Discovery of magnetic anti-skyrmions in tetragonal inverse Heusler compounds

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Magnetic skyrmions (Sks) and anti-skyrmions (aSks) are topologically protected nanoscopic vortices of magnetization that can be stabilized in magnets with broken inversion symmetry. Skyrmions have the potential to be used as magnetic bits in high density storage devices such as racetrack memories as well as for neuromorphic computing systems that go beyond Moore's law. These non-collinear topological spin textures arise as a result of a competition between the Heisenberg exchange mechanism and antisymmetric Dzyaloshinskii-Moriya exchange Interactions (DMI). The chirality of magnetic Sks essentially depends on the lattice symmetry and the sign of DMI. Depending on the helicity of spin rotation, different types of Sk textures (Fig. 1) have been observed, such as Bloch and Néel Sks and very recently aSks that have been discovered for the first time in a collaboration between Parkin's department at the MPI-MSP and Felser's department at the MPI-cpfs.

It has been shown that skyrmions can be laterally displaced by very low current densities (10^5-10^6 A/m^2) and, thus, have been promoted as promising for energy efficient, information storage devices. However, the bottleneck for Sk-based spintronics is the identification of novel materials hosting dense Sks at ambient temperature and in zero magnetic field. Also, the size of Sk is a critical parameter. So far Sk lattices have been experimentally realized at low temperatures in various bulk cubic B20 transition metal compounds e.g. MnSi, $Mn_{1-x}Fe_xSi$, $Mn_{1-x}Co_xSi$, $Fe_{1-x}Co_xSi$, FeGe, MnGe and the multiferroic insulator Cu_2OSeO_3. Only β-Mn-type bulk cubic chiral, $Co_8Zn_8Mn_4$ has been found to host Sks at ambient temperature, but in the presence of high magnetic fields. Another major drawback in these systems is that, magnetic Sk lattices are found to be stable only in a narrow regime of the temperature-magnetic field phase diagram, which limits their application. Therefore, it is our prime goal to conceive, design, and characterize novel materials that can host dense Sk/aSk lattices at ambient temperature and in zero magnetic fields.

Accomplished projects:

- **Discovery of an anti-skyrmion lattice in a tetragonal inverse Heusler material**

For the application of Sks in spintronics, key requirements are that the material should retain a magnetic ordering beyond room temperature and the Sk state should be stable over an extended temperature range. In addition, it is extremely important to control the magnetic anisotropy, which can influence the competition between helical magnetic states, skyrmion textures, and collinear spin states. In this context, tetragonal magnetic Heusler compounds, X_2YZ (where X, Y are transition metals and Z is a main group element) with multiple magnetic sublattices are potential candidates for the design of anisotropic and non-centrosymmetric room temperature magnets which provide a unique route to the formation and stabilization of aSk lattices. Besides, in Heusler compounds it is possible to manipulate the chemical composition as a means of controlling magnetic anisotropy and DMI.

We have discovered the existence of an anti-skyrmion lattice beyond room temperature in a tetragonal inverse Heusler material ($Mn_{1.4}Pt_{0.9}Pd_{0.1}Sn$) by direct imaging with Lorentz Transmission Electron Microscopy (LTEM) using an FEI TITAN G2 80-300 and double-tilt stages with variable temperature capabilities (100-

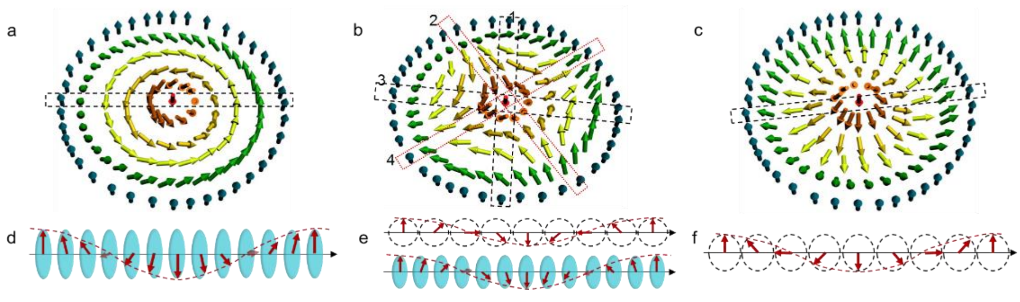

Figure 1. Different types of topological spin textures (a) Bloch skyrmion, (b) Anti-skyrmion and (c) Néel skyrmion. Cross-sections of respective spin textures along the radial directions indicated by the dashed rectangles show (d) a transverse helix for the Bloch skyrmion, (f) a cycloid for the Néel skyrmion, and (e) an alternating cycloid and transverse helix for the anti-skyrmion. Skyrmions and anti-skyrmions have opposite topological charge.

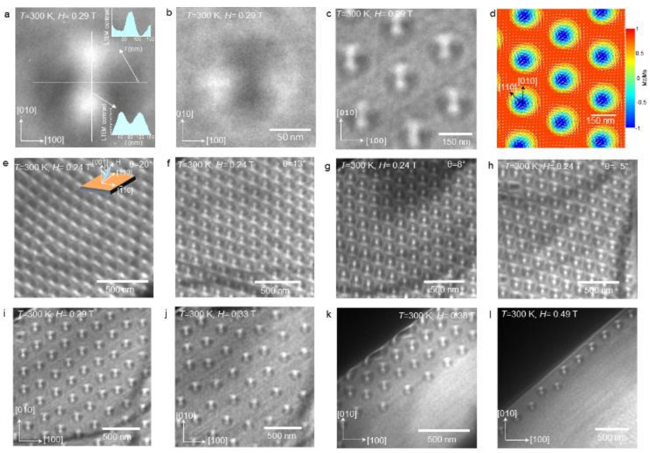

Figure 2. Anti-skyrmions in $Mn_{1.4}Pt_{0.9}Pd_{0.1}Sn$ at 300 K. (a) Under-focused LTEM image of a field-stabilized single anti-skyrmion. The lower and upper insets show the intensity profiles of the in-plane magnetization along the vertical and horizontal direction, respectively. (b) Over-focused LTEM image of the field-stabilized single anti-skyrmion in (a). (c) Under-focused LTEM image showing a hexagonal lattice of anti-skyrmions. (d) Simulation of an anti-skyrmion lattice in an oblique field. The colour represents the magnetization component normal to the sample plane. (e)–(h), Under-focused LTEM images of anti-skyrmions taken at 300 K and in H = 0.24 T with tilting the sample at an angle of θ = 20° (e), θ = 13° (f), θ = 8° (g) and θ = − 5° (h). (i)–(l), Under-focused LTEM images of anti-skyrmions taken at fields applied along the c-axis of 0.29 T (i), 0.33 T (j), 0.24 T (k) and 0.49 T (l).

400 K and 10-300 K). In the Lorentz mode we have the capability of nearly magnetic field free TEM imaging. It is a powerful method for real space imaging of skyrmions due to its high spatial resolution, and due the fact that it measures the *B*-field throughout the thickness of the sample. LTEM can identify transformations from a helical magnetic phase to the aSk phase and eventually to the fully aligned magnetic state over a wide range of temperature and magnetic field. A helimagnetic ground state, propagating in the tetragonal basal plane, transforms into an aSk lattice state under magnetic fields applied along the tetragonal axis [001] over a wide range of temperatures (100 - 400 K). In LTEM imaging, aSks appear as two bright and two dark lobes, as shown in Fig. 2(a). Due to the presence of alternating chiral boundaries which consist of helicoid and cycloid spin propagations, as shown in Fig. 1(b), the transmitted electron beam will converge towards the centre of the aSk in the vertical direction, whereas it will diverge in the horizontal direction. This assignment has been verified by micromagnetic simulations of the LTEM pattern. The most interesting findings in this study are that stable and metastable aSk lattice states are realized over a large region in the *B*–*T* phase diagram over the whole temperature range. The field stabilized aSk lattice at 300 K is very promising for applications.

Projects in progress:

- **Design of new Heusler compounds for Antiskyrmions and Skyrmions**
  - **Untwinned single crystals**

The undoped compound $Mn_{1.4}PtSn$ possesses the structural ingredients to display anti-skyrmions. Large single crystals are needed to carry out small angle neutron scattering to explore anti-skyrmions in the bulk. However, untwinned single crystals are difficult to synthesize due to the martensitic phase transition. Recently we established a successful method to grow micron sized twin-free single crystals of $Mn_{1.4}PtSn$ based on a self-flux method. Our structural refinement shows that the compound crystallizes in the non-centrosymmetric space group $I\bar{4}2d$ and that the Mn vacancies form an ordered arrangement as a superstructure of the tetragonal Heusler parent compound. The direction-dependent magnetic properties show a ferromagnetic transition at T_C = 392 K as well as a spin-reorientation transition at 170 K with a saturation moment of 4.7 μ_B/f.u. at 2 K. These transitions are also reflected in the temperature dependent resistivity which is metallic to the lowest temperatures explored.

  - **Thin films**

The presence of non-trivial spin structures can give rise to real space Berry curvatures and thereby topological/anomalous Hall or Nernst effects. We have experimentally observed a non-collinear spin configuration in the acentric tetragonal Heusler compound Mn_2RhSn [3]. Apart from the bulk materials, we have also studied epitaxial Mn_2RhSn films prepared by magnetron sputtering [4,5]. The tetragonally distorted Mn_2RhSn films show a topological contribution to the Hall resistivity of about 50 nΩ cm, which is ~5% of the anomalous Hall effect, for temperatures below 100 K. The topological Hall effect signifies the presence of a chiral magnetic structure induced by the non-collinear magnetic structure that Mn_2RhSn is known to exhibit. In thin films of Mn1.5PtSn, we observe a large topological Hall effect below the spin reorientation temperature, close to the value for bulk Mn1.5PtSn. Additionally, the topological Nernst effect NT = 125 nV K^{-1} measured in the same microstructure has a similar magnitude as reported for bulk MnGe. It is possible to extract the topological features in nanostructures as well as

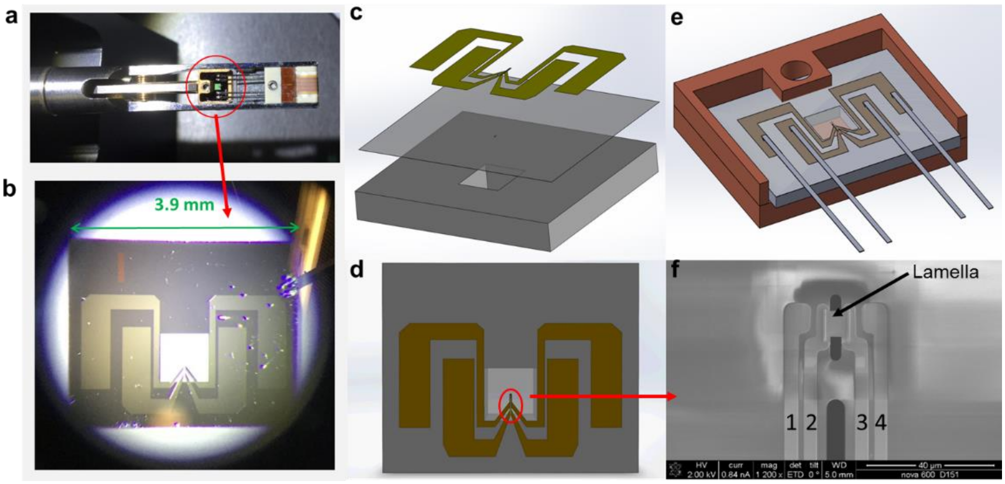

Figure 4. Device structure of the TEM lamella on a biasing holder for in situ current injection. a, DENS solutions double tilt TEM biasing holder. b, Magnified view of the biasing chip. c, Schematics of biasing chip consists of 200 nm Au contact pad (top) and 200 nm Si_3N_x membrane (middle) supported on 0.4 mm Si substrate (bottom). d, Schematic view of biasing chip. e, Schematic view of four terminal (connected to source meter/pulse generator) attached on the Au contact pad. f, Scanning electron microscopy (SEM) image of lamella placed on the Au contact pad. The numbers indicate four contact pads.

measuring the topological Nernst effect in thin films.

- **Compensated ferrimagnetic tetragonal Heusler compounds**

Moving the anti-skyrmions one-step forward, a long-term goal is the realization of a new class of skyrmions, known as antiferromagnetic skyrmions, which are tiny magnetic objects that actually have no net magnetic moment. Skyrmions in high-moment magnetic materials are generally too large for spintronics applications. While antiferromagnetic skyrmions are more difficult to observe, but have unprecedentedly small diameters and unique properties that make them very interesting for several future applications. Via compensated ferromagnetic inverse tetragonal Heusler compounds such as Mn3-xPtxGa [6,7] we might be able to tune anti-skyrmions into antiferromagnetic skyrmions. Mn2.4Pt0.6Ga is a compensated ferrimagnet which behaves like an artificial antiferromagnetic; small deviations from this composition leads to ferrimagnets with small magnetic moments.

- **In-situ current driven motion in LTEM**

Previously we have shown that, domain wall velocities of ~1,000 m/s can be achieved in racetracks that are formed from atomically engineered synthetic antiferromagnets for current densities of ~10^{12} A/m^2 [8]. Anti-skyrmions have significant potential to move with even higher velocities at the same current densities or similarly high domain wall velocities, but at lower current densities. For the potential application of aSks in spintronic devices, we would like to study the current-driven motion of aSk using LTEM. Moreover, aSks in this tetragonal Heusler alloy ($Mn_{1.4}Pt_{0.9}Pd_{0.1}Sn$) is stable beyond room temperature, and this makes them potential for room temperature experiments. For *in-situ* current injection we transfer the sample onto a DENS solutions double tilt sample holder (Fig. 4a). Corresponding samples consist of thin lamellae, which will be positioned on a biasing chip containing electrical contacts (Fig. 1b-f). Then the biasing chip together with the lamella can be mounted on the DENS solution holder for the experiment. For injecting current, we connect the TEM biasing holder with a pulse current generator that applies current pulses of different widths and amplitude. The parameters of the current pulse is adjusted so that we can avoid joule heating that can damage the lamella.

- **Preparation of high-quality of TEM lamella**

LTEM investigations of thin crystal plate hosting (anti)skyrmions largely depends on the quality of sample preparation. For the preparation of lamella, we have two state of the art Focused Ion Beam (FIB) systems, TESCAN GAIA 3 (Ga ion source) and TESCAN FERA 3 (Xe plasma source) which have been installed recently in the MPI Halle. The TESCAN GAIA 3 (Ga ion source) is best for surface analysis, microstructure analysis and TEM lamella preparation from a small structure (of the order of 10 microns). On the other hand, the Xe plasma source FIB column is capable of milling large structures (order of few hundreds of microns), a process which is either time-consuming or impossible with Ga ion FIB. We will use the Xe plasma source to prepare samples only in case to reduce the chance of surface amorphisation and Ga ion implantation. An additional advantage of the Xe plasma source is that it is inert, hence there is no chance of forming any intermetallic compound with the alloy. We have been optimising the various conditions for the preparation of high quality of wedge or ultra-thin lamella using Xe FIB.

Publications based on accomplished projects:

1. *A. K. Nayak, V. Kumar, T. Ma, P. Werner, E. Pippel, R. Sahoo, F. Damay, U. K. Rößler, C. Felser, and S. S. P. Parkin, Magnetic antiskyrmions above room temperature in tetragonal Heusler materials, Nature 548, 561 (2017).*

2. *R. Saha, A. K. Srivastava, T. Ma, J. Jena, P. Werner, V. Kumar, C. Felser, and S. S. P. Parkin, Enhanced stability of magnetic anti-skyrmions in tetragonal inverse Heusler compounds, (2018), Submitted.*

3. *K.G. Rana, O. Meshcheriakova, J. Kübler, B. Ernst, J. Karel, R. Hillebrand, E. Pippel, P. Werner, A.K. Nayak, C. Felser and S. S. P. Parkin, Observation of topological Hall effect in Mn2RhSn films, New J. Phys. 18, 085007 (2016).*

4. *O. Meshcheriakova, A. Köhler, S. Ouardi, Y. Kondo, T. Kubota, S. Chandra, J. Karel, C. V. Barbosa, R. Stinshoff, R. Sahoo, S. Ueda, Eiji Ikenaga, S. Mizukami, S. Chadov, D. Ebke, G. H. Fecher, and C. Felser, Structural, electronic, and magnetic properties of perpendicularly magnetised Mn2RhSn thin films, J. Phys. D: Appl. Phys. 48, 164008 (2015).*

5. *O. Meshcheriakova, S. Chadov, A. K. Nayak, U. K. Rößler, J. Kübler, G. André, A. A. Tsirlin, J. Kiss, S. Hausdorf, A. Kalache, W. Schnelle, M. Nicklas, and C. Felser, Large noncollinearity and spin reorientation in the novel Mn2RhSn Heusler magnet, Phys. Rev. Lett. 113, 087203 (2014).*

6. *R. Sahoo, L. Wollmann, S. Selle, T. Hoche, B. Ernst, A. Kalache, C. Shekhar, N. Kumar, S. Chadov, C. Felser, S. S. P. Parkin, and A. K. Nayak, Compensated ferrimagnetic tetragonal Heusler thin films for antiferromagnetic spintronics, Adv. Mater. 28, 8499 (2016).*

7. *Ajaya K. Nayak, Michael Nicklas, Stanislav Chadov, Panchanana Khuntia, Chandra Shekhar, Michael Baenitz, Yurii Skourski, Veerendra K. Guduru, Alessandro Puri, Uli Zeitler, Claudia Felser, Design of compensated ferrimagnetic Heusler alloys for giant tunable exchange bias, Nature Materials 14, 679 (2015) 679.*

8. *Yang, S.-H., Ryu, K.-S. & Parkin, S. S. P. Domain-wall velocities of up to 750 ms−1 driven by exchange-coupling torque in synthetic antiferromagnets. Nat. Nano. 10, 221-226, (2015).*