Scaling analysis of anomalous Hall resistivity in the Co$_2$TiAl Heusler alloy

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Abstract
A comprehensive magnetotransport study including resistivity ($\rho_{xx}$), isothermal magnetoresistance, Hall resistivity ($\rho_{xy}$) and magnetization have been carried out at different temperatures on the Co$_2$TiAl Heusler alloy. Co$_2$TiAl alloy shows a paramagnetic to ferromagnetic (FM) transition below the Curie temperature ($T_C$) $\sim$ 125 K. In the FM region, resistivity and magnetoresistance reveal a spin flip electron–magnon scattering and the Hall resistivity unveils the anomalous Hall resistivity. Scaling of anomalous Hall resistivity with resistivity establishes the extrinsic scattering process responsible for the anomalous Hall resistivity; however, skew scattering is the dominant mechanism compared to the side-jump contribution. A one to one correspondence between magnetoresistance and side-jump contribution to anomalous Hall resistivity verifies the electron–magnon scattering being the source of side-jump contribution to the anomalous Hall resistivity.

Keywords: Heusler alloys, magnetotransport, electron magnon scattering, anomalous Hall effect, skew scattering, side jump scattering

(Some figures may appear in colour only in the online journal)

1. Introduction

Ferromagnetic Heusler alloys (HA) are known for their multifunctional properties such as shape memory effect, magnetocaloric effect, large magnetic field induced strain, high spin polarization, topological properties, large anomalous Hall effect etc [1–4]. In recent times, the cobalt based Heusler alloys have attracted the attention of researchers due to functional properties like half metallicity, spin polarization and anomalous Hall effect (AHE) which makes them possible candidates for spintronics applications [5–8].

Anomalous Hall effect is observed in the ferromagnetic materials resulting from spontaneous magnetization of the material hence present even in absence of magnetic field and can be described by the equation:

$$\rho_{xy} = \rho_{xy}^{OH} + \rho_{xy}^{AH} = R_0 H + 4\pi M_S R_S$$

where $\rho_{xy}^{OH}$ and $\rho_{xy}^{AH}$ are the ordinary and anomalous Hall resistivity respectively, and $H$ is the applied magnetic field. The coefficients $R_0$ and $R_S$ are characterized by the strength of ordinary ($\rho_{xy}^{OH}$) and anomalous ($\rho_{xy}^{AH}$) Hall resistivity [9]. The ordinary Hall effect (OHE) is classically explained by the Lorentz force deflecting the moving charge carriers whereas, for anomalous Hall effect, in general, three scattering mechanisms are considered in the literature that explain the origin of anomalous Hall resistivity. One of them is referred to as Smit asymmetric scattering or skew scattering mechanism [10, 11], which follows a quadratic dependence with longitudinal resistivity,

$$\rho_{xy}^{AH-SK} \propto \rho_{xx}^2.$$ 

The second one is side-jump mechanism proposed by Berger [12], which follows a quadratic dependence with longitudinal resistivity,

$$\rho_{xy}^{AH-SJ} \propto \rho_{xx}^2.$$ 

Both these mechanisms have extrinsic origin, while the third mechanism is the intrinsic one which arises from spin orbit coupling and depends on the band structure inherent to the material, leading to

$$\rho_{xy}^{AH-I} \propto \rho_{xx} \rho_{xx}^{2}.$$ 

This was discovered by Karplus and Luttinger [13, 14]. Recently, Karplus Luttinger (KL) mechanism has been interpreted in the language of Berry curvature formalism [15–17].
Hall effect has been studied in cobalt based Heusler alloys also, in order to understand their scattering mechanism. For example, a temperature independent anomalous Hall effect proportional to magnetization has been observed in Co$_2$CrAl suggesting the intrinsic mechanism in this alloy [18]. The AHE study on the thin films of Co$_2$FeAl and Co$_2$FeSi by Imort et al suggested the skew scattering mechanism influenced by crystalline quality [19], whereas in another detailed study by Hazra et al on Co$_2$FeSi thin films, side-jump and skew scattering are found to be the dominant mechanism in comparison to the intrinsic contribution to anomalous Hall resistivity [20]. For Co$_2$MnSi$_{1-x}$Al$_x$ alloys the AHE scaling with resistivity showed the contributions both from skew scattering as well as intrinsic mechanism [21]. The phenomenon of anomalous Hall effect is intriguing due to the rich physics, complexity involved in understanding its origin and the possibility of applications in electronic devices.

Co$_2$TiAl is a cobalt based Heusler alloy having L2$_1$ crystal structure with space group Fm3m. It undergoes a PM to FM second order phase transition at ∼125 K. Cobalt atom contributes to the magnetic moment of this alloy and Co$_2$TiAl shows the typical characteristic of soft ferromagnetic material with saturation magnetic moment ∼0.7 μB [22, 23]. In this paper, we report for the first time a comprehensive magneto-transport study on Co$_2$TiAl alloy including resistivity, magnetoresistance and Hall resistivity along with magnetization study. Co$_2$TiAl is lesser explored cobalt based Heusler alloy specially in case of transport studies, whereas there are few magnetization studies present in the literature [24]. A detailed magnetoresistance and AHE analysis explains the transport mechanism in this alloy.

2. Experimental details

Co$_2$TiAl is synthesized by arc melting the stoichiometric amounts of Co, Ti and Al in high purity argon atmosphere. The as-cast ingot obtained is vacuum annealed (better than 5 × 10$^{-6}$ mbar) at 1073 K for a week. The sample is characterized by powder x-ray diffraction (XRD) using the Bruker D8 Advance diffractometer (Cu K$_\alpha$, λ = 1.54 Å) for which a part of the sample was crushed using agate mortar to obtain the powdered specimen. A rectangular bar shaped piece from the remaining sample was cut for transport measurements. Resistivity and magnetoresistance are measured by standard four probe technique whereas a five probe method is used for measuring the Hall resistivity using the AC-transport option of PPMS down to 2 K and magnetic fields up to 9 T. For resistivity measurements, voltage is measured along the direction of applied current (x-direction) on the sample surface plane, whereas for Hall resistivity the Hall voltage is measured transverse (y-direction) to the direction of current and magnetic field is applied perpendicular to the sample surface and current direction, for both resistivity and Hall measurements. The current, voltage, and Hall contacts are made on the sample through fine copper wires using indium. The dimensions of the sample used are 6.4 × 4.1 × 1.2 mm$^3$. DC magnetization is measured in the 16 T vibrating sample magnetometer (VSM) option of PPMS, QD USA.

![Figure 1](image-url) - Figure 1. (a) Red circles show the room temperature powder x-ray diffraction pattern along with its Rietveld refinement (black line). The blue line shows the difference between observed and calculated data along with the Bragg peaks (vertical green lines). (b) Crystal structure of Co$_2$TiAl.

3. Results and discussion

3.1. X-ray diffraction

Figure 1(a) shows the x-ray powder diffraction pattern along with its Rietveld refinement, the difference between observed and fitted pattern along with the Bragg peak positions and figure 1(b) shows the cubic crystal structure of the sample. The XRD pattern shows no impurity peaks. Rietveld refinement of the XRD data confirms the single phase nature of the sample and its crystallization in cubic L2$_1$ structure having space group Fm3m. Cobalt occupies ($\frac{2}{2}$, $\frac{1}{2}$, $\frac{1}{2}$) whereas Ti and Al occupy (0, 0, 0) and ($\frac{2}{2}$, $\frac{1}{2}$, $\frac{1}{2}$) Wyckoff positions respectively. The lattice parameter obtained from Rietveld refinement is found to be 5.85 Å which is in agreement with the literature [22]. The Rietveld refinement also suggests the stoichiometric formation of the sample i.e. Co$_2$:Ti:Al = 2.04(1):0.99(1):1.03(2).

3.2. Resistivity and magnetoresistance

Figure 2(a) shows the temperature dependent resistivity in zero magnetic field, taken during cooling and heating cycles and figure 2(b) shows the temperature dependent magnetization curve in the presence of a field of 500 Oe. Resistivity decreases gradually as the temperature is decreased from 300 K down to 5 K signifying the metallic nature of the sample in this temperature range. Resistivity changes slope at ∼125 K which corresponds to the temperature driven phase transition from PM to FM state at the Curie temperature ($T_C = 125$ K) [23]. $T_C$ here is determined as the point of inflection in the derivative of zero field resistivity curve as shown in the inset of figure 2(a). The residual resistivity ratio (RRR) value, defined as $\rho_{300K}/\rho_{5K}$, is 6.75 for the sample. Figure 2(b) shows magnetization as a function of temperature at 500 Oe and its derivative (dM/dT) is shown in the inset which features a clear transition at ∼123 K and corroborates well with $T_C$ obtained from resistivity measurements.

According to Matthiessen’s rule, the total resistivity of crystalline metallic sample is the sum of all the resistivity contributions resulting from various scattering processes which can
be expressed as:

\[ \rho(T) = \rho_0 + \rho_{e-m} + \rho_{e-p} + \rho_{e-e} \]  

where \( \rho_0 \) is the residual resistivity, \( \rho_{e-m}, \rho_{e-p} \) and \( \rho_{e-e} \) are the electron–magnon, electron–phonon and electron–electron scattering contributions to the total resistivity. \( \rho_0 \) is temperature independent part arising from lattice defects, imperfections or disorder. \( \rho_{e-p} \) varies linearly with temperature whereas \( \rho_{e-m} \) and \( \rho_{e-e} \) have quadratic temperature dependence. The temperature dependent resistivity is analyzed in the temperature range of 6 K–50 K, 50 K–110 K and 150 K–300 K in order to extract the different scattering contributions to the resistivity. Figure 3(a) shows the temperature dependent resistivity at zero-field along with the theoretical fits and figure 3(b) shows \( \rho_{xx}(T) \) at various constant magnetic fields. In the high temperature range i.e. 200 K \( \leq T \leq 300 \) K, a linear temperature dependence of resistivity is observed as a result of the electron–phonon scattering. Below \( T_C \), in the temperature range of 50 K–110 K, a combination of linear and quadratic temperature dependence fits well. The values of the coefficients of linear and quadratic terms in the temperature region 50 K–110 K are \( 4.8(2) \times 10^{-8} \Omega \text{ cm K}^{-1} \) and \( 2.43(1) \times 10^{-9} \Omega \text{ cm K}^{-2} \) respectively. This linear dependence has been identified as the Bloch–Gruneisen behaviour in earlier reports [25]. Below 50 K, only a \( T^2 \) dependence is obtained. These fittings have also been applied for the data in presence of magnetic fields but for the sake of clarity these fittings are not shown in the figure 3(b). Scattering from both electron–electron and electron–magnon follows the \( T^2 \) relation but, the strength of scattering resulting from electron–electron is insensitive to magnetic fields whereas, the strength of electron–magnon scattering gets suppressed with external applied magnetic fields. To determine the origin of \( T^2 \) dependence, the magnetic field dependence of such scattering strength has been determined. Coefficient of \( T^2 \) term \( (B) \) obtained from the fitting between 6 K–50 K and 50 K–110 K are plotted against the respective fields in figures 4(a) and (b) showing a decreasing trend with the increase in magnetic field; suggesting the \( T^2 \) behaviour originates from electron–magnon scattering. The indication of electron–magnon scattering has also been reported by earlier specific heat and resistivity measurements [23, 26].
magnetization curves at various temperatures demonstrating the equation (3) at 30 K and 50 K. Inset shows the enlarged view of Δρ_{xx} at 10 K and 2 K.

state. This reduces the total amount of magnons and thus the electron–magnon scattering. The decrease in the magnitude of resistivity drop at lower temperatures is an outcome of reduced electron–magnon scattering. The high field dependence of resistivity due to electron–magnon scattering in the PM state is known as anomalous Hall effect. The anomalous Hall coefficient (R_S) is determined from the magnetization data (fig 6(b)). The temperature dependence of R_S and ρ_{xx} is shown in figures 7(a) and (b) respectively. It is observed that the R_S increases with temperature similar to that of the longitudinal resistivity. The anomalous Hall resistivity (ρ_{xy}^{AH}) or R_S and the longitudinal resistivity (ρ_{xx}) follow a scaling relation based on the scattering mechanisms involved. In ferromagnets, the anomalous Hall coefficient R_S scales with longitudinal resistivity ρ_{xx} as:

\[ R_S = a ρ_{xx} + b ρ_{xx}^2, \]

where the first and second terms on right-hand side arise from skew scattering (SK) and side-jump (SJ) or intrinsic (I) contributions respectively to the anomalous Hall effect [10–14].

It has been shown that the temperature independent residual resistivity (ρ_{xx0}) and temperature dependent part of the longitudinal resistivity (ρ_{xxT}) have distinct role in driving the AHE [28, 29]. Therefore, the scaling between the R_S and ρ_{xx} has been revisited taking ρ_{xx0} and ρ_{xxT} both into account i.e. by replacing ρ_{xx} by (ρ_{xx0} + ρ_{xxT}), equation (4) can be written as:

\[ R_S(T) = (a_0 ρ_{xx0} + a_1 ρ_{xxT}) + β_0 ρ_{xx0}^2 + γ ρ_{xx0} ρ_{xxT} + β_1 ρ_{xxT}^2. \]

Here, the term (a_0 ρ_{xx0} + a_1 ρ_{xxT}) is the total skew scattering (R_{SK}^S) contribution to R_S (T), γ ρ_{xx0} ρ_{xxT} is the cross-term arising from competition between different scatterings and (β_0 ρ_{xx0}^2 + β_1 ρ_{xxT}^2) is the total contribution from the side-jump scattering and/or intrinsic effect (R_{SI}^S). Equation (5) depicts the temperature dependence of anomalous Hall coefficient. We
We have scaled $R_S$ with $\rho_{xx}$ in order to extract the scattering contributions that drive the AHE in this system. To begin, the general scaling for $R_S(T)$ defined in equation (4) is used to scale AHE but it does not provide a good fit to the data suggesting that it does not completely address the inherent mechanism involved in the AHE of this system. Individual linear and quadratic relations also do not provide the good fit. In order to separate the temperature dependent and temperature independent contributions to $R_S$, scaling of $R_S$ with $\rho_{xx}$ discussed in equation (5) is used to fit the data and a good fit is obtained which is shown in figure 7(c).

Figure 8(a) shows the dependency of each individual term used in equation (5) on temperature, which are basically the different contributions to $R_S(T)$. The terms $\alpha_0 \rho_{xx}$ and $\beta_0 \rho_{xx}^2$ are temperature independent, whereas the terms $\alpha_1 \rho_{xx} T$, $\gamma \rho_{xx} \rho_{xx} T$ and $\beta_1 \rho_{xx}^2 T$ are temperature dependent terms. The cross-term $\gamma \rho_{xx} \rho_{xx} T$ contributes weakly to $R_S(T)$ whereas the term $\beta_1 \rho_{xx}^2 T$ has a weak temperature dependence up to 50 K and increases in magnitude above this temperature, but $\alpha_1 \rho_{xx} T$ contributes dominantly to $R_S(T)$ compared to all other terms as seen in figure 8(a). Irrespective of magnitude, a finite contribution from all these terms is obtained at all the temperatures. The scaling relation defined in equation (5) also reproduces the temperature dependence of $R_S$ as shown by the solid line in figure 8(a). Figures 8(b) and (c) show the total contribution to the $R_S$ arising from skew scattering ($R_{SK}^S$) and side-jump/intrinsic effect ($R_{SJ,I}^S$) respectively and their temperature dependence. In terms of magnitude, the skew scattering is nearly four times larger than the side-jump/intrinsic contributions thereby dominating the $R_S(T)$. Experimentally, it has been a challenge to distinguish the intrinsic and side-jump contributions as both show the scaling $R_S \propto \rho_{xx}^2$. The intrinsic AHE has been shown to be weakly temperature dependent where $\sigma_{AH}^{xx}$ is almost constant even though $\rho_{xx}$ has a temperature dependence [9, 30].

Further it has been proposed that the electron–magnon scattering can lead to the temperature dependence of the side-jump contribution [31]. As discussed earlier, our system has electron–magnon contribution to resistivity and the magnetoresistance is governed by electron–magnon spin flip scattering. Therefore, magnons could be responsible for side-jump contribution asserting the theoretical prediction by Yang et al. [31]. To confirm this possibility, we have scaled the temperature dependence of change in resistivity with field ($\Delta \rho_{xx}$) and $R_{SK}^S$ as shown in figure 9. Inset of figure 9 shows the linear relation
between $\Delta \rho_{xx}$ and $R_S^{\text{xy}}$. A good correlation between MR and $R_S^{\text{xy}}$ as well as the linearity between them confirms that the $R_S^{\text{xy}}$ ($T$) originates from the spin flip electron–magnon scattering.

Figure 10 shows Hall resistivity as a function of temperature at 1 T. As temperature is reduced $\rho_{xy}$ slowly increases with a peak at $T_C \sim 125$ K, which is clearly seen in the inset of figure 10, followed by reduction in $\rho_{xy}$. Large $\rho_{xy}$ at $T_C$ could be due to spin fluctuations where PM to FM order sets in. The temperature dependent $\rho_{xy}$ values agree well with those obtained from the $\rho_{xy}$ isotherms at 1 T in the FM region.

4. Summary

In conclusion, a detailed study of magnetotransport properties of cobalt-based Heusler alloy Co$_2$TiAl by resistivity, magnetoresistance and Hall resistivity has been carried out. Analysis of temperature dependent $\rho_{xx}$ shows the manifestation of electron–magnon scattering below $T_C$. In this regime the magnetoresistance is negative and governed by spin flip electron–magnon scattering. Hall resistivity shows a large change at PM to FM transition. Anomalous Hall resistivity is observed below $T_C$ which scales with longitudinal resistivity. Scaling of anomalous Hall resistivity shows that AHE in this system is driven by extrinsic mechanism viz skew scattering and side-jump scattering mechanisms, however skew scattering dominates the $R_S$ ($T$). Side-jump contribution to the anomalous Hall resistivity correlates well with the magnetoresistance, confirming the origin of side-jump contribution to be the electron–magnon scattering.

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