Anomalous Hall Effect and Magnetoresistance in Sputter-Deposited Magnetic Weyl Semimetal Co$_2$TiGe Thin Films

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Herein, sputter-deposited ferromagnetic Weyl semimetal (WSM) and full-Heusler compound Co$_2$TiGe is investigated. Crystal quality is analyzed using X-ray diffraction and reflectivity. In addition, temperature-dependent transport and magnetization measurements are carried out. The sample shows indications on the formation of L2$_1$ crystal structure. Magnetization measurements show a saturation magnetization of 1.98 $\mu_B$ (f.u.$^{-1}$) and 1.45 $\mu_B$ (f.u.$^{-1}$) at 50 and 300 K, respectively. This is in close agreement to the calculated value of 2 $\mu_B$ (f.u.$^{-1}$) at 0 K using the Slater–Pauling rule. The obtained Curie temperature is 378.5 K, which is close to prior results for bulk samples. The residual resistivity of 142.7 $\mu\Omega$ cm is mainly dominated by disorder scattering. At temperatures above 60 K, the Coulomb interaction dominates the resistance. The residual resistance ratio is around 1.49. Hall measurements show positive ordinary Hall and anomalous Hall constants and a positive dependence on temperature. Skew scattering and side jumps or intrinsic mechanisms contribute in similar amounts to the anomalous Hall resistivity, which indicates a higher than usual intrinsic contribution, which is expected for WSMs. The expected relation between the longitudinal and the anomalous Hall conductivity of $\sigma_{xy}^A \propto \sigma_{xx}^0$ is not met.

1. Introduction

Weyl semimetals (WSMs) have attracted large interest in recent years, as their band structure gives rise to nontrivial topological states. Their excitations (Weyl fermions) are expected to have a large mobility, and thus there are also potential applications of such materials in electronics. Their band structure shows a linear dispersion around Weyl nodes which results in exotic transport properties as well as a topological protection.\[1\] The WSM state is realized either by breaking time-reversal symmetry due to magnetism or crystal inversion symmetry. The latter was discovered in TaAs, whereas the magnetic counterparts remain elusive.\[4,6,7\]

Heusler alloys are a class of highly versatile materials, which provide a wide variety of properties such as ferrimagnetism, metallic or semiconducting character, and tailorable band structure.\[6–10\] Some of the cobalt-based Heusler alloys show a semimetallic behavior and have been thoroughly studied in recent years.\[11–15\] The Co$_2$TiGe (CTG) compound is one of those candidates and is promising, as it has a Curie temperature ($T_C$) above room temperature which is crucial for application. Several structural, magnetic, and transport studies on bulk CTG have been conducted and confirm the expected properties.\[16–25\] A recent study on CTG grown by molecular beam epitaxy (MBE) showed differing results.\[26\] In this work, we analyze thin films of CTG grown by sputter deposition, which is crucial for later real-life application and mass production.

2. Results and Discussion

From X-ray fluorescence, the stoichiometry of the investigated sample system is found to deviate less than 1% from the target value of 50:25:25 of Co:Ti:Ge for each of the constituents. The X-ray reflectometry (XRR) and diffraction (XRD) measurements are carried out to find the best parameters for the film growth. The lattice constant of CTG is around 5.831 Å, thus, growth is expected to take place 45° rotated, i.e., parallel to the [110] direction on the substrate (MgO, lattice mismatch \(\approx 2\%\)).\[27\] This is confirmed by texture measurements (not shown here).

The crystallographic analysis is shown in Figure 1. Figure 1a shows the XRR result with a simulation of the experimental data using the Parratt recursion formula to simulate specular reflectivity. A very low roughness is obtained (standard deviation \(\sigma\) of the nominal thickness of 0.4 nm) and the fitted thickness \(d = 20.4\) nm corresponds very well to the 20 nm value calculated via the deposition rates. The resulting density \(\rho\) is slightly higher than the expected value. Figure 1b shows the XRD results of the same sample on an MgO substrate with an adjacent MgO seed layer.

The spectrum shows the MgO (002) substrate double peak (Cu $K_{\alpha 1}$ and $K_{\alpha 2}$) at $2\theta \approx 42.9^\circ$, as well as the small Cu $K_{\beta}$ feature at $2\theta \approx 38.6^\circ$. The CTG thin films is presented by the (004) fundamental peak at $2\theta = 63.76^\circ$ and the order dependent (002) peak

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Figure 1. a) X-ray reflectivity measurement (blue circles) and simulation (red line) of a 20 nm CTG thin film on top of (001) MgO substrate. The extracted density $\rho$, thickness $d$, and roughness $\sigma$ are also stated. b) $\theta$/$2\theta$ scan in the [001]MgO direction of the same sample system. The inset shows the [111]CTG peak from a $\theta$/$2\theta$ scan along the [111]CTG direction.

The vibrating sample magnetometer (VSM) results displayed in Figure 2a show a ferromagnetic response and an in-plane magnetization orientation for good crystalline quality.[17,25] The presence of the (002) reflection gives indication of at least a B2 structure. Moreover, at least partial L2$_1$ crystal structure is indicated by the presence of the (111) superlattice peak, which is shown in Figure 1b inset. Furthermore, the XRD spectrum does not reveal any unexpected reflection, thus no undesired phases are present in the thin film.

Transport measurements are carried out in four probe geometry at temperatures between 2 and 300 K. The results are shown in Figure 3. The inset in Figure 3a shows the Hall bar geometry with six side contacts (three on each side) with $l = 1150 \mu$m and $w = 200 \mu$m, which is used to measure the longitudinal and transverse responses. Electrical resistivity of the thin film as a function of the temperature during a cooldown process is shown in Figure 3a. As a rough measure of sample quality, the residual-resistance ratio $RRR = \rho(300K)/\rho(2K)$ can be used. It shows a value of 1.49, which is in good agreement with prior works and a confirmation for good crystalline quality.[17,25] Electrical resistivity of semimetallic materials

Figure 2. Magnetometry results of the same 20 nm-thick CTG sample. a) Vibrating sample magnetometry measurements of the in-plane magnetic moment at different temperatures as a function of the external magnetic field. b) Experimental data (blue dots) with error bars of the magnetic moment in saturation as a function of temperature and a simulation (red line) to estimate the saturation magnetization $M_0$ and $T_C$ of CTG.
Figure 3. a) Longitudinal resistivity of CTG as a function of temperature (black) with the fits (red) for temperatures below 60 K (light red area, Equation 2), between 60 and 100 K (yellow area, Equation 3) and above 100 K (cyan area, Equation 4). The inset shows the Hall bar schematic. b) Hall resistivity $\rho_{xy}$ as a function of external magnetic field ($B \| [001]$) taken at different temperatures between 2 and 300 K. The inset shows the extracted carrier density $n$ as a function of temperature.

constitutes of different types of scattering mechanisms. They can be summarized by $\rho(T) = \rho_0 + \rho_{\text{e-e}} + \rho_{\text{e-ph}} + \rho_{\text{e-mag}}$.\[13] Here, $\rho_0$ is the temperature-independent residual resistivity, due to impurities. The other components contribute as follows: $\rho_{\text{e-e}} \propto T^2$-dependent contribution due to charge carrier Coulomb interaction, $\rho_{\text{e-ph}} \propto T$-dependent contribution from electron–phonon interaction, and $\rho_{\text{e-mag}}$: electron–magnon interaction contributes with $\propto T^2$ for one magnon scattering, whereas two magnon scattering contributes with $\propto T^{9/2}$ and $\propto T^{7/2}$ for low and high temperatures, respectively. According to Kubo and Ohata, $\propto T^2$ one magnon scattering is forbidden in semimetallic materials.\[32]

In addition, a $\propto T^{1/2}$ dependence can contribute to the resistivity in disordered systems. In our case, the slope analysis ($d\rho/dT$) suggests that the $\rho$–$T$ curves are divided into three distinct regions for a proper description. These regions are indicated by different background colors. A low temperature region up to 60 K (light red), followed by a short region up to 100 K (yellow), where a change in carrier density $n$ occurs (see the inset in Figure 3b), and a high temperature region up to room temperature (300 K, cyan). The data are then fitted with the following equations

$$\rho(T) = \rho_0 + AT^{1/2} + CT^2 + ET^{9/2} \quad (\text{for } T < 60 \text{ K}) \quad (2)$$
$$\rho(T) = \rho_0 + CT^2 + ET^{9/2} \quad (60 \text{ K} < T < 100 \text{ K}) \quad (3)$$
$$\rho(T) = \rho_0 + BT + DT^{7/2} \quad (100 \text{ K} < T) \quad (4)$$

The results of the fits are shown in Table 1. In the lowest regime, the impurity-driven temperature-independent residual resistivity $\rho_0 = 142.68 \, \mu\Omega \text{ cm}$, which is lower than literature values on bulk samples. This can be attributed to a very high crystal quality. The temperature-dependent disorder scattering $\propto T^{1/2}$ contributes predominantly with a factor $A = -0.097 \, \mu\Omega \text{ cm K}^{-1/2}$, the electron–electron interaction term $\propto T^2$ is weaker at low temperatures, with $C = -2.03 \times 10^{-4} \, \mu\Omega \text{ cm K}^{-2}$, and becomes the dominating term above 63 K. The negative constants in the lowest regime account for the increase in resistivity toward lower temperatures, which can be attributed to multiple reasons. This kind of behavior is observed by Obaida et al. in different Co-based Heusler compounds.\[13] There, the upturn in resistivity in amorphous or crystalline metals with structural disorder is interpreted as weak localization or electronic correlation effects.\[14,15] The two magnon scattering is not contributing significantly, $E = 15.5 \times 10^{-9} \, \mu\Omega \text{ cm K}^{-9/2}$. The second temperature region is heavily dominated by electron–electron $C = 6.45 \times 10^{-4} \, \mu\Omega \text{ cm K}^{-2}$, with a negligible contribution by two magnon scattering $E = 1.84 \times 10^{-9} \, \mu\Omega \text{ cm K}^{-9/2}$. The high temperature region above 100 K is dominated by electron–phonon interaction, $B = 0.19 \, \mu\Omega \text{ cm K}^{-1}$, whereas two magnon scattering only contributes with $D = 5.6 \times 10^{-9} \, \mu\Omega \text{ cm K}^{-7/2}$.

In addition, Hall resistivity measurements have been carried out using a six terminal Hall bar geometry and driving an external magnetic field pointing out-of-plane. A direct current (DC) current of 0.5 mA has been applied along the Hall bar and transverse and longitudinal voltage have been recorded. Exemplarily, a few of the resulting Hall measurements are shown in Figure 3b. It shows the transverse resistivity in dependence of the external magnetic field in the range of $\pm 4$ T for temperatures between 2 and 300 K. The Hall resistivity depends proportionally on the temperature. Applying a field alters the resistivity due to change

| Temp. [K] | $\rho_0$ [\(\mu\Omega \text{ cm}\)] | $A \times 10^9$ [$\mu\Omega \text{ cm K}^{-1/2}$] | $B \times 10^9$ [$\mu\Omega \text{ cm K}^{-2}$] | $C \times 10^{-4}$ [$\mu\Omega \text{ cm K}^{-2}$] | $D \times 10^{-4}$ [$\mu\Omega \text{ cm K}^{-7/2}$] | $E \times 10^{-9}$ [$\mu\Omega \text{ cm K}^{-9/2}$] |
|----------|---------------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| 2-60     | 142.68              | -0.097          | -2.03           |                | 15.5            |
| 60-100   | 140.10              | 6.45            | 5.65            |                | 1.84            |
| 100-300  | 129.23              | 0.19            |                |                | 5.65            |
in the magnetization until the saturation occurs. The transverse response as a function of an external magnetic field $\mu_0 H$ can be described by $\rho_H(\mu_0 H) = \rho_0(\mathcal{R}_\text{OHE} + \mathcal{R}_\text{AHE})$, where $\rho_0$ is the vacuum permeability, $\mathcal{R}$ the spontaneous magnetization $\mathcal{R}_\text{OHE}$ and $\mathcal{R}_\text{AHE}$ the ordinary Hall and anomalous Hall constants, respectively.\[16\] Small misalignments and imperfect lithography can falsify the results, thus an antisymmetrization, $\rho_H(\mu_0 H) = (\rho_H(\mu_0 H) - \rho_H(-\mu_0 H))/2$, is carried out on the data. The slope of the high field part $\mathcal{R}_\text{OHE}$ gives information about the carrier density $n = 1/(R_\text{OHE}e)$, where $e$ is the charge of an electron. Furthermore, it can be used to calculate the Hall mobility, $\mu_H = \sigma_\text{xx} R_\text{OHE}$. In our case, the carrier density varies between $1.6 \times 10^{22}$ cm$^{-3}$ at low temperatures and $2.4 \times 10^{22}$ cm$^{-3}$ at room temperature, with a rapid change between 80 and 100 K (Figure 3a inset). Consequently, the mobility shows an inverted shape and drops from 2.65 to $1.6 \times 10^{22}$ cm$^{-3}$ (not shown here). Figure 4a shows the anomalous Hall resistivity as a function of temperature. The data resemble the longitudinal resistivity measurements in shape, but the anomalous Hall resistivity is two orders of magnitude lower. To get some additional information on the contributions in the anomalous Hall resistivity, it is analyzed in dependence on the longitudinal resistivity. The following equation, based on Vidal et al., is used to analyze the data

$$\rho_{\text{AHE}} = (a + b\rho_{\text{xx},0})\rho_{\text{xx},0} + (a + 2b\rho_{\text{xx},0})\rho_{\text{xx}} + b\rho_{\text{xx}}^2$$ \hspace{1cm} (5)

Here, the first term is the residual anomalous Hall resistivity, $a$ is the skew scattering parameter, $b$ is the parameter for quadratic intrinsic mechanism, and $\rho_{\text{xx},0}$ is the residual longitudinal resistivity.\[17\] The fitted data are shown in Figure 4b. The analysis shows a skew scattering parameter $a = 6.56 \times 10^{-4}$, and the intrinsic mechanism parameter $b = 1.23 \times 10^{-3}$ (µΩ cm)$^{-1}$. In WSMs, it is expected that the intrinsic contribution dominates over the extrinsic contributions, due to their chiral anomaly. In this case, the contributions are of similar magnitude, but in comparison with other Co-based Heusler alloys the skew scattering contribution is by two orders of magnitude smaller.\[38\] The quadratic part is more prominent, though it can result from a higher contribution from side jumps.

A method to verify the Weyl semimetallicity in a material is the analysis of the anomalous Hall conductivity (AHC) $\sigma_{\text{xy}}^a = \rho_{\text{xy}}/(\rho_{\text{xx}}^2 + \rho_{\text{xy}}^2)$ as a function of the longitudinal conductivity $\sigma_{\text{xx}}$. Figure 5 shows the dependency of $\sigma_{\text{xy}}^a$ on $\sigma_{\text{xx}}$. The expected $\sigma_{\text{xy}}^a \propto \sigma_{\text{xx}}$ constant dependency is not observed in the present sample. A possible explanation is that the intrinsic contribution of $\sigma_{\text{xy}}^a$ is reciprocally dependent on the distance between the Weyl nodes and the Fermi energy, which is around 0.3 eV in the case of Co$_2$TiGe.\[39\]

3. Conclusions

In conclusion, thin films of epitaxial Co$_2$TiGe have been successfully grown using sputter deposition. φ-scans, confirm the formation of at least partial L2$_1$ structure and a good crystalline quality with low surface roughness. The magnetization orientation in the present 20 nm sample is in-plane and the magnetic
moment of nearly 1.43 μB (f.u.)−1 at 300 K agree with the literature for bulk samples and comply with the Slater–Pauling rule for half-metallic ferromagnetic full-Heusler alloys (2 μB (f.u.)−1 at 0 K).

Similarly, the extrapolated Curie temperature $T_C = 378.5 \pm 1.5$ K corresponds to prior publications and confirm the overall good quality of the films. Transport measurements show an RRR of 1.49 and a residual resistivity of $\approx 142.7 \mu \Omega \cdot \text{cm}$ at 2 K. The carrier density is in the range between $1.6 \times 10^{21}$ and $2.4 \times 10^{22} \text{cm}^{-3}$. The measurements show a positive Hall constant and a positive dependence on the temperature. The well-ordered film exhibits an anomalous Hall conductivity $\sigma_{xy} = 60 \text{S cm}^{-1}$ at low temperatures. Deeper analysis shows that the dominant conductivity mechanism is skew scattering, though it is less prominent than in other cobalt-based Heusler alloys. This indicates a higher contribution from intrinsic mechanisms, which is expected in WSMs.

This shows that conventional sputtering technique can be used to grow thin films of WSMs for further investigation into spintronic application. The films show behavior closely resembles the physical properties of bulk samples. The upturn in resistivity is intriguing and deserves deeper research but it is not the focus of this work.

4. Experimental Section

Epitaxial thin films were grown using a UHV magnetron sputtering system with a base pressure $p_0 < 3 \times 10^{-9}$ mbar. DC and radio frequency (RF) sources were used to cosputter from 3 in. targets of highly pure Co, Ti (both DC), and Ge (RF) targets. MgO (001), MgAl2O4 (001), and SrTiO3 (001) substrates were initially used to find the best possible foundation. All substrates were chemically cleaned prior to insertion into the sputtering chamber. The procedure consisted of 15 min of ultrasonic treatment in acetone, followed by 15 min in ethanol and 5 min in deionized H2O. TiN and Cr seed layers were also used to promote good crystalline growth, due to their very smooth growth and low lattice mismatch of $\approx 1\%$, respectively. Deposition temperature ($T_D$) of the Co2TiGe was varied between 200 and 900°C. All samples were protected by a 3 nm electron-beam-evaporated MgO capping layer to prevent degradation. Carrying out transport measurements without compromising the results, required an insulating buffer, unlike Cr and TiN. Thus, this article focuses on a sample grown on a MgO substrate with an adjacent homoepitaxial layer of $\approx 5$ nm of MgO. The substrate was annealed for 30 min at 1000°C to ensure a clean surface. Subsequently, the substrate was cooled down to 450°C, and the MgO was deposited by e-beam evaporation at a deposition pressure $p_{de} = 4 \times 10^{-8}$ mbar. The MgO deposition was followed by annealing the substrate at 850°C for 30 min to enhance the crystalline quality of the MgO film. The CTC deposition followed, after cooling down the sample to 700°C and waiting for 30 min, at a process pressure $p_{CTC} = 3.4 \times 10^{-5}$ mbar of Ar as sputtering gas. The film composition was verified by X-ray fluorescence spectroscopy.

The crystallographic properties of the CTC samples were analyzed at room temperature in a Philips X’Pert Pro MPD X-ray diffraction system, equipped with a Cu source and Bragg–Brentano optics. Magnetization measurements were carried out in a VSM at temperatures between 50 and 370 K with magnetic fields of up to 4 T. Transport measurements were carried out in a closed cycle helium cryostat at temperatures between 2 and 300 K and fields up to 4 T.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

anomalous Hall effect, ferromagnetic half-metals, Heusler compounds, Weyl semimetals

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