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WORKSHOP

"FUTURE SPINTRONICS: CHALLENGING THE LIMITS"

**COMRAD Innovative Training Network** • October 08-10, 2024 Barcelona (Spain)



The **COMRAD** Innovative Training Network (Cold Opto-Magnetism for Random Access Devices) is proud to announce the workshop **"Future Spintronics: challenging the limits"** focused on the approaches that challenge established fundamental limits faced by nowadays magnetic recording, g, data storage and information processing technologies. In particular, the workshop will cover several recently emerged and rapidly developing fields in spintronics, including attosecond spintronics, orbitronics, superconducting spintronics and altermagnetism.

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# Ultrafast On-Chip Spintronics

#### J. Bokor

Department of Electrical Engineering and Computer Sciences, University of California, Berkeley, California, USA

#### jbokor@berkeley.edu

The field of spintronics involves the study of both spin and charge transport in solid-state devices. magnetism involves the Ultrafast use of femtosecond laser pulses to manipulate magnetic order on subpicosecond time scales, including helicity-independent all-optical switching. We have united these phenomena by using picosecond charge current pulses generated on-chip using an ultrafast photoconducting (Auston) switch (Fig. 1) to induce deterministic, repeatable ultrafast reversal of the magnetization of a ferromagnetic GdFeCo thin film [1]. Using 9 ps duration current pulses, the magnetization reverses in ~10 ps, which is more than one order of magnitude faster than any other electrically controlled magnetic switching, and demonstrates a fundamentally new electrical switching mechanism that does not require spinpolarized currents or spin-transfer/orbit torques. (Fig. 2) Furthermore, the energy density required for switching is low, projecting to only 4 fJ needed to switch a (20 nm)<sup>3</sup> cell.

This ultrafast magnetization reversal phenomenon through a nonequilibrium thermal excitation is primarily limited to Gd based ferrimagnets, such as the GdFeCo alloy used in the experiments shown in Figs. 1 and 2. In order to integrate this fast switching with a readout, a magnetic tunnel junction exhibiting high tunneling magneto resistance (TMR) is desired. Yet, the TMR value reported for devices using GdFeCo is too small ( $\approx 0.6\%$ ) for practical applications [2]. Thus, it is of great interest to switch a ferromagnet with helicity independent optical pulses, which could then be implemented as a storage layer in a high TMR memory cell.

We have shown how to transfer the ultrafast switching of GdFeCo to a ferromagnet (in our case Co/Pt multilayers) using Ruderman–Kittel–Kasuya– Yosida (RKKY) exchange coupling mediated HI-AOS of the ferromagnet layer driven by the HI-AOS of the ferrimagnet layer [3, 4]. This technique is generally applicable to other ferromagnets that could then be used for MTJ readout of the state of the switched magnetic structure with high TMR.

We also show that 6-10 ps duration electric current pulses can be used to directly and deterministically switch the out-of-plane magnetization of a ferromagnetic thin cobalt film via spin–orbit torque (SOT) [5]. The current pulses were applied to a heavy metal/ferromagnet thin film heterostructure in the presence of an in-plane symmetry breaking magnetic field. Depending on the relative current

pulse and in-plane magnetic field polarities, we observe either SOT switching or an ultrafast demagnetization and subsequent recovery, but no switching. (Fig. 3) The short current pulse induces the ultrafast demagnetization of close to 30% due to transient Joule heating. This heating plays a crucial role in promoting the SOT switching in the presence of the in-plane magnetic field. Nevertheless, we also project low energy (~fJ range) for switching of a (20 nm)3 cell using this mechanism. We use a macromagnetic simulation model coupled with an ultrafast heating model to analyze the effect of ultrafast thermal anisotropy torque (intimately related to the ultrafast demagnetization) and current-induced SOT the observed dynamics. Good agreement in between our experimental results and the macrospin model shows that the switching dynamics are coherent rather than involving domain nucleation and growth, even though our device dimensions are as large as 4 X 5 um2.

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#### **Figures**



**Figure 1.** (A) Schematic of electrical switching experiment. The photoswitch is illuminated with laser pulses while being biased with a dc source. Magnetization dynamics of GdFeCo is monitored with time-resolved MOKE. Left: Optical image of the photoswitch. During laser illumination, photoexcited carriers in low-temperature GaAs conduct current across the gap, generating a transient electrical pulse propagating in both directions. Right: Optical image of the GdFeCo section of CPS. Scale bars, 20  $\mu$ m. (B) Calculated temporal current density profile through the GdFeCo section, based on the temporal current profile measured with a Protemics Spike probe positioned 1 mm before the GdFeCo section.



**Figure 2.** Single-shot optical and electrical switching of GdFeCo. (A) Differential MOKE images of bare GdFeCo film after sequential 6.4-ps optical pulse irradiation. Absorbed fluence is 1.8 mJ/cm2. After each optical pulse, the magnetization of GdFeCo toggles to the opposite direction. The contrast indicates change in magnetization. (B) Differential MOKE images of GdFeCo CPS section after sequential 9-ps electrical pulse excitation. After each electrical pulse, the magnetization of GdFeCo toggles. Yellow and blue dashed lines indicate gold CPS and GdFeCo sections, respectively. Scale bars, 5  $\mu$ m.



**Figure 3.** Ultrafast current pulse–induced magnetization switching dynamics. The approximately 9 picosecond current pulse–induced time-resolved magnetization dynamics measured in the presence of (A) positive and (B) negative in-plane symmetry-breaking magnetic field and reversing the direction of the current pulses starting from positive and negative magnetic saturation. The dotted lines show the theoretical analysis by solving a macroscopic LLG equation and combining ultrafast one-temperature heating model due to a 9-ps current pulse with a current density of  $\pm 7.3 \times 10^{12} \text{ A/m}^2$ .

### Non-equilibrium effects in superconducting thin films probed by time-domain THz spectroscopy

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Chiara Ciccarelli <sup>1</sup>University of Cambridge, UK

cc538@cam.ac.uk

Superconducting logic holds the promise of significant improvement compared to traditional semiconductor logic, both in terms of computational power and energy consumption, due to the lack of ohmic losses when transferring bits. However, most of these advantages are negated by the lack of a suitable cryogenic memory that can be coupled to the superconducting logic.

The ambition of superconducting spintronics is to integrate magnetic and superconducting elements in a unique device that combines permanent storage and low power logic characteristics. The new opportunities that superconductivity offers to spintronics were realised only quite recently. Instead, conventional superconductivity and magnetism were long believed to be mutually exclusive since in s-wave superconductors, Cooper pairs are in a singlet state and are therefore inadequate for carrying spin-information. But recently, new discoveries have shown new routes for superconductivity and magnetism to co-operate, enabling novel device concepts based on the interplay between spin, charge and superconducting phase coherence. This includes the condensation of spin-polarised triplet Cooper pairs in conventional superconductors interfaced by heavy metals [1-5].

THz transmission spectroscopy is an extremely powerful tool to characterise conventional superconductors because the superconducting gap falls within the bandwidth of the THz pulse that is generated for example by optical rectification in a ZnTe crystal. The great advantage of THz transmission spectroscopy, in comparison to other gap-spectroscopy techniques such as STM, resides in the fact that it is time-resolved. We can therefore apply it to quantify the melting/recovery dynamics of the superconducting condensate subject to a fast perturbation and to detect new transient superconducting correlation states. In this talk I will give an overview on the non-equilibrium studies carried in superconductors via time-domain spectroscopy and show our very recent effort in using time-domain THz spectroscopy to resolve the interactions at the interface between thin-films superconductors and normal metals and how these affect the superconductor's dynamics.

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# Neuromorphic and probabilistic computing with spintronic devices

#### Giovanni Finocchio<sup>1</sup>

<sup>1</sup>University of Messina, Viale F. Stagno D'Alcontres 31, Messina, Italy

gfinocchio@unime.it

The development of more efficient and high performance spintronic devices and the efforts to have co-integration of spintronics with CMOS technology is driving the development of hybrid CMOS-spintronic solutions for applications where one can take the advantages of both technologies while minimizing their disadvantages. In this talk, I will present our recent developments on new potential applications of magnetic tunnel junctions (MTJs) in neuromorphic and probabilistic computing. For neuromorphic computing, I will discuss how to implement spiking neurons and on-chip training taking advantage of MTJ properties.

I will also focus on probabilistic computing with probabilistic-bits (p-bits) which is emerging as a computational paradigm able to be competitive in solving NP-hard combinatorial problems. I will show how to map hard combinatorial optimization problems (Max-Sat, Max-Cut, Traveling Salesman problem) into probabilistic Ising machine. We will discuss the potential of advanced annealing schemes comparing simulated annealing, paralleltempering, and simulated-quantum-annealing and how it will be possible to implement an efficient probabilistic co-processor.

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### HAMR and Beyond: a journey from Research Concept to Data Storage Products

#### **Ganping Ju**<sup>1</sup>, Mike Seigler<sup>2</sup>, Stephanie Hernandez<sup>3</sup> Mike Kautzky<sup>2</sup>, Mark Gubbins<sup>4</sup>, Jan-Ulrich Thiele<sup>1</sup> and, Ed Gage<sup>3</sup>

<sup>1</sup>Seagate Recording Media Research Center, Seagate Technology, 47488 Kato Rd, Fremont, CA 94538, USA, <sup>2</sup>Seagate Recording Head Group, Seagate Technology, Bloomington, MN 55408, USA <sup>3</sup>Seagate Research Group, Seagate Technology, Bloomington, MN 55408, USA, <sup>4</sup>Seagate Technology, Springtown, Northern Ireland, BT480LY Ganping.A.Ju@Seagate.com

#### Abstract

Data is growing faster than the world's ability to store it. In 2024, the people of Earth will generate 30 zettabytes of data each year-but only 2 zettabytes of storage capacity are manufactured each year. To address such demand for data storage, Heat Assisted Magnetic Recording (HAMR) has proven to be the primary successor to perpendicular magnetic recording - productization is well underway in 3 TB/disk configurations [1,2]. HAMR addresses the magnetic recording "trilemma" [3-5] more effectively than other energy assist schemes by incorporating high anisotropy FePt-based media in hard disk drive (HDD) platforms. FePt media can be scaled to small grain sizes, enabling areal density growth while maintaining thermal stability. HAMR heads feature a near field transducer that converts far field light incident from a laser to near field light at the media surface [3-5]. These apply high temperature to FePt media grains during the write process, thereby lowering the anisotropy of the grains enough to be written by the magnetic fields supplied by the HAMR head.

High areal density HAMR demonstrations have been published through the years [6],[7], including recent demonstrations presented at TMRC 2024, experimental spin-stand recording data in excess of 5.5 TB/disk a demonstration of 4 TB/disk in a fully formatted, factory-processed drive [8].

In the talk, we will present the research and development journey for HAMR technology, from the research concepts to the data storage products. Potential prospects will also be discussed on technologies extending the recording density growth and critical challenges before it could become a viable technology path.

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**Figure 1.** Critical Technology Innovations enabling Seagate's MOZIC3+ HAMR Product platform.



**Figure 2**. Areal Density Capability (ADC) over time for Seagate longitudinal recording products, perpendicular recording products, and HAMR laboratory demonstrations.

# Photonic Integration from a spintronic and magnetic perspective

#### Bert Koopmans

<sup>1</sup>Department of Applied Physics / Eindhoven Hendrik Casimir Institute, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands b.koopmans@tue.nl

This presentation will address the opportunities for the integration of spintronics with photonics (Figure 1). 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). 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. Other motivations for such integration come from the inherent difficulty to make a dense, fully photonic memory, and challenges faced in realizing an integrated optical isolator, for which solutions might be provided by magnetism and magneto-optics, respectively.

After a brief introduction into photonic integration different integrated and identifying photonic platforms, opportunities for spintronic-photonic integration will be sketched. Next, recent progress on scientific issues that are considered key for realizing the envisioned technology will be discussed [1]. Examples of progress towards integrated spintronic-photonic devices will be presented, including current-induced domain wall motion in Pt/Co/Gd-based conduits that display efficient AOS [2] with domain-wall velocities over 2000 m/s [3], AOS of MTJs [4], as well as on-chip magneto-optical reading of 300 x 400 nm<sup>2</sup> magnetic elements structured on top of InP photonic waveguides [5]. Moreover, a first proof-of-concept demonstration of on-chip AOS as realized in a Si<sub>3</sub>N<sub>4</sub> photonic integrated circuit will be discussed. Finally, to further reduce the device footprint and increase data densities, near-field plasmonic approaches will be inevitable. Recent device simulations on using photonic cavities and plasmonic nano-antennas for sub-diffraction limited optical writing and reading [6], as well as using wavelength division multiplexing strategies [7], provide inside into pushing the ultimate performance.

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**Figure 1.** Schematic representation of spintronic-photonic integration.

#### Ultra-fast all optical switching in spintronic devices

J. Gorchon<sup>1</sup>, T. Hauet<sup>1</sup>, M. Hehn<sup>1</sup>, J. Hohlfeld<sup>1</sup>, G. Malinowski<sup>1</sup> and S. Mangin<sup>1</sup> <sup>1</sup> Université de Lorraine, CNRS, Institut Jean Lamour, F-54000 Nancy, France Stephane.mangin@univ-lorraine.fr

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 subpicosecond 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 helicityindependent 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 singlepulse 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 currentinduced 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.

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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**Figure 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]

# Neuromorphic Computing for Science

#### Johan H. Mentink

<sup>1</sup> Radboud University, Institute for Molecules and Materials, Nijmegen, The Netherlands

#### johan.mentink@ru.nl

Solving the major challenges in computational science rapidly increases the demand for high-end digital computing infrastructures, leading to unsustainable growth of the corresponding energy costs. Neuromorphic Computing is an emerging computing paradigm that co-locates memory and processing and thereby enables faster and much more energy-efficient computations. Therefore, neuromorphic computing might enable a more sustainable future of scientific computing.

However, understanding how to identify potential advantages of neuromorphic hardware is a nontrivial task and may depend strongly on the computational problem at hand. In this talk, we will present our recent efforts aimed at identifying computational advantage for quantum simulation of magnetic systems with artificial neural network networks [1,2]. This computational task features two widely used computational problems: (i) the inference of feedforward neural networks and (ii) Markov-chain Monte Carlo simulation. We explore the potential advantage of these tasks on two established neuromorphic paradigms: in-memory computing [3] and stochastic Ising machines [4].

Taking the energy cost and latency as key performance metrics, we predict that computational advantage appears for sufficiently large systems, which is intimately related to the massive parallelization that is present in the neuromorphic hardware. We also find that computational advantage depends strongly on the properties of the network being realized in physical hardware. Hence, no universal computational advantage exists, and we present a methodology to quantify the possible advantage of neuromorphic Monte Carlo sampling directly from simulations on digital hardware.

We expect that our findings will stimulate further exploration of neuromorphic hardware for computational science, which ultimately will lead to the breaking of existing computational barriers. Moreover, given the generality of the computational tasks, our findings may also stimulate adoption of neuromorphic hardware for non-scientific applications.

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# Terahertz coherent magnonics in canted antiferromagnets

Rostislav Mikhaylovskiy <sup>1</sup> Lancaster University

r.mikhaylovskiy@lancaster.ac.uk

Magnonics aims to employ quanta of spin waves, magnons, to carry, transport and process information, avoiding the dissipation of energy inherent to electronics. Experiments on magnons in regular (ferro)magnets have yielded demonstrations of basic logic devices, albeit macroscopic (mmscale) in size and operating at GHz frequencies. Recently, the spotlight has shifted towards the use of antiferromagnets, in which neighbouring spins are aligned antiparallel to each other. This alternating order leads to significantly higher spin wave propagation velocities and might enable devices operating at terahertz (trillion of hertz) clock-rates. However, the absence of the net magnetisation also makes antiferromagnets magnetically 'invisible': it is and hard to detect influence the verv antiferromagnetic Yet. order. in some antiferromagnets strong spin-orbit coupling results in canting of the spins, thereby producing net magnetization. The canted antiferromagnets combine antiferromagnetic order with phenomena typical for ferromagnets and hold a great potential for spintronics and magnonics. In this way they can be identified as closely related to the recently proposed novel class of magnetic materials, called altermagnets. In my talk I will discuss a new canted antiferromagnets and functionality of altermagnets for magnonics and show that these materials facilitate mechanisms allowing to nonlinearly generate. detect and convert propagating magnons at the nanoscale [1-3].

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# Orbital Magnetism By Light

#### Yuriy Mokrousov<sup>1,2</sup>

<sup>1</sup>Peter Grünberg Institut, Forschungszentrum Jülich, Jülich, Germany <sup>2</sup>Institute of Physics, University of Mainz, Germany

y.mokrousov@fz-juelich.de

Orbital degree of freedom in solids is attracting ever increasing attention owing to possible applications the diverse in areas of magnetization control and angular momentum current generation [1,2]. So far, the properties of non-equilibrium orbital magnetism and orbital currents has been explored predominantly in the regime of weak perturbations, while the orbital physics is expected to be particularly rich in the realm of excitations brought by light. From the fundamental point of view, the interaction of light with matter is mediated predominantly by the orbital properties of quantum states. As we shall see, in various materials this gives rise to strong orbital response in terms of orbital magnetization, which imprints a subdominant spin response, discussed and studied intensively in the past. We will show that when structural inversion symmetry is broken either intrinsically, upon deposition, or by magnetism, strong orbital coupling to the electric field of the pulse will give rise to currents of orbital angular momentum, with optical currents of spin dragged by spin-orbit interaction [3]. The currents of angular momentum can be detected in THz emission experiments [4], providing a unique insight into the temporal and spatial properties charge, spin and orbital of interconversion processes [5]. We will demonstrate that among various material structurally platforms, complex antiferromagnets present a unique niche for achieving best orbital performance with direct implications for the magnetic order control [6,7], will and discuss the prospects and repercussions light-induced of orbital magnetism.

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### Superconducting Electronics and Spintronics for Energy-Efficient Computing

Oleg A. Mukhanov SEEQC, 150 Clearbrook Rd. Elmsford, NY, USA

omukhanov@seeqc.com

According to the International Energy Agency (IEA), electricity consumption from data centers, artificial intelligence and the cryptocurrency sector could double by 2026 and exceed 1,000 TWh [1]. Reducing the power dissipation at the highest processing speed is the central objective for any information processing circuit technology.

Superconducting Single Flux Quantum (SFQ) digital electronics based on Josephson junction circuits with its unparallel clock speed at tens to even hundreds of GHz at very low-power dissipation [2,3] has been long considered as the most promising technology for variety of high-end applications including high-performance computing and quantum computing. However, the low integration density and the lack of high capacity, high operational margin superconducting memory technology compatible with digital superconducting circuits has been a major limiting factor in utilizing superconducting electronics for many decades.

Superconductor/ferromagnet heterostructures including Josephson junctions with ferromagnetic materials are capable of addressing these challenges and even potentially enable new applications in neuromorphic, reservoir, and quantum computing. In superconductor-ferromagnet Josephson junctions, the spin-singlet Cooper pairs perform phase oscillations as they go through a ferromagnetic material in the presence of the exchange field. As a result, the phase difference across such Josephson junctions oscillates between 0 and  $\pi$  as with the thickness of the ferromagnetic material [4]. This phenomenon is used to build the static phase shifters and switchable junctions in superconducting digital circuits to increase their operational tolerances, reduce total bias current, and enable new functionalities.

In particular, half-flux-quantum (HFQ) circuits are based on  $0-\pi$  superconducting quantum interference devices (SQUIDs). Comparing the HFQ toggle flip-flop (TFF) with its rapid single-flux quantum (RSFQ) counterpart under the same fabrication process, it is anticipated that the HFQ TFF can achieve ~70% reduction in both static and dynamic power dissipation [5].

Another area where superconductor/ferromagnet heterostructures can make an impact is the development of non-reciprocal devices – superconducting diodes. This can be used in dcbiased SFQ circuits to avoid the rise of total bias current with the number of cells limiting the circuit scalability. This is also important for SFQ circuits used for qubit control and readout while minimizing thermal load and electromagnetic noise [6]. The developed superconducting diodes recently demonstrated high efficiency and tunability [7]. This enabled the construction of the first superconducting rectifiers with superconducting diodes made of thinfilm bilayers of V/EuS. The rectifier operated with frequencies up to 40 kHz and efficiencies up to 43%. The efficiency of the individual diodes reaches up to ±50% by combining the effect of edge asymmetry in the superconducting vanadium (V) and stray fields from a ferromagnetic insulator (EuS) in the V/EuS bilayers [7].

The most significant impact is expected in using superconductor/ferromagnet elements for memory applications. If a junction contains multiple ferromagnetic layers whose relative magnetization directions can be controlled by application of a relatively small magnetic field (e.g., using current lines) or by spin torque, then this device can serve as a memory element. There are several challenges in ferromagnetic material choices to ensure a success [4]. Multiple memory cell designs were proposed and implemented. The key is to design a memory element which can be integrated to form a density, highly energy hiah efficient. fullv addressable memory array compatible with SFQ digital technology.

A hybrid vertically integrated superconductingferromagnetic device can also serve as an addressable memory cell, in which the ferromagnetic junction stores the information, while superconducting Josephson junction acts as a readout device [8]. Electric current applied along the superconducting electrode can change the magnetization of the ferromagnetic layer in such a way that, for one current direction, a magnetic flux penetrates the junction perpendicular to the layers, whereas for the opposite direction, the perpendicular magnetic flux can be removed. In the former state, the modulation pattern of the Josephson critical current in the magnetic field may acquire minimum near zero field and restores its usual shape with maximum in the second state. This possibility of electric control, and a large discrimination between the Josephson critical current levels for the two states which will be preserved upon reduction of the device dimensions, is important for integration of memory arrays with high parameter margins.

For quantum computing,  $\pi$  junctions have been proposed as circuit elements qubit designs. The characteristic hysteretic behavior of the ferromagnetic barrier provides an alternative and intrinsically digital tuning of the gubit frequency by of magnetic field pulses means [9]. The functionalities and limitations of this device were tested by coupling to a readout resonator. The possibility to use the qubit as a noise detector and its relevance to investigate the subtle interplay of magnetism and superconductivity is envisaged. To make a real impact in superconducting qubit technology, ferromagnetic Josephson junction have to be transferred to aluminum technology - the

dominant material for fabrication of Josephson junctions for all superconducting qubits. The recent results demonstrated that the ferromagnetic layer does not affect the quality of the junction in qubits.

In summary, we believe that cryogenic spintronic elements integrated with superconducting electronics can play an important enabling role in many new functionalities, performance capabilities, and practical working characteristics not attained with other technologies.

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### **Neuromorphic Magnonics**

Katrin Schultheiss<sup>1</sup>, Christopher Heins<sup>1</sup>, Lukas Körber<sup>1,2</sup>, Zeling Xiong<sup>1</sup>, Tobias Hula<sup>1</sup>, Joo-Von Kim<sup>3</sup>, Thibaut Devolder<sup>3</sup>, Sonia Thlang<sup>3</sup>, Johan Mentink<sup>2</sup>, Attila Kákay<sup>1</sup>, J. Fassbender<sup>1</sup>, H. Schultheiss<sup>1</sup>

<sup>1</sup>Helmholtz-Zentrum Dresden-Rossendorf, Bautzner Landstrasse 400, Dresden, Germany <sup>2</sup>Radboud University, Institute of Molecules and Materials, Heyendaalseweg 135, 6525 AJ Nijmegen, The Netherlands <sup>3</sup>Centre de Nanosciences et de Nanotechnologies, CNRS,

Université Paris-Saclay, 91120 Palaiseau, France

k.schultheiss@hzdr.de

Neural networks are powerful tools to learn patterns and make inferences in complex problems. However, they rely on a massive number of neurons and interconnecting weights which require extensive training using a large dataset. To compensate for this, reservoir computing is based on recurrent neural networks with randomly fixed weights. Thereby, only the output weights require training for a particular task, reducing the training to a simple linear regression. Recently, there has been a shift towards physical reservoir computing, offering potential advantages in speed, energy efficiency, simplicity. Physical reservoir and hardware computing utilizes the inherent nonlinearity of physical systems to map the input into a higherdimensional space in which different input patterns become linearly separable. New advancements and experimental implementations use diverse physical substrates, including mechanical structures, optical systems, and spintronic devices.

In our work, we take advantage of the rich nonlinear dynamics inside magnetic vortices. Their eigenmode system comprises the gyrotropic motion of the vortex core as well as magnon modes with welldefined radial and azimuthal quantization in the vortex skirt. A few mode profiles are depicted in Figure 1. By harnessing the nonlinear interactions between these different vortex eigenmodes in reciprocal space, it is possible to perform temporal information processing and pattern recognition without relying on information transport in real space [1]. This presentation will give a comprehensive overview of experimental results and numerical simulations demonstrating the capabilities and advantages of magnon reservoir computing inside a magnetic vortex.

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### Figures



**Figure 1.** Working principle of a magnon-scattering reservoir. Radiofrequency pulses with different temporal order but the same average frequency content are used to trigger nonlinear scattering between the magnon eigenmodes in a magnetic vortex disk. The dynamic response is experimentally detected using Brillouin-light-scattering microscopy. In contrast to a linear system, the magnon-scattering reservoir produces different outputs depending on the temporal order of the input.

# Femtosecond dynamics of quantum materials

#### Sangeeta Sharma<sup>1</sup>, J. K. Dewhurst<sup>2</sup>

<sup>1</sup>Institute for theoretical solid state physics, Freie Universität Berlin, Arnimallee 14, Germany, and *Max Born Institute for Nonlinear Optics and Short Pulse Spectroscopy, Max-Born-Str. 2A, 12489 Berlin, Germany* <sup>2</sup>Max Planck instute for microsctructure physics, Halle, Germany

sharma@mbi-halle.de

From the outset of research into femtomagnetism, the field in which spins are manipulated by light on femtosecond or faster time scales, several questions have arisen and remain highly debated: How does the light interact with spin moments? How is the angular momentum conserved between the nuclei, spin, and angular momentum during this interaction? What causes the ultrafast optical switching of magnetic structures? What is the ultimate time limit on the speed of spin manipulation? What is the impact of nuclear dynamics on the light-spin interaction?

In my talk I will advocate a parameter free *ab-initio* approach to treating ultrafast light-matter interactions, and discuss how this approach has led both to new answers to these old questions but also to the uncovering of novel and hitherto unsuspected spin dynamics phenomena [1,2]. In particular I will highlight following aspects of ultrafast dynamics:

(a) Femto- phono- magnetism: an extra degree of control over spin dynamics can be obtained by selective excitation of phonon modes [3].

(b) Pulse design: control of spin- valley- tronics via pulse shaping [4,5].

(c) Spin vacuum switching: full reversible switching using spin-currents[6].

(e) Exciton dynamics: strong correlations between excitons and free carriers.

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# Altermagnetism: spin symmetry prediction and experimental evidence

#### Libor Šmejkal<sup>1,2,3</sup>

<sup>1</sup>Max Planck Institute for the Physics of Complex Systems, Nöthinzer Str.38, 01187 Dresden, Germany <sup>2</sup>Institut für Physik, Johannes Gutenberg Universität Mainz, D-55099 Mainz, Germany <sup>3</sup>Institute of Physics, Czech Academy of Sciences, Cukrovarnická 10, 162 00, Praha 6, Czech Republic

#### lsmejkal@pks.mpg.de

Magnetic materials are traditionally classified as either ferromagnets or antiferromagnets. Here, we will show that a systematic spin symmetry classification of collinear magnets introduces a third class-altermagnetism[1]. Altermagnets feature even-parity-d-, alternating g-, or i-wave magnetization in real space and corresponding unconventional spin polarization in momentum space that breaks time-reversal symmetry without generating net magnetization [1-6]. This unconventional time-reversal symmetry breaking allows altermagnets to generate strong anomalous Hall and spin currents of unconventional symmetry, opening up exciting possibilities for spintronics applications. We will also present experimental evidence of altermagnetic time-reversal symmetry breaking in MnTe through: (i) momentum-space photoemission spectra showing altermagnetic band splitting [7-8], and (ii) real-space XMCD and XMLD mapping of altermagnetic domains [9]. Finally, we will discuss the extension of this classification to noncollinear systems, unveiling an unconventional odd-parity-wave magnetic class [10]."

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Figure 1. Visualisation of anomalous Hall current in an altermagnet.

# Precessional all optical switching and deterministic switching outlook

Ricardo C. Sousa<sup>1</sup>, D. Salomoni<sup>1</sup>, Y. Peng<sup>2</sup>, L. Farcis<sup>1</sup>, S. Auffret<sup>1</sup>, M. Hehn<sup>2</sup>, G. Malinowski<sup>2</sup>, S. Mangin<sup>2</sup>, B. Dieny<sup>1</sup>, L.D. Buda-Prejbeanu<sup>1</sup>, I.L. Prejbeanu<sup>1</sup> <sup>1</sup>Spintec (UGA/CNRS/CEA), 17 rue des Martyrs, 38054 Grenoble, France <sup>2</sup>Institut Jean Lamour, UMR CNRS 7198, Université de Lorraine, 54011 Nancy, France

ricardo.sousa@cea.fr

In the pursuit of faster, more efficient memory, alloptical switching (AOS) technology seeks to achieve magnetization reversal through energy-efficient ultrafast writing with single-femtosecond laser pulses. This work addresses the objective of merging helicity-independent HI-AOS and MRAM devices. The materials investigated are rare-earth transition metal multilayer systems, with a particular focus on Tb/Co-based structures. Through a combination of experimental investigations and numerical simulations, we unveiled the intricate dynamics and precessional character of AOS in these multilayer systems, using them to develop pulse switchable nanometer-sized AOS laser magnetic tunnel junctions (AOS-MTJs). These results provide an understanding of the AOS reversal process and explore its applications in nonvolatile data storage and energy-efficient computation at the nanoscale.

Exploring HI-AOS in thin films of rare-earth transition-metal multilayer systems allows for reliable magnetization toggle reversal, relying on the response to different pulse duration and fluence. Key new findings include the observation of concentric rings with opposite magnetic orientations and the independence of fluence on pulse duration, challenging previous understanding. A hypothesis emphasizing the role of local anisotropy decrease favoring a precessional type of helicity-independent AOS is introduced. The development of a macrospin model, incorporating principles of a two-temperature explains the precession-driven model (2TM), switching mechanism observed in the Tb/Co-based multilaver system. In this framework our investigations. allowing simulating the rapid response of our system to ultrafast laser pulses and gain valuable insights into the underlying physics. One of the central findings of the study is the remarkable independence of HI-AOS from the pulse duration, a characteristic that was consistently observed experimentally and can be attributed to the unique dynamics of the system. This independence implies that the core mechanisms driving the reversal process occurs over longer timescales than the laser pulse duration, primarily influenced by the absorbed laser fluence and the subsequent cooling of the system to its thermal equilibrium. The model considered various parameters such as temperature-dependent uniaxial anisotropy and the tilted anisotropy shedding light on the axis,

intricacies of the reversal mechanisms within the system. Our simulations have demonstrated how parameters like damping, anisotropy angles, and the Q factor at equilibrium influence the threshold fluences necessary for HI-AOS. While the 2TM provided the foundational knowledge, it is clear that the full complexity of HI-AOS requires further investigation.

Similar to our initial research [1-3], the AOS behavior appears exclusively in the cobalt-rich region. It is important to note that the regions 'rich' in terbium and cobalt are temperature dependent. When the cobalt-rich region, is usually referred in relation to room-temperature conditions (300 K). Put differently, our system allows for ultra-fast reversal only when the initial temperature (usually RT) is slightly above the compensation temperature ( $T_{Comp}$ ). In such cases, the total magnetization increases as the temperature increases.

The implementation of a stable hard reference layer and optimization of the MgO natural oxidation has allowed for tunnel magnetoresistance (TMR) values, reaching a maximum value of 74%, representing a significant improvement over previous realizations [4]. The main achievement was the demonstration of field-free HI-AOS on 100 nm diameter patterned [Tb/Co]x5 p-MTJ devices, using 50 fs laser pulses with an estimated absorbed energy of approximately 68.6 fJ per bit reversal. These findings hopefully pave the way toward nanoscale devices for optospintronic embedded memories combining nonvolatility, ultrafast and energy-efficient writing. We acknowledge financial support from the Agence Nationale de la Recherche (ANR) [Grant No. ANR-17-CE24-0007 UltraFast Opto-magneto-spintronics for Future Nanotechnologies (UFO) project] and the European Union (EU) Horizon 2020 research and innovation program under Marie Skłodowska-Curie Grant Agreement No. 861300 ["Cold Opto-Magnetism Devices" for Random Access (COMRAD)].

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**Figure 1.** Multilayer thickness of [Tb/Co]<sub>5</sub> showing helicity independent switching in cobalt rich highlighted green region under 3.8mJ/cm<sup>2</sup> 50fs laser pulse.



**Figure 2.** Illustration of the all-optical writing and electrical reading of an AOS MTJ stack showing SEM image of a 100-nm diameter pillar. Out of plane field hysteresis loop and resistance measurement after application of laser-pulse demonstrating toggle switching.

# Spin and Orbital Magnetism by Light in Rutile Altermagnets

T. Adamantopoulos<sup>1,2,3</sup>, M. Merte<sup>1,2,3</sup>, F. Freimuth<sup>3</sup>, D. Go<sup>3</sup>, L. Zhang<sup>1</sup>, M. Ležaić<sup>1</sup>, W. Feng<sup>4,5</sup>, Y. Yao<sup>4,5</sup>, J. Sinova<sup>3,6</sup>, L. Šmejkal<sup>3,6,7</sup>, S. Blügel<sup>1</sup>, and Y. Mokrousov<sup>1,3</sup> <sup>1</sup>Peter Grünberg Institut, Forschungszentrum Jülich, 52425 Jülich. Germanv <sup>2</sup>Department of Physics, RWTH Aachen University, 52056 Aachen, Germany 3Institute of Physics, Johannes Gutenberg University Mainz, 55099 Mainz, Germany <sup>4</sup>Centre for Quantum Physics, Key Laboratory of Advanced Optoelectronic Quantum Architecture and Measurement (MOE), School of Physics, Beijing Institute of Technology, Beijing 100081, China <sup>5</sup>Beijing Key Lab of Nanophotonics and Ultrafine Optoelectronic Systems, School of Physics, Beijing Institute of Technology, Beijing 100081, China <sup>6</sup>Institute of Physics, Czech Academy of Sciences, Cukrovarnická 10, 162 00 Praha 6, Czech Republic <sup>7</sup>Max Planck Institute for the Physics of Complex Systems, Nöthnitzer Str. 38, 01187 Dresden, Germany

t.adamantopoulos @fz-juelich.de

While the understanding of altermagnetism is still at a very early stage, it is expected to play a role in various fields of condensed matter research, for example spintronics, caloritronics and superconductivity [1]. In the field of optical magnetism, it is still unclear whether altermagnets can exhibit magnetisation dynamics effects distinct from ferromagnets and antiferromagnets. Here we choose RuO2, a prototype metallic altermagnet with a giant spin splitting, and CoF2, an experimentally known insulating altermagnet, to study the inverse Faraday effect (IFE) in altermagnets from firstprinciples [2]. We predict large and canted induced spin and orbital moments after the optical excitation which are distinct on each magnetic sublattice. By resorting to microscopic tools, we interpret our results in terms of the altermagnetic spin splittings and of their reciprocal space distribution. Overall, in accordance with our symmetry analysis, we demonstrate that the behavior of altermagnets when exposed to optical pulses incorporates both ferromagnetic and antiferromagnetic features.

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# Terahertz-driven magnetoelectric torque in the collinear antiferromagnet Cr<sub>2</sub>O<sub>3</sub>

Vladislav Bilyk<sup>1</sup>, Roman Dubrovin<sup>2</sup>, MengXing Na<sup>1</sup>, Anatoliy Zvezdin<sup>3</sup>, Alexey Kimel<sup>1</sup> <sup>1</sup>Radboud University, Nijmegen, The Netherlands <sup>2</sup>loffe Institute, St. Petersburg, Russia <sup>3</sup>A.M.Prokhorov General Physics Institute, Moscow, Russia

vladislav.bilyk@science.ru.nl

Magnetoelectric effect, facilitating a control of spins in magnets with the help of electric field, has long been seen as a cornerstone for future energy efficient and nano-scalable technologies for magnetic writing and processing of magnetically stored information. In contrast to spin-polarized currents, a control of spins with the help of electric fields promises much lower dissipations and in contrast to magnetic fields, electric fields are easier to apply to a nanoscale bit. Understanding temporal evolution of the magneto-electric effect, revealing how fast spins respond to an electric field is thus crucial for revealing the fundamental limit on the operational frequency of magnetoelectric devices. Here we report on ultrafast magnetoelectric effect in antiferromagnetic Cr<sub>2</sub>O<sub>3</sub>.

Cr<sub>2</sub>O<sub>3</sub> prototypical is Chromium oxide а magnetoelectric antiferromagnet with the corundum trigonal crystal structure (s.g. R3c) and the Néel temperature T<sub>N</sub>=307.6 K. The studied sample in the form of a thin plane plate cut such that the antiferromagnetically ordered spins of Cr<sup>3+</sup> ions lie sample plane (1010) in the parallel to crystallographic c-axis of the crystal. The sample was excited with the THz pulses electrical field of which were able to rotate in the plane of sample surface with the set of two wire grid polarizers as described in ref. [1]. THz-induced dynamics excited were detected with a balance photodetector as a probe polarization rotation. Microscopical image (fig. 1a) of the domain structure was visualized with circularly polarized probe pulses [2].



**Figure 1.** a) SHG image of the domain pattern gained with the circularly polarized probe with the energy of fundamental pulses 2.1eV. b) The amplitude of the oscillations excited at different angles of the THz electric

field observed in a single domain. The angle values stand for orientation of electric field  $\mathbf{E}_{THz}$  of the THz pulse .

The THz-induced transient signals (fig. 2) for the probe polarization rotation measured in  $Cr_2O_3$  at temperature 77K for various polarizations of the THz pump pulse with respect to the antiferromagnetic Néel vector **L** show oscillations at the frequency of the antiferromagnetic resonance at 0.165 ps [3,4] and is related to the THz pump driven spin dynamics.

When the magnetic field is orthogonal to the antiferromagnetic Néel vector  $H_{THz} \perp L$  [5,6] Zeeman torque excitation mechanism is expected to reach the maximum values. In contrast, when the magnetic field of the THz pump pulse is parallel to the antiferromagnetic Néel vector  $H_{THz} \parallel L$ , and consequently  $E_{THz} \perp L$ , the efficiency of this excitation mechanism must be equal to zero. It is seen in Fig. 1b that the spin dynamics in Cr<sub>2</sub>O<sub>3</sub> is excited with comparable efficiency both by the Zeeman torque at  $H_{THz} \perp L$ , and at the orthogonal to it polarization  $H_{THz} \parallel L$  ( $E_{THz} \perp L$ ).

The amplitude of the oscillations depends on the THz pump polarization angle (fig. 1b) and have a maximum in between  $H_{THz} \perp L$  and  $E_{THz} \perp L$  which indicates interference between two torques that can excite spin dynamics. The amplitude of oscillations is a linear function of the THz magnetic and electric field strengths when the magnetic field  $H_{THz}$  is parallel and perpendicular to the antiferromagnetic Néel vector **L**.



**Figure 2.** THz-induced spin dynamics measured for domains with oppositely oriented Néel vectors  $L_+$  and  $L_-$  for Zeeman and magnetoelectric geometries. different angles between the THz electric field and antiferromagnetic L vector at 77K.

In order to distinguish between these two excitation mechanisms we performed an experiment (fig. 2) where probe rotation dynamics were measured in two domains with oppositely oriented L vector and the THz electric field  $E_{THz} \parallel L (H_{THz} \perp L)$  was oriented either parallel or perpendicular  $E_{THz} \perp L$  to Néel vector which can be ascribed to Zeeman and magnetoelectric torque. When the THz magnetic

field and the antiferromagnetic Néel vector are mutually perpendicular  $H_{THz} \perp L$  (Zeeman torque), a reversal of the L vector upon transition to opposite domain does not affect the detected spin dynamics. In case of the THz magnetic field and the antiferromagnetic Néel vector are mutually perpendicular  $H_{THz} \parallel L$  which is equivalent to  $E_{THz} \perp L$  (magnetoelectric torque), a reversal of L is changes the phase of the oscillations by 180°.

Such behavior can be explained taking into account thermodynamical potential  $\Phi$  for magnetoelectric antiferromagnet having term a term  $M_x E_x L_y$ . The effective magnetic field that triggers the dynamics of Néel vector would be  $H_{eff} \sim E_x L_y$  and thus provides the phase change of the oscillations on reversal of both either electric field or Néel vector in the observable magnetization dynamics for the magnetoelectric mechanism of excitation. At the same time excitation by the Zeeman torque dL/dt ~ LxH does not depend on the orientation of L and changes the phase of the oscillations upon the reversal of magnetic field.

We have demonstrated that antiferromagnetic resonance in  $Cr_2O_3$  can be driven using nearly single cycle THz electromagnetic pulse. It is shown that the electric component of the THz pulse plays in the excitation significant role that exert a magnetoelectric along the Zeeman torque. Hence, our experiments also reveal a fundamentally new mechanism for ultrafast control of spins in antiferromagnets.

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#### Diversity of Ultrafast Spin Dynamics Near the Tricritical Point in a Ferrimagnetic Gd/FeCo Multilayer

T.G.H. Blank<sup>1,2</sup>, B.D. Muis<sup>3</sup>, T. Lichtenberg<sup>2</sup>, B. Koopmans<sup>2</sup>, A.V. Kimel<sup>1</sup>
 <sup>1</sup>Radboud University, Institute for Molecules and Materials, 6525 AJ Nijmegen, the Netherlands.
 <sup>2</sup>Department of Applied Physics, Eindhoven University of Technology, P.O. Box 513, Eindhoven 5600 MB, the Netherlands.
 <sup>3</sup>Kavli Institute of Nanoscience, Delft University of Technology, 2600 GA Delft, the Netherlands.

#### thomas.blank@ru.nl

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 relatively strong exchange interaction between the sublattices, the critical field is strong and the studies required exceptionally high magnetic fields 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 1) 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], where the phase transition changes from being discontinuous to continuous. We highlight exchangedriven reversal as a particularly interesting type of

dynamics, providing new insights into the tricritical point. The latter 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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**Figure 1.** The six distinct types of laser-induced magnetization dynamics in Gd/FeCo measured by the time-resolved magneto-optical Faraday rotation using 100 fs probe pulses at a central wavelength of 800 nm, recorded at different temperatures and external magnetic fields applied along the out-of-plane easy axis of anisotropy. The observed Faraday effect is known to be proportional to the out-of-plane magnetization of the FeCo sublattice. The Faraday effect was normalized such that a full 180-degree reversal of the FeCo magnetization corresponds to a signal value of 2. The arrows provide a pictorial description of the FeCo magnetization based on our interpretation of the data.

# Slow (relativistic) and fast (exchange) switching in Gd/FeCo multilayers

Aleksandr Buzdakov<sup>1</sup>, Thomas Blank<sup>2</sup>, Konstantin Zvezdin<sup>3</sup>, Alexey Kimel<sup>2</sup> <sup>1</sup>Istituto Italiano di Tecnologia, Italy <sup>2</sup>Radboud University, Netherlads <sup>3</sup>Istituto PM, Italy

alex.buzdakov@gmail.com

Ferrimagnets are highly appealing in ultrafast magnetism. The discovery of all-optical magnetic switching in GdFeCo alloys nearly two decades ago spurred interest in this field, revealing phenomena like toggle switching and current-induced switching rare-earth transition metal alloys. in Rareearth/transition-metal multilayers further enhance this field by allowing control over interlayer exchange interaction, magnetization, and magnetic anisotropy. By tuning multilayer thicknesses, one can manipulate compensation and Curie temperatures, leading to diverse laser-induced spin dynamics. This work draws inspiration from recent findings on tunable laser-induced spin dynamics in Gd/FeCo multilayers [1]. We theoretically explored laser-induced magnetization dynamics in Gd/FeCo multilayers, focusing on the effects of applied magnetic field and temperature. Unlike GdFeCo alloys, these multilayers exhibit unique magnetic parameters, which we defined through an H-T phase diagram model fitted to experimental data. Using these parameters and modified Landau-Lifshitz-Bloch equations, we simulated transverse and longitudinal magnetization dynamics, successfully reproducing a wide range of experimental observations. This work advances our understanding of ultrafast magnetization dynamics in multisublattice materials with canted spins.

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 Figures



**Figure 1.** Schematic picture of studied Gd/FeCo multilayered structure.

### Ultrafast Imaging of Electrically-Controlled Laser-Induced Spin Dynamics

# **T.T. Gareev**<sup>1</sup>, N.E. Khokhlov<sup>1</sup>, A.P. Pyatakov<sup>2</sup>, A.V. Kimel<sup>1</sup>

<sup>1</sup>Radboud University, 6525AJ, Nijmegen, the Netherlands <sup>2</sup>Lomonosov Moscow State University, 119991, Moscow, Russia

#### timur.gareev@ru.nl

Mechanisms allowing to control of magnetization using electric rather than magnetic fields have long been in a focus of fundamental research in magnetism and considered as an essential step toward the development of future energy-efficient spintronic nanodevices [1,2]. Aiming to increase the operational frequency of the devices, it is natural to focus on electric control of ultrafast magnetization dynamics, in particular.

Here we report on an experimental study of how an applied electric field can control ultrafast laserinduced spin dynamics in the epitaxial film of ferrimagnetic iron garnet known to show strong magneto-electric effects [3]. The studied (BiLu)<sub>3</sub>(FeGa)<sub>5</sub>O<sub>12</sub> film was grown on a (110) oriented Gd<sub>3</sub>Ga<sub>5</sub>O<sub>12</sub> substrate. Applying a nearly in-plane magnetic field and exciting the sample with a femtosecond laser pulse, we launched 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 0.5 MV/m (Fig. 1). Surprisingly, the effect of the electric field on the amplitude of the dynamics seems to be as large as in the case of 2D materials, which are known to be strongly susceptible to electric fields [4].

We have shown that the electric field plays the role of the effective external magnetic field due to the linear magnetoelectric effect, and thus gives control over the amplitude and frequency of oscillations. Moreover, the electric field was found to affect the spatial distribution of magnetization dynamics (Fig. 1a, b). In particular, we discovered that without any applied electric field, laser pulse induces a nonhomogeneous pattern of spin dynamics in a form of rings. We believe the formation of such rings is a very general phenomenon originating from the interplay of several contributions to magnetic anisotropy. The electric field, being responsible for an additional effective magnetic field, can thus affect this interplay and result in changes of the inhomogeneous pattern.

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**Figure 1.** (a, b) Time-resolved magneto-optical images of electric field-controlled laser-induced spin dynamics. The images were captured at different moments of time without (a) and with the applied electric field (b). (c) Magnetization dynamics traces extracted from the time-resolved magneto-optical images without (black curve) and with an applied electric field (orange curve). A magnetic field of Hext = 30 mT was applied in all experiments. The laser fluence was 36 mJ/cm<sup>2</sup>.

### Micromagnetics Of All-Optical Switching Dynamics In A Tb/Co Bilayer

Paul-Iulian Gavriloaea<sup>1</sup>, David Salomoni<sup>2</sup>, Elías Saugar<sup>1</sup>, Ioan Lucian Prejbeanu<sup>2</sup>, Liliana D. Buda-Prejbeanu<sup>2</sup>, Oksana Chubykalo-Fesenko<sup>1</sup>

<sup>1</sup>Instituto de Ciencia de Materiales de Madrid, CSIC, Cantoblanco, 28049 Madrid, Spain <sup>2</sup>University Grenoble Alpes, CEA, CNRS, Grenoble INP, SPINTEC, 38000 Grenoble, France

paul.gavriloaea@csic.es

Ultra-fast heating alone drives helicity independent, all-optical switching (HI-AOS), predominantly in Gdbased ferrimagnetic alloys and multi-layers [1-3]. Governed by the inter-sublattice exchange coupling, this mechanism requires a careful adjustment of the laser fluence in relation to the pulse length, thus undermining its technological potential. A different HI-AOS mechanism was recently shown to govern laser-induced magnetisation switching in Tb and Dybased ferrimagnetic multilayers, bilayers or trilayer systems [4]. Several of its characteristics remain theoretically unexplained: (a) the energy threshold to switching is independent of the pulse duration up to several ps; (b) the excitation area presents a concentric ring domain pattern; (c) the magnetisation exhibits a slow recovery process [4,5]. The switching is believed to occur due to an in-plane reorientation of the magnetisation under a temperature induced reduction of the perpendicular anisotropy at the interface and the presence of a local distribution of easy-axes directions arising from the granular thin film texture which could favour the slow, in-plane precession dynamics [4,5]. Here we provide a high temperature micromagnetic model which captures in full detail this behavior.

We examine this problem in the framework of the Landau-Lifshitz-Bloch (LLB) micromagnetic model [6] considering a Tb(1nm)/Co(1nm) bilayer as reference system. The key ingredient enabling the AOS is the assumption of a distribution of easy-axes (EAs) directions perpendicular to the thin film. We assume that it follows a Gaussian distribution within a cone with an average value set to  $0^{\rm 0}$  and a standard deviation of  $2.5^{\rm 0}$  for the polar angle  $\theta_{\text{EA}}$  – see Fig. 1(a). The laser heating is described via a two-temperature model which includes a laser power source with a Gaussian profile in time as well as in space, inside the Oxy plane. The EAs distribution has the advantage of preserving an out-of-plane perpendicular magnetisation at room temperature whilst providing a torque on magnetisation favouring the precession during the ultra-fast laser excitation. Our model indicates that a 50-fs pulse induces a rapid heating of the sample along with an initial fast switching on a timescale of several ps, followed by a subsequent slow recovery in the ps-ns time domain as seen in subplot (c) of Fig. 1. During the laser

pulse heating, the maximum electron temperatures corresponding to the squared regions in (b) satisfy the relationship: T<sub>1</sub>>T<sub>2</sub> due to the Gaussian spatial profile. This leads to a slower reversal in region 1 due to a competition between transverse and longitudinal relaxation mechanisms where a strong demagnetization followed by a slow precession can be observed, in comparison to the faster dynamics in region 2. Our model recovers the characteristic, ring domain-like pattern - shown for the Co mz component in subplot (b) 1.5 ns after exposure to the laser pulse. We explore this switching mechanism closely following the results presented in Refs. [4,5]. Thus, we investigate the role of the  $\theta_{EA}$ maximum tilting, the exchange coupling between the Co and Tb layers as well as the cooling rate in fitting the experimental switching curves. Although the results of Fig. 1 are obtained in zero applied field, we separately assess the role of in-plane and out-ofplane Zeeman contributions in the switching process. Our results contribute towards the fundamental understanding of recent advancements in the ultrafast magnetism community which so far lacked a supporting, theoretical picture.

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**Figure 1.** (a) EA anisotropy distribution: average  $\theta_{EA}$  is zero with standard deviation of 2.5<sup>o</sup> and  $\varphi_{EA}$  varies uniformly in the range [0<sup>o</sup>: 360<sup>o</sup>]. (b) Concentric ring domain pattern in the Co sublattice along Oz, 1.5 ns after exposure to a 50-fs laser-pulse. (c) Average <m<sub>z</sub>>dynamics extracted for Co and Tb from the two squared spatial regions in (b).

### Coherent control of THz-scale spin resonances using optical spin—orbit torques

Julian Hintermayr<sup>1</sup>, Paul M. P. van Kuppevelt<sup>1</sup>, Bert Koopmans<sup>1</sup> <sup>1</sup>Eindhoven University of Technology, Department of Applied Physics, P.O. Box 513, 5600 MB Eindhoven, Netherlands

j.hintermayr@tue.nl

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 spintransfer 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 (see Fig. 1 Employing a). time-resolved magneto-optical demonstrate studies, we 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, as shown in Fig. 1 b. Extending this concept, we find that ferrimagnetic exchange resonances in Co/Gd-based systemsoffering 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 exchangedriven modes, we identify features that challenge the current understanding of optically generated spinorbit torques, and we discuss possible explanations. These insights hold great promise for the advancement of ultrafast spintronic computation devices.

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#### Figures



**Figure 1. a** Schematic representation of the double pump excitation scheme and spin excitation in Pt to drive FMR dynamics in the FM layer. **b** TR-MOKE measurements of OSOT-driven FMR dynamics and demonstration of coherent control.

# Dual-pulse laser excitation and MFM probing of a Pt/Co/Pt trilayer

**D. Khusyainov**<sup>1</sup>, R. Liefferink<sup>1</sup>, MX. Na<sup>1</sup>, F. Kammerbauer<sup>2</sup>, R. Frömter<sup>2</sup>, M. Kläui<sup>2</sup>, J.H. Mentink<sup>1</sup>, D. Afanasiev<sup>1</sup>, A.V. Kimel<sup>1</sup>, Th. Rasing<sup>1</sup>

<sup>1</sup> Radboud University, Institute for Molecules and Materials, 6525 AJ Nijmegen, The Netherlands

<sup>2</sup> Institute of Physics, Johannes Gutenberg University of Mainz, 55099 Mainz, Germany

Dinar.khusyainov@ru.nl

#### Abstract

Recent studies shown have that manipulating out-of-equilibrium matter with light is more efficient than controlling equilibrium states. For example, dual-optical pumping, with a preparational pump pulse getting matter out of equilibrium followed by a switching pump pulse, is more energyefficient for optical switching between insulator-tostates [1]. Similarly, manipulating nonmetal equilibrium spin systems leads to new energyefficient ways to control magnetic states. A recent study demonstrated that a preparational laser pulse could switch an antiferromagnet to a transient nonequilibrium ferromagnetic state, which could be controlled with a second excitation pulse [2]. The dual-pulse optical technique has also been shown to lead to very efficient switching of ferromagnetic Pt/Co/Pt trilayers. [3]. However, the mechanism behind the switching of this ferromagnetic material is still under discussion.

Here, we use a scanning probe magnetic force microscope (MFM) with dual-pulse laser excitation (see Fig. 1a) to study a Pt/Co/Pt trilayer. The laser beam is adjusted to the scanning area of the microscope to conduct in situ measurements following nucleation. By scanning the excitation area for various delays between two pump laser pulses, we can discern magnetic nanopatterns and monitor the switching area of nucleated magnetic domains as function of delay time. When the delay between the two pumps is zero, the nucleated magnetic pattern exhibits the highest switched area (Fig. 1b, lower panel). Introducing a delay between the pulses results in a lower switched area value for the nucleated pattern. The switched area as a function of the delay between the two pulses is plotted in Fig. 1b. The switched area is approximately 120  $\mu$ m<sup>2</sup> at  $\Delta t=\pm 5ps$  and approximately 250  $\mu m^2$  in the overlap. We fitted the relaxation of the switched area with an exponential fit with a time constant of 1.4±0.8 ps. According to the 3-temperature model, this dynamics corresponds to the time of the relaxation of the spin system in the Pt/Co/Pt trilayer. [4].

To conclude, we demonstrate that combining MFM with dual-pulse excitation, offers a new approach to study All-optical switching in ferromagnetic Pt/Co/Pt with high temporal and spatial resolution.

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Figure 1. Dual-pulse excitation and MFM probing of Pt/Co/Pt. a) Schematics of the experimental setup of dualpulse excitation and MFM probing. b) Switched area as a function of pump-pump delay. The lower panel shows MFM images of the nucleated patterns after dual-pulse excitation. The scale bar is  $20 \ \mu m$ .

### Switching the In-Plane Magnetization in Gd-Based Spin Valves Using Optically Excited Spin Currents

J.-X. LIN<sup>1</sup>, Y. LE GUEN<sup>1</sup>, J. HOHLFELD<sup>1</sup>, J. IGARASHI<sup>1</sup>, Q. REMY<sup>2</sup>, T. HAUET<sup>1</sup>, J. GORCHON<sup>1</sup>, G. MALINOWSKI<sup>1</sup>, S. MAMGIN<sup>1</sup>, and M. HEHN<sup>1</sup> <sup>1</sup>Université de Lorraine, CNRS, Institut Jean Lamour, F-54000 Nancy, France <sup>2</sup>Department of Physics, Freie Universität Berlin, 14195 Berlin, Germany

jun-xiao.lin@univ-lorraine.fr

The discovery of magnetization manipulation using laser pulse-induced spin currents generated by laser-induced demagnetization [1] has attracted significant attention for its potential application in magnetic memories, as it future enables magnetization reversal within a few hundred femtoseconds [2]. Previous studies have primarily focused on collinear spin valve structures composed of GdFeCo/Cu/ferromagnetic (FM) layer, where the Gd sublattice in GdFeCo predominantly generates the spin currents, and the aim is to switch the ferromagnet's magnetization via the spin current pulse [3,4]. Most research has concentrated on reversal magnetization perpendicularly in magnetized spin valves [5]; however, this configuration does not allow for the disentanglement of the impact of layer thickness on the reversal process since these ultrathin layers exhibit a Curie temperature (T<sub>c</sub>) that varies significantly with film thickness.

In this talk, I will present findings from experiments using a similar structure with in-plane magnetic anisotropy, which allows us to isolate the effects of Tc and FM layer thickness on magnetization switching of the FM layer. The results show that (1) a higher T<sub>c</sub> in the FM layer requires a larger laser threshold for complete fluence reversal. Furthermore, (2) for samples with a fixed T<sub>c</sub>, an increase in FM layer thickness also demands a higher laser fluence threshold. Beyond a certain thickness, multiple pulses are necessary to switch the entire FM layer. I will also discuss (3) the maximum thickness at which full switching is achieved, depending on the Gd concentration in the spin current source layer, GdCo. In summary, this switching behavior can be explained by the need for substantial demagnetization of the FM layer upon the arrival of the current pulse. These findings improve our understanding of magnetization reversal induced by ultrafast spin current pulses and contribute to the design of energy-efficient magnetic memory devices [6].

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**Figure 1.** Schematic illustrating the parameters studied in this in-plane magnetized ferromagnet (FM)/Cu/GdCo spin valve structure, including the FM Curie temperature (T<sub>C</sub>), FM layer thickness (t), and Gd concentration (x) in the current source GdCo alloy. This study aims to investigate the factors influencing the switching of the FM layer. It is concluded that the criterion for observing switching is that the angular momentum loss (J<sub>s</sub>, referred to as optically excited spin currents) from the Gd sublattice must exceed the remaining magnetization ( $\Delta$ M) in the FM layer, both of which are dependent on the laser fluence (F).

#### Disentangling spin and charge contributions to the THz emission from ultrathin antiferromagnetic heterostructures

Thomas Metzger<sup>1</sup>, Takkashi Kikkawa<sup>2</sup>, Eiji Saitoh<sup>2</sup>, Alexey Kimel<sup>1</sup>, Davide Bossini<sup>3</sup> <sup>1</sup>Institute of Molecules and Materials, Radboud University, Nijmegen, Netherlands <sup>2</sup>Department of Applied Physics, The University of Tokyo, Tokyo, Japan <sup>3</sup>Department of Physics, Konstanz University, Konstanz, Germany

thomas.metzger@ru.nl

Antiferromagnetic (AF) spintronics is highly promising for fundamental research and future applications. Particularly, the potential of antiferromagnets lies in their robustness against perturbing external electromagnetic fields and their dynamics, intrinsically faster than in ferromagnets, as the magnetic eigenfrequencies enter the terahertz (THz) range [1-2]. The ability to convert the spin dynamics into a charge signal is key to spintronics inspiring recent studies on heavy metal / AF heterostructures [3].

In particular, thin film Pt/NiO heterostructures have become a model system in antiferromagnetic spintronics. Laser-induced THz emission as a result of spin-current injection from NiO [4], optically triggered torque on NiO spins as a consequence of femtosecond laser heating of Pt [5] and magnetooptical pump-probe experiments investigating the ultrafast demagnetization of antiferromagnetic NiO sublattices [6] were reported. This rich variety of the reported and sometimes discrepant effects in the literature sparks controversy and arises from the complexity of these heterostructures rendering the identification of the microscopic origin of the laserinduced THz emission to be challenging.

Here, we suggest that disentangling authentic magnetic from non-magnetic contributions to the THz emission, requires measurements as a function of an external magnetic field intense enough to modify the spin configuration in the ground state. Unprecedented measurements as a function of an external magnetic field up to µHext =7 T allow us to reveal two distinct mechanisms of THz emission from Pt/NiO heterostructures. The first contribution to the emitted THz radiation does not depend on the applied magnetic field, which excludes laser-induced spin dynamics while suggesting optical difference frequency generation. The second contribution is characterized by a linear dependence on the externally applied magnetic field disclosing a magnetic origin. In particular, we interpret this contribution in terms of ultrafast laser-induced quenching of the magnetization, originating from a magnetic field-induced canting of the spin-sublattices in NiO. Our work constitutes an unambiguous experimental approach to access the debated nature

of THz emission observed from heavy metal antiferromagnetic heterostructures.

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# Controlling the type II multiferroic Nil<sub>2</sub> via orbital excitations

M. Na<sup>1</sup>, V. Radovskaia, J. Groefsema<sup>1</sup>, N. Khoklov<sup>1</sup>,
D. Khusyainov<sup>1</sup>, F. Pabst<sup>2</sup>, A. Isaeva<sup>2</sup>,
S. Acharya<sup>3</sup>, A. V. Kimel1, T. H. Rasing<sup>1</sup>, and D.
Afanasiev<sup>1</sup>
<sup>1</sup>Radboud University, Institute for Molecules and Materials, Nijmegen, The Netherlands
<sup>2</sup>University of Amsterdam, Van der Waals-Zeeman

Institute, Amsterdam, The Netherlands

<sup>3</sup>National Renewable Energy Laboratory, Golden, Colorado 80401, USA

mengxing.na@ru.nl

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

Recently, the type II multiferroic Nil<sub>2</sub> has gained significant interest for retaining its multiferroicity down to the monolayer limit [3]. Nil<sub>2</sub> is a quasi twodimensional van der Waals material in which the Ni<sup>2+</sup> ions sit on a trigonal lattice inside an octahedra of I2ions. Through interlayer exchange, Nil<sub>2</sub> becomes an antiferromagnet (AFM) at  $T_N = 80$  K, here, spins ferromagnetically coupled within each layer, and anti-aligned between layers (Fig. 1a) [4]. In addition to interlayer exchange, a significant amount of geometric frustration arises from trigonal lattice of Ni<sup>2+</sup> ions [4]. This frustration outcompetes interlayer exchange below 60 K, leading to the emergence of an incommensurate helical spin-spiral (Fig. 1b) approximately 7 unit cells (~30 nm) in periodicity. The spin-spiral is stabilized by the inverse Dzyaloshinskii-Moriya interaction (DMI), which - in combination with spin-lattice coupling - leads to an asymmetric shift of the I<sup>2-</sup> ion positions [5]. This shift breaks inversion symmetry, resulting in ferroelectric polarization P and giving a rise to a multiferroic phase which can be probed optically using second harmonic generation.

To control the multiferroic phase in Nil<sub>2</sub>, we tune femtosecond laser pulses to target *d*-*d* orbital excitations of the Ni<sup>2+</sup> ions. These electronic excitations involve change in the orbital state of the magnetic Ni<sup>2+</sup> ions and can affect the inverse DMI through spin-orbit coupling. Behaving similarly to conventional excitons, they can be either localized (e.g. Frenkel-like) or delocalized (Wannier-Mott-like), extending across multiple lattice sites. The localized *d*-*d* excitations have been previously shown to efficiently generate THz coherent spin dynamics in collinear Ni<sup>2+</sup> based antiferromagnets [6]. In contrast to collinear AFM systems, we find that the localized <sup>3</sup>A<sub>1g</sub>  $\rightarrow$  <sup>3</sup>T<sub>2g</sub> orbital excitation does little to perturb the non-collinear multiferroic phase of Nil2. Instead, we find that the multiferroic phase can be resonantly quenched by delocalized  ${}^{3}A_{1g} \rightarrow {}^{3}T_{1g}$  orbital excitations, which manifest as a sudden quench of the multiferroic order and the appearance of critical slowdown of the recovery dynamics (see Fig. 2a-c) [7]. The delocalized excitation distorts the electronic wavefunction over a distance larger than the periodicity of the spin-spiral (~30 nm), which likely quenches the inverse DMI more efficiently than localized excitations (Fig. 2c).

Furthermore, our ab-initio microscopic DFT simulations show that the excitation affects wavefunctions in neighbouring van der Waals layer (Fig. 2b). This delocalization may lead to an enhancement of the interlayer exchange which favors the AFM phase of Nil<sub>2</sub> over the multiferroic phase stabilized by the inverse DMI. By tuning polarization, fluence, and wavelength, we aim to enable ultrafast optical control of ferroelectricity and magnetism in 2D vdW multiferroics.

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**Figure 1**. (a) Nil2 crystal and magnetic structure in the antiferromagnetic phase. Interlayer exchange leads ferromagnetically coupled layers that are anti-aligned between each other, red and blue correspond to up and down magnetic moments, respectively. (b) The helical spin-spiral in Nil<sub>2</sub> in the multiferroic phase colors and arrows represent the in-plane component of the magnetic moment. The incommensurate spin-spiral has an approximate periodicity of seven unit cells. The ferroelectric polarization induced along the b-axis breaks inversion symmetry. (c) Second harmonic intensity as a function of temperature is proportional to the multiferroic order parameter P.



**Figure 2.** (a) The time-resolved ultrafast dynamics of the second harmonic (SHG) intensity in response to pump pulses resonant to the localized  ${}^{3}A_{1g} \rightarrow {}^{3}T_{2g}$  orbital excitation (purple) and the delocalized  ${}^{3}A_{1g} \rightarrow {}^{3}T_{1g}$  orbital excitation (orange). The quench in intensity is followed by a relaxation on the order of picoseconds and nanoseconds. Solid lines are fits to the data. (b) The relaxation timescale extracted from the fit of the orange curve in (a). A critical slowdown is observed for the delocalized  ${}^{3}A_{1g} \rightarrow {}^{3}T_{1g}$  excitation. Right side shows the abinitio calculation of the electron-hole wavefunction for the delocalized  ${}^{3}A_{1g} \rightarrow {}^{3}T_{1g}$  orbital excitation, showing the extent of delocalization in the layer (top) and between layers (bottom). (c) The relaxation timescale extracted from the fit of the purple curve in (a). Right side shows the ab-initio calculation of the electron-hole wavefunction.

### The role of the magnetic sublattices on the picosecond spin current generation in ferrimagnetic GdCo

Guillermo Nava Antonio<sup>1</sup>, Quentin Remy<sup>2</sup>, Jun-Xiao Lin<sup>3</sup>, Yann Le Guen<sup>3</sup>, Dominik Hamara<sup>1</sup>, Jude Compton-Stewart<sup>3</sup>, Joseph Barker<sup>4</sup>, Thomas Hauet<sup>3</sup>, Michel Hehn<sup>3</sup>, Stéphane Mangin<sup>3</sup>, and Chiara Ciccarelli<sup>1</sup> <sup>1</sup>Universtity of Cambridge, Cambridge, UK <sup>2</sup>Freie Universität Berlin, Berlin, Germany <sup>3</sup>Université de Lorraine, Nancy, France <sup>4</sup>University of Leeds, Leeds, UK

E-mail: gn312@cam.ac.uk

Ultrafast spintronics aims to exploit the electronic spin to develop unprecedently fast and energyefficient data storage and processing technologies. Rare earth-transition metal (RE-TM) ferrimagnets have emerged as one of the most promising types of systems in this research field, since their magnetization can be directly switched with light [1]. Furthermore, these materials are versatile sources of picosecond spin current when they are excited with ultrashort laser pulses. Such spin current has been employed to reverse the magnetization of an adjacent ferromagnet in spin valve structures [2]. In devices containing ferrimagnetic GdFeCo, it has been claimed that the spin current produced by the Gd sublattice plays a crucial role in the switching of the nearby ferromagnet [3]. On the other hand, other studies have argued that Gd cannot produce a spin current at THz frequencies [4].

In this work, we examine the strength and time scales of the RE and TM contributions to the picosecond spin current generated by GdCo via THz emission spectroscopy. We systematically investigate the temperature and alloy composition dependences of the THz signal, which is proportional to the generated spin current in the frequency domain. As shown in Figs. 1(a) and 1(c), we find that the THz emission vanishes at the magnetization compensation point, when an external magnetic field is applied. However, in case there is no external field, the spin current remains across compensation (see Figs. (b) and (b)).

Previously, this vanishing of the THz emission from RE-TM ferrimagnets close to compensation has been attributed to the cancellation of the spin current produced by the two magnetic sublattices [5]. Our experiments indicate that this explanation does not hold for GdCo since the THz emission persist under zero applied magnetic field. Instead, through magneto-optic Kerr effect imaging and the analysis of the external field dependence of the THz signal, we demonstrate that the suppression of the spin current stems from the formation of domains in the GdCo static magnetic configuration. Subsequently, we investigate why the Gd and Co spin currents do not cancel out. By considering the changes of the THz emission as the Gd atomic concertation (x) is varied (see Fig. 2), we determine that, at room temperature, the picosecond spin current is dominated by hot spin-polarized electrons excited from the Co sublattice. However, at low temperature, we detect a substantial contribution to the THz spin current from Gd, with a spectrum containing, average, lower frequency on components. We ascribe this activation of the Gd spin current to the increase of the spin polarization of the Gd 5d bands with lowering temperature [6]. Therefore, we show that the Gd and Co spin currents cannot fully offset or compensate each other due to their different time scales.

Lastly, we investigate how the spin current spectrum changes across magnetization compensation. Our experiments reveal a blueshift of the THz emission associated with the multi-domain structure formed close to compensation. This effect, together with previously discussed vanishing of the THz signal, underscores the importance of the equilibrium micromagnetic state in the laser-induced spin current generation.

Our results showcase the tunability of RE-TM ferrimagnets as sources of THz spin current and are relevant for the design of optically controllable spin valve devices.

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**Figure 1.** THz emission intensity and GdCo static magnetization as a function of temperature. The THz intensity is defined as the integral of the absolute value of the THz pulses. (a) and (b) THz emission intensity measured under in-plane applied magnetic fields of 8.5 kOe and 0 Oe, respectively. The dashed line in (a) indicates the temperature at which the THz emission is the weakest, denoted  $T_{THz}$ . (c) and (d) In-plane magnetization measured by superconducting quantum interference device (SQUID) magnetometry under magnetic fields of 8.5 kOe and 0 Oe, respectively. The dashed lines in (c) and (d) mark the magnetization compensation temperature  $T_{M}$ .



**Figure 2.** THz emission for different Gd concentrations. (a) and (b) THz signal at 300 K and 6 K, respectively. The multiplicative factors on the left indicate by how much the pulses were rescaled to have the same amplitude as the Co emission (x = 0%) at the corresponding temperature. In (b), the black dashed lines are linear combinations of the Gd and Co signals. The purple (green) dashed lines are the corresponding Co (Gd) components in these linear combinations.

# Electrically assisted photo-magnetic sub-switching regime in garnet films

#### Lukáš Nowak

*T. Zalewski, A. Stupakiewicz* <sup>1</sup> University of Bialystok, Poland

I.nowak@uwb.edu.pl

In the last decade, a plethora of fundamental mechanisms for magnetization dynamics induced by ultrashort laser pulses has been actively discussed. Recently, it was discovered that only by a single laser pulse, nonthermal and reversible photomagnetic switching in Co-doped yttrium iron garnet films (YIG:Co) can be obtained [1]. Microscopically, in this mechanism, an incident linearly polarized pump pulse excites strongly anisotropic garnet ions, generating an effective field of photo-induced magnetic anisotropy [2]. This photo-induced magnetic anisotropy is an essential component of the effective field anisotropy, which can drive the magnetization dynamics described bv LLG formalism, enabling an all-optical impact on the magnetic state at ultrashort timescales. The photomagnetic strong contribution is short-lived (about 30 ps) and is closely tied to an external light pulse, which can trigger the precession or even switch the magnetization if the conditions are sufficient [3]. However, this contribution must be differentiated from other external and internal contributions to the anisotropy which further affect the overall anisotropy and therefore the magnetization dynamics as well. Here, we examine the potential of using an electric field to modify the magnetic anisotropy in YIG:Co films. Employing a garnet patterned with a goldplated comb-like structure of electrical contacts, we apply the electric field to influence the anisotropy. subsequently affecting the frequency of the photomagnetic precession. We investigate the dependency of the amplitude and orientation of the electric field with respect to the crystallographic axes in garnets. Conducted measurements reveal a precession frequency decrease of up to 10% with the external magnetic field parallel to the electric field with the amplitude < 2 MV/m. Additionally, we observe both frequency decrease and increase dependent on the amplitude of the external magnetic field in the case of a perpendicular electric field. This opens new opportunities for controlling photomagnetic dynamics and switching through anisotropy design and manipulation via an electric field. Appropriate design of anisotropy leads to the possibility of optimized magnetization state control by a single ultrashort laser pulse and is the key to using the photomagnetic effect in applicable devices.

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#### Ultrafast Antiferromagnetic Switching of Mn<sub>2</sub>Au with Laser-induced Optical Torques

Jackson Ross<sup>1,2</sup>, Paul Gavriloaea<sup>3</sup>, Frank Freimuth<sup>4,5</sup>, Theodoros Adamantooulos<sup>5,6</sup>, Yuriy Mokrousov<sup>4,5</sup>, Richard Evans<sup>1</sup>, Roy Chantrell<sup>1</sup>, Rubén M. Otxoa<sup>7,8</sup>. Oksana Chubykalo-Fesenko<sup>3</sup> <sup>1</sup>School of Physics, Engineering and Technology, University of York, York, UK <sup>2</sup>School of Physics and Astronomy, University of Edinburgh, Edinburgh, UK <sup>3</sup>Instituto de Ciencia de Materiales de Madrid, CSIC, Madrid, Spain <sup>4</sup>Institute of Physics, Johannes Gutenberg University Mainz, Mainz, Germany <sup>5</sup>Peter Grünberg Institut and Institute for Advanced Simulation, Jülich, Germany <sup>6</sup>Department of Physics, RWTH Aachen University, Aachen, Germany <sup>7</sup>Hitachi Cambridge Laboratory, Cambridge, UK <sup>8</sup>Donostia International Physics Center, Donostia San Sebastian, Spain

#### Jackson.ross@york.ac.uk

Recent *ab initio* work [1] has suggested a novel option for direct Nèel vector control of antiferromagnets (AFMs): the induction of staggered fields using direct optical laser excitation. The frequency dependence of the induced staggered magnetic fields was calculated for optical and THz excitations, and is shown to generate a net non-staggered torque. This torque could potentially switch the AFM order parameter.

We present atomistic spin dynamics simulations of an optical frequency excitation from ultrafast laser pulses on Mn<sub>2</sub>Au [2] using the coupling scheme suggested in Freimuth *et al.* [1]. To distinguish between the other laser excitation torques acting through spin transfer techniques [3], we call this generated torque a laser optical torque (LOT). We focus our work here on demonstrating the possibility to switch the Nèel vector in AFM using purely LOTs in optical frequencies. Additionally, we provide a method using the LOT symmetry to preferentially control the switching direction of the Nèel vector, allowing for deterministic, non-toggle all-optical switching (AOS) in AFM.

Optically-induced torques show strong crystal symmetry and frequency dependent coupling to the polarised electric field components of the laser. A full analysis of the symmetry requirements in the Mn<sub>2</sub>Au bulk crystal is based on the Keldysh non-equilibrium formalism in [1]. The magnitude and spatial symmetry of the predicted torque depends both on the local orientation of the Nèel vector L, as well as the electric field E direction of the applied optical pulse.

The torque tensors depend on the vector components of the electric polarisation and AFM order parameter (Fig. 1). Our chosen laser geometry can be approximated to follow the trigonometric relation  $\sin(2\varphi - 2\phi)$ , with  $\varphi$  the azimuthal angle of the electric field polarisation of the laser, and  $\phi$  the

azimuthal angle of the magnetisation. Taking the physical constants into the variable  $\tau(I, \omega)$ , the induced field from the laser can be written as

$$\boldsymbol{H}_{LOT} = \frac{1}{\mu_c} \tau(\boldsymbol{I}, \boldsymbol{\omega}) \sin(2\varphi - 2\phi) \hat{\boldsymbol{z}} \times \hat{\boldsymbol{S}}$$
(1)

The staggered fields then lead to a non-staggered effective torque. A linearly polarised pulse with  $E \parallel \langle 110 \rangle$  and intensity *I*=10 GW/cm<sup>2</sup>, set at a photon energy of hv = 1.55 eV, will produce a LOT of magnitude  $\approx 12 \times 10^{-24}$  J, which corresponds to an effective field of 17.3 mT on each magnetic moment, canting the local Nèel vector out-of-plane.

Unlike traditional Nèel SOT, the LOT has the additional feature of changing sign during the switching: the intrinsic spatial symmetry defined in Eq. (1) ensures the induced LOT changes its sign for any 90-degree rotation of the Nèel vector (Fig. 1). This allows for both clockwise and counter-clockwise switching by means of the same laser polarisation.

The sin2 reliance on the electric field polarisation of the generated torque in Eq. (1) allows a shift of the maximal torque away from the easy axis by rotation of the laser polarisation vector. Contrary to the toggle switching caused by  $E \parallel \langle 100 \rangle$  or  $\langle 010 \rangle$ , shifting the azimuthal angle of the laser polarisation will create an asymmetric torque profile (Fig. 1). Thus, the magnetisation will experience a larger torque when starting from only two of the four easy axis directions, giving a preference between clockwise and counterclockwise switching. This generation of a quadrantasymmetric torque introduces an additional level of control to the switching process, allowing for preferential, non-toggle switching.

Since Mn<sub>2</sub>Au is a metallic conductor, strong laser intensities will induce transient heating to the system. We include this effect using the common twotemperature model (TTM). Fig. (2) shows the switching probability for different laser polarisations and different starting easy axis directions. Fig. (2a) confirms robust toggle switching for a wide range of pulse durations with no overshooting. Fig. (2b) demonstrates the preferential switching nature between starting easy axes, with Fig. (2c) confirming the ability to toggle switch both starting easy axes with the same laser polarization, changing only the pulse fluence and duration. Thus, a combination of pulses of the same laser polarisation could deterministically switch the Nèel order. That is, could intentionally direct the Nèel order between starting axes without knowing the initial position. This effect is a result of crystal and non-centrosymmetric symmetry breaking, and is not unique to Mn<sub>2</sub>Au. Recent ab initio results have shown this LOT for the altermagnets RuO<sub>2</sub> and CoF<sub>2</sub> as well [4]. This suggests that laser optical torques could be a new and critical feature for AFM spintronics.

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*Figure 1.* Diagram of torque symmetry described by Eq. 1 for various laser polarisations.



**Figure 2.** Switching probabilities including the TTM for different laser polarisations and starting easy axes showing toggle, preferential, and deterministic switching.

# Antiferromagnetic spin-orbit torque oscillator

**P. K. Rout**<sup>1</sup>, J. A. Vélez<sup>2,3</sup>, J. Godinho<sup>1</sup>, F. Vilsmeier<sup>4</sup>, R. Salikhov<sup>5</sup>, Z. Šobáň<sup>6</sup>, D. Lazore<sup>7</sup>, C. H. Back<sup>4</sup>, O. Hellwig<sup>5,8</sup>, R. M. Otxoa<sup>9,2</sup>, and J. Wunderlich<sup>1,6</sup>

<sup>1</sup>Institute of Experimental and Applied Physics, University of Regensburg, Germany

<sup>2</sup>Donostia International Physics Center, San Sebastián, Spain

<sup>3</sup>Polymers and Advanced Materials Department: Physics, Chemistry, and Technology, University of the Basque Country, San Sebastián, Spain

<sup>4</sup>Fakultät für Physik, Technische Universität München, Germany

<sup>5</sup>Institute of Ion Beam Physics and Materials Research, Helmholtz-Zentrum Dresden-Rossendorf, Germany

<sup>6</sup>Institute of Physics, Czech Academy of Sciences, Czech Republic

<sup>7</sup>Instituto de Alta Investigación, CEDENNA, Universidad de Tarapacá, Chile

<sup>8</sup>Institute of Physics, Chemnitz University of Technology, Germany

<sup>9</sup>Hitachi Cambridge Labyoratory, Cambridge CB3 0HE, United Kingdom

Pradeep-Kumar.Rout@physik.uni-regensburg.de

Antiferromagnetic materials have unique properties due to their alternating exchange-coupled magnetic moment arrangements, leading to exchange-field enhanced fast and complex spin dynamics [1, 2]. A nonvolatile antiferromagnetic memory mimicking an artificial synapse with extremely reproducible synaptic weights has been realized in a synthetic antiferromagnet (SAF), in which the reconfigurable synaptic weight is encoded in the ratio between reversed antiferromagnetic domains [3]. The nonvolatile memory is "written" by spin-orbit torquedriven antiferromagnetic domain wall motion and "read" by nonlinear magneto-transport. We also realize a spin-orbit torque driven antiferromagnetic oscillator inside a nano-constriction patterned from a SAF multilayer. By exploiting the spin rectification effect (SRE), we identify spin-orbit torque-driven excitations of optical and acoustic antiferromagnetic modes. Near the spin-flop transition, additional resonant modes appear in the SRE signal when applying a dc current above a critical current density. We associate these additional modes with spin-orbit torque driven antiferromagnetic self-oscillations that are injection locked to SRE the detection frequencies. Macro-spin and micromagnetic simulations of our nano-constriction spin orbit-torque oscillator confirm antiferromagnetic self-oscillations in the studied applied magnetic field regime. The simulations show, also in agreement with our experimental findings, a chaotic behavior of the selfoscillations around the antiferromagnetic spin-flop transition.

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#### THz-driven coupled dynamics of 4f orbitals and 3d spins in rare-earth orthoferrites

M. R. Vovk<sup>1</sup>, O. Y. Kovalenko<sup>1</sup>, R. Dubrovin<sup>2</sup>, R.V. Pisarev<sup>2</sup>, A. Kalashnikova<sup>2</sup>, R. V. Mikhaylovskiy<sup>1</sup> <sup>1</sup> Department of Physics, Lancaster University, Bailrigg, Lancaster, United Kingdom <sup>2</sup>loffe Institute, Saint Petersburg, Russia m.vovk1@lancaster.ac.uk

The rare-earth orthoferrites (REO) have been attracting significant attention since their discovery in the 1940s due to unique magnetic properties, such as spin reorientation phase transitions (SRPT), strong magneto-optical effects, THz-frequency spin dynamics, multiferroicity, and quantum magnonics. Recent advancements in the development of THz time-domain spectroscopy (THz-TDS) techniques allowed the study of various types of excitations with meV energies, including spin dynamics, by directly interacting with atomic spins using THz radiation. Regarding the REOs, an interesting idea was to use intense THz fields to achieve control over nonlinear SRPT and switch of iron spins due to the changing crystalline magnetic anisotropy via the REO subsystem, driven by THz pulses [1-2]. This raises the question regarding the exchange interaction between rare-earth and iron orbitals in a non-equilibrium state. Moreover, different scenarios for dynamic coupling are possible depending on the type of a rare-earth ion.

To address this issue, we have formulated a theoretical model based on the microscopic approach [3] to elucidate the magnetic switching in REOs with non-Kramers ions subjected to strong THz excitation [1-2]. Using the archetypical orthoferrite TmFeO3 as a model system, we investigated the static properties of the R and Fe subsystems across the SRPT. Employing an adiabatic approximation, we determined the resonance frequencies for the Fe and R magnetic sub-lattices as a function of temperature, aligning our findings with experimental data from [1]. We then performed numerical modelling to accurately describe the behaviour of its anisotropy functions vs temperature. Through numerical modelling, we described the behaviour of anisotropy functions relative to temperature, identified threshold fields for spin switching-whether via Zeeman mechanisms or anisotropy-driven torques altered through the Rsubsystem—and estimated the energy dissipation during the switching process, achieving excellent correlation with the experimental values from [2].

Furthermore, we extended our model to interpret our recent experimental results on TbFeO<sub>3</sub>, a unique member of the orthoferrite family. In our experimental work we employed ultrashort terahertz (THz) pulses to excite low-energy excitations of Fe<sup>3+</sup> spins and Tb<sup>3+</sup> atomic-like transitions to reveal the character of their coupling and hybridization. The magnetisation in TbFeO<sub>3</sub> mainly arises from the

spins of the Fe<sup>3+</sup> ions, whereas the interaction of the spins with the Tb<sup>3+</sup> electronic orbitals sets the character of the magnetic configuration. The TbFeO<sub>3</sub> exhibits a second-order magnetic phase transition of the Jahn-Teller type at temperatures below 8.5 K. This SRPT originates from the interplay between two anisotropy energies: Tb-Fe exchange and crystalline anisotropy energies. As a result, TbFeO3 exhibits a cross-over (i.e. equality) of the frequencies of the antiferromagnetic resonance mode and atomic-like mode of Tb, which leads to their dynamical repulsion and avoided crossing effect. The coupling is so strong that the frequency of the low-lying hybrid mode is pushed to zero. Using our model, we managed to model dynamics between interacting Fe and Tb magnetic subsystems, and to fit it with an excellent match to our experimental findings (Fig.1).

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**Figure 1.** Temperature dependence of the resonance frequencies  $\omega_{qFM}$  and  $\omega_{Tb}$  in TbFeO<sub>3</sub>: panel (a) the Fourier map plot of resonance frequencies obtained from polarisation rotation measurements; panel (b) comparison between experimental data and theoretical curves, accounting for the dynamical repulsion. The dashed lines indicate phase transitions temperaturas T<sub>1</sub> and T<sub>2</sub>.



EDITED BY PHANTOMS FOUNDATION Alfonso Gómez 17

28037 Madrid, Spain www.phantomsnet.net