Thickness dependence of magnetic anisotropy and intrinsic anomalous Hall effect in epitaxial Co₂MnAl film

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Abstract: We have investigated the thickness dependence of magnetic anisotropy and intrinsic anomalous Hall effect (AHE) in single-crystalline full-Heusler alloy Co₂MnAl (CMA) grown by molecular-beam epitaxy on GaAs(001). The magnetic anisotropy is the interplay of uniaxial and the fourfold anisotropy, and the corresponding anisotropy constants have been deduced. Considering the thickness of CMA is small, we ascribe it to the influence from interface stress. The AHE in CMA is found to be well described by a proper scaling. The intrinsic anomalous conductivity is found to be smaller than the calculated one and is thickness dependent, which is ascribed to the influence of chemical ordering by affecting the band structure and Fermi surface.

Keywords: Spintronics; Heusler alloy; Molecular-beam epitaxy; Magnetic properties

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I. INTRODUCTION

Co-based full-Heusler alloy in a chemical form of Co_2YZ have attracted much attention in recent years due to high Curie temperature and high spin polarization, which are promised properties for the technological development of spintronics [1, 2]. These ferromagnets possess a gap at Fermi level in the minority band but exhibit metallic behavior in the majority band, therefore the spin-polarization at the Fermi level is considered to be 100% [3-5]. Recently, some of these alloys including Co_2MnSi , Co_2FeAl and $\text{Co}_2\text{Fe}_x\text{Mn}_{1-x}\text{Si}$ have been incorporated into magnetic tunnel junctions or current-perpendicular-to-plane giant magnetoresistance as ferromagnetic electrodes, achieving relatively high magnetoresistance [6-9]. Therefore the integration of Cobalt-based Heusler alloys as a ferromagnetic electrode in spintronic devices requires detailed investigation of their magnetic properties such as magnetic anisotropy and spin-dependent transport properties such as anomalous Hall effect (AHE).

In connection with spintronics, the AHE is presently receiving new attention. Electrons moving through a ferromagnet will acquire a transverse velocity with opposite directions for different spin orientations due to spin orbit coupling (SOC), since the charge currents have usually a net polarization, this spin-dependent transverse velocity will result in a net transverse anomalous Hall voltage [10, 11]. A major challenge in this field is to clarify the microscopic origin of the AHE, which has been a controversial subject for more than half a century. Karplus and Luttinger have proposed that the intrinsic AHE arises from the transverse velocity of Bloch electrons

induced by SOC together with interband mixing [12-17]. On the other hand, the extrinsic mechanisms including skew scattering and side jump come from the asymmetrical scattering of conduction electrons due to SOC [18-20]. To explore and distinguish the possible mechanisms of AHE, a unified scaling describing the AHE resistivity ρ_{AH} in terms of the longitudinal resistivity ρ_{xx} has always been investigated. In contrast to the conventional picture that ρ_{AH} scales with the total resistivity irrespective of its sources, recent experimental studies have revealed that both ρ_{AH} and the scaling relation are qualitatively different for various types of electron scattering.

In the previous work on investigating the AHE in Heusler alloys, a common practice is tuning the longitudinal resistivity of ferromagnetic material by varying the content or annealing at different temperatures to study the scaling between ρ_{AH} and ρ_{xx} [21-23]. This approach suffers from the deficiency that the different content not only provides additional scattering centers that modulate the extrinsic AHE contribution, but also could modify the electronic structure and make the interpretation of intrinsic AHE difficult. Utilizing the finite size effect on electric resistivity in the thickness regime where the band structure is preserved with little modification, this deficiency can be overcome. Working on epitaxial GaAs/Fe films, Tian *et al* have limited the scattering of electrons to two sources, one by interface roughness and another by phonons, with independent control on their strengths through the film thickness and sample temperature [24]. A comprehensive scaling between the anomalous Hall resistivity and longitudinal resistivity has been finally

established, giving

$$\rho_{AH} = \alpha\rho_{xx0} + \beta\rho_{xx0}^2 + b\rho_{xx}^2 \quad (1)$$

where ρ_{xx0} is the residual resistivity induced by impurity scattering, ρ_{xx} denotes the longitudinal resistivity, α is the parameter of the skew scattering, β denotes the side jump, and b the intrinsic anomalous Hall conductivity. Using this scaling, Wu *et al* have investigated the AHE in the ultrathin film regime for Fe (001) (1–3 nm) films epitaxial on MgO(001) [25]. They have identified that the coefficient of skew scattering has a reduction from metallic to localized regime, while the contribution of side jump has inconspicuous change except for a small drop below 10 K. Furthermore, the intrinsic anomalous Hall conductivity decreases with the reduction of thickness below 2 nm. It seems that the mechanisms of AHE and the influence from localization can be clearly explored with varying the thickness of the films. The effect of the interface is similar to that of doping the bulk material with layers of impurities, and the density of these impurities can be easily controlled by means of the film thickness [26]. Therefore, the advantage of a thin film approach in AHE study is that by tuning the film thickness, the impurity density can be continuously manipulated. The investigation and clarification of these intriguing issues are of fundamental importance for better understanding the underlying physics of AHE.

In this paper, we have investigated thickness dependence of magnetic anisotropy and AHE in single-crystalline full-Heusler alloy Co_2MnAl (CMA) grown by molecular-beam epitaxy on GaAs(001). The magnetic anisotropy is ascribed to the competition between the uniaxial and fourfold anisotropy, and the corresponding

anisotropy constants have been deduced. Considering the thickness of CMA is small, we ascribe the uniaxial anisotropy to the influence from interface stress. The origin of the fourfold interface anisotropy seems to be the modification of the electronic structure at the interface. The AHE in CMA is found to be well described by the scaling given by Tian *et al.* The intrinsic anomalous conductivity is also found to be thickness dependent, which is ascribed to the influence of chemical ordering by affecting the Fermi surface. On the other hand, the variance of α and β with temperature have a small impact on the total anomalous Hall conductivity, and the localization correction is weak.

II. EXPERIMENTAL DETAILS

Co₂MnAl films with the thickness of $t=3, 5, 8, 10, 12$ nm were deposited on the more spatially isotropic $c(4\times4)$ reconstructed GaAs (001) surface by molecular-beam epitaxy at 260 °C. In order to emphasize the effect from interface and localization, the thickness of CMA in this paper is in the range of 3~12 nm. Streaky RHEED patterns emerged during deposition as shown in Figure 1(a), indicating epitaxial growth. In order to protect the surfaces from oxidation, films were capped with 2 nm of aluminum. The samples were first characterized using double crystal x-ray diffraction (DCXRD) to check the crystal structure. DCXRD patterns of the 12-nm-thick CMA film is taken as an example and shown in Fig. 1(b). Besides the (004) and (002) diffractions of GaAs substrate, we can only observe the (004) diffraction peak of CMA, indicating the A2 structure as discussed in our previous work [27]. Assuming the lattice constant to be that of GaAs is 0.5654 nm, the effective cubic lattice

constant of CMA is about 0.58 nm. The magnetic properties were investigated by a superconducting quantum interference magnetometer. The films were then patterned into Hall bars with a nominal length l of 2.5 mm and a width w of 0.2 mm using photolithography and ion-beam etching, and the transport properties were carried out in a physical property measurement system (Quantum Design PPMS-9T system).

III. RESULTS AND DISCUSSION

Hysteresis loops of CMA films measured at 300 K under the external magnetic field along the [110] and [1-10] directions are shown in Fig. 1(c) and (d) respectively, and the magnetization is normalized to the saturation magnetization M_S . In-plane uniaxial magnetic anisotropy with easy axis parallel to the [110] direction is found. All the films have small coercive field, which is not evidently changed with varying thickness. In the [1-10] direction, discontinuities appear at the so-called split field H_S which is a consequence of the superposition of the uniaxial and the fourfold anisotropy with the hard axis of the uniaxial anisotropy coinciding with an easy axis of the fourfold component [28]. The split field H_S reduces from 200 to 10 Oe as increasing t from 3 to 12 nm. It is noted that the magnetic anisotropies of all the films should be attributable to the competition between uniaxial and the fourfold anisotropy. Considering the shape of hysteresis loops, both uniaxial anisotropy constant K_U and cubic anisotropy constant K_C were deduced using the method mentioned in Ref [28]. Firstly, the inverse slope of $M(H)$ for $M=0$ is calculated by $\frac{1}{s} = \frac{\partial H}{\partial M} \Big|_{M=0}$, then K_C and K_U can be calculated using $K_C = \frac{1}{2} \frac{M_S(-1+H_S s)}{(H_S^3 s^3 + H_S^2 s^2 + H_S s + 1)s}$ and $K_U = \frac{1}{2} \frac{M_S H_S (H_S^2 + 2 + H_S s)}{H_S^3 s^3 + H_S^2 s^2 + H_S s + 1}$ respectively. The results are shown in Fig. 2, it is found that K_C increases while K_U

decreases as t increases, resulting smaller split fields. On the other word, the effect from the uniaxial anisotropy which mostly depends on the interface stress has become weak as increasing t .

For clarity we normalize the resistivity of all the films by the value at 300 K and plot the temperature dependence of $\rho_{XX}(T)/\rho_{XX}(300\text{ K})$ in Fig. 3(a). According to the Matthiessen rule, ρ_{XX} can be decomposed into the residual resistivity ρ_{XX0} at 5 K originating from impurity scattering and ρ_{XXT} caused by finite temperature excitation by entities such as phonons and magnons. In low temperature regime, the resistivity in all the films has increased obviously as the temperature decreases, which can be ascribed to the localization. As shown in Fig. 3(b) the residual resistivity ρ_{XX0} increases with decreasing film thickness. The finite size effect on the resistivity is present in the thin film, and the interface scattering become significant. Because the interface scattering is inelastic, the electron mean free path is shortened by the presence of the interface and the resistivity is enhanced. The ρ_{AH} in all the films were obtained in the same way by subtracting the ordinary Hall component determined from a linear fit to the high-field region up to $\pm 4T$. Fig. 3 (c) demonstrates a representative set of hysteretic ρ_{AH} curves for the 12-nm-thick CMA film measured at various temperatures from 5 to 300 K. The temperature dependence of ρ_{AH} is plotted in Fig. 3(d), which shows an increasing trend with temperature. In the low temperature regime, the increase of ρ_{AH} is similar to the behavior of longitudinal resistivity, and the influence of localization on the AHE will also be investigated.

Considering that the conductivity of ultrathin CMA films is small enough, it is

adequate to use the scaling given by Tian *et al* for describing the AHE in this system, which has been justified by theories and verified by experiments [24, 25]. Using this scaling, we can extract the intrinsic contribution from the total anomalous Hall conductivity. In Fig. 4(a), the ρ_{AH} is plotted versus the longitudinal resistivity square ρ_{XX}^2 . By linear fitting we found that for all the films these plots show good linearity from high temperature to low temperature, and the slope will be the intrinsic anomalous Hall conductivity b shown in Equation (1). The intrinsic anomalous Hall conductivity b of different thicknesses obtained by the linear fitting in Fig. 4(a) are plotted together in Fig. 4(b). The intrinsic anomalous Hall conductivity noticeably decreases with thickness decreasing, which indicates that the size effect can change the Berry curvature contribution in CMA. The value of b for 12-nm-thick film is about $1340 \Omega^{-1}\text{cm}^{-1}$. Kübler *et al* have computed Berry curvatures for a set of Heusler compounds using density functional calculations and the wave functions that they provide, and the anomalous Hall conductivity is obtained from the Berry curvatures [29]. Using this method, the anomalous Hall conductivity in CMA is calculated to be about $2000 \Omega^{-1}\text{cm}^{-1}$. In our work, however, the value of b for 12-nm-thick film is smaller than the calculated bulk intrinsic anomalous Hall conductivity, while the intrinsic anomalous Hall conductivity for 3 nm film drops down to about $1100 \Omega^{-1}\text{cm}^{-1}$. It is found that the anomalous Hall conductivities can dramatically influenced by the band structure and the Fermi-surface topology. The smaller intrinsic anomalous Hall conductivity is most likely that the sample is in disordered A2 crystal structure and does not have the ideal Heusler L_{21} crystal structure. On the other hand,

with varying the thickness of CMA, the chemical ordering will also be changed due to interface stress. Meanwhile, as the material approaching ultrathin films, the Berry curvature from bulk may gradually vanish. The intrinsic anomalous Hall conductivity, as the integral of all the Berry curvatures over the whole Brillouin zone, is supposed to decrease as observed in experiment. The influence of film thickness on intrinsic AHE needs further theoretical analysis in the future. On the other hand, to investigate the influence from localization in low temperature range, the skew scattering coefficient α and side-jump contribution β are deduced by tuning thickness at different specific temperatures using the method in Ref [25]. Firstly, it should be pointed out that the Berry curvature in the CMA films could be temperature independent according to the nearly liner relationship of $\rho_{\text{AH}} \sim \rho_{\text{XX}}^2$. We now focus on the low temperature regime 5~100 K and $\rho_{\text{AH}} \ll \rho_{\text{XX}}$, taking the approximation $\sigma_{\text{XX}} \approx 1/\rho_{\text{XX}}$ and $\sigma_{\text{AH}} \approx -\rho_{\text{AH}}/\rho_{\text{XX}}^2$. Then the scaling Eq. (1) can be transformed to:

$$\frac{(-\sigma_{\text{AH}}(T)-b)\sigma_{\text{XX}0}^2}{\sigma_{\text{XX}}(T)^2} = \alpha\sigma_{\text{XX}0} + \beta \quad (2)$$

Using Eq. (2), α and β have been calculated are shown in Fig. 4(c) and (d) respectively. In the range from 5 to 100 K, the value of α is about 0.0352 and the value of β is about $-2.1 \times 10^{-4} \mu\Omega^{-1}\text{cm}^{-1}$, the opposite signs of α and β indicate that the extrinsic skew scattering contribute to AHE in an opposite way to that of the side jump contribution. Both α and β have not evidently changed, indicating both skew scattering and side jump contribution have negligible change. Therefore, the variance of α and β with temperature has a small impact on the total anomalous Hall conductivity, and the localization correction is weak. It is clear that all three

mechanisms contribute to the total AHE in comparable magnitudes at low temperature, but at high temperature the intrinsic contribution becomes dominating.

IV. SUMMARY

In summary, we have investigated the thickness dependence of magnetic anisotropy and intrinsic AHE in single-crystalline full-Heusler alloy CMA. The magnetic anisotropy is the interplay of uniaxial and the fourfold anisotropy, and the corresponding anisotropy constants have been deduced. Considering the thickness of CMA is small, we ascribe it to the influence from interface stress. The AHE in CMA is found to be well described by the proper scaling $\rho_{\text{AH}} = \alpha\rho_{\text{xx}0} + \beta\rho_{\text{xx}0}^2 + b\rho_{\text{xx}}$. The intrinsic anomalous conductivity is found to be smaller than the calculated one and is thickness dependent, which is ascribed to the influence of chemical ordering by affecting the band structure and Fermi surface.

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Figure Captions

Fig. 1 (a) The RHEED pattern of CMA film. (b) DCXRD pattern of 12-nm-thick CMA film. (c) Normalized in-plane hysteresis loops of CMA films with varying thickness, and the field is applied along [110] direction. (d) Normalized in-plane hysteresis loops of CMA films with the field applied along [1-10] direction.

Fig. 2 (a) Effective cubic magnetic anisotropy constant K_C and (b) effective uniaxial anisotropy constant K_U for CMA films with varying thickness.

Fig. 3 (a) The temperature dependence of the longitudinal resistivity ρ_{XX} for the six samples. The data are normalized by the values at 300 K. The inset figure is the longitudinal resistivity of 3-nm-thick CMA in low temperature range. (b) The residual resistivity ρ_{XX0} plotted against the film thickness. (c) ρ_{AH} -H curves at different temperatures for 12-nm-thick CMA film. (d) ρ_{AH} as the function of temperature for all the films.

Fig. 4 (a) ρ_{AH} vs ρ_{XX}^2 for different thicknesses of CMA films. (b) The thickness dependence of the intrinsic anomalous Hall conductivity b . (c) and (d) skew scattering coefficient α and side-jump contribution β vs temperatures respectively.