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Influence of disorder on magnetic properties and intrinsic anomalous hall effect in epitaxial Co_2FeAl film

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ABSTRACT

We have investigated the influence of disorder on magnetic properties and intrinsic anomalous Hall effects in epitaxial single crystalline full Heusler alloy Co_2FeAl. The magnetic properties in both ordered and disordered films are proved by X ray absorption spectroscopy and X ray magnetic circular dichroism measurements. Using a proper scaling, we have extracted the intrinsic anomalous Hall conductivity (AHC) of the films. The intrinsic AHC in the as deposited films is thickness dependent, but in the annealed ones the value is nearly constant, which is ascribed to modified the Fermi surface due to disordering.

1. Introduction

Full-Heusler alloys belong to a group of ternary intermetallics with the stoichiometric composition X_2YZ ordered in an $L2_1$ type structure, in which X and Y are transition metals and Z is usually a main group element [1–3]. These ferromagnets (FM) 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%. The high degree of spin polarization is interesting for various applications in magnetoelectronics. Recently, Co-based full-Heusler alloy in a chemical form of Co_2YZ are of particular interest due to their high Curie temperatures and high spin polarization [4–6]. Some of these alloys including Co_2MnSi, Co_2FeAl and $Co_2Fe_xMn_{1-x}Si$ have been incorporated into magnetic tunnel junctions or current-perpendicular-to-plane giant magnetoresistance as ferromagnetic electrodes, achieving relatively high magnetoresistance [7–10]. Therefore, the electronic band structure and magnetic properties of the Co-based Heusler compounds have been intensively investigated both theoretically and experimentally in recent years. Notably, the Co_2YZ can transform into disordered structures, such as $B2$ type disorder due to mixing between Y and Z, $A2$ type disorder due to complete mixing of Co, Y and Z [11]. The magnetic properties will be affected by the mixing of the atoms, for example in Co_2CrAl alloy, the total magnetic moment decreases with increasing Co-Cr type disorder owing to the antiferromagnetic coupling of the anti-site Cr with the first nearest-neighbor ordinary-site Cr [12].

Furthermore, this kind of phase separation into two or more disordered phases is expected to result in reduced spin polarization because of the additional minority density of states at the Fermi level [13]. The topology of Fermi surface will correspondingly become complex, resulting in a modified spin-dependent transport properties such as anomalous Hall effect (AHE) [14]. The AHE which occurs in FM due to spin-orbit coupling (SOC) has resisted theoretical and experimental assaults for almost a century [15]. It has various origins resulting from either the extrinsic impurity scattering processes or the intrinsic mechanism which can be interpreted as the Berry curvature of the occupied Bloch states [15]. The possible mechanisms of AHE can be explored through giving a unified scaling describing the AHE resistivity ρ_{AH} in terms of the longitudinal resistivity ρ_{xx}. Karplus and Luttinger have proposed that the intrinsic AHE arises from the transverse velocity of Bloch electrons induced by SOC together with interband mixing, giving $\rho_{AH}\sim \rho^2_{xx}$ [16–21]. On the other hand, the extrinsic mechanisms including skew scattering and side jump come from the asymmetrical scattering of conduction electrons due to SOC, giving $\rho_{AH}\sim\rho_{xx}$ and $\rho_{AH}\sim\rho^2_{xx}$, respectively [22–24]. Recent experimental work on $Co_2FeSi_{0.6}Al_{0.4}$ and $Co_2FeGa_{0.5}Ge_{0.5}$ films revealed that the intrinsic AHE can be significantly influenced by chemical ordering with affecting the topology of Fermi surface [25].

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 temperatures [26]. They developed the experimental strategy neglecting the contribution of phonon skew scattering, giving the scaling:

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

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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 (AHC). This scaling has been justified by theories and verified by experiments. Hou et al. have investigated the AHE in epitaxial face-centered-cubic Co film grown by molecular beam epitaxy on MgO(001), and the intrinsic AHC can be separated from the scattering related extrinsic contributions and is found to be temperature independent [27]. Ye et al. have proposed a general experimental method to explore the unusual temperature dependence of AHE in Ni, which can extract the intrinsic AHC over the whole temperature range [28]. Totally speaking, 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. As an attempt, we want to use this scaling to investigate the AHE in Co-based full-Heusler alloy.

In this paper, we have investigated the influence of disorder on magnetic properties and intrinsic AHE in epitaxial single-crystalline full-Heusler alloy Co_2FeAl with comparing the as-deposited films (in *A*2 structures) and the annealed ones (in *B*2 structures). The magnetic properties are proved by x-ray absorption spectroscopy (XAS) and x-ray magnetic circular dichroism (XMCD) measurements. The magnetic moments of Co and Fe elements have been calculated. Using Eq. (1), we have also extracted the intrinsic AHC of the films. It is found that the AHC in the as-deposited films is thickness dependent, while in annealed ones the value is thickness independent. We ascribe this feature to the influence of chemical ordering by affecting the Fermi surface.

2. Results and discussion

Co_2FeAl films with the thickness of t=4, 8, 12 nm were deposited on the more spatially isotropic *c*(4×4) reconstructed GaAs (001) surface by molecular-beam epitaxy at room temperature. Reflection high energy electron diffraction was applied to in situ monitor the surface reconstruction during growth. In order to protect the surfaces from oxidation, films were capped with 2 nm of aluminum. After the deposition of the films, they were annealed at 400 °C and 500 °C in high vacuum system for 1 hour to improve their crystal structure and the atomic order. Here, we selected the thickness to be relatively small, because with tuning the film thickness the impurity density can be continuously manipulated and the electronic structure has fully developed in this thickness range. We can utilize this finite size effect to achieve independent control of the extrinsic and intrinsic contributions to the AHE.

The X-ray diffraction (XRD) profiles of the 12-nm-thick Co_2FeAl films as-deposited and annealed at various temperatures are shown in Fig. 1. Only the (004) Co_2FeAl peak and (002) and (004) GaAs substrate peaks were detected in the as-deposited films, indicating a *A*2 order structure. The (002) Co_2FeAl peak appeared after annealing at 400°C and became slightly larger after annealing at 500°C, corresponding to a *B*2 order structure or a mixing order structure of *B*2 and *A*2. In this paper, we will just compare the films of as-deposited films and the one annealed at 500 °C.

XAS and XMCD measurements are powerful techniques for obtaining microscopic information about the element-specific electronic and magnetic states [12]. Fig. 2 shows the example of XAS and XMCD spectrum at the Co-$L_{2,3}$ and Fe-$L_{2,3}$ edges in as-deposited 5-nm-thick Co_2FeAl film. All XAS spectrums are measured using total electron yield method at room temperature, which directly detects sample electron current while scanning the photon energy and strongly depended on the content of the element in the samples. We have calculated the magnetic moments of Co and Fe elements using the sum rules according to the size of the XMCD spectrum [12]. The obtained magnetic moments for Co and Fe atoms in the as-deposited films were 1.01 ± 0.1 μ_B and 2.18 ± 0.1 μ_B, while in the annealed films the values

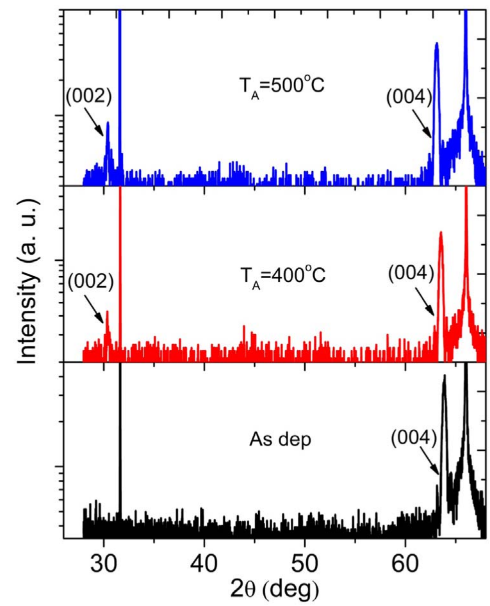

Fig. 1. XRD for Co_2FeAl films with various annealing temperature T_A.

changed to be 1.31 ± 0.1 μ_B and 2.21 ± 0.1 μ_B respectively. It indicates that the annealing has dramatically increased the magnetic moments of Co, but the value of Fe has not been evidently changed as varying the disorder [12]. Both XMCD measurements and theoretical prediction have revealed that the magnetic moment of the Co element in Co-based alloys is always smaller than that in the single Co layer, in which there is only Co-Co interaction. Therefore, in the relatively ordered Co_2FeAl film, the increased Co-Co interaction may lead to a larger magnetic moment of Co element.

In order to investigate the influence of disorder on the AHE in Co_2FeAl, 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. The transport measurements were carried out in a physical property measurement system (Quantum Design PPMS-9T system). We have normalized the resistivity of all the films by the values measured at 300 K and plot the temperature dependence of ρ_{XX} (T)/ρ_{XX} (300 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 extreme low temperature range, the resistivity in all the films has increased obviously, which has always been found in Co-based full-Heusler alloys and can be ascribed to the weak localization. The residual resistivity ρ_{XX0} of all the films increases with decreasing film thickness as shown in Fig. 3(b), which can be ascribed to the significant interface scattering due to finite size effect. Considering the interface scattering is inelastic, the electron mean free path is shortened by the presence of the interface and the resistivity is enhanced. On the other hand, the residual resistivity in the annealed films is all larger than the as-deposited ones due to increased ordering and interface mixing, but the value became almost the same in 12-nm-thick films and the interface contribution becomes weak. The transverse resistivity was measured by varying external magnetic field at different temperatures between 5 K and 300 K, and the anomalous Hall resistivity ρ_{AH} for each temperature is obtained as the extrapolation from the high field data to zero field. We have also normalized the ρ_{AH} of all the films by the values measured at 300 K and plot the temperature dependence of ρ_{AH} (T)/ρ_{AH} (300 K) in Fig. 3(c). The increase of ρ_{AH} in the low temperature regime is similar to the behavior of longitudinal resistivity. The residual AHE resistivity ρ_{AH0} has a similar behavior as ρ_{XX0} and decreases with increasing film thickness as shown in Fig. 3(d). Considering that the resistivity of all Co_2FeAl films is large enough, it is adequate to use the scaling in Eq. (1) to

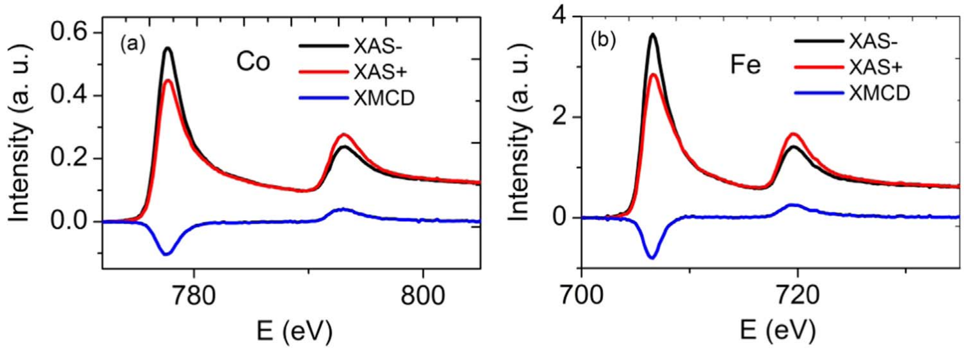

Fig. 2. XAS and XMCD spectrums at the Co-$L_{2,\ 3}$ and Fe-$L_{2,\ 3}$ edges in as-deposited 5-nm-thick Co_2FeAl film. XMCD is defined as the difference between the two XAS spectra for opposite magnetic-field directions XAS+ and XAS-.

describe the AHE in this system [26]. Using this scaling, we can extract the intrinsic contribution from the total anomalous Hall conductivity. The ρ_{AH} in as-deposited and annealed films are plotted versus the longitudinal resistivity square $\rho^2{}_{XX}$ as shown in Fig. 4(a) and (b) respectively. 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 AHC b shown in Eq. (1). The intrinsic AHC b of all the films in different thickness obtained by the linear fitting in Fig. 4(a) and (b) is plotted together in Fig. 4(d). In the as-deposited films, the intrinsic AHC noticeably decreases with thickness decreasing, which indicates that the size effect can change the Berry curvature contribution in the disordered Co_2FeAl films. On the contrary, the values of b in annealed films have not been dramatically changed. Previous studies have shown that the AHC can dramatically influenced by the band structure and the Fermi-surface topology [14]. 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 $L2_1$ crystal structure. On the other hand, with varying the thickness of Co_2FeAl, the chemical ordering will also be changed due to interface stress, and as the material approaching ultrathin films the Berry curvature from bulk may gradually vanish. The intrinsic AHC in disordered Co_2FeAl films, as the integral of all the Berry curvatures over the whole Brillouin zone, is supposed to decrease as observed in experiment. However, in the relatively ordered $B2$ type structure, the Berry curvature seems to be more stable. Finally, it should be pointed out that the Berry curvature in the Co_2FeAl films could be temperature independent according to the nearly liner relationship of $\rho_{AH} \sim \rho^2{}_{XX}$.

$$\rho_{AH0} = \alpha\rho_{xx0} + \beta\rho^2{}_{xx0} + b\rho^2{}_{xx0} \tag{2}$$

Now we attempt to further distinguish the different anomalous Hall contributions from the overall experimental data. The ρ_{AH0}/ρ_{XX0} versus ρ_{XX0} set at 5 K in the as-deposited and annealed films have been shown in Fig. 4(c). According to Eq. (1), in this case the scaling can be described by Eq. (2). The α term is identified as the skew

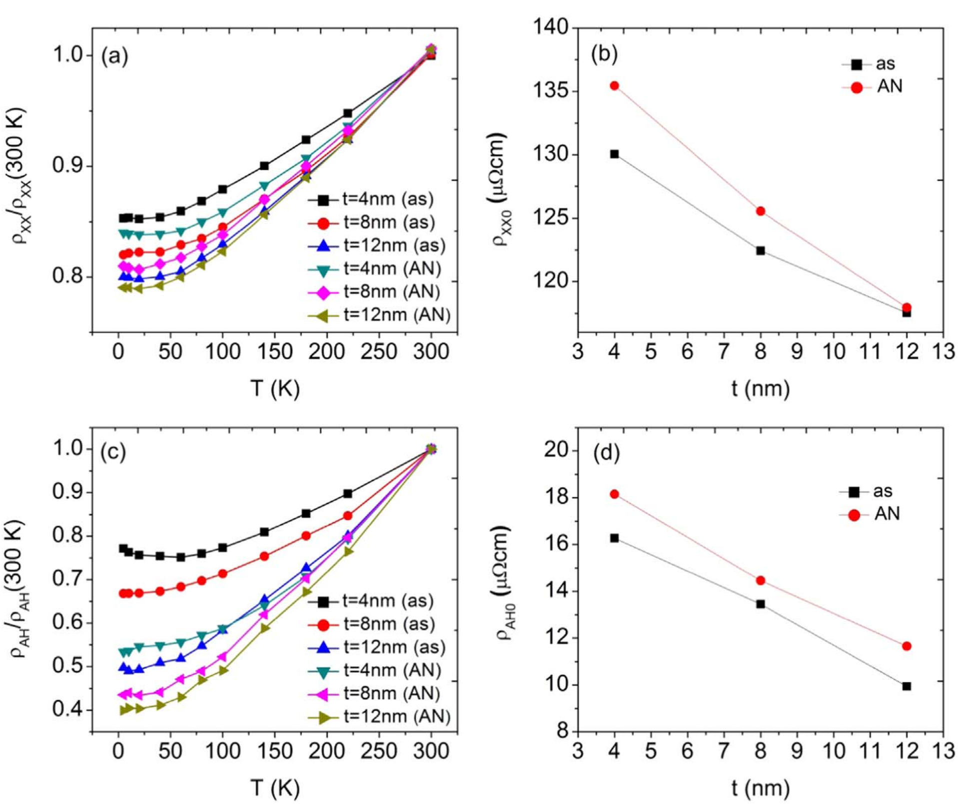

Fig. 3. (a) The temperature dependence of ρ_{XX} for the films. The data are normalized by the values at 300 K. "as" denotes as-deposited films and "AN" denotes the films annealed at 500 °C. (b) The residual resistivity ρ_{XX0} plotted against the film thickness. (a) The temperature dependence of ρ_{AH} for the films. The data are normalized by the values at 300 K. (b) The residual ρ_{AH0} plotted against the film thickness.

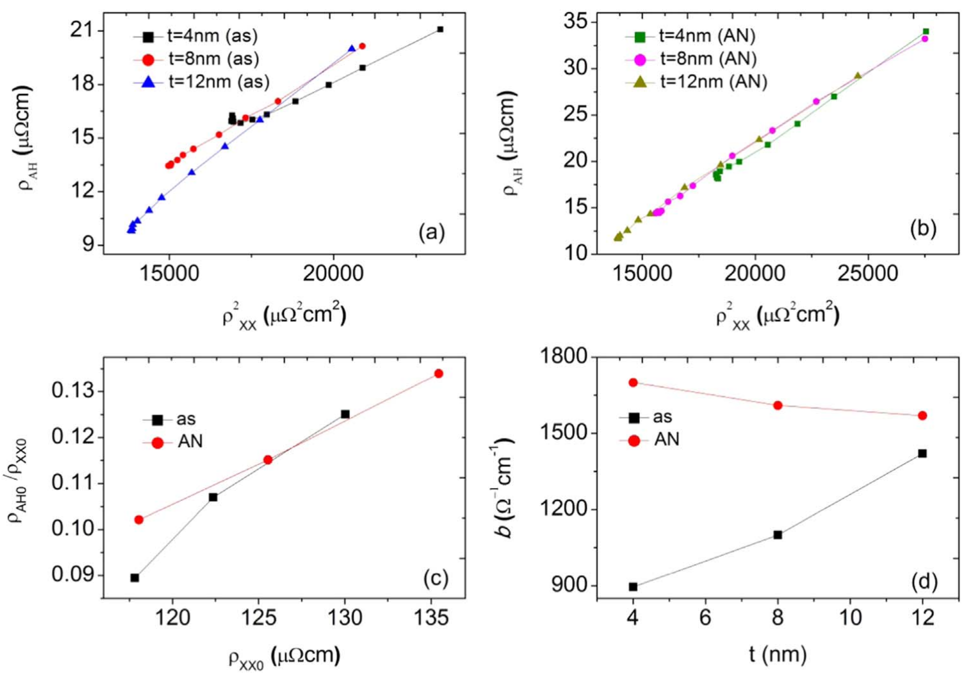

Fig. 4. (a) ρ_{AH} vs ρ^2_{XX} for different thicknesses of as-deposited Co_2FeAl films. (b) ρ_{AH} vs ρ^2_{XX} for different thicknesses of annealed Co_2FeAl films. (c) The ρ_{AH0}/ρ_{XX0} versus ρ_{XX0} in the as-deposited and annealed films. (d) The thickness dependence of the intrinsic AHC b in as-deposited and annealed films.

scattering, and considering the nearly linear dependence of ρ_{xx0} the values can be deduced to be −0.245 and −0.115 in as-deposited and annealed films respectively. β is extrinsic origin and very likely the AHC of the side jump mechanism. Based on the slope in Fig. 4(c) and the values of b in Fig. 4(d), the β in annealed films is calculated to be a constant of 300 Ω^{-1} cm^{-1}. However, in the as-deposited films the value is changed as varying the thickness, indicating that the thickness also influence the side-jump contribution in the disordered systems.

3. Summary

In summary, we have investigated the influence of disorder on magnetic properties and intrinsic AHE in Co_2FeAl by comparing the as-deposited and annealed films. The XAS and XMCD measurements have shown that in the relatively ordered films the magnetic moments of Co have been increased. The AHE in Co_2FeAl is found to be well described by the proper scaling $\rho_{AH}=\alpha\rho_{xx0}+\beta\rho^2_{xx0}+b\rho_{xx}$. Then intrinsic AHC in the disordered films is found to be thickness dependent, while in the annealed films the value has not changed evidently. We ascribe this feature to the influence of chemical ordering by affecting the Fermi surface.

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