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Habilitation à Diriger des Recherches : Ultrafast spintronics

J. Gorchon

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J. Gorchon. Habilitation à Diriger des Recherches : Ultrafast spintronics. Other [cond-mat.other].
Université de Lorraine, 2023. tel-03966866

HAL Id: tel-03966866

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Habilitation à Diriger des Recherches

Spécialité: *Physique*

Présentée par

Jon Gorchon

Habilitation soutenue publiquement le 26 janvier 2023 à Nancy devant le jury composé de:

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30 OCTOBRE 2023

Table of contents

1. Introduction.....	6
2. Short bio	7
3. Curriculum Vitae.....	9
4. Research work.....	12
a. Manipulating spins and speed limits.....	12
b. All-optical magnetization switching (AOS)	16
i. AOS in ferrimagnetic materials	17
ii. AOS in ferromagnetic materials	19
iii. AOS in combined ferri and ferromagnetic heterostructures	20
iv. AOS in fully ferromagnetic spin-valves.....	23
c. THz spintronics	26
i. THz spin heating	27
ii. THz spin injection	30
iii. Spintronic THz emission	34
d. Ongoing / Future work	36
e. Active collaborations.....	37
5. Experimental developments	38
6. Mentoring of students.....	42
7. Responsibilities and other works	44
8. References.....	48

Acknowledgments

I am very fortunate to have the job I have, with the colleagues that I have. For this, I have to specially thank all the people who've trained me, worked with me and guided me for years: Vincent Jeudy, Jacques Ferré, Javier Curiale, Jeffrey Bokor, Richard Wilson...

I also have to deeply thank all the members of my team, the SPIN TEAM. First, and foremost I would like to thank the people who got me here: Stéphane and Gregory. Eighth years ago I couldn't have imagined coming to Nancy, even less as a CNRS researcher. I couldn't be happier now. Stéphane, thanks for everything you do for me and for the group, I know it's not easy, I do not know how you can possibly find the energy to do everything you do, and I admire you for it. And Greg, you know how much of a pleasure it is for me to work with you...

Since I arrived to the lab, the person who has helped me the most to grow into a proper advisor and who has pushed me forward in my career, has been Michel Hehn. In addition, Michel has also grown all of my samples, and every single time in less than 24 hours. Such a pleasure to work with somebody like you. Thank you so much for your constant support Michel, I can't thank you enough.

Big thanks to all my friends/colleagues in the lab that make my day to day so much fun, whether we discuss science, drink coffee/beer or play ping-pong. Thanks Karine, Seb, Thom, Olivier, Carlos, Dan, François and all of the students.

And last but not least, a big thank you to my lovely wife and son, Djeyda and Oihan. Milesker familia! Muxu handi bat!

1. Introduction

The *Habilitation à Diriger des Recherches* (HDR) is the highest degree diploma in France. It was created* in 1988 as a way of recognizing a researcher's high scientific level, the originality of his/her work as well as his/her ability to master a research strategy within a broad scientific field and to mentor students. Moreover, the HDR allows researchers to access the positions of full professor within a University or that of Director of Research within the CNRS.

It is actually not all that clear what an HDR manuscript should or should not be. The format is loosely defined, but there are a few guidelines†: Importantly, an HDR manuscript is not a thesis or a memoire. The HDR should be a *light* synthesis of the research activities since the PhD, highlighting the role of the researcher and the quality of the research. Finally, the HDR should also present the mentoring activities of the researcher and his capability to guide students.

Therefore, in the following manuscript I will try to briefly present both my research and non-research activities, as well as their impact in a broader context, including my mentoring experience. This manuscript is aimed at a scientific jury of my choosing, and I will therefore try my best to keep it interesting for them !

* See ordinance: *arrêté* de 23 novembre 1988

† See circular: *circulaire relative au doctorat et à l'habilitation à diriger des recherches (ministère de l'éducation nationale et de la culture 27 Octobre 1992)*

2. Short bio

I was born in the UK, raised in San Sebastian (Spain) and have spent most of my academic career in France. I now carry my work at the Institut Jean Lamour (IJL), within the Université de Lorraine, as a CNRS scientist. I am a member of the SPIN Team which focuses on growth and characterization of magnetic materials.

I obtained an engineering master's degree in 2006 from the Institut National des Sciences Appliquées in Toulouse, with a major in Physics. During my bachelor I was lucky enough to spend a year at the University of Tohoku in Japan, where I was affiliated to a magnetic recording lab and was introduced for the first time to the academic world of research. Following my master's degree, I embarked on a PhD at the Université Paris-Sud (now rebranded as Paris-Saclay), at the Laboratoire de Physique des Solides, under the supervision of Pr. Vincent Jeudy. My PhD topic consisted on the study of current and field induced *slow* magnetic dynamics. For example, we studied the *creep* regime of magnetic domain walls pushed by a magnetic field as a function of temperature and tested the robustness and universality of such regime. During this time, I also dedicated some time to a science outreach program under Pr. Julien Bobroff's guidance, in which I visited science fairs and schools in order to talk to the general public about science and magnetism. I thoroughly enjoyed my PhD time.

In 2015, I joined the Lawrence Berkeley National Lab and the University of California Berkeley for a postdoctoral stay in the group of Pr. Jeffrey Bokor. There I transitioned to the study of *ultrafast* magnetic dynamics. As a postdoc, I also took a more involved role in the mentoring of Jeff's students. During my time in the lab, I learned extensively about femtosecond lasers and optics, nanoscale thermal transport, and electronics and material science in general. The weekly talks, open classes and extremely dynamic atmosphere of the Berkeley Bay area constituted an incredible environment for me during this period of time. Together with a number of students in the group, we had a pretty prolific time. We were able to demonstrate a number of new phenomena in the realm of light-induced ultrafast magnetization dynamics and unlocked a few speed world records along the way. Also during this time, we started exploring new ways of manipulating magnetization via ultrafast means. Particularly, we began to use short electrical terahertz (THz) bursts generated with photoconductive switches and demonstrated THz heating induced demagnetization and switching. This new line of work perfectly coincided with the rise of THz spintronics, a relatively young but exciting field where THz dynamics in magnetic systems are explored.

Following these works I obtained a CNRS position in France end of 2017. I moved to Nancy, to the IJL, joining one of the world-leading groups in ultrafast magnetism. A few years later, I had setup my own experimental platform, and with my first PhD student, Kaushalya Jhuria, we were generating THz burst on our devices. Soon after, we were able to demonstrate for the first time that ultrashort spin currents in

the picosecond range could be used to reverse the magnetization, and got to observe the dynamics due to the spin torques with sub-picosecond resolution. These experiments bring up many interesting questions, which will likely form the bedrock of my future work.

Besides the various scientific works and collaborations that I have been a part of since my joining of the CNRS, I have obtained a ANR young-researcher funding for 270k€, I have lead one of the main work-packages of a national 38M€ spintronics ministerial program (PEPR exploratoire), I have been invited to various conferences and seminars, I have received two scientific prizes, and I have been the lead web-developer and editor of the team's website, among other things.

So far, I have been extremely lucky during my career, for which I am extremely grateful. The people I met, the scientist that advised me, the students I mentored, have all played a major part in my career, and I couldn't be where I am without all of them. Today, with all the world instabilities and the challenging aspects of academia, I believe it is more than ever extremely difficult to complete a PhD (particularly in 3 years!). This is why I consider mentoring and accompanying students as the most critical part of my job. By obtaining my HDR I wish to take in students, ideally in collaborations with other scientists/advisors, to give them all the chances for success, and hopefully discover interesting new things together along the way.

Finally, I would just like to add a word about a certain new aspect challenging my life as a researcher, which is the carbon budget of what I do. As researchers we are meant to exchange internationally, to travel, build new setups... but we are also well aware of the science of climate change, the impact of our work, and the example we should be giving. I first thought that our emissions were minimal and necessary for our jobs and that doing research was a greater good anyway. Also, selfishly, I wanted to secure my career by following the *classical* path. Nowadays though, I feel these arguments no longer hold. There are similar things to be said with the ethics of the publishing system, grants system... that I do not fully agree with, but partake in anyway. These aspects do not only affect me, but also my colleagues and students, which make it more difficult to change. I hope that I (we) can adapt and hold the right values while keep being involved in science at the highest level.

3. Curriculum Vitae

Education & Professional experience

- **CNRS permanent researcher** @ Institut Jean Lamour (Nancy) 12/2017 – present
- **Postdoctoral fellow** @ University of California, Berkeley (Berkeley, USA) 01/2015– 12/2017
- **PhD in physics** @ Laboratoire de Physique des Solides, Université Paris-Sud (France) 2011 – 2014
- **Engineering diploma** @ l’Institut National des Sciences Appliquées (Toulouse, France) 2006 – 2011

Funding

- **PEPR Exploratoire: SPIN** (program board, work-package lead) – 38 M€ 2022 – 2028
[project includes 47 french labs and 443 permanent researchers]
- **ANR JCJC: SPOTZ** (coordinator) – 269 060€ 2021 – 2024
- **ANR: UFO** (participant) – 540 000€ 2021 – 2024
- **University of Lorraine grant (for “pôle M4”)** (coordinator) – 14 500€ 2020
- **FEDER (EU) + regional « plateforme nanoTHz » fund** (participant) – 269 060€ 2018 – 2022
- **ANR: MISSION** (participant) – 202 000€ 2018 – 2022
- **French-Berkeley Fund** (coordinator) – \$12 000 for mobility 2017 – 2019
- **C’Nano Ile-de-France PhD grant** (beneficiary) – 3 year salary + 20 000€ 2011 – 2014

Awards

- **Prix Zivi** for young researchers associated with a laboratory of the Lorraine University - given by the Stanislas Academy – 2 000€ 2021
- **Prix Edouard Branly** for young researchers who have made a remarkable contribution in physics, particularly in the fields of optics or electronics, with possible future applications - funded by the French Federation of Scientific Societies (F2S), French Physics Society (SFP) and IEEE – 2 500€ (shared with Jean Comtet) 2021

Student mentoring

- Jeremy Létang, **Postdoc (co-advisor)**: “*optical detection of spin accumulation*” 11/2021 – 06/2022
- Eva Luisa Díaz González, **PhD student (co-advisor)**: “*picosecond spintronics*” 10/2021 – present
- Bekim Fazliji, **M1 internship (advisor)**: “*design de lignes RF pour les THz*” 10/2020 – 06/2021
- Kaushalya, **PhD student (co-advisor)**: “*magnetic control with on-chip THz pulses*” 10/2018 – 10/2021
- Akshay Pattabi, **PhD student (mentor)**: “*optical detection of the spin Hall effect*” 01/2015 – 12/2017
- Yang Yang, **PhD student (mentor)**: “*ultrafast magnetism and spintronics*” 01/2015 – 03/2017
- Sajant Anand, **L2 internship** : “*layer transfer of GaAs films onto various substrates*” 06/2016 – 08/2016
- Romain Faubert, **M1 internship** : “*STT on magnetic bubbles in (Ga,Mn)(As,P)*” 06/2013 – 09/2013







Other responsibilities

· Developer , designer, author & photographer of team’s website : https://spin.ijl.cnrs.fr	01/2020 – present
· Expert evaluator of European projects (Marie Sklodowska-Curie)	10/2019 – 12/2019
· Organization assistant for the Tohoku-Mainz-Lorraine Workshop	09/2019
· Journal reviewer : Nat. Elec., Nat. Com. Phys. Rev. X, Phys. Rev. Lett., Adv. Sci...	12/2017 – present
· Co-responsible of 2 “ <i>ultra-fast optics</i> ” experimental rooms @ IJL (Nancy, France)	01/2018 – present
· Member of workgroup “ <i>international relations</i> ” @ IJL (Nancy, France)	04/2018 – 01/2020
· Member of workgroup “ <i>lab website design</i> ” @ IJL (Nancy, France)	01/2018 – 12/2020
· Member of workgroup “ <i>diversity & inclusion</i> ” @ LBNL (Berkeley, USA)	09/2016 – 09/2017

Oral Contributions

Over 20 invitations to international conferences, workshops and seminars, including SPIE: Spintronics, Interomag, the Online Spintronic Series Seminars, Spin Argentina...

Selected Publications

→ 32 articles (3 PRL, 1 Nat. Elec, 1 Sci. Adv., 1 Adv. Sci., 2 Adv. Mat...), ~900 citations	
<i>Picosecond spin-orbit torque induced coherent magnetization switching in a ferromagnet</i>	2022
D. Polley, Akshay Pattabi, Ashwin Rastogi, Kaushalya Jhuria, Eva Diaz, Hanuman Singh, Aristide Lemaitre, Michel Hehn, J. Gorchon , J. Bokor	
<i>In preparation</i>	
<i>Sub-picosecond magnetization switching in ferromagnetic spin valves</i>	2022
J. Igarashi  , W. Zhang, Q. Remy, E. Díaz, J.X. Lin, J. Hohlfeld, M. Hehn, S. Mangin, J. Gorchon  , G. Malinowski	
<i>under review</i> ( corresponding author)	
<i>Accelerating ultrafast magnetization reversal by non-local spin transfer</i>	2022
Q. Remy, J. Hohlfeld, M. Vergès, Y. LeGuen, J. Gorchon , G. Malinowski, S. Mangin, M. Hehn	
<i>under review</i>	
<i>Is terahertz emission a good probe of the spin current attenuation length?</i>	2022
J. Gorchon  , S. Mangin, M. Hehn, G. Malinowski 	
<i>Appl. Phys. Lett.</i> 121 , 012402 ( corresponding author)	
[Perspective article] <i>Progress toward picosecond on-chip magnetic memory</i>	2022
D. Polley, A. Pattabi, J. Chatterjee, S. Modal, K. Jhuria, H. Singh, J. Gorchon , J. Bokor	
<i>Appl. Phys. Lett.</i> 120 , 140501	
<i>Spin-orbit torque switching of a ferromagnet with picosecond electrical pulses</i>	2020
K. Jhuria, J. Hohlfeld, A. Pattabi, E. Martin, A.Y. Arriola Córdova, X. Shi, R. Lo Conte, S. Petit-Watelot, J.C. Rojas-Sánchez, G. Malinowski, S. Mangin, A. Lemaître, M. Hehn, J. Bokor, R.B. Wilson, J. Gorchon  , ( corresponding author)	

Nature Electronics **3**, 680-686 (2020)

[Review article] *Progress towards ultrafast spintronics applications* 2019
Amal El-Ghazaly, **J. Gorchon**, R.B. Wilson, A. Pattabi, J. Bokor,
J. Magn. Magn. Mater. **502**, 166478 (2019)

Single-shot multi-level all-optical magnetization switching mediated by spin transport 2018
S. Iihama, Y. Xu, M. Deb, G. Malinowski, M. Hehn, **J. Gorchon**, E.E. Fullerton, S. Mangin,
Advanced Materials **30**, 1804004 (2018)

Single-shot ultrafast magnetization switching of ferromagnetic Co/Pt multilayers 2017
J. Gorchon* C.H. Lambert*, Y. Yang*, A. Pattabi, R.B. Wilson, S. Salahuddin, J. Bokor *equal contribution
Appl. Phys. Lett. **111**, 042401 (2017)

Ultrafast magnetization reversal by picosecond electrical pulses 2017
Y. Yang*, R.B. Wilson*, **J. Gorchon***, C.H. Lambert, S. Salahuddin, J. Bokor *equal contribution
Science Advances **3**, 11, (2017)

Ultrafast magnetic switching of GdFeCo with electronic heat currents 2017
R.B. Wilson*, **J. Gorchon***, Y. Yang, C.H. Lambert, S. Salahuddin, J. Bokor *equal contribution
Phys. Rev. B. Rapid Communications **95**, 180409(R)

Electric current induced ultrafast demagnetization 2017
R.B. Wilson*, Y. Yang*, **J. Gorchon**, C.H. Lambert, S. Salahuddin, J. Bokor *equal contribution
Phys. Rev. B **96**, 045105 (2017)

Model for multishot all-thermal all-optical switching in ferromagnets 2016
J. Gorchon, Y. Yang, J. Bokor
Phys. Rev. B. Rapid Communications **94**, 020409(R) (2016)

Pinning-dependent field driven domain wall dynamics & thermal scaling in an ultrathin Pt/Co/Pt magnetic film 2014
J. Gorchon, S. Bustingorry, J. Ferré, V. Jeudy, A.B. Kolton, T. Giamarchi
Phys. Rev. Lett. **113**, 027205 (2014)

Patents

-
1. *Writing of a magnetic memory with electric pulses* 2019
Y. Yang, **J. Gorchon**, R.B. Wilson, C.H. Lambert, S. Salahuddin, J. Bokor
US Patent 10 388 349

4. Research work

My most recent research has mainly focused on the control and understanding of magnetization at fast timescales. To this aim, we use femtosecond lasers and THz sources, and try to induce fast magnetization dynamics. In the next section, I will give a brief and general introduction to magnetization dynamics under a few different conditions, and the expected speed limits. In the following sections I will discuss some major results in the field and present some of my contributions. For further reading, there are three recent review/perspective articles that have recently been published which discuss part of my results¹⁻³. I would like to emphasize that the presented literature is not meant to be complete, but should constitute the minimum necessary to give context to my work.

a. Manipulating spins and speed limits

Magnetic field induced dynamics:

Spins (S) and their associated magnetization (M) can be controlled by a number of different stimuli such as magnetic fields, currents, electric fields, heat, strain, light... We can understand the influence of these stimuli on the magnetization in different ways. We'll first start here with a macrospin picture, where we consider an ensemble of N parallel spins. This results in a net magnetization \vec{M} of constant amplitude M_s . A common and convenient way to express its dynamics is as a function of the torques that act upon \vec{M} , as is described by the Landau-Lifshitz-Gilbert (LLG) equation:

$$\frac{d\vec{M}}{dt} = -\gamma\vec{M} \times \vec{H} + \frac{\alpha}{M_s}\vec{M} \times \frac{d\vec{M}}{dt} \quad \text{Eq. 1}$$

Where γ is the free electron's gyromagnetic ratio and α is the damping parameter. Naturally, all possible torques can be decomposed into two orthogonal components, which lead to the rotation of \vec{M} . In the original LLG equation an effective magnetic field induces two such orthogonal torques: a (Larmor) precessional or field term ($\propto \vec{M} \times \vec{H}$) and a damping term ($\propto \vec{M} \times \frac{d\vec{M}}{dt}$), which can also be written as a function of the applied field ($\propto \vec{M} \times \vec{M} \times \vec{H}$). The precessional term includes any conservative forces (a gradient of some energy landscape) that affects the magnetization such as the exchange interaction, the magnetic anisotropy or external stimuli like an external magnetic field. This term however is, again, conservative, meaning it doesn't allow for energy dissipation nor angular momentum variations in the system. Such dissipation is allowed only by the damping term which enables the coupling of the spin angular momentum bath to the exterior (ex: to the crystal lattice via the spin-orbit interaction). Therefore, when applying a magnetic field, the magnetization first precesses around the field, and slowly dissipates

energy and falls into a new equilibrium position (parallel to the field), where it stops its motion. In the situation where the field is almost antiparallel to the magnetization and induces its full reversal, clearly the change in spin angular momentum in the system needs to be supplied by some exterior bath. For example, if it's the lattice that supplied it, this should result in the Einstein-de Hass effect, which induces a rotational motion of the lattice. But the system could also change its angular momentum by other means: emission/absorption of circular polarized light, spin transport away from the system... The damping term is a phenomenological parameter that englobes all of these possible mechanisms.

The speed of the dynamics can be separated in two categories: the precessional speed, given by the product γH , and the damping speed given by $\alpha\gamma H$, which determines the time required to align the magnetization along the magnetic field H . Therefore, if we want to control the magnetization fast, we will have to use these terms to our advantage. On the one hand, one can exploit the damping term to determine the final state of the magnetization. In order to increase its speed one has to increase the magnetic field H (to increase the $\overline{dM/dt}$ term), and the damping parameter α . However, because $\alpha < 1$ the damping dynamics will always be slower than the precessional ones. On the other hand, in order to reverse the magnetization using the faster precessional term, one needs to have a perfect control on the timing of the field application in order to stop the dynamics when the magnetization has performed about half a precessional period. Also, since the intensity of the field sets the precession period, using an inhomogeneous field with a fixed pulse duration can lead to a spatially dependent outcome, as was nicely demonstrated⁴ by Back et al (see Fig. 1).

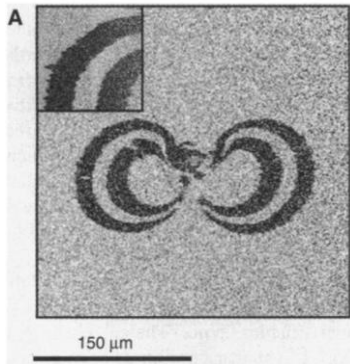


Fig. 1: Magnetization pattern after a single 4.4 ps magnetic field pulse. Because the magnetic field is non homogeneous (stronger in the center) a bull's eye pattern appears, where the magnetization has turned by 180° in the black outer ring, then 360°, 540°, and so on. Figure extracted from Ref⁴.

There are two interesting questions that one can wonder at this point: first, if we can switch the magnetization by exploiting the “dissipation-less” precessional term, where is the angular momentum coming from? And second, how fast could theoretically one reverse the magnetization using such precessional term? The answer to the first question is, from the torque. The angular momentum is changed by the torque, and the torque is generated by the magnetic field H . This means that the angular momentum

should be supplied by the electromagnetic field (or inversely, the time-varying magnetization should be giving back angular momentum to the field). In fact, this is not an obvious question, and the problem seems to be a topic of quantum electro-dynamics⁵ which seems to involve virtual photons that mediate the electromagnetic interaction between the coil and the spins and allow such exchanges of angular momentum. I will definitely not delve into this aspect here (I am not really able to discuss it any further) but I do believe it is interesting to think about. Concerning the second question, from Eq. 1 it appears that as long as we can increase the magnetic field H , the dynamics should just keep getting faster. Even if there is a very strong internal field, such as the magnetic anisotropy, if a stronger external field is applied, dynamics would be governed by such dominant field. Fundamentally, since no interaction with the lattice is necessary (such as in the case of the damping term), the speed should not be limited by the spin-orbit interaction timescale (from Heisenberg's uncertainty principle, for a 50 meV spin-orbit energy, this timescale is estimated⁶ to >20 fs). This means there could even be a regime where precession happens faster than any damping can take place. The upper speed limit though is not trivial to predict, and is potentially related to the interaction between the electromagnetic field and the spins as discussed earlier⁵. In practice though, it is extremely difficult to generate such a short and intense magnetic field. Take for example a patterned single spire with a relatively small 1 μm radius r and a whopping 1 A of current I , the magnetic field in the center would be $\mu_0 I / 2r \sim 0.6$ T, which would result in dynamics in the $\gamma H \sim 17$ GHz range and require a 30 ps timing control.

Spin-transfer torques:

Since magnetic fields are generally inefficient to generate, we can use other stimuli to manipulate magnetization at fast timescales. One of the first discovered alternatives is that of electrical currents. More precisely, when electrical currents are used to induce spin transport, the injection of spin angular momentum in a magnet can lead to dynamics. This is known as the spin-transfer torque (STT) effect⁷. Depending on how this injected spin interacts at the interface of the magnet, two possible torques can appear: a field-like torque ($\propto \vec{M} \times \vec{\sigma}$) that induces a precession of the magnetization around the injected spin density $\vec{\sigma}$, or a damping-like torque ($\propto \vec{M} \times \vec{M} \times \vec{\sigma}$) that aligns the magnetization with the spin density. Both of these torques are due to the exchange scattering of conduction spins with the localized magnetization, and generally, it is the damping-like torque that is dominant. The speed of a magnetization reversal depends on, among other parameters, the field to damping-like ratio and on the direction of the injected spin density. If the spin density is perfectly antiparallel to the magnetization, no torques should be present. In practice though, there is always some finite angle, which can be further enhanced by thermal fluctuations, and so a small torque can start acting on the magnetizations and grow as the magnetization is carried away from its original equilibrium position. This initial lag is often called the thermal activation delay. However, if the spin density that is injected is orthogonal to the magnetization, then an immediate torque should act on \vec{H} . Other than these differences about the geometry of the torques, as with magnetic

fields, the STT is sped up as the intensity (the spin injection or current) is increased. However, one should keep in mind that the amplitude of this current may also be limited by possible damaging of the material.

Non-coherent dynamics:

Before closing of this introduction, we need to briefly consider the dynamics in real systems, where the magnetization has some spatial dependence. Unlike in the macrospin approximation, where all spins point in the same direction, in actual magnetic systems the spins are coupled via exchange and dipolar interactions, and the external stimuli are not necessarily homogeneous. These facts result in non-homogeneous dynamics of the magnetization. LLG models can still capture most of the dynamics, but in this case the systems need to be divided into smaller volumes and a single macrospin model is used for each sub-volume (down to the atomic dimensions in atomistic LLG models). Even in the smallest devices that have been studied experimentally (around 20 x 20 nm), reversal is governed by a two-step process that includes magnetic domain nucleation first and sub-sequent domain wall propagation^{8,9}. These processes have therefore important consequences on the speed of reversal of a magnetic system. In addition, importantly, as soon as there is a spatial distribution of spins, magnons can be populated. The macrospin Larmor precession around the magnetic anisotropy field, called the ferromagnetic resonance mode, is in fact non-other than the lowest wave-number magnon (largest wavelength magnon, the homogeneous mode). Higher coherent magnon modes can lead to THz dynamics (mostly driven by the exchange interaction). If the magnons are incoherent, they are related to an effective higher magnetic temperature and lead to a reduction in the saturation magnetization M_S .

This introduction has only brought up a few of the many elements that need to be considered when attempting to control the magnetization at fast timescales. In the following sections, I will focus on ultrafast optics and THz as a way to affect the magnetization and will further dig into the physics at play.

b. All-optical magnetization switching (AOS)

The dream of controlling magnetization with light has long been sought after. The first results in this direction were mostly concentrated in semiconductor materials, where direct bandgap absorption of circularly polarized photons can lead to a net spin polarization in the materials¹⁰. However, to this day, there is no ferromagnetic semiconductor material with a room temperature Curie temperature. For this reason, the field of magnetism has mostly focused on metals and insulators. The first discovery of a drastic and ultrafast change in the magnetization of a ferromagnetic metal was made¹¹ by Beaurepaire et al. in 1996. This work has led to numerous studies on the ultrafast interaction of light with magnetic systems, which attempted to answer questions such as *where is the angular momentum going, via which mechanism, and how can we control or use this demagnetization to our advantage*. Since, light induced heating has been considered as a possible way of lowering the energy barrier for reversal^{12,13}, known as heat-assisted magnetic recording (HAMR), but this reversal method still requires some magnetic field of some sort. Another direction explored, particularly by the group of Pr. Rasing at Nijmegen in the Netherlands, has been that of using non-thermal effects such as the inverse Faraday effect. In 2005 the group published a paper¹⁴ where they demonstrated helicity-dependent dynamics in an antiferromagnetic oxide system (TmFeO_3) that they attributed to an opto-magnetic torque. The group had to wait a few more years before finally discovering a light induced, magnetic field-free, reversal of the magnetization: the all-optical magnetization switching, AOS (shown in Fig. 2).

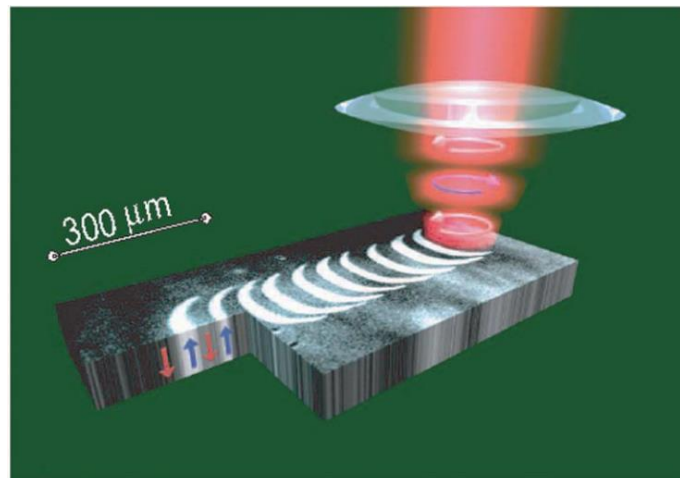


Fig. 2: Schematic of first demonstration of the all-optical switching, without any magnetic field assistance. The laser writes magnetic domains of alternating polarization by using the helicity of the light. Figure extracted from ref¹⁵.

i. AOS in ferrimagnetic materials

In 2006, the group of Nijmegen discovered that GdFeCo could reverse its magnetization due to exposure to circularly polarized light¹⁵. Initially it was believed to be due to the inverse Faraday effect. However, they soon realized that even a single pulse of linearly polarized light could induce switching^{16,17}. The reversal was then attributed to the ultrafast heating of the electron bath, and to the peculiar properties of the ferrimagnetic material. Initially, it was suggested that the very high electronic temperatures following light absorption allowed each sub-lattice (Gd and FeCo) to demagnetize freely at their own rates, and once the FeCo would be demagnetized, the exchange interaction could enable exchange of angular momentum between the sub-lattices leading to a final reversed state. A few subsequent works tried to distinguish between thermal and non-thermal contributions to the switching¹⁸⁻²⁰ and varied some of the experimental parameters to gain more insight.

In 2016, we started looking at the question of how hot the electrons and/or lattice really needed to get for switching to take place. To this aim, we performed²¹ single pulse reversal experiments with linear polarized light (which should eliminate non-thermal effects) at various initial temperatures, and with different pulse durations. Surprisingly, we found that we could obtain AOS even with 15 ps long pulses, with only a slight increase in the threshold fluence. When estimating the electronic temperatures, we found that the electronic temperature could reach 1600 K for short pulses, but only around 550 K for longer pulses, meaning high electron temperature requirements were off the table (see Fig. 3). This work seemed to point towards a mechanism where non-thermal electrons (electrons with ~ 1.5 eV energy, such as those excited by the laser) may not play an important role in AOS as previously thought, and that only the fast heating of the electrons with respect to the lattice would be important.

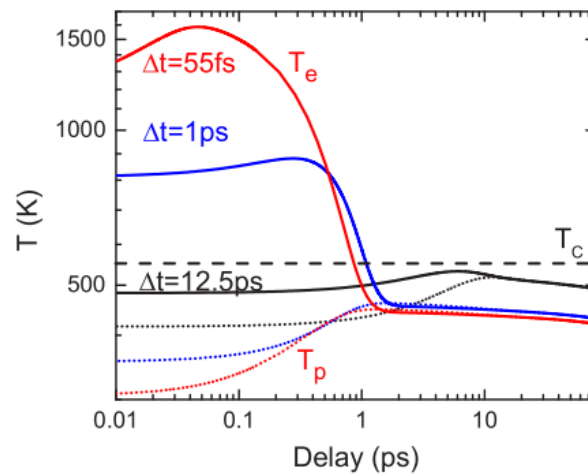


Fig. 3: Simulation of temperature evolution of electrons and phonons for various switching conditions (Δt being the pulse duration) using a 1D 3 temperature model (spin temperatures not plotted). Figure extracted from ref²¹.

In order to gain more knowledge on the question of the electronic distribution, we made similar experiments on samples with a variable thickness of gold on top and a thin platinum capping layer²², as shown in the inset of Fig. 4. These top layers largely reduced optical absorption of photons by the GdFeCo layer, particularly for thick Au layers (see the left Fig. 4). In the thicker Au samples, heat could only reach the GdFeCo via electronic transport through the Au layer, largely reducing the amount of high-energy electrons and the non-thermal character (unless purely ballistic transport was present). We found that indirect-AOS was still possible even with the thickest of Au spacers (up to 200nm), induced by fast electronic heat transport. The dynamics got slightly slowed down due to the electronic transport lag and pulse dispersion, but was still in the picosecond range. Heat-diffusion calculations allowed us to estimate a constant threshold energy absorption for all Au thicknesses, when including both direct optical absorption and diffusive heat absorption (see right Fig. 4). The thermal model could also estimate the observed picosecond lags in the dynamics. These calculations supported a fully diffusive transport scenario with low energy electrons, and motivated us for a future experiment: Attempting to switch GdFeCo with \sim meV THz photons, using short current bursts (see section i. THz spin heating).

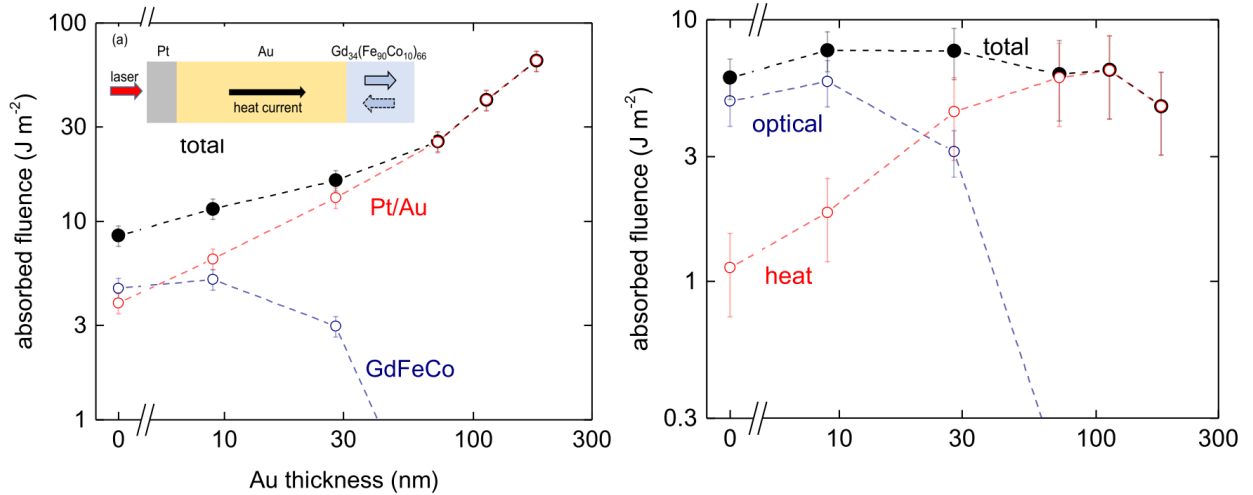


Fig. 4: Left) calculated absorbed fluence as a function of Au thickness. Inset, experiment schematic with Pt/Au/GdFeCo structure. Right) calculated absorbed fluence within the GdFeCo layer when including both optical absorption and heat current absorption. Figure extracted from ref²².

As a side comment, it is interesting to note that in recent years a number of different ferrimagnetic systems that present AOS have been discovered: Gd/Co multilayers²³, MnRuGa²⁴, Tb/Co multilayers²⁵... The full understanding of AOS in all these systems is still evolving.

ii. AOS in ferromagnetic materials

In 2014 my current group (I joined only in 2018) in collaboration with Eric Fullerton at UCSD, made the discovery^{26,27} that not only GdFeCo could be switched optically, but a whole family of materials, including ferromagnets. However, very quickly it was understood that the mechanism was distinct from the GdFeCo one. The reversal could only happen with helicity and after numerous femtosecond laser pulses. The initial understanding of the community was that the helicity of the light could impart some angular momentum, or act like a field (such as in the inverse Faraday effect¹⁴) and that combined with laser heating, eventually the magnetization could be reversed. In 2016 I proposed²⁸, almost at the same time as Roy Chantrel and Eric Fullerton²⁹, an alternative explanation: that magnetization could be *deterministically* reversed purely by *stochastic* thermal activation, if sufficient magnetic circular dichroism was present. In this model, I first assumed that magnetic cells were isolated from each other (thermally and magnetically), which is almost the case in granular FePt where AOS had been observed²⁶. After light absorption, thermal activation leads to switching with a certain probability, which is higher if the cell absorbs more light (see simple model schematic in Fig. 5). This probability for a single event can never be higher than 50% though. Only when calculating the cumulative probability after many pulses, if there is enough asymmetry in the heating of up and down magnetized cells, we get that the switching probability approaches 1. Since, various works have confirmed the possibility that both dichroism and/or inverse Faraday effect could indeed play a role in the switching³⁰⁻³². The down-side of a thermally activated process however, is that it will necessarily require multiple pulses and will inevitably be slow. Many groups have since focused on trying to use inverse Faraday or other coherent effects³³⁻³⁵ to impart torques on the magnetization, with the hopes of fully reversing the magnetization in a single shot. We, in Berkeley and Nancy, took a different approach, were we combined GdFeCo with a ferromagnetic layer to *extend* the AOS capabilities to the ferromagnet.

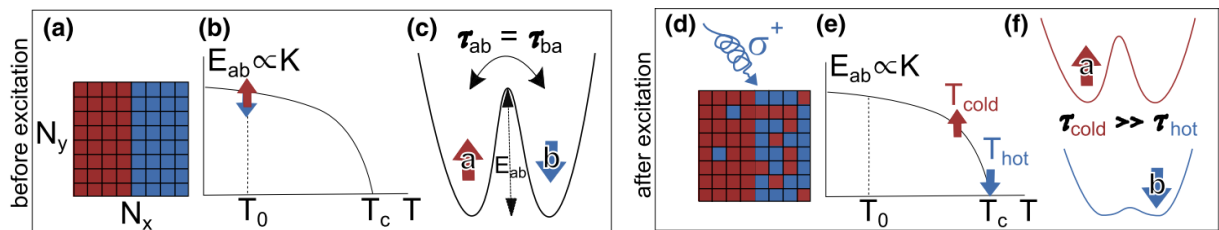


Fig. 5: Schematic of model for MCD driven AOS. (a-c) state of magnetic cells before and (d-f) after excitation with a circularly polarized laser pulse. (a,d) Shows the distribution of magnetic states. (b,c) shows the energy barriers as a function of temperature, and the temperature of up and down cells. (c,f) represent the well barrier and hopping time for non-excited (c), as well as hot and cold cells (f). Figure extracted from ref²⁸.

iii. AOS in combined ferri and ferromagnetic heterostructures

In 2016, while in Berkeley, we first studied what happened to a Co/Pt multilayer when grown on top of GdFeCo and irradiated by a femtosecond laser pulse³⁶. The magnetic layers were exchanged coupled (through the Pt layer), and after a single laser shot we could reverse both the magnetizations of the GdFeCo layer and the Co/Pt layer on top. We monitored the dynamics with a depth-sensitive MOKE technique³⁷, and showed that the reversal of the Co/Pt happened within the first ~ 7 ps. This was the fastest reversal of a ferromagnet at the time, and demonstrated that GdFeCo could be used as a driving force to induce reversal in different magnets.

A few years later, we extended this idea in Nancy³⁸ to samples with a thicker non-magnetic spacer of Cu (see left Fig. 6). The magnetic layers in this case were statically decoupled (no bias was visible in the minor hysteresis loops), so that the coupling if any could only be dynamic, i.e. appear during or after laser exposure. After a single laser shot we could again reverse the GdFeCo and/or the Co/Pt, depending on the fluence (see series of laser shots in right Fig. 6). Since the layers were decoupled we could reach 4 distinct levels, with the ferromagnetic and ferrimagnetic layers being parallel (P) or antiparallel (AP). The reversal of the Co/Pt magnetization was attributed to the spin currents leaving the GdFeCo layer³⁹ during its reversal.

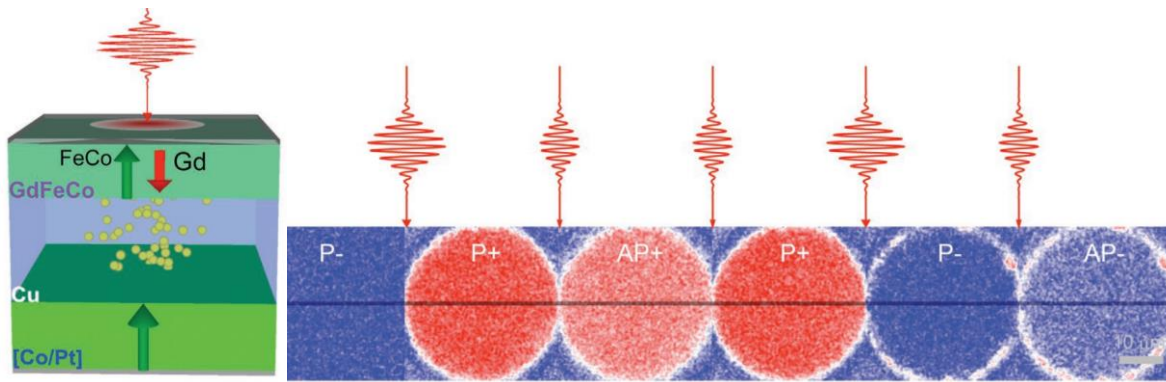


Fig. 6: (left) GdFeCo/Cu/[Co/Pt] structure indicating a polarized current in the Cu spacer (as yellow circled electrons) and (right) magnetization state observed by a MOKE microscope after a series of single laser shot. The large amplitude optical pulses switch the GdFeCo layer AND makes the Co/Pt layer parallel P ($P^- \rightarrow P^+$ or $P^- \rightarrow P^+$). The lower amplitude pulses switch only the GdFeCo layer (for ex: $P^+ \rightarrow AP^+$ or $P^- \rightarrow AP^-$). Figure extracted from ref⁴⁰.

Finally, a few more years later we realized that in some similar structures of GdFeCo/Cu/[Co/Pt] with higher Gd content, we could reverse the magnetization of the Co/Pt layer even without reversing the GdFeCo via AOS^{41,42}. In fact, we worked with GdFeCo layers that had such a composition that they should never exhibit AOS on their own (far away from their compensation temperature). However, even a mere demagnetization of GdFeCo was able to induce enough spin currents to reverse the magnetization of

Co/Pt, and surprisingly the magnetization reversed within less than a picosecond. Observing the dynamics was quite challenging here, since we used a depth-sensitive time-resolved MOKE microscope, where multiple calibration steps were needed to make sure we were really probing the right layer's magnetization. The sub-picosecond speed of the reversal is a further confirmation that an ultrafast transfer of angular momentum, due to spin currents, is taking place.

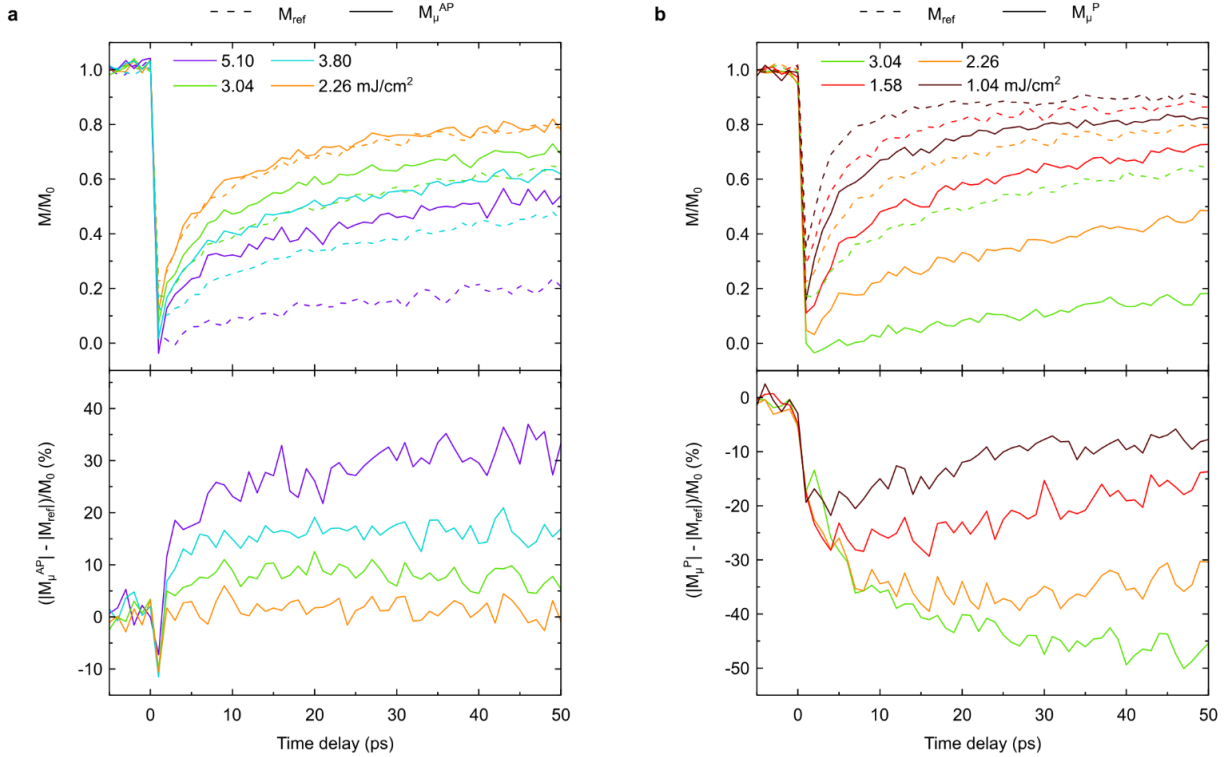


Fig. 7: The demagnetization of the GdFeCo layer (left) induces a spin current in the direction of the Gd spins that can reverse the Co/Pt layer (right). The image on the right is a magneto-optical image of the reversed domain by the laser pulse. Figures extracted from ref⁴².

Further studies⁴², allowed us to understand that such a fast and intense injection of spin into the ferromagnet, not only alters drastically its dynamics but also the effective spin temperatures⁴³. Indeed, when comparing the demagnetization of the Co/Pt layer within the heterostructure with that of a simple Co/Pt ferromagnet reference (see Fig. 7), the magnetization dynamics were noticeably different due to the presence of the spin currents (from the GdFeCo layer). When using a high fluence (5.10 mJ/cm²) on the reference sample (Co/Pt), we observed a magnetization reaching almost zero and a slow recovery of the magnetization (see purple dashed line in Fig. 7.a). Such slow-down of the dynamics is due to[‡] the diverging of the magnetic heat capacity close to its Curie temperature, resulting in a slow-down of energy transfer rates. However, when using a fluence that results in a similar demagnetization on the heterostructure in an AP state (see purple solid line in Fig. 7.a), the recovery was much faster. One could first think that the more

[‡] There are different models and ways of thinking about such critical slowdown^{80,81}

efficient cooling here is mostly due to the increase in metallic volume, since a bigger volume implies a bigger heat capacity, and if such volume absorbs little light (ex: Cu), then it would suck heat out of the Co/Pt. However, this hand wavy explanation breaks down when reversing the magnetization of the GdFeCo layer, and going into a P state. As shown in Fig. 7.b, the recovery was then slower in the heterostructure than in the reference layer. Our interpretation, supported by modelling, is that the spin density injected by the GdFeCo affects the actual spin temperature. This means that you can “reach T_c ” without going through a critical slowdown, the thermal dynamics being dominated by the angular momentum injection rate, instead of thermal energy exchanges. Once the spin injection rate goes to zero (after equilibration of out-of-equilibrium spin density and magnetization) conventional thermal dynamics take over. In the bottom part of Fig. 7, the percentage of cooling (panel a) and heating (panel b) with respect to the reference layer is calculated, which can be as large as 50%.

Additionally, this experiment hinted for the first time to the possibility of using non ferrimagnetic systems, as a possible source of spin for ultrafast reversal.

iv. AOS in fully ferromagnetic spin-valves

After seeing that demagnetization within a GdFeCo source layer could generate enough spin current to induce switching in a second ferromagnetic layer, we soon wondered if a ferromagnet would suffice as the spin current source. We have since tried⁴⁴ a number of spin-valves systems that include Co/Pt as a hard layer, a Cu spacer up to 80 nm thick and a thin Co/Pt as a free layer (see Fig. 8), structures that are very similar to the ones used during typical STT experiments⁷ (see Fig. 8.a).

Normally, for AOS within a ferrimagnet, the two magnetic sublattices need to be antiparallel (AP) to exchange opposing angular momentum and drive the reversal of each sublattice. This has been a clear and well accepted criterium for AOS in alloys and multilayers⁴⁵. We note here that an advantage of using ferromagnetic layers is that there is no possible confusion between the meanings of AP and parallel (P), in contrast to ferrimagnetic layers where P could indicate the parallel alignment of the magnetizations or that of the sublattices. With ferromagnetic layers, if one of the magnetic layers was pinned and AP to the free layer's magnetization, it would be relatively easy to imagine how the demagnetization of the pinned layer could lead to the reversal of the free layer and make it P (see Fig. 8.b). However, in our ferromagnetic structures, as we increase the laser fluence we first see a P to AP reversal of the Co/Pt layer (see Fig. 1Fig. 9.b). This is extremely surprising, since there is no obvious bath for such angular momentum in the structure that could help the Co/Pt switch. We note that if we start in an AP configuration, the sample remains in a AP state after the same excitation (see Fig. 1Fig. 9.b) which means that the pinned Co/Pt layer is able to interact with top free layer despite the 80 nm of Cu, similarly to what we have seen in previous sections. Interestingly, at higher fluences the dominant mechanism seems to change, and a "partial" AP to P switching starts taking over.

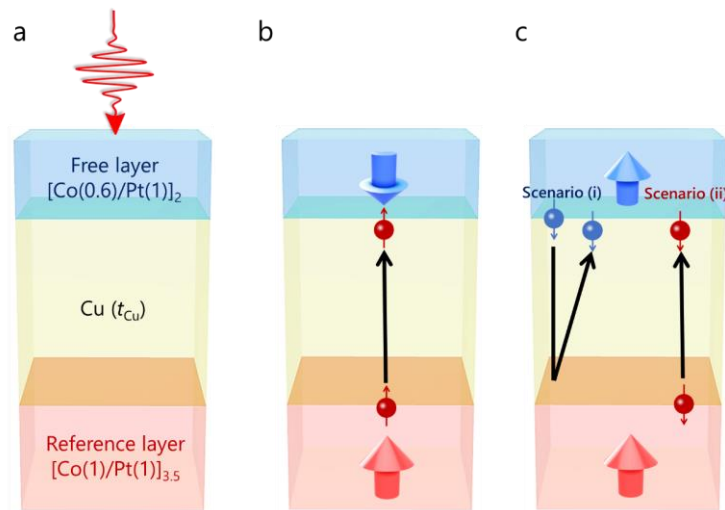


Fig. 8: Sample structure and schematic illustrations of the switching observed in this study. a) Sample stack. Values in parentheses are layer thicknesses in nm. Schematic illustration of from the process for b) AP-to-AP switching and c) P-to-AP switching based on scenario (i) and (ii) explained in the text. Blue (Red) spins are sourced from the free (reference) layer.

In our analysis, we first made sure we could safely eliminate any magnetostatic couplings as possible reasons for the switching. The use of thick Cu spacers (up to 80nm) allowed us to eliminate the exchange interaction, and the observed speed of reversal for the Co/Pt layer (less than 1 ps, see Fig. 9), among other reasons (we performed various other experimental tests⁴⁴), allowed us to eliminate the dipolar coupling as a possible mechanism (for >1 THz dynamics an unrealistic $\gg M_s$ dipolar field would be required). Once again, we could only think of some dynamic, long-range coupling mechanism such as spin currents to explain the observed reversal. The major question remained: where is the angular momentum coming from? We believe that at short time-scales it can only come from either (scenario i) the free layer itself due to a reflection mechanism, or from (scenario ii) the pinned layer (see Fig. 8.c). Eventually however, the lattice needs to supply the missing angular momentum. One major mechanism candidate for scenario ii was the possibility that after the demagnetization of the reference layer, its magnetization recovery during cool-down could result in a “suction” of angular momentum (i.e. emission of a minority spin current) from the reference layer, and induce its reversal. We therefore tested such a scenario with an s-d model based on Beens et al.⁴⁶, and could demonstrated that the remagnetization spin currents could indeed lead to a reversal, but at a longer time-scales than the observed switching. As shown in Fig. 9.a, the remagnetization of the reference layer always starts after reversal of the free layer. Moreover, we observed a reliable switching of the free layer even with the 80 nm thick Cu sample when almost no optical absorption takes place in the reference layer, which casts doubts into scenario ii where the reference layer needs to be excited. This reasoning, pushes us to lean towards a scenario i mechanism, in which we see strong parallels with STT, where P-to-AP switching is possible due to the reflection of minority spins and absorption of majority spins at the interface with the second ferromagnet (the reference layer). However, the actual origin and mechanism is still very much uncertain.

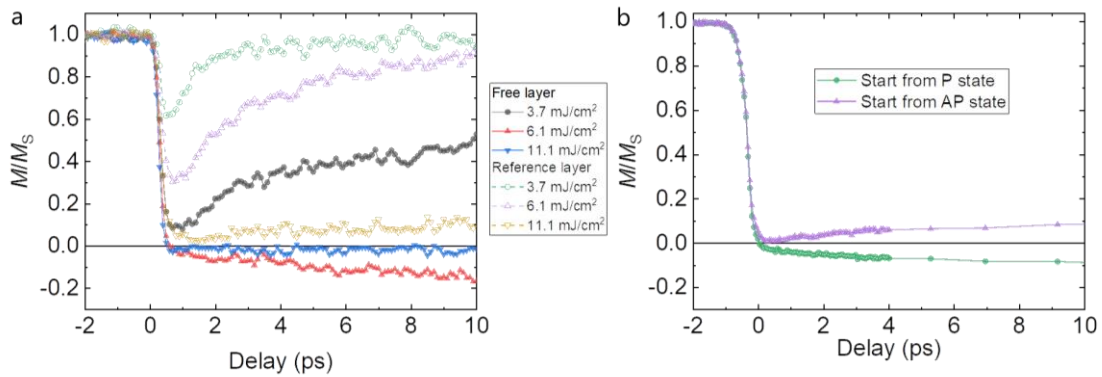


Fig. 9: Magnetization dynamics obtained from TR-MOKE. a) Magnetization dynamics for a Cu thickness of 10 nm measured for several fluences starting from a P configuration. Utilizing a quarter wave-plate enables separating the signal from the free layer and from the reference layer. An external magnetic field larger than the coercive field of the reference layer is applied during the measurement. b) Magnetization dynamics for a Cu thickness of 80 nm starting from the P+ (green) and AP+ (purple) states with $F = 10.4$ mJ/cm². An external magnetic field (H) whose magnitude lies between the coercive fields (H_C) of free layer and the reference layer (H_C of free layer $< H < H_C$ of reference layer) is applied during the measurement

Our results have been very consistent over a wide range of ferromagnetic layers (with different thicknesses, compositions...), and have been verified under many conditions (with/out magnetic fields, in single shot, in stroboscopic experiments...). More experiments and simulations are under way to pinpoint the exact mechanism and make this phenomenon more accessible. Again, we are encouraged by the analogy we find with conventional STT. We believe this work might help to bridge even further the fields of spintronics and ultrafast magnetism.

c. THz spintronics

Magnetism has been studied with gigahertz sources for many years, generally focusing on the ferromagnetic resonance modes. As discussed earlier, in order to look into faster processes, ultrafast optics has been successfully employed since 1996. However, due to the complexity of sources and detectors in the THz spectrum, little work has been performed in the intermediate THz regime. One of the first works to combine THz radiation and magnetism was actually published⁴⁷ by Beaupaire and colleagues in 2004. They realized that if the magnetization was suddenly changing in the sample, Maxwell equations predicted a radiative THz field, which they could detect. During the same period, a few others works started looking into THz radiation generated by the magnetization change in epitaxial ferromagnetic metals and semiconductors^{48,49}. From 2002 to 2013 we also saw the first works using THz to control the magnetization, either by irradiating THz on magnets and using the B field to induce precession^{50,51} or, similarly, by guiding THz current pulses with transmission lines and using the Oersted field^{52,53}. In parallel, the idea that the angular momentum loss during ultrafast demagnetization could lead to ultrafast THz spin currents started emerging^{43,54}. Finally, in 2013, the now known *spintronic emitters* were discovered by the group of Tobias Kampfrath^{55,56}. In contrast to the THz generated by the change in magnetization (related to B), these spintronic emitters use the variations in charge density (related to E), and result in a much more intense emission. In order to convert the magnetization change in an electric field change, the spin currents generated by the ultrafast demagnetization are converted to charge currents by mechanisms such as the inverse spin Hall effect⁵⁷. The arrival of such emitters, which have many interesting properties in comparison to other THz sources (tunability, low cost and ease of fabrication on arbitrary substrates, broadband excitation, large bandwidth of emission...), combined with the growing interest with antiferromagnetic materials which have natural resonances in the THz range, ignited the start of a new field in magnetism: *THz spintronics*⁵⁸.

i. THz spin heating

Around 2013 the group of Ali Shakouri published a paper⁵⁹ where they looked into fast electronic heating by picosecond (THz) current pulses. They generated a few picosecond wide pulses with photoconductive-switches[§], and performed time-resolved thermo-reflectance measurements in order to probe the fast temperature rises along the transmission line. This work partially motivated us to attempt a similar experiment with magnetic materials, in order to demonstrate THz current induced ultrafast demagnetization. During the same period, similar works were performed with free-space THz by Bonetti et al.⁶⁰ and Shalaby et al.⁶¹

We obtained our first demagnetization results⁶² in 2016 on out-of-plane magnetized Pt/Co/Pt samples using home-made photoconductive switches (see top left Fig. 10). We first confirmed that the peak demagnetization induced by the pulses were quadratic with the current, i.e. linear with the dissipated power, as expected (see top right Fig. 10). We then compared the demagnetization induced by 5.5 ps wide current pulses (~ 4 ps in power) vs 2.6 ps wide optical pulses. We noted that the dynamics were not the same: the optically induced dynamics showed an important hump, whereas the electrical ones did not (see bottom Fig. 10). In order to fit these dynamics, we used a 3 temperature model (electrons, phonons and spins) where most of the parameters were fixed from the literature or characterized separately. In practice, the spin temperature follows almost exactly the electron temperature due to the long ps duration of the pulses. One of the most critical parameters was the electron-phonon coupling constant g_{e-p} . The optical data (bottom left Fig. 10) was best fitted with $g_{e-p} \sim 7 \text{ Wm}^{-3}\text{K}^{-1}$ whereas the current/THz data (bottom right Fig. 10) required a $g_{e-p} \sim 15 \text{ Wm}^{-3}\text{K}^{-1}$, in best agreement with the value derived from scattering theory⁶³. We attributed the difference in g_{e-p} to the difference in initial distribution of the electrons. In the case of 800 nm light, the initial distribution contains electrons with ~ 1.5 eV of energy and electrons are not thermalized (do not follow a Fermi-Dirac distribution). As time goes by, electrons scatter with each other and thermalize. If the system absorbs THz photons instead, electrons are only excited by a few meV. To absorb the same amount of total energy in both optical and electrical experiments, many more photons and electrons would need to be involved when using current (x1000). Also, because this energy is lower than the thermal background ($k_B T \sim 25$ meV), the distribution of electrons would remain thermalized. Intuitively, if more electrons are excited above the Fermi level, there are more chances for electron-phonon collisions to take place, and therefore g_{e-p} should increase, in agreement with our fits

[§] A photoconductive switch uses femtosecond optical pulses to change the conductivity of a low temperature grown GaAs substrate region surrounded by biased electrodes, and releases the energy stored with picosecond-lived transient current⁸².

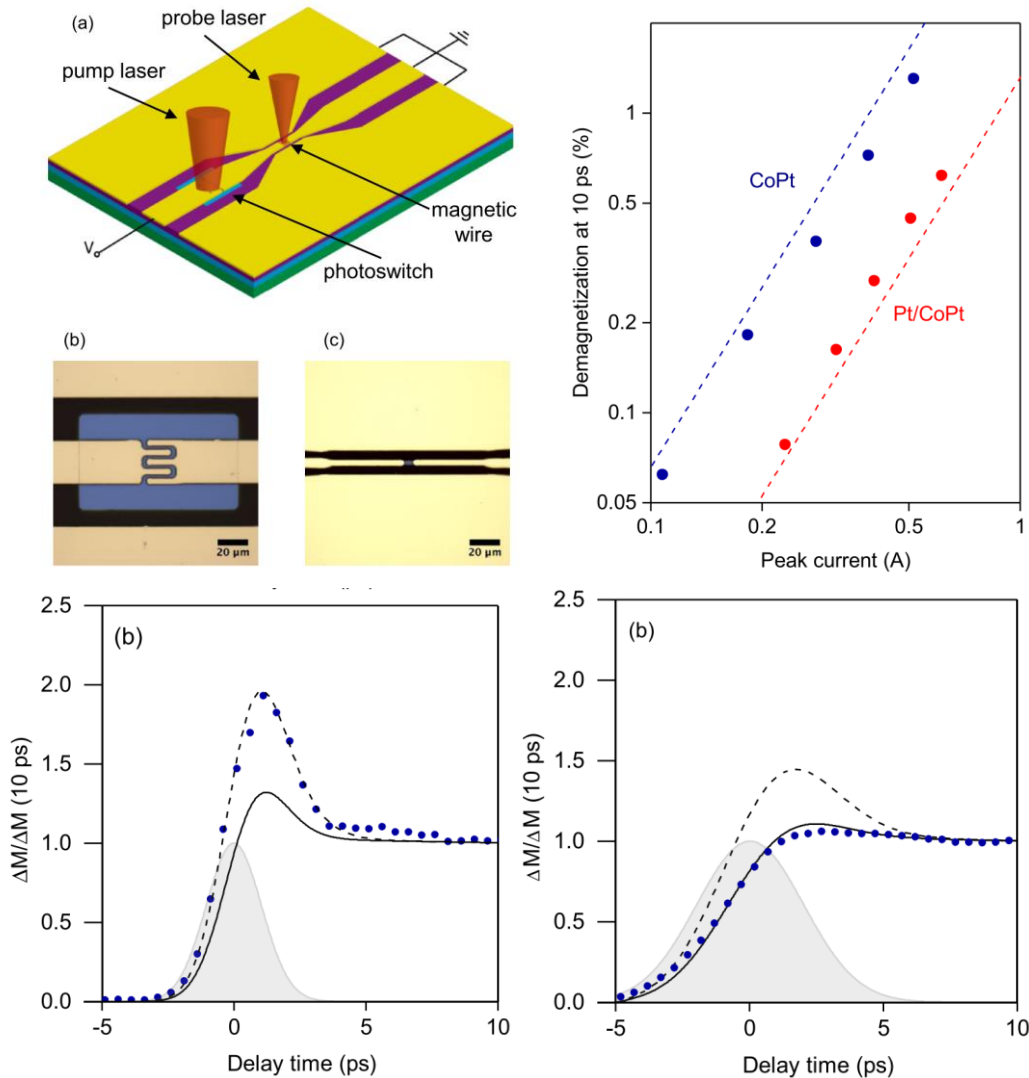


Fig. 10: Top left) a) Schematic of experiment and sample b) image of photoconductive switch c) image of magnetic device. Top right) demagnetization 10ps after arrival of electrical pulse as a function of the peak current within the pulse. Bottom left) polar MOKE response after a 2.6 ps optical pulse. Bottom right) polar MOKE response after a 5.5 ps electrical current pulse. Figures extracted from ref⁶².

Recently, the group of Kampfrath performed a similar comparison between optical and THz demagnetizations⁶⁴ in in-plane magnetized Fe films. In their work they used much shorter pulses, 500 fs for the THz pulses and 200 fs for the optical pulses, and even used a deconvolution method to push the resolution of the THz pulse up. Interestingly, the major conclusion of their analysis and modelling is that there is no major difference in the induced demagnetization dynamics due to a THz vs an optical pulse, somewhat in disagreement with our results. The reasons for this discrepancies are not totally clear at the moment. In Kampfrath's work both torques (linear in E) and heating effects (quadratic in E) are experimentally disentangled, and then the dynamics for both pump energies are plotted on top of each other, revealing the extremely similar response. This simple methodology is quite convincing. Our work on the other hand is based on the assumption that we can neglect the torques (spin-orbit torques and

Oersted fields) according to literature-based estimations, which if wrong, could somewhat change our conclusion. To this aim, further experiments where we monitor the angle of the magnetization might be required. Only changing the direction of the electric current might not be enough, since we are only measuring the projection of the magnetization along one axis, and therefore cannot distinguish a torque that would bring the magnetization towards the plane in a clockwise manner vs anti-clockwise (at least not without breaking the symmetry with some kind of in-plane field... which would introduce additional torques).

After demonstrating that picosecond current pulses could induce a strong demagnetization, we exchanged the Co/Pt film for a GdFeCo one. We first made sure the ferrimagnet's magnetization could be switched when using 10 ps wide optical pulses. We then sent a single 9 ps wide current pulse and, at a certain current threshold, we observed a full reversal of the magnetic area within the transmission line. As with the AOS, the switching was independent of the polarity of the current, toggling after each pulse (see Fig. 11). The reversal was induced by pure current induced Joule-like heating. Furthermore, the dynamics showed that the reversal (crossing of zero magnetization) took place within ~ 10 ps, making this the fastest current induced magnetization switching ever probed.

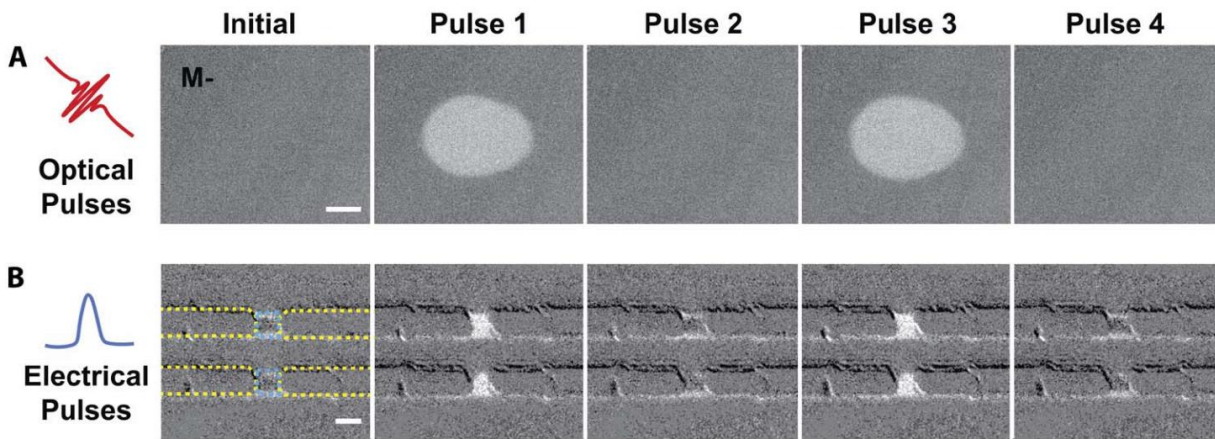


Fig. 11: Fast electronic heating induced toggle switching of the magnetization in a GdFeCo sample, using a) optical pulses and b) 9 ps wide electrical pulses. Figure extracted from ref⁶⁵.

ii. THz spin injection

As discussed earlier, one well established way of generating fast spin current transients with THz frequencies, consists in demagnetizing a magnetic layer with a femtosecond optical beam. The demagnetization, causes spin currents to leave the layer, which can then be utilized to, for example, exert a torque on a second magnetic layer^{37,66}. However, this method only allows for experiments in a particular geometry (spin currents are always perpendicular to the plane) and the generation of the spin current is a complex process that involves non-thermal distributions, ballistic electrons, heat diffusion ... making its amplitude extremely difficult to predict and quantify.

Alternatively, we can use THz pulses to induce spin transport in a more controlled manner. The THz electric field, which we can quantify, will directly induce charge currents which can subsequently be polarized. Not only does this method simplify the mechanism for spin current generation, but since the Drude model still works well at the THz frequencies, we can make comparisons with DC experiments more reliably^{67,68}, and make predictions based on low frequency measurements. Moreover, these THz pulses can be almost unipolar, allowing for a change in polarity of the spin currents.

To the best of my knowledge, one of the first and only works dealing with spin transport and THz was published⁶⁹ in 2015 by a german collaboration, including once again the group of Tobias Kampfrath. This work demonstrated that the giant magneto-resistance can still work up to the THz frequencies, and may allow us to access scattering parameters with a non-contact method. THz have since become a very useful probe of spintronic properties, particularly with the advent of spintronic emitters (see section iii).

In 2019, we started applying our own picosecond pulses to ferromagnetic structures (see Fig. 12)⁷⁰. For this experiments, we used Pt and Ta as charge-to-spin converters, exploiting the spin Hall effect. The incoming picosecond charge current pulse resulted in a picosecond wide spin current, which was injected into an out-of-plane magnetized Co magnet. The resulting spin torque is known as a spin-orbit torque, since the spin Hall effect requires a large spin-orbit to create the spin current. Previously, the shortest spin-orbit torque mediated switching reports had used ~ 200 ps wide pulses^{71,72}. Here, we wanted to use 30-50x

shorter pulses, reaching timescales much shorter than natural precession frequencies of the magnetic systems at study.

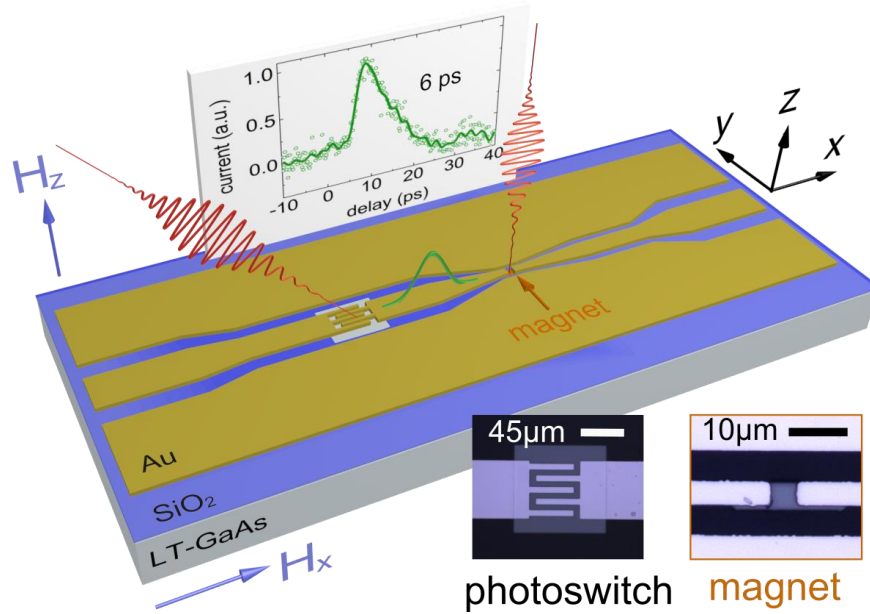


Fig. 12: Picosecond current pulse generation platform with photoswitch, coplanar waveguide and ferromagnet. The ferromagnet stack used here experiences spin-orbit torques due to the spin Hall effect. The THz pump – optical probe configuration allows the investigation the dynamics of the magnet. Figure extracted from ref⁷⁰.

Our experiments were similar to those performed on GdFeCo, consisting of a transmission line with a photoconductive switch activated by femtosecond laser pulses. However, for this experiments, an additional in-plane magnetic field was required to break the in-plane symmetry of the ferromagnet and enable spin-orbit torque switching⁷³. We performed two different types of experiments: First, we checked what a single high intensity picosecond pulse would do to the magnetization, by looking at the after-pulse magnetic state with a MOKE microscope. We could see that after a single high intensity pulse, the magnetization would switch, for a given in-plane field and current polarity, but would remain the same if either the field or current polarity was changed. The final state was also independent of the initial state. The observed outcome agreed with observations from spin-orbit torque experiments with longer microsecond-wide pulses. Secondly, to confirm the ultrafast character and presence of spin-orbit torques, we performed time-resolved MOKE experiments. For this experiments we used sub-threshold current amplitudes and observed the “kick” from the spin-orbit torques and subsequent precession and relaxation around the equilibrium position (see Fig. 13). Depending on the initial out-of-plane magnetization direction, applied in-plane field or current polarity, the dynamics followed different dynamics. We were able to reproduce most features of these dynamics using a macrospin model coupled to a single-bath thermal diffusion model, and temperature dependent magnetizations and anisotropies. From these simulations we could also estimate the ratio of damping-like / field-like torques. The most interesting

feature of these dynamics is the observation of simultaneous torques and demagnetization, which are normally not present in quasi-static experiments. Moreover, the demagnetization, also induces new torques by changing the equilibrium field position (the anisotropy gets lowered by the ultrafast heating, which triggers precessional dynamics).

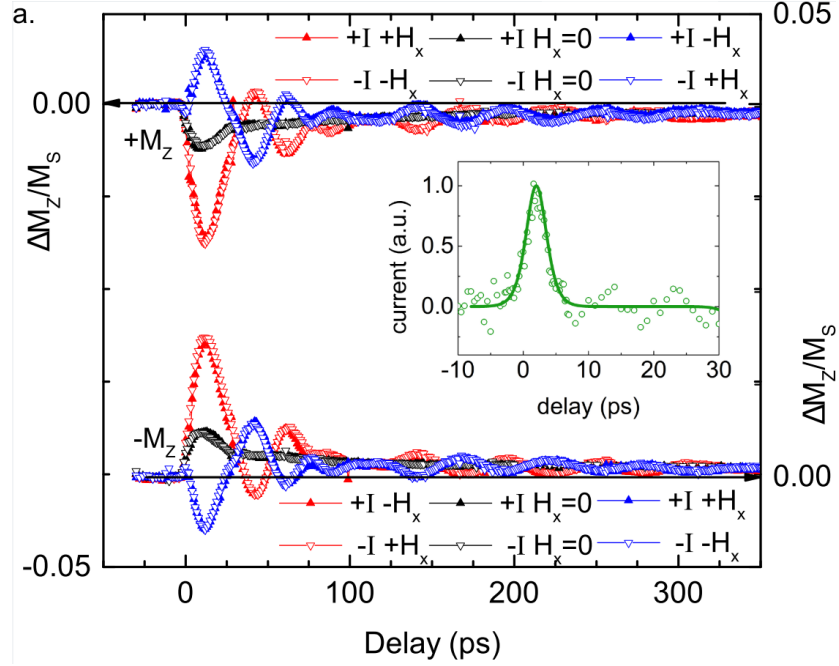


Fig. 13: Normalized change in the out-of-plane component of the magnetization $\Delta M_z/M_s$ (probed via time resolved polar MOKE) after excitation by a 3.8 ps wide electrical current pulse (shown in the inset). Current and in-plane field polarities are shown in the labels. Figure extracted from ref⁷⁰.

More recently, we were able to probe the dynamics during reversal after a ps current pulse in the same samples (see Fig. 14), in collaboration with the group of Jeff Bokor in Berkeley. The dynamics show that the reversal (zero-crossing) happens in about 60 ps, way after the action of the picosecond pulse. However, this is not entirely surprising: if the magnetization is “kicked” out of equilibrium (via a short burst of SOT and a change in the equilibrium field due to heating) we expect a precessional swing around the equilibrium field, which can induce a switching on a timescale comparable with the FMR⁷⁴. So far, a simple macrospin model which includes temperature changes of M and the anisotropy seems to confirm that the magnetization switches coherently (see dashed lines in Fig. 14). Moreover, given the large dimensions of the magnetic film ($5 \times 4 \text{ } \mu\text{m}^2$) and short picosecond pulse, the domain walls velocities would need to be in the range of $\sim 25 \text{ km/h}$, for a classical nucleation + domain wall propagation scenario to be possible. Further studies will attempt to detect in-plane dynamics of the magnetization to confirm the proposed coherent scenario. Also, we will attempt to speed up the reversal by increasing the SOT intensity, and/or changing

the spin Hall material and/or the magnetic material. Finally, a full characterization of the ps pulses, in amplitude, and about all the propagation characteristics (attenuation, dispersion, reflections) is underway, in order to get to know the exact power delivered to the sample.

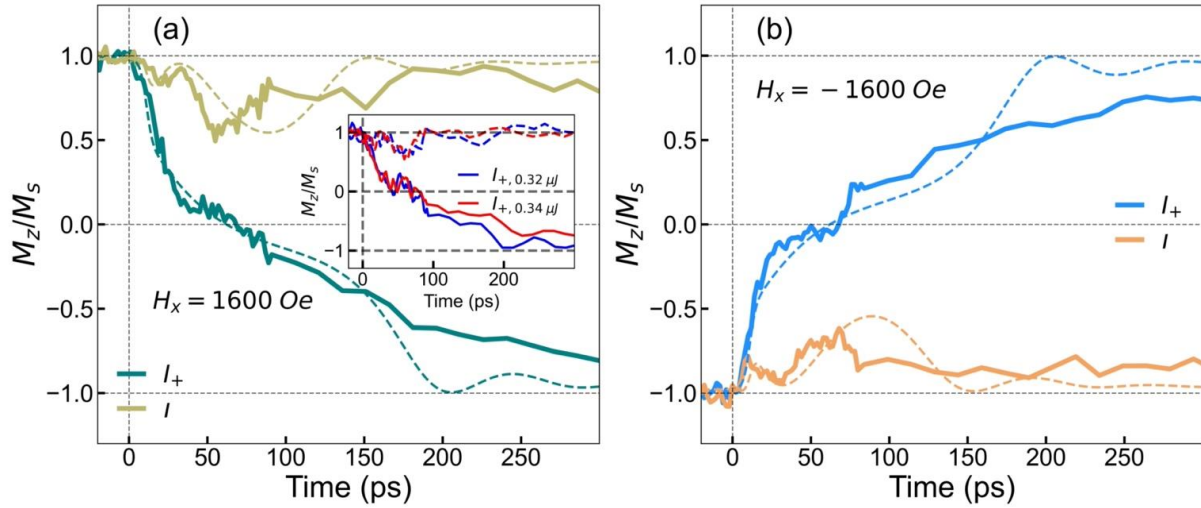


Fig. 14: Evolution of the out-of-plane component of the magnetization of a Pt/Co/Cu/Ta film after injection of a ~ 6 ps current pulse. A) starting with a magnetization pointing up, and for two current polarities b) starting with a magnetization pointing down and for two current polarities. The in-plane magnetic field is also flipped between these experiments.

iii. Spintronic THz emission

Recently, we built a THz time-domain spectroscopy setup allowing us to measure the emitted THz of a sample. As explained earlier, magnetic samples can emit THz when excited with an ultrafast laser due to various mechanisms. Analyzing the emitted THz of different series of samples and in different configurations (changing magnetic fields and/or excitation side) allows us to learn more about such mechanisms. Moreover, the technique requires no patterning, the experiments are completely non-contact, which allows for high-throughput measurements (measurements of many samples fast). For example, we have recently started exploring spin-to-charge conversion mechanisms in crystalline samples by using this THz detection setup. In fact, it is much easier to study the emission as a function of the sample angle here (simple sample rotation and/or magnetic field rotation is enough) than with classical transport measurements where multiple devices need to be patterned at different crystallographic angles.

With this new setup we recently attempted to answer the question of how far photo-excited spin currents can go in Cu. Indeed, when looking into the literature, some works extract extremely short spin current attenuation lengths on the order of a few nanometers^{37,75,76}, whereas some other works (including our recent works on spin-valves) report significant spin transport over 10s or 100s of nanometers^{42,66}. We remind here that the equilibrium spin diffusion length in Cu is typically above > 50 nm, closer to several hundred nanometers.

In order to attempt to explore this question we wanted to use THz emission experiments, where the Cu layer would separate the spin current source (a Co layer) from the spin-to-charge converter layer (a Pt layer.). However, we soon realized that adding Cu would drastically change the THz emission, as already reported and explained by Seifert et al.⁵⁶. We therefore designed two sets of samples, one with a Cu layer of variable thickness in between the Co and Pt layers, and one with the Cu on top of the bilayer (see Fig. 15.a). We expected that between these sets of samples, THz optical characteristics would remain similar, unveiling any changes in spin transport between the two sets.

Emission data is shown in Fig. 15.b for different thicknesses and the two types of samples. We first notice that the emitted THz in both sets of samples is very similar and is drastically reduced by the increase in Cu thickness. Both types of samples show very small differences even for a thick 16 nm Cu layer (see inset in Fig. 15.b). We performed some transfer matrix method calculations with an embedded THz source term to evaluate the emitted THz in the different structures, and were able to fit all of the peak emitted intensities with a single set of optical THz parameters taken from the literature (see Fig. 15.c). We also added an exponential decay term as a function of Cu thickness to fit the Co/Cu(t)/Pt emission data, in order to include spin transport attenuation. In order to properly fit the data we had to set this spin current attenuation length parameter (λ) to around 50 nm, while a sub-10 nm value gave clearly wrong results.

However, it may also be the case that this small difference between the two types of samples is related to some other subtler variations of, for example, the multilayers' THz response or the spin current amplitude.

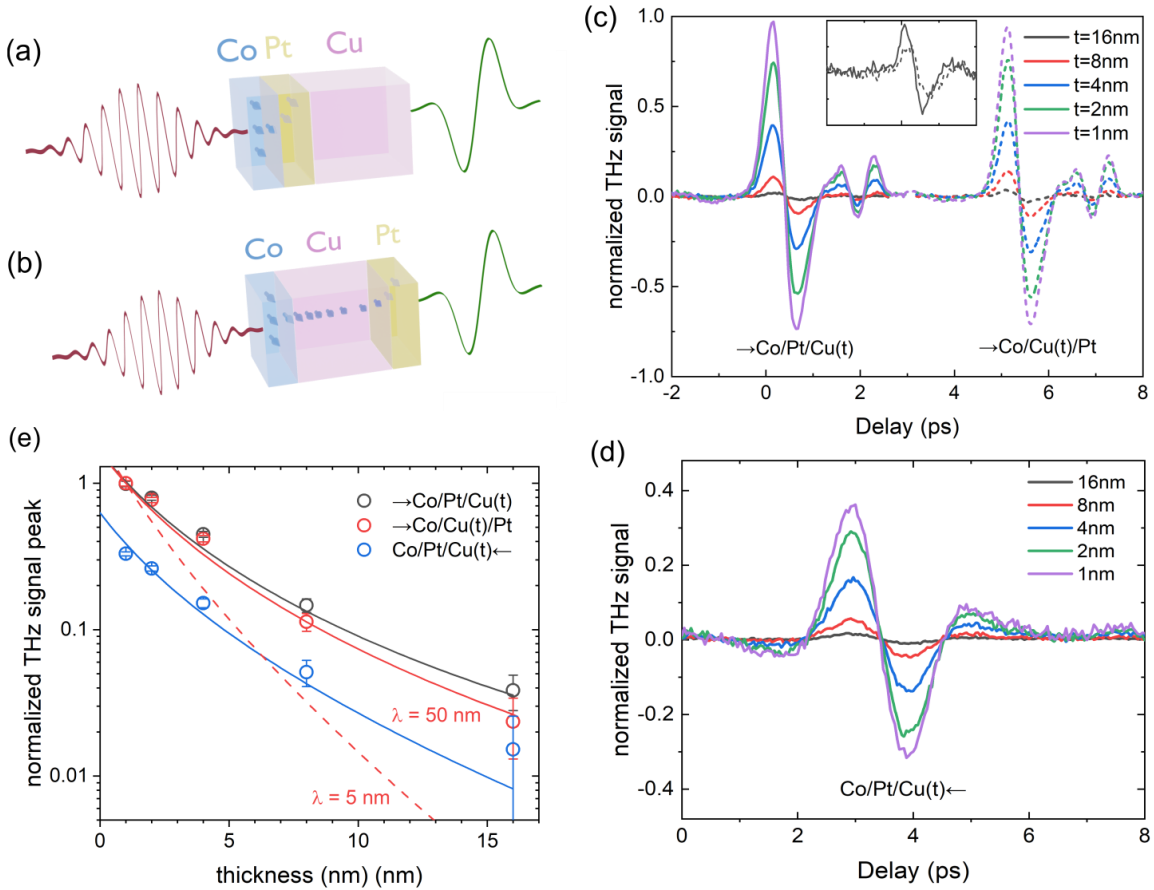


Fig. 15: Schematics of (a) Co/Pt/Cu(t) and (b) Co/Cu(t)/Pt structures with different Cu thicknesses (t), that result in (c-d) THz emission. In (c) the samples are excited through the glass substrate and in (d) samples are excited through the top side (THz emission travels through the glass substrate and gets attenuated and broadened) (e) Peak THz emission as a function of Cu thickness, and model predictions assuming long (50 nm) or short (5 nm) attenuation lengths. Extracted from Ref⁷⁷.

In conclusion, this simple but careful study allowed us to a) confirm that spin currents can travel far longer distances than 10 nm in Cu and b) highlights some of the challenges in using this technique to extract spin transport properties.

d. Ongoing / Future work

Better understanding of spin currents:

One major objective we strive for, is to better characterize and understand the spin currents generated by optical excitation of our heterostructures. We will attempt to develop / combine various methods for spin current characterization from the literature in our lab: THz emission, TR-MOKE of demagnetization, TR-MOKE of spin accumulation⁶⁶, TR-MOKE of spin-torques³⁷. With these methods, we will try to understand the origin of the reversal observed in the parallel ferromagnetic spin-valves, among other things.

Expanding possibilities of Auston switches:

Currently, and in the near future, I would like to continue to grow our THz platform so that it allows us to inject **well controlled** THz current and/or spin current pulses by using Auston switches **into any kind of system**. To this aim, we have already developed devices that allow us to reduce all parasitic reflections, that allow us to combine GHz and THz pulses, we should soon be able to characterize the amplitude of the current pulses precisely, and we are developing methods to situate Auston switches on any target substrate/sample. A future goal is to increase the bandwidth up to $\sim 4-5$ THz, the physical limit for LT-GaAs. The goal, is that we can then collaborate with various groups, and help them to work with Auston switches for their projects. We will be able to fabricate and characterize the devices with switches for them, guide them in their use, and therefore get to work with many different and exotic systems.

With this platform, we will also continue to carefully study the reversal of magnets via spin-transfer torque and/or spin-orbit torque in the picosecond timescales. We will attempt to understand the differences with the ns regime, the stochasticity, coherence of the rotation and carefully separate thermal from torque effects. Subtler but interesting phenomena such as inertia or nutation will also be sought for.

Finally, we would like to start working with antiferromagnets, where sub-THz resonant modes are naturally found. The challenges of course are numerous: magnetic state setting (and resetting), magnetic state probing, sub-room temperature Néel temperatures are common... We will start working on these challenges together with our colleagues in Saclay in the realm of the PEPR-SPIN program.

e. **Active collaborations**

Role of magnons during ultrafast demagnetization with P. Pirro (Kaiserslautern University)

Ultrafast thermal transport with R.B. Wilson (University of California Riverside)

Picosecond spintronics with J. Bokor (UC Berkeley), R.B. Wilson (UC Riverside) & Aristide Lemaître (C2N, Orsay)

Detection of inertia in ferromagnets with J.E. Wegrowe (ENS)

THz antiferromagnetism with J.Y. Chauleau, M. Viret (SPEC/CEA), R. Lebrun (CNRS/Thales) ...

5. Experimental developments

During my career I have always been very interested in setup development. I enjoy understanding the in and outs of every setup I use, and always try to improve them somehow. Since my arrival to IJL, I have developed a number of unique experimental setups. I also help maintain some of the optic equipment in the group, and in particular the ultrafast optics labs.

Optical and THz setups:

The *picoSOT* setup is a multi-purpose setup I have built, and improved over the years. This setup combines a MOKE microscope with a femtosecond pump-probe setup, all while enabling electrical access and magnetic field application. We can use the setup either for all-optical experiments or for intense THz pumping of samples in combination with an optical readout. The name of the setup refers to the possibility of studying Spin-Orbit Torques with picosecond-wide electrical pulses.

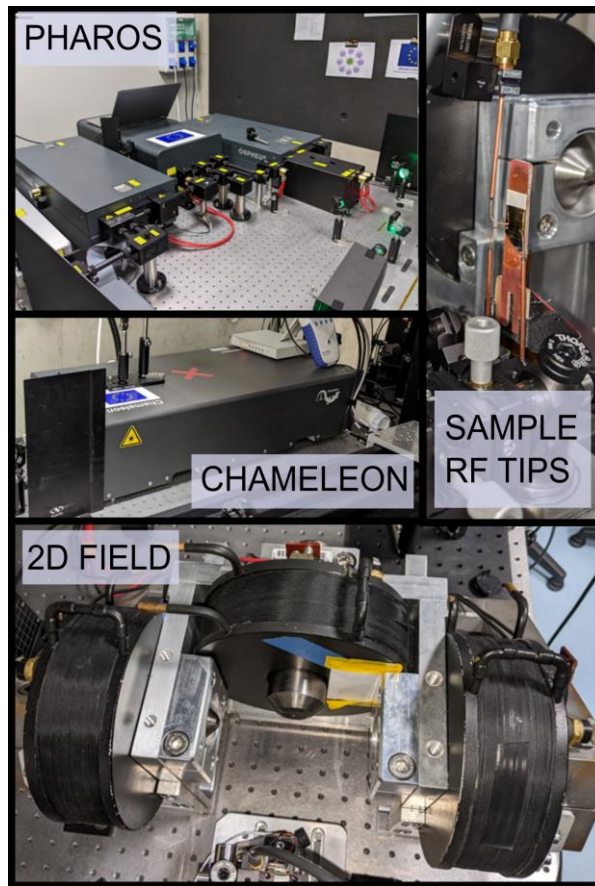


Fig. 16: Two available laser systems, sample holder with high-frequency electrical probe tips and 2D electromagnet.

In this setup we are using two different laser systems: A PHAROS system from Light Conversion and a CHAMELEON system from Coherent. The PHAROS is a Yb amplified system, with single-shot to 100kHz repetition rates, 20 W output, and ~ 300 fs pulses at 1035 nm. It is setup so that 90% of the power is diverted into a pair of optical parametric amplifiers, that enable to a) shorten the pulse duration down to 40 fs, and b) cover the 400nm to 2600 nm wavelength ranges. The CHAMELEON system, is a Ti:Sa oscillator, working at 80MHz, 2.6W and 75 fs at 800 nm. On the one hand, the amplified system enables high fluence experiments, which allow for important temperature excursions on the sample (up to the damage point). The low power oscillator on the other hand is used for small excitations, of only a few K maximum, but with a much better signal-to-noise ratio, enabling the detection of a fraction of a percent of demagnetizations.

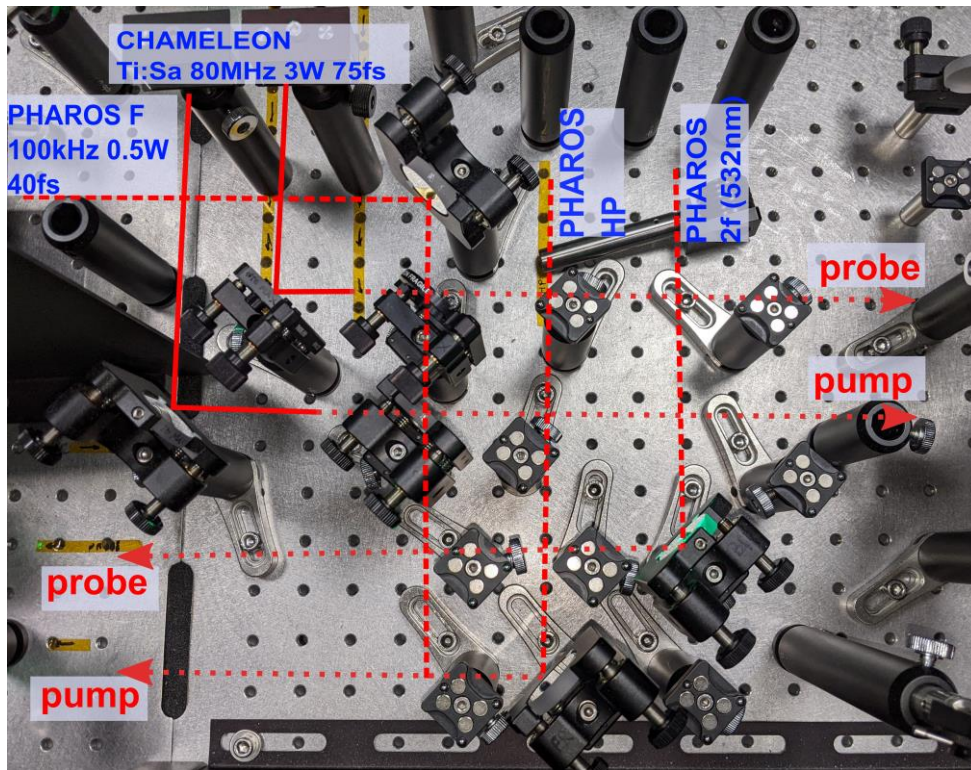


Fig. 17: Beam array configuration. The various kinematic mirrors allow to select any beam as a probe or pump for any of the two setups (right top, bottom left). Both PHAROS and CHAMELEON lasers are fed into the matrix.

Both laser systems can be rooted to two different setups (a time-resolved MOKE microscope to the left and the setup presented in this section to the right in Fig. 17). Pumps and probes can be selected among the various available beams by using an array of kinematic mirrors (see Fig. 17).

We use a home-built MOKE microscope in the Köhler configuration (for homogeneous illumination) with a 633 nm LED. We typically work with a single $f=10$ cm lens, a 10x or a 50x objective (the latter with an numerical aperture of 0.47), so we reach, at best, a $\lambda/2NA \sim 0.7$ μm resolution. We are often changing the lens to switch between the high resolution and a wider field of view for alignment purposes or magnetic domain observation. Finally, we use a beamsplitter to bring in the laser probe beam through the microscope objective and focus the laser spot down to around ~ 1 μm size.

The sample is on a XYZ motorized stage, with 10 nm resolution, includes a Hall probe with XYZ field detection, and can be excited both from the front and/or back.

The sample platform also holds three RF electrical GSG (50 Ohm) probes, rated up to 20 GHz, for either DC or RF measurements. These tips move with the sample, when the stage is activated.

Surrounding the sample, we have a 2 axis electromagnet, for out-of-plane and/or in-plane magnetic field, up to ~ 100 mT (OOP) and ~ 200 mT (IP).

A few other special features of the setup are the following: an extremely long delay line (up to 13 ns), the possibility to sync external electrical sources with the laser (with ~ 50 ps jitter) for combined optical / electrical experiments, and the use of an electro-optic modulator for lowering the noise down (almost) to the shot-noise limit of the laser.

In addition, as discussed earlier, we have recently built a *TD-THz spectroscopy setup* for detection of THz radiation emitted from spintronic structures. Some other setups are currently in the works: A low temperature time-resolved + THz detection setup, a NV-center + femtosecond laser setup...

Labview programs for all optics experiments:

All of the optical setups in the lab are using Labview codes developed by me. The structure of the code is supposed to allow for growth. There is a main window (see Fig. 18) that allows to store experimental parameters, the name of the person working on the bench, the date and time of the experiment and a universal data counter. It also logs a line in a datalog with each new data-set taken. This program then allows a user to start many instruments. Each instrument has its own window (see Fig. 19), and runs continuously, enabling real-time feedback and/or control. *Pointers* to each of the outputs of each instrument are shared across the programs so that any program can access any instrument output at any time. The main program is also used to start *programs* (jobs, routines, loops), that command instruments and log data into text or image files. Currently 4 computers use a version of this program. See below for some screenshots.

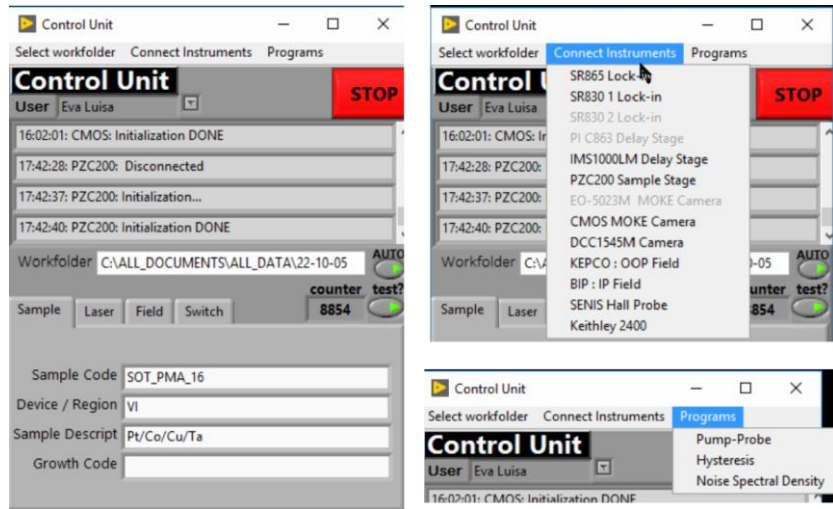


Fig. 18: Labview Control Unit used in the optics labs by all PCs. It contains a message list, a single counter identifier for experiments, some logging fields (user, samples, laser type and conditions...), which are all stored in a digital logbook. This interface also gives access to open access to instruments and programs (jobs or loops), see right panels.

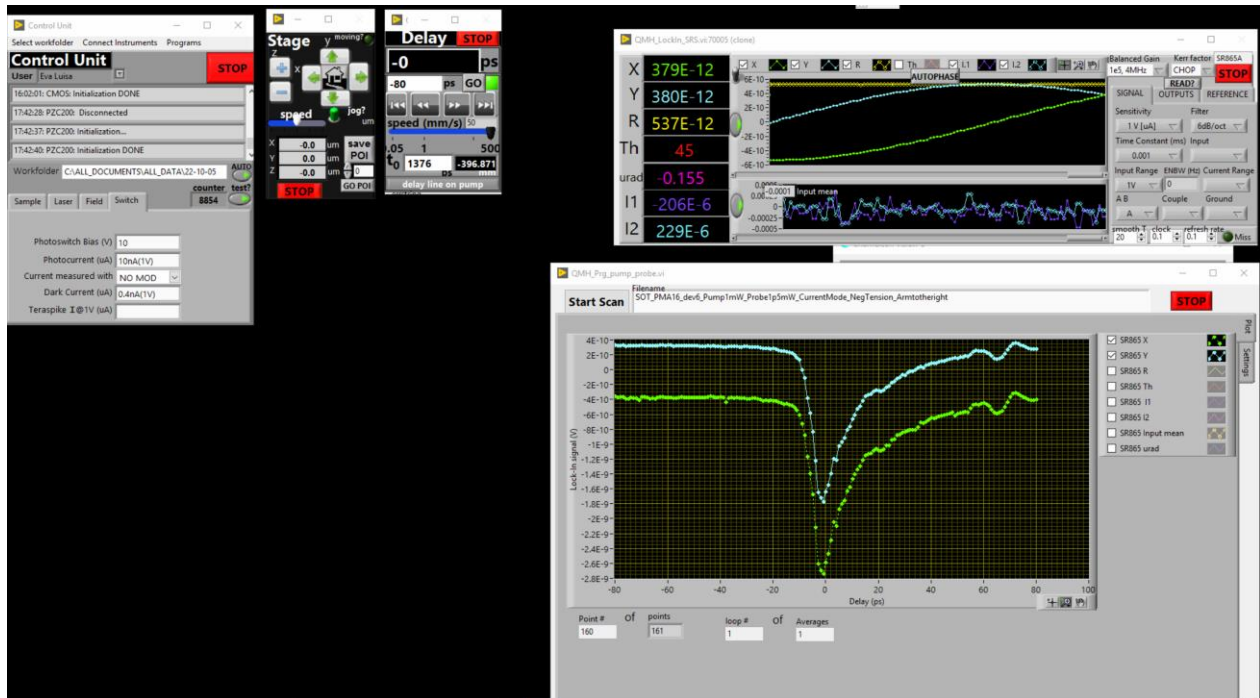


Fig. 19: Labview typical interface, with various windows opened that enable control of the sample stage, delay line and lock-in, as well as a pump-probe program that communicates with the delay line and lock-in.

6. Mentoring of students

After more than 8 years as a student, about 3 years as a postdoc and 5 years as a permanent researcher, I still consider I do not have enough mentoring experience. I believe that student mentoring is still widely considered a “natural” thing one needs to master with time, which is why teachings are limited (two two-day courses are available at Lorraine University). This is somewhat unfortunate, because if one is not very aware of their own quality of mentorship, I do not think one can improve all that much. The problem also, I believe, is that as researchers we have more and more reports, proposals and different administrative obligations to work on which reduce our time with students and their projects, reducing altogether the quality of our mentoring.

In the following paragraphs I will explain some of my experience with students and a few of the key points that I’ve learned, that I find important for my future advising.

PhD mentoring experience in the US:

During my three years as a postdoc in the US, I worked together with two PhD students and a few interns. We would meet our common advisor, Jeff Bokor, once a week to discuss our progress, but during the rest of the week I would unofficially mentor the students.

The first student was Akshay Pattabi, a first year grad student who had just arrived from India. I followed Akshay during his first three years, and he then came for a month to our IJL lab as an exchange student. He successfully graduated a few years back now. Akshay was initially in charge of magnetic second harmonic generation (SHG) measurements. The setup used an amplified Ti:Sa laser beam to induce MSHG at the surface of materials where the inversion symmetry was broken, and coupled with a set of polarizers, it could detect magnetization. This method is also sometimes known as non-linear MOKE. I helped Akshay to build and characterize the setup, and with it we were able to detect spin accumulation in normal metals such as Pt⁷⁸. A follow-up project consisted in detecting the influence of an optical pump on such accumulation, but these efforts were never successful (the samples got damaged before we could detect any noticeable signals). We then worked on some of my own projects together, such as the all-optical control of ferromagnetic layers via exchange coupling them to GdFeCo films, where he was quite invested.

The second student I worked with was Yang Yang, a Chinese student who was in his third year when I joined the group. He had started his PhD working in magnetic MEMS, but due to numerous fabrication challenges, the project fell down. He then started building up the time-resolved MOKE setup, without prior optical knowledge. When I arrived, I first guided him into building a MOKE microscope and with it, we started taking some micrographs of magnetic domains generated by AOS. From there, together with Richard Wilson and Charles-Henri Lambert, we all embarked on many of the projects I presented in this manuscript.

Working with Akshay and Yang, as a colleague and friend, was very useful to learn about how to guide young researchers, and to learn about the sometimes difficult separation between work and personal life. We complemented well each other which allowed us to often split the work, and having short term goals helped us being efficient in our work. Clearly, I found that having a small group of people working on the same subject was extremely beneficial.

PhD mentoring experience in France:

Back in France, as a permanent researcher, the relationship with my students has drastically changed. When I joined the group, I was still only a few years older than some of the PhD candidates in the group, and enjoyed sharing leisure time with them, but now had the responsibilities of an advisor, a boss. This has been a challenge, a learning experience, but also a rewarding part of my work. Training students through experiments, the literature, the data analysis and presentation methods, has been long, particularly because there is no one else in the group who can do it or help me in some of these tasks, particularly the most technical ones concerning the setup. In the future, I hope that students can teach each other, train each other, at least partially, once I have a big enough sub-group working on similar topics. So far, with only one student at a time (+ covid) it has been a real challenge to really kick-start many of my projects.

My first PhD student was Kaushalya. She was officially advised by Michel Hehn, and co-advised by me, and worked on the picosecond SOT experiments. We developed the new setup together, and she took most of the data for the experiments. Then, due to COVID and thesis writing, we only got some extra preliminary data that we still need to treat and publish. But her PhD was considered a big success by her peers, due to the Nature Electronics publication and exciting experiments she was involved in. She got a postdoc position in the Lawrence Berkeley National Laboratory to work on Si q-bits.

Now, my current PhD student is Eva Diaz, from Chile. During her first year we have improved the experimental setup, and are performing picosecond current injection experiments in a number of different systems. We are also trying to better characterize and understand our systems, pulses and induced-dynamics.

Additionally, I've mentored a few postdocs, which were hired by Stéphane Mangin for short periods of time (3-5 months each time).

All this experience has proven me that I can manage a small team, guide a few students, and keep doing good science. I believe that having a few students working on the same topics will help, as it will create a sort of "team spirit", and hopefully increase the efficiency of all of us. I am aware of the possible problems about authorship this strategy can create, and will of course sufficiently distinguish topics between students, and make very clear the ownerships from very early on.

Another important aspect I can see affecting students is the stress induced by a PhD, which is getting worse every day. The stress is mostly related to the difficult prospects of academia and pressures for obtaining good publications, but not only. This stress is particularly enhanced for foreign students that need to a) adapt to a new country's culture b) live far from home and c) navigate through all of the immigration and French speaking administrative hick-ups.

I obviously cannot summarize all of the challenges I may/will encounter as an advisor, but believed it was important to lay out some of my observations and sensibilities about the topic for the HDR diploma. Moreover, I personally care a lot about being a good mentor to students and hope I can learn from my mistakes and keep improving over all my career.

7. Responsibilities and other works

Lab responsibilities: At IJL I have been a part of a few work groups that attempt to increase the visibility of the lab: The international relations group and the lab website group. In addition, I am co-responsible of two ultrafast optic rooms, and in charge of two femtosecond laser systems. I also impart a laser-safety class for non-french speaking students.

Expertise / reviewing work: In the past years I have worked as a reviewer for various international peer-reviewed journals and European projects (Marie Skłodowska-Curie grant proposals).

Education: In September 2022 I will be in charge of part of the European School of Magnetism's practical activities.

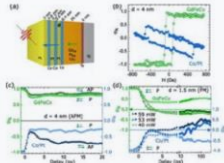
Conference organization: I have participated to the organization of a few conferences: The Tohoku-Lorraine-Mainz workshop and the Ultrafast Magnetism Conference 2022.

Science dissemination: Together with scientific publication, I have written/edited/shared media articles to disseminate my works and reach a wider audience. I have, for example, written/edited a few articles published by the CNRS (ex: <https://www.inp.cnrs.fr/fr/cnrsinfo/accelerer-la-vitesse-decriture-sur-memoire-magnetique>, see bottom of Fig. 20), written a blog article for the Nature Engineering Community (see <https://engineeringcommunity.nature.com/posts/exploring-pico-spintronics>) and edited PR articles later shared by online aggregators and/or media outlets (such as Phys.org, see center of Fig. 20). Some of my articles have been highlighted by, being covered by a News & Views article⁷⁹ or leading to a large interest (reaching the top viewed and cited articles in spintronics from APL, see top of Fig. 20).

Top Articles in Magnetism and Spintronics

By utilizing electron spins to process and store information, magnetism and spintronics research is fundamentally transforming computation and information technology. *Applied Physics Letters* reports the latest innovations, both theoretical and experimental, in magnetic and spintronic materials, interfaces, devices, and more. With over **22,000 views** and nearly **280 citations**, the articles below are among the most viewed and most cited magnetism and spintronics research in *Applied Physics Letters*.

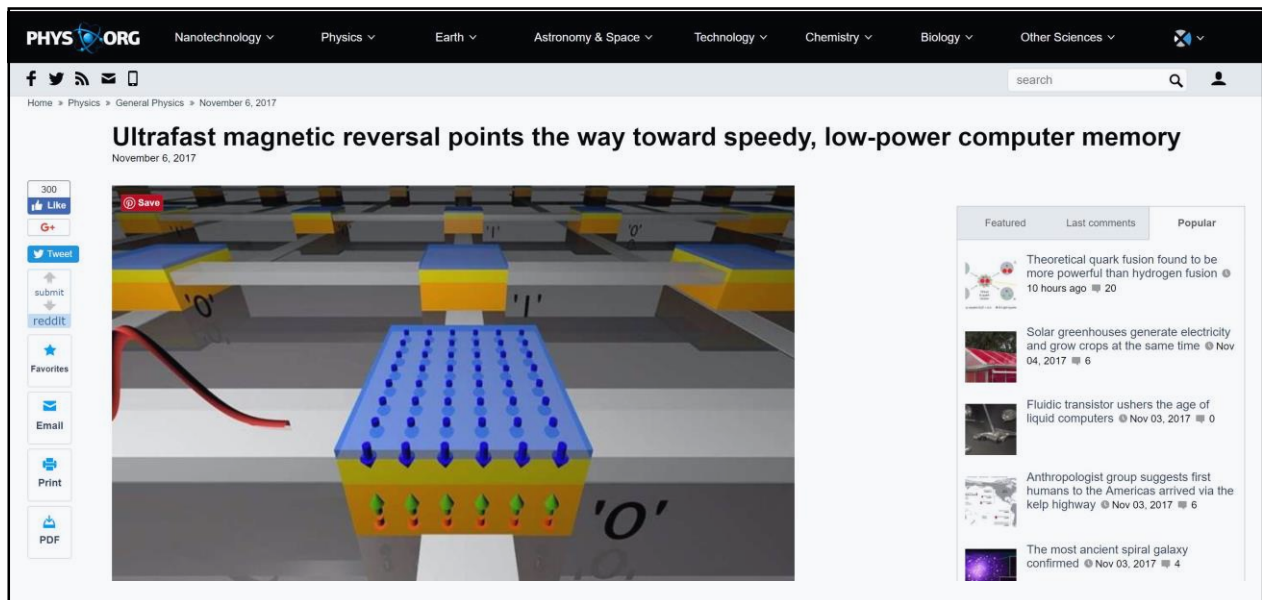
Does your research belong among the top magnetism and spintronics research published in APL?



Single shot ultrafast all optical magnetization switching of ferromagnetic Co/Pt multilayers

Jon Gorchon, Charles-Henri Lambert, Yang Yang, Akshay Pattabi, *et al.*

[Read More](#)

The screenshot shows the Phys.org website interface. At the top, there is a navigation bar with categories like Nanotechnology, Physics, Earth, Astronomy & Space, Technology, Chemistry, Biology, and Other Sciences. Below the navigation bar, there is a search bar and a user profile icon. The main content area features an article titled "Ultrafast magnetic reversal points the way toward speedy, low-power computer memory" dated November 6, 2017. The article includes a 3D visualization of a magnetic memory structure with blue and yellow layers and red arrows indicating the direction of magnetization reversal. To the left of the article, there are social media sharing options (Like, G+, Tweet, submit, reddit) and a Favorites section. To the right, there is a "Featured" section with a list of other articles, including "Theoretical quark fusion found to be more powerful than hydrogen fusion", "Solar greenhouses generate electricity and grow crops at the same time", "Fluidic transistor ushers the age of liquid computers", "Anthropologist group suggests first humans to the Americas arrived via the kelp highway", and "The most ancient spiral galaxy confirmed".

Accueil > Actualités

Accélérer la vitesse d'écriture sur mémoire magnétique

18 décembre 2020

RÉSULTAT SCIENTIFIQUE

Fig. 20: Examples of science dissemination and impact of our work.

Team responsibilities: Since beginning 2020 I have created and maintained the team’s website: www.spin.ijl.cnrs.fr I have personally taken care of all the administrative and technical aspects: authorizations, legal notices, hosting and domain name, web architecture, design, photography, analytics, calendar, project page creation, job postings, news postings... Since the publication of the website we have in average 300 new users per month visiting the website (see Fig. 21), and receive messages for the job applications that we post. Moreover, we host information useful for our group members (almost 50 in average, with lots of regular changes), such as upcoming seminars, links of interest & project details. Below, you’ll find some screenshots of the website (Figs 13-15).

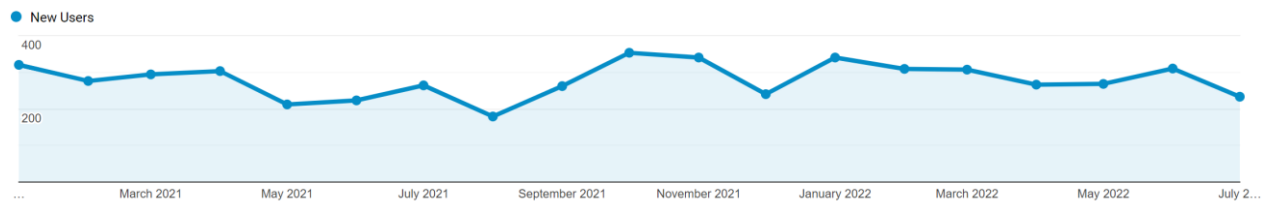


Fig. 21: Average number of users per month from January 2021 to July 2022.

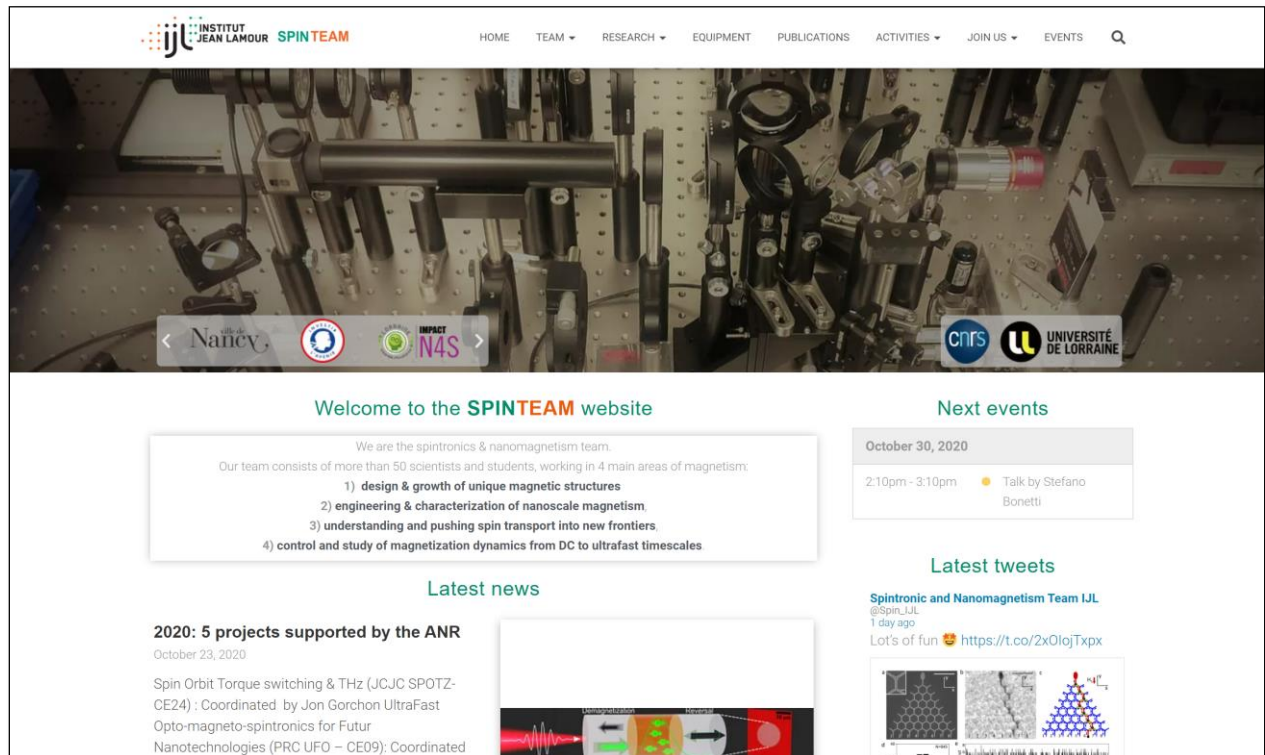


Fig. 22 Home page with sponsors, team presentation, news, twitter feed, calendar...

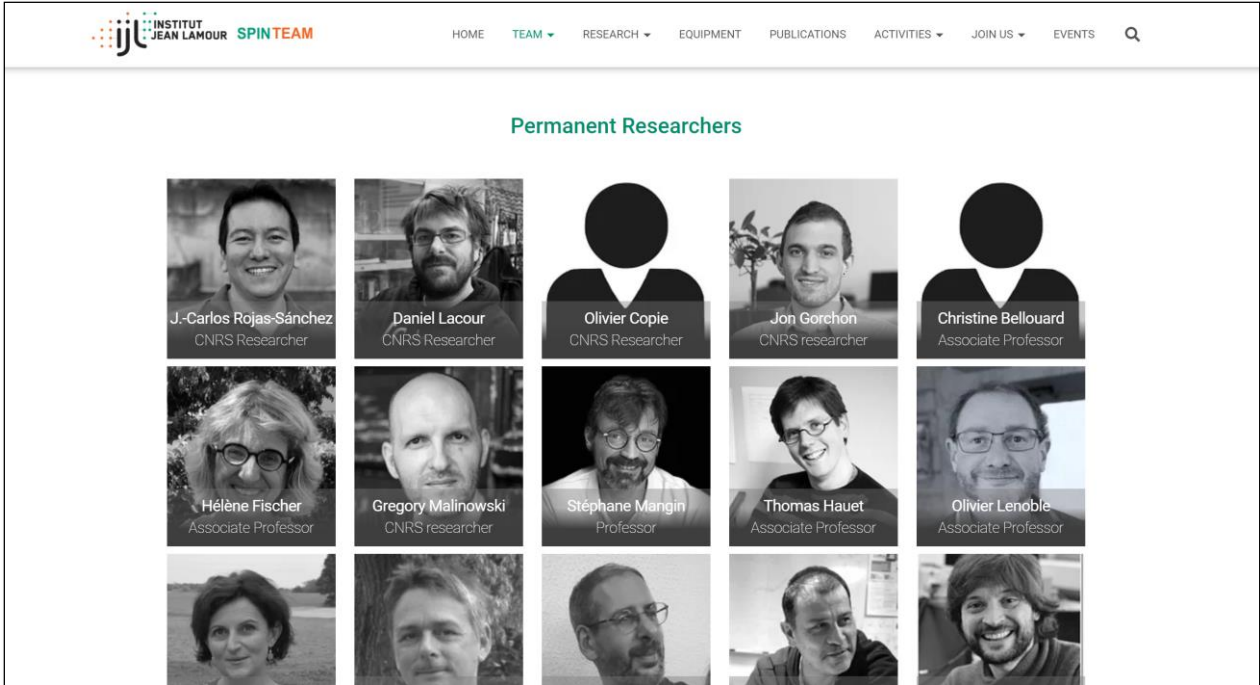


Fig. 23: Team page, permanent researcher section.

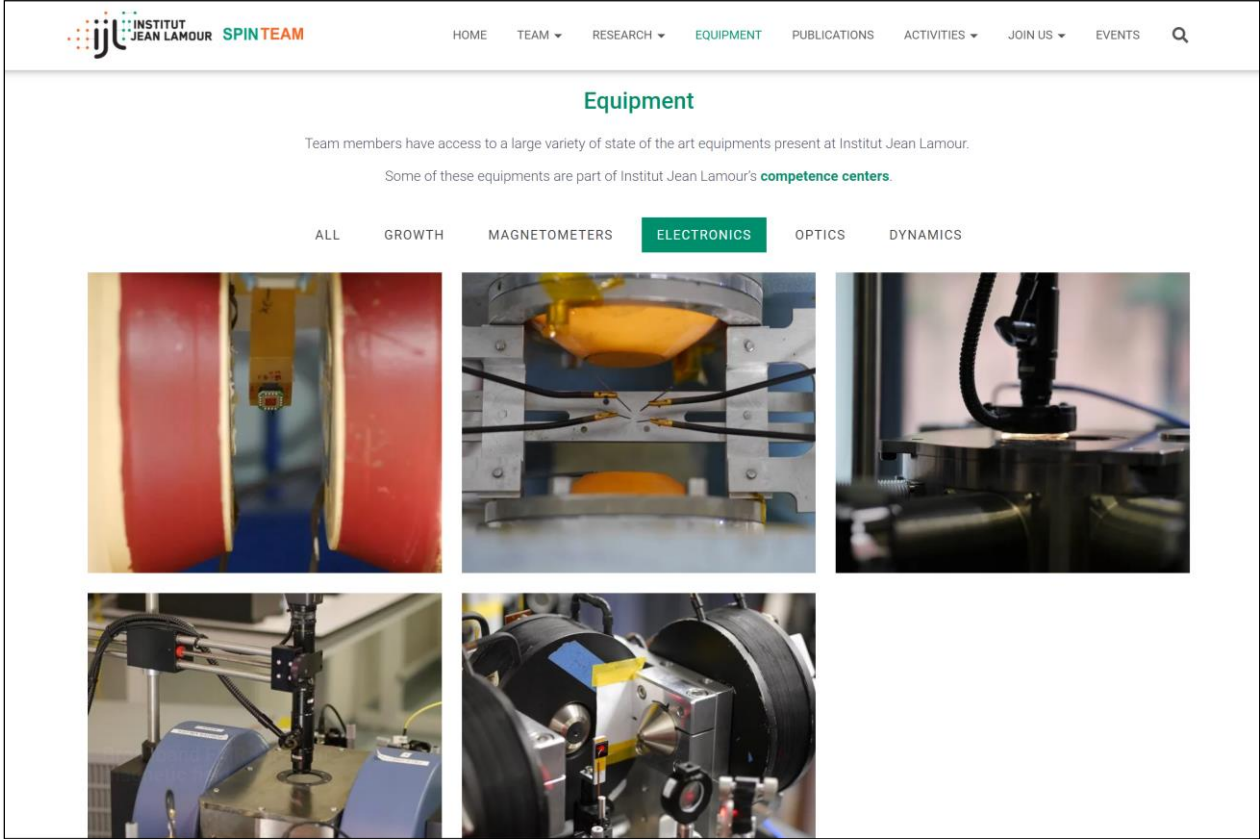


Fig. 24:: Equipment page.

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