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2012 J. Phys.: Condens. Matter 24 482001

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The anomalous Hall effect in epitaxial face-centered-cubic cobalt films

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Received 21 September 2012
Published 22 October 2012
Online at stacks.iop.org/JPhysCM/24/482001

Abstract
The anomalous Hall effect in epitaxial face-centered-cubic Co film grown by molecular beam epitaxy on MgO(001) is investigated. The intrinsic anomalous Hall conductivity is separated from the scattering-related extrinsic contributions and determined to be \(727 \pm 1 \text{ cm}^{-1}\) and temperature independent. This result provides well defined experimental data for further theoretical justification.

(Some figures may appear in colour only in the online journal)
bulk Co is strongly anisotropic along the $c$-axis and in the $a$–$b$ plane [12]. In a study on polycrystalline film it was declared that the AHE in Co films is dominated by the intrinsic contribution, and the intrinsic anomalous Hall conductivity is determined to be $240 \, \Omega^{-1} \, \text{cm}^{-1}$ [13, 14]. These works share the common weakness that due to the polycrystalline nature, the intrinsic anomalous Hall conductivity obtained is an effective average of different crystal orientations, while the intrinsic term is only well defined for single crystal. Theoretically the intrinsic anomalous Hall conductivity has been calculated for Co in hcp and fcc structures [15], but so far the previous experimental works cannot deliver a justification for the calculation values. Therefore experimental effort on single-crystalline samples with well defined Berry curvature is highly desired. The metastable fcc Co(001) film turns out to be a nice system as yet unexplored as regards the AHE. The fourfold in-plane symmetry ensures the equivalence of the directions of longitudinal and Hall measurement. The epitaxial fcc Co can be stabilized in the thin film regime within $100 \, \text{nm}$ [16], which permits us to tune the resistivity over a large thickness range, for independent control of the extrinsic and intrinsic contributions to the AHE [7, 10]. In this paper we report our measurement of the AHE in Co(001) films and determine the intrinsic anomalous Hall conductivity, which provides substantial experimental data for further theoretical investigation.

There have been many works on the epitaxial growth of Co [16–20], but the growth of fcc Co(001) on an insulating substrate which is suitable for in-plane transport measurement is still lacking. We have achieved production of an insulating template for fcc Co(001) film by the growth of $0.8 \, \text{nm Cu}$ on MgO(001) with post-annealing at $420 \, ^\circ\text{C}$ for 30 min. The sheet resistance of $0.8 \, \text{nm Cu}/\text{MgO(001)}$ is larger than $200 \, \Omega$. Figure 1(b) shows the RHEED pattern of the annealed $0.8 \, \text{nm Cu}/\text{MgO(001)}$ sample, which indicates the formation of an fcc (001) surface. Then the Co layer is grown at room temperature with a rate of $2 \, \text{Å} \text{min}^{-1}$. The evaporation rate is monitored using a quartz microbalance. Figure 1(c) is the RHEED pattern of $30 \, \text{nm Co}$, which shows little difference from figure 1(b) indicating the fcc structure. Comparing the RHEED line profile of figure 1(a) with that of figure 1(c), the lattice constant of Co is determined to be $3.56 \, \text{Å} \pm 0.02 \, \text{Å}$. To minimize the systematic error in the film thickness in sample growth, six steps with different thicknesses of Co were evaporated on the same substrate using a shadow mask, as illustrated in figure 1(d). Before the sample was transferred out of the chamber, $4 \, \text{nm MgO}$ was evaporated as a protection layer [7, 10]. Standard Hall bars were patterned on each of the steps with different thicknesses by photolithography. The transport measurement was carried out in an Oxford cryo-free low temperature system (TeslatronPT, 9 T). The magnetic field was applied perpendicular to the film plane. The measurements of longitudinal and transverse resistances were carried out by the standard four-probe method with a Keithley 2400 source meter and a Keithley 2182 nanovolt meter.

The data sets in figure 2(a) show the temperature dependence of the longitudinal resistivity $\rho_{xx}$ for various film thicknesses $d$ from 5 to 300 K. According to the Matthiessen rule, $\rho_{xx}$ can be decomposed into the residual resistivity $\rho_{xx0}$ at 5 K originating from impurity scattering and $\rho_{xxT}$ caused by finite temperature excitation by entities such as phonons and magnons, $\rho_{xx} = \rho_{xx0} + \rho_{xxT}$. In figure 2(b) it is shown that the residual resistivity $\rho_{xx0}$ increases with decreasing film thickness. This finite size effect on the resistivity is present in the thin film sample, and the physical picture is explained as follows [21]. When the electron is confined in a thin film with thickness comparable to the mean free path, the electrons should be scattered by the interface boundary. If the scattering at the interface is ideally elastic and no kinetic energy is lost, the electron will be bounced by the interfaces until it is scattered diffusively in the volume as illustrated in figure 2(c). Under these conditions, the resistivity of thin film equals that for the bulk material, for the film interface does not contribute extra diffusive scattering to the electron. However for a real system as in figure 2(d), the interface scattering is inelastic, so the electron mean free path is shortened by the presence of the interface and the resistivity is enhanced. For the equivalent path illustrated in figure 2(d), 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. The advantage of a thin film approach in AHE study is that by tuning the film thickness, the impurity density can be continuously manipulated; meanwhile the electronic structure has fully developed in this thickness range. We utilize this finite size effect to achieve independent control of the extrinsic and intrinsic contributions to the AHE.

The transverse Hall resistance is measured in the Hall bars of each thickness. In figure 3(a) the magnetic field

Figure 1. (a)–(c) The RHEED pattern of MgO(001) substrate, 0.8 nm Cu/MgO(001) and 30 nm Co/0.8 nm Cu/MgO(001); for detailed sample conditions please see the text. (d) An illustration of the fcc Co thickness step sample.
Figure 2. (a) The temperature dependence of the longitudinal resistivity \( \rho_{xx} \) for different film thicknesses. (b) The residual resistivity \( \rho_{xx0} \) plotted against the film thickness \( d \). The blue line is a guide for the eyes. (c) An illustration of electron motion in the film without diffusive interface scattering. The yellow spot denotes the scattering in the volume area. The solid part shown in deep blue represents the real sample and the translucent part shows the equivalent electron path. (d) An illustration of electron scattering motion in the film with diffusive interface scattering. The green spot denotes the scattering at the interface. For the detailed explanation please see the text.

Figure 3. (a) The Hall resistance measured in 8 nm Co film versus magnetic field at representative temperatures. The dashed line is a linear fit of high magnetic field data extrapolated to zero field to obtain the anomalous Hall resistance. (b) The anomalous Hall resistivity versus temperature for various film thickness.

dependence of the transverse Hall resistance is plotted for 8 nm Co at selected temperatures. On increasing the magnetic field from zero field, the Hall resistance first increases quickly, mainly due to the anomalous Hall effect before magnetization saturation, and then decreases linearly with the magnetic field due to the ordinary Hall effect. The opposite sign of the two
contributions arises because the anomalous Hall effect in Co is of hole type, similar to the case for Fe, but the ordinary Hall effect in Co is produced by electron carriers. The anomalous Hall resistance is obtained by extrapolating the high field data to zero field, as shown by the dashed line in figure 3(a). The temperature dependence of the anomalous Hall resistivity $\rho_{\text{AH}}$ is plotted in figure 3(b), which shows an increasing trend with temperature.

Now we attempt to distinguish the different anomalous Hall contributions from the overall experimental data. In figure 4(a) we plot the data set at 5 K $\rho_{\text{AH0}}/\rho_{\text{xx0}}$ versus $\rho_{\text{xx0}}$, and find the linear dependence that can be described by

$$\rho_{\text{AH0}}/\rho_{\text{xx0}} = \alpha + \beta' \rho_{\text{xx0}},$$  

and we multiply $\rho_{\text{xx0}}$ on both sides and obtain

$$\rho_{\text{AH0}} = \alpha \rho_{\text{xx0}} + \beta' \rho_{\text{xx0}}^2.$$  

The $\alpha$ term is identified as the skew scattering because of its linear dependence of $\rho_{\text{xx0}}$ as it is pointed out theoretically that phonons make a much smaller contribution to skew scattering [22–24] and this conclusion is verified experimentally [7]. The $\beta'$ term shows the quadratic dependence on $\rho_{\text{xx0}}$. The skew scattering coefficient is comparable to those for the Fe/GaAs(001) and Ni/MgO(001) systems [7, 10].

Next we move to analyzing the relation between $\rho_{\text{xx}}$ and $\rho_{\text{AH}}$ from 5 to 300 K. In figure 4(b) the anomalous Hall resistivity $\rho_{\text{AH}}$ is plotted versus the longitudinal resistivity square $\rho_{\text{xx}}^2$. By linear fitting we find that the data sets share the same slope within the error bar as is shown in figure 4(b) inset. From this sample-independent feature we identify the fitted slope as the intrinsic anomalous Hall conductivity $b$, and the temperature response of $\rho_{\text{AH}}$ simply comes from the $b \rho_{\text{xx}}^2$ term. At 5 K, $b \rho_{\text{xx}}^2$ is reduced to $b \rho_{\text{xx0}}^2$, and considering the finite difference between $b$ and $\beta'$ we find that $\beta'$ in equation (2) should be decomposed into two terms: $\beta' = \beta + b$, in which $\beta$ is obviously of extrinsic origin and very likely the anomalous Hall conductivity of the side jump mechanism. So the AHE scaling over the whole temperature range is formulated as

$$\rho_{\text{AH}} = b \rho_{\text{xx}}^2 + \alpha \rho_{\text{xx0}} + \beta \rho_{\text{xx0}}^2,$$

which is in the same form as the proper scaling realized in our early work on Fe and Ni [7, 10]. It should also be noted that the intrinsic anomalous Hall conductivity in the fcc Co(001) system is constant from 5 to 300 K and the behavior is very different from that for the temperature-dependent case of fcc Ni. The intrinsic anomalous Hall effect has the origin of spin–orbit interaction, which can also be reflected in the magnetocrystalline anisotropy. Indeed, the magnetocrystalline anisotropy in Ni has strong temperature dependence [25, 26], while the bulk component of the magnetocrystalline anisotropy in fcc Co shows a much weaker temperature effect [27].

By averaging the fitting result shown in figure 4(b) inset, we determine the intrinsic anomalous Hall conductivity of fcc Co to be $727 \pm 43 \, \Omega^{-1} \, \text{cm}^{-1}$, and $\beta$ as $-311 \, \Omega^{-1} \, \text{cm}^{-1}$. The theoretical value of the intrinsic anomalous Hall conductivity for fcc Co(001) is $249 \, \Omega^{-1} \, \text{cm}^{-1}$ [15] which is much smaller than our experimental result. We notice that the lattice constant used in calculation [15] is $3.53 \, \text{Å}$, which is smaller than the value $3.56 \, \text{Å}$ for our sample, and the intrinsic anomalous Hall conductivity calculated for hcp Co(0001) shows about 40% difference from the experimental value [15, 12]. In a recent theoretical work it has been pointed out that the LDA/GGA method neglecting the on-site Coulomb interaction may cause incorrect estimation of the spin–orbit interaction induced phenomena. By including the on-site Coulomb interaction, the LDA/GGA $+ U$ calculation gives an intrinsic anomalous Hall conductivity close to the experimental values for fcc Ni and hcp Co [28]. Thus more theoretical calculation for fcc Co is needed for comparison with our experimental result.

In summary, we explored the anomalous Hall effect in fcc Co(001) film experimentally. The anomalous Hall effect in fcc Co is found to be well described by the proper scaling $\rho_{\text{AH}} = b \rho_{\text{xx}}^2 + \alpha \rho_{\text{xx0}} + \beta \rho_{\text{xx0}}^2$. The intrinsic anomalous conductivity

\begin{figure}[h]
\begin{center}
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\end{center}
\caption{(a) $\rho_{\text{AH0}}/\rho_{\text{xx0}}$ versus $\rho_{\text{xx0}}$. $\rho_{\text{AH0}}$ and $\rho_{\text{xx0}}$ are the longitudinal resistivity and anomalous Hall resistivity measured at 5 K. The blue line is the linear fit. (b) $\rho_{\text{AH}}$ versus $\rho_{\text{xx0}}^2$ fitted with equation (2). The slope is the intrinsic anomalous Hall conductivity $b$. The fitting result for $b$ is given in the inset.}
\end{figure}
is obtained as $727 \pm 43 \, \Omega^{-1} \, \text{cm}^{-1}$ with little temperature
dependence from $5 \, \text{K}$ to $300 \, \text{K}$.

This work was supported by MOST (No. 2009CB929203
MSTC and No. 2011CB921802) and NSFC (No. 10834001).

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