Skyrmions and hard magnets

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Magnetic skyrmions are topologically protected nanoscopic vortices of magnetization that can be stabilized in a magnetic field. Magnetocrystalline anisotropy (K_1) in uniaxial crystal structures is required to be strong enough to resist self-demagnetization in the shape of a thin lamella or film. We discovered antiskyrmions in Mn_{1.4}PtSn with uniaxial anisotropy and spin-reorientation transition at low temperature. Inspired by Mn_{1.4}PtSn, we further discovered new skyrmion bubble materials with similar features of perpendicular anisotropy and spin-reorientation in traditional hard magnetic materials, MnBi and Nd₂Fe₁₄B, which exhibit a large topological Hall effect at room temperature. However, spin-reorientation transition is unfavourable in hard magnets. To avoid this, new rare-earth-free hard magnetic materials with a strong K_1 at room temperature, including Rh₂CoSb and (Fe,Co)₂(P,Si), were discovered at Felser's department.

Background

Skyrmions can be potentially used as magnetic bits in high density storage devices, such as racetrack memories, and neuromorphic computing systems that operate beyond Moore's law. The bottleneck for skyrmion-based spintronics is the identification of novel materials that host dense skyrmions at ambient temperature and in low magnetic fields. Thus far, skyrmion lattices have been experimentally realized at low temperatures in various bulk cubic B20 transition metal compounds (such as MnSi) and Cu₂OSeO₃, a multiferroic insulator. Only β -Mn-type bulk cubic chiral Co₈Zn₈Mn₄ has been found to host skyrmions at ambient temperature.

The topological Hall effect (THE) is a key feature of skyrmions. For mesoscopic topological structures, such as skyrmions, THE is induced by the Berry curvature that originates from non-zero scalar spin chirality. By probing THE via magnetotransport measurements, we can gain insights into system that contain skyrmions without conducting expensive studies with neutron diffraction or low-temperature Lorentz transmission electron microscopy (L-TEM).

Accomplished projects

THE in Mn_{1.4}PtSn with antiskyrmions

Inverse Heusler $Mn_{1.4}$ PtSn is a hard magnet at room temperature whose moment is aligned along the *c*-axis. Below the spin-reorientation transition temperature of 135 K, the collinear spin becomes non-coplanar [1, 2]. A complex interrelation of ferromagnetic and antiferromagnetic exchange, combined with Dzyaloshinskii-Moriya interaction due to its D_{2d} symmetric structure, renders this compound a host of multiple magnetic textures, including antiskyrmions.

Based on THE [3], antiskyrmions were found in inverse Heusler $Mn_{1.4}$ PtSn at ambient temperature [4]. We report new insights from in-situ magnetotransport,

magneto-optical Kerr-effect and magnetic force microscopy studies, ferromagnetic resonance, and first-principle calculations on the intriguing magnetic properties of this unique compound. We directly detected THE contributions of antiskyrmions at room temperature in micron-sized devices, prepared from single crystals by focused ion beam patterning. We demonstrate the finite-size effect on its magnitude and reveal the peculiar temperature that depends on the magnetic properties of the host compound.

Skyrmion bubbles in traditional hard magnetic materials, MnBi and Nd₂Fe₁₄B

Uniaxial crystal structures that host skyrmions require strong magnetocrystalline anisotropy, K_1 , to resist selfdemagnetization in the shape of a thin lamella or film. All hard magnetic materials have strong enough K_1 and are good candidates to find skyrmion or skyrmion bubbles, depending on the symmetry. Therefore, we expect to find new hard magnetic materials to expand the implementation area for magnetic recording and hard magnetic applications. MnBi is known to be a hard magnet at room temperature with a large magnetic moment of 3.94 μ_B/Mn and high Curie temperature $(T_{\rm C} > 630 \text{ K})$; it has a magnetocrystalline anisotropy, K_1 , of approximately 1 MJm⁻³ at room temperature, which is sufficient to resist self-demagnetization into in-plane domains in the case of thin lamella and stabilize bubbles. Below the spin-orientation transition temperature $T_{SR1} = 137$ K, the magnetic moment gradually rotates away from the *c*-axis upon cooling until it abruptly locks into the *ab*-plane at $T_{SR2} = 87$ K.

Nd₂Fe₁₄B is another famous hard magnetic material; this magnet has the largest energy product. this magnet has the largest energy product. It has a high $T_{\rm C}$ of 588 K and large K_1 of 4.9 MJm⁻³ at room temperature, which is strong enough to resist self-demagnetization. The ferromagnetically coupled moments from Nd and Fe become non-collinear below the spin-reorientation



Fig. 1: Spin-reorientation transition, topological Hall effect, and skyrmion (bubble) for (a-c) $Mn_{1.4}PtSn$, (*d-f)* MnBi, and (*g-i*) $Nd_2Fe_{14}B$, respectively.

transition temperature, $T_{SR} = 135$ K, and the net moment is gradually tilted away from the *c*-axis, thus forming an easy-cone structure at low temperature. Most previous studies focused on hard magnetism in the bulk at room temperature. However, little attention has been paid to the mesoscopic-sized crystals or films, especially at low temperatures below T_{SR} . In mesoscopic-sized samples, such as thin lamellas, additional dipole-dipole interaction results in a stripe domain structure rather than the two-branched pattern in bulks. This stripe domain is the ground state for skyrmions or bubbles, which are stabilized by an applied magnetic field. The saturation magnetization of Nd₂Fe₁₄B at room temperature ($\mu_0 M_s = 1.6$ T) is the largest among all hard magnets, thereby providing a good opportunity for bubble memory with a small size [5].

Both MnBi and $Nd_2Fe_{14}B$ exhibit the same feature of $Mn_{1.4}PtSn$ of perpendicular anisotropy and spin-

reorientation transition, which are good candidates for skyrmions. We studied the domain structure of MnBi [6] and Nd₂Fe₁₄B [7] thin lamella by Kerr microscopy and L-TEM, respectively. Magnetic bubbles with a minimum diameter of approximately 100 nm were observed, accompanied with a large THE. New types of bubbles were first observed below T_{SR} in Nd₂Fe₁₄B. Magnetic bubbles result in THE, which depends on thickness and temperature.

New hard magnetic materials

It is noteworthy that spin-reorientation transition is unfavorable in hard magnets. The parallel alignment of ferromagnetic interaction with all moments along the c-axis is the key for a large magnetic moment and magnetocrystalline anisotropy. Therefore, to develop new hard magnetic materials, spin-reorientation transition should be avoided. Based on this concept, we developed two new hard magnetic materials, including Rh₂CoSb and Co, Si co-doped Fe₂P, which can be used in heat assisted magnetic recording (HAMR) for films and bulk magnets, respectively.

Rh₂CoSb for HAMR

The development of high-density magnetic recording media is limited by superparamagnetism in significantly small ferromagnetic crystals. Hard magnetic materials with strong perpendicular anisotropy offer stability and high recording density. HAMR was developed to overcome the difficulty of writing media with a large coercivity; in HAMR, the writing medium is rapidly heated to the Curie temperature, $T_{\rm C}$, before writing, followed by rapid cooling. Furthermore, it requires a suitable $T_{\rm C}$, coupled with anisotropic thermal conductivity and hard magnetic properties.

Rh₂CoSb was introduced as a new hard magnet with potential for thin film magnetic recording [8]; it includes a magnetocrystalline anisotropy of 3.6 MJm⁻³ with saturation magnetization of $\mu_0 M_s = 0.52$ T at 2 K (2.2 MJm⁻³ and 0.44 T at room temperature, respectively). It exhibits a magnetic hardness parameter, κ , of 3.7 at room temperature, which is the highest value observed for any rare-earth-free hard magnet.



Fig. 2: (a) Magnetization curves at 2 K and 300 K with the field along the a- or c-axes. The inset demonstrates a pattern of branched domains at the surface perpendicular to the c-axis, typical of a strong uniaxial ferromagnet. (b) Magnetocrystalline anisotropy calculated from magnetization curves at different temperatures. The blue line shows the magnetic hardness parameter, κ , at different temperatures. (c) Comparison of κ , with other hard magnets. The light grey plane marks the threshold, $\kappa = 1$.

Projects in progress

Rare-earth-free Fe₂P-based hard magnets

Permanent magnets are crucial components in modern science and technologies because they are applied abundantly in large-scale and emerging applications, such as hard disks, electronic devices, wind turbines, and electric vehicles. At present, the most used high performance permanent magnets include rare-earth metallic compounds of Sm-Co (SmCo₅ and Sm₂Co₁₇) and Nd₂Fe₁₄B. Despite their extraordinary properties, their applications are significantly limited by the high price of strategic rare-earth elements owing to limited resources. Commercial rare-earth-free alternatives include ferrites (e.g., BaFe₁₂O₁₉ or SrFe₁₂O₁₉) and alnico magnets, which are produced on a large scale. However, their maximum energy product, $(BH)_{max}$, is approximately only 40 kJ/m⁻³ due to either low M_s or small K_1 . Therefore, it is essential to find gap magnets with low cost to fill the gap between rare-earth and rare-earth-free magnets.

Because Fe is the most abundant 3d element on earth with the lowest price, it is desirable to find Fe-based rare-earth-free magnets. Pure iron is body-centered cubic, whose high-symmetric crystal structure is undesired for a large anisotropy. Hence, alloys or compounds with a uniaxial crystal structure, such as hexagon or tetragon, are preferred. The key aim involves determining "the partner" of iron. Generally, light elements are abundant on earth; therefore, they are economically friendly, especially for the first three periods. Intermetallic compounds consisting of iron and alkali (Li and Na) or alkaline-earth metal (Mg) do not exist. Fe-based compounds with halogen (F and Cl), chalcogen (O and S) are antiferromagnetic or ferrimagnetic, whose small moment is undesirable. Ferromagnetic compounds with the boron group (B and Al), carbon group (C and Si), and pnictogens (N and P) are ferromagnetic, which are potential candidates for hard magnets.

Among pnictides, Fe₂P simultaneously exhibits a large K_1 (2.32-2.68 MJm⁻³) and $\mu_0 M_s$ of 1.03 T. The value of K_1 is one of the largest in compounds without rare earths. In addition to its large K_1 , its large saturation magnetization ($\mu_0 M_s$) of 1.03 T makes it promising for hard magnetic applications.

We are currently working on Co- and Si-doped Fe₂P to improve the room temperature hard magnetic properties [9]. The theoretical value of $(BH)_{max}$ has been found to be 204 kJ/m⁻³, which is more than four



Fig. 3: Comparison of magnetic properties in rareearth-free materials with hardness parameter $\kappa > 1$ at 300 K.

times larger than that of commercially available ferrites (e.g., BaFe12O19 or SrFe12O19) and alnico magnets. This material combines the advantages of high performance and low cost and it is promising for commercial applications [10].

External Cooperation Partners

Our external cooperation partners include Bernd Rellinghaus (IFW Dresden, Germany) and Rudolf Schaefer (IFW Dresden, Germany).

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