Strong anomalous Nernst effect in collinear magnetic Weyl semimetals without net magnetic moments

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We predict a large anomalous Nernst effect in the inverse Heusler compensated ferrimagnets Ti2MnX (X=Al,Ga,In) with vanishing net magnetic moments. Though the net magnetic moment is zero, the Weyl points in these systems lead to a large anomalous Nernst conductivity (ANC) due to the lack of a global time-reversal symmetry operation that inverts the sign of the Berry curvature. In comparison to the noncollinear antiferromagnets Mn3Sn and Mn3Ge, the high ANC stems almost entirely from the Weyl points in this class of compounds, and thus, it is topologically protected. This work shows for the first time a large ANC with zero net magnetic moments in collinear systems, which is helpful for comprehensive understanding of the thermoelectric effect in zero-moment magnetic materials and its further applications.

In the ordinary Hall effect, a longitudinal electron current generates a transverse voltage drop by the Lorentz force in the presence of external magnetic fields. Contrary to this, the transverse electron current in the anomalous Hall effect (AHE) is induced by intrinsic magnetic moments and spin-orbit coupling (SOC) [1,2]. It is also possible to apply a temperature gradient, instead of an electric field, in combination with a magnetic field to generate a transverse charge current, which is known as the Nernst effect [3,4]. Analogously, a temperature-gradient-induced transverse charge current can also exist in the absence of external magnetic fields, referred to as the anomalous Nernst effect (ANE) [5,6]. In ferromagnets, the imbalance of carriers with spin-up and spin-down leads to a spin-polarized transverse charge current. Therefore, the magnitude of the AHE and ANE were historically considered to be proportional to the magnitude of the magnetic moments in the system [7].

In the last decade, a more fundamental understanding of the intrinsic AHE from the Berry phase has been established [2,7]. Because the Berry curvature (BC) is odd with respect to the time reversal operation, the intrinsic AHE can only exist in magnetic systems. In collinear antiferromagnets (AFMs), though the time reversal symmetry is broken, the combination of a magnetic field with a time reversal operation can generate a transverse charge current, which is known as the Nernst effect [3,4]. Analogously, a temperature-gradient-induced transverse charge current can also exist in the absence of external magnetic fields, referred to as the anomalous Nernst effect (ANE) [5,6]. In ferromagnets, the imbalance of carriers with spin-up and spin-down leads to a spin-polarized transverse charge current. Therefore, the magnitude of the AHE and ANE were historically considered to be proportional to the magnitude of the magnetic moments in the system [7].

In this work, we have theoretically studied the ANE in the compensated ferrimagnetic WSM Ti2MnX (X=Al, Ga, and In) and complemented our results with a minimal model. We predict that a large anomalous Nernst conductivity (ANC) can exist over a large temperature range. These results which indicate a strong ANE have contributions from both occupied and unoccupied bands near the Fermi level, which leads to two distinct behaviors of these effects.

In this investigation we performed DFT calculations using the vasp package [21]. We employed a plane wave basis set with pseudopotentials and used the generalized gradient approximation (GGA) [22] for the treatment of the exchange-correlation energy. From the DFT band structure, Wannier functions were generated using WANNIER90 [23] with initial projections to the s-, p-, and d-orbitals of Ti and Mn and to s- and p-orbitals of X.
To evaluate the BC $\Omega$, the tight-binding Hamiltonian $H$ was set up from the Wannier functions and used with the Kubo formula \[ \text{[2, 7, 24]} \]

$$\Omega_{ij}^n = \sum_{m \neq n} \frac{\langle n | \frac{\partial H}{\partial C_k} | m \rangle \langle m | \frac{\partial H}{\partial C_k} | n \rangle}{(E_n - E_m)^2} (E_n - E_m), \quad (1)$$

where $\Omega_{ij}^n$ denotes the $ij$ component of the BC of the $n$-th band, $|n\rangle$ and $|m\rangle$ are the eigenstates of $H$, and $E_n$ and $E_m$ are the corresponding eigenvalues. From this we calculate the $ij$ component of the AHC $\sigma_{ij}$ as

$$\sigma_{ij} = \frac{\epsilon^2}{\hbar} \sum_n \int \frac{d^3k}{(2\pi)^3} \Omega_{ij}^n$$

(2)

and of the ANC $\alpha_{ij}$ as proposed by Xiao et al. \[3, 7\]

$$\alpha_{ij} = \frac{1}{T} \frac{e}{\hbar} \sum_n \int \frac{d^3k}{(2\pi)^3} \Omega_{ij}^n [(E_n - E_F) f_n +$$

$$+ k_B T \ln (1 + \exp \frac{E_n - E_F}{-k_B T})], \quad (3)$$

where $T$ is the actual temperature, $f_n$ is the Fermi distribution, and $E_F$ is the Fermi level. To realize integrations over the Brillouin zone, a k mesh of $251 \times 251 \times 251$ points was used.

FIG. 1. (a) Left: AFM structure with a time reversal + slide symmetry leading to a vanishing AHE. Right: Ferrimagnet with broken slide symmetry. (b) Inverted Heusler FCC crystal structure of Ti$_2$MnX ($X=$Al, Mn, Ga). The magnetic moments of Ti and Mn are all aligned along the (001) direction and compensate each other. (c) Band structure, AHC, and ANC (at 300 K) of the model Hamiltonian for the AFM, FiM, and FiM (phv) case (for parameters see main text). The FiM systems show a non-zero AHC, but to get a finite ANC additionally the particle-hole symmetry has to be broken (see inset of the ANC). Note the change of the energy scale for AHC and ANC.

As previously mentioned, the net Berry phase of an AFM is zero due to the combined symmetry of a space group operation $\hat{O}$ and the time reversal operation $\hat{T}$, that changes the sign of the Berry curvature (BC). As an example, this can be observed in the combination symmetry $\hat{T}\hat{O}$ of a glide operation to the center of the unit cell and time reversal, see the left panel in Fig. 1(a). A simple and effective way to remove this symmetry is by replacing the equivalent atoms lying on the other sublattice with a different element, see the right panel in Fig. 1(c), which is just a compensated FiM, and a nonzero Berry phase from the whole BZ is allowed. In both panels the flat arrows (BC$_{\text{ANE}}$) depict the electron flow from cold (blue) to hot (red) in the ANE. The FiM model is deduced from the compensated FiM Ti$_2$MnX ($X=$Al, Ga, In) [see Fig. 1(b)]. Since the charge carrier density is relatively small in most compensated FiMs, the net Berry phases are normally very close to zero. However, the Berry phase can be strongly enhanced by some topologically protected band structures, such as nodal lines and Weyl points [see Fig. 1(c)].

To achieve a deeper understanding of the underlying mechanisms, we first study a minimal effective four-band model derived from the two-band model by Lu et al. \[25\] to analyze the effect of Weyl points on the ANE. The Hamiltonian reads as

$$H = M_0 \tau_z \otimes \sigma_z + \alpha (\sin k_x \tilde{\tau} \otimes \sigma_x + \sin k_y \tilde{\tau} \otimes \sigma_y)$$

$$+ t C(\tilde{k}) \tilde{\tau} \otimes \sigma_x + t' C(\tilde{k}) \tau_x \otimes \sigma_x$$

(4)

with $C(\tilde{k}) = \cos k_x + \cos k_y + \cos k_z$, where $\tau$ and $\sigma$ are the Pauli matrices for lattice and spin, respectively, and $M_0$, $\alpha$, $t$, and $t'$ are model parameters. The matrix $\tilde{\tau}$ is used to switch between AFM and FiM and is defined below. This minimal model describes a pair of Dirac/Weyl nodes at $k_x = k_y = 0$ with all their topological properties. As model parameters we used $M_0 = 2$ eV, $\alpha = 0.1$ eV, $t = -1$ eV, and $t' = 0.2$ eV and the results discussed in the following are all shown in Fig. 1(c). To investigate the AFM case (see Fig. 1(a) left panel) we set $\tilde{\tau} = \tau_x$ and $t' = 0$, which leads to a band structure with two Dirac points and vanishing AHC and ANC due to the presence of the $\hat{T}\hat{O}$ symmetry. To break this symmetry we make a transition to the FiM case (see Fig. 1(a) right panel) by setting $\tilde{\tau} = \frac{1}{2} \begin{pmatrix} 1 & 0 \\ 0 & 0.5 \end{pmatrix}$ to model different atom types at the lattice sites. We also set $t' = 0.2$ eV to switch on hopping between the sites. The resulting band structure exhibits two Weyl points at the Fermi level and a large AHC connected to them. However, the ANC at the Fermi level is still zero due to the preserved particle-hole symmetry (PHS) in the band structure. Therefore, it is crucial to get to a PHS violation (phv) to find a finite ANC. In our model we achieve this by shifting the onsite energy of the last two orbitals up by 1 eV simulating the difference between the atom types. Just like in the symmetry-preserving case, the model shows two Weyl points on
the line \( k_x = k_y = 0 \). However, since the last term now breaks the PHS, the band structure and thus the AHC are no longer symmetric around the Fermi level, leading to a nonzero ANC at zero energy. In both FiM cases the ANC vanishes when the AHC reaches its maximum value. It is important to note, that SOC (model parameter \( \alpha \)) is necessary to get a finite AHC. However, the strength of the AHC mainly is proportional to the distance of the two Weyl points in \( k \) space \(^{20}\) and shows to be independent of \( \alpha \), when SOC is large. This minimal model captures all important properties regarding Weyl points, AHE, and ANE, and motivates us to study the compensated FiMs \( \text{Ti}_2\text{MnX} \) \((X=\text{Al}, \text{Ga}, \text{In})\).

These compounds have an inverse Heusler lattice structure with space group \( F43m \) (No. 216) \(^{11, 20}\). They exhibit an isotropic ferrimagnetic structure, where magnetic moments are located at the Ti \([\mu \approx 1.3(1.2) \mu_B \text{ for first(second) atom}]\) and Mn \([\mu \approx -2.5 \mu_B]\) atoms. The net magnetic moment in \( \text{Ti}_2\text{MnX} \) vanishes because of the compensated magnetic sublattices formed by Ti and Mn. In total, there are twelve pairs of Weyl points. Their positions in the Brillouin zone are depicted in Fig. 2(a) and they are located slightly above the Fermi level \([34 \pm 40(27 - 36, 13 - 27) \text{ meV for } \text{Ti}_2\text{MnAl(Ga,In)}, \text{respectively}]\), as indicated in Fig. 2(b). The influence of the SOC becomes evident: While in its absence all Weyl points are at the same energy, the different SOC strength \( [\Delta \text{SOC} = 0.2(5.5, 40.2) \text{ meV for Al(Ga,In)}] \) leads to a larger spread in energy as the atoms get heavier.

At low temperature, the ANC can be obtained from the Mott relation as the derivative of AHC with respect to energy \(^6, 7\),

\[
\alpha_{ij} = \frac{\pi^2 k_B T}{3} \frac{\partial \sigma_{ij}}{\partial E}(E_F).
\]

To determine the ANE at low temperature, we first calculated the energy-dependent AHC for the three compounds with moments aligned along the \((001)\) direction. As shown in Fig. 2(c) for \( \text{Ti}_2\text{MnAl} \), the AHC \((\sigma_{xy}^z)\) can reach up to \(253(268, 133) \text{ S cm}^{-1}\) for \( \text{Ti}_2\text{MnAl(Ga,In)}, \text{respectively}\). The AHC varies sharply in the energy space around the Fermi level, which leads to a large ANC \((\alpha_{xy}^z)\) \([\text{see Fig. 2(d)}]\). At 50 K, the ANCs for \( \text{Ti}_2\text{MnAl(Ga,In)} \) are approximately \(0.2(0.16, -0.11) \text{ A m}^{-1}\text{K}^{-1}\) as large as that in the noncollinear AFM \( \text{Mn}_3\text{Sn} \)\(^{13, 14}\). The ANC from the BC formalism in equation (5) and from the Mott relation in equation (5) are compared in Fig. 2(d) at 50 K for \( \text{Ti}_2\text{MnAl} \) and show a very good agreement.

From an application point of view, the ANC at room temperature is interesting. In Fig. 3(c) we see the energy dependence of \( \alpha_{xy}^z \) at 300 K. At the Fermi energy, the value shows a peak of \( \alpha_{xy}^z = 1.31(0.94) \text{ A m}^{-1}\text{K}^{-1}\) for \( \text{Ti}_2\text{MnAl(Ga)} \), which is a high value in comparison to \( \text{Mn}_3\text{Sn} \)\(^{13, 14}\). In an analogous manner to the minimal model established in the beginning, the ANC also drops to zero at an energy above the Fermi level, which coincides with a maximum of the AHC. The different behavior of \( \text{Ti}_2\text{MnIn} \) can be understood in terms of the stronger SOC induced by the higher atomic mass.
FIG. 4. (a) BC in the \(k_x-k_y\) plane at \(k_z = 0\), showing only contributions near the Weyl points (marked as green dots). (b) BC integrated along \(k_x\) \((\Omega_{xy}^{k_x}(k_x, k_y) = \int \Omega_{xy}(k_x, k_y, k_z) dk_z)\), again the highest contributions come from the Weyl points. The eight more central maxima can be associated to the same Weyl points but from the neighboring BZs and are included due to the projection procedure. (c) ANC at 300 K in the \(k_x-k_y\) plane at \(k_z = 0\), showing only contributions near the Weyl points. (d) ANC at 300 K integrated along \(k_z\). A clear connection to the BC in (b) is visible.

of In. This leads to a larger spread in energy of the Weyl points and strongly influences the behavior of the AHC [see Fig. 3 (a) and (b)], and consequently, the ANC near the Fermi level. Based on this concept, this indicates a strong connection between the Weyl points and AHE/ANE in these systems. The sign of the ANC is related to the slope of the AHC at the Fermi level via the Mott relation. This slope is positive for Ti\(_2\)MnAl(Ga) and negative for Ti\(_2\)MnIn, which is also the case for the respective ANCs at low temperature.

We also investigated the temperature dependence of the ANC [see Fig. 3 (d)] using equation (3). The temperature dependent ANC from the Mott relation and equation (3) shows good agreement at low temperatures. The divergence between the two at high temperatures imply the Mott relation only applies to low temperatures. The ANC shows a broad peak around \(T = 300\) (350) K for Ti\(_2\)MnAl(Ga). This high value in this temperature range means that both Ti\(_2\)MnAl and Ti\(_2\)MnGa are interesting candidate materials for room-temperature thermoelectric applications. Ti\(_2\)MnIn behaves differently due to a larger SOC. More importantly, the sign of the ANC changes near 175 K.

Since the ANC can be understood as the integration of the BC over the BZ with the inclusion of temperature effects, we further examined the origin of the ANC by investigating the BC distributions in BZ. Structurally and energetically all three compounds share similar properties, therefore Ti\(_2\)MnAl with magnetization along the (001) direction is suitable for the \(k\) dependent BC analysis. In Fig. 4(a) the \(k_x-k_y\)-plane at \(k_z = 0\) is shown. There are eight spots with a non-negligible \(\Omega_{xy}\) component. These points correspond to the positions of the Weyl points near \(k_z = 0\) (marked as green dots). Examining the BC integrated along \(k_z\) \((\Omega_{xy}^{k_z}(k_x, k_y) = \int \Omega_{xy}(k_x, k_y, k_z) dk_z)\), again there are clear maximums located at the projected positions of the Weyl points. The eight points with a high BC contribution which are closer to the center of the Brillouin zone can be referred to the same eight Weyl points but in the neighboring Brillouin zones, and result from the projection.

Taking temperature into consideration, as indicated in Fig. 4(c) a cut through the Brillouin zone at \(k_z = 0\) is shown with the \(z\)-component of the ANC at 300 K. Moreover, the highest contributions to the ANC stem from the eight Weyl points near the plane in this case. Examining the projection of the ANC along \(k_z\) [see Fig. 4(d)] a clear relation to the BC in Fig. 4(b) is visible. Thus, the ANC stems mostly from the Weyl points in these materials.

In summary, we have theoretically investigated the ANE in the compensated ferrimagnetic WSMs Ti\(_2\)MnX (X=Al,Ga,In). Although the net magnetic moments are zero, all three compounds exhibit strong ANEs due to the large BC of the Weyl points around the Fermi level. In comparison to the noncollinear AFMs Mn\(_3\)Sn and Mn\(_3\)Ge, which also exhibit nonzero ANC in the absence of net magnetic moments, the ANC in Ti\(_2\)MnX (X=Al,Ga,In) is dominated by the Weyl points. The temperature-dependent ANC shows a broad plateau around \(T = 300\) K in the compounds Ti\(_2\)MnAl and Ti\(_2\)MnGa. Due to the large ANC and the vanishing net magnetic moment, they would allow for possible application and detection at ambient temperature. In studies of topological bands in AFM systems it has been shown that there is an anisotropic response with the direction of the magnetization, which we believe should also be present in our systems due to the similarities [9, 27]. This work demonstrates, for the first time, a large ANC in fully compensated collinear ferrimagnetic systems that is due to the topology of the electronic structure.

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