Thin films and devices

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The nontrivial topology of magnetic quantum materials has come to the forefront of solid state research. These materials can provide a large Berry curvature through a suitable distribution of their topological Weyl nodes or nodal lines, and further novel physics with exotic transport responses. Heusler compounds show great potential due to their range of crystal structures and high chemical tunability. Our work focuses on the growth of thin-film topological materials, lithographic structuring of devices, and characterization of the resulting properties. We aim to push forward the frontiers of the field of topology, enhancing the fundamental insights into physical phenomena such as topological magnetism, and making progress towards the utilization of such materials in devices. Several new phenomena and devices are being studied, including materials that display unique magnetic textures, like noncollinear structures and antiskyrmions.

Magnetic topological Weyl semimetals

Magnetic topological semimetals are of great interest, along with their associated transport signatures for next-generation spintronic applications with low power consumption. Recently, only a few magnetic compounds, such as the kagome-lattice $Co_3Sn_2S_2$ and cubic Co_2MnGa, have shown a strong intrinsic anomalous Hall effect (AHE), observed as an additional Hall voltage transverse to the direction of charge-carriers flow in magnetically ordered materials. The intrinsic contribution to the AHE is due to the finite Berry curvature of the topological electronic structure. This topological electronic structure for the above-mentioned compounds consists of two entangled topological bands that are degenerate at the Weyl points position at the Fermi occupation level. Their linear dispersion near the Weyl points leads to dynamic and exotic transport properties, such as the AHE and its thermal counterpart, the anomalous Nernst effect (ANE).

To fully harness these transport phenomena arising from the topology in thin-film materials for future spintronic devices, a good understanding of their dependence on the film thickness is necessary. Within the thin-film limit, the properties of the surface play a significant role due to the intrinsic topology nested in the electronic structure. As a model system, we have grown high-quality epitaxial (001)-oriented Co_2MnGa films with varying thickness (10–80 nm) [1, 2]. These films show a combination of a large anomalous Hall conductivity (AHC) of 1138 S cm^{-1} and anomalous Hall angle of 13%, which are orders of magnitude larger than those of topologically trivial metals. Further, there is a good agreement between our experimental results (80 nm film) and bulk first-principles calculations of the AHE, which suggests that the experimentally observed AHE is primarily intrinsic (Fig. 1(a)). In thinner films, although the scattering introduced from the surfaces reduces the magnitude of the AHE, the overall trend does not change.

When a temperature gradient replaces the applied current, the ANE can be measured instead of the AHE, which originates from extrinsic and intrinsic mechanisms due to the electronic structure. We systematically studied the anomalous Nernst coefficient (ANC) for epitaxial Co_2MnGa films of different thickness, and found a strong variation with temperature [3]. On-chip thermometry techniques were used to quantify the thermal gradient during the thermoelectric measurements, enabling direct evaluation of the ANC. A 50-nm Co_2MnGa film exhibited a large ANE of -2 μV K^{-1} at 300 K (Fig. 1(b)), whereas a 10-nm film exhibited a significantly smaller ANC, despite having similar volume magnetization. These findings suggest that the microscopic origin of the ANE in Co_2MnGa is complex and may contain multiple contributions (skew-scattering, side-jump, intrinsic Berry curvature). Interestingly, the total ANE of Co_2MnGa is large compared that of other film materials.

Further, in collaboration with our topological transport theory group, we predicted that in the well-known $X$$Pt_3$ compounds (X = V, Mn, Cr), a new type of electronic structure simultaneously entangles multiple sets of bands, thereby increasing the AHE and ANE. We specifically focused on the electronic structure of VPt_3, $CrPt_3$, and $MnPt_3$ that harmoniously combine four topologically entangled electronic bands to produce much larger Berry curvature responses than in $Co_3Sn_2S_2$ and Co_2MnGa. The compounds seamlessly combine the physics of simple metals (e.g., Fe) and complex topological metals. We confirmed our theoretical predictions in thin films of $CrPt_3$ with an

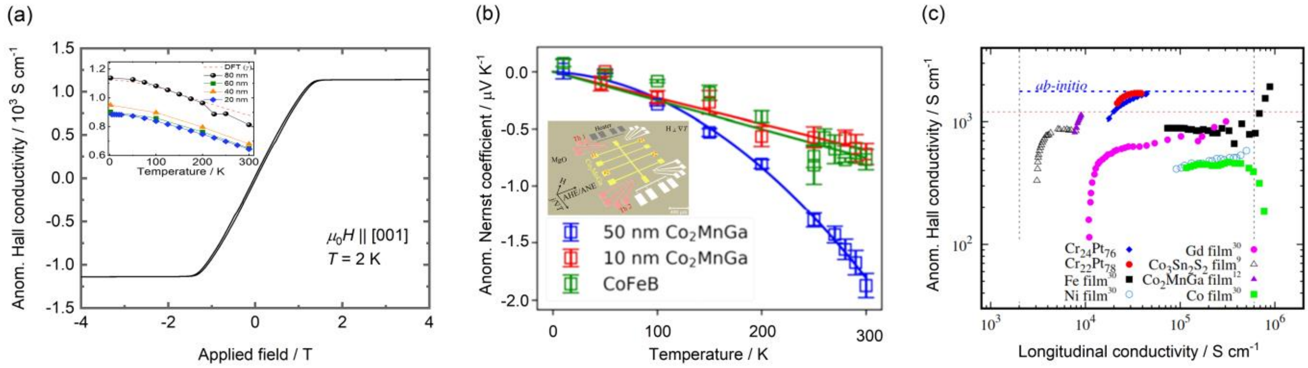

Fig. 1: (a) AHC of an 80-nm thick Co_2MnGa film at 2 K. The inset shows the temperature-dependent Hall conductivity of Co_2MnGa films with different thickness. The red dashed line represents the constant γ broadening from ab-initio calculations. (b) Temperature dependence of ANC for Co_2MnGa films with different thicknesses. The green curve corresponds to the ANC for the trivial ferromagnet CoFeB. The inset shows an optical microscopy image of the measurement device. (c) Log–log plot of the AHC as a function of the longitudinal temperature-dependent conductivity of experimental $CrPt_3$ films compared to well-known metallic and semi-metallic films. The dashed blue line shows the zero K ab-initio result for the AHC. The horizontal dashed red line shows the theoretical value for the topological kagome-lattice $Co_3Sn_2S_2$.

intrinsic AHE that lies far above those of thin films of both known topological semimetals and elemental metals [4].

We have grown high-quality $CrPt_3$ thin films with (111) orientation. Along this crystallographic direction, the films display perpendicular magnetic anisotropy with large coercivity and a high Curie temperature. In lithographically patterned Hall bar devices, we measured a robust AHC of 1750 S cm^{-1}, and fit the experimental data to determine the intrinsic Berry curvature contribution. The linear scaling of the impurity-density-independent terms was quite large, of the order of the theoretically calculated values. To further understand the underlying mechanisms in $CrPt_3$, a longitudinal conductivity (σ_{xx}) versus AHC (σ_{xy}) curve was plotted (Fig. 3(c)). Remarkably, the results for $CrPt_3$ films lie in the middle regime with a clear intrinsic AHC that reaches the upper echelons of known trivial and nontrivial materials published in the literature. The significant transverse magnetotransport response is due to the combination of two gapped nodal lines that synergistically work to generate Berry curvature.

Topological signatures of nontrivial magnetic textures in tetragonal inverse Heusler compounds

Magnetic materials that host topological spin textures are being studied as potential candidates for future spintronic applications. Of these spin textures, skyrmions and antiskyrmions are mesoscale whirling objects with distinct chiral magnetic boundaries and opposite topological charges. Of particular interest is the antiskyrmion (Fig. 2(a)), which was theoretically predicted to exist in certain tetragonal materials with acentric crystal structures with, e.g., D_{2d} symmetry. Recently, some tetragonal Heusler compounds (Fig. 2(b)) were shown to host antiskyrmions, in addition to other topological spin textures of interest. Spin chirality in metallic materials with noncoplanar spin structure gives rise to a Berry phase and induces a topological Hall effect (THE), which can be used to screen materials for magnetic textures useful for device applications.

We have grown high-quality epitaxial thin films of Mn_xPtSn. The Mn content (x) and thickness were tuned, which enables microscopic control of the magnetic exchange parameters that can be detected electrically with the THE [5, 6]. Specifically, we showed that the role of x is to tune the ratio of antiferromagnetic and ferromagnetic exchange interactions, which is crucial for the stabilization of antiskyrmions. Fine tuning of x can enable discrete adjustment of the antiskyrmions below the spin-reorientation temperature T_{SR} (Fig. 2(c)). This effect is observable from low temperatures up to ~ 200 K, depending on x. As expected, the magnetic dipole–dipole interactions play the largest role in determining the thin-film limit. We found that the THE changes substantially with x and the topological Berry curvature can be fine-tuned by modifying the film composition.

The topological origin of the observed effects can only be confirmed by the combination of magnetotransport and independent magnetization measurements. In collaboration with Goennenwein's group, we proposed an alternative method for determining the topological signatures, whereby the anomalous Hall and Nernst effects are measured for micropatterned thin films. For $Mn_{1.8}PtSn$ films (Fig. 2(d)), below $T_{SR} \approx 190$ K, a large THE and TNE of $S_{xy}^{T} = 115$ nV K^{-1} was measured in the same microstructure [7], which has a similar magnitude to that of bulk MnGe ($S_{xy}^{T} \approx 150$ nV K^{-1}), the only material to date for which a topological Nernst was reported. Our data serve as a model system to introduce a topological quantity, which enable the detection of the presence of topological transport effects without independent magnetometry data.

Further, we studied another member of the Heusler family with D_{2d} symmetry that hosts antiskyrmions with smaller magnetization compared to Mn_xPtSn. In collaboration with Parkin's group, we found two distinct peaks in the THE of Mn_2RhSn thin film grown by our group [8]. Using a phenomenological approach and electronic transport simulations, these THE features are considered direct signatures of two topologically distinct chiral spin objects, namely, skyrmions and antiskyrmions (Fig. 2(e)). THE studies allow us to determine the presence of these two topological objects over a wide range of temperatures and magnetic fields. In particular, it was observed that skyrmions are stable at low temperatures, suggesting an increased importance of dipolar interactions.

Noncollinear antiferromagnetic Mn_3Sn films

Noncollinear antiferromagnets exhibit novel Berry-curvature-driven phenomena that are of interest to the cutting-edge field of topological spintronics. In particular, bulk samples of hexagonal Mn_3Sn have been shown to exhibit room-temperature AHE and ANE related to Weyl points. Prior to the implementation of these materials into antiferromagnetic spintronic applications, there is the need for a deeper exploration of the topologically driven physics of Mn_3Sn in thin-film form.

We have grown epitaxial thin films of Mn_3Sn with both (0001) *c*-axis orientation and $(40\bar{4}3)$ texture [9,10]. Magnetometry measurements of the resulting thin-film magnetic properties revealed a small uncompensated magnetization, which arises from the Mn spins canting towards the magnetocrystalline easy axes. The Mn spins can be reversed by applying an external magnetic field, which also reverses the chirality of the inverse triangular spin texture. We further investigated this uncompensated magnetic moment using X-ray magnetic circular dichroism (XMCD), and used XMCD sum rules to deconvolute the spin and orbital components. The small uncompensated moment cannot account for the large AHC simultaneously observed in $(40\bar{4}3)$ textured films with different

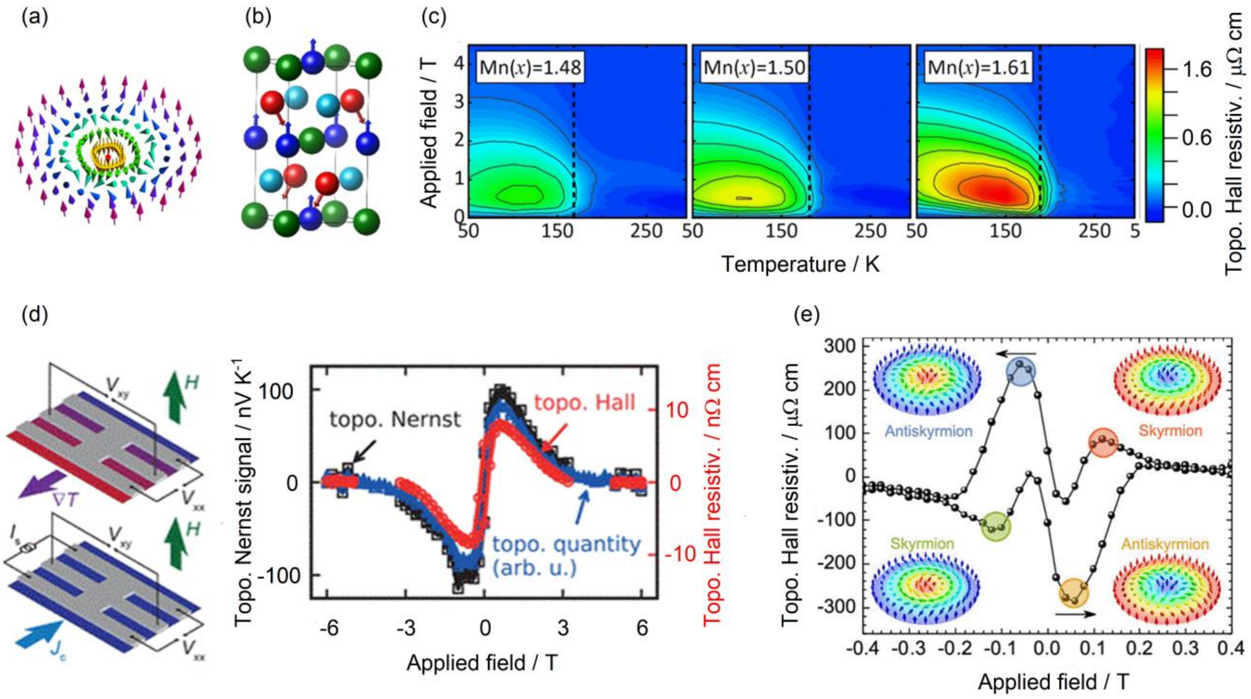

Fig. 2: (a) Spin texture of an antiskyrmion. (b) Tetragonal Heusler structure with D_{2d} symmetry. (c) Topological Hall resistivity as a function of temperature and applied magnetic field for Mn_xPtSn (x = 1.48–1.61) films with a thickness of 38 nm. (d) Contact geometry for the Nernst (top panel) and Hall measurements (bottom panel) of a patterned $Mn_{1.8}PtSn$ film. The topological quantity (blue triangles) indicates the magnetic-field dependence of the TNE (black squares) and THE (red circles). (e) Topological Hall resistivity of a Mn_2RhSn film, where the two peaks per hysteresis branch correspond to two distinct magnetic phases with the same topological charge.

thickness, even at room temperature, with a magnitude σ_{xy} ($\mu_0 H$ = 0 T) = 21 S cm^{-1} (Fig. 3(a)). We attribute the origin of this AHE to momentum-space Berry curvature arising from the symmetry-breaking inverse triangular spin structure of Mn_3Sn.

In addition, a low-temperature THE was observed in Mn_3Sn thin films, which is thought to arise from either chiral domain walls or magnetic skyrmions. As shown in Fig. 3(b) the sign of this THE, and thus the handedness of the non-trivial magnetic structure driving it, can be controlled by the field cooling conditions, which provides new strategies for manipulating the chiral spin texture in thin films.

In collaboration with Goennenwein's group, we demonstrated that the magnetic structure in lithographically patterned devices based on epitaxial Mn_3Sn (0001)-oriented thin films can be spatially mapped [11]. We used a magnetic imaging technique based on a laser-induced local thermal gradient combined with detection of the ANE (Fig. 3(c)). Fig. 3(d,e) show that the observed contrast is of magnetic origin. At room temperature and below, the magnetic structure is insensitive to magnetic fields up to 6 T. We further showed that domains can be written into the magnetic structure by local heating in combination with moderate magnetic fields (± 0.5 T).

Outlook

Spin-to-charge conversion is key to the realization of spintronic devices. Therefore, we are motivated to pursue research in novel topological materials (TMs) that promise large spin-charge conversion through intrinsic mechanisms arising from topology, Weyl physics, or crystal structure symmetry-breaking. Examples include (magnetic) topological insulators (TIs) with spin-momentum-locked surface states, magnetic Weyl semimetals (WSMs) in the Heusler family with Weyl nodes or nodal lines, noncollinear antiferromagnets, and large Fermi-arc chiral B20 compounds. Using the resources available within our institute, we plan to quantify the spin Hall effect (SHE) using the spin pumping technique. Along with our collaborators, we can measure the SHE via magnetotransport techniques, such as spin torque ferromagnetic resonance. Further, we plan to study terahertz (THz) emission from WSM/Pt bilayers with Kampfrath's group, where a laser-induced spin current

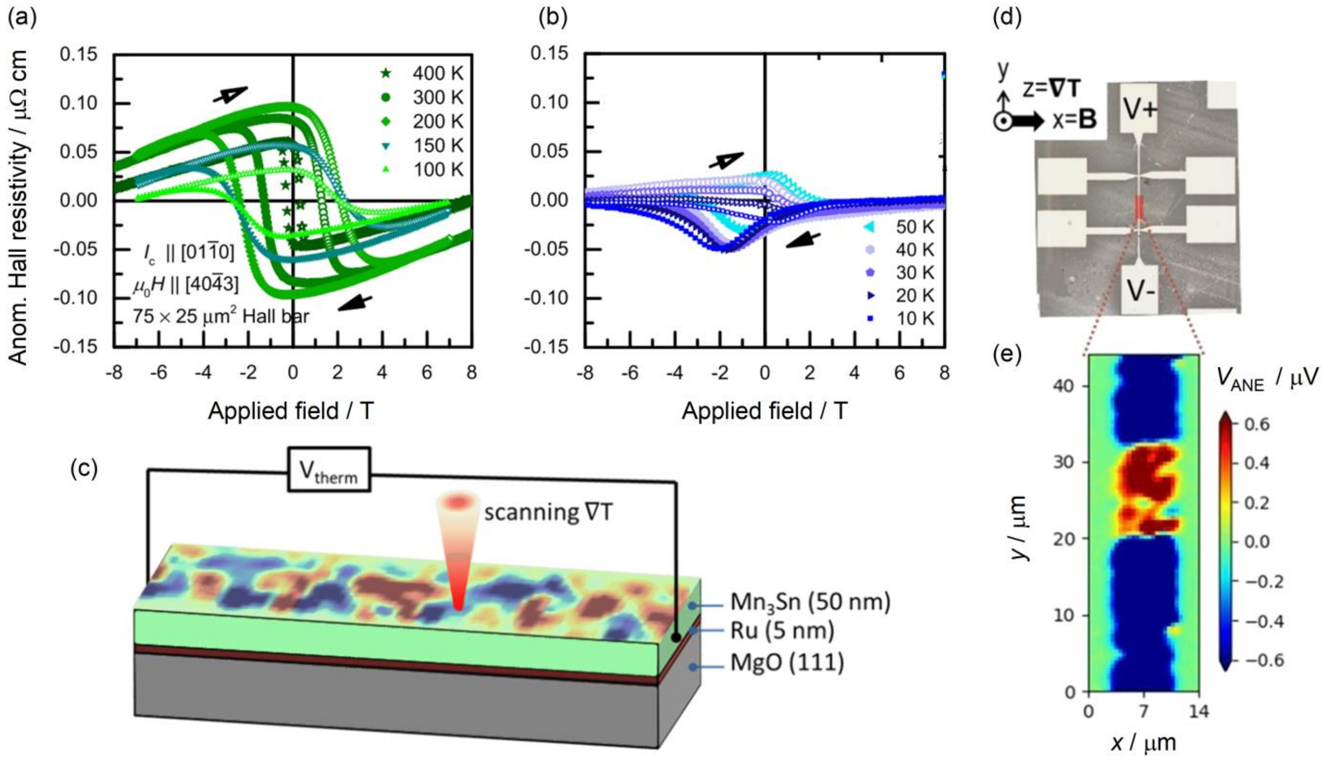

Fig. 3: (a) Isothermal Hall resistivity measured as a function of magnetic field applied out of plane in the (a) higher-temperature and (b) lower-temperature regimes. In the lower-temperature regime the THE appears. (c) Schematics of a scanning thermal gradient microscopy set-up. The laser beam is scanned over the sample surface and the resulting local thermo-voltage sign and magnitude reflect the local magnetic properties. (d) Optical microscopy image of a typical device and the experimental geometry. (e) Spatially resolved thermo-voltage obtained in the region indicated by the red rectangle, showing a clear magnetic contrast.

is injected from the WSM into the Pt layer and consequently converted into a THz pulse via the spin-orbit interaction inside Pt.

A topic we are particularly interested in is the combination of different quantum materials, e.g., topological materials (insulator, semimetal, metals)/ (anti)skyrmion, ferroelectric/(anti)skyrmion. We aim to understand the heterostructures with high-quality interfaces such as new properties, large SHE and spin-transfer torque. We will focus on the cross-fertilization between different topological classes of materials to realize devices with intertwined electronic spin and topology. In particular, we aim to combine two topological classes, one existing in real space associated with topological solitons, and a second defined in the reciprocal space associated with TM, such as TI and WSM. For this, we will use the intrinsically high spin-polarized states naturally arising from spin-to-momentum locking in such TMs. This will enable the formation of spin-currents from charge-currents via the SHE. A significantly larger spin Hall angle can be expected from TMs compared to common heavy metals, such as Pt, Ta, and W. We will demonstrate the current-induced magnetization switching of the skyrmionic layer by the spin-orbit torque (SOT) originating from current flowing in the TM layer. Such functional heterostructures can be further used for SOT nucleation and manipulation of skyrmions.

We plan to study the doping-dependent evolution of the electronic band structure using electrical transport in $Sb_{2-x}V_xTe_3$ thin films prepared by molecular beam epitaxy. The tunability by doping and the coexistence of the surface states with ferromagnetism make $Sb_{2-x}V_xTe_3$ thin films a promising platform for energy-band engineering. In this way, topological quantum states may be manipulated to transition from the quantum Hall effect to quantum anomalous Hall effect. We anticipate that combining such devices with conventional superconductors may give rise to Majorana fermions, which are suitable for braiding devices for use in topological quantum computers.

External Cooperation Partners

Stuart S. P. Parkin (Max Planck Institute of Microstructure Physics); Sebastian T. B. Goennenwein (Universität Konstanz); Jacob Gayles (University of South Florida); Tobias Kampfrath (Freie Universität Berlin).

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