Publishable Summary for 17FUN08 TOPS
Metrology for topological spin structures

Overview

In recent years, topology (the study of the properties of geometric configurations which are unaltered by certain elastic transformations such as a stretching, bending or twisting) has emerged as a fascinating phenomenon in solid state research from both fundamental and applied perspectives. This is particularly the case for certain magnetisation configurations, where the topology protects the spatial magnetisation or spin arrangement. Due to their unique properties, such topologically-protected spin structures (TSS) have the potential to revolutionise the Information and Communications Technology sector. This project has supported fundamental research in this active field by developing metrological tools and methods for the characterisation of TSS. This includes accurate measurements of the Dzyaloshinskii-Moriya interaction (DMI) constant, spatially-resolved methods for the identification and manipulation of multiple and individual TSS, and reliable concepts for the investigation of dynamical properties of TSS. With these project results, the scientific community has now more information as well as better tools and techniques at hand, which will be helpful for the possible realisation of microwave and magnonics related applications based on TSS. Moreover, the DMI-measurement comparison has created awareness in the scientific community of the reliability and unreliability of measurement results.

Need

Fundamental research in the field of spintronics has led both to the recognition of scientific merits at the highest level and to extremely fast development of a huge market sector dealing with mass production of consumer and industrial electronics, such as hard disk storage devices and sensors for mobile phones and cars. The search for new materials featuring room temperature operation, ultralow power consumption, full electrical control, and scalability continues apace. Several years ago, the study of spin structures with a certain topologically-protected spin arrangement, has moved into worldwide focus. Despite intensive research on TSS (such as *chiral domain walls* which are boundaries between regions of uniform magnetisation with a certain gradual rotation of the magnetisation or *skyrmions,* which are vortex-like spin arrangements with diameters typically ranging between several nm to several 100 nm), there have been several high-level requirements in this field connecting basic research, metrology, and ability to exploit these structures in novel devices that needed to be addressed:

- The quest for new materials and systems with stable TSS requires a precise understanding and knowledge of relevant material parameters such as the Dzyaloshinskii-Moriya interaction (DMI) constant. However, validated metrology tools for these parameters did not exist.

- Due to their nanoscale size, it is difficult to experimentally probe some types of TSS. Therefore, validated measurement methods were needed enabling the identification and manipulation of multiple and individual TSS.

- Reliable concepts for the investigation of current- and field-induced dynamics of TSS at GHz and THz frequencies had to be developed. In addition, experimental high-risk-high-gain research was required to verify whether TSS enable the realisation of novel quantum standards.

- Additional research on TSS required the fabrication of TSS with reproducible topological characteristics. Micromagnetic simulations and analytical tools were required to validate experimental results and to reliably predict novel material properties.

Objectives

The overall objective of this project was to develop and establish metrological and scientific tools for the characterisation of TSS. This work was expected to significantly contribute to the development of new magnetic storage, spin-logic, and microwave devices in the future well as new quantum standards.

EMPIR EURAMET
The EMPIR initiative is co-funded by the European Union's Horizon 2020 research and innovation programme and the EMPIR Participating States

The specific objectives of this project were:

1. To develop and validate metrology tools and methods for reliable determination of key material parameters of TSS, i.e., the Dzyaloshinskii-Moriya interaction (DMI) constant.
2. To develop, compare and validate measurement techniques capable of unambiguously identifying and manipulating specific nanometre-scale TSS, such as domain walls, bubbles, and skyrmions in different magnetic materials. These methods would be applicable to both multiple and individual TSS.
3. To develop methods for the investigation and analysis of novel dynamical and quantisation effects in TSS. This work would capture the dynamics of TSS at GHz and THz frequencies and explore whether TSS might serve as quantum standards at room temperature and low magnetic fields.
4. To provide protocols for the reproducible growth of materials for experiments on TSS and reliable micromagnetic simulations and analytical tools for the modelling of TSS. The simulations would allow for a comparison with and an interpretation of experimental results.
5. To implement a research network on TSS in Europe with complementary infrastructure. To develop guidelines for accurate characterisation of TSS and to implement new measurement services on the DMI constant.

Progress beyond the state of the art

The accuracy of the determination of the DMI constant is crucial for designing materials with TSS and modelling TSS. Different strategies for DMI measurement have been attempted leading in part to contradictive results. It was therefore not yet clear how DMI depends on material properties such as composition, interface quality, or layer thicknesses. The project undertook, for the first time, a systematic comparison of different methods for the determination of the DMI constant and developed a measurement setup suitable for establishing a measurement service for the DMI constant.

TSS such as skyrmions can have a diameter ranging from several nm to several 100 nm. Skyrmions have already been identified using several high-resolution measurement techniques. However, especially with regard to thin films the exact phase transitions of skyrmions are still under deep discussion. The project developed quantitative measurement techniques and techniques capable of unambiguously identifying multiple and individual TSS. Moreover, novel emergent thermoelectric and electrodynamic phenomena, e.g., spin-charge coupling, was studied and led to new insight into the electrical manipulation of TSS.

So far, characteristic dynamical modes, which typically occur at several GHz have been identified in TSS by comparing experiments with simulations. Yet, little is known about ultrafast dynamics of topological spin structures at GHz and THz frequencies. This project investigated high-frequency properties of TSS by direct imaging of the characteristic modes at GHz frequencies and by performing time-resolved magneto-optic Kerr effect measurements on TSS. Moreover, feasibility studies towards novel quantum resistance standards operating at room temperature and low magnetic fields was explored.

Several bulk crystals have been identified as prototypical materials for TSS and different thin films allow for the existence of TSS. Modelling of TSS has already been used to predict novel properties, however, some topics still lack a general theory. This project developed protocols for the reproducible growth of materials for TSS with the aim of obtaining stable high-quality and large-size materials. Moreover, the experiments in this project were supported with micromagnetic simulations, which was essential for the success of the project.

Results

Reliable determination of key material parameters of TSS (objective 1)

Different methods have been proposed to evaluate the interfacial Dzyaloshinskii-Moriya interaction (DMI). The two most popular methods employed by the scientific community are the asymmetric bubble expansion method using the magneto-optic Kerr effect (MOKE) and the determination of spin wave non-reciprocity exploiting Brillouin light scattering (BLS).

In a worldwide first international round robin comparison, the project partners and collaborators compared these two methods and focused, in particular, on the MOKE method, which was envisioned as a possible candidate for a measurement service. Since the evaluation of the DMI constant from the MOKE measurement is an ongoing discussion in the community, the most promising theoretical models, applied to a range of different magnetic multilayers with perpendicular magnetic anisotropy (PMA), have been investigated for the extraction of the DMI constant using measured MOKE data. The asymmetric bubble expansion method can be applied to a wide variety of heterostructures with magnetic bubble domains. However, its applicability depends strongly on the interface quality. It was found that models based on the standard creep hypothesis are not able to reproduce the domain wall (DW) velocity profile when the DW roughness is high. The project results demonstrate that the DW roughness and the interface roughness of the sample layers are correlated. The rougher the bubble DW, the more difficult is it to identify the DW velocity and its minimum in order to evaluate the DMI constant.

The automatised determination of the DW velocity by using a software package, which was published at GitHub, may be a future tool for a more reliable analysis. Regarding the identification of the minimum velocity by models, samples characterised by a lower DW roughness can be modelled by the standard creep model, while the arbitrary angle propagation model provides insights on the presence of magnetic effects from neighbouring bubbles. Samples with induced defects by irradiation show different behaviour though. The DMI value is increasing with increasing irradiation. As a result of the investigations, guidance was given on how to obtain reliable results for the DMI value with this popular method, including corresponding measurement uncertainties. A comparison of the results with BLS measurements on the same samples showed that the BLS approach often results in higher measured values of DMI. This fact is topic of ongoing research. A corresponding paper has been submitted to a peer reviewed journal and uploaded to the arXiv (https://arxiv.org/abs/2201.04925).

Based on these results, this project objective was successfully achieved.

Unambiguous identification and manipulation of specific nanometre-scale TSS (objective 2)

Magnetic force microscopy (MFM) is a widespread technique for imaging magnetic structures with a resolution of some 10 nm. MFM can be calibrated to obtain quantitative (qMFM) spatially resolved magnetisation data in units of A/m by determining the calibrated point spread function of the instrument, its instrument calibration function (ICF), from a measurement of a well-known reference sample. The project partners have together with collaborators explored how a Ti/Pt/Co multilayer stack can be used as a suitable reference material (https://arxiv.org/abs/2201.09763). Additionally, the consortium has used qMFM to analyse the radial dependent phase response of skyrmions in chiral magnetic multilayers. A simple model has been developed that treats the skyrmion size and domain wall as independent parameters, from which one can easily explore the role of each on the calculated expected phase response of qMFM. The results highlight how qMFM can be used to shed light on the underlying magnetization structure of skyrmions, and for the numerical or micromagnetic simulations results validation. It is anticipated that this work will help to expedite the process to optimise the magnetic parameters used for modelling skyrmionic systems, accelerating the technological development of skyrmionic systems. A corresponding paper detailing these findings has been submitted to a peer-reviewed journal.

Moving from qMFM techniques to other methods, a paper on the identification of Neel and Bloch type skyrmions by Lorentz Transmission electron microscopy (LTEM) which details the procedure to identify the skyrmion size using this method has been published on the arXiv (https://arxiv.org/abs/2002.12469) and as a peer-reviewed paper in Ultramicroscopy.

Another work performed by project partners has been devoted to comparing the results of different experimental methods namely magnetic force microscopy (MFM) and LTEM on nominally identical samples grown in the same batch. Since MFM and LTEM measurements require different substrates (electron transparent membranes in the case of LTEM and standard Si substrates in the case of MFM), no exact consensus on the appearance of skyrmions in temperature and magnetic field phase space could be reached, probably due to different strain in the deposited films. The paper has been submitted to a peer-reviewed journal and uploaded to the arXiv (https://arxiv.org/abs/2111.12634).

In addition, a large variety of thermoelectric and transport measurements have been undertaken. From this work, a paper on individual skyrmion manipulation by local magnetic field gradients has been published in Communication Physics (https://arxiv.org/abs/1903.00367). An additional paper on the thermoelectric signature of individual skyrmions has been published in Phsical Review Letters (https://arxiv.org/abs/2001.10251v3). Moreover, a manuscript on deterministic field-free skyrmion nucleation has been published in Nano Letters (https://arxiv.org/abs/1902.10435).

Finally, the project partners together with collaborators, studied skyrmions in Pt/CoB/Ir multilayers by means of scanning transmission x-ray microscopy at the Swiss Light Source at the Paul Scherrer Institute. By this means the average velocity and skyrmion Hall angle for each skyrmion of a known diameter could be determined. The work has been published in Nature Communications (https://doi.org/10.1038/s41467-019-14232-9).

Based on these results, this project objective was successfully achieved.

Analysis of novel dynamical and quantisation effects in TSS (objective 3)

Dynamic magnetoelectric modes in TSS have been extensively studied using ferromagnetic-resonance-(FMR)-based techniques and Brillouin light scattering (BLS). BLS measurements have been performed on He+ irradiated Co/Pt multilayers hosting skyrmions and the results have been compared to FMR measurements, achieving a careful determination of the first- and second- order anisotropy constants, as well as of the DMI constant, that are important parameters to tune the characteristics of skyrmion formation in these samples. The main results have been published in Scientific Reports (https://arxiv.org/abs/2105.03976).

Ferromagnetic resonance (FMR) studies have been performed for different alloys of FeCoSi as well as for Cu_2OSeO_3. These studies have been realised at different institutes and within a researcher mobility grant (RMG). Using X-ray magnetic circular-dichroism-based techniques (XFMR), the dynamic modes for Cu_2OSeO_3 have been unambiguously identified since this material showed the lowest damping parameter making the identification of the modes straight forward. The findings have been published in Physical Review Letters (https://arxiv.org/abs/1909.08293). Additionally, the identification of skyrmion modes in the low temperature skyrmion phase of the bulk B20 material Cu_2OSeO_3 has been achieved and published in Physical Review Letters (https://arxiv.org/abs/2011.07826).

Recently, it has been shown that certain materials possess a so-called metastable skyrmion phase (MSkL), where skyrmions exist in an extended parameter range. The project partners have studied the skyrmion dynamics in the MSkL for the first time. Using $Fe_{1-x}Co_xSi$ and employing time-resolved magneto-optical Kerr effect (TR-MOKE) measurements, the higher potential of the MSkL for applications as compared to the equilibrium skyrmion phase was demonstrated and it was shown that the MSkL is well suited to investigate generic properties of skyrmion dynamics. The studies have been summarised in a manuscript submitted for peer review and published on the arXiv (http://arxiv.org/abs/2202.04182).

Finally, some feasibility studies with respect to new quantum standards have been performed. The experimental high-risk-high-gain research carried out with respect to this deliverable had the goal of studying the predicted Hall quantization depending on ultra-clean interfaces between a graphene monolayer and a TSS. Unfortunately, despite of the high quality of the available graphene material it turned out that required excellent interface properties could not be achieved by placing the skyrmion hosting materials on top of the graphene layers. Instead, the project partners have studied an ultra-clean hybrid bilayer system consisting of graphene mono- and bilayers and ultra-thin metallic films. The growth and structural properties of this novel hybrid material are discussed in a paper published in Phys. Rev. Materials (https://arxiv.org/abs/1905.12438).

Based on these results, this project objective was successfully achieved.

Protocols for reproducible growth of materials and reliable micromagnetic simulations and analytical tools (objective 4)

Protocols for reproducible growth of TSS have been developed. To this end, different growth parameters have been optimised with the aim of obtaining stable high-quality thin-film materials and large-size high-quality bulk materials. The size aspect is especially important for future applications since at present the bulk material size is restricted a few millimetres. In total, a variety of multilayer and bulk samples have been grown and characterised. Samples have been shipped to partners for further experiments completing this task successfully.

Despite previous theoretical work, the development of new models was an essential prerequisite to validate the experimental results obtained in this project. Several phenomena such as the anomalous Hall effect, magnetoelectric modes, and spin-charge coupling could only be reliably interpreted and assigned to TSS characteristics with the availability of accurate simulations. To this end the project partners performed joint micromagnetic simulations.

A first study concentrated on the spin wave eigenmodes of isolated elliptical nanodots, showing the effect of DMI on both the eigenmodes frequencies and their spatial character. The results have been published Appl. Sci. (https://doi.org/10.3390/app11072929).

A second micromagnetic study was concerned with magnonic crystals consisting of a ferromagnetic film supported by an array of heavy-metal stripes. The effect of DMI on the magnonic bands of the magnonic crystal was investigated, suggesting a way to take advantage from the artificial periodicity of the magnonic crystal to achieve a better sensitivity in the determination of the DMI constant by BLS. The work has been published in the Journal of Magnetism and Magnetic Materials (https://arxiv.org/abs/2112.05360).

A third micromagnetic analysis concentrated on the eigenmodes of single and coupled skyrmions, showing that the band structure of a chain of skyrmions can be interpreted starting from the eigenmodes of a single isolated skyrmion and then considering the effects of both dipolar and exchange coupling. The work has been published in published in IEEE Magn. Lett. (https://arxiv.org/abs/2112.04967).

Finally, the project partners have employed a non-abelian gauge theory to derive the relation between the potential form of the magnetic DMI energy term. The key outcome of the theory is that the traditional spatial derivative appearing in the micromagnetic energy functional, must be substitution by a gauge covariant derivative which includes a gauge field. This theory has implications for determining surface-DMI and bulk-DMI terms.

Based on these results, this project objective was successfully achieved.

Impact

This project has created impact for the scientific, metrological, industrial and end user communities through fundamental investigations on spintronics of magnetic nanolayers and nanosystems with the first steps towards traceable measurements on such devices, initial research towards future metrological applications, and availability of a good practice guide for accurate characterisation of topological spin structures. The latter will form the basis for a possible future measurement service in Europe.

Key dissemination activities to date are (i) 71 presentations at international conferences such as MMM-Intermag, CLEO, CPEM, JEMS, or SPIE Photonics West; (ii) 22 published articles in peer-reviewed journals such as Nano Letters, Communication Physics, Physical Review Letters, Physical Review B, Applied Physical Letters; additionally, 4 papers have been submitted to peer-reviewed journals; (iii) four newsletters and several interactive training courses/talks accessible form the project webpage. These achievements outperform the initial targets, despite institute closures due to COVID-19.

Impact on industrial and other user communities

ICT is an important sector for economic development in Europe and affects economic growth across the economy. The research in this project has enabled a fundamental understanding of novel spintronic effects, possibly enabling promising new device concepts.

Close cooperation between NMIs and leading research institutes has provided unique expertise to support the existing research networks on topological spin structures in Europe. Such cooperation might also lead to a follow-on collaboration after the project lifetime and, thus, strengthen the research in Europe. European researchers pioneered this field and are at present among the leading experts. Moreover, several national programmes on topological spin structures are still running. Interaction with different national programmes was an important dissemination activity, e.g., through jointly organised workshops. The DMI already has an influence on today's magnetic nanoscale devices. With an ongoing decrease of the size of such devices, the influence will even become more important. The successful implementation of the project concerning a Round-Robin comparison and guidelines for accurate measurements of the DMI constant is an achievement from which industry will profit in the short- and long term, respectively.

The project yielded information about charge-spin coupling in TSS, single skyrmion detection, and dynamics of TSS. Such information will be essential for future applications of TSS, e.g., for racetrack memory devices, high density storage devices, or logic devices.

Impact on the metrology and scientific communities

The accuracy of the determination of the DMI strength is crucial for the design of future applications employing TSS, e.g., novel types of magnetic memories. Only if the DMI strength is uniquely defined and measured, a clear relation with intrinsic or extrinsic material properties can be established. This project has taken a major step to resolve this key scientific controversy by organising a round robin (RR) comparison measurement involving the world-leading groups in the field. For the first time the effects of different measurement setups on the determination of the DMI constant could be pinpointed. A good practice guide for the measurement of the DMI constant by Brillouin light scattering was written. Brillouin light scattering appears to be currently the more reliable one, having a straightforward analysis of the measured data, not depending on the sample material or quality. A discussion of the measurement uncertainties and sources for systematic errors are included in the report This guide will form the basis for a possible future measurement service for the DMI constant in Europe.

The TOPS consortium was co-organiser of the Skymag 2020 workshop, which was supposed to take place in Paris from 6-9 April 2020. Due to COVID restrictions, the workshop was postponed to April 2021 and held as an online event. Additionally, two partners of the consortium (Brian Hickey, ULE and Massimo Pasquale, INRIM) chaired a session at the Joint European Magnetic Symposia (JEMS, which took place in Lisbon in December 2020) with the title “Magnetic based metrology tools and techniques”. This session included two talks from the TOPS consortium. A dissemination workshop had been held online together with the final meeting. During the dissemination workshop, Giovanni Finocchio from the University of Messina gave the plenary talk.

Impact on relevant standards

So far, no relevant standards exist related to the measurement of the DMI constant. The consortium was in regularly contact with IEC TC 113 and IEC TC 68 to explore the possibility of implementing an IEC standard. However, especially due to the DMI measurement comparison, it was shown that a standard would be too premature and additional studies and comparisons will be necessary before a new standard can be feasible.

Longer-term economic, social and environmental impacts

Several possible industrial products for TSS devices are currently envisaged in the scientific community. First, TSS allow for discrete magnetic states being of smaller size and energetically more stable than their single-domain counterparts. For this reason, it is envisaged that TSS may be used as bits to store information in future memory and logic devices, where the state of the bit is encoded by the existence or non-existence of the TSS. This will have significant economic impact since the global digital storage device market is anticipated to reach $ 6.27 billion by 2022. Second, the position of TSS within a nanostructure may be manipulated using low current densities. Thus, TSS also provides promising candidates for future racetrack-type storage or logic devices. Third, TSS exhibit strong gyrotropic and breathing modes at GHz frequencies, which might open the avenue for TSS-based microwave applications.

A major goal of the EU is a 20 % increase of energy efficiency and a corresponding reduction of CO_2 gas emissions by 2020. The use of low power magnetic logic and storage devices based on TSS could lead to more energy efficient ICT and CE devices enabling a significant reduction of global energy consumption.

Society needs technologies based on innovative and disruptive products and concepts. TSS offers the potential to create novel spin-based electronic devices with improved speed, reliability, and significantly decreased power consumption.

The research carried out in this project supports the above longer-term impacts.

List of publications

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This list is also available here: https://www.euramet.org/repository/research-publications-repository-link/

Project start date and duration:		01 June 2018, 42 months
Coordinator: Mark Bieler, PTB	Tel: +49-531-592-2540	E-mail: mark.bieler@ptb.de
Project website address: https://www.ptb.de/empir2018/tops		
Internal Funded Partners: 1 PTB, Germany 2 INRIM, Italy 3 NPL, United Kingdom	External Funded Partners: 4 Singulus, Germany 5 TUM, Germany 6 ULE, United Kingdom 7 UNIPG, Italy	Unfunded Partners: -
RMG1: INRIM, Italy (Employing organisation); PTB, Germany (Guestworking organisation)		