Role of magnetic exchange interactions in chiral-type Hall effects of epitaxial Mn\textsubscript{x}PtSn films

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ABSTRACT

Tetragonal Mn-based Heusler compounds feature rich exchange interactions and exotic topological magnetic textures, such as antiskyrmions, complimented by the chiral-type Hall effects. This makes the material class interesting for device applications. We report the relation of the magnetic exchange interactions to the thickness and Mn concentration of Mn\textsubscript{x}PtSn thin films, grown by magnetron sputtering. The competition of the magnetic exchange interactions determines the finite temperature magnetic texture and thereby the chiral-type Hall effects in external magnetic fields. We investigate the magnetic and transport properties as a function of magnetic field and temperature. We focus on the anomalous and chiral-type Hall effects and the behavior of the dc-magnetization, in relation to chiral spin textures. We further determine the stable crystal phase for a relative Mn concentration between 1.5 and 1.85 in the \textit{I\textsubscript{4}2d} structure. We observe a spin-reorientation transition in all compounds studied, which is due to the competition of exchange interactions on different Mn sublattices. We discuss our results in terms of exchange interactions and compare them with theoretical atomistic spin calculations.

Introduction

Magnetic spin textures are of vital interest for technological applications due to the ease of manipulation via external electromagnetic fields\textsuperscript{1,2} and detection purely through electrical means\textsuperscript{3}. One such method for the detection for complex spin textures arises from the Hall effects, i.e. transverse electric fields arise in response to an applied external electric field. The well-known ordinary (OHE) and anomalous (AHE) Hall effects scale with external magnetic field and the alignment of magnetic domains in the absence of a magnetic field, respectively\textsuperscript{4}. In addition, there are transport effects that are independent of the external magnetic field and net magnetization, which we group into the chiral-type Hall effects\textsuperscript{5–8}. These chiral-type Hall effects are experimentally determined by the subtraction of the AHE and OHE from the total Hall effect\textsuperscript{9} and are related to the spatial variation of the local magnetic lattice in real space\textsuperscript{3}. The relative spatial variation gives rise to an emergent electromagnetic field, termed the Berry curvature, which is finite for localized magnetic textures with spin chirality. Electromagnetic fields couple to those spin textures through the conduction electrons and in turn the ground state electronic properties\textsuperscript{10}.

The ground state electronic properties determine the magnetic exchange interactions necessary to stabilize spin textures at finite temperatures and magnetic fields. These exchange interactions arise in a hierarchical competition of energy scales: the symmetric Heisenberg exchange which is nominally the largest and determines the ordering temperature; the anti-symmetric Dzyaloshinskii-Moriya interaction (DMI) attributed to the absence of inversion symmetry and strong spin-orbit coupling (SOC)\textsuperscript{11}; the dipole-dipole interactions; the magnetocrystalline anisotropy. The competition of these interactions can lead to magnetic textures that are noncoplanar with a finite chiral Berry curvature. One example is the skyrmion crystal arising in a competition of the Heisenberg exchange with the DMI, found in B20 compounds\textsuperscript{9,12–15}, or the antiskyrmion in inverse tetragonal Heusler compounds\textsuperscript{16}. The Hall effect from such magnetic textures is nominally the topological Hall effect (THE) since it can be directly related to the topological winding of the skyrmion lattice\textsuperscript{3}. Systems with electron correlation can also show this type of response, which can sometimes be associated with magnetic domains and bubbles\textsuperscript{17}. Lastly, there are the noncollinear spin arrangements that can originate in the competition of ferromagnetic and antiferromagnetic exchange interactions between neighboring spins\textsuperscript{18}.

The class of tetragonal inverse Mn-based Heusler compounds (\textit{Mn}\textsubscript{X}YZ, \textit{Y} is a transition metal and \textit{Z} a main-group element), associated with a broken inversion symmetry (\textit{D}\textsubscript{2h}) that enables the stabilization of noncoplanar spin textures, received significant attention following the observation of antiskyrmions with an anisotropic winding over a broad temperature and field
The Mn concentration has a profound effect on the competition of the Heisenberg exchange interactions augmented plane-wave method within the generalized gradient approximation (GGA) exchange correlation (XC) functional.

We carried out density functional theory (DFT) calculations using the full potential linearized First principle calculations. MgO[010]) were recorded simultaneously. In order to determine the longitudinal and the Hall resistivity, Lithography and electrical transport measurements. Magnetization measurements were performed on a SQUID vibrating sample magnetometer (MPMS 3, Quantum Design). In order to infer the magnetization of the films, we subtracted the diamagnetic substrate contribution as well as a low-temperature paramagnetic contribution from the raw data. Here, the paramagnetic contribution is attributed to impurities in the MgO substrate. The diamagnetic susceptibility of MgO was estimated from the high field slope at contribution as well as a low-temperature paramagnetic contribution from the raw data. Here, the paramagnetic contribution is attributed to impurities in the MgO substrate. The diamagnetic susceptibility of MgO was estimated from the high field slope at

The stoichiometries of the films were confirmed by energy-dispersive x-ray spectroscopy (EDXS), with an experimental uncertainty of 5 at.%. Structural characterization was carried out on a PANalytical X’Pert PRO x-ray diffractometry (XRD) system employing Cu-Kα radiation (λ = 1.5406 Å). The film thicknesses were determined by x-ray reflectivity (XRR) measurements.

Hard x-ray photoelectron spectroscopy. The experiments were performed at beamline P22 of PETRA III (Hamburg, Germany) using linearly polarized photons with an energy of hv = 5226.1 eV for excitation. Horizontal (p) polarization was obtained directly from the undulator. Grazing incidence (α = 89°) – normal emission (θ = 1°) geometry was used ensuring a polarization vector nearly parallel to the surface normal. The energy resolution was set to 250 meV, verified by spectra of the Au valence band at the Fermi energy EF. The spectra were acquired using a Phoibos 225HV photoelectron spectrometer with a hemispherical analyzer, delayline detector and wide angle lens (SPECS, Berlin). The spectra are not influenced by the 3 nm thick Si capping layer, due to the large electron mean free path and low cross section of its valence states.

DC magnetometry measurements. Magnetization measurements were performed on a SQUID vibrating sample magnetometer (MPMS 3, Quantum Design). In order to infer the magnetization of the films, we subtracted the diamagnetic substrate contribution as well as a low-temperature paramagnetic contribution from the raw data. Here, the paramagnetic contribution is attributed to impurities in the MgO substrate. The diamagnetic susceptibility of MgO was estimated from the high field slope at 300 K. The paramagnetic contribution was fitted and subtracted from the raw data using the Brillouin function.

Lithography and electrical transport measurements. In order to determine the longitudinal and the Hall resistivity, the thin films were patterned into 8-contact Hall-bar devices (width = 50 µm; length = 250 µm) using optical lithography and subsequent Ar ion etching (Fig. 2b). The electrical transport measurements were conducted on a physical properties measurement system (PPMS, Quantum Design) with fields up to 7 T applied along the out-of-plane (z) direction (MgO[001]). An in-plane current, Ix = 1 mA (along MgO[100]), was applied along the Hall-bar while the voltages along the current direction (Vxx, x-direction corresponding to MgO[100]) and perpendicular to the current direction (Vxy, y-direction corresponding to MgO[010]) were recorded simultaneously.

First principle calculations. We carried out density functional theory (DFT) calculations using the full potential linearized augmented plane-wave method within the generalized gradient approximation (GGA) exchange correlation (XC) functional. We converged the unit cell with 12x12x8 k-point mesh and 4.0 bohr⁻¹ cutoff radius for a spin full calculation. Spin spiral
Symmetric radial ($\omega - 2\theta$) x-ray diffractometry scans are employed to determine phase purity, verify the previously reported structure, and investigate changes in the lattice parameters. We show in Fig. 1a the $\omega - 2\theta$ scans of 110 nm thick Mn$_x$PtSn films at various Mn($x$). Between Mn($x$)=1.5 and Mn($x$)=1.85 all films grow epitaxially without any additional phases. In this range, we observe only the (h00) Bragg reflections, attributed to the Mn$_x$PtSn film with the structure as mentioned above. Above Mn($x$)=1.85 a shoulder forms at the (200) peak at around 27.0°, while below Mn($x$)=1.5 additional peaks start to appear at around 40.2° and 60.3°. The out-of-plane $a$-lattice parameter and the unit cell volume increase with increasing Mn($x$), depending sensitively on the stoichiometry up to Mn($x$)=1.6 and followed by saturation towards Mn($x$)=1.85 (see Fig. S3).

Figure 1b depicts the $\omega - \theta$ scans of the film thickness dependence at Mn($x$)=1.6. Here, we confirm phase purity and epitaxial growth for all thicknesses. The bulk values for the lattice parameters ($a=6.3651$ Å, $c=12.2205$ Å for Mn$_{1.44}$PtSn) suggest a lattice mismatch with the MgO substrate of 6.4% and 2.5% along the two distinct in-plane directions of Mn$_x$PtSn. This results in an in-plane compressive strain and in turn an elongation of the out-of-plane lattice parameter ($a$). Upon relaxation of the compressive strain with increased $t$, the $a$-lattice parameters, therefore, decrease towards the bulk limit (see Fig. S3). Further information on the structure can be found in the supplemental material.

### Photoemission
We investigate the core levels and the valence bands of Mn$_x$PtSn films by hard x-ray photoelectron spectroscopy (HAXPES). An analysis of the peak heights of the Mn 2$p$, Pt 4$d$, and Sn 3$d$ states (not shown here) confirms the trend of the relative Mn($x$) between the films. Fig. 1c compares the Mn 2$p$ photoelectron spectra of Mn($x$)=1.48 and Mn($x$)=1.76. The 2$p$ states exhibit a clear spin-orbit splitting of 12.4 eV, independent of the composition. Further, the spectra are dominated by an exchange splitting due to an interaction between the 2$p$ core hole with the valence 3$d$ states resulting in a pronounced multiplet spectrum, well known for Mn$_32$. Since the Mn atoms carry a localized magnetic moment related to the magnetic splitting of the 3$d$ states, the multiplet structure of the Mn 2$p$ state reflects the magnetic state. It is seen from the inset in Fig. 1c that the multiplet splitting at the Mn 2$p_{3/2}$ derived peak is weaker at a higher Mn($x$).

### Magnetic spin reorientation
The magnetic structure of Mn$_x$PtSn displays interesting magnetic transitions as a function of temperature and external field$^{19,27}$. In the absence of an external magnetic field and as the temperature decreases from the paramagnetic state, there is a transition...
Fig. 2. (a) Crystal structure of Mn$_x$PtSn. (b) Magnetic spin texture prior and after field polarization at $\mu_0 H_s$. (c) Optical micrograph of the Hall-bar device. The black scalebar is 200 $\mu$m wide. (d) Zero field longitudinal resistivity of a 110 nm thick Mn$_{1.61}$PtSn film as a function of temperature. The dashed line at $T_s = 195$ K indicates the spin reorientation transition temperature, while the dashed line at $T_C = 377$ K indicates the Curie temperature. (e)-(i) 1$^st$ quadrant of the measured Hall data for 110 nm thick Mn$_x$PtSn films with varying Mn($x$) at 300 K, 200 K, 175 K, 150 K and 50 K, respectively.

to a collinear state at the Curie temperature ($T_C$). Further decreasing the temperature leads to a first-order transition, from the collinear to a noncoplanar magnetic state, at the spin reorientation transition temperature ($T_s$) $^{24}$. In Fig. 2b, we show a schematic of the spin reorientation as a function of an external magnetic field below $T_s$. There are two distinct magnetic sublattices of the Mn ions (Mn$_{I-4a}$ and Mn$_{II-8d}$) shown in gold and in red in Fig. 2a-b. In the demagnetized state, the localized spins within the unit cell form a noncoplanar structure. As an external magnetic field is applied, the magnetic domains and the noncoplanar spin texture orient along the direction of the applied magnetic field. The increase of the external field allows for a spin reorientation at $\mu_0 H_s$, where the spins show a second-order transition from the noncoplanar arrangement into a collinear ferromagnetic alignment. These transitions are evident in the resultant transport phenomena in the presence of an electric field $^{27}$ and in $dM/dH$.

**Magnetotransport**

In Fig. 2c, we show the lithographically patterned 8-contact Hall-bar device. We measure the longitudinal resistivity ($\rho_{xx}$) in the absence of an external field as a function of temperature (Fig. 2d). Here, the slope changes at $T = 195$ and 377 K indicate $T_s$ and $T_C$, respectively. The magnetic structure adds linearly to the other resistivity sources (e.g., phonons and impurities) due to Matthiessen’s rule. Above $T_s$ the slope of $\rho_{xx}(T)$ decreases as compared to the noncoplanar configuration, where the magnetoresistance decreases due to the collinear spin arrangement. The residual resistivity ratio (RRR) of our samples ranges from 1.58-2.79 as a function of thickness and Mn($x$)(see Fig. S8). The chiral textures also further contributes to the Hall effects, due to the associated Berry curvature$^4$. Hence, the total Hall resistivity can be described as$^{4,27,33}$,

$$\rho_{xy} = \rho_{xy}^{\text{OHE}} + \rho_{xy}^{\text{AHE}} + \rho_{xy}^{\text{THE}}.$$  

(1)

Equation (1) comprises, first, the ordinary Hall resistivity ($\rho_{xy}^{\text{OHE}} = R_0 H$) with the scaling parameter $R_0$ as the Hall constant, depending on the carrier density. Secondly, the anomalous Hall resistivity ($\rho_{xy}^{\text{AHE}} = R_SM$) associated with the spontaneous magnetization and the scaling parameter $R_S$ that originates from the scattering mechanisms and the intrinsic Berry curvature. Lastly, the topological Hall resistivity ($\rho_{xy}^{\text{THE}}$) that originates from the chiral magnetic texture and is independent of the net magnetization. Therefore, it is straight-forward to extract $\rho_{xy}^{\text{THE}}$ by subtracting the contributions to $\rho_{xy}$ that scale with the external field and the saturation of magnetization$^{4,9,27}$ (see Fig. S9).

We show the total Hall resistivities in Fig. 2e-i measured on the micro-patterned devices under the application of an external magnetic field and as a function of Mn($x$) and temperature. At 300 K and 200 K, $\rho_{xy}$ portrays the typical behavior for a
Fig. 3. (a)-(c) $M(H)$ loops and (d)-(f) magnetic field dependence of $dM/dH$ calculated from the dc-magnetization. The field is applied along the film normal at 300 K, $T_s$ and 50 K, respectively, for Mn($x$)=1.48 and Mn($x$)=1.71 films from the 110 nm Mn($x$) series and $t=20$ and 100 nm films from the Mn($x$)=1.6 thickness series. Squares and circles in (f) indicate the magnetic transitions. Insets right: $M(H)$ loops and $dM/dH$ with the field applied in the film plane. Inset top (c): Enlarged area around 0 T. Inset left (f): Magnetic spin texture below and above $\mu_0H_s$.

ferromagnet following a saturating magnetization in an external magnetic field (Fig. 2e-f). We determine $\rho_{AHE}$ as the linear extrapolation of the resistivity above 2.5 T to zero field. The size of $\rho_{AHE}$ varies with concentration related to the electron occupation determining the Berry curvature and the sources of scattering centers (see Fig. S10). In the graphs, we observe the largest $\rho_{AHE}$ for Mn($x$)=1.61. Upon further decreasing the temperature (Fig. 2g-i), we see a nonlinear, hump-like, behavior in $\rho_{xy}$ in the range of 0.5-2.5 T and a more considerable hysteresis$^{19,22,27}$. At 175 K, in Fig. 2g, the hump-like feature is seen for all Mn($x$) except Mn($x$)=1.48, where it appears at 150 K. The overall magnitude of $\rho_{AHE}$ decreases with the decrease in temperature, as expected from the reduction in scattering mechanisms. For $\rho_{THE}$ the following trend is apparent: zero above $T_s$; finite around $T_s$; maximal for temperatures below $T_s$. Further, there is a unique dependence of $\rho_{xy}$ on Mn($x$), which can only point to the magnetic interactions that determine the magnetic ordering of the system.

DC magnetic susceptibility
Changes in the magnetic texture can be inferred from the static magnetic susceptibility ($dM/dH$)$^{13,14,34}$. Hence, we perform dc-magnetometry measurements in order to extract the magnetization of the unpatterned samples. In Fig. 3a-c we show the out-of-plane (OOP) magnetizations as a function of an externally applied field ($M(H)$ loops) at 300 K, $T_s$ and 50 K. The insets on the lower right are the in-plane (IP) $M(H)$ loops for the respective films. We show two samples with Mn($x$)=1.48 and Mn($x$)=1.71 from the 110 nm Mn($x$) series as well as two samples at $t=20$ and 100 nm from the Mn($x$)=1.6 thickness series. The upper right inset of Fig. 3c depicts a small hysteresis pocket around 0 T. The OOP $M(H)$ loops are reminiscent of hard-axis behavior with a small coercive field. The IP $M(H)$ loops display that the magnetic easy-axis lies within the film plane due to the anisotropy of the system and in agreement with the tetragonal $c$-axis orientation. Both, the coercive fields and the saturation magnetization increase with decreasing temperature.

In Fig. 3d-f we plot the OOP $dM/dH$ as a function of the externally applied field. The static magnetic susceptibility is calculated from the numerical derivative of the experimental magnetization. The insets show the IP $dM/dH$. At 300 K (Fig. 3d), the demagnetized textures rapidly converge to a saturated state at increasing fields of about 1 T. Below $T_s$ (Fig. 3f) there are two transitions, close to 0 T (open circles) and ~2.5 T (open square), both determined from peaks in the second
Fig. 4. (a) Density of states as a function of the Fermi energy for Mn$_2$PtSn (grey dashed), Mn$_{1.5}$PtSn (solid red), Mn$_{1.6}$Pt$_0.9$Sn (solid blue), and Mn$_{1.6}$PtSn$_{0.9}$ (solid black). Positive (negative) DOS are the majority (minority) projected states. Inset: Enlarged area around the Fermi energy. (b) The band-structure of Mn$_2$PtSn around the Fermi energy. (c) The AHE calculated as a function of the Fermi energy for Mn$_2$PtSn (grey dashed) and Mn$_{1.5}$PtSn (solid red). The points show the experimental anomalous Hall conductivities. (d) Calculated $M(H)$ loops for Mn$_{1.5}$PtSn with the out of plane magnetic field above and below the spin reorientation. The insets show the magnetization for in plane magnetic field. (e) $dM/dH$ derived from the $M(H)$ loops.

derivative of the magnetization with respect to the external magnetic field. In this range, the magnetic domains begin to align along the field direction. The noncoplanar magnetic texture is dominant where $dM/dH$ slope is decreased compared to the slope close to 0 T. For fields larger than $\mu_0H_s$, the magnetic structure is collinear and $dM/dH$ is a constant zero. The IP $dM/dH$ (Fig. 3f (top inset)) shows two peaks that are symmetric around 0 T, which is due to the square loop hysteresis. In the spin reorientation(Fig. 3e), $dM/dH$ is a mixture of the two states above and below $T_s$. Further, the hysteresis displayed in the inset of Fig. 3c, does not belong to the magneto-crystalline anisotropy, but to the energy required to orient noncoplanar spins with respect to one another$^{13,35}$. This is a first-order transition highlighted by the circles close to 0 T ($\mu_0H_{nc}$). The smooth second-ordered transition from the noncoplanar to the collinear spin arrangement is shown as schematic insets (Fig. 3f). These effects originate in the magnetic exchange interactions, specifically, the competition between antiferromagnetic and ferromagnetic interactions on the Mn sublattices.

**First principle calculations**

We carry out first-principle calculations on Mn$_{1.5}$PtSn within the FLAPW method using the FLEUR code$^{30}$. Here, we find that the moments on the Mn$_I$ and Mn$_II$ sublattice are 3.72 and 3.4 $\mu_B$, respectively$^{19}$. Due to the hybridization, there is a small moment induced on the Pt and Sn sites of 0.3 and 0.1 $\mu_B$, respectively. We allow the directions of the moments to relax with SOC and find the angle between the Mn$_I$ and Mn$_II$ sublattices to be 52°. This shows that the ground state is a noncoplanar spin structure. The collinear ferrimagnetic (FiM) structure has an energy that is 60 meV larger while the collinear ferromagnetic state is 140 meV larger.

Upon substitution of Mn onto a Pt-8c (Sn-8d) site, the symmetry of the system is lowered, and the average canting angle of the Mn sublattices is decreased (increased) to 47° (56°). In Fig. 4a, we plot the spin decomposed density of states (DOS) for the ferrimagnetic (dashed), ferromagnetic (solid black), a single Pt substituted by Mn, and a single Sn substituted by Mn per unit cell. The positive values are for the majority spins, and the negative for the minority spins. For the Mn substituted onto a Pt or Sn site, the Mn prefers to align antiferromagnetically to the net moment, thereby decreasing the total moment. For the substitution at the Sn site the DOS at the Fermi energy decreases. $\Delta$ denotes the exchange field generated by each Mn sublattice ($\perp$). In Fig. 4b we plot the bands of Mn$_2$PtSn, which are significantly less complicated than that of Mn$_{1.5}$PtSn (see Fig. S7). We calculate the anomalous Hall conductivity (AHC) for the collinear ferromagnetic structures where we assume the external magnetic field has aligned the moments of Mn$_{II}$-8d and Mn$_I$-4a along the $a$-axis. In the Mn$_2$PtSn we find an AHC.
of 95 S/cm (273 S/cm) along the b-axis (c-axis) and in \( \text{Mn}_1.4\text{PtSn} \) we find an AHC of 155 S/cm (232 S/cm) along the b-axis (c-axis). Along with that, we plot the AHC as a function of energy for \( \text{Mn}_2\text{PtSn} \) and \( \text{Mn}_1.3\text{PtSn} \) for the collinear magnetic spin texture along with the experimental anomalous Hall conductivities (Fig. 4c).

**Atomistic spin calculations**

The origin of the noncollinear magnetic textures is due to a competition of classical exchange interactions between the inter-site interactions of \( \text{Mn}_4\text{PtSn} \) and \( \text{Mn}_1\text{PtSn} \) and the intra-site interactions. This can be modeled by a classical exchange Hamiltonian \( \mathcal{H} = -J_1 \sum_{i>j} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{i<j} \mathbf{S}_i \cdot \mathbf{S}_j + J_3 \sum_{i>j} \mathbf{S}_i \cdot \mathbf{S}_j \). Here, \( \mathbf{s} \) is the unit vector of spin for each sublattice, \( J_1 \) is the inter sublattice interaction, \( J_2 \) and \( J_3 \) are the \( \text{Mn}_4 \text{PtSn} \) and \( \text{Mn}_1\text{PtSn} \) intra-site interactions for each sublattice, respectively. All interactions together are necessary for a noncollinear spin texture. The positive \( J_\text{MnPtSn} \) and \( J_\text{Mn1PtSn} \) directions and an open boundary in the a crystal axis. In Fig. 4d we plot the \( M(H) \) loops, below and above the spin reorientation for a magnetic field along the a-axis. The inset shows the \( M(H) \) loops for the field along the c-axis. Fig. 4e shows the \( dM/dH \) curves of the theoretical calculations. Similar to the experimental curves, the theoretical \( dM/dH \) shows the transition field, \( \mu_0H_c \) (open square), of the magnetic texture. Below \( \mu_0H_c \) the magnetic textures induce a change in the Berry curvature, thereby adding a chiral product (\( \chi \)) contribution to the Hall effect.

**Chiral-type Hall effect from magnetic texture**

In Fig. 5a-h we combine the results of the extracted \( \rho_{xy}^{\text{THE}} \) and the transition fields of \( dM/dH \) as a function of temperature and field for the \( \text{Mn}(\chi)=1.8 \) thickness series (top row) and the \( t=110 \) nm \( \text{Mn}(\chi) \) series (bottom row). The dashed line indicates \( T_s \), the squares indicate the second-order transition and the circles indicate the first-order transition. For both (\( \text{Mn}(\chi)=1.6 \) and \( \text{Mn}(\chi)=1.8 \)) thickness series, we plot the maximum topological Hall resistivity (\( \rho_{xy}^{\text{THE, max}} \)), the magnetization at the corresponding field of \( \rho_{xy}^{\text{THE, max}} \) and \( T_s \) in Fig. 5i-k, respectively. The dashed lines are asymptotic fits of the experimental data. As can be expected, \( T_s \) shows little variation with thickness (Fig. 5j). Further, \( \rho_{xy}^{\text{THE, max}} \) saturates above 40 nm ((Fig. 5i). When compared with the magnetization dependence in Fig. 5k, the origin of this mechanism can be related to the magnetic dipole-dipole interaction, where the magnetization increases with a decrease in film thickness. 

In Fig. 5l-n, we show the dependence of \( \rho_{xy}^{\text{THE, max}} \), the magnetization at the corresponding field of \( \rho_{xy}^{\text{THE, max}} \) and \( T_s \) as a function of \( \text{Mn}(\chi) \) for \( t=38 \) nm (black) and 110 nm (red) films. The saturating evolution of \( \rho_{xy}^{\text{THE, max}} \) as function of \( \text{Mn}(\chi) \) is distinct from the thickness dependence of \( \rho_{xy}^{\text{THE, max}} \), mainly determined by anisotropies. Here, \( \rho_{xy}^{\text{THE, max}} \) is determined by the variation of exchange constants as a function of \( \text{Mn}(\chi) \) that in turn determine the size of the chiral product. The magnetization in Fig. 5n shows a decrease with \( \text{Mn}(\chi) \), while \( T_s \) (Fig. 5m) increases, both saturating above \( \text{Mn}(\chi)=1.75 \). This indicates an increase of the chiral product with \( \text{Mn}(\chi) \). The increase is related to a higher \( \text{Mn} \) sublattice occupation that prefers an antiparallel alignment of the magnetic moments due to the antiferromagnetic exchange, in turn increasing the overall canting angle (see Fig. 2b). Furthermore, due to only slight changes in the electronic structure and Berry curvature (Fig. 4a), the origin of the variation in the \( \rho_{xy}^{\text{THE, max}} \) is due to the described canting of the spins in the magnetic field.

In Fig. 6, we show the \( \rho_{xy}^{\text{AFE}} \) versus \( \rho_{xy}^{\text{THE, max}} \) on a logarithmic scale for three classes of materials: skyrmion, chiral and correlated. Within these classes, we select several relevant systems. The Heusler compounds, known to have chiral spin textures and to host antiskyrmions, \( \text{Mn}_1.3\text{Pt}_{1-x}\text{Rh}_{x}\text{Sn} \), \( \text{Mn}_2\text{RhSn} \), \( \text{Mn}_2\text{CoAl} \), \( \text{NiMnGa} \) and \( \text{NiMnIn} \). The well known B20 skyrmion systems, \( \text{MnGe} \), \( \text{MnSi} \), \( \text{Mn}_{1-0.9x}\text{Fe}_{0.9x}\text{Si} \), \( \text{Fe}_{1-0.9x}\text{Co}_{0.9x}\text{Ge} \) and \( \text{Mn}_{0.9x}\text{Co}_{0.04}\text{Si} \). The oxide materials, with many being correlated and showing magnetic domains/bubbles, \( \text{EuO} \), \( \text{EuTiO}_3 \), \( \text{Ca}_{1-0.9}\text{Cu}_{0.9}\text{MnO}_3 \), \( \text{SrRuO}_3 \) and \( \text{La}_{0.5}\text{Sr}_{0.5}\text{MnO}_3 \). Lastly, antiferromagnetic compounds, frustrated systems, and systems that we classify as others. Interestingly, Mn is one of the
most frequented elements in these compounds. Furthermore, on a logarithmic plot, all these compounds straddle the one to one ratio line for $\rho_{xy}^{\text{AHE}}$ and $\rho_{xy}^{\text{THE, max}}$. The correlated materials comparatively show both large $\rho_{xy}^{\text{AHE}}$ and large $\rho_{xy}^{\text{THE, max}}$. Whereas, the skyrmion materials show the most substantial variation away from the one to one ratio line. Lastly, the chiral compounds show a significant variation across the entire graph.

Skyrmionic textures are nominally considered to be long-range textures that give rise to the THE, due to the emergent field of the skyrmion lattice. This is usually pictured as an electron that couples adiabatically to a smoothly varying magnetization lattice that can be several orders of magnitude larger than the crystal unit cell. Whereas, in correlated systems the density of free carriers can be reduced orders of magnitude to the comparative systems. Here, the coupling of the conduction electron to the magnetic sublattice is enhanced and not only the Berry phase (adiabatic) but also non-adiabatic processes should be taken into consideration. The chiral magnetic structure suffices in both regimes. The overall strength of the THE depends on how
strongly the conduction electrons couple to the magnetic sublattice, wherein these systems the magnetic lattice is on the order of the crystal unit cell. This results in two effects: i) the rapid variation in the sublattice leads to non-adiabatic effects; ii) Berry phases in both real and momentum space emerge. Specifically, Mn$_x$PtSn shows clearly that as the canting of the spins increases, the THE is also enhanced, and the non-adiabatic mechanisms become crucial.

Nominally, the exciting contribution to the AHE is the intrinsic adiabatic Berry curvature. The requirement for the AHE is the lack of time-reversal symmetry and the splitting of orbital degeneracy. In ferromagnets, this is realized by a finite magnetization and the SOC. Whereas, in the noncollinear antiferromagnets the time-reversal symmetry is broken by the all in or all out structure. The orbital degeneracy is lifted by the conduction spin coupling to the magnetic lattice. Likewise, for the THE the time-reversal symmetry is broken by the direction of the cone angle of the noncoplanar spins. The SOC is replaced by the relative change in the direction of the magnetization in the magnetic sublattice. This leads to very similar but distinct origins for the AHE and THE. While the overall magnitude of the effects depends on the intimate nature of the electronic structure, the values of the two will always be comparable when sharing the same electronic structure. An exception may occur when there is a topological Fermi surface where the THE has a weaker dependence on the Fermi sea contribution in the weak
In summary, Mn$_x$PtSn and the host of compounds in this Heusler family, are of particular interest because the fundamental exchange parameters are tunable by chemical substitution. Specifically, we show that the role of Mn($x$) is to tune the ratio of antiferromagnetic and ferromagnetic exchange, which is crucial in the stabilization of antiskyrmions. Fine-tuning of Mn($x$) would allow for discrete adjustments of the antiskyrmions below the spin reorientation transition temperature. Furthermore, we find that this effect should be observable from low temperatures up to $\sim$200 K depending on Mn($x$). As expected, the magnetic dipole-dipole interactions play the most significant role in the thin film limit. The most substantial change in the THE is due to the Mn($x$), which tunes the relation of the exchange couplings and thereby the topological Berry curvature. The increase of Mn($x$) increases the unit cell volume, therefore we can infer that the noncoplanar spin texture is also coupled to the lattice size, and can be tuned as in multiferroic heterostructures.

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