetic order. Here, I present our recent theoretical predictions of novel phenomena arising from chiral phonon driving, in particular light-induced magnetization in antiferromagnets through magnonic rectification [1] and light-induced transient multiferroicity in a nonmagnetic nonpolar material [2]. Furthermore, I will show how light can be used to make achiral materials chiral through a phononic rectification process that breaks all improper rotation symmetries of the crystal structure [3]. Finally, I provide suggestions of how to measure these phenomena in a variety of materials.

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Abstracts
of
Poster Session I

Ultrafast demagnetization through femtosecond generation of non-thermal magnons

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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 [1]. We have developed a fully ab initio parameterized out-of-equilibrium theory based on a quantum kinetic approach to describe magnon occupation dynamics due to electron-magnon scattering [2]. This model is applied to perform quantitative simulations on the ultrafast, laser-induced generation of magnons in iron and demonstrates 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 toward higher energy states via a so-far-overlooked scattering term. It is demonstrated that the simple relation between magnetization and temperature computed at equilibrium does not hold in the ultrafast regime. The ensuing Gilbert damping, furthermore, becomes strongly magnon wavevector dependent and requires a description beyond the conventional Landau-Lifshitz-Gilbert spin dynamics. The ab initio parameterized calculations show that ultrafast generation of non-thermal magnons provides a sizable demagnetization within 200 fs in excellent comparison with experimentally observed laser-induced demagnetizations. This investigation thus establishes the importance of non-thermal magnon excitations for the ultrafast demagnetization process.

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Diversity of ultrafast spin dynamics near the tricritical point in a ferrimagnetic Gd/FeCo multilayer

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Ultrafast magnetism is a rapidly evolving field of physics that explores spin dynamics launched in magnetically ordered materials by ultrashort stimuli lasting only a few picoseconds or less. Among all classes of magnetic materials studied in ultrafast magnetism (ferro-, ferri-, and antiferromagnets), ferrimagnets are most promising for the ultrafast and efficient control of magnetic properties. Ferrimagnets, characterized by antiferromagnetically coupled spins with non-equivalent magnitudes, exhibit a nonzero net magnetization dependent on temperature and can have their magnetic structure tuned by external magnetic fields, resulting in a rich H-T phase diagram. In particular, when reaching a large critical field, the spins undergo a spin-flop transition to a noncollinear spin configuration [1]. Prior studies of ferrimagnetic rare-earth (RE) transition-metal (TM) alloys have revealed a new dimension in ultrafast magnetism related to this noncollinear phase [2]. However, due to the relatively strong exchange interaction between the sublattices, the critical field is strong, and the studies required exceptionally high magnetic fields of up to 30 T, hampering further detailed investigations.

Here, we overcome this obstacle by fabricating a high-quality synthetic ferrimagnet – a Gd/FeCo multilayer [3]. In such a heterostructure, the reduced number of nearest neighbors from different species (RE/TM) compared to the alloy is expected to significantly lower the RE-TM exchange interaction as well as the critical field. We found that subtle changes in temperature and magnetic fields below 1 Tesla result in dramatic changes in the ultrafast response of spins to a femtosecond laser excitation. Six distinct types of spin dynamics (see Figure) were identified and explained by considering the spin-flop transition to the noncollinear phase and the concept of a “tricritical point” [4, 5] in the H-T phase diagram [6]. Notably, we highlight exchange-driven reversal as a particularly interesting type of dynamics, providing new insights into the tricritical point, which is found to separate two thermodynamically distinct noncollinear phases with the TM magnetization pointing on adjacent sides of the anisotropy plane.

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The ultrafast magnetization dynamics between optics, magnetism, and quantum mechanics.

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The T-MOKE setup of the pump-probe experiment is often utilized to measure the laser-induced ultrafast magnetization dynamics at femtosecond time scales. The pump laser initiates the dynamics through direct interaction with the electronic degrees of freedom, while the probe pulse triggers the electronic excitation from the filled core states to the empty conduction states above the Fermi level. Interpreting the induced magnetization dynamics is intricate due to the interplay between magnetism and optics that couple several degrees of freedom.

The magnetic asymmetry, the relevant experimental observable, correlates with the off-diagonal components of the dielectric tensor. The matrix elements of the dielectric tensor are energy-dependent due to the selection rules dictated by the quantum numbers of the initial (3p) and the final (3d) states triggered by the probe pulse.

The pump-induced change in the magnetic asymmetry at a given energy is often interpreted as a change in the magnetization ignoring its energy dependence. This can lead to critical misinterpretations of the measurements especially for multicomponent magnetic alloys with overlapping valence bands where optical intersite spin transfer is expected and debated. Furthermore, recent ultrafast experiments probed the dynamics of transition metals and magnetic alloys at more than one probing energy and obtained dramatically variant results [1-3]. Therefore, a comprehensive microscopic picture is crucial to explain the seemingly different dynamics and to unambiguously distinguish the signatures of the induced dynamics by the pump laser from the artifacts of using optical probes as a measurement of the magnetization. To this end, I will present a theoretical framework based on real-time time-dependent density functional theory (RT-TDDFT) to calculate the static and transient energy-dependent reflectivity of two different polarizations and directly compare it to the experiment. We apply this framework to compare the induced magnetization dynamics in hcp Co, and the two Heusler half metals Co2MnGa and Co2MnGe as a prototype for different dynamics measured at a certain probe energy. We also explain the irregular enhancement of the magnetic asymmetry at a given energy of a simple magnetic element like Co and Ni [2]. The crucial role of the quantum mechanics selection rules in the energy-dependent magnetic asymmetry is also emphasized. Finally, we give general guidelines for the interpretation of time-resolved magneto-optical data.

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Magnetization dynamics after noncollinear dual optical excitation

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The ability to manipulate magnetization on ultrafast timescales holds promise for next-generation technologies. Currently, there are significant efforts focused on finding more efficient ways to manipulate spin arrangement with ultrashort laser pulses to obtain the desired state of the quantum systems. For optically isotropic materials, it is believed that optical excitation with two noncollinear ultrashort laser pulses from the same source would result in the same response from the material as a single excitation pulse with equivalent fluence. Specifically, in ultrafast magnetization dynamics, we would expect to see nearly the same demagnetization and recovery dynamics when pumped with two noncollinear pulses compared to excitation with one pump pulse with nearly double fluence. However, our preliminary results show a different trend. Results on magnetization precession in permalloy, demagnetization dynamics in Pt/Co/Pt and TbCo, and all-optical magnetization switching experiments in GdFeCo show that dual optical excitation triggers magnetization dynamics significantly different from those triggered by only one pump pulse. The observed discrepancy in magnetization dynamics could not be explained within the current state of knowledge and must originate from altered magnetic behavior of the material due to optical excitation with interfering light—a new physical effect that implies the possibility to vary the behavior of electrons. This intriguing effect is not limited to a single material; it extends across various aspects of magnetization dynamics, challenging our current understanding of light-magnetic media interactions. We will present our experimental results in detail and discuss their implications for future research.

Kohn-Sham-Proca equations for ultrafast exciton dynamics

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Opto-excitonics i.e. ultrafast laser induced creation, dynamics, and control of excitons, is a potential alternative to conventional charge-based electronics. The most prominent theoretical method of choice for treatment of light-matter interaction, time-dependent density functional theory (TDDFT), however, is unable to describe the dynamics of excitons due to lack of exchange-correlation functional capable of capturing this physics. In our work we introduce an extension to TDDFT in the form of Kohn-Sham-Proca scheme. We demonstrate that this scheme (i) correctly reproduces the linear response excitonic effects in the weak pump regime in excellent agreement with experiment and, (ii) captures excitonic physics in the highly non-linear regime of ultrafast strong laser pumping, in particular "bleaching" (i.e. reduction) of the excitonic weight and the appearance of excitonic side bands.

Laser induced detection, transformation and control of magnetic spin textures in Fe/Gd multilayers

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Materials exhibiting magnetic spin textures provide intriguing opportunities for manipulating static and dynamic magnetic properties via femtosecond laser excitation. We investigate ferrimagnetic [Fe(0.35 nm)/Gd(0.40 nm)]160 multilayers hosting a rich diversity of magnetic spin textures including stripe domains, a dense bubble/skyrmion (B/SK) lattice, and a single domain state [1, 2]. Using fs Kerr spectroscopy, we unambiguously identify the different magnetic spin textures by observing distinct nanosecond coherent spin wave dynamics in response to weak laser excitation. Strong laser excitation allows us to achieve versatile control of the coherent spin dynamics via a non-equilibrium transformation of magnetic spin textures by both creating and annihilating B/SKs, as corroborated by micromagnetic simulations and Lorentz transmission electron microscopy with in-situ optical excitation [3]. Finally, we use a two pump excitation scheme with variable delay to to demonstrate control of the coherent spin dynamics for the B/SK lattice state in amplitude and phase [4].

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Light-induced complete valley polarization in pristine graphene

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For any 2D material where the valley acts as the fundamental quantum index, achieving valley polarization is a key requirement for a functional valleytronic device. Over the past decade, transition metal dichalcogenides (TMDs) have demonstrated valley polarization using circularly polarized light, where light of helicity σ_+ (σ_-) interacts with the K (K$_-$) valleys to induce charge excitations [1]. Consequently, various 2D gapped systems have been investigated to enhance valley contrast [2]. However, achieving this in graphene is difficult due to its intrinsic Dirac-Weyl topology, which leads to zero Berry curvature and inhibits light-valley coupling by circularly polarized pulses. In our work, we have demonstrated that a hybrid femtosecond laser pulse, combining an infrared circularly polarized pulse with a terahertz (THz) frequency linearly polarized pulse, can generate valley-polarized charge states in pristine graphene. The magnitude and direction of these states can be precisely controlled by the fluence of the circular pulse and the polarization angle of the linear THz pulse, respectively. By optimizing the parameters of this hybrid pulse, we can achieve a complete (95%) valley polarization, importantly achieved without the widespread Brillouin zone excitation found in other approaches [4-6].

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Direct versus indirect excitation of ultrafast magnetization dynamics in FeNi alloys

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Ultrafast demagnetization can be induced either coherently by direct interaction with the optical light field or indirectly via excitation of the electron system, which subsequently couples to the magnetization. An intriguing direct process is optical intersite spin transfer (OISTR), which changes the local magnetic moments during the optical absorption process itself and leads to a potentially coherent net spin transport between two magnetic subsystems.

In this contribution, we present transverse magneto-optical Kerr Effect (T-MOKE) measurements in the extreme ultraviolet (XUV) spectral range and compare the ultrafast response of an FeNi alloy after direct optical laser pulse excitation with indirect excitation via injection of a hot electron current.

For both excitation geometries, we find a delayed onset of the loss of magnetic asymmetry of Ni with respect to Fe as well as an increase of magnetic asymmetry for photon energies below the Ni M2,3 edge, signatures that have been interpreted in recent literature as evidence for OISTR, i.e. a form of spin manipulation directly driven by light. Our results are consistent with recent theoretical and experimental investigations that propose alternative explanations for these experimental observations that do not rely on a direct and coherent interaction between light and spin. Instead, the distinctly different magnetization dynamics of Fe and Ni may be governed by intersite spin transfer driven by electron scattering, or may be the result of element-specific microscopic properties such as inhomogeneous spin-orbit coupling or electron-magnon scattering.

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https://doi.org/10.1103/PhysRevResearch.6.013270

Electronic scattering dynamics in altermagnetic materials

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Among the various intriguing properties of altermagnetic materials [1] are their anisotropic band-structures with a characteristic alternating spin arrangement. We explore the impact of these spin structure electronic dynamics as it is driven by ultrashort optical pulses. We present a numerical investigation of electronic dynamics in a prototypical altermagnetic band structure. We determine the optical excitation for the ab-initio calculated band structure of Kru4O8 using Fermi's Golden Rule [2,3]. For these optical excitation conditions we calculate the microscopic momentum-dependent scattering dynamics throughout the whole Brillouin zone. A minimal tight-binding model with a two-dimensional k-space and optimized parameters is employed to capture the essential momentum-space characteristics of the altermagnetic phase. Using an efficient implementation of the equations of motion for the distribution functions, we determine the electron dynamics resulting from electron-phonon [2] and electron-electron interactions. We characterize the influence on charge and spin dynamics throughout the entire Brillouin zone and highlight the importance of Elliot-Yafet spin flips. Our results reveal fingerprints of altermagnetic features on the ultrafast timescales, which may guide future experiments.

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udkm1Dsim – a Python toolbox for simulating 1D ultrafast dynamics in condensed matter

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The investigation of electronic, magnetic, and structural dynamics in solid-state physics has made significant progress during the last decades due to the increasing availability of ultrashort electron and light pulses in a broad spectral range from THz to hard X-rays at large-scale facilities as well as in the laboratory. One of the primary goals of these experiments is to follow the coupling of different degrees of freedom on the relevant time and length scales. To understand and interpret such experimental data, scientists rely on a pool of simulations for modelling and fitting, which are available as software toolkits or as published formalisms. The implementation of these formalisms or the usage and adaption of available external software packages are very time-consuming, and each piece of software covers only very limited aspects of real-world, time-resolved experiments. To that end, the need for a generic, modular, and open-source toolbox that combines different functionalities is obvious.

The udkm1Dsim toolbox [1] allows for creating arbitrary one-dimensional (1D) structures made of crystalline and/or amorphous layers, including stoichiometric mixtures, typically on the nanometer length scale. These 1D structures hold all relevant material information, such as structural, elastic, thermal, magnetic, and optical parameters. The toolbox allows for calculating thermal, structural, and magnetic dynamics on these 1D structures utilizing an N-temperature model (NTM) and multi-layer absorption formalism, a linear masses-and-springs model, as well as a Landau-Lifshitz-Bloch (LLB) and micromagnetic 3-temperature model, respectively. Different types of light-scattering theories can be applied to retrieve the static as well as the transient response from these sample structures due to the dynamics mentioned above, similar to actual pump-probe experiments. Currently, kinematical and dynamical X-ray theories, which can also include polarization-dependent magnetic scattering, are supported. With that, the generally non-linear dependence of the actual observable (scattered light intensity) and the physical quantity of interest (temperature, strain, magnetization, ...) can be revealed. This includes also methods to apply realistic instrumental broadening to the simulated results for better comparison with experimental data.

The udkm1Dsim is freely available at github.com/dschick/udkm1Dsim. It includes complete version control, issue and feature tracking, and project management capabilities to allow for better collaboration between users and developers. This also includes automatic code validation and unit testing, as well as source-code-based generation of the documentation at udkm1Dsim.readthedocs.org as part of the continuous integration (CI) concept.

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Element-resolved detection of ultrafast loss of magnetic order in epitaxial CoO/Fe bilayers

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We study the ultrafast spin dynamics in an epitaxial ferromagnetic/antiferromagnetic Fe/CoO bilayer on Ag (001) by time-resolved resonant soft-x-ray reflectivity. In this system, the axis of the collinear antiferromagnetic spin structure of CoO can be aligned by the magnetization direction of the Fe layer [1]. After excitation with 60 fs light pulses of 800 nm and 400 nm wavelengths, we analyze the transient x-ray magnetic linear dichroism (XMLD) in reflection at the Co L2 edge and the x-ray magnetic circular dichroism (XMCD) at the Fe L3 edge. Both layers demagnetize in less than 200 fs at 800 nm. Since the photon energy of the pump is smaller than the CoO band gap, the excitation of the CoO must arise indirectly from the Fe layer. An atomistic energy-conserving spin model indicates that energy transfer from the hot electrons in Fe to CoO accounts for the ultrafast loss of AFM order in the latter. For excitation at 400 nm, we speculate that the above bandgap excitation and spin transport between Fe and CoO could be possible mechanisms.

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Nutation: separating the dynamics of magnetization from its angular momentum

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The interaction of ultrashort laser pulses with ferromagnets 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 precessional motion of the spin system. In this intermediate time window, the direction of the magnetic moment and the angular momentum are transiently decoupled due to inertia, resulting in additional oscillations, known as nutation, superimposed on the usual precession, with higher frequencies but smaller amplitudes and relaxation times. Using ultrafast time-resolved magneto-optical Kerr effect (TR-MOKE) experiment, we show that a sudden incoherent 'kick' of the spin system of a ferromagnet by a femtosecond optical pulse does not only trigger the well-known precession, but also on top, a much faster nutation. We find a characteristic frequency of nutation in the range of ~ 0.1 THz in permalloy thin films, which has a negligible dependence on magnetic field and film thickness. By comparison with atomistic spin dynamics simulations, we reveal that this experimental observation cannot be explained by the well-known LLG equation of motion, but can be attributed to inertial contributions leading to nutation of the magnetization vector around its angular momentum similar to the wobbling motion of a gyroscope. We show that the separation of the angular momentum from its magnetic moment is an inherent phenomenon occurring during the relaxation of an ultrafast non-equilibrium excitation of a magnetic system using ultrashort optical pulses. Our results enable us to manipulate the interplay of various magnetic processes - from rapid demagnetization to nutation to precession - within a single experiment, promising exciting avenues for future research and technological advancement.

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Laser-driven resonant magnetic soft-X-ray scattering and spectroscopy with 9ps temporal resolution

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Time-resolved resonant magnetic spectroscopy and scattering in the soft-X-ray range are powerful tools for accessing the element-specific and spatially-resolved spin dynamics in magnetic materials. So far, these photon-hungry techniques have been limited to large-scale facilities. However, upgrades to diffraction-limited storage rings supporting only X-ray pulses beyond 100 ps and the shift of X-ray free-electron lasers towards attosecond pulses aggravate the competition for beamtime in the picosecond time window, which is of utmost relevance for magnetism research.

Here, we present the development of a lab-based instrument providing sufficient photon flux for resonant scattering and spectroscopy up to 1.5 keV photon energy covering the soft-X-ray resonances of transition and rare-earth metal atoms. Our setup features the mandatory tunability in energy- and reciprocal space combined with 9 ps temporal resolution by exploiting the broadband emission of a laser-driven plasma X-ray source.

We set up two dedicated beamlines for scattering experiments in transmission and reflection, and for absorption spectroscopy [1]. Here, we present a series of experiments highlighting the feasibility and flexibility of our approach. At the scattering beamline, we followed the dynamics of the magnetic and structural Bragg peaks of an antiferromagnetically-coupled Fe/Cr superlattice at the Fe L-edges at about 700 eV photon energy [2]; we probed the picosecond reorganization of lateral magnetic domains in a Gd/Fe superlattice by magnetic small-angle X-ray scattering (SAXS) in transmission at the Fe L (700 eV) and Gd M-edges (1200 eV) [3]; and we observed the temperature-dependent evolution of the spin spiral of the non-collinear antiferromagnet Dy in reciprocal space. Regarding spectroscopy applications, we have demonstrated that it is possible to measure high-quality X-ray magnetic circular dichroism (XMCD) spectra in spite of the plasma source being intrinsically unpolarized [4]. The generation of partially circularly polarized soft X-rays is realized by utilizing magnetic thin-film polarizers, e.g. at the Fe L-edges. Time-resolved spectroscopy experiments will be performed at a dedicated beamline currently being set up, where we aim to collect broadband, high-resolution X-ray-absorption and XMCD spectra with 9-ps temporal resolution.

Our laser-driven approach enables a variety of resonant magnetic scattering and spectroscopy techniques in the soft-X-ray range on a laboratory scale. It combines high availability with maximum flexibility in sample handling, as well as environmental and excitation conditions. The temporal resolution of 9 ps is well suited to access photoexcited dynamics of, e.g., coherent magnons and phonons, de- and remagnetization processes, including domain dynamics and all-optical magnetic switching.

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Magnetization dynamics in magnetic trilayers with a wedged antiferromagnetic spacer layer at ultrafast timescales

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We studied the influence of an antiferromagnetic (AFM) layer at the interface to a ferromagnetic (FM) layer on the FM magnetization dynamics. For this, we looked at the time-resolved magnetization dynamics of epitaxially grown heterostructures comprised of an AFM Mn wedge sandwiched between two FM Co layers on Cu(001). The two FM layers are coupled indirectly by the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction and directly by exchange through the AFM spin structure, leading to parallel or antiparallel alignment of the FM layers, depending on the Mn thickness [1]. Deposition of Mn in a wedge allows for access to different coupling regimes on the same sample. Magnetization dynamics was probed after excitation with 60 fs laser pulses with a wavelength of 800 nm by X-ray magnetic circular dichroism in resonant soft-X-ray reflectivity. For weak parallel coupling between the two FM layers, a difference in demagnetization time between parallel and antiparallel alignment of the FM layers is observed. We explain this by differences in the AFM spin structure leading to presence or absence of optically induced intersite spin transfer [2] between Mn and Co.

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Characterizing the dynamics of a generalized LLG equation

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Recently, Anders et al. [1] have proposed a generalized stochastic Landau-Lifshitz-Gilbert (LLG) equation which takes into account coloured thermal fluctuations, memory effects and obeys the fluctuation-dissipation theorem. Here we will present some recent results that characterize the spin dynamics in different parameter regimes. First, we show that the generalized LLG equation is well-described by the Bloch equations [2] in the white-noise and Markovian limit of the spin dynamics. Second, going beyond the white-noise and Markovian limit, we present numerical results that show strong ultrafast and inertial effects in the spin dynamics, such as a faster relaxation and the appearance of nutation oscillations.

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Ultrafast magnetization dynamics in NiCo thin films presenting weak stripes domains

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Controlling magnetization without using magnetic fields is a technology-driven strong motivation in the quest for new electronic devices allowing for fast control with low energy consumption. A lot of results exist for ultrafast demagnetization in pure material (as pure Ni for example) but it is essential to understand this phenomenon in alloys or hetero-structures since this systems present the highest potential for applications. We describe how we are using the chemical selectivity and the optical properties of the high harmonics to study the demagnetization induced

by femtosecond laser pulses in thin nickel cobalt films presenting weak stripes magnetic domains.

Recently, all optical switching has emerged as a powerful technique to drive the magnetization of thin films. We are trying to adopt such an all-optical strategy to control the magnetization state of magnetostrictive nanowires grown inside a photostrictive matrice. The first step is to investigate the demagnetization of thin films of a nickel rich (> 80%) cobalt nickel alloy that will compose the nanowires. The femtosecond magnetization dynamics has already been studied in several ferromagnetic alloys and multi-layers such as iron-nickel or cobalt-platinum but there have been contradictory results concerning possible different demagnetization time or delays in the onset of the demagnetization of the different elements composing the system. Ultrafast demagnetization has not been studied so far with element selectivity in cobalt-nickel alloys and we hope to contribute to the development of a unified picture of this phenomenon in alloys. The sample we have studied is a 100 nm thick cobalt-nickel grown on a silicon substrate. Because of the magnetostriction of this alloy, a weak perpendicular magnetic anisotropy exists inside the thin layer. Therefore, above a threshold thickness, a magnetic structure consisting of alternating nanometric magnetic domains with out of plane component of opposite directions will appear. The structure will act as a diffraction grating for photon energies in resonance with the M absorption edges of cobalt (60 eV) and nickel (67 eV). We can then follow the intensity of the scattering spots for these energies as a function of the delay between the pump and the probe and thus reconstruct the magnetization dynamics of cobalt and nickel simultaneously. The preliminary results indicate that there is a delay of about 20 to 30 fs between the onset of demagnetization of cobalt and nickel and that the cobalt demagnetizes less than nickel.

Field-like torque in rare earth-transition metal alloys

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Recently, the magnetization switching driven by spin orbit torque (SOT) in ferrimagnetic materials has attracted widespread attention. It is especially interesting for rare-earth/transition metals alloys that posses a point of angular compensation. In this work we simulate the dynamics of ferrimagnetic materials when excited by SOT close to the angular compensation. In particular, we demonstrate through a mathematical analysis the predominance of the field-like torque (FLT) over the damping-like torque (DLT) in ferrimagnetic materials close to angular compensation. Furthermore, we unveil the particular role of the damping constant in the switching process for such materials.These findings pave the way for the practical applications of the ultrafast magnetization in ferrimagnetic materials.

Energy-resolved spin polarization and magneto-optic response in Ni

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Ultrafast magnetization dynamics were discovered using magneto-optic methods and since then have been intensively studied with them. Recent experiments show a strong dependence of the magneto-optic response on the photon energy and indicate an inter-energy transfer of spins on the timescale of the laser in a pure metallic ferromagnet [1].

We investigate the non-equilibrium spin dynamics on these early timescales by full spin-resolved Boltzmann collision integrals. We trace the temporal evolution of the individual distribution functions of up and down electrons, where spin-flips due to electron-electron and electron-phonon collisions are taken into account.

From the dynamics of the distributions, we extract the spin polarization of both electrons and holes at various energies, similar to an approach we recently applied for spectral electron densities in Au [2]. The energy-resolved spin polarization can vary significantly depending on the considered energy range. It can deviate from the overall magnetization dynamics, both in terms of the qualitative behavior and the timescales involved. We show that signatures usually attributed to ultrafast optically induced inter-energy transfer of spins can also originate from secondary population dynamics. Additionally, we present results on the non-equilibrium magneto-optic response calculated from the distribution functions. It exhibits features similar to the spin polarization and we discuss their different origin. To that end, we compare the spin polarization with the spin-resolved hole dynamics.

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Understanding the experimental observables in magneto-optical Kerr effect spectroscopy in the extreme ultraviolet spectral range

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Time-resolved, magneto-optical Kerr effect spectroscopy (MOKE) in the extreme ultraviolet spectral range (XUV) can provide element-specific information on ultrafast magnetization dynamics of multi-element magnets. In this work, we present a detailed characterization of the experimental observables for three different geometries that allow access to the magnetization for both in-plane and out-of-plane magnetized films using linearly polarized radiation. [1] Besides the most common configuration where the magnetic field is transverse to the incoming p-polarized light (T-MOKE), we show that large asymmetries in the magnetic reflectance is also found in a longitudinal (L-MOKE) as well as in a polar (P-MOKE) configuration. [2] In addition, our analysis shows that pronounced deviations from a linear relationship between magnetic asymmetry and magnetization can occur. These observations are based on static and time-resolved experimental data in combination with simulations based on a wave propagation algorithm. In particular, we find that the reflectance of complex heterostructures in the vicinity of the Brewster angle, which is the most popular geometry in the literature, can exhibit a complex, nonmonotonic, and nonlinear response. This includes increasing values of the magnetic asymmetry for decreasing magnetization.

Finally, we describe sample structures and experimental geometries which are particularly useful in that a linear response of the MOKE observable in the XUV spectral range remains a valid approximation.

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Ultrafast melting of antiferromagnetic order in Cr_2O_3

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Antiferromagnets, discovered only in the 20th century, represent the largest and the least explored class of magnetically ordered materials. Due to the record high (THz) frequency of spin resonance in antiferromagnets, it is believed that one can write bits on antiferromagnets 1000 times faster than on their ferromagnetc counterparts [1]. However, the mechanism and scenarios allowing to manipulate spins in antiferromagnets have been a challenge from the very discovery of antiferromagnetism.

Here we experimentally study spin dynamics triggered in compensated antiferromagnet Cr2O3 by ultrafast heating with the help of femtosecond laser pulse. Using second harmonic generation we visualized antiferromagnetic domains with mutually opposite antiferromagnetic Néel vectors [2]. We study dynamics of the antiferromagnetic domain pattern with sub-picosecond temporal resolution after the optical excitation. The central wavelength of the pump was tuned to a strong absorption in the sample. The experiments show that the pump can cause a melting of the antiferromagnetic order. We discover that depending on the fluence the melting can follow two different routes. The conventional “slow” route is realized at low pump fluences. The mechanism has been previously described for magnetic dielectrics. The characteristic melting time is practically defined by the characteristic time of the spin-lattice interaction, the kinetics reveals a critical slow down upon approaching the Néel temperature, while the amplitude of the laser-induced changes increases [3]. At high fluences, the mechanism is dramatically different. The kinetics does not show any critical slow down and the amplitude decreases upon approaching the Néel temperature. We argue that the “fast” regime reveals the bottleneck in the process of energy transfer between the spin and lattice subsystems.

Our work reveals a new route for melting spin order in dielectric antiferromagnets. The route facilitates picosecond switching between antiferromagnetic and paramagnetic phases and can, in principle, facilitate Heat Assisted Antiferromagnetic Recording at the record-breaking rates.

The work is supported by ERC Grant Agreement No.101054664 (SPARTACUS).

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Field-free laser-induced exchange bias reversal in synthetic ferrimagnetic structures

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Exchange bias (EB) at a ferromagnetic/antiferromagnetic interface is extensively applied in magnetoresistive (MR) sensors for providing a robust reference axis. For application in navigation, robotics or biomedicine1,2, it is required to detect multiple magnetic field components, and do so preferably at a single point in space3. Hence, we are interested in methods for dynamically changing the reference axis orientation by directly acting on the EB layer, allowing for on-demand reprogramming of the reference axis. A candidate of interest for this is all-optical helicity-independent switching4, which was first harnessed by Guo et al.5 to demonstrate sign reversal of EB in an IrMn/GdCo bilayer with single femtosecond laser pulses. In this work, we demonstrate field-free reversal of EB using the method from Guo et al.5 in stacks containing IrMn and Co/Gd multilayers6, mimicking the pinned layer of an MR sensor. With each successive laser pulse we are able to toggle switch between positive and negative EB, and there is always only one stable equilibrium at zero field which toggles between M up and M down, satisfying the unidirectionality required for referencing in MR applications.

To deepen our understanding of the underlying mechanisms we used a mixture of experiments and simulations. Experimentally, we extensively mapped out a phase space of three main parameters: laser pulse fluence, IrMn layer thickness and external magnetic field. The pulse fluence dependence revealed different stages in the reversal process, from which we could identify threshold fluences FAFM and FFM corresponding to exceedance of critical temperatures of the IrMn and Co/Gd, respectively. In order to validate our interpretation, we varied the IrMn layer thickness and verified that FAFM is shifted accordingly while FFM remains unaffected. Moreover, applying external fields that either assist or hinder the optical switch only affects FFM and not FAFM, as expected. Using models describing exchange-driven and temperature-driven dynamics on both the ultrashort and longer timescales we obtained good qualitative agreement with behaviors from both our own experimental parameter space, as well as past measurements by others5. Our findings provide us with indicators to favorable parameters and/or stack configurations for delivering optimized performance for application in MR sensors.

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Two-photon photoemission of antiferromagnetic $LaFeO_3$ with a tunable fs laser system

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Antiferrmomagnetic materials are of high interest for spintronics as they should enable faster magnetic switching as convential ferromagnetic materials [1]. LaFeO3 is an antiferromagnet in the family of perovskite oxides that exhibits weak ferromagnetism through spin canting. It has a high Néel temperature of 740K. The band gap opens between oxygen 2p levels and Fe 3d t2g states. Optical absorption measurements have determined the size of the band gap to be 2.3eV [3]. We use two-photon photoemission (2PPE) with electron excitation across the band gap into the unoccupied Fe states to probe the electronic structure. Using a tunable fs laser system with UV-UV pump-probe experiments, we aim to gain insights into the lifetime and relaxation pathways of electrons at the conduction band minimum. Similarly, time-resolved pump-probe measurements of NiO(100) showed a remarkable short electron lifetime of <10fs at the conduction band minimum, relaxing in a many-body in-gap state [4]. This state shows intensity oscillations at THz frequency by varying the pump-probe delay, which scales with the strength of the antiferromagnetic superexchange interaction [4].

Here 9nm LaFeO3 on SrRuO3/DyScO3 is prepared by pulsed laser deposition, with SrRuO3 as bottom electrode for photoemission experiments. Low energy electron diffraction reveals a (2x2) superstructure, which is a superposition of a c(2x2), (2x1) and (1x2) magnetic order due to spin canting in LaFeO3. The perovskite structure has been identified by the three characteristic surface phonon-polarition modes in high-resolution electron energy loss spectroscopy (HEELS).

We find the onset of photoemission at 1.2 eV below EF, belonging to the oxygen 2p states. Using single-colour 2PPE with photon energies of 4.1, 4.2, and 4.4 eV we find a broad state at 2.0 eV above the Fermi level, which is assigned to photoexcitation into Fe t2g states forming the conduction band onset. Longer exposures to X-rays and UV light shift the LaFeO3 work function to higher values, with X-rays introducing defect states, leading to one-photon photoemission from the Fermi level.

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Laser induced ultrafast magnetization in FeRh using table-top HHG source and T-MOKE scheme

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FeRh is a complex magnetic material that exhibits antiferromagnetic behavior at room temperature, transitioning to a ferromagnetic phase at approximately 340 K. The mechanisms driving laser-induced magnetization in this material remain not fully understood, with potential contributing factors including the nucleation of magnetic domains and lattice expansion. At the CITIUS beamline, part of the Laboratory of Quantum Optic at the University of Nova Gorica, Slovenia, a time-resolved transverse magneto-optical Kerr effect (MOKE) setup has been developed. This setup utilizes femtosecond infrared pulses to pump the sample, while a high-harmonic generation (HHG) source probes the magnetic elements at their M-edges.

In this study, we present results on the laser-induced antiferromagnetic-to-ferromagnetic phase transition in FeRh, monitored through the resonant MOKE signal reflected from the sample. Additionally, we investigate element-specific demagnetization of FeRh in its ferromagnetic phase, facilitated by external heating. This approach allows us to decouple the exchange coupling between the species within this complex system. Our findings provide valuable insights into the dynamic processes governing the magnetization changes in FeRh, advancing the understanding of its magnetic behavior under ultrafast laser excitation.

Nanoscale control of magnetism through nonlinear photon pumping and phonon excitation: First principles calculations

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Intense photon pumping, carefully selected to drive matter to transient or quasistatic states beyond normal conditions, opens avenues for potential advancements in nanoelectronic device development by harnessing nonlinear effects in unconventional materials. Irradiation at THz frequencies can yield high field intensities capable of exciting lattice vibrations into a nonlinear regime, thereby profoundly impacting magnetic properties or material structure, particularly in insulators. This influence may induce changes in magnetic order or insulator-to-metal transitions. Additionally, controlling the excitation of phonons may disrupt symmetry and lead to transitions between different magnetic states [1]. An essential area of study revolves around understanding the transfer of angular momentum and its effects on resulting magnetization. Our research focuses on the modifications of magnetic order attributed to specific non-equilibrium phonon populations [2], particularly in MXenes. MXenes, a family of 2D transition-metal carbides, nitrides, and carbonitrides, have garnered significant attention since 2011 for their potential in advanced applications. This work explores the latest progress in MXene research, focusing on structural modifications and unique opto-electro-magnetic properties [3]. The study underscores the versatility and promise of MXenes as a 2D nanoplatform for future technological innovations. Their potential in nonlinear photon pumping enhances optoelectronic performance, while efficient phonon excitation in MXenes contributes to improved thermal management and device stability. The ability to understand and control these properties in different Mxene monolayers could open doors for advancements in nanoelectronic devices and facilitate the harnessing of nonlinear effects in unconventional materials. Leveraging ab initio simulations, we aim to comprehend how these populations influence magnetic exchange interactions and uncover potential nonlinear effects. Concurrently, our work investigates the electronic properties of different multilayers or monolayers (e.g: V2C), encompassing the optimization of atomic structures, computation of electronic band structures, and the analysis of vibrational properties using frozen phonon method.

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Laser-induced magnetostriction in Gd, Tb and Dy probed by UXRD

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In ultrafast X-ray experiments, we compare the laser-induced magnetostriction in Gd, Tb and Dy upon femtosecond laser excitation at temperatures above and below the magnetic ordering temperatures. All rare earth elements exhibit giant spontaneous magnetostriction and consequently negative thermal expansion (NTE). The coupling between magnetic order and the lattice below the Curie temperature leads to a competition between the expansive stresses from electrons and phonons and the contractive stress due to magnetic order. Thus, upon femtosecond laser excitation, the ultrafast dynamics show a variety of interesting results, such as transforming a typical bipolar strain wave into a unipolar pulse [1, 2].

We compare the thermal lattice expansion and heat capacities of Gd, Tb, and Dy in equilibrium to separate the electronic, phononic and magnetic contributions to the stress in terms of an extended Grüneisen model, which relies on the energy density in the three subsystems: electron, phonons and magnetic excitations. This yields the driving stresses in a linear chain model in order to fit the ultrafast strain dynamics and to identify the coupling constants between the subsystems. We aim to identify the cause of the slower increase of magnetic pressure in Gd compared to Tb and Dy seen in the time resolved measurements. Although these elements are neighbours in the periodic table of elements, we observe different dynamics, which can be explained by the different occupation of the 4f orbitals, which influences the spin-lattice coupling [3, 4].

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Extracting strain contributions to the trMOKE response in $REFe_2$ compounds

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We present time-resolved magneto-optical Kerr effect (MOKE) experiments on $TbFe_2$, $DyFe_2$ and $Dy_{0.7}Tb_{0.3}Fe_2$ thin films which strongly differ in their magnetocrystalline anisotropies. All $REFe_2$ materials exhibit giant (inverse) forced magnetostriction, with more than 10-3 lattice constant change when applying a magnetic field [1], and should therefore respond strongly to picosecond strain pulses created by the ultrafast laser excitation. The Rare Earth element determines the anisotropy for the large effective magnetic moment and the Iron content yields a Curie temperature considerably above room temperature. This makes $REFe_2$ materials interesting for magneto-acoustic transducer applications that could potentially be extended to picosecond ultrasonics [2] or even strain driven magnetization switching [3].

We analyze the signatures of picosecond strain pulses and quasi-static strain created by the (thermal) expansion upon ultrafast laser excitation. We model the strain in the probed near-surface region, which contains both quasi-static and coherent contributions. These simulations can be validated by ultrafast X-ray diffraction experiments, which give direct access to the change of the lattice constant [4]. Strain pulses are clearly visible in polar trMOKE experiments, and therefore the quasi-static strain must also contribute to the total signal. By changing various experimental parameters such as pump fluence or external field strength, we explore the conditions where the (quasi-static) strain contribution is the largest and discuss if the signal is caused by the magnetization dynamics.

Even though the effect is expected to be the strongest in highly magnetostrictive materials, the same principles also apply to other materials with smaller magnetoelastic coupling [5–7]. For a complete model, besides the effects of demagnetization and temperature-dependent anisotropies, the magnetoelastic coupling must also be considered.

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Identifying the non- and near-equilibrium pathway of the laser-induced antiferromagnetic-to-ferromagnetic phase transition in FeRh

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The abrupt change of structural, electronic and magnetic properties of materials featuring first-order phase transitions predestines them for ultrafast laser control of functionalities. Additionally, the complex kinetics including domain nucleation, growth and coalescence associated with a co-existence of multiple phases triggered extensive research on the involved fundamental processes and dynamics of the strongly coupled degrees of freedom.

We use time-resolved x-ray diffraction (UXRD) to study the dynamics of the prototypical laser-induced first-order antiferromagnetic (AFM) to ferromagnetic (FM) phase transition in FeRh that is accompanied by a gigantic expansion of the unit cell. While time-resolved experiments probing the macroscopic magnetization are mainly sensitive on the alignment and coalescence of the nucleated FM domains on hundreds of picoseconds [1], UXRD experiments can directly access the speed of the emergence of the FM phase since the associated lattice expansion is independent of the orientation of the local magnetization. However, these kinetics remained unclear and controversial as previous experiments reported rise times ranging from 15 to 90 ps depending on the excitation strength and the probing depth for inhomogeneously excited FeRh films [2,3].

To clarify these puzzling results, we studied different thicknesses and excitation scenarios of FeRh films. For thin homogeneously excited FeRh films upon direct photoexcitation, we identified an 8 ps timescale for the nucleation of ferromagnetic domains irrespective of the excitation strength [4]. Notably, this time scale is not determined by the propagation of the lattice expansion through the FeRh layer at sound velocity as stated previously. By adding an optically opaque Pt cap layer, FeRh is only indirectly heated above the transition temperature via near-equilibrium heat transport, which leads to a subsequent rise of the ferromagnetic phase on a distinctively larger 50 ps timescale. Further experiments with varying pump pulse durations spanning the electron-phonon relaxation time, revealed the crucial role of photo-excited non-thermal electrons to enable the non-equilibrium pathway of the phase transition in FeRh [5] via an ultrafast modification of the electronic band structure [6]. Finally, we demonstrated that both the non- and near-equilibrium pathways of the AFM-to-FM phase transition occur in inhomogeneously excited FeRh films [4] explaining the previously reported different rise times for different excitation strength and probing depths.

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Laser induced magnetization dynamics in Nickel: Demagnetization and precession

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We compare the laser-induced magnetization dynamics in Nickel for different excitation conditions and geometries of the external magnetic field. The observed time-resolved magneto-optical Kerr effect (trMOKE) experiments have signatures from magnetization precession and demagnetization. By combining trMOKE and ultrafast x-ray diffraction (UXRD) measurements we are able to calibrate the strain and temperature in the modeling of the laser induced dynamics. The temperature and strain response are used to model ultrafast demagnetization and precession.

We identify the role of quasi-static strain, strain pulses and demagnetization driving the precession in two Ni films for different excitation conditions. The relatively homogeneously excited 20 nm thin nickel film exhibits a maximum precession amplitude for an out-of-plane field orientation (angle $\xi = 0°$). In contrast, exciting 200 nm thick nickel films from the backside and probing from the frontside yields the maximum precession amplitude at about $\xi = 45°$. Exciting and probing the thick film from the front side yields trMOKE signals that resemble the thin film, since the probe signal originates from the laser-excited part of the sample.

The magnetization response can be captured using a macrospin model that describes the magnetization dynamics using a modified Landau-Lifshitz Gilbert equation that includes demagnetization as well as contributions from the quasi-static strain and propagating strain pulses on the effective field. These laser-induced change of the magnetoelastic field control the magnetization precession. Two scenarios are identified: for the 20 nm and 200 nm nickel in direct excitation, mainly quasi-static strain drives the precession; in contrast, the backside excitation of the 200 nm sample sends out strain pulses that lead to a resonant enhancement of the magnetization precession. Our findings underscore the significance of the magnetoelastic contributions in the precession response and elucidate the underlying mechanisms responsible for precession.

The ultrafast demagnetization and remagnetization is further investigated by combining time-resolved MOKE and UXRD under identical conditions. We model the temperature and strain response calibrated by UXRD and test the input-parameters of the microscopic three-temperature model (m3TM). In particular we attempt to systematically model fluence dependent measurements to clarify the participation of the phonon system in the energy flow.

X-ray view on light-driven spin-reorientation in orthoferrite $TmFeO_3$

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Antiferromagnets (AFMs) are gaining increasing attention in spintronics and magnonics applications for their potential to overcome limitations associated with traditionally used ferromagnetic (FM) materials. A key advantage of AFMs is their faster spin dynamics, which being not constrained by the rate of angular momentum transfer to the lattice, reside in the THz frequency range. However, the absence of net magnetization in the ground state of AFMs significantly hinders their integration into modern technologies, which require manipulation and probing of AFMs. Consequently, ongoing research is focused on both fundamental understanding and developing methods to efficiently control and probe the spin dynamics in AFM systems.

Among various AFMs, rare-earth (RE) orthoferrites are known for their ultrafast laser-driven spin reorientation phase transitions (SRT). A single femtosecond laser pulse has been shown to launch a 90° reorientation of the AFM Néel vector in thulium orthoferrite TmFeO31. Although optical probes such as magnetic linear birefringence enable the observation of time-resolved spin dynamics during the reorientation, they are not element-specific, lacks spatial information and are frequently affected by unwanted changes in optical properties due to the pump pulse. Ultrashort X-ray probes could offer element-specific insights into the dynamics of both RE and TM ions, providing unprecedented spatial and temporal resolutions. However, studying AFM dynamics with the conventional X-Rays probes, such as X-ray Magnetic Circular Dichroism is challenging as it requires a substantial magnetization absent in the ground state of TmFeO3.

In this work, we use X-ray Magnetic Linear Dichroism (XMLD), which is directly sensitive to the Néel vector, to selectively probe the dynamics of Fe^{3+} spins in $TmFeO_3$. Although XMLD is a well-established tool for studying antiferromagnets (AFMs) in their ground state, its application out-of-equilibrium on the ultrafast timescale remains unexplored to the best of our knowledge. To trace the dynamic pathway of the Néel vector in TmFeO3 as it undergoes the laser-driven SRT, we utilized ultrashort (Δt ~ 120 fs) X-Ray pulses at the BESSY-II slicing facility. A c-cut single crystalline sample of $TmFeO_3$ was used, and an IR pulse (Δt ~ 50 fs) was employed to induce the SRT. By selecting the crystallographic direction and thereby orienting the Néel vector relative to the x-ray polarization, we mapped the Néel vector dynamics on the relevant time scales intrinsic to the SRT. Our results show that the Néel vector undergoes a full rotation through the ac-plane of the crystal, completing its transition from the c-axis to the a-axis in approximately 20 ps. This study demonstrates the XMLD technique's potential for capturing the dynamics of local spin moments in AFM systems on sub-picosecond timescales and provides a detailed insight into the spin reorientation process in $TmFeO_3$.

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The ultrafast dynamics of magnetic anisotropy and magnetic structure in ferrimagnetic alloys CoTb and FeTb.

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30 years ago, Beaurepaire made a groundbreaking discovery that initiated the field of Femtomagnetism. Through a pump-probe experiment on a pure nickel layer, Beaurepaire observed an ultrafast demagnetization. While numerous theories have been proposed to explain this phenomenon, there is still no consensus within the scientific community.

Additionnaly to this fundamental quest, the ultrafast magnetization dynamic has grown in interest due to its potential applications, such as All Optical Switching (AOS). AOS could significantly decrease information encoding times, and is usually obseved in ferrimagnetic rare-earth (RE)-transition metal (TM) alloys. These systems exhibit distinct ultrafast magnetization dynamics when subjected to a femtosecond infrared pulse. To bring more information about the dynamics of such alloys with a spatial resolution, we carried out a Time resolved Small Angle X-ray Scattering (Tr-SAXS) experiment on Co-Tb and Fe-Tb. This experience was carried out at the beamline DIPROI at FERMI. We used the capacity of FERMI to deliver simultaneously two X-ray pulses of different energies to probe the 3d Fe, Co and 4f Tb valence electrons, using the absorption edges M2,3 (52.7eV or 58.9eV) and N4,5 (150.5eV) of Fe or Co and Tb, respectively.

Our samples are amorphous beacause we grow them using magnetron sputtering. They have a high perpendicular magnetic anisotropie due to local order, and possesses magnetic stripes domains. The idea is to scatter x-rays on magnetic domains, and to use two CCD camera in such a way as to get light scattered by Fe or Co and by Tb, at the same time. The CCD camera for Tb is placed three times further away than the one for Fe, corresponding to the factor of three difference in the wavelengths at their respective absorption levels. At the end we were able to recover the demagnetization curve of Fe or Co and Tb simultaneously, for a pump laser of 800 nm with different fluences. By looking at the azimuthal integration of the scattering ring as function of the time delay, we can extract the time resolved evolution of magnetization, domain size and domain distribution of both TM and RE elements. The first results will be presented and discussed.

Investigation of polarization- and magnetic field-dependent transient reflectivity of the 2D antiferromagnetic semiconductor CrSBr

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Since the successful creation of graphene, two-dimensional (2D) materials have captivated substantial and enduring interest over the past two decades due to their unique properties, such as effortless stackability, enhanced interaction with light, and easy tunability of the Fermi level [1]. Among these, CrSBr, an antiferromagnetic semiconductor, stands out as particularly promising for spintronic applications, combining the beneficial properties of a 2D semiconductor with antiferromagnetic order. CrSBr exhibits Wannier excitons and coherent magnons, which can lead to a strong exciton-magnon coupling as demonstrated by Bae et al. [2]. This coupling facilitates optical control of magnetic order and the manipulation of excitonic properties via magnonic excitations.

We conducted transient reflectivity measurements on cleaved CrSBr crystals using an optical pump-supercontinuum probe setup. This approach allowed us to investigate the femtosecond optical response of CrSBr across a wide wavelength range (450-990 nm), where CrSBr exhibits several excitonic resonances. By varying the light polarization vector and applying an out-of-plane magnetic field, we could identify the origins of the detected coherent modes. Our results provide insights into the coupling of quasiparticles such as excitons, phonons, and magnons within this broad wavelength range.

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Exploring magnetic strain as a general proxy for magnetic order in heavy rare-earth elements

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The magnetic exchange interaction is the phenomenon at the heart of both collinear and non-collinear magnetic order. Its distance dependence inevitably induces a change of the equilibrium lattice constant resulting in a magnetic strain contribution [1]. This exchange striction effect is dominant in the thermal expansion of heavy rare-earths, resulting in negative thermal expansion in the temperature range hosting magnetic order [2]. Previous ultrafast hard X-ray diffraction studies of the strain response in rare earth thin films show a competition of the expansion driven by electron and phonon excitations with the contraction due to the loss of magnetic order [3,4].

Using a two-pump pulse excitation scheme [5] in ultrafast hard X-ray diffraction experiments, we experimentally extract the time dependence of the magnetic strain contribution of a 40nm thin dysprosium film for both the helical antiferromagnetic and ferromagnetic phase. We relate our results to time-resolved observations of resonant soft X-ray diffraction and x-ray magnetic circular dichroism (XMCD), which probe the helical antiferromagnetic and ferromagnetic order of the same sample under similar laser-excitation conditions.

We find that the magnetic strain contribution probed in the structural diffraction recovers faster than the diffraction intensity from the helical spin system. This can be rationalized by a transient state where the magnetic order recovers locally, but restoring the long-range helical spin order throughout the thin film takes longer. The recovery of the time-resolved XMCD response on the other hand is observed to be on the same timescale as the recovery of the magnetic strain. This could be rationalized such that the ferromagnetic order is less affected by transient multidomain states and its recovery could be guided by the external magnetic field.

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Studying all-optical magnetization switching of GdFe by double-pulse excitation

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All-optical-switching (AOS) is a potential solution for developing faster and more energy-efficient magnetic data storage devices [1-3]. To study the re-switching of the magnetization and enhance our understanding of the speed of reading and writing processes, we performed experiments using two single laser pulses with an adjusted time delay between them. The double-pulse experiments are performed on a Gd26Fe74 ferrimagnetic alloy with an-out-of-plane easy axis of magnetization and a magnetic compensation temperature of about 120 K. The switching was studied using X-ray magnetic circular dichroism photoelectron emission microscopy (PEEM) and optical Kerr microscopy. At the PEEM setup, laser pulses of 800 nm wavelength and pulse duration of 120 fs excited the sample, while a laser with 1030 nm wavelength and 250 fs pulse duration was used for Kerr microscopy.

AOS was measured as a function of the fluence of the first and second pulse for different time delays between the pulses. At room temperature, when the fluence of the first pulse is above the threshold, in a time-delay window between 3 and 20 ps, the sample re-switches by the second pulse for specific fluence ratios of the two pulses. However, for longer time delays, the sample comes back to regular toggle switching.

Atomistic spin dynamics (ASD) simulations were carried out to describe double-pulse switching in the sample. These simulations successfully describe the behavior of the re-switching as a function of laser fluences and time delay between the two pulses.

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Magnetization-dependent electronic structure and ultrafast electron dynamics in CrGeTe3

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2D magnets present new possibilities to develop sizable and versatile nanodevices that work by manipulating the spin current, making it more efficient than charge-based devices. One of these promising 2D magnets is CrGeTe3 (CGT)[1] with a Curie temperature of ≈ 65K. Besides the magnetic order, the dimensionality allows us to study heterostructures with other 2D materials and make use of magnetic proximity effects. Here, we study bulk crystals of CGT using angle-resolved photoemission spectroscopy (ARPES) and femtosecond time-resolved ARPES (trARPES) to understand the magnetic-order induced changes to the electron distribution and the exchange splitting dynamics at different temperatures. By comparing data at the ASTRID synchrotron with data from our trARPES setup, we discuss the temperature-dependent band structure modifications and their ultrafast dynamics in bulk CGT. We further discuss the implication of our results for thin layers, monolayers, and heterostructures.

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Giant Faraday rotation in atomically thin semiconductors

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Faraday rotation is a fundamental effect in the magneto-optical response of solids, liquids, and gases. Materials with a large Verdet constant find applications in optical modulators, sensors, and non-reciprocal devices.

We measure Faraday rotation spectra around the neutral and charged exciton lines in hBN-encapsulated monolayers of WSe2 and MoSe2 under moderate magnetic fields (B < 1.4 T) [1]. Interestingly, we find that the plane of polarization rotates by several degrees, resulting in a giant Verdet constant of -1.9×107 deg T-1 cm-1 and -2.3×107 deg T-1 cm-1, respectively. To our knowledge, these are the largest Verdet constants measured in the visible/near-infrared regime up to now. The giant Faraday rotation is due to the large oscillator strength and large g-factor of the excitons in the monolayer semiconductors. A heterostructure of a 2D semiconductor with a 2D ferromagnet could further increase the Verdet constant, because in such a heterojunction, strong magnetic exchange interaction effects between the ferromagnetic layer and the excitons in the TMDC are expected.

For our measurements, we use a novel charge-coupled device-based method for performing high-quality Faraday rotation spectroscopy on 2D materials and thin magnetic films on the microscale [2].

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Femtoslicing HR: A new fs-slicing branchline for high spectral resolution XAS and XMCD at BESSY II

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After 2 decades of successful experiments with 100 fs X-ray pulses with variable polarization of 400 - 1400 eV at the UE56/1 ZPM at BESSY II, we report here on an upgrade, which will allow experiments with about a factor 10 higher energy resolution ($E/\Delta E > 4000$) while maintaining the high time resolution of 100 fs and the variable (elliptical) polarization. For this purpose, a novel beamline branch is being set up. The temporal fs x-ray pulse elongation due to diffractive optics is circumvented by performing pump-probe experiments in an intermediate focus in white light prior to monochromatization employing one out of an array of up to 11 blazed VLS gratings (produced by means of e-beam-lithography). This experimental scheme allows for XAS / XMCD measurements in transmission geometry. The detector approx. 37 m behind the source point is a TimePix3 detector in combination with a gateable MCP stack. Furthermore, an increase of the repetition rate (now 6 kHz) is planned, made possible by a new filling pattern, making use of sequential generation of 100 fs X-ray pulses from multiple electron bunches of the BESSY II storage ring. This new beamline is designed for fs magnetization dynamics studies in thin films, where laser-induced magnetic and electronic dynamics can be investigated by XAS and XMCD in transmission geometry for previously unresolvable spectral changes at core-hole-lifetime limited spectral resolution. We show the conceptual design of the entire experiment, the optical layout, raytracing and the performance of the optics and detectors, as well as first results from the commissioning.

Substrate-controlled helicity-dependent magnetization reversal

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The urgent need to reduce energy consumption for electronic devices has led to intense research dedicated to discovering more energy-efficient and ultrafast methods for reversing magnetization. One promising research direction involves all-optical switching of magnetization. Thus far, a variety of different approaches have been identified, ranging from single-shot helicity-independent toggle-switching in GdFeCo alloys [1,2] and helicity-dependent multi-pulse switching in ferro- and ferri-magnetic heterostructures [3,4] to polarization-dependent switching in cobalt-doped iron-garnets [5].

All of the aforementioned mechanisms utilize light in the visible to near-infrared spectral ranges to manipulate the magnetic order. Recently, it was found that excitation with mid- and far-infrared light pulses tuned to match the frequency of the LO phonon mode could also reverse magnetization in iron-garnet samples [6]. More broadly, in the infrared spectral range, one can pump optical phonons at resonance and thereby create magnetization and effective magnetic fields [7,8]. In this direction, we recently demonstrated that circularly-polarized optical phonons driven at resonance in various dielectric paramagnetic substrates generate magnetization via the ultrafast Barnett effect. This enables the directional switching of magnetic order in a nearby overlayer [9].

However, there still remain open questions regarding the necessary degree of circular polarization needed to efficiently drive the switching, and also the universality of this newly-discovered switching mechanism. To address these questions, we have built a setup at the FELIX facility for transient-grating spectroscopy. In this experimental scheme, we interfere two orthogonally-polarized infrared laser pulses, generating spatially-varying polarization. This scheme allows us to freely access a broad range of pumping wavelengths. Moreover, this setup allows us to indirectly extract the lifetime of the excited phonons by changing the temporal overlap of our laser pulses.

Our results reveal that the phonons in the sapphire substrate have a lifetime on the order of several picoseconds. Furthermore, our investigation into the effects of circular polarization quality revealed that when the pump wavelength is on-resonance, the circularity of our light is not critically important. Conversely, when we are slightly off-resonance, a small decrease in circularity causes a significant reduction in switching quality. We have already observed magnetization reversal using sapphire, glass-ceramic, and zinc oxide substrates. The presence of switching in these various substrates points to a universal mechanism. By examining the effects of these substrates, we aim to understand what makes certain substrates more effective. This could yield key insights into the differences in switching quality across the various substrates.

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Magnetic field-induced terahertz photocurrents flowing on the topological surface state of Bi_2Se_3

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We study ultrafast magneto-photocurrents in a three-dimensional topological insulator. For this purpose, we excite (Bi1-rInr)2Se3 thin films with a femtosecond laser pulse (wavelength 800 nm) in the presence of an external magnetic field (Bext) of less than 0.3 T parallel to the film plane. The resulting photocurrent is measured by detecting the terahertz (THz) electromagnetic pulse it emits. We observe an in-plane photocurrent that scales linearly with and is perpendicular to Bext. Strikingly, for r ≥ 4%, we observe an abrupt reduction of the magneto-photocurrent, which coincides with the Indium-induced quenching of the topological surface states of (Bi1-rInr)2Se3. The rise time, decay time, and amplitude of the THz magneto-photocurrent can consistently be explained by a scenario in which a spin-polarized electron photocurrent propagates toward the film surface where it is converted into an in-plane charge current due to spin-velocity locking.

Ultrafast magnetism at the Spectroscopy and Coherent Scattering Instrument of the European XFEL

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The Spectroscopy and Coherent Scattering (SCS) Instrument of the European X-ray Free Electron Laser provides time-resolved X-ray spectroscopy tools to investigate the electronic structure of complex materials and reveal material dynamics on nanometer length scales using soft x-rays. Two experiment stations are provided, FFT and XRD, for solid state sample environments, facilitating forward- and back-scattering geometries for thin films, complex samples and single crystals, respectively. The main classes of experiments are time-resolved and comprise X-ray Absorption Spectroscopy (tr-XAS) in transmission geometry, Small Angle X-ray Scattering (tr-SAXS) from thin films and nanostructures, Coherent Diffraction Imaging and (sub)-microsecond X-ray photon correlation spectroscopy, Resonant Inelastic X-ray Scattering (tr-RIXS) from thin films and crystals and X-ray Diffraction (tr-XRD) of electronic superstructures and lattices in crystals. With the operation of the Apple X afterburners this summer, full polarization control will become available. This will unleash XAS and XMCD studies of complex materials dynamics on femtosecond time scales.

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Ultrafast spin transport in metallic multilayers: excitation and damping of spin waves

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Optical generation of ultrashort spin current (SC) pulses attracts a lot of attention since it pushes the rapidly emerging field of spintronics to the THz domain. As a model system for our studies, we have chosen high-quality epitaxial stacks of Fe and Au layers at MgO(001). This allows to generate shortest (down to 200 fs) SC pulses at Fe/Au interfaces, which propagate over long (~100 nm) distances in Au at time scales corresponding to the Fermi velocity. Combining the magneto-induced second harmonic generation (SHG) and magneto-optical Kerr effect (MOKE) techniques in the back pump-front probe scheme, we have demonstrated long-range nearly-ballistic spin transport in Fe/Au bi-layers [1,2]. Then, in Fe/Au/Fe tri-layers, this approach allowed us to characterize 250 fs-short SC pulses and reveal the SC-induced contribution to the SHG response [3]. Here, we control the SC pulse shape and duration by varying the Fe emitter thickness and show that they are determined by the parameters of hot electron emission and diffusive spin transport in Fe [4]. Using a simple model of emission we develop a description of superdiffusive spin transport in Au and extract electron velocity and scattering rates from experimental data. The crucial role of spin- and energy-dependent electron transmittance of Fe/Au interface and electron-magnon scattering in Fe is discussed.

Furthermore, in Fe/Au/Fe tri-layers, the interface-confined spin transfer torque exerted on the second Fe layer (collector) by these SC pulses, is capable of exciting perpendicular standing spin waves (PSSW), which are probed optically in the time domain [5]. The ultimately small SC pulse duration and asymmetric pulse shape with steep rising part are crucial for the excitation of high-frequency PSSWs: by continuously reducing the collector thickness from 17 to 1 nm, we tuned the frequencies up to 2.4 THz [6]. However, not the SC pulse spectrum limits the observed PSSW frequency but the damping arising from the spin transport in Fe layers with transiently non-uniform magnetization. Here we demonstrate that at sufficiently large wave vectors k the damping is dominated by transverse spin transport effects scaling with 4th power k. Although this contribution is known to originate in the spin diffusion, we argue that at even larger k more general description is necessary and extend the data obtained by MOKE with those obtained by spin-polarized electron energy loss spectroscopy (SPEELS) for the spin wave damping at large k in a similar system, Fe/Ir(001), where the k-dependency of damping saturates. We develop a model describing both MOKE and SPEELS data sets on equal footing, where the 'transverse spin mean free path' is the key parameter, which is estimated to be ~0.5 nm.

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Writing a CoFeB/MgO magnetic tunneling junction with picosecond electrical pulses

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Spintronics holds immense potential for the development of energy-efficient and high-speed memory and logic devices. Magnetic tunnel junctions (MTJs) are key elements of spintronics and have been the focus of extensive research aiming at reducing power consumption and increasing speed. Despite these efforts, the electrical writing speed of MTJs in some applications has yet to surpass 100 ps. Developing new electrical writing mechanisms is essential for exploring the speed limits of MTJs and bridging the gap between ultrafast spintronics and ultrafast magnetism. This challenge necessitates not only precise fabrication techniques but also the use of powerful ultrashort electrical pulses, which are difficult to achieve with commercial pulse generators. Building on previous research that demonstrated 6 ps on-chip spin-orbit torque (SOT) writing in ferromagnetic cobalt [1], we have made advancements by fabricating Auston switches as standalone modules capable of generating robust 10 ps electrical pulses. Utilizing this novel experimental setup, we report for the first time the field-free electrical writing of an 300nm*1000nm MTJ on a 8-inch wafer with 10 picosecond pulses. We demonstrate that the in-plane magnetization of CoFeB/MgO can be switched with a train of picosecond electrical pulses, with switching behaviors depending on the polarity of spin-orbit torques. We believe that the energy-consumption could be less than 1pJ/bit for a 100nm dot. We also characterize the ultrafast magnetization dynamics using a high-bandwidth oscilloscope. The dynamic measurements reveal a multi-resistance state switching regime, where numerous resistance transition events occur before the final decisive one. These transitions are statistically random in their timing. Our work not only explores the speed limit of in-plane MTJs but also suggests new functionalities such as probabilistic computing.

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THz filed excitation of noncollinear antiferromagnet Mn_3Sn

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One of the directions of spintronics development is to clarify the properties of non-standard classes of materials, such as non-collinear antiferromagnets. This class of materials has certain advantages in contrast to classical spintronic materials, namely negating the effect of stray fields and availability of magnetization dynamics in terahertz (THz) frequency range, which makes it possible to record information at higher frequencies in future spintronic devices. Mangetotransport properties due to the topological band structure effects are also distinguished.

The representative of this materials class are noncollinear antiferromagnets Mn3Sn, the antiferromagnetic Weyl semimetals with a noncollinear spin structure in a kagome lattice. Epitaxial thin films of Mn3Sn have attracted a lot of attention due to their potential application in topological spintronics. For example, in such films it was shown that there is a contribution from the topological Hall effect arising from the nonzero scalar spin chirality, which generates the Berry phase in real space [1]. It is important to note that spin-orbit torque with unusual spin orientation was also observed, which in future could lead to efficiently switching of perpendicular magnetizations in spintronic devices [2].

Theoretical studies of magnetization dynamics in non-collinear antiferromagnets show the presence of three separable dynamic modes—one uniform and two optical modes, which allows for resonant excitation by THz radiation [3]. In this work ultrafast magnetization dynamics of epitaxial noncollinear antiferromagnet Mn3Sn thin films, excited by 0.5 ps THz optical transient with the peak electric field of up to 1 MV/cm, is investigated experimentally by means of magnetooptical Kerr and Faraday effects.

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Improved modelling of ultrafast excitation in 2D van der Waals magnets

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Since the discovery of magnetic ordering in two-dimensional systems, the family of two-dimensional van der Waals magnetic materials (2D vdW) has been considerably enriched, comprising now all types of magnetic ordering, ferromagnets, antiferromagnets and even ferrimagnets. Recently the interaction of 2D materials with ultrafast laser pulses has been more and more investigated both experimentally and theoretically Refs. [1-4].

We propose an improved model that is able to reproduce the type-II ultrafast demagnetisation dynamics observed in 2D systems. The spin system is coupled to the electronic thermal bath and it is treated within the atomistic spin dynamics via the software package VAMPIRE[5], while the electron and phonon heat baths are described phenomenologically by coupled equations, also known as the two-temperature model.

Our proposed two-temperature model takes into account the effect of the heated substrate, which for 2D systems results in a slow demagnetisation regime. Since the substrate has different specific heats than the magnetic system, it will heat up more slowly and will lead to a slow demagnetisation of the

magnetic system. We applied the framework to a generic 2D system, CrI3, and we are able to observe a type-II demagnetisation process, characterised by two steps, the first step being attributed to the free electrons generated when the system behaves as a quasi-metal under optical excitation, and the second step, the slower demagnetisation region, due to the heated substrate. After laser excitation, we are able to observe domain formation, similarly to experimental observation. Finally, are able to study the domain formation and type-II ultrafast magnetisation dynamics for various fluences and values of the spin damping.

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Ultrafast dynamics of chiral magnetic domain wall in synthetic antiferromagnets

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Synthetic antiferromagnetic (SAF), which consist of FM layers periodically interleaved with non-magnetic spacers, have attracted considerable interest in spintronics as an important component for magnetic sensors or recording heads in hard disk drives. The SAFs are considered materials with properties in between ferromagnet and antiferromagnet that originate from the constituent ferromagnetic layer and antiferromagnetic interlayer exchange coupling [1]. Therefore, engineering and exploiting their tunability will lead to new physics and applications beyond their conventional use as a pinning layer in spin valves. The negligible stray field can lead to the room-temperature stabilization of chiral domain walls (DW) and cancel the lateral deviation of skyrmions [2]. However, the absence of net magnetization in SAF systems gives a challenge to their observation using standard measurement techniques.

Here, we employ X-ray resonant magnetic scattering (XRMS) to gain access to the internal spin texture of chiral domain walls in SAF multilayers. The element-specific soft X-ray measurements provide the ultimate information on the spin state and depth profiling of the magnetic materials [3]. By using infrared laser pump - XUV probe measurements based on free electron laser, we achieve femtosecond temporal resolution, making time-resolved XRMS a powerful tool for studying ultrafast spin texture changes [4]. The static XRMS of SAFs has performed at a half-odd integer multiple of the Bragg peak. The temperature variation of spin spiral optimized SAF samples has been performed and shown no variation in the magnetic texture periodicity. It indicates a similar temperature dependence of the exchange stiffness and Dzyaloshinskii-Moriya interaction (DMI) [5]. Under the excitation of the femtosecond laser, we studied the magnetization dynamics of the SAF structure. In ferromagnetic multilayers, our ultrafast model proposed to explain the higher demagnetization in the DWs assumes that the domains are far bigger than the DWs [6]. However, in a spin spiral SAF multilayer, we observe a faster demagnetization and coinciding dynamics of magnetism (SUM) and chirality (DIFFERENCE) signal, which accord with spin-polarized hot electron-induced demagnetization mechanism.

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All-phonon control of ultrafast magnetization across first-order Morin phase transition

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Intense pulses of mid-infrared radiation, tuned in resonance with infrared (IR) active phonons, known as phonon pumping, can induce non-equilibrium structural distortions in the crystal lattice, significantly affecting their electronic properties. Recent years have witnessed a rapid advance of this approach for ultrafast control of ordered spins. Starting from the excitation of coherent spin precession [1] and subtle demagnetization [2] the phonon-pumping has led to tantalizing demonstrations of ultrafast coherent switching between magnetic orders [3], and complete reversal of the spin orientation [4].

The spontaneous Morin phase transition in dysprosium orthoferrite (DyFeO3) is an interesting example of a first-order magnetic phase transition, where the magnetic system can be thermally switched between antiferromagnetic and weakly ferromagnetic spin orders. Recently, we demonstrated that by resonantly pumping IR-active phonons this phase transition can be driven nonthermally on an ultrafast timescale. Our findings showed that the transition proceeds via a transient metastable state not accessible in equilibrium, suggestive of a dynamic competition and thus inhomogeneous spin dynamics.

In this study, we visualize the spatio-temporal evolution of the phonon-driven phase transition in DyFeO3 using the unique capabilities of single-shot pump-probe magneto-optical imaging at Nijmegen's Free Electron Laser (FELIX). We reveal not only spatio-temporal snapshots of the phonon-induced magnetization dynamics but also demonstrate that the direction of the magnetization inherent to the phonon-induced weakly ferromagnetic order can be unambiguously controlled by the polarization of the IR-driven phonon. We further discuss potential pathways enabling such control.

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How carrier screening changes the ultrafast lattice dynamics in MoS_2

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We employ femtosecond electron diffuse scattering (FEDS) to investigate the momentum-resolved phonon dynamics in bulk MoS2. Our data reveal that the non-equilibrium lattice dynamics after photo-excitation with a 2.1 eV pump pulse occurs in two successive stages involving qualitatively distinct non-thermal phonon populations before the lattice finally reaches a hot but quasi-thermal state. The first of these populations is established on the 500 fs timescale, and represents K and M phonons generated through electron-phonon thermalization. A second nonthermal phonon distribution emerges with a timescale of roughly 3 ps, featuring hot Q phonons in addition to K and M phonons. Finally, over a timescale of around 40 ps, a hot but quasi-thermal phonon distribution is observed.

To rationalize the observed phonon dynamics, we combine ab initio computations of the structure factor with time-dependent Boltzmann equations. To account for the large excitation densities employed in the experiment, we introduce an approach to account for screening effects of the electron-phonon matrix elements. By combining these tools, we are able to reproduce the two-step relaxation process of the nonequilibrium lattice in MoS2 seen in the FEDS experiments. For the first step, we identify some particularly strongly coupled acoustic modes at M and K which are coupled to the electrons.

These results demonstrate the richness of nonequilibrium lattice dynamics in 2D materials.

Two-dimensional spectroscopy setup at optical frequencies for the investigation of 2D materials

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The exciton is a central quasiparticle in transition metal dichalcogenides (TMDCs), but also in the recently much investigated 2D semiconductor magnets. A detailed understanding of excitons and how they are influenced by magnetic order is essential to assess the potential of TMDC/2D magnets as a platform for optical control of valley polarization and spintronic devices. Two-dimensional spectroscopy is a well-established method to probe exciton properties in nanoscale materials such as quantum dots or quantum wells. Here we introduce a coherent two-dimensional spectrometer tunable over the visible spectral range to investigate the exciton dynamics of 2D materials. The output of a commercial Ti:Sapph amplifier (<100 fs, 800 nm, 1 kHz) is focused into a hollow-core fiber for broadband visible continuum generation. Pumping the fiber with the output of a commercial OPA provides tunability over the visible spectrum and thus can be tuned to the specific requirements of a sample, such as specific excitonic resonances. The broadband visible pulses coming out of the fiber are subsequently sent to programmable pulse shapers, where phase-locked pulse sequences are produced with phase stability lasting one day or more, as measured with spectral interferometry. The phase-locked pulses are employed in the 2D spectroscopy experiments. A custom sample area has been designed specifically for 2D materials, typically featuring high degrees of spatial inhomogeneity and small sizes (few tens of microns). With our setup we aim to reveal insights into exciton dephasing mechanisms and deliver quantities such as spin-dependent charge-transfer time in TMDC/2D magnet heterostructures.

Nonlinear optical probing of a strongly correlated phase transition in epitaxial $NdNiO_3$ thin films

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The family of rare-earth nickelates is a prime example of strongly correlated oxide materials displaying a wealth of physical phenomena from antiferromagnetism to superconductivity. NdNiO3 specifically exhibits a metal-to-insulator phase transition upon cooling that is concomitant with the emergence of antiferromagnetic order. In epitaxial thin-films, the electronic degrees of freedom can be tailored to fundamentally change the character of the phase transition and to enable an antiferromagnetic metal phase within the nickelate family [1].

In this work, we use nonlinear optical second harmonic generation (SHG) in the near-infrared regime to gain insight into the symmetry and ultrafast dynamics of this correlated electronic and magnetic phase transition. In static measurements, we investigate the origin of the SHG response by performing a systematic analysis of the signal as a function of laser polarization, temperature, and photon energy. The optical second-harmonic response enables time-resolved pump-probe experiments as a next step to elucidate the out-of-equilibrium behavior and specifically the processes ensuing an ultrafast quenching of the antiferromagnetic order. Our study highlights the capabilities of nonlinear optics and advances the understanding of strongly correlated quantum materials.

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Magnetic order-dependent ultrafast magnetization dynamics in 4f-based intermetallics

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The speed of magnetic devices is limited by the dissipation channels available for Angular Momentum Transfer (AMT). While Ferromagnets (FM) rely on lattice for AM transfer, Antiferromagnets (AF) offer potentially faster and more robust devices through inter-sublattice spin-spin AMT. However, the microscopic understanding of these channels, especially in Lanthanides with indirect (RKKY) exchange interaction, remains limited. Recent work on LnTm2Si2 (varying Ln) series demonstrated a scaling of AMT rate with RKKY interaction strength, with inter-sublattice channel dominating the magnetization dynamics.

Following this, a complimentary study on the effect of changing the conduction electrons (by varying Tm) while fixing the Ln is of strong interest. To this end, we perform Time-Resolved Resonant X-Ray Diffraction (TrRXD) on Gd based AFMs (GdTm2Si2, Tm=Co, Rh, Ir) as well as Eu based FMs (EuRu2P2, EuFe2P2). Interestingly, the Gd series reveals a similar scaling of AMT rate with the 5d spin polarization of the conduction electrons. These results highlight conduction electrons as an additional tuning parameter for Ln-based devices. Furthermore, a comparison of the AFM dynamics with Eu-FMs provides information about the AMT to the lattice.

Expanding the horizons of angular momentum: Beyond spin

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While spin angular momentum is often the first type of angular momentum that comes to mind, orbital angular momentum (OAM) is another crucial and distinct form. Recent advances in materials science and experimental techniques, such as ultrafast spectroscopy, are revealing new aspects of angular momentum dynamics beyond spin. This expanding field of research can be divided into three main areas: i) Orbital Current: This area focuses on propagating wave packets consisting of hybridized atomic orbitals, which play a vital role in transporting angular momentum in materials. ii) Ultrafast Einstein–de Haas and Barnett Effects: In this domain, spins lose angular momentum to nonthermalized chiral phonons, demonstrating the microscopic origin of the ultrafast Einstein–de Haas effect. Conversely, the ultrafast Barnett effect involves spontaneous magnetization gained temporarily through angular momentum transfer. iii) Manipulation of Magnetism with OAM: This research investigates how light or electron pulses carrying orbital angular momentum can influence and control magnetic properties in materials.

Following this broad introduction to the field of angular momentum beyond spin, I will focus on our own experimental results, demonstrating how photonic OAM affects the ultrafast demagnetization dynamics of a thin nickel film within the first few hundred femtoseconds. The results reveal that the photonic OAM, depending on the excitation geometry, can either accelerate or decelerate the demagnetization process. This effect is observed to be delayed with respect to the initial excitation of the system, suggesting it is due to a secondary process.

Efficient CoPt-based orbitronic terahertz emitters via orbital-to-charge Conversion

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Orbitronics exploits the orbital degree of freedom and paves a new route for developing ultrafast magnetic devices. The orbital source is critical for the performance of ultrafast orbitronic devices. While the orbital generation from Ni upon ultrafast pumping has recently been studied extensively, the application of Ni-based devices is hindered by its low generation efficiency and the low Curie temperature. Here, we present a more effective method of generating ultrafast light-induced orbital currents using CoPt alloy. The resulting orbitronic terahertz emission from CoPt/Cu/MgO structures exhibits terahertz radiation comparable to benchmark spintronic terahertz emitters. Through systematic adjustments in the composition of CoPt alloy, the thickness of Cu, and the capping layer, we verify that terahertz emission primarily originates from the generation of ultrafast orbital accumulation within CoPt material, which propagates through the Cu layer and achieves orbital-to-charge conversion at the Cu/MgO interface via the inverse orbital Rashba-Edelstein effect. This work provides a new opportunity for the developing ultrafast orbitronic devices, paving the way to efficient orbital terahertz emitters.

Ultrafast laser-induced magnetization in a two-dimensional semiconductor

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The quest for rapid control of magnetism has propelled advancements in the field of femtomagnetism. In this study, we theoretically demonstrate that a net magnetic moment can be induced in a two-dimensional (2D) non-magnetic semiconductor on a femtosecond time scale, even in the absence of spin-orbit coupling. Two-dimensional hexagonal semiconductors, such as transition metal dichalcogenides, possess a valley degree of freedom, where valleys are local minima in the conduction bands having opposite Berry curvatures at opposite corners of the Brillouin zone. Optical excitations with circularly polarized laser pulses in these materials result in valley-polarized excitations, a cornerstone of the field of valleytronics. We show how these valley-polarized excitations lead to the induction of an orbital magnetic moment on sub-femtosecond time scales, under both resonant and multi-photon resonant conditions. Furthermore, in the presence of spin-orbit coupling, the net magnetic moment is determined by competing contributions from valley-polarized spin and orbital magnetic moments. Moreover, we show that the induced spin and orbital magnetic moments can be distinctly controlled by tuning the polarization properties of the laser pulse. This work opens new avenues for ultrafast information processing and storage technologies based on the orbital degree of freedom of magnetism.

Coupling phonons, magnons and orbitals: an x-ray view

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Manipulating the magnetization of antiferromagnets (AFs) is of great importance for spintronic applications. To do that with photons on ultrafast timescales, one either requires special materials or special concepts as the magnetic field is usually not a driving parameter in antiferromagnets. Here we address opportunities driving the spins on ultrafast timescales exciting magnons, phonons or orbitals. Discussed experiments are using ultrashort X-ray pulses created by an X-ray free-electron-laser (SwissFEL) that probe the sublattice magnetization, the orbitals and/or the crystal structure.

In the first example, I will show how one can resonantly drive an electromagnon and how to probe the phononic wave function of the electromagnon based on time resolved X-ray diffraction. This can be combined with the response of the spin system using time resolved resonant magnetic x-ray diffraction, which allows us to disentangle lattice and magnetic excitation in this composite mode. [1] The second example addresses the properties of direct driving of an orbital excitation using the magnetic field of the THz pulse. The response after the excitation results in quantum interference of the "two levels" seen through a time dependent antiferromagnetic signal with the frequency of two-level energy separation. [2] (See Fig.) Last, it is shown how static experiments using circularly polarized x-rays can transfer orbital angular momentum to chiral phonons, which are intrinsically carry magnetization (break time reversal symmetry). [3]

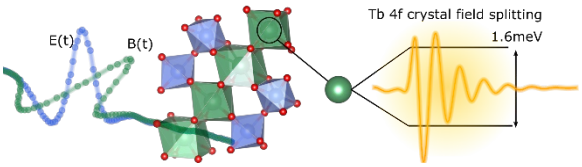

Fig: Sketch of THz driven orbital excitation and its quantum coherent magnetic response.

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The role of angular momentum in ultrafast spintronics

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Starting with the famous discovery of Einstein and de Haas it has been common knowledge that magnetic moment and angular momentum are physical properties which are fundamentally connected. As a consequence, electrodynamics is coupled to quantum mechanics and the transfer and control of angular momentum is a key aspect for the understanding of magnetic materials and the development of future spintronic applications. As a further consequence, precession dominates magnetization dynamics in analogy to the mechanical motion of a spinning top.

When a ferromagnetic film is excited by strong, ultrashort laser pulses, it can lose its magnetic order almost completely on femtosecond time scales. Only recently it was demonstrated that, on the same time scale, the spin angular momentum is transferred to the lattice via the ultrafast creation of chiral phonons that absorb the angular momentum of the spin system [1]. In this talk, I will focus on the understanding of coupled spin-lattice dynamics. Furthermore, I will show how magnetic inertia [2] can separate the dynamics of the magnetization from its related angular momentum [3], an effect that in mechanics is known as nutation.

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Driving and probing terahertz spin and orbital currents

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The field of spinorbitronics exploits the electron orbital momentum L and its spin S for future data-processing applications [1]. Terahertz (THz) time-domain spectroscopy is a powerful tool to reveal spinorbitronic phenomena with femtosecond time resolution [2]. Here, I will present two complimentary approaches: Studying spinorbitronic dynamics using THz emission spectroscopy (TES) as well as by driving with strong THz pulses.

In TES, we optically trigger ultrafast angular-momentum transport in thin-film stacks [3,4]. Varying the thin-film materials allows us to focus on either S or L-dominated ultrafast transport and distinguish different S/L-to-charge-current conversion mechanisms. I will highlight our recent advances in improving spintronic THz sources based on the -channel of transport to obtain THz fields of about 1 MV/cm with a frequency content up to 12 THz [5].

In a second approach, we use these strong THz driving fields to establish spinorbitronic THz detectors by sensing the THz-induced spin accumulation in a bilayer sample. Eventually, I will show how we initiate nonlinear THz magnetization dynamics in the antiferromagnet Mn2Au by driving Néel spin-orbit torques through the strong THz electric fields [6].

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Dynamic control of antiferromagnetic spin-wave spectrum via light-driven electronic excitation

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Antiferromagnetic (AFM) materials feature intrinsic terahertz (THz) spin dynamics, rendering them ideal candidates for future ultrafast and low-energy magnonic and spintronic devices aimed to use spin instead of conventional charge degrees of freedom. The THz response of AFMs is defined by the spectrum of their collective spin excitation known as spin-waves or magnons. Manipulating the spin-wave spectrum is thus the most direct way to control AFM's response times and thus device's operational rates. Achieving this control efficiently, however, is challenging as it necessitates an equally fast as well as long-lasting access to the strength of microscopic interactions (e.g. exchange and/or magnetic anisotropy) governing AFM state.

Here we demonstrate that optical pumping of charge-transfer electronic transitions in prototypical AFM insulator DyFeO3 leads to a appearance of a continuum of the thermally inaccessible in-gap SW states. Examining their coherent dynamics we reveal that the states are consequence of a formation of a transient nearly flat SW band hosting SWs with energies below the quasiparticle magnon gap and almost independent of their momentum. Using numerical simulations, we show that the magnon band emerges as a consequence of long-lasting impact of the electronic excitation on fundamental Dzyaloshinskii-Moria and/or superexchange interactions. Our research not only unveils new avenues for antiferromagnetic magnonics and dynamic control of AFM states, but also paves the way for exploring the exotic flat-band physics in AFMs.

Interplay of electron-magnon scattering and spin-orbit induced electronic spin-flip scattering in a two-band Stoner model

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We present a theoretical investigation of the influence of electron-magnon scattering processes on the ultrafast demagnetization in the framework of a ferromagnetic model system. We compute the microscopic spin-dependent dynamics of electrons in itinerant Bloch states including electron-magnon and electron-electron interactions. While the electron-magnon interactions lead to spin-flip transitions due to the creation/destruction of magnons, the electron-electron scattering processes allow for a different kind of spin-flip processes due to the influence of spin-orbit coupling, which exchanges angular momentum with the lattice.

Our treatment of the combination of these two electronic spin-flip processes constitutes a microscopic dynamical description of the ultrafast demagnetization process, which also underlies the dM/dt model introduced by Beens, Duine and Koopmans in 2020 (PRB 102, 054442).

We show, in particular, that the interplay of the two different electron spin-flip scattering processes leads to the creation of non-equilibrium magnons and makes the transfer of angular momentum from the electronic spins to the lattice in this dynamical scenario particularly effective. In addition, we present evidence that magnonic relaxation mechanisms, such as a magnon-phonon interactions, need to be included in order to describe the remagnetization process.

Optical control of the 4f electronic state in rare-earth metals

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Rare-earth 4f metals play a key role in materials allowing all-optical switching (AOS) of magnetization, 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) experiments at EuXFEL and FLASH, we studied ultrafast changes in the 4f electronic state of different 4f-metals after optical pumping. For Tb metal, we show that 5d-4f inelastic electron-electron scattering leads to selective excitation out of the 4f groundstate multiplet, involving a change of the total angular momentum J. As a consequence, the MCA and thus the coupling of the 4f spins to their environment is manipulated on fs-timescales. In the case of Dy metal the fluence dependent magnetization dynamics shows signatures of an altered 4f-electronic state at large excitation fluence.

The so far disregarded 5d-4f scattering mechanism revolutionizes the fundamental understanding of 4f magnetization dynamics and provides a new perspective to the discussion on AOS in Tb based materials [1].

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Ultrafast angular momentum transfer in RKKY-coupled 4f antiferromagnets

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When photoexcited by an ultrafast laser pulse, antiferromagnets allow direct angular momentum transfer between opposing spins, promising faster functionality than ferromagnets, which are intrinsically limited because their net angular momentum must dissipate to another degree of freedom. The process of angular momentum transfer is closely linked to the nature of magnetic coupling in the system. In lanthanides, 4f magnetic exchange is mediated indirectly through the conduction electrons (the Ruderman–Kittel–Kasuya–Yosida interaction, RKKY), and the effect of such conditions on the antiferromagnetic direct spin transfer is largely unexplored.

In a first set of experiments, we used resonant ultrafast X-ray diffraction to study ultrafast magnetization dynamics in a series of 4f antiferromagnetic intermetallics of type LnRh2Si2, and systematically varied the 4f occupation by substituting the different Lanthanides Ln (Ln=Pr, Nd, Sm, Gd, Tb, Dy, Ho) - thereby altering the magnitude of the on-site RKKY coupling strength. By combining time-resolved soft X-ray diffraction with ab initio calculations, we find that the rate of direct transfer between opposing moments is directly determined by this coupling. [1]

In addition, the influence of the non-local conduction electrons on the RKKY coupling has been explored in the Series GdT2Si2, by varying the transition metal T (T=Co, Rh, Ir). Here, we find a more than twofold increase in ultrafast angular momentum transfer between the materials, which we associate with modifications in the conduction electron susceptibility due to differences in their density of states. [2]

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Disentangling the ultra-fast demagnetization and spin accumulation in magneto-optical detection

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Efficient generation and detection of ultrafast electrical signals can lead to significant advancements in material science, data processing and communication technology. In this sense, THz spin currents from magnetic materials provide a promising framework to overcome some of the challenges of current THz technology, primarily due to their large bandwidth and tunability [1]. Therefore, accurately quantifying and understanding such ultrafast spin currents is a major challenge in the field of ultrafast magnetism. For this reason, the spintronics community has invested vastly into studying both the process of ultrafast demagnetization [2] and the transport of spin currents into other layers either by THz spectroscopy [1], analysis of ultrafast spin torques [3] or even direct optical detection of the resulting spin accumulation [4–6]. In this work, we carefully analyze magneto-optical signals in a series of ferromagnetic samples, in order to deconvolute the signals resulting from the changes in local magnetization and angular momentum transport across layers. During this talk, we will show different signatures of the magneto-optical signals, we will compare rotation and ellipticity, and will present a simple model that describes the detected signals. These observations could shed some light into the longstanding controversy on the differences observed in the rotation and ellipticity in the magneto-optical signals at ultra-short time scales [7–10].

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Ultrafast spintronics

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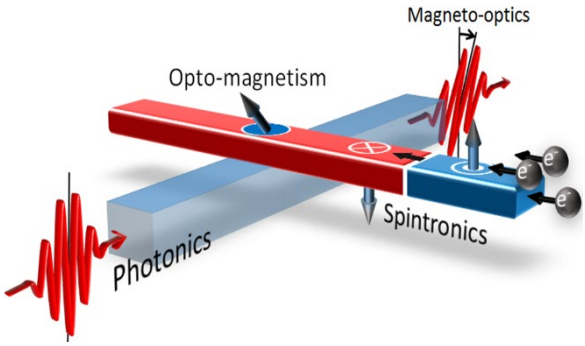
Novel schemes for controlling the ferromagnetic state at femtosecond time scales by pulsed laser excitation have received great interest. By driving systems into the strongly non-equilibrium regime, it has been shown possible not only to quench magnetic order, but also to switch the magnetization by single laser pulses – so-called all-optical switching (AOS). In parallel, it has been found that pulsed laser excitation can also induce spin currents over several to tens of nanometers, which can act as an additional source of sub-picosecond magnetization dynamics. Thereby, a scientifically exciting link between the fields of 'femtomagnetism' and spintronic transport physics has emerged. Moreover, it is being envisioned that combining the two fields could pave the way to a new class of hybrid spintronic-photonic devices, in which data is copied between photonic and magnetic (spintronic) domain without any intermediate electronic steps, leading to ultrafast and highly energy-efficient IT solutions.

In this tutorial, the underlying phenomena will be introduced, and a selection of experimental results combining AOS and spin transport will be presented. Simple models will be introduced that allow for treating all phenomena on equal footing, and providing an intuitive feeling of the various phenomena and their interplay. Finally, I will discuss recent progress towards realizing integrated spintronic-photonic devices, covering the realization of on-chip magneto-optical reading and optically induced switching, reprogrammable sensors based on optically switchable exchange bias, and coherent control of terahertz exchange resonances using optical spin-orbit torques.



Spin-torque-driven ultrafast dynamics of noncollinear antiferromagnet Mn_3Sn

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Noncollinear antiferromagnets, such as Mn_3Sn, stand out for their unique topological properties and potential in antiferromagnetic spintronics. This emerging field aims to harness ultrafast magnetization dynamics of noncollinear antiferromagnets through spin torques. To achieve this goal, it is essential to experimentally measure the time-domain spin-torque-driven magnetization dynamics and theoretically understand the underlying spin transfer physics for noncollinear antiferromagnets, both of which have remained unexplored. Here we report the time-resolved dynamics of Mn_3Sn on a picosecond timescale, driven by an optically-induced spin current pulse. Our results reveal that the magnetization of Mn_3Sn tilts immediately after the spin current pulse and subsequently undergoes 70 GHz precession. This immediate tilting underscores the predominant role of damping-like torque stemming from spin current absorption by Mn_3Sn. By analyzing the tilt in relation to Mn_3Sn thickness, we determine the spin coherence length of Mn_3Sn to be ~15 nm. This value significantly exceeds that of ferromagnets, highlighting a distinct spin-dephasing process in noncollinear antiferromagnets. Our results hold promise for ultrafast applications of noncollinear antiferromagnets and enrich our understanding of their spin transfer physics.

Unraveling optically induced ultrafast modification of nanoscale magnetic textures

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Ultrafast control of magnetization has emerged as a new paradigm for the next generation memory and data storage devices. However, it has been recently recognized that nanoscale heterogeneities can play a critical role in dictating the ultrafast behavior. I will discuss our recent experimental results at x-ray free electron laser (XFEL) sources where we uncovered magnetic texture dependent magnetization dynamics at ultrafast timescales. Our results shows that the symmetry of magnetic texture dictates the magnitude and timescale of the ultrafast response. We also observed fluence threshold dependence for distortions of diffraction pattern which are not seen for magnetization quenching, consistent with a picture of domain wall motion with pinning sites. Supported by simulations, we show that a speed of ~66 km/s for highly curved domain walls can explain the experimental data. We show that far from equilibrium behavior can be used to manipulate spin degrees of freedom at mesoscopic lengthscales.

THz spectroscopy of spin-Hall magnetoresistance in fully metallic systems from 0.3 up to 30 THz

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The manipulation of magnetic moments by current through the spin-orbit torque (SOT) revolutionised the modern spintronics and became a promising ingredience for high-frequency and energy-efficient electronics [1, 2] . Moreover, SOT can be applied to control not only ferromagnets but also antiferromagnets [3] . The recent demonstration of a mutual correlation between SOT and the spin-Hall magnetoresistance (SMR) [4] makes the SMR a suitable tool for indirect experimental investigation of SOT.

Inspired by the ultrafast study of THz anisotropic magnetoresistance [5] which has provided new insights into the ultrafast spin transport in ferromagnets, we applied the same technique on the SMR effect in the THz spectral range. Here, we report on the first THz measurements of the SMR in a fully metallic system in the frequency range from 0.3 to 30 THz. This is acheived by using a differential detection of changes in the THz conductivity for perpendicular orientations of magnetization of the ferromagnetic layer. Our results show the persistence of the SMR up to 30 THz and we discuss them in the framework of the recent THz SMR theory [6] .

Our observation of SMR in the THz spectral range opens the door towards the investigation of SOT in the ultrafast regime or magnonic contributions to the SMR in both ferromagnets and antiferromagnets.

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Tuning of the ultrafast demagnetization by ultrashort spin polarized currents in multi-sublattice ferrimagnets

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Femtosecond laser pulses can be used to induce ultrafast changes in the magnetization of magnetic materials [1]. Several microscopic mechanisms have been proposed to explain the observations, including the transport of ultrashort spin-polarized hot-electrons (SPHE) [2,3]. Such ultrafast spin currents attract growing interest because of the recent challenges in ultrafast spintronics. One of the key challenges in this emerging field is to characterize the actual role of the spin-polarization of such ultrafast current, especially in the specific case of technologically relevant ferrimagnetic alloys. Among different ferrimagnetic materials, the rare-earth transition-metal (RE-TM) alloy thin films, such as FeGd, are interesting for spintronic applications due to their antiferromagnetically coupled 3d and 4f sublattices and the possibility of manipulating the magnetization on ultrafast time scales in a controlled way. Additionally, these are also among systems that show single pulse all-optical switching at a subpicosecond time scale, an important property for applications using ultrafast spintronics [4,5].

In this work, we will present the combined experimental and theoretical results evidencing the spin-dependent hot electron (SPHE) induced demagnetization on the ultrafast time scale on FeGd alloy in a specifically designed spin valve structure. The experimental results were obtained by time-resolved X-ray magnetic circular dichroism (TR-XMCD) at the transition-metal (TM) L3 and rare-earth (RE) M5 edges at the BESSY II Femtoslicing source of the Helmholtz-Zentrum Berlin [6]. This experimental method combines element and magnetic sensitivity with femtosecond time resolution, resolving the ultrafast magnetization dynamics in ferrimagnetic Fe74Gd26 alloys. In addition, relying on experimentally defined geometry and composition as well as interatomic exchange, theoretical modeling based on atomistic spin-dynamics simulations [7] reproduces the experimental ultrafast dynamics of this system. This fact allows identifying the microscopic process of spin angular momentum transfer at the shortest time scale. Our study shows that SPHE drives the demagnetization of the two sub-lattices of the Fe74Gd26 films. This behavior is explained based on two physical mechanisms, i.e., spin transfer torque and thermal fluctuations induced by the SPHE. We provide a quantitative description of the heat transfer of the ultrashort SPHE pulse to the Fe74Gd26 films responsible for the observed magnetization dynamics [8].

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Ultrafast spin transport in layer structures with MgO tunnel barriers

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Ferromagnet/heavy metal bilayers represent a central building block for spintronic devices where the magnetization of the ferromagnet can be controlled by spin currents generated in the heavy metal. The efficiency of spin current generation is essential. Equally important is the efficient transfer of spin current across ferromagnet/heavy metal interfaces. We study this for Pt/Ferromagnet and Ta/ferromagnet interfaces and show atomic scale control of spin current transmission by the insertion of ultrathin MgO layers.

Next, we use these results to improve the performance of spintronic terahertz emitters. Specifically, we demonstrate that stacking spintronic emitters separated by insulating MgO barrier layers can greatly enhance their performance compared to individual emitters. This effect can be fully exploited for terahertz emitters in the on-chip configuration.

Finally, we study the tunneling process across MgO barriers on the ultrafast time scale. For this, we measure the spin conductance across the barriers at terahertz frequencies. As these are the natural frequencies of spin-transport dynamic we obtain important insights into ultrafast spin transport with MgO barriers.

Terahertz antiferromagnetic dynamics induced by ultrafast spin currents

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Insulating antiferromagnets have emerged as significant players in the realm of ultrafast spintronics. Their intrinsic terahertz spin dynamics (see for example [1]) and their capacity to convey spin currents (see for example [2]) make them highly promising for processing spin information at terahertz rates. This capability opens up new perspectives for tomorrow's ultrafast devices. However, the direct transfer of spin angular momentum to an antiferromagnetic insulator at picosecond and sub-picosecond time scales has yet to be conclusively demonstrated. In this presentation, I will address this issue, specifically discussing our experimental evidence that the transfer of ultrafast spin angular momentum to an antiferromagnetic texture can generate coherent terahertz antiferromagnetic excitations [3]. We achieve this through a combination of advanced time-resolved magneto-optical experiments on ferromagnetic/antiferromagnetic bilayers and state-of-the-art mastering of thin epitaxial layer antiferromagnetic textures [4,5]. These findings underscore the efficiency of ultrafast spin transfer torque in triggering antiferromagnetic dynamics and, more broadly, confirm that magnetic information can indeed be propagated into antiferromagnetic spin waves at picosecond timescales.

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Terahertz spin-conductance spectroscopy: probing coherent and incoherent ultrafast spin tunneling

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We introduce a method to measure the ultrabroadband spin conductance across an interlayer X between a ferromagnetic-metal layer F=CoFeB and a heavy-metal layer H=Pt at terahertz frequencies, which coincide with the intrinsic frequencies of spin-transport dynamics. Our approach is applied to X=MgO tunneling barriers of varying thicknesses d=0-6 Å. In the time domain, the spin conductance exhibits two distinct components: an instantaneous feature, which is attributed to coherent spin tunneling, and a longer-lived component, which indicates incoherent resonant spin tunneling mediated by MgO defect states. Notably, the relaxation time of the latter component increases monotonically with d, reaching up to 270 fs at d=6.0 Å. These findings are corroborated by an analytical model. They suggest that terahertz spin-conductance spectroscopy can provide novel insights into ultrafast spin transport in a manifold of spintronic nanostructures.

Probing ultrafast spin transfer effects in Heusler alloys with extreme ultraviolet high harmonics

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The complex band structures of Heusler alloys can be engineered to exhibit a wide range of tunable behaviors. Recently, several such alloys have been predicted theoretically and proven experimentally to demonstrate spin transfer effects in response to laser excitation, on timescales of <100 fs. We compare ultrafast spin dynamics in two Co-based alloys, Co2MnGa and Co2MnGe, as well as pure Co, using the transverse magneto-optical Kerr effect (TMOKE) probed with extreme ultraviolet (EUV) high harmonics. By tuning our driving laser central wavelength, we are able to probe across the entire M-edge of the resonant magnetic sublattices in each material. We find that Co2MnGa exhibits an enhancement of the TMOKE signal across the entire Co M-edge in the first ~100 fs following laser excitation, whereas the enhancement in Co2MnGe and pure Co TMOKE signals is limited to a narrow range of energies. By combining our experimental measurements with time-dependent density functional theory, we determine that ultrafast laser excitation induces a transfer of minority electrons from Co to Mn in Co2MnGa, resulting in the transient increase in Co magnetization. However, in Co2MnGe and pure Co, the very short spin lifetimes prevent a detectable transient TMOKE enhancement resulting from spin transfers. We demonstrate that laser-induced spin transfer effects in Heusler alloys depend on a complex interplay between spin states and lifetimes, but can nevertheless be uncovered by careful measurements with EUV probes aided by theoretical analysis.

Unraveling light-driven spin transfer and hot carrier dynamics by EUV magneto-optical spectroscopy

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The control of spin dynamics by light offers a unique opportunity to control the magnetic state of matter and bring spintronics into the femtosecond regime. Recently, light-driven spin dynamics have been shown to evolve on <10 femtosecond timescale via the process of optical inter-site spin transfer (OISTR) [1-4]. However, due to nonlinearities and optical artefacts in the ultrafast measurement methods used, a quantitative comparison between experiment and theory has not been possible and doubts have been cast on the main experimental observations.

Here, we address these challenges and present a quantitative measurement of femtosecond light-driven inter-site spin dynamics in FeNi alloys. From incidence angle-resolved EUV T-MOKE measurements, we recover the femtosecond dynamics of the full dielectric tensor [5]. In a study on laser-excited nickel, permalloy and iron-nickel alloy [6], we isolate unambiguous signatures of few-femtecond spin dynamics that can be compared to ab-initio theory calculations directly. These results verify the OISTR effect in iron-nickel alloy, and allow us to elucidate how the previously found delayed demagnetization of Ni in FeNi alloys arises as a consequence of the OISTR effect.

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Extracting spin from an antiferromagnet at picosecond timescales

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Interfaces in heavy metal– antiferromagnet heterostructures have recently become highly investigated and debated systems in the effort to create spintronic devices that function at terahertz frequencies. Such heterostructures have great technological potential because antiferromagnets can generate sub-picosecond spin currents which the heavy metal can convert into charge signals. In this talk I will present our recent work on the optically induced picosecond spin transfer from antiferromagnets to Pt using time-domain THz emission spectroscopy. We will focus on two studies in antiferromagnetic insulators KCoF3 and KNiF3, and in antiferromagnetic metal FeRh. Through our studies, we are able to shine light on the microscopy of spin transfer at picosecond timescales and identify key figures of merit for its efficiency. Our results are important for progressing in the fundamental understanding of the highly discussed physics of the heavy metal/antiferromagnet interfaces, which is the necessary cornerstone for the designing of femtosecond antiferromagnetic spintronics devices with optimized characteristics.

Abstracts
of
Poster Session II

Magnetic vortex dynamics probed by time-resolved magnetic helicoidal dichroism

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The laser excitation of magnetization dynamics without applying external fields is a topic of high interest for its potential applications. We explore the use of a ultrashort infrared laser pulse for triggering transient changes in a magnetic vortex structure, probing the induced dynamics by magnetic helicoidal dichroism (MHD) in resonant scattering of extreme ultraviolet light from a free-electron laser carrying orbital angular momentum (OAM). MHD describes the optical response of a magnetic sample upon reversal of the OAM sign or of the magnetization sign, and has been shown to be sensitive to the sample magnetic topology. Here, we provide a proof-of-principle time-resolved MHD experiment, showing that in addition to the well-known ultrafast de-magnetization and re-magnetization laser-induced processes, the analysis of the MHD signal supported by micromagnetic simulations provides a direct evidence of important transient reorganization of the spin texture. In particular, we find that a 50 fs laser pulse of sufficient intensity can temporarily reverse the vortex curling on a 20 ps time scale.

Magnon terahertz spin transport in metallic Gd|Pt thin-films

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Transport of spin angular momentum is a fundamental operation required for future spin-electronic devices. To be competitive with other information carriers, it is required to push spin transport to ultrafast time scales [1]. Here, we use femtosecond laser pulses to trigger ultrafast spin transport in prototypical F N bilayers from a ferromagnetic layer F into a nonmagnetic metal layer N [2]. Following absorption of the pump, a spin current in F is launched and converted into a transverse charge current in N, where it gives rise to the emission of a THz electromagnetic pulse [2]. Two driving forces can occur: (i) a temperature gradient (Seebeck-like effect) [3] and (ii) a spin-voltage gradient [4]. In metallic F, (ii) dominates and relies on conduction electrons, while (i) is found for insulating F [3,4]. Remarkably, in the fully metallic ferromagnet Gd, we find Seebeck-type dynamics and, thus, spin transport due to magnons. This finding highlights the great importance of magnon-mediated spin transport, in particular in metallic systems.

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Influence of Metal/Organic-Molecules Interface on Magnetic Anisotropy in Co Thin Films: A Time-Resolved MOKE Investigation

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We investigated influence of metal/organic-molecules interfaces on the magnetic anisotropy in thin polycrystalline Co thin films grown by electron-beam evaporation. To assess the magnetic anisotropy we measured temperature and magnetic field dependence of the femtosecond-laser induced coherent spin waves by means of the time-resolved magneto-optical Kerr effect (MOKE) spectroscopy [1]. We compare the effects of interfacing the Co films to a nonmagnetic metal (Al), metalorganic complexes tris(8-ydroxyquinoline)gallium (Gaq3) and M-phthalocyanines (M=Cu, Co) as well as Buckminster-fullerene (C60) molecules.

In general, the transient MOKE signals were found to exhibit damped coherent spin wave oscillations (CSWO) with frequencies up to several tens of GHz. Detailed analysis of the spin-wave temperature and magnetic field dependences allowed us to compare the influence of different molecular interfaces.

We found that the thin Co films interfaced with molecular layers display strong hardening of the CSWO frequency at low T with a rather sharp transition in the 150 K - 200 K range, depending on the molecular species. Interfaces with different molecules show qualitatively similar behavior despite different molecular shapes. The hardening is attributed to increase of the interface induced anisotropy due to the hybridization between the molecular-orbitals and the Co d-orbital derived interface states. [2]

Strong damping of the CSWO observed in the molecular-layers interfaced Co films is attributed to the dephasing due to the molecular layer disorder.

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Ultrafast imaging of electric field-controlled laser-induced magnetization dynamics in iron garnet

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Mechanisms allowing to control of magnetization with the help of an electric field, instead of electric currents or magnetic fields, have long been in the focus of fundamental research in magnetism promising eventually future data storage and memory devices with significant reductions in switching power and scalable down to the nanoscale [1]. Hence understanding electric control of magnetization dynamics in magnetic materials is an essential step for the development of ultrafast spintronic devices.

Here we report about an experimental study of how an applied electric field affects ultrafast laser-induced spin dynamics in the epitaxial film of ferrimagnetic iron garnet known to show strong magneto-electric effects [2]. The studied (BiLu)3(FeGa)5O12 film was grown on a (110) oriented Gd3Ga5O12 substrate. Applying a nearly in-plane magnetic field and exciting the sample with a femtosecond laser pulse, we launched large amplitude spin dynamics, which was monitored with the help of an all-optical pump-probe technique. We found that the excited spin dynamics is strongly affected by an external electric field of 1 MV/m applied along the [-110] crystallographic axis. The field strength is by far weaker than in similar experiments on 2D materials, which are known to be strongly susceptible to electric fields [3].

The study demonstrated that the electric field has a significant impact on the dynamics. In particular, due to the magneto-electric coupling, the frequency of spin oscillations changes up to nearly by a factor of 2, the amplitude changes up to nearly by a factor of 5, and the damping – by a factor of 1.5. We propose a model which can explain all these changes in terms of electric field-induced magnetic anisotropy.

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Spin and orbital Rashba-Edelstein effect induced by a femtosecond laser pulse: theory for Au(001)

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Rashba-type spin-orbit coupling has various notable effects on surfaces and interfaces, including spin-momentum locking and spin splitting. A key phenomenon in nonequilibrium situations is the spin Edelstein effect (SEE), where an electric current induces a homogeneous spin polarization perpendicular to the current. This effect is complemented by the orbital Edelstein effect (OEE), where the current induces an orbital polarization. While these effects are well understood in steady-state conditions, their behavior under ultrafast dynamics remains less explored.

In this study, we theoretically investigate the ultrafast SEE and OEE induced by a femtosecond laser pulse on an Au(001) surface. Utilizing a tight-binding model within our EVOLVE framework, we simulate the time evolution of the electron dynamics described by the von Neumann equation.

Our results provide insights into the transient nature of the ultrafast Edelstein effects, revealing significant differences between the spin and orbital components. We analyze the induced spin and orbital polarizations and their respective currents within the Au sample.

Additionally, we explore the generation of spin and orbital Hall currents and evaluate the magnitudes of laser-induced spin and orbital angular momentum. This work contributes to understanding deeper ultrafast spin-orbit-driven phenomena and highlights the intricate dynamics of spin and orbital responses under femtosecond laser excitation.

Ultrafast generation of pure spin- and valley-current in 2D valley active materials

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Pure currents comprise the flow of a two state quantum freedom -- for example the electron spin -- in the absence of charge flow. Radically different from the charge currents that underpin present day electronics, in two dimensional materials possessing additional two state freedoms such as valley index they offer profound possibilities for miniaturization and energy efficiency in a next generation spin- and valleytronics. Here we demonstrate a robust multi-pump lightwave [1,2] protocol capable of generating both pure spin and valley currents on femtosecond times. We combine a linearly polarized pulse component with two circularly polarized components, correlating in this way the crystal momentum (controlled by the linearly polarized component) with the spin excitation (controlled by the circularly polarized components). The generation time is determined by the 2d material gap, with the creation of pure spin current in WSe2 at 40 fs and pure valley current in bilayer graphene at 200 fs. Our all-optical approach demands no special material design, requiring only a gapped valley active material, and is thus applicable to a wide range of 2d materials.

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Theory of magnon mediated electric current drag from nonlocal spin-Hall magnetoresistance in the ac regime

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The spin-Hall effect (SHE) allows the conversion between an electric and spin current in normal metals with spin-orbit coupling. In bilayer systems consisting of a normal metal (N) and a ferromagnetic insulator (F), this gives rise to a small correction to the electrical resistance of a charge current through N due to a spin-transfer torque acting on the magnetization in F, called spin-Hall magnetoresistance (SMR). It has been proposed and measured that the same effects can cause a nonlocal charge current in N F N trilayers, whereby the charge current is transmitted through an electrical insulator by magnons. Here, we present a theory for the nonlocal SMR for the first time in the ac regime up to terahertz driving frequencies and derive its local and nonlocal electrical conductivity up to second order in the applied electric field. Our unified theoretical approach to a variety of bilinear effects – namely Joule heating, spin-torque diode effect, and magnonic unidirectional spin-Hall magnetoresistance – allows for their quantitative comparison and aims to disentangle the various contributions measured in experiments.

Ultrafast phase transition dynamics in the rare-earth orthoferrite ErFeO3

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The prospering field of ultrafast magnetism seeks to explore the picosecond dynamics of promising materials to fulfil the ever-increasing demand for faster read-write operations in magnetic memory. One of the most promising avenues is the light-induced reversal of magnetization by ultrashort laser pulses. Currently, the fastest recorded write-read event time is 30 ps, as demonstrated in GdFeCo [1]. To further reduce this time, materials with switching dynamics at ever-faster timescales are sought after, often characterized by a high anisotropy field.

Here, we investigate the phase transition dynamics in the rare-earth orthoferrite ErFeO3. Previous studies have reported on the dynamics near the spin reorientation transition by employing optical pump-probe [2], THz transmission [3] and THz emission [4, 5] techniques. However, it is still unknown what happens around the compensation temperature under an applied magnetic field.

By employing optical pump – THz emission probe spectroscopy, we study the phase transition at the compensation point. In particular, we apply an external magnetic field along the magnetization axis and track the dynamics of the quasi-antiferromagnetic (q-AFM) mode. Hereby, a switching of the magnetization can be observed as indicated by phase reversal in the time domain. Moreover, our temperature dependent THz emission experiments reveal a hysteresis in the switching dynamics upon heating/cooling. These results are promising for establishing ultrafast switching in ErFeO3. [6]

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Terahertz spin-orbit torques in Fe|GaAs(001)

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The Fe GaAs(001) system is a model system for the study of spin-orbit torques (SOT), i.e., the manipulation of the ferromagnetic order of Fe by an applied electric rather than magnetic field. However, these SOTs have not yet been studied at the femtosecond scale, which is the natural time scale of electronic processes in metals. In this study, we utilize intense ultrabroadband phase-locked single-cycle terahertz electromagnetic pulses to drive SOTs in Fe GaAs(001). The resulting spin dynamics is interrogated by a magneto-optic probe pulse. Our results reveal the presence of a damping-like spin-orbit torque of Dresselhaus-type symmetry. The temporal dynamics of the magnetic order provides crucial insights into spin-orbit interactions in this material system.

Coherent control of terahertz-scale exchange resonances using optical spin–orbit torques

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The ultrafast manipulation of spins on the nanoscale poses one of the core challenges in spintronics [1]. Recent breakthroughs have revealed the potential of optically generating spin currents in non-magnetic heavy metals, which can be injected into neighbouring ferromagnetic layers, exerting a spin–transfer torque [2, 3]. This novel phenomenon was termed optical spin-orbit torque (OSOT), as it exploits spin-orbit interactions. Notably, the spin current's polarization can be reversed by changing the circular polarization of the laser pulse. This offers a more versatile approach for exciting spin dynamics compared to alternatives like ultrafast demagnetization of a neighboring layer, where the spin current's polarization is predetermined by the magnetization direction of said layer [4, 5].

This study explores coherent control of spin resonance modes using multiple pump beams with adjustable delays and polarization states [6]. Employing time-resolved magneto-optical studies, we demonstrate that ferromagnetic resonance modes in Pt/Co/Pt can be triggered by the first pump pulse, with the second pulse amplifying or suppressing the mode based on its delay and polarization. Extending this concept, we find that ferrimagnetic exchange resonances in Co/Gd-based systems—offering much higher frequencies (~THz) and obviating the need for external fields—can likewise be manipulated through this method. Furthermore, investigating phase and amplitude of the exchange-driven modes, we identify features that challenge the current understanding of optically generated spin–orbit torques, and we discuss possible explanations. These insights hold great promise for the advancement of ultrafast spintronic computation devices.

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Probing ultrafast demagnetization with 20 fs resolution reveals dynamics beyond the Stoner model

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Optically induced ultrafast magnetization dynamics (UMD) of ferromagnetic transition metals is a complex phenomenon because it involves many degrees of freedom (electrons in different orbital and spin states, and the crystal lattice) as well as different temporal and spatial scales [1,2].

In this work, we excite ferromagnetic transition metals with a femtosecond laser pulse and use terahertz emission spectroscopy to simultaneously probe the dynamics of the induced spin voltage and UDM [3] with a time resolution of 20 fs. Remarkably, we find that the rise time of the spin voltage is longer than the rise time of the electronic energy following optical excitation. This observation cannot be explained by microscopic models of the Stoner type where the rate of demagnetization instantaneously follows the deposited laser-pulse energy [3,4]. Instead, our measurements indicate that the spin voltage arises from an exchange of angular momentum with electronic states that do not contribute to electronic transport.

We can explain these findings with a generalized Stoner model that includes localized states following the spirit of the s-d model [5, 6]. In this model, electrons are divided into localized states (d), which carry most of the magnetization, and s electrons, which transport spin. Therefore, another time scale arises, on top of spin dissipation in the crystal lattice, due to the exchange of angular momentum between s and d electrons which explains the non-zero rise time of the demagnetization rate. The dynamics of the measured UDM is consistent with angular momentum transfers happening in less than 100 fs for both processes. Our results provide new insights into the physics of UDM at the shortest timescales and reveal a limit for the maximum speed of demagnetization.

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Integrating a multi-terahertz source with active shot-to-shot jitter correction for terahertz-pump optical-probe rxperiments

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High-power terahertz (THz) sources, such as spintronic THz emitters or nonlinear optical crystals [1], are commonly employed to drive the motion of electron, phonons and spins into a nonlinear regime. Difference-frequency generation (DFG) by near-infrared pulses in GaSe provides a compact and straightforward method for generating sub-picosecond high-field pulses at frequencies >10 THz [2]. For example, a pair of frequency-detuned infrared pulses from a dual optical parametric amplifier (OPA) can be used to generate THz pulses by DFG in GaSe [3]. The generated pulses are intrinsically phase-stable with a tunable central frequency ranging from 10 to 70 THz, making them well-suited for resonantly driving infrared-active optical phonons [4].

The time resolution and signal-to-noise ratio of the optical probe signals can be improved by utilizing laser pulses from the seed oscillator of the amplified laser system for probing. However, this approach suffers from time-delay fluctuations between the terahertz pulse and the seed pulses, which arise from optical-path-length fluctuations inside the amplifier. The resulting shot-to-shot timing jitter, typically in the range 10-20 fs, poses a significant obstacle that makes the measurement of small high-frequency pump-probe signals challenging.

Here, we present a DFG-based multi-THz source at frequencies >10 THz whose THz pulses are sampled by the electro-topic sampling using the seed pulses of the laser system. Our setup is combined with a shot-to-shot jitter correction scheme based on the cross correlation of the seed pulse and a chirped amplifier pulse inside a beta barium borate (BBO) crystal [5]. The center frequency spectrum of the generated sum-frequency generation (SFG) signal reports on the relative timing of the two pulses. The time shift can be extracted at the 1 kHz repetition rate of the Ti:Sapphire regenerative amplifier and used to actively correct the time axis of the recorded pump-probe transients. We achieve a temporal resolution better than 1.5 fs.

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Spin vacuum switching

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While physical mechanisms underpinning spin switching are established for picosecond time scales, we here present a physical route to magnetization toggle control – i.e. multiple switching events – at less than 100 femtosceonds. A minority spin current injected into a ferromagnet is shown to generate rapid depopulation of the minority channel below the groundstate Fermi level, creating a minority "spin vacuum" that then drives rapid charge redistribution from the majority channel and spin switching. We demonstrate this mechanism reproduces many of the features of recent sub-picosecond switching of ferromagnetic Co/Pt multilayers, and provide simple practical rules for the design of materials via tailoring the electronic density of states to optimize "spin vacuum" control over magnetic order.

In press, Science Advances.

Mapping of nanoscale terahertz magnetic fields by ultrafast Zeeman torque

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In terahertz spintronics, studying spin torques requires precise understanding of the local electric and magnetic field distribution across metallic thin-film structures. Here, we implement spatially resolved magnetic-field sampling by inserting a thin ferromagnetic layer within heavy metal film of 10 nm thickness. Achieving a resolution of 1 nm and 100 fs, we find that the magnetic field amplitude drops linearly from 1.6 to 0.5 with respect to the incident field over only 10 nm, i.e., over scales much smaller than the vacuum wavelength of about 300 µm. With optimized heterostructures, we expect to approach magnetic-field gradients of 100% per nanometer soon, resulting in gradients of the order of 1 T/nm with state-of-the-art high-field terahertz sources. Such nanoscale terahertz magnetic fields are highly relevant in spintronics and spectroscopy. In particular, they cannot be neglected in the study of interfacial spin-orbit torques at terahertz frequencies. As an example, we show that they can excite perpendicular standing spin waves in a ferromagnetic Fe Ni80Fe20 bilayer.

Single-shot detection of ultrabroadband terahertz electric-field transients

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The detection of terahertz (THz) electromagnetic pulses is of utmost importance for techniques such as THz emission spectroscopy, where THz pulses are generated following the excitation of a sample by a femtosecond laser pulse [1]. To detect the emitted THz wave, electro-optic sampling (EOS) is typically utilized [2]. In this multi-shot pump-probe scheme, the timing between the THz pump and probe is modified to measure the temporal change in the refractive index of the material, which is proportional to the THz electric-field amplitude [3]. In this approach, the shot-to-shot fluctuations (noise) of the multiple sampling pulses may easily exceed small signal amplitudes. Single-shot EOS (SEOS) could overcome these drawbacks because it requires only one probe pulse to measure the time-resolved change in the refractive index of the material over the whole duration of the THz pulse [4, 5].

This work showcases the successful implementation of a single-shot approach for the of THz pulses from a spintronic THz emitter [6, 7] over with a record bandwidth of 0.3-15 THz. The efficacy of this approach is ascertained through a comparison with the established multi-shot EOS (MEOS) methodology. To gain deeper insights into the underlying dynamics at play, theoretical modeling of the SEOS technique is also carried out. These investigations provide a comprehensive understanding of the advantages and limitations offered by the SEOS approach in THz-field detection.

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THz spin and charge currents in a non-centrosymmetric ferromagnet

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Investigating spin-to-charge conversion (SCC) on ultrafast time scales is important for understanding its physical mechanisms and the development of terahertz (THz) sources. SCC may arise from, e.g., the inverse spin Hall effect and spin-galvanic effect, whose experimental separation is challenging. Here, we use a femtosecond laser pulse to excite thin films of the ferromagnetic Heusler compound NiMnSb and measure the emitted THz electromagnetic pulse.

We observe THz signals and, thus, ultrafast photocurrents that can be ascribed to SCC by contributions with Rashba- and Dresselhaus-like symmetry. Based on a symmetry analysis, we separate both contributions and find that the related photocurrents exhibit different temporal dynamics. Further, by changing the amplitude of an external magnetic field, we observe that the Rashba-like current is mainly induced by the ordinary Hall effect, whereas the Dresselhaus-like current is independent of the magnetic field strength. We discuss possible microscopic scenarios of the Dresselhaus-like contribution.

Sub-wavelength localised all-optical helicity-independent switching in GdTbCo using plasmonic gold nanostructures

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All-optical helicity-independent switching (AO-HIS) of the magnetization is envisioned as an ultrafast and energy efficient mechanism for future magnetic storage and spintronic applications [1]. Such applications rely on a high magnetic bit density, with good control over the size, shape, and position of the magnetic domains. In rare earth transition metal alloys such as GdTbCo the high magnetic anisotropy of Terbium(Tb) stabilizes domains down to the less than 100 nm width [2,3]. Metallic nanostructures that support localized surface plasmon polaritons allow electromagnetic confinement well below the diffraction limit and placed on a magnetic multilayer system are a promising way to achieve sub wavelength sized magnetic bits.

Here, we deposit gold nanostructures on Gd10Tb12Co78 films by electron beam lithography and probe the high-resolution magnetization state using magnetic force microscopy (MFM). We toggle the magnetization state of the magnetic film by optically exciting the gold nanostructures with single, intense ultrashort laser pulses at a center wavelength of 1030 nm. To this end we couple the laser pulses into a hollow-core photonic crystal fiber, transport them into the encapsulated MFM and focus the out-coupled laser pulses onto the sample. The low dispersion of the hollow core fiber allows the transmission of ~350 fs short laser pulses with minimal pulse width distortions, crucial to observe AO-HIS.

This novel setup allows us to observe localized AO-HIS in the sample after resonant excitation of gold nano-waveguides by a single 370 fs long laser pulse. We demonstrate that the strong localization of optical fields through resonant excitation of plasmonic waveguides enables nanoscale AOS-HIS of ~240nm width. We report a reproducible reversal of magnetization upon irradiation of the sample with consecutive single pulses with a fluence of 4.5 mJ/cm2. Moreover, we study the influence of the localized electromagnetic field enhancement by the plasmonic nanostructures on the required fluence to switch the magnetization. We find that the fluence required for switching of the magnetization is slightly smaller compared to the bare GdTbCo film of 4.8 mJ/cm2 which is due to the strong confinement of electric field around the nano-structures. We also demonstrate that different shapes of plasmonic nanostructures yield differences in the shape of the switched area due to the electromagnetic field scattering from the nanostructures.

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Fabrication of a hybrid spintronic-photonic device for demonstrating of on-chip all-optical switching of magnetization

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In today's information era, where the demand for computational power is rapidly increasing, conventional charge-based electrical memory devices can hardly meet the high requirements for speed and energy consumption. Non-volatile spintronic memory devices, which encode information through the local magnetization of materials, is a promising candidate to solve these challenges. Their ability to function without a constant current supply might be the catalyst for the shift from traditional electronics to the next-generation computing systems.

Despite the current development of non-volatile memory, such as magnetic random-access memory (MRAM), the reliance on intermediary electronic steps and conversion of data still imposes limitations on energy efficiency and operation speed. Integrated photonic circuitry is anticipated to play a growing role in transferring data on-chip between memory and CPU, as it is capable of processing high-bandwidth information with ultrafast speed and less energy consumption. Therefore, a direct bridge between spintronic devices and photonic circuits would allow us to bypass the energy consuming intermediate electronic steps.

We propose an integrated spintronic-photonic circuit, on which the local magnetization can be efficiently switched via femtosecond laser pulses, the so-called all-optical switching (AOS) process. We propose a proof-of-concept device, in which an optically switchable ferrimagnetic Co/Gd thin film is fabricated in a post-processing step on a Si3N4 photonic chip provided by LioniX International. The magnetic pattern is precisely aligned to the underneath photonic stage with a cross shape. By injecting successive fs laser pulses to the waveguide, the magnetization can be toggle-switched under the ultrafast energy-transfer process from waveguide to magnetic film.

For the whole process, the bare integrated photonic chip is prefabricated by UV light lithography, by which Si3Nx waveguides are formed as the bottom stage of our magnetic layers. A Co/Gd Hall cross is structured on the center of the stage by means of E-beam lithography and a lift-off process. By coupling continuous laser light into the chip via an edge coupler, the scattered light around the metal Hall cross region can already optically prove the good coupling of light from free space to the photonic chip. The magnetic and magneto-transport properties (employing the Anomalous Hall Effect) of the Hall crosses after nanopatterning were carefully analyzed. Presently, experiments on “writing magnetic bits” on our chip by feeding femtosecond single pulses into the waveguide are in progress. Our results demonstrate the possibility of on-chip ultrafast switching of magnetization, and thus open a path to future ultrafast spintronic memory with high energy efficiency.

Exploiting phononic manipulation of magnetization to probe magnetoelastic interactions

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The energy efficient manipulation of magnetization on ultrafast time scales has long been a challenge of critical interest for the development of advanced technologies such as sensors and data recording technologies. In this context, optical means using ultrafast lasers have been increasingly investigated, ranging from helicity dependent switching of Gd-based ferrimagnetic alloys [1] and ferromagnetic multilayers [2] to helicity independent switching [3,4] and exploiting angular momentum transfer in spin valve devices [5,6].

Other recent approaches aim to excite crystal deformations, i.e. phonons, to ultimately influence the magnetic state. In our work, mid-infrared optical pulses emitted from a free-electron laser at the FELIX laboratory are tuned in frequency to match the optical phonon modes of the material. So far, phononic manipulation of magnetic properties has been demonstrated in ferrimagnetic iron garnet [7-9] and antiferromagnetic iron borate [10]. The peculiar macroscopic patterns of the switched domains could be qualitatively explained and reproduced by micromagnetic solutions considering a light-induced elastic strain equivalent to that of a non-uniformly heated object with an axially symmetric temperature distribution [7,8]. However, questions remain about the quantitative distribution of the magnetoelastic interaction.

In this poster, we present the study of a Co-doped iron garnet (Co-YIG) exhibiting labyrinth-like magnetic domains. Upon illuminating the film with narrow-band infrared pulses targeting optical phonon modes at resonance, we observe new permanent domains consisting of stripes and magnetic bubbles. Since the shape of these magnetic patterns after irradiation directly reflects the underlying magnetoelastic interactions, we aim to quantitatively characterize the magnetoelastic interactions and effective fields that drive the dynamics leading to these peculiar magnetic domains.

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Experimental evidence for circular polarization via X-ray magnetic circular dichroism at the 3d transition metal L-edges at FLASH 2

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Absorption spectroscopy, as well as magnetic circular dichroism with circular polarized soft X-rays (XAS and XMCD), is a powerful tool to probe magnetic dynamics in magnetic materials element- and site selectively. Groundbreaking results have been achieved, for instance, for magnetic alloys, which helped to fundamentally advance the field [2,6]. Only recently, a better understanding of helicity dependent XAS opened the opportunity to deduce ultrafast changes in the spin-dependent valence state occupation to deduce spin-dynamics at its very roots [4,5].

Since 2007, the Slicing Facility at BESSY II has been the first and, for a long time, the only facility to offer circular polarized X-rays [3]. Only in the recent years, FEL sources like LCLS or SwissFEL began to implement helical undulator techniques to provide circular polarization.

In a recent upgrade of FLASH, a variable polarization afterburner undulator at FLASH2 was installed at the beginning of 2024. This upgrade allows the generation of circularly polarized X-ray pulses, including the L-edges of the important 3d transition metal elements [1].

In this work, we present first time-resolved experimental results with circularly polarized pulses at the FL23 beamline using XMCD at L2,3 edges of 3d elements, Co, Fe, and Ni. We obtain significant XMCD, indicating a clearly circular polarized beam, and see magnetic dynamics. With an improved pump laser system and the planned SpinFLASH end station, FLASH will offer interesting research possibilities for ultrafast spin dynamics experiments from 3d transition metals.

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Applicability of semi-phenomenological temperature models to ultrafast magnetization dynamics in 2D van-der-Waals ferromagnets

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Sparked by the discovery of sub-picosecond magnetic response of 3d-transition metals to femtosecond laser excitations [1], ultrafast magnetization dynamics have been a focus topic of interest for over two decades now. Still, the microscopic origin of this phenomenon is a hot matter of debate. Semi-phenomenological temperature models have shown themselves successful for a quantitative description of ultrafast magnetization dynamics. The Microscopic Three Temperature Model (M3TM) is a prominent candidate to explain the rapid loss of ferromagnetic order by a transferal of angular momentum from spins to lattice via spin-flip assisted electron- phonon scattering events [2, 3].

In the works to be presented here, we focus on the applicability of the M3TM to two-dimensional van-der-Waals ferromagnets of metallic Fe3GeTe2 (FGT), as well as semimetallic CrI3 and Cr2Ge2Te6 (CGT) [4] . We show that the ultrafast magnetic response of these materials can be very well described and predicted by an extended M3TM, supported by experimental data. In semimetellic systems, a prolonged lifetime of photo-excited electrons in the conduction band give rise to electron-phonon scattering events on picosecond timescales. With experimental results and modeling analysis of itinerant electrons, phonons and spins, we reveal non-thermal phonon dynamics long after electron relaxation in FGT, yielding a complete and comprehensive understanding of the energy dynamics in this material, opening new paths to investigate the processes involved in angular momentum transfer between spins and phonons.

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Femtosecond microscopy of ultrafast spin dynamics at magnetic domain walls

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The dynamics of domain walls and spin excitations are crucial in a variety of magnetic systems, where processes occur on nanometer and femtosecond scales. Recent experimental observations reveal exciting possibilities for manipulation of magnetic domain walls on femtosecond time scales via optical excitation [1-3]. However, comprehensive and systematic studies of nanoscale-femtosecond phenomena in real space remain challenging due to the early development stage of experimental tools that offer simultaneous nanometer and femtosecond resolutions [4-6].

In this work, we introduce a table-top ultrafast microscopy system designed for real-space imaging of magnetic textures, achieving record high spatio-temporal resolution, of sub-15 nm and sub-40 fs. The system utilizes a high harmonic generation (HHG) source with tunable polarization to access X-ray magnetic circular dichroism (XMCD) contrast. The intrinsic, drift-free nature of this real-space imaging tool allows us to explore femtosecond evolution of magnetic domain walls with unmatched angstrom-level precision.

Our study focuses on the direct real-space investigation of the key aspects associated with demagnetization of thin magnetic films exhibiting nanoscale magnetic domains with out-of-plane magnetization. We assess domain wall width, position, relevant temporal scales, fluence dependence, and the potential rearrangement of domain walls during ultrafast demagnetization. Apart from the general magnetization suppression, our data show no discernible spatio-temporal domain wall dynamics for moderate pump fluences that induce up to 50% demagnetization within a 20 ps temporal window for the investigated CoPd multilayers and TbCo alloys. However, higher pump fluences result in pronounced inhomogeneous demagnetization patterns, with domain melting and rearrangement occurring at specific sites, likely related to defects and pinning centers. At very high pump fluences, inducing over ~80% transient magnetization suppression, we observe irreversible alterations in the magnetic domain structure across the field of view.

Our experimental technique addresses the long-standing challenge of simultaneous access to nanometer spatial and femtosecond temporal resolution and provides new insights into the understanding of ultrafast demagnetization in the presence of nanoscale magnetic domain network. We believe that the exceptional performance and accessibility of the developed technique, alongside rapid advances in ultrafast laser and HHG technologies, will position this tool as essential for advancing ultrafast magnetism and spintronics research, particularly where spatial information is of critical importance.

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All-dielectric metasurface for THz control of spins in ferrimagnetic Bismuth doped Gadolinium iron garnet

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Plasmonic antennas have long been seen as a promising tool to facilitate the efficient coupling of light to spins. They have been used in the cutting-edge technology of Heat Assisted Magnetic Recording [1, 2], facilitating magnetic writing with the help of light at the nanoscale well below the diffraction limit. These plasmonic antennas can enhance electromagnetic fields in a broad spectral range from visible light to THz radiation [3]. In particular, the antennas designed for the THz spectral range boosted the effect of a nearly single cycle THz pulses on the spins in TmFeO3 [4, 5]. This revealed the temporal and spectral all-coherent spin switching in TmFeO3.

Here we report that the coupling efficiency of terahertz electromagnetic fields to spins in a ferrimagnetic iron garnet film can be boosted by structuring the surface with a non-magnetic substrate, effectively creating a 2D metasurface. In particular, we have identified the optimal dimensions of the two- dimensional metasurface in the form of stripes using simulations. We verified the amplification of a ferrimagnetic spin resonance by performing pump-probe experiments with a terahertz pump and a visible probe. For these measurements we employed the magneto-optical Faraday effect and Second Harmonic Generation, which are sensitive to the magnetic and electric components of the amplified electromagnetic field, respectively. These measurements reveal the presence of an out-of-plane magnetic field component created by the metasurface at the fundamental and first overtone frequency of the ferrimagnetic spin resonance.

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Time-resolved measurements with THz-STM on semiconducting $MoTe_2$

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The dynamics of elementary excitations on surfaces occur on the scale of nano- to picoseconds, requiring ultrafast measurement techniques to probe them effectively. To achieve this, a pump-probe method using ultrashort laser pulses is preferred. This approach, when combined with scanning tunneling microscopy (STM), not only achieves the necessary time resolution but also provides sub-nanometer spatial resolution [1-2]. Here, we utilize a home-built THz setup in combination with an STM operating at 5K in ultra-high vacuum to investigate the charge dynamics of a semiconducting bulk MoTe2 sample. Our THz pump-probe measurements reveal a temporally decaying oscillatory signal, indicating the presence of coherent excitations [3-4]. The energies of these excitations suggest coherent lattice vibrations or phonons. Additionally, we observe that local defects influence these excitations, as certain excitations are detected only when probing specific defect states.

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Ultrafast magnetization dynamics in vertically aligned nanocomposite thin films

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Exploring optically-induced processes on femto-/picosecond timescales in different types of nanometer-sized magnetic structures might help to develop new design strategies for light-controlled high-density data storage devices. Recent studies have for example scrutinized the ultrafast manipulation of nanoscale domains in thin films [1] and magnetic granular media with perpendicular magnetic anisotropy [3] or tried to identify the fundamental spatial limits of magnetic all-optical switching [2].

In this contribution, we present preliminary results obtained on a novel type of fully epitaxial two-phase system, which consists of ultrathin vertically aligned transition metal wires (with diameters below 10 nm), embedded in a $SrTiO_3$ matrix [4]. After providing an in-depth description of the self-assembly processes that give rise to such heterostructures, we demonstrate how their magnetic anisotropy can precisely be modified using matrix-induced strain [5]. We then turn to a systematic quantitative description of time-resolved MOKE experiments performed on these nanocomposites, with special emphasis on the impact of the pump polarization on the de- and remagnetization behavior. Eventually, we highlight promising future routes to grow multifunctional vertically aligned composites and further analyze light-pulse induced dynamical processes on fs-ps timescales in epitaxial nanostructures.

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Phenomenological modelling of laser-induced nucleation and growth of magnetic domains in Co/Pt thin films

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Ultrafast control of nanoscale magnetic domains is of key fundamental interest and increasingly relevant for storage technologies and emerging alternative computing paradigms. Following pioneering studies of all-optical switching in ferrimagnetic GdFeCo systems [1,2], it has also been discovered that domains much smaller than the spot size of the laser can be nucleated, for example in Co/Pt multilayers hosting magnetic skyrmions [3,4]. Moreover, multipulse switching was discovered in few-layer Co/Pt systems [5], which shows that the helicity of the laser can be used to control the nucleation and induced growth of domains.

Recently, experimental results on laser excitation with consecutive pulses obtained with high-resolution magnetic force microscopy show that the nucleation of domains in Co/Pt occurs stochastically, while the average pulse-to-pulse growth happening on the nanoscale is deterministic and polarization dependent. Interestingly, systematic variation of the laser fluence discloses that the stochastic nucleation depends on the domain structure that is already present. We developed a simple phenomenological model based on nanoscale domain switching energy barriers, that accounts for the helicity dependent absorption of light and moreover depends on the relative orientation of adjacent domains. The latter is key to include the dependence on the existing texture. These barriers lead to temperature dependent switching probabilities which, combined with the inhomogeneous laser heating, allows control over domain evolution consistent with the experiments as well as existing literature. In our model, effective domain growth is achieved via nucleation and coalescence, in contrast to previous explanations in terms of temperature gradient induced domain wall motion [6,7]. These results suggest new pathways utilizing inhomogeneous heating for the control and investigation of ultrafast nanoscale magnetism and photo-excitation across first-order phase transitions in general.

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Terahertz spin currents across a metal|semiconductor interface: Does the photon energy matter?

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Efficient spin current injection into semiconducting materials at terahertz (THz) rates is crucial for future ultrafast spintronic devices.Here, we report on spin injection from a metallic ferromagnetic thin film (F) into a few-monolayer thick semiconducting transition metal dichalcogenide (TMDC), which are known for their efficient spin-to-charge current conversion (S2C). We use ultrashort laser pulses to induce ultrafast spin voltage in F, which triggers a spin current from F into the TMDC layer. Using terahertz-emission spectroscopy, we resolve the photo-induced ultrafast in-plane charge that results from S2C inside the TMDC layer [1, 2]. Surprisingly, upon varying the pump photon energy from 1.5 to 3 eV, we find no change in the photo-induced charge current dynamics while the spin-to-charge conversion (SCC) efficiency increases by a factor of 4. This yields to question the existing models for spintronic THz emission in such heterostructures [1].Our study might enable a more profound understanding of laser-driven spin currents across metal semiconductor interfaces.

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Distortion-free sampling of ultrabroadband terahertz electric fields by spin accumulation

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In spintronics, FM HM stacks consisting of a ferromagnetic-metal (FM) and a heavy-metal (HM) layer are versatile model systems with elementary functionalities like spin transport, spin-charge interconversion and torque [1]. Excited by femtosecond laser pulses, FM HM stacks emit ultrabroadband terahertz electromagnetic pulses that have sizeable amplitude and carry unique information on ultrafast processes inside the stack [2,3]. However, in contrast to the generation of terahertz fields, their spintronic detection has not yet been addressed.

Here, we apply intense terahertz pulses to FM HM stacks and probe the resulting optical birefringence. Strikingly, the signal odd in the FM magnetization agrees excellently with the shape of the incident terahertz electric field over the full bandwidth 1-13 THz. Analysis indicates that the birefringence arises from the terahertz-field-driven spin accumulation at the FM/HM interface through the spin Rashba-Edelstein effect [4], which decays by electron velocity relaxation in <10 fs. A possible contribution by the spin Hall effect is minor as it would be longer-lived.

Our results demonstrate spintronic detection of intense ultrabroadband terahertz fields with extremely flat amplitude and phase response and, thus, no need for deconvolution procedures. Our experiment provides time-domain signatures of the spin Rashba-Edelstein effect and can be viewed as an implementation of interface-specific terahertz-optical sum-frequency generation.

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Potential of spintronic THz emitters for chiral/helical field generation

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We present a proposal for using spintronic THz emitters for the generation of THz fields with localized strong chirality density. This is done by taking advantage of the intrinsic magnetostatic interactions in the magnetic part of the THz emitters. In particular, the shape anisotropy in nanostructured magnetic layers is used to design new type of metamaterials based on spintronic THz emitters that allow to control key properties of the emitted fields such as their local polarization states and orbital structures. As an example, we present full photonic simulations for the generation and propagation of the fields combined with analytical elaboration on the underlying physical processes. The spintronic-based metamaterial is contrasted to conventional cases and some applications of the proposed fields to steer chiral objects will be presented.

Spin-current enabled unidirectional invisibility in non-Hermitian magnonics

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Magnetic excitations are perfectly suited to demonstrate fundamental concepts in quantum field theory, Hermitian and non-Hermitian mechanics, and in nonlinear dynamic. Here, we discuss a setup for realizing unidirectional invisibility of low-energy excitations in a non-Hermitian systems based on magnetic stripes attached to structured current-carrying heavy metal contacts and structured biased fields. The heavy metal serves in essence as a source for charged current-controlled spin-orbit torque. A key point is to engineer the heavy metal and the biased fields as to create a textured PT-symmetric system, a situation hardly realizable experimentally for an electronic quantum mechanical system. For the proposed setup we study the features of the low-energy excitations (magnons) and their propagation.

We identify the conditions necessary for the formation of unidirectional invisibility and find that it requires a specific combination of bias layers and current amplitudes in the heavy metals such that the system exhibits a non-Hermitian degeneracy or exceptional point (EP). The unidirectional invisibility at EP has a larger spectral range for periodic PT-symmetric systems. We analyzed the effect of intrinsic damping on PT-symmetric unidirectional invisibility and confirmed the existence of such phenomenon in experimentally feasible structures by micromagnetic simulations. The potential of this new phenomenon for chiral and non-linear magnonic is demonstrated with numerical simulations.

Ultrafast energy flow in the van der Waals ferromagnet Fe_3GeTe_2 (FGT)

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Two dimensional materials have been the focus of intense study in recent years, with extensive effort invested in transition metal dichalcogenides. A recent addition to this is the study of 2D magnetic materials. Here we focus on one such ferromagnetic material: Fe3GeTe2 (“FGT”). We present an overview of its sub-picosecond response to photoexcitation by employing three experimental probes: trARPES (probes carriers), time-resolved XMCD (probes spins), and ultrafast electron diffraction (UED; probes phonons). The main focus will be on lattice dynamics using UED. We will demonstrate that by probing all three sub-systems, we can identify shortcomings of the conventional M3TM model, in particular misrepresenting lattice dynamics. We demonstrate how slightly modifying the model can reliably reproduce the combined response of all three experiments.

Fingerprints of spin-order in the electronic structure of magnetic 2D materials

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Recent discoveries of 2D materials with intrinsic ferromagnetic/antiferromagnetic ordering show great potential for new spintronics applications. The van der Waals ferromagnets, Fe3GeTe2 (FGT), NiPS3, are particularly promising for their high and electrically tunable curie/Néel temperature. We have recently observed using ARPES that both local magnetic moments and itinerant spin excitations play a cruicial role in describing the electronic ground state and the lowest energy excitations in these materials. In this work, we provide a comprehensive investigation of the electronic structure of FGT, NiPS3 using angle-resolved photoemission spectroscopy (ARPES) with a special attention to creating a pipeline between the DFT results and the observed ARPES signal. This is enabled by wannierization of the DFT states, followed by the construction of a tight-binding model. We demonstrate that, including spin orbit coupling, our DFT bands show good agreement with the experimental ARPES bands.The further use of chinook [Day, R.P.,et al. npj Quantum Mater. 4, 54 (2019)] allows to compare calculations and experiments in the photoemission signal.

Lattice dynamics in a 2D ferromagnet (CGT) on ultrafast time scales

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$Cr_2Ge_2Te_6$ (CGT) is a fascinating material due to its potential applications in spintronic devices and its unique magnetic properties. In this study, we investigate the ultrafast lattice dynamics in CGT using ultrafast electron diffraction (UED). By employing a femtosecond pump-probe method, we observe changes in lattice behavior before and after the magnetic transition at ~63K. Our observations also imply distinctive dynamics between different directions. These findings suggest notable variations in lattice dynamics across the transition temperature and directions, potentially providing insights into the coupling mechanisms among the subsystems in CGT.

Increasing terahertz spintronic emission with planar antennas

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Spintronic terahertz emitters constitute a promising alternative to semi-conductor based devices to rectify femtosecond laser pulses for terahertz generation[1]. It has recently been showed that such emitters could be integrated in larger antenna structures to influence their radiation. [2] In this communication, we build upon this idea and report about the design, simulation, fabrication and characterization of small-size spintronic THz emitters equipped with planar electromagnetic antennas of different shapes. We observe an increased THz emission for all designs, with a particularly strong effect in the sub-THz range and for the broadband antennas. Simulations were performed using CST Studio Suite. The spintronic terahertz emitter (width 10µm, length 7 to 15 µm) is modelled as an excitation port with an internal impedance in the range of 30-100 Ohms, in agreement with the sheet resistance of the layers used. Using open boundary conditions and neglecting losses in both sapphire substrate and gold antenna arms, we calculate the reflection coefficient seen from the port and use an optimization algorithm to minimize it around 1THz. This procedure was used for three different geometries, namely dipole, bow-tie and spiral antennas. As expected, for the dipole geometry, the optimization returns an antenna length close to half a wavelength (57 µm), and a relatively narrow-band response. For bow-tie and spiral geometries, we obtain much larger dimensions (217-920 µm) and a very broadband response starting from 0.1-0.3 THz. To fabricate the corresponding structures, we first deposit a Ta(3)/Co(5)/Pt(3) nm trilayer onto a sapphire substrate by magnetron sputtering. Then we pattern it into rectangles of appropriate size by laser lithography and ion beam etching. Finally we pattern the antenna arms by a second laser lithography step and lift-off of a Ti(10)/Au(60nm) layer. The THz emission for the different designs has been characterized using a pump-probe technique. A DC magnetic field of ∼350mT was permanently applied along the sample plane to align the magnetization of the cobalt layer in-plane, perpendicular to the antenna arms. Femtosecond pump pulses (800nm@35fs), delivered by an amplified 10kHz laser system, are focused on the single selected device using a X10 objective lens. The spot size of 30 µm ensures the homogeneous pump fluence of 2.8 mJ/cm2 over the emitter surface, maximizing the spin current generation. The generated sub-ps electromagnetic pulses are collected from the substrate side and focused onto a ZnTe crystal via two parabolic mirrors. Electro-Optical Sampling is used to retrieve the electric field profile of the THz pulse. We observe a systematic increase of amplitude for emitters equipped with antennas with respect to single emitters of the same size. The increase of pulse amplitude is up to a factor of ten, the largest enhancement being observed for a large bow-tie antenna. Comparing the spectral contents for the different geometries, we observe stronger low frequency component for the broadband designs than for the dipole one, in agreement with simulations. [1] T. Seifert et al., Nature Photonics 10, 483 (2016). [2] U. Nandi et al., Appl. Phys. Lett. 115, 022405 (2019).

Effect of resonant pumping on the spin dynamics in Co/C60 thin films

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We demonstrate the manipulation of spin dynamics in a nano-scale metallic/organic hybrid unit in a deterministic way. Our work represents the all-optical control of the spin precession in thin cobalt films interfaced with C60 molecules. The excitons in the C60 layer, excited by ultrashort laser pulses at specific photon energies, influence the GHz spin dynamics in ferromagnetic cobalt. Our analyses reveal that the modifications in the precession frequency is up to 60% between resonant and off-resonant excitation. Furrthermore, the precession frequency increased fivefold at low temperatures, compared to pristine Co. Transient reflectivity spectra emphasize the pivotal role of resonant exciton formation within the C60 layer in determining the observed frequencies, highlighting the complex interplay between exciton dynamics and magnetic anisotropy in Co/C60 samples. By analyzing the ultrafast spin dynamics both with and without excitons, we establish a direct link between exciton formation in C60, modulation of magnetic anisotropy in Co thin films,and the resultant changes in GHz spin dynamics. This research advances our understanding of nano-scale hybrid units based on metallic ferromagnetic substrates and lays the groundwork for the development of highly efficient, ultrafast spintronic devices that capitalize on the distinctive properties of molecular materials.

Investigation of coherently excited magnons and phonons using high repetition rate (1 GHz) femtosecond laser comb

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Recent progress in the field of magnonics and spintronics has sparked significant interest in studying phenomena related to magnons and phonons, both from a fundamental research perspective and for potential applications in information and communication technology [1]. It is well known that the magnons have well-defined angular momentum, phonons were also described to carry intrinsic angular momentum through coupling with the magnetism [2]. So far, most of the studies related to magnon-phonon interaction use conventional way such as interdigitated transducers (IDTs) and piezoelectric substrates for the electrical excitation of phonons which often limits the frequency range as well as the efficiency of the excitation. Hence, there is a need to develop an efficient approach for phonon excitation in the material systems without much-complicated fabrication and understand the coupling mechanism with other quasi-particles such as magnons.

Here, we adopt a novel approach based on high repetition rate (1 GHz) femtosecond (fs) laser combs combined with micro-focused Brillouin light scattering microscopy (μ-BLS) to investigate the coherently excited phonons and magnons in NiFe thin films. The versatility of frequency comb enhanced μ-BLS [3] has proven to be an efficient way for ultrafast generation of sustained spin waves [4-6] and spin-wave caustics [7]. In this work, films of permalloy (Ni80Fe20) were sputtered onto c-plane sapphire substrates by DC magnetron sputtering under a 3 mTorr Ar atmosphere in an ultrahigh vacuum chamber. In the μ-BLS experiment, the sample is excited with a 1 GHz rep-rate fs-laser of wavelength 816 nm and probed with a continuous wave laser of wavelength 532 nm. The scattered beam from the sample is passed through a polarizer and guided to the tandem Fabry-Pérot interferometer for spectral decomposition of the light. In stark contrast to thermally excited incoherent phonons/magnons, photons scattered from the coherently excited phonons/magnons exhibit a giant rotation of their polarization angle [8]. The rotation is frequency and field-dependent, showing a resonant behavior around the NiFe ferromagnetic resonance. Our study highlights the effectiveness of frequency combs for generating and investigating sustained spin and acoustic excitation in magnetic thin films at several GHz frequencies.

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Ultrafast changes in the M5,4 branching ratio in Terbium

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All optical switching (AOS) to reverse magnetic orientation on ultrafast time scales is a promising concept for future generations of magnetic memory storage [1]. Many of the materials showing AOS are containing 4f rare earth metals. Thus, it is important to understand the dynamics of magnetic and electronic properties upon optical excitation in those materials.

As reported recently, inelastic 5d-4f electronic scattering in 4f rare earth metals transiently alters the 4f orbital state and therewith the total angular momentum J [2]. These transitions transiently change the electronic and magnetic properties in 4f metals on ultrafast time scales.

Within an X-ray absorption study performed at the FemtoSpeX slicing facility at BESSY II we show, that the observed changes of J also affect the M5,4 branching ratio on ultrafast timescales in Terbium, proving the 3rd rule of Thole and Van der Laan [3] to be applicable in non-equilibrium. The change of the branching ratio can be used, to calculate the percentage of 4f-excited atoms and extract a 4f temperature.

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Epitaxial growth of Mn_3Sn thin films using magnetron sputtering

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The theoretical predictions and subsequent experimental demonstration of a large anomalous Hall effect (AHE), driven by non-vanishing Berry curvature in antiferromagnets with non-collinear spin textures such as Mn3X (X = Ir, Sn or Ge), have generated considerable interest in the field of spintronics (1-4). In addition to the AHE and the anomalous Nernst effect, which highlight their unique topological properties, these non-collinear antiferromagnets (NCAFM) also exhibit ultrafast spin dynamics in the terahertz (THz) frequency range, significantly exceeding the spin dynamics observed in traditional ferromagnets (5). These extraordinary attributes make Mn3X compounds highly promising for the development of next-generation spintronic technologies. However, the challenge lies in growing high-quality thin films, a crucial requirement for further studies and practical applications.

In this work, we present our recent progress in the growth of thin Mn_3Sn films by magnetron sputtering. We successfully synthesized epitaxial films, with thicknesses ranging from 5 nm to 60 nm, by co-sputtering high purity Mn and Sn targets in an ultra-high vacuum chamber, with a base pressure of 1 x 10-9 Torr. The stoichiometry of the films was analyzed using Rutherford backscattering spectroscopy, confirming precise elemental ratios. Our thin films also show very smooth surface morphology, with a root mean square roughness of less than 0.5 nm. Mn3Sn crystallizes in the hexagonal space group P63/mmc and transitions to a non-collinear antiferromagnetic phase below 420 K, and it can exhibit both in-plane and out-of-plane magnetic anisotropy, depending on its crystallographic orientation. Here we demonstrate that epitaxial Mn3Sn can be grown with in-plane anisotropy along the (0001) orientation on Al2O3 (0001) substrates, and with out-of-plane anisotropy along the (20-20) orientation on MgO (110) substrates. Additionally, we have made significant progress in fabricating free-standing layers of single-crystalline Mn3Sn grown on a water-soluble sacrificial layer, which allows us to explore the intrinsic properties of these materials without substrate-induced constraints. Finally, we present our findings on out-of-plane magnetic domain imaging using resonant magnetic circular dichroism (RMCD), which provides insight into the antiferromagnetic domains structure in Mn3Sn films.

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Ultrafast soft X-ray magnetic holography at SwissFEL

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X-ray imaging techniques have enabled a significant advancement in the understanding of the physics driving magnetic systems, thanks to the possibility of combining element sensitive imaging, sensitivity to the magnetic and antiferromagnetic ordering through contrast mechanisms such as X-ray magnetic circular (XMCD) and linear (XLD) dichroism, and high spatial and temporal resolutions. Up to now, most of the X-ray imaging research on spintronics has concentrated on ferromagnetic materials where both the geometric dimensions and characteristic timescales are accessible with synchrotron-based microscopes. However, the constant push towards the investigation of faster processes occurring at smaller length scales is now encouraging the development of novel time-resolved X-ray imaging techniques able to accommodate such requirements.

For X-ray imaging at the ultrafast timescales, free-electron lasers become a necessity. Here, we present the first results of the magnetic imaging setup recently commissioned at Maloja endstation of soft X-ray Athos beamline at SwissFEL (PSI, Switzerland). The experimental setup is based on X-ray holographic imaging [1], which is a lensless imaging technique where an interference pattern between the X-ray beams crossing the sample and a set of defined references is recorded on a 2D detector. Both amplitude and phase information of the sample transmission function then can be recovered by performing a Fourier transform of the recorded interference pattern. Since X-ray holography is an intrinsically drift free technique and a full-field microscopy technique, it is well suited for free-electron lasers, where shot-to-shot variations render scanning microscopy techniques unsuitable [2, 3]. Furthermore, the holographic reconstructions can be utilised as a first guess for a phase retrieval algorithm to further improve the image resolution.

Athos beamline of SwissFEL, with its 16 Apple-X undulators, provides a full control over X-ray polarisation (250 to 1800 eV, covering all the relevant edges of the magnetic elements). This makes it particularly suitable for ultrafast X-ray magnetic imaging, where XMCD and XLD are employed as contrast mechanisms for both ferromagnetic and antiferromagnetic samples, respectively. During the commissioning phase the static images of the labyrinth magnetic domain structures in thin films with perpendicular magnetic anisotropy were measured. This is the first step towards our goal – ultrafast time-resolved soft X-ray magnetic imaging. Moreover, we were able to demonstrate the feasibility of coherent correlation imaging [4] with single shot holograms at SwissFEL, which paves the way for sub-picosecond imaging of stochastic processes.

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Helicity-dependent population dynamics in the unoccupied states of Sb_2Te_3

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We investigated helicity-dependent population dynamics in the unoccupied states of the topological insulator Sb2Te3 upon excitation with near-infrared circular pump pulses of 1.55 eV for different crystal orientations. To this end, we employed combined time-and angle-resolved photoemission spectroscopy (tr-ARPES) and circular dichroism in the angular distribution (CDAD) to record photoelectrons E(kx,,ky) for the complete Dirac cone.

An excitation with 1.55 eV pump pulses results in an anisotropic population in the warped part of the topological surface state and higher-lying surface resonance states hybridizing with bulk states. The dichroism mainly shows a three-fold symmetric modulation that is connected to the bulk crystal symmetry of Sb2Te3 . Our results indicate a strong involvement of the out-of-plane spin texture of the Dirac cone, as we observe an energy-dependent modulation of the dichroic contrast in the warping region. For an excitation along the ΓK direction, we identify an anti-symmetric dichroic pattern, perpendicular to the laser incidence direction that is supported by one-step photoemission calculations and was similarly seen for a mid-infrared excitation in Ref. [1]. This anti-symmetric signature represents a signature of a macroscopic spin-polarized photocurrent. We show that the initially excited anisotropic population vanishes on a timescale of 200 fs, as electrons from higher-lying conduction band states scatter into the surface state.

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Electron dynamics in the van-der-Waals magnet $Cr_2Ge_2Te_6$

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The electron dynamics in the vicinity of the Brillouin zone center of $Cr_2Ge_2Te_6$ has been investigated by time-resolved ARPES. Optical excitation with a photon energy of 1.55 eV leads to a transient shift of bands in the region of the valence band maximum where multiple bands with different dispersions parallel and perpendicular to the surface overlap. Additionally, an excited electron population is created which is initially concentrated at the energy range of 0.7 to 1.3 eV above the Fermi level and continuously relaxes towards states at 0.5 eV within 1ps. According to their long lifetime of >3 ps we attribute these states to the conduction band minimum which theory predicts to be located close to the K point of the surface Brillouin zone [1,2]. The broad momentum distribution in the conduction band suggests enhanced scattering in the excited material.

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Magnetic circular dichroism in darkfield laser PEEM imaging

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Imaging magnetic circular dichroism (XMCD) for core level photoemission in photoemission electron microscopy (PEEM) is well established for magnetic domain characterization on the nanoscale [1,2]. This technique requires synchrotron radiation, which limits the temporal resolution when imaging dynamics of ultrafast processes. To enable ultrafast imaging on nanometer length and femtosecond time scales, laser-induced photoemission in combination with laser excitation can be used in a pump-probe fashion. However, MCD is required for magnetic imaging in near-threshold laser photoemission, which demands deeper understanding of the valence-band photoemission for magnetic thin films.

Here we report threshold MCD PEEM on epitaxial thin films of Fe(001) on MgO(001) and Ni(001) grown on Cu(001) near the photoemission threshold. By excitation with circularly polarized light from a tunable femtosecond laser (4 – 4.8 eV) or from a mercury discharge lamp, we report substantial MCD asymmetries for in-plane magnetization. These asymmetries exhibit a pronounced dependence on both photoelectron momentum and energy, accurately captured by relativistic one-step photoemission calculations [3] for the specific experimental conditions. Energy- and momentum-selective measurements in darkfield PEEM therefore enhance the contrast for domain imaging (up to 3% for Fe). Comparison of Fe and Ni thin films emphasizes the influence of their different band structures. This approach allows laser pump-probe experiments for revealing the dynamics in real space of in-plane and out-of-plane magnetization in ferromagnetic thin films on a femtosecond time scale.

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Narrow band THz frequency emission from micropatterned THz emitters

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THz frequency devices have gained significant attention driven by widespread application potential ranging from communication to nondestructive characterization techniques to astrophysics applications. Recent efforts by the community centered around Spintronic THz Emitters (STEs) have produced substantial experimental work alongside the development of theoretical models describing the relevant physics. Such STEs show great potential to complement conventional semi-conducting materials due to their straightforward and cost-effective fabrication techniques resulting in seamless on-chip integration with modern device technologies[1].

While typical STEs exhibit a very broad emission spectrum[2], in some cases a narrow emission band may be preferred.

In this work we present a micropatterned spintronic THz emitter based on CoFeB/Pt bilayer, which creates a THz burst corresponding to a narrow band emission at the burst's fundamental frequency. The samples were fabricated by DC sputtering and optical lithography. The THz emission measurements were performed with a wide band detection range up to 40 THz. These devices demonstrate a viable and straightforward method to produce controlled emission in a well-defined THz frequency band.

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Nonlinear and chaotic domain wall dynamics induced by ultrafast laser pulses

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In this study, we present a method to deploy ultrafast laser pulses to induce chaos and non-linearity in single-domain wall dynamics within a magnetic nanowire containing size defects i.e. two symmetrical notches. To quantify the perturbation effects of these ultrafast laser pulses, we expanded the existing unidimensional model to incorporate the influence of the laser pulses. Bifurcation diagrams and the largest Lyapunov exponents have been calculated for magnetic and laser shot properties to reveal the degree of chaos and optimal parameters needed to achieve a purely chaotic regime.

In addition, we demonstrate how such a nano-oscillator can be utilized as a neuromorphic hardware device stimulated by laser shots, producing non-linear responses capable of being trained for time series prediction. This device exhibits purely random domain wall dynamics, which can be exploited as the next technology dedicated to random number generators.

Gate-tunable magnons in CrSBr probed by time-resolved reflectivity

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The twodimensional layered antiferromaget CrSBr is a platform to study several coupled quasiparticles such as excitons, magnons and polaritons [1]. A femtosecond laserpulse can excite coherent magnons with frequencies of 24 and 34 GHz in this material [2,3]. These magnons can travel several micrometers, making them promising for future magnonic devices [2]. Their coupling to excitons allows us to monitor the spin waves employing time-resolved reflectivity measurements with a temporal resolution of less than 200 fs. Using a double-graphite gated device and applying a combination of perpendicular electric field and doping, we tune the magnon frequencies in few-layer CrSBr by up to 10%. We can connect the gate-dependent behavior of the magnon modes to the charge carrier density in the separate layers of the CrSBr device by employing gate-dependent photoluminescence measurements. In future experiments, this may allow us to bring the magnons into resonance with other magnonic or phononic modes, leading to hybridization [3].

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Stability and dynamics of magnetic skyrmions in FM/AFM heterostructures

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The recent growth in artificial intelligence has created a demand for energy-efficient, non-conventional computational techniques. Spintronics based devices are one of the possible targets as they can satisfy the required qualities such as low power usage, nonvolatility, high speed etc. Skyrmions are ideal information carriers due to their small size, ability to be closely packed, and their interaction with spin currents via spin transfer torque. Compared to magnetic domain walls, skyrmions require significantly less current, reducing power consumption. Skyrmions can be easily detected by magneto-transport effects such as magnetoresistance and electrical effects like the topological Hall effect. This ease in transport and detection makes them ideal for logic and data storage applications. A fundamental unit of a device utilizing skyrmions would be a skyrmion-racetrack, a channel in which the skyrmion can be stabilized and moved in a controlled manner. However, two major challenges exist in the design of such a racetrack: the skyrmion Hall effect (SkHE) which results in the vertical drift of Skyrmion under an applied current and the requirement of a strong out-of-plane field to stabilize the skyrmion. A practical solution to mitigate the SkHE is to couple two skyrmions antiferromagnetically so their individual Hall angles cancel each other out. Zero-field skyrmions in exchange bias systems, where the exchange bias field substitutes the external field, have already been experimentally realized. Numerical simulations can provide more fundamental information about the physics including the dependence of the skyrmion properties on the material parameters and the atomic arrangement at the interface. In this work, we use atomistic spin dynamics and Monte Carlo simulation to study the stability and dynamics of skyrmions at FM/AFM interfaces. We show that exchange bias skyrmions can be more stable than field-stabilized skyrmions due to interfacial effects. Additionally, we propose the design of a skyrmion transport device made by stacking two skyrmion-hosting layers between two AFM layers, where the skyrmion can move without Hall effects at speeds on the order of a few kilometers per second. This device can overcome existing limitations and has significant potential in spintronic

applications.

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The effect of intrinsic vs. extrinsic scattering in charge and spin dynamics of monolayer WSe_2

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Our goal is to understand the mechanisms causing spin scattering in TMD materials. While the mechanisms such as phonon-related scattering or exchange-related scattering are well studied and understood, the role of defect-related scattering remains unclear. To address this, we generate localized states in WSe2 using electron beam exposure. From the power dependence of localized excitons related to these defects, we estimate the defect density of 1012 cm-2. We then determine charge and spin dynamics in samples with controlled defect density using a combination of time-resolved reflectivity and Kerr measurements. While the time constant determining charge dynamics depends on the defect density only weakly, the spin accelerates when the defect density exceeds a critical value, likely the density of the intrinsic defects. We suggest a potential defect-mediated pathway. Overall, our results provide a view of previously unexplored spin depolarization mechanisms and suggest approaches to improve TMD-based spin/valleytronic devices.

Optical control of the magnetization emerging from the magnetic part of the optical field in the Landau-Lifshitz-Gilbert equation

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It is well known that the relaxation time of a magnetic moment towards an externally applied field scales inversely with the amplitude of the field, H, and the Gilbert damping parameter, α. Therefore, in ultrashort optical pulses, where H can temporarily reach high amplitudes of several Teslas, the Gilbert relaxation time can momentarily be extremely short, reaching even picosecond timescales. Recently, we have reported that ultrashort optical pulses may control the magnetization state by merely considering the optical magnetic field in the Landau-Lifshitz- Gilbert (LLG) equation [1]. The principle behind the interaction is that the magnetization is incrementally affected within each optical cycle, such that a significant net torque can build up over the entire pulse duration in typical experimental conditions. We find the strength of the interaction to be determined by $\eta = \alpha\gamma/f$opt, where fopt is the optical frequency and γ is the gyromagnetic ratio. Accordingly, the loss of spin angular momentum to the lattice, which is represented by α, is key to the interaction. Moreover, we show that under circularly polarized (CP) pulses, the polarity of the optically-induced torque is determined by the optical helicity. From a quantitative analysis, we find that a sizable effective out-of-plane field is generated, which is comparable to that measured experimentally in ferromagnet/heavy-metal (FM/HM) material systems [2]. Our results provide an additional torque to the all optical helicity-dependent switching (AO-HDS) [3,4] that has been considered on grounds of thermal [5], photomagnetic [6], and optomagnetic [2,7] mechanisms. Moreover, our conclusions are supported by an analytical two-level-system model for the magnetization dynamics in which we demonstrated the coupling between the helicity of the driving field and the polarity of the resulting torque [1].

Figure 1(a) presents the calculation for experimental settings following Refs. [7,8], where only the optical magnetic field is substituted into the LLG equation. In agreement with the observations of the AO-HDS, pulses of the opposite helicity induce an opposite transition, as shown in Fig. 1(b).

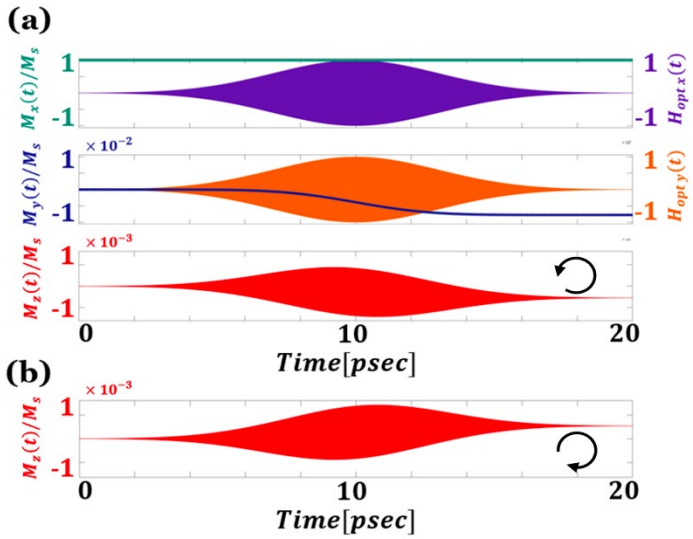

Fig. 1. (a) Magnetization reversal induced by an RCP Gaussian pulse for α = 0.035, M_S = 3×10^{5}A/m, η = 2.5×10^{-4} and t_{peak} = 10 psec. Top and middle panels depict the temporal evolution of the x and y components of $\vec{M}$ and $\vec{H}$ in normalized units. Bottom panel depicts M_Z/M_S . (b) M_Z/M_S , for the application of an LCP pulse.

Our results show the relevance of the LLG equation to the optical regime and attributes the optically-induced torque measured in the AO-HDS also to the magnetic part of the optical beam.

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Ultrafast control over Spin-valley dynamics in TMDC heterostructures

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In this poster, I will present our results on the ultrafast spin manipulation at the interface of 2D Transition Metal Dichalcogenides (TMDCs). We study the effect of out-of-plane electric field on the spin-valley dynamics in the MoS2/MoSe2 heterobilayer at low temperatures. Ultrafast Time-resolved Kerr Rotation (TRKR) and time-resolved reflectivity measurements reveal a new scattering channel for spin-polarized carriers in this system. Critically dependent on the temperature and excitation power used, we discover rise-times much slower than the typical ultrafast charge transfer in these type-II heterostructures. While most previous studies have focused on spin-valley lifetimes, we present a tunable channel for rise-dynamics of spin-valley polarization over an order of magnitude in this system.

Electronic structure for large-scale chiral magnet

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In solid systems, the existence of competing interactions, including Heisenberg exchange coupling, magnetic anisotropy, Dzyaloshinskii-Moriya (DM) interaction, dipole interaction, and others, leads to the emergence of various rich spin textures and many intriguing magnetic structures such as helical magnetism, vortex, and the well-known skyrmions [2]. These magnetic structures typically scales at the nanometer or sub-micrometer level and usually include thousands of unit cells, making first-principles calculations based on density functional theory (DFT) impossible. However, based on the ultra long range (ULR) DFT method developed by T. Müller et al [1]., it has become possible to study these large-scale special magnetic structures from a first-principles perspective. By relying on the ULR ansatz, we can further map the electronic structure details of these large-scale magnetic structures, such as band struc tures, back to the first Brillouin zone, thus enabling detailed and even quantita tive first-principles studies of these large-scale magnetic systems. In this work we have extended the ULR to study such large length scale chiral spin structure in microscopic detail from ab-initio theory.

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Strain-induced pseudomagnetic field to study excitons in 2D semiconductors

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Excitons in transition metal dichalcogenides (TMDs) exhibit large binding energies and long coherence times, making them promising for optoelectronic and spintronic applications. A key feature of TMD excitons is direct optical access to pseudospin, associated with valleys in the band structure. We present a novel technique to control pseudospin using tunable uniaxial strain in TMDCs. Our experiments demonstrate Zeeman splitting of exciton peaks in MoSe2 and WSe2, with shifts up to 1.2 meV, and the generation of large pseudomagnetic fields reaching 30 Tesla. We also show polarization of pseudospin and Larmor precession, confirming the 'magnetic' nature of the field. Furthermore, by manipulating doping levels, we reveal the transition of charged excitons from fermionic to bosonic behavior. This method significantly advances the control of pseudospin dynamics, facilitating the implementation of high pseudomagnetic fields on-chip and opening new avenues for valleytronics and quantum technologies.

Ultrafast control of Rashba interactions in a TMD heterostructure

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We unveil a novel spin relaxation mechanism in a TMD heterobilayer induced by the Rashba effect. In such a system, Rashba interactions arise from an out-of-plane electric field due to photo-generated interlayer excitons inducing a phonon-assisted intravalley spin relaxation. We develop a theoretical description based on a microscopic approach to quantify the magnitude of Rashba interactions and test these predictions via time-resolved Kerr rotation measurements. We demonstrate robust and tunable Rashba interactions on an ultrafast time scale, where the spin dynamics is controlled by varying the laser fluence. Our work identifies a previously unexplored spin depolarization channel in heterostructures which can be used for ultrafast spin manipulation.

Modelling nonlinear effects in nonlinear photonic devices

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Nonlinear photonics, where light-matter interactions deviate from linear behaviors, accommodates a plethora of advanced modern optical technologies, enabling a wide range of applications [1-2] from quantum computing to nonlinear imaging, photonic logic gates, etc. Despite these advantages, studying nonlinear photonic devices involves several challenges due to the complex nature of light-matter interactions in material media. Accurately obtaining and understanding material parameters, such as the nonlinear response to light, adds significant complexity to designing the next-generation photonic devices. In this work, we have designed a nanometer-scale photonic device made of silicon and studied its nonlinear optical effects using our in-house code, NLIME (Non Linear Maxwell Equation solver). We utilized time-dependent density functional theory to extract the nonlinear response of silicon to a given optical pulses using ab-initio ELK code. The nonlinear effects were modeled by solving the nonlinear Maxwell equations. Furthermore, we thoroughly discuss the methods used to solve Maxwell's equations to determine the electromagnetic steady-state of linear photonic nanodevices, and how these methods extend to nonlinear regimes. Our study provides a comprehensive approach to understanding and designing the nonlinear photonic devices, paving the way for advancements in optical technologies.

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All about two-dimensional materials

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Two dimensional (2D) materials have introduced to condensed matter both new degrees of freedom – such as the valley index and moire length – as well as dramatically enriching physics well known from 3D materials. This has birthed new thriving fields, for example valley- and twist-tronics, devoted to exploring and utilizing the valley and moire freedoms, as well as provided great new impetus to existing fields such as spintronics and topological physics. While the early

hopes of the field in the years immediately following the discovery of graphene in 2005 of an "all carbon coherent electronics" have not been fulfilled, the potential for paradigm shifts in technology from 2D materials remains. In this overview talk, I will review the fundamental physics of 2D materials from graphene to the latest investigations involving light-matter interaction.

I will first explore valley physics, the role of Berry curvature, and effects such as the anomalous Hall effect that flow from these, paying particular attention to the transition metal dichalcogenide and graphene families and the (as yet unrealized) potential that this holds for applications. The remarkable physics of deformation in graphene, and the much richer physics of few layer assemblies of "moire materials", both represent examples of an emergent electronic structure in 2D materials that has no counterpart in its 3D cousins. I will review both of these paying attention to possible applications involving one of the latest classes of 2D materials to be discovered, 2D magnets.

Finally, I will describe early attempts to control quasi-particles in 2D materials by ultrafast pulses of laser light – both free carriers as well as the rich excitonic physics of 2D semiconductors. Early explorations of this "femtosecond universe" indicate a rich potential for uncovering a remarkable non-equilibrium yet coherent physics of 2D materials.

Ultrafast dynamics in the ferroelectric and antiferromagnetic phase of a van der Waals multiferroic NiI_2

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Multiferroics are a class of materials in which different ferroic properties – such as electricity and magnetism – are coupled [1]. When taken to the two-dimensional (2D) limit, the strong light-matter interactions inherent to 2D systems are further enhanced by the magneto-electric interaction, enabling a direct coupling of the electric component of light to magnetic degrees of freedom. This in turn, makes 2D-multiferroics a particularly appealing platform for realizing highly efficient optical control of magnetism [2].

In the recently discovered 2D van der Waals (vdW) multiferroics NiI2 and CoI2, the onset of magnetism promotes the emergence of ferroelectricity, which has been observed down to the monolayer limit [3, 4]. Here, we study the antiferromagnetic and ferroelectric phase of NiI2 -with a combination of optical techniques, including second harmonic generation, linear dichroism and magneto-optics to image the domain structure. Following a detailed characterization of the phases in equilibrium, we employ 100 fs light pulses to study the ultrafast dynamics of NiI2 using pump-probe magneto-optical techniques. By tuning polarization, fluence, and wavelength, we aim to enable ultrafast optical control of magnetism in 2D vdW multiferroics, in which ferroelectric and magnetic orders coexist and are mutually coupled.

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Insights in the magnetic excitation spectrum of Fe_3GeTe_2 from femtosecond bandstructure dynamics.

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Iron Germanium Telluride (FGT) is a van der Waals metallic ferromagnet that has shown a wealth of exotic phenomena, ranging from 2D ferromagnetism with gate-tunable Curie temperature [1] and skyrmion formation[2], to heavy fermion behaviour[3]. Despite its interest for advanced manipulation of electronic and spin states in devices, its electronic structure and the microscopic interactions giving rise to magnetic order are still poorly understood: evidence exists of both itinerant and localized magnetic behaviour, an anomaly in a purely 3d electronic system. By employing time-resolved angle-resolved photoemission spectroscopy (trARPES), we observe clear evidence of the closure of the Stoner exchange gap, a clear indication of an itinerant character of the electronic excitations. This is in contrast with previous results of quasi-equilibrium studies, which ascribed FGT demagnetization to localized excitations[4]. We also observe, by employing frequency domain ARPES, the impact of phononic excitations, resulting in an A1g phonon strongly coupled with the bandstructure at the Brillouin zone centre.

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All-optical control of spins in van der Waals magnets

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Recently discovered two-dimensional (2D) van der Waals (vdW) magnets have revealed new opportunities for control of magnetism via mechanisms such as strain, voltage and the twistronics. Ultrafast laser pulses provide the fastest way of manipulation of magnetic properties but their influence on the spins in 2D vdW magnets is largely unknown. In this talk, I will highlight the latest advancements in all-optical control of 2D vdW magnets, focusing specifically on our experimental findings with semiconducting ferromagnets CrI3 [1] and Cr2Ge2Te6 [2].

Our research demonstrates that integrating a thin CrI3 flake with a monolayer of transition metal dichalcogenide WSe2 allows for both helicity-dependent and helicity-independent all-optical switching (AOS) down to a single laser pulse [1]. The AOS can be explained by the spin-dependent charge transfer across the CrI3/WSe2 interface, which is expected to begin within a few to hundreds of femtoseconds and should allow for control of magnetic properties on unprecedented ultrafast timescales. Next, I will show that optical pumping can lead to formation of various spin textures, including reversible transformations between stripe and bubble/skyrmion phases in Cr2Ge2Te6 [2].

Finally, I will discuss thickness dependent remagnetisation dynamics in Cr2Ge2Te6 observed via time-resolved beam-scanning Kerr microscopy. Our findings reveal that reducing the thickness of the 2D magnet enhances heat dissipation to the substrate, significantly shortening the magnetization recovery time from several nanoseconds to a few hundred picoseconds. Finally, I will demonstrate how the low cross-plane thermal conductivity of Cr2Ge2Te6 can be exploited in retrieving the original domain structure, even after a complete loss of magnetic order in the uppermost layers.

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[2] M. Khela, M. Dąbrowski, et al. Nat. Commun. 14, 1378 (2023)

Optical Excitation of Spin Polarization in the Altermagnet RuO_2

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The discovery of altermagnets as a new class of magnets opens new ways to access and manipulate the spin degree of freedom in magnetic materials with compensated spin structures. Ultimately, this opportunity arises from the momentum-split band structure of altermagnets that was recently observed in experiments and is responsible for novel current-driven spin functionalities, as spin-splitter currents.

In this contribution, we explore the ultrafast response of altermagnetic materials after optical excitation with fs light pulses. For the exemplary case of RuO2, we employ theoretical ab-initio calculations to predict the spin polarization of the optically excited carriers. We find a clear correlation between the direction of the light polarization vector of the exciting light pulses with respect to the RuO2 crystal structure and the sign and magnitude of the optically generated spin polarization. Our theoretical predictions are confirmed by time-resolved MOKE experiments, which demonstrate the existence of highly spin-polarized carrier distributions in RuO2 for characteristic excitation geometries, i.e., light polarization vector orientation. In this way, our findings show a clear pathway to optically generate and control spin polarized charge carriers in materials with altermagnetic band structures.

Optical excitation effects in altermagnetic RuO_2

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Recently, altermagnets have gained interest due to their novel magnetic properties that combine those of known ferromagnets and antiferromagnets. Since altermagnetic materials exhibit momentum dependent spin polarization in reciprocal space without net magnetization, they are particularly well suited as potential building blocks for faster and more robust spintronic devices [1,2]. These properties also make altermagnets an interesting candidate for the study of optical properties on ultrashort timescales. In principle, the existence of spin-polarized states allows a direct optical manipulation of the spin system, despite being in a fully compensated material. Therefore, we focus on the optical response of these materials as a key property. We model the optical excitation of the d-wave altermagnet RuO2 and investigate how the underlying symmetries affect the spin dependent populations of excited states. We employ a hybrid approach that combines Density Functional Theory (DFT) [3] and optical transition probabilities in a Fermi's Golden Rule approach [4,5] to simulate the excitation with momentum, spin and band resolution. The calculations give rise to a polarization angle dependent spin polarization induced by the laser field, which is supported by time-resolved MOKE measurements [6].

References

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THz lattice and spin dynamics in a 2D antiferromagnet driven by d-d transitions

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d-d transitions between localized 3d states of transition metal ions within crystalline solids play a pivotal role in diverse phenomena across solid-state physics, materials science, and chemistry. They contribute to the optical properties of transition metal oxides, catalytic activity on oxide surfaces, high-temperature superconductivity, and magnetic behaviors, facilitating spin-crossover transitions and linking optical excitation to quantized phenomena such as phonons and magnons. The discovery of unique effects in 2D antiferromagnets, such as electron-phonon bound states, sub-terahertz frequency magnon modes, and hybridized phonon-magnon modes, highlights the complex phenomenology driven by d-d transitions.

I will discuss our recent investigations into FePS3, a van der Waals antiferromagnetic semiconductor scalable to the 2D limit. Initially, pump-probe magneto-optical measurements were conducted to observe laser-driven lattice and spin dynamics. Pumping in resonance with a d-d transition induced a coherent phonon mode oscillating at 3.2THz. This mode is connected to long-range magnetic order and enables optical excitation of a phonon-magnon hybrid mode in an external magnetic field [1]. Additionally, we utilized time-resolved angle-resolved photoelectron spectroscopy (tr-ARPES) to probe the electronic structure [2] and to capture the ultrafast dynamics of selected spin-allowed and spin-forbidden d-d transitions in FePS3 [3]. The insights from magneto-optical experiments, juxtaposed with ARPES findings, shed light on the intricate quasiparticle dynamics underpinning d-d transitions in FePS3, offering a deeper understanding of their role in quantum material behaviors.

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