Giant room-temperature anomalous terahertz Faraday rotation in the magnetic Weyl semimetal Co$_2$MnGa

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(Dated: December 23, 2021)

We report measurement of terahertz anomalous Hall conductivity and Faraday rotation in the magnetic Weyl semimetal Co$_2$MnGa thin films as a function of the magnetic field, temperature and thickness, using time-domain terahertz spectroscopy. The terahertz conductivity shows a thickness-independent anomalous Hall conductivity of around 600 Ω$^{-1}$·cm$^{-1}$ at room temperature, and it is also frequency-independent from 0.2-1.5 THz. The magnitude of both the longitudinal and Hall conductivity, the weak spin-orbit coupling and the position of Weyl points very close to the chemical potential all satisfy the criteria for intrinsic anomalous Hall conductivity. First-principle calculation also supports the frequency-independent intrinsic anomalous Hall conductivity at low frequency. We also find a thickness-independent Faraday rotation of 59 (±6) mrad at room temperature, which comes from the intrinsic Berry curvature contribution. In the thinnest 20 nm sample, the Faraday rotation divided by the sample thickness reaches around 3 mrad/nm due to Berry curvature, and is the largest reported at room temperature. The giant Verdet constant of the order of 10$^6$ rad m$^{-1}$ T$^{-1}$ at room temperature indicates that Co$_2$MnGa is of great potential for applications as optical isolator and modulator in the THz spectral range. The Hall angle from 0.2-1.5 THz is around 8.5 % at room temperature, which is promising for THz spintronics.

Introduction

The last decade witnessed an explosion of research on topological states of matter characterized by the topological properties of the bulk wave-functions\(^1\)-\(^3\). Topological insulators are robust to adiabatic perturbation as long as the bulk gap is not closed\(^4\)-\(^3\). Weyl semimetals are newly discovered topological states of matter without a bulk gap and with open Fermi surface arcs when either time-reversal or inversion symmetry is broken\(^4\)-\(^1\).\(^1\)-\(^1\)\(^1\). These materials have accidental band touching at pairs of points with different chirality in the momentum space. Near these points, the quasi-particles (low-energy excitations) can be described by Weyl equations first proposed by Hermann Weyl in 1929\(^12\). As a result, these touching points in the band structure are called Weyl points, and the quasi-particles near them resemble Weyl fermions. The bulk wave functions in Weyl semimetals acquire a Berry phase as they move around the Weyl point because each Weyl node behaves like a fictional magnetic field known as Berry curvature. These Weyl points (monopoles in k space) are also topologically protected because translation-invariant perturbations are identical to moving Weyl points in the momentum space, unless they meet in the zone boundary and annihilate with each other.

Weyl semimetals host many exotic phenomena including interesting temperature and frequency dependence in optical conductivity\(^13\)-\(^15\), novel quantum oscillations related with Fermi arcs\(^16\), giant second harmonic generation\(^17\),\(^18\), as well as a chiral magnetic effect and intrinsic anomalous Hall effect\(^19\). Experimental signatures of Weyl semi-metals are still far behind the advance of various theoretical proposals. Thus far, the widely experimentally studied Weyl semi-metal materials are non-magnetic, but with broken inversion symmetry\(^9\)-\(^11\),\(^20\).

Fermi arcs in magnetic Co$_2$Sn$_2$S$_2$ and Co$_2$MnGa were identified recently\(^21\),\(^22\), which established direct evidence for the magnetic Weyl semimetals. Among them, Co$_2$MnGa is particularly interesting as it is a room temperature ferromagnetic with a large anomalous Hall effect and a high curie temperature at $T_C$=690 K\(^23\),\(^24\). Bulk Co$_2$MnGa is a Heusler compound, which has a cubic face-centered structure with space group $Fm3m$ (No. 225). Large anomalous Nernst effects are also observed in both bulk and thin films\(^25\)-\(^27\).

The anomalous Hall effect (AHE) has been studied extensively in conventional ferromagnetic materials\(^28\),\(^29\). The intrinsic AHE is a scattering-independent process first proposed by Karplus and Luttinger\(^30\), which depends only on the topological band structure with the contribution of the Berry curvature in the momentum space\(^28\),\(^31\),\(^32\). The AHE can be greatly enhanced when Fermi energy is close to band (anti-)crossings such as the Weyl points\(^33\)-\(^35\). Due to its exotic band structure, magnetic Weyl semimetals (WSMs) such as Co$_2$MnGa are a good platform to study the intrinsic AHE\(^36\),\(^37\). When the chemical potential is close to the Weyl points, the nonzero net Berry curvature effect is dominating, and a large intrinsic anomalous Hall effect can be calculated by the Kubo formula and compared with experiments\(^28\).

Another advantage of Co$_2$MnGa is that thin films, which are ideal for device applications, can be fabricated by sputtering. The samples studied in this work are grown by sputtering on MgO substrates and capped with 3 nm Al, the growth details of which were the subject of previous study\(^24\). At room temperature, transport measurement on thin films show a large anomalous Hall conductivity (AHC) of 814 Ω$^{-1}$·cm$^{-1}$ and a large anomalous Hall angle (AHA) of 10.5%\(^24\). However, the spectrum of the anomalous Hall conductivity, $\sigma_{xy}(\omega)$, and Hall an-
the intrinsic contribution. Using first-principle calculations, we show that the AHC and Faraday rotation both are dominating from the intrinsic contribution.

Results and Discussion

We present the zero-field terahertz conductivity on these films first. Utilizing TDTS, we obtain the complex longitudinal conductivity spectra, $\sigma_{xx}(\omega)$, as we measure the magnitude of the Faraday rotation is proportional to the polarizability at high frequency. The flat spectra in the real part (solid lines) and the small linear spectra of Im $\sigma_{xx}(\omega)$ (dashed lines) both indicates a short $\tau$. Fitting shows $1/\tau$ is around 3-5 THz in the whole temperature range. The magnitude of the real part of the longitudinal conductivity falls into one of the criteria for intrinsic AHE as discussed below.

Besides the intrinsic contribution to AHE, there are two other kinds of extrinsic contributions: skew scattering and side jump. In ferromagnets, the combination of the total anomalous Hall and longitudinal conductivity show a crossover behavior in three regions according to the magnitude of longitudinal conductivity: dirty regime ($\sigma_{xx} < 10^4 \, \Omega^{-1} \cdot \text{cm}^{-1}$), intermediate regime ($\sigma_{xx} = 10^4 - 10^6 \, \Omega^{-1} \cdot \text{cm}^{-1}$), and extreme conducting regime ($\sigma_{xx} > 10^6 \, \Omega^{-1} \cdot \text{cm}^{-1}$). The intrinsic contribution always dominates in the intermediate regime. The magnitude of the real part of the longitudinal conductivity in Co$_2$MnGa falls at the lower boundary in the intermediate regime. Often, to separate the three contributions, the scaling relation between longitudinal conductivity and anomalous Hall conductivity is used. The contrition of the skew scattering to the AHC is proportional to the square of longitudinal conductivity $\sigma_{xy}^2 \propto \sigma_{xx}^2$, while the intrinsic and side-jump contributions are both independent of the longitudinal conductivity. Previous DC transport on Co$_2$MnGa bulk crystal used the scaling relation and revealed that the intrinsic AHC is around $10^3 \, \Omega^{-1} \cdot \text{cm}^{-1}$. Nevertheless, it is very difficult to separate the intrinsic contribution and side jump in DC transport. Interestingly, Co$_2$MnGa also satisfies the other two criteria that favors intrinsic contribution: 1) The AHC is on the order of 1000 $\Omega^{-1} \cdot \text{cm}^{-1}$. The intrinsic AHC value in the thin film and bulk crystal was reported to be 1138 $\Omega^{-1} \cdot \text{cm}^{-1}$ and 1164 $\Omega^{-1} \cdot \text{cm}^{-1}$ respectively. 2) The anticrossing point (the Weyl point) is only around 80 meV above the chemical potential. Because of these three criteria, one can perform first-principles calculations to
reliably predict the frequency-dependent intrinsic contribution and compare it with experiments.

With a set of freestanding wire-grid polarizers as shown in Fig. 2A, our TDTS can resolve the polarization state of the THz signal and measure the frequency dependent AHC. Three THz wire-grid polarizers (P1,P2,P3, extinction ratio >2000 at 1 THz) are used to measure the Faraday angle. P1 aligns the incident polarization vertically. P2 is mounted on a rotation stage (not shown in the figure) to selectively pass the vertical electric field \(E_x\) or horizontal electric field \(E_y\). P3 is fixed at 45° so that \(E_x\) and \(E_y\) have the same response at the detector. The polarization change before and after the Co₂MnGa film is the Faraday rotation \(\theta_F(\omega) = \frac{E_y(\omega)}{E_x(\omega)}\).

These measurements were performed under an out-of-plane magnetic field up to 7 T. To exclude the nonmagnetic effects such as birefringence from the windows, we apply ±B to get a symmetrized Faraday angle spectra \(\theta_F(\omega,H) = \frac{\theta_F^{\text{meas}}(\omega,H) - \theta_F^{\text{meas}}(\omega,-H)}{2}\). Fig. 2B shows the Faraday angle \(\theta_F(\omega)\) of the 40 nm sample at 290 K. The rotation has a weak dependence on the frequency. The field dependence is similar to the known magnetization curve with a saturation field \(H_s \approx 1.5\) T as shown in Fig. 2C. Note that usually the Faraday rotation is proportional to the thickness and characterized by the Verdet constant \(\theta_V\). The large \(\theta_F\) around 60 mrad at room temperature is nearly thickness independent as in Fig. 2D, which mainly contribute from the anomalous terahertz Hall effect. \(\theta_V\) increases monotonically and reaches 80 mrad as the temperature cools down to 2K (see Fig. 2E). Below 1 Tesla, the Verdet constant of the 40 nm sample reaches \(10^6\) rad m⁻¹T⁻¹, which is of similar size of the giant magneto-optical effect in topological insulators HgTe \(^{34}\) and Bi₂Se₃ \(^{40,45}\) at low temperature. In Fig. 2F, we show the normalized Faraday rotation by thickness at room temperature, Co₂MnGa has the largest value of 3 mrad/nm to our best knowledge. It is 30 times larger than the value in the Weyl antiferromagnet candidate Mn₃Sn \(^{46,47}\). Even if we compare it with other reported values at low temperature, 3 mrad/nm is larger than any material report and is similar to another magnetic Weyl semimetal Co₃Sn₂S₂ \(^{18}\).

The terahertz Hall conductivity spectra \(\sigma_{xy}(\omega)\) can be extracted from the Faraday rotation by the relation

\[
\sigma_{xy}(\omega) = \frac{\theta_F(\omega)}{H_s} \frac{n+1}{d} Z_0,
\]

where \(n\) is the refractive index of the substrate, \(Z_0\) is the vacuum impedance and \(d\) is the thickness of the film. Fig. 3A shows the terahertz Hall conductivity \(\sigma_{xy}(\omega)\) of 40 nm Co₂MnGa. Consistent with the flat spectra of longitudinal conductivity and Faraday rotation, it is also independent of frequency. The mean value between 0.2 and 1.5 THz under each field is plotted in Fig. 3B, again scaling with the magnetization \(^{24}\). According to the Hall conductivity formula:

\[
\sigma_{xy} = R_0 H + \sigma_{xy}^A
\]

The total Hall conductivity \(\sigma_{xy}(\omega)\) consists of the or-
The large scattering rate in Co$_2$MnGa makes it difficult to separate the intrinsic and side jump contributions via the scaling of $\sigma_{xy}$ versus $\sigma_{zz}^2$ at terahertz frequency, as the measured THz frequency range is comparable to the scattering rate. Nevertheless, as we discussed above, because Co$_2$MnGa satisfies the three criteria for intrinsic anomalous Hall conductivity in terms of longitudinal conductivity, Hall conductivity, and chemical potential, it is quite accurate to use density functional theory (DFT) to calculate and identify the intrinsic contribution. We calculate the electronic band structure based on DFT by employing the full-potential local-orbital code (FPLO) with localized atomic basis. The exchange and correlation energies were considered in the generalized gradient approximation (GGA) level following the Perdew–Burke–Ernzerhof parametrization scheme. Following experimental results, we set a ferromagnetic structure with magnetic moment along $z$ direction. We projected the Bloch wavefunction into high symmetric atomic-orbital-like Wannier functions and constructed the tight-binding model Hamiltonian by the Wannier function overlap. Based on the tight-binding model Hamiltonian, the DC anomalous Hall conductivity and THz conductivity were computed by following the Kubo formula approach in linear response approximation and clean limit, with AHC
and terahertz conductivity

\[ \sigma_{xy}(\hbar\omega) = ie^2\hbar \int_{BZ} \frac{d^3k}{(2\pi)^3} \sum_{m \neq n} \frac{f_m(k) - f_n(k)}{E_m(k) - E_n(k)} \]

\[ <u_n(k)|\hat{v}_x u_n(k)> <u_m(k)|\hat{v}_y u_m(k)> \]

\[ \frac{E_m(k) - E_n(k) - (\hbar\omega + \eta)}{E_m(k) - E_n(k)} \]

(4)

where \( f_n(k) \) is the Fermi-Dirac distribution, \( E_n(k) \) is the eigenvalue of \( n \)th band with eigenstate \( |u_n(k)\rangle \), \( \hat{v}_x(k) = \frac{1}{\hbar} \frac{\partial H(k)}{\partial x} \) is the velocity operator, \( \omega \) is the transition energy, and \( \eta \) is a smearing parameter to avoid numerical divergence. (Here we set \( \eta = 0.1 \) meV.) We used a dense \( k \)-grid of \( 240^3 \) for the numerical integration. As shown in FIG. 3D, when the Weyl points are around 80 meV above the chemical potential, it matches the anomalous THz Hall conductivity, which also agrees with previous transport and ARPES studies. FIG. 3D shows the intrinsic anomalous THz Hall conductivity over a larger frequency range with resonant features associated with Berry curvature contribution, which we hope future experiments could explore.

We also measure the Hall angle at THz frequency, \( \theta_H = \frac{\sigma_{xy}(\omega)}{\sigma_{xx}(\omega)} \), as it will be useful for field-effect transistors around 1 THz. A large Hall angle \( \sim 8.5\% \) is observed at 290 K, as shown in FIG. 3F. It increases as temperature decreases and reaches a maximum with 10 \% at 2 K. Co2MnGa is one of few materials that exhibits large AHE and large Hall angle at the same time. Since AHE and spin Hall effect are generated by the same mechanisms, the large AHE in Co2MnGa guarantees a large spin Hall effect, which was reported recently. Looking forward, we believe that our observation of large THz anomalous Hall conductivity, Faraday rotation, and Hall angle from the intrinsic contribution at room temperature will be critical to use Co2MnGa for future applications such as optical isolator, modulator, and topological spintronics at the THz frequencies.

X.H., J.S. and L.W. acknowledge the support from the ARO under the Grants W911NF1910342 and W911NF2020166. X.H. and J.S. are also partially supported by the Gordon and Betty Moore Foundation’s EPiQS Initiative, Grant GBMF9212 to L.W. The acquisition of the laser for the THz system is support from a seed grant at National Science Foundation supported University of Pennsylvania Materials Research Science and Engineering Center (MRSEC)(DMR-1720530).

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