Spontaneous anomalous Hall effect arising from antiparallel magnetic order in a semiconductor

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Abstract

Recently, a distinct mechanism of spontaneous anomalous Hall effect, the so-called crystal anomalous Hall effect, was predicted to arise from antiparallel magnetic moments surrounded by cages of nonmagnetic atoms [Šmejkal et al., Sci. Adv. 6, eaaz8809(2020)]. Here, we observe this spontaneous anomalous Hall effect in thin films of semiconducting MnTe with well-established room-temperature collinear antiparallel ordering. We show that this Hall signal is consistent with the measured experimental magnetic easy axis and with the calculated unconventional time-reversal symmetry broken band structure.

The anomalous Hall effect (AHE) is one of the most prominent phenomena of relativistic spintronics. Usually, it refers to voltage generated transverse to electric current and the pseudovector related to magnetic order [1, 2]. Unlike the ordinary Hall effect, which is proportional to the external magnetic field, the AHE is determined by the magnetic order and, therefore, can be saturated. Consequently it can have finite value at zero magnetic field and exhibit hysteresis [1]. This spontaneous character of the AHE makes it also distinct from the ordinary Hall effect. From a symmetry point of view, the AHE relies on breaking of time reversal symmetry (TRS) in the band structure. Therefore, until recently, it was commonly believed that the AHE could be found exclusively in ferromagnets [2]. It was however predicted, based on symmetry considerations [3, 4], that certain non-collinear antiferromagnets could also exhibit AHE and estimated to be measurable based on calculations of momentum space Berry curvature contributions to the effect. This was experimentally confirmed [5, 6], triggering a fast development in the field when effects such as anomalous Nernst effect [7, 8], magneto-optical Kerr effect [9] or magnetic spin Hall effect [10] were observed in non-collinear antiferromagnets. The anomalous Hall effect was also observed in other metallic antiferromagnets with multiple magnetic sublattices (≥ 3) and its relation to magnetic multipoles was studied [11–15].

Recently, a distinct mechanism of spontaneous AHE was predicted to arise in compensated antiparallel ordered collinear magnets [16]. The mechanism relies on TRS broken band structure due to the combined effect of antiparallel magnetic sublattices and nonmagnetic atoms. The two antiparallel magnetic sublattices surrounded by nonmagnetic atoms cages are then related by crystal-rotation symmetries [17, 18]. These compensated magnets were delimited via nonrelativsitic spin symmetries as separate magnetic phase, so
called altermagnetism\cite{17}. Altermagnetism in this respect refers to magnets with time reversal symmetric broken band structure and non-relativistic spin momentum locking \cite{17}. It is in stark contrast with the common antiferromagnets with spin degenerate band structure. Metallic RuO$_2$ was identified as a prime member of this new material class. The particular mechanism was referred to as crystal TRS breaking due to its strong sensitivity to the nonmagnetic atoms, which leads to sizable coherent spin currents\cite{19,20} and was also predicted in several other magnetically compensated systems \cite{21,24}. Very recently these predictions were supported by the observation of the AHE \cite{25} and spin torque \cite{26,28} in RuO$_2$. However, RuO$_2$ exhibits easy axis incompatible with spontaneous AHE and requires thus application of external spin reorientation magnetic fields. Additionally, the spontaneous Hall effect in collinear, magnetically compensated semiconductors with potentially strong spin coherence, attractive for antiferromagnetic semiconducting and topological spintronics, remained hitherto elusive\cite{29}.

Here we show that the spontaneous anomalous Hall effect can be observed in semiconducting MnTe with collinear antiparallel magnetic order. The MnTe structure fulfills the symmetry requirements of altermagnetism, can host an spontaneous AHE and, at the same time, is exceptionally well understood \cite{30,38}. Moreover, the experimentally accessible spin flop field, the room temperature magnetic ordering with T$_N \sim$310 K\cite{31} and its semiconducting character\cite{33,39,40} make MnTe an ideal model system to study the anomalous Hall response in this new class of materials. In this paper, we prepare high quality thin films by molecular beam epitaxy, systematically study their anisotropic magneto-resistance and find agreement with the theoretical predictions based on \emph{ab initio} calculations and symmetry analysis. We show that, despite its zero net magnetic moment measured in a magnetometer, a spontaneous anomalous Hall effect is present, which is particularly pronounced for magnetic field sweeping along the electrical current. This AHE signal is absent above the MnTe Néel temperature. Our results show that MnTe indeed exhibits TRS broken band structure and related effects. Due to the truly semiconducting character of MnTe our work opens prospects of breaking TRS in an intrinsic semiconductor material which is one of the long-term ambitions of the field of semiconducting and topological spintronics.

\emph{α-MnTe} with NiAs structure (space group P6$_3$/mmc #194\cite{41}), as shown in Fig. 1b, fulfills the symmetry requirements outlined before to host a spontaneous anomalous Hall effect\cite{16,17}. In MnTe magnetic moments arrange in ferromagnetically ordered Mn sheets.
which are stacked antiparallelly along the $c$-axis\cite{32,37}. The MnTe magnetic crystal breaks both the TRS coupled with inversion, and TRS coupled with partial unit cell translation\cite{16}. The symmetry breaking is provided by the Te atoms that occupy non-centrosymmetric Wyckoff position 2$c$ and create octahedra around the Mn atoms as we show in Fig. 1a similarly as in RuO$_2$\cite{16}. However, unlike in RuO$_2$, the octahedra share a face and the two opposite Mn sublattices are connected by crystallographic rotational symmetry $C_6$ coupled with half-unit cell translation along $c$-axis as can be seen from the blue and red color-shaded octahedra in Fig. 1a,\cite{17,42}. The TRS is thus broken in the electronic band structure and depending on the sublattice magnetization orientation with respect to the crystal axes MnTe can exhibit spontaneous Hall effect $\cite{2,16,17,42}$. Figure 1b,c shows two possible moment configurations within the $c$-plane. In the thin films the moments are oriented 30 degree rotated around the $c$-axis (point along $[1\bar{1}00]$) $\cite{43,37}$. In this case, the magnetic point group generators are inversion and a single two-fold screw-axis along $c$ $\cite{42}$, that allow for the existence of a pseudovector along $c$ and therefore enables the AHE $\cite{16}$. However, in case of the perfectly antiparallel moments oriented along the $a$-axis ($[2\bar{1}10]$ direction) the magnetic unit cell has an orthorhombic shape $\cite{42}$. The presence of two-fold rotation and screw-axis along the unit cell axes prevents the existence of an AHE for this moment orientation.

First-principles calculations based on density-functional theory (DFT) were used to study the electronic structure and transport properties of the MnTe. We have used the VASP (Vienna \textit{ab-initio} simulation package) code $\cite{44}$ and GGA+U (we use $U=3.03$ eV to reconstruct well previous theory and experimental results$\cite{37,42,45,46}$). The electron wave function was expanded in plane waves up to cut-off energy of 500 eV and a $12\times12\times16$ $k$-mesh has been used for the Brillouin-zone integration. By using maximally localized Wannier functions $\cite{47,48}$, we have found an effective tight-binding Hamiltonian to calculate the anomalous Hall conductivity (AHC) tensor. Integration of the Kubo formula was carried out within a $400^3$ $k$-grid with the Wannier Tools code $\cite{49}$. The found AHC for various Néel vector orientations within the $c$-plane supports our symmetry consideration above. For the magnetic moment configuration shown in Fig. 1b and current along the $a$-axis the energy dependent AHC is shown in Fig. 1d. Values above 300 S/cm are found for states deep in the valance band, but also near the valance band edge a finite AHE is expected. Here, we orient the $a$-axis or $[2\bar{1}10]$, and the $c$-axis or $[0001]$ along the $x, z$-axis of the used coordinate system,
FIG. 1. Theoretical calculation of spontaneous Hall signal in collinear MnTe: (a) Atomic configuration of Mn (violet) and Te (gold) with hexagonal NiAs structure. (b,c) Magnetic moment configurations of the hexagonal $c$-planes with magnetic moments oriented along [1$ar{1}$00] and [2$ar{1}$10], respectively. The magnetic unit cell shape (black line) for the two electron spin (red arrows) orientations is shown. For the moment configuration of (b) and current along $x$ the anomalous Hall conductivity in panel (d) is found. (e) Transversal conductivity components for an energy of 0.25 eV below the valence band maximum in dependence of the Néel vector orientation. The angle $\phi$ is defined with respect to the $a$-axis.

respectively. The $y$-axis therefore points along [01$\bar{1}$0]. For a particular energy, the transversal conductivity components $\sigma_{ij}^{AHE}$ are individually calculated for a Néel vector rotation within the $c$-plane (Fig. 1e), where we define $\phi$ as the angle between the Néel vector and the $a$-axis, $i$ as the current direction and $j$ as the voltage detection direction. Although
a perfect antiparallel arrangement of moments was used, an AHC of up to \( \sim 100 \) S/cm is found for \( \sigma_{xy} \). Other components (\( \sigma_{yz} \), and \( \sigma_{zx} \)) remained zero in agreement with our symmetry expectations. The symmetry restrictions also force \( \sigma_{xy}(\phi) \) to zero every 60 degree, i.e. where the magnetic moments are along the \( a \)-axis or equivalent directions. The resulting \( \sigma_{xy}^{\text{AHE}} \) therefore shows a harmonic variation proportional to \( \sin 3\phi \).

The calculations above only considered the AHE, but in experiments we expect to find also other contributions upon a Néel vector rotation. Symmetry considerations outlined in more detail in the SI lead us to expect a series of anisotropic magnetoresistance (AMR) contributions. In particular when considering terms up to the sixth-order the conductivity \( \sigma_{yy} \) (in the coordinate system of Fig. 1) is expected to contain terms proportional to \( \cos n\phi \), with \( n = 2, 4, 6 \). The transversal conductivity \( \sigma_{yx} \) can contain terms proportional to \( \sin n\phi \), with \( n = 2, 3, 4 \).

In order to study these effects experimentally, epitaxial thin films of MnTe were grown by molecular beam epitaxy on single crystal InP(111)A substrates at a substrate temperature of 380 °C using a Mn and Te beam flux source, with a layer thickness of around 50 nm. As indicated by reflection high energy electron diffraction, two-dimensional layer-by-layer growth occurs. X-ray diffraction data indicated excellent crystallographic quality and a film thickness of 48 nm. For growth on InP(111)A \( \alpha \)-MnTe the epitaxial relationship of \((0001)_{\text{MnTe}} \parallel (111)_{\text{InP}}\) is forming. Details of the growth, transmission electron microscopy, and neutron diffraction studies of these films can be found in Refs. 36, 37. Given their crystalline orientation, these films provide an ideal platform for studying the transport coefficients within the \( c \)-plane.

For magnetotransport studies, Hall bars, as shown in Fig. 2, were produced by electron beam lithography, Argon milling, and Cr/Au contacts produced by a liftoff process. Similar to previous studies we find the longitudinal resistivity with the characteristic temperature variation typical for MnTe (Fig. 2b). Although MnTe is a semiconductor with \( \sim 1.4 \) eV band gap 33, 39, 40 the resistivity is decreasing below 250 K and only raises again below 50 K. This was previously attributed to spin dependent scattering processes 35. In our case the decrease of \( \rho_{yy} \) at temperatures above 250 K is governed by the thermal activation of carriers in the semiconducting InP substrate and can not be solely attributed to the MnTe thin film. Magnetotransport studies therefore focus on lower temperatures where the substrate can be considered isolating. The compensated magnetic nature of these thin films can be seen from
FIG. 2. Magnetic, and basic transport properties. (a) Microscopy image of the Hall bar together with electrical schematics of our measurement. For all transport measurements a moderate current density $j$ of $\sim 5 \cdot 10^6$ A/m$^2$ is used. (b) Temperature dependent resistivity and DC susceptibility. The bulk Néel temperature $T_N$ is indicated at 310 K. The upturn in susceptibility at low temperatures is attributed to paramagnetic oxygen. (c) Field orientation dependent longitudinal resistivity for inplane magnetic fields at $T = 50$ K. (d) Field strength dependence of $\cos(n\alpha)$ components.

the low susceptibility variation shown in Fig. 2b, which only shows a weak anomaly near the expected magnetic transition temperature and is otherwise dominated by the diamagnetic substrate.

The longitudinal resistivity $\rho_{yy}$ of the MnTe thin film for magnetic field rotated within the $c$-plane is shown in Fig. 2c. The angle of the magnetic field $\alpha$ is defined with respect to the $a$-direction and is along the current ($y$) at $\alpha = 90^\circ$. The signal can be fitted by a series of
AMR terms proportional to $\cos n\alpha$, with even $n$. Note that we use the magnetic field angle $\alpha$ here instead of the Néel vector angle $\phi$ and that the moments under an applied magnetic field above the spin-flop typically arrange nearly perpendicular to the applied field. By analyzing the various harmonic contributions, we find that the spin flop field is below $\sim 3$ T since the non-crystalline AMR (proportional to $\cos 2\alpha$) is largely saturated at this field. At a temperature of 50 K, however, the largest amplitude is found for the six-fold term, i.e. proportional to $\cos 6\alpha$, which results from the six-fold crystal symmetry. Additional experiments with a Hall bar along the $x$ direction [42] show that this term is determined by the orientation of the magnetic moments with respect to the crystal direction, and therefore commonly termed crystalline AMR. The strong crystalline AMR also proves that our thin films are perfectly aligned with the single crystal substrate in the inplane direction. The shape of $\rho_{xx}(\alpha)$ lets us also conclude about the magnetic easy axis of our material. In Fig. 2c, the minima found every 60 degree appear wider than the maxima which means that the magnetic easy axes for the magnetic moments are along $\langle 1\bar{1}00 \rangle$ (moment configuration as in Fig. 1b) in agreement with previous neutron diffraction studies [37]. We also note that a vertical offset between the curves recorded for different field magnitudes is caused by an isotropic magnetoresistance contribution, which we find to be $\sim 4 - 6\%$ at 8 T [42].

Since we have shown the single crystalline nature of our thin films, we further focus on the detection of the predicted AHE signal. For this purpose, one could study $\rho_{yx}(\alpha)$. It turns out, however, that the transversal AMR (or planar Hall effect) contribution is largely dominating the signal, which masks the interesting AHE [42]. Therefore, we focus on inplane magnetic field sweeps with magnetic field at various angles. An example of the collected $\rho_{yx}(H)$ for magnetic field along the current is shown in Fig. 3a. In such measurements, it is more straightforward to isolate the AHE. Since AMR is invariant under a reversal of the magnetic moments and therefore mostly even in magnetic field we can eliminate it by separating the component symmetric in magnetic field $\rho_{yx}^{\text{even}}$ and obtain the remaining purely odd in magnetic field $\rho_{yx}^{\text{odd}}$ by subtraction [42].

In the even part (Fig. 3b) we find AMR variation which at high fields is largely saturated consistent with the expected ordering control at these field strength. The signal odd in magnetic field $\rho_{yx}^{\text{odd}}$ (Fig. 3c) shows a clear S-shape contribution which saturates around the spin flop field. Further, it includes some ordinary Hall contribution. Note that an ordinary Hall voltage here is only arising because of slightly out of plane misalignment of
FIG. 3. Transversal magnetoresistance and magnetization for inplane magnetic field sweeps recorded at $T = 50$ K. (a) As measured transversal resistivity data for field along $y$ ([0110]). (b) Even in magnetic field signal $\rho_{xy}^{\text{even}}$ obtained by symmetrizing the average of up and down field sweep. (c) Odd contribution $\rho_{xy}^{\text{odd}}$ obtained from the raw data by subtraction of the even contribution. (d) Magnetization obtained for inplane magnetic field, which is dominated by the diamagnetic substrate. The inset shows the signal after subtraction of the substrate contribution in units of Bohr magnetron per Mn.

the magnetic field. The ordinary Hall effect for fully out of plane magnetic field is found to be more than 40 times larger and corresponds to a hole carrier density of $4.9 \cdot 10^{18}$ cm$^{-3}$ [42]. Most notable, however, the odd signal shows a hysteresis loop with an opening around zero magnetic field. While some splitting at finite field could originate from the difference of the spin flop transition upon applying and remove the magnetic field (cf. Ref. 50), the
opening at zero magnetic field shows the spontaneous nature of the AHE. Figure 3 shows that no such splitting can be observed in the magnetization.

The direction and temperature dependence of $\rho_{yx}^{\text{odd}}$ is shown in Fig. 4. Figure 4a-c show $\rho_{yx}^{\text{odd}}$ for three inplane magnetic field directions. While an S-shaped signal is found in all cases, only for magnetic field along the current direction (here the $y$ direction) an open hysteresis loop is observed. The shape of the loop, and in particular the size of the hysteresis loop in comparison to the difference between the saturated states, is influenced by the population of the various inplane easy axis. In Ref. 37 it was found that after the application (and removal) of a saturating field, still all three possible equivalent axis are somewhat populated. Here for the AHE, in contrast to AMR (and other effects even in magnetization) not only the Néel axis, but also its polarity matter. We suggest that the imbalance of the two Néel vector polarities is caused by the Dzyaloshinskii–Moriya interaction arising from the non centrosymmetric Te atoms or the thin film interfaces.

In Fig. 4d we show the temperature dependence of the shift between saturated trajectories at positive and negative fields. The open-loop vanishes when one approaches the magnetic ordering temperature. Note that the spontaneous AHE requires that the inplane anisotropy can’t be overcome by thermal excitation, which in agreement with previous works requires a temperature below the onset of antiparallel order as detected by neutron diffraction. In Fig. 4d we have further converted the measured AHE into conductivity units to enable comparison with theory calculations. The values found in the experiments are significantly lower than the calculated quantities. However, the calculated values do strongly dependent on the considered energy shift and are only a few S/cm near the valence band maximum. In addition, a potential mixture of domains with effective $c$-axis pointing up and down, i.e. corresponding to a sign change of the AHE, and incomplete polarization of the magnetic order can lower the experimentally determined AHE.

Finally, we highlight that experimentally an open hysteresis loop is found for magnetic field oriented along the current for current directions along $y$ (Fig. 4b), and $x$ [42]. This indicates that the inplane anisotropy can be influenced by the patterning. Most importantly, this excludes any contribution from the field-induced magnetization, which does not contribute to the transversal signal in this configuration.

In conclusion, we have measured the spontaneous anomalous Hall effect in the compensated collinear antiferromagnetic semiconductor MnTe. In magnetic field sweeps within the
FIG. 4. Anomalous Hall contribution to the transversal resistivity for field sweeps in various directions. (a-c) Odd contribution to the transverse resistivity at $T = 50$ K for magnetic field along $x$ ([2110]), $y$ ([0110]) and $-x$ ([2110]), respectively. Note that here a linear background originating from ordinary Hall effect was subtracted. (d) Temperature dependence of the extracted anomalous Hall contribution for current and magnetic field along $x$.

c-plane we identified open hysteresis loops which saturate above the spin flop field, while magnetometry yields no measurable magnetization. Theoretical considerations corroborate our experiments and allow us to assign this anomalous Hall effect to the asymmetric crystal environment around the magnetic atoms, which break the symmetry and in combination with favorable easy axis generates the AHE. Future studies are needed to address the re-orientation mechanisms of the magnetic order and curious magnetic field dependence of the anomalous Hall signal. Our work opens prospects of TRS-broken room temperature
compensated magnetic semiconductors.

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[42] See Supplemental Material at [URL] for additional experimental data and figures.

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