Project title: Optical Control of Nanoscale Spin Textures (OptiSPIN)
Project number: K162/2018

Executive Summary

In the framework of the Leibniz Collaborative Excellence project OptiSPIN, the Max Born Institute, Berlin (MBI), the Max Planck Institute for Intelligent Systems, Stuttgart (MPI-IS) and the Peter Grünberg Institute of the Research Center Jülich (FZJ) have formed a collaboration to study optical control of magnetization on a nanometer scale. Specific research questions include investigations on how light pulses couple to magnetization with particular view on topologically non-trivial textures and chiral interactions. Experimentally, the project is based on the integration of a specialized in-situ picosecond laser developed by MBI into the scanning x-ray microscope MAXYMUS operated by MPI-IS at the BESSY II synchrotron-radiation source. The FZJ group supports the project with expertise in theoretical modeling including the ab-initio description of the electronic structure, surface and interface effects, topologically non-trivial spin textures and their dynamics as well as response to external stimuli.

The OptiSPIN collaboration was working successfully over the funding period from 2019 until 2023. Even beyond the funding period, the project partners still cooperate in the scientific network established, which includes also new external partners. Two of the researcher positions financed by the project were filled with PhD students (both female) who both have received their degree in 2023. Two positions were filled with PostDocs.

Technically, the program has reached its main objective with the commissioning of the custom-developed laser system at the x-ray microscope. After installation, the laser was continuously upgraded with particular attention to shortening the pulse duration and increasing the laser power. We could demonstrate excellent performance of the laser system in single-shot applications investigating laser-induced modifications as well as in picosecond time-resolved experiments. The integration of the laser into an x-ray microscope provides an experimental platform to study the interaction of materials with ultrashort laser pulses which is still unique world-wide and reaches beyond magnetic materials.

Difficulties in the project have emerged from the restrictions due to the Covid-19 pandemic, reducing the amount of experimental work that could be carried out. Most importantly, access to the large-scale facilities, where the experimental work was predominately carried out, was restricted. The project was therefore extended by one year. As a result, the scientific project objectives could be reached and several remarkable scientific results have been obtained.

Most noteworthy, we were able to demonstrate ultrafast optically induced magnetic skyrmion nucleation and annihilation on the nanometer-scale for the first time. Based on time-resolved x-ray scattering experiments and atomistic modeling, we revealed the time-scale of the nucleation and its microscopic mechanism. Proceeding from this fundamental understanding, we have developed application schemes including efficient means to control skyrmions on the nanometer scale. The theoretical work within OptiSPIN is similarly successful focusing on the question how chiral magnetic textures can be created by optical pulses via exploiting the electron dynamics, under the influence of thermal fluctuations and electronic scattering.

The scientific work in the project has already led to 20 peer-reviewed articles, including publications in *Nature Materials*, *Nano Letters* and *Nature Communications*. Furthermore, a patent has been submitted. We expect that more results will be published in the near future with manuscripts already being submitted and data currently under evaluation. Given the technical opportunities created and the scientific network established, all project partners will continue research in the field of controlling magnetism with lasers on the nanometer scale.

1. Achievement of objectives and milestones

Experimentally, the project builds on the development of a specialized picosecond laser system from MBI and its integration into the x-ray microscope MAXYMUS operated by MPI-IS at the BESSY synchrotron-radiation source at Helmholtz Zentrum Berlin (HZB). This system has been put into operation and was commissioned successfully in 2019. Optimized for operation with asynchronous sampling mode for time-resolved experiments with the full bunch pattern of BESSY, this new experimental platform integrating optical laser excitation and x-ray microscopy is highly performant, and, to our knowledge, a unique instrument world-wide. This achievement constitutes the most important milestone of the project as the experimental work strongly depends on it. While the construction of the laser was entirely financed by MBI, we have requested funds for the upgrade of the laser within the OptiSPIN project. We have shifted the focus of the upgrade from higher repetition rate as outlined in the application towards a reduction of the pulse length to below 1 ps, and, at the same time, to a significant increase of the laser power, resulting in a single-shot fluence of several 100 mJ/cm^2 on the sample. These laser parameters unlock access to magnetization dynamics passing through highly excited states. The following relaxation dynamics and the nanoscale magnetic states finally reached are of high technological importance and can now uniquely be investigated at MAXYMUS. With the upgrade, we have reached all technical objectives of the project.

The project worked also scientifically very successful with in total 20 publications in peer reviewed journals to date. Regarding the three experimental work packages (WP1–3), almost all objectives have been accomplished and have led to very noteworthy scientific results, as detailed in section 3. We have already published a number of articles on the laser-induced nucleation of topological spin textures, in particular skyrmions (WP1). This work includes research on nanoscale materials processing to control the dynamics of magnetic skymions directly (WP2) or locally tailor the optical excitation amplitudes and the induced heat (WP1). Results on *time-resolved* measurements of the skyrmion dynamics have been published in a PhD thesis and article manuscripts are in preparation (WP3). A second focus of our experimental work was on the nanoscale control of photo-induced demagnetization and switching in ferrimagnets, where we have published a time-resolved study and are currently compiling a manuscript on a patterning approach (WP2). The theory work in WP4 was equally productive, making significant advances in theoretical description of interaction of chiral textures with light. The project group has reached all goals in the work package with no less than 11 peer-reviewed publications.

The project partners closely adhered to the original financial planning. All four positions funded by the project have been staffed. However, MPI-IS and PGI-1/FZJ have decided to fill their position with PostDocs in order to better execute the ambitious work program. The investment budget was used for the laser upgrade as requested and was complemented by in-kind contributions from MBI. Due to delays particularly in the experimental part as a result from the Covid-19 pandemic, the project was extended by one year almost without additional funding (the exception being the subsidy to PhD student salaries kindly offered by the Leibniz Association). Part of the travel funds were used to extend the contracts of the PhD students.

2. Activities and obstacles

On a technical level, the installation and commissioning of the laser system at the MAXYMUS endstation was a crucial precondition for the execution of the scientific work in the project. This work was carried out in very close cooperation between MBI and MPI-IS, resulting in a very successful demonstration of the in-situ laser operation already during the very first beamtime in January 2020.

The laser was then regularly used in beamtimes allocated by BESSY II after recommendation of our scientific proposals by the review panel. We could perform experiments almost every semester, only interrupted by Covid-19-related shutdowns and access restrictions. The breaks between beamtimes at BESSY II were partially compensated by performing additional experiments at other large-scale facilities. Using holographic x-ray imaging, we were able to demonstrate laser-induced nucleation of nanometer-scale skyrmions in ferromagnetic multilayers for the first time. The experiments at PETRA III (DESY, Hamburg) were made possible by equipping the MBI x-ray holography instrument with a sub-ps commercial fiber laser system. In addition, time-resolved investigations of the skyrmion nucleation were carried out at free-electron laser sources as European XFEL in Hamburg and FERMI in Trieste. Combining experiment and theory we found that the nucleation proceeds via a high-temperature fluctuation phase mediating the topological phase transition to form skyrmions.

Published at the end of 2020 in *Nature Materials*, this is a highly visible result which was met with very large interest in the topological spin structure community.
The laser development, installation and technical support at BESSY II is predominately carried out by permanently employed staff in order to ensure reliable laser operation and support also beyond the project end. One senior scientist and one technician at MBI take this responsibility, interfacing with the beamline scientists employed by MPI-IS and HZB. In 2019, the MBI has employed two PhD students financed by the project to work on the specific scientific topics proposed (WP1 and WP2). At the start of the project, the MPI-IS group had difficulties in finding a suitable candidate for their position in the project. In addition, one of the project leaders on the MPI-IS side left the group in 2020. Together with the general difficulties associated with the Covid-19 pandemic, this shortage of qualified personnel led to a delay of the scientific work in WP3. In order to accelerate progress for the second half of the project, the project coordinators decided to fill the WP3 scientist position with a PostDoc who is a very experienced researcher for time-resolved imaging via scanning x-ray microscopy. The PGI-1/FZJ theory group also decided to staff the position associated with WP4 with a PostDoc to work on the theory program in the project.
The PGI-1/FZJ group covered the theoretical treatment of magnetic materials in the project including ab-initio description of the electronic structure, surface and interface effects, topologically non-trivial spin textures and their dynamics as well as response to external stimuli. The key idea is to understand how chiral textures can be created by optical pulses via exploiting the electron dynamics effects, under the influence of thermal fluctuations and electronic scattering. The theory team has developed and implemented the quantum-classical solver that combined the exact quantum evolution of electronic states with the more classical evolution of local magnetic moments to which electrons are coupled. This has been used to establish the profoundly non-thermal mechanism of fluctuations-mediated coherent chiral electronic excitations exerting ultrafast torques on local moments in response to light, as a driving force behind formation of transient chiral states on a ps-time scale. The relevance of the novel mechanism for generating complex textures has been demonstrated for a spin-spiral order creation as well as two-dimensional meron-antimeron spin lattice creation by light in antiferromagnets which are collinear in their ground state. Generally, the conclusions of the work concerning the importance of fluctuations for driving complex states in magnets are in accord with the experimental effort which was conducted in other work packages. The unexpected role of coherent electronic dynamics opens new perspective in educated design of optically driven chiral dynamics.

3. Results and successes

In the course of the OptiSPIN project, we have published 20 peer-reviewed articles, so far, including publications in high-profile journals, namely *Nature Materials*, *Nature Communications*, and *Nano Letters*. We are already preparing and have submitted more manuscripts and expect to publish more noteworthy results within in the OptiSPIN frame work in the near future. Two PhD theses were completed as well as one Master's thesis and two Bachelor theses. In the following, we will briefly report on the results of main scientific topics addressed in then project:

Laser-driven generation, manipulation and annihilation of skyrmions: We have covered this topic with great success. Using x-ray imaging, we were able to demonstrate skyrmion nucleation and annihilation on the nanometer-scale. Concepts for technological application of these findings were published in *Applied Physics Letters* **118**, 192403 (2021). A patent on one of the developments is currently in the disclosure phase. Moreover, time-resolved x-ray scattering at European XFEL in concert with atomistic modeling led to a microscopic understanding of the dynamics of the process, in particular, with respect to the topology of the system [*Nature Materials* **20**, 30 (2021)]. More data providing deeper insights into the transient high-temperature fluctuation state mediating the topological transition have been acquired in experiments at the XFEL FERMI in Trieste with a manuscript already under review [arXiv:2212.03143 (2022)].

Laser-driven magnetic switching at inhomogeneities and nanostructures: Based on the results from the previous topic, we investigated strategies to control the position of the skyrmion nucleation on the nanometer-scale. We successfully developed a scheme based on reliable shaping of the optical excitation strength by a proximity mask [*Phys. Rev. B.* **106**, 054435 (2022)]. Alternatively, we use patterning with a focused beam of He-ions to locally alter the magnetic properties [*Beilstein Journal of Nanotechnology* **12**, 304 (2021)] and steer the skymion dynamics on the nanoscale [*Nano Letters* **22**, 4028 (2022)]. The MPI-IS team was additionally involved controlling skyrmions in a 2D material [*Nano Letters* **22**, 9236 (2022)] and did first successful measurements on the laser-based skyrmion nucleation in these materials. This data is currently under evaluation.

Laser-driven magnetic switching on the nanoscale: We have shifted the focus of these activities from ferromagnetic materials to rare-earth–transition-metal ferrimagnets. Here, we have demonstrated time-resolved imaging of the laterally inhomogeneous demagnetization at the maximum repetition rate of the laser of 50 MHz [*Structural Dynamics* **10**, 024301 (2023)]. To our knowledge, this is the highest repetition rate used in pump-probe measurements of this kind, so far. This achievement was possible by applying careful measures to manage the heat load from the laser excitation. Furthermore, we have again used He-ion patterning to define nanoscale areas for all-optical switching in these materials which we then investigated with lab-based MOKE microscopy and at MAXYMUS. These results have been achieved within a Master's thesis and an article manuscript is currently in preparation.

Skyrmion dynamics: Based on the control of the skymrions by material modification with He ions, we have developed a new platform for repetitive pump-probe imaging to study the pico and nanosecond dynamics of skyrmions. We performed time-resolved measurements at beamline P04 of PETRA III and MAXYMUS where we imaged spin-orbit torque and laser-induced magnetization dynamics. The results were already published in a PhD thesis and manuscripts for journal articles are in preparation. Studying the interaction of skyrmions with spin-wave in a direct imaging approach as proposed in our application has turned out to be challenging. The MPI-IS group is still very active in developing this field [*Crystals* **11**, 546 (2021*), IEEE Trans. Magn.* **58**, 1 (2022)] and in joint efforts from MBI and MPI-IS we are working on a platform to optically excite coherent spin waves. These are ongoing activities.

Theoretical description of topological spin structures and defects: The PGI-1/FZJ group made significant advances in theoretical description of interaction of chiral textures with light (WP4). (i) The coherent description of the interplay between spin torques, dynamical orbital magnetism and non-equilibrium Dzyaloshinskii-Moriya interaction in dynamical magnetization textures has been provided [*Phys. Rev. B* **120**, 245411 (2020)]; (ii) The prominent role that the chirality can play for mediating colossal optical torques on the magnetization has been established [*Phys. Rev. B* **103**, 054403 (2021)]; (iii) the properties of magnetic chirality as a robust and viable variable of magnetic systems in ultrafast chiral magnetism have been demonstrated. The foundation for time-resolved ultrafast experimental detection of chirality with the help of FEL pulses has been established. [*Nature Comms*. **11**, 6304 (2020)]; (iv) The first steps towards current response of ultrafast chiral dynamics in terms of photocurrents have been made by developing the methodology of non-linear magnetoresistance, non-linear Hall effects and photocurrents have been made [*J. Phys.: Condens. Matter* **34**, 055301 (2022), *Phys. Rev. B* **103**, 075408 (2021)]. At the same time, the interplay of chiral magnetism with orbital degrees of freedom has been reviewed [*Solid State Physics* **71**, 1-38 (2020)]. (v) The methodology for time-dependent treatment of electron dynamics in combination with the magnetization dynamics in response to ultrashort pulses has been successfully developed, implemented and applied [*Phys. Rev. Research* **5**, L022007 (2023), *Comm. Physics* **5**, 69 (2022)].

4. Equal opportunities, career development and internationalisation

All three institutes participating in this Leibniz Collaborative Excellence project are certified as family friendly via the audit "berufundfamilie" and have special measures in place to foster careers of women in science. Specifically concerning OptiSPIN, we have reached our goal of filling at least half of the positions funded by OptiSPIN with female scientists. All positions were internationally advertised and one of the four scientists employed is a non-German scientist. Both female scientists were attracted to come back to Germany after M.Sc. programs abroad. Both PhD students were members of graduate schools (Integrated Research Training Group of the CPC/TRR 227 “Ultrafast Spin Dynamics” and Berlin School of Optical Sciences and Quantum Technologies) in order to promote scientific exchange across institutions, provide networking opportunities and soft-skill training. We foster and encourage the participation of the PhD students and PostDocs on national and international conferences and workshops in order to build up a professional network.

5. Structures and collaboration

The collaboration between the three institutes as outlined in the application for this Leibniz Collaborative Excellence project was firmly established and the project partners still cooperate in the scientific network established. We actively extended the collaboration beyond the OptiSPIN team for specific projects where we benefited from external expertise (sample fabrication, instrumentation or modeling), and scientific exchange.

6. Quality assurance

Publications are only submitted to peer-reviewed scientific journals. Preferably, we publish as open access. If this is not possible, author versions of the manuscripts are made public via arXiv or university archives. Submissions are routinely checked by plagiarism software prior to submission. Where reasonable, we deposit the data underlying the publications on public data archives. In any case, all raw data, analysis code etc. is archived for at least ten years locally by the institutes. All participating groups are bound by the rules of good scientific practice as set out by the *Deutsche Forschungsgemeinschaft*.

7. Additional resources

Improvements, extensions and maintenance of the laser system installed at MAXYMUS were cofinanced via the MBI institutional budget. This effort included permanently employed personnel (one senior scientist 0,25 FTE×48 months ≈ 90 T€, one technician 0,25 FTE×48 months ≈ 60 T€) and funds for investment and consumables (≈150 T€). Dr. Pfau who is permanently employed by MBI was responsible for the project coordination and daily supervision of the PhD students and carried out part of the scientific research in the project (0, 25 FTE×48 months ≈ 90 T€).

8. Outlook

With the end of this project, the laser installed at MAXYMUS is fully developed and his entirely integrated into the research at the microscope. The laser will be further operated and maintained by MBI and usage is also open for other groups via cooperation agreements. In the future, we will continue working on controlling the interaction of laser pulses with magnetic materials, in particular, by using the flexible platform of He-ion based patterning developed within OptiSPIN. We also have ongoing activities to study the laser-induced nucleation of skyrmion in order to achieve a better understanding of the physics behind the new kind of topological phase transition found in this project. This research is continuously supported by theory further pursuing the vision that the properties of electronic dynamics can be utilized for driving various spin states in magnetic materials as mediated by fluctuations, in two as well as in three spatial dimensions. We will also push forward the recently acquired understanding of the key role that the orbital degree of freedom of electrons plays for magnetization dynamics, potentially unravelling the role of orbital dynamics and orbital momentum redistribution for ultrafast topological dynamics of spin textures reaching into the regime of three-dimensional spin structure dynamics.