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Anomalous Hall effect behavior in (100) and (110) CrO₂ thin films

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First and second order magnetic anisotropy constants have been determined in (110) and (100) CrO₂ films using magnetometry and anomalous Hall effect measurements. Higher in-plane anisotropy is observed in strain-free (110) CrO₂ films as compared to strained (100) CrO₂ films, while out-of-plane magnetic anisotropy (OPMA) is stronger in (100) films. Temperature-dependent OPMA is particularly striking for (110) films with a sharp drop below 200 K, whereas for (100) films the anisotropy increases as the temperature decreases. These results are consistent with changes in the magnetization orientation with decreasing temperature, possibly caused by differences in the thermal expansion coefficient between the substrate and film.

I. INTRODUCTION

Half-metallic materials (spin polarization \( P = 100\% \)) are attractive candidates for spintronic applications when integrated into magnetoresistance devices such as magnetic tunnel junctions. These devices, in principle, should show very high magnetoresistance effects according to the Julliere model. CrO₂ is one of the most interesting half-metallic oxides, with a theoretical bandgap of about \( \sim 2.0 \) eV in its minority density of states, and it has been experimentally verified to have nearly 100% spin polarization. Moreover, the Curie temperature of CrO₂ is well above room temperature (around 395 K). All of these characteristics make CrO₂ a promising candidate for spintronic applications, as well as a magnetic material of significant fundamental interests.

Recently, the epitaxial growth of CrO₂ thin films on TiO₂ substrates, as well as on Al₂O₃ substrates, has been demonstrated via chemical vapor deposition using a two zone furnace. The crystallographic orientation of the substrate has a significant effect on the growth and the structural and magnetic properties of CrO₂. CrO₂ films deposited on (100) TiO₂ substrates exhibit a large substrate-induced strain even up to 200 nm in thickness, whereas no significant strain is induced even on 25 nm CrO₂ films deposited on (110) TiO₂ substrates. It has also been reported that this substrate-induced strain significantly affects the finite temperature saturation magnetization in (100) CrO₂ films.

Although the temperature dependence of the magnetization in strained (100) CrO₂ films has been well investigated, magnetic anisotropy has not been completely elucidated, especially for strain-free, low indexed (110) CrO₂ films. Because the magnetic anisotropy is directly associated with the thermal stability of spintronic devices, it is very important to investigate the magnetic anisotropy in CrO₂ thin films. In recent developments of magnetic recording, the anomalous Hall effect (AHE) method for studying magnetic anisotropy has been quite relevant and popular, because perpendicular anisotropy in very small features—for example, in an array of nano-sized dots—can be studied using this technique. In the present study, the temperature dependence of out-of-plane magnetic anisotropy (OPMA) values, \( K_{11} \) and \( K_{12} \), are determined and reported based on the AHE observed in strained (100) and strain-free (110) CrO₂ films. AHE measurements also qualitatively demonstrate the temperature dependence of the magnetic anisotropy in the (100) and (110) CrO₂ films. The in-plane magnetic anisotropy (IPMA) in our films is determined using standard magnetometry techniques.

II. EXPERIMENT

All CrO₂ films of (100) and (110) orientations on respective TiO₂ substrates were fabricated using a previously reported method. For our study, we prepared CrO₂ films 25 and 70 nm in thickness for both orientations. Detailed x-ray diffraction studies revealed the lattice parameters in strained (100) and relatively strain-free (110) CrO₂ films at room temperature. CrO₂ films of (110) and (100) orientations have [110] and [100] axes along the film normal direction, respectively. The common easy axis of magnetization in both (100) and (110) CrO₂ is [001], i.e., the c-axis. The other in-plane directions are [010] and [110] for (100) and (110) CrO₂ films, respectively.

In all of our CrO₂ films, temperature-dependent IPMA measurements were performed using a Quantum Design superconducting quantum interference device (SQUID) magnetometer. AHE measurements were performed in a Quantum Design physical property measurement system (PPMS) with a constant current of 300 \( \mu \text{A} \) applied along the c-axes.
of the films at temperatures of 100 to 350 K. Data were analyzed using the generalized Sucksmith-Thompson (GST) method.\textsuperscript{14,15} This method, originally derived by Sucksmith and Thompson,\textsuperscript{16} is well known for its use in the study of magnetic anisotropy, and some details of this method are provided later in this paper. The measurement geometry for the AHE in this study is shown in Fig. 1. The AHE voltage is proportional to the vertical component of the magnetization. Together with this vertical component, the AHE measurements also contain contributions from the normal Hall effect (NHE) and anisotropic magnetoresistance (AMR) in the films. The NHE component from the experimental data was subtracted as a straight line component in the higher magnetic field region where the magnetization was saturated along the film normal direction, and the AMR signal was subtracted as an even function of the applied magnetic field.

III. RESULTS AND DISCUSSION

The magnetization curves of the CrO\textsubscript{2} films at room temperature are shown in Fig. 2. The magnetization curves along the c-axis saturate at a lower magnetic field in all of the CrO\textsubscript{2} films, which indicates that the c-axis is the easy axis of magnetization. However, the magnetization curve along [010] is essentially the same as that along [001] in a 25 nm (100) CrO\textsubscript{2} film, which suggests that there is small IPMA in 25 nm (100) CrO\textsubscript{2} film. In comparison, (110) CrO\textsubscript{2} films show a distinct difference between in-plane magnetization curves in both 25 nm and 70 nm films. These results indicate that the IPMA of (110) CrO\textsubscript{2} films is much larger than that of (100) CrO\textsubscript{2} films, and this is consistent with previously reported results.\textsuperscript{7,11} However, the OPMA shows the opposite tendency—(100) CrO\textsubscript{2} films exhibit a higher magnetic anisotropy than that of (110) CrO\textsubscript{2} films (as described later in this paper). The OPMA of these films has been measured as a function of temperature in order to obtain the temperature dependence of the magnetic anisotropy.

Magnetization versus temperature behavior of the films are shown in Fig. 3 and they were obtained in a SQUID when a constant magnetic field of 5 kOe was applied along the in-plane easy direction (i.e., [001]) of the films. All films showed a magnetization value of \(~650\text{ emu/cc}\) at 10 K, which is in very good agreement with the previously reported low temperature magnetization of CrO\textsubscript{2}.\textsuperscript{11} Therefore, we considered the magnetization values of the films at different temperatures to be correct within 2\% of the measured values, with error mainly arising from small temperature fluctuations. As the temperature increased, (100) CrO\textsubscript{2} films exhibited a more noticeable decrease in the magnetization due to substrate induced strain (details are published elsewhere\textsuperscript{9,10}).

Following the GST method, the equilibrium magnetization angle can be obtained as a condition of \(\Delta M_{\theta}/\Delta \theta = 0\) (\(\epsilon\) denotes the magnetic energy per unit volume) and expressed as follows:

\[
\frac{2K_{U1}^{\text{eff}}}{M_S} + \frac{4K_{U2}}{M_S}\cos^2 \theta = \frac{\sin(\theta - \phi)}{\cos \theta \sin \theta} H, \tag{1}
\]

where \(K_{U1}^{\text{eff}}\) is the effective first order magnetic anisotropy, which includes shape anisotropy \((K_{U1}^{\text{eff}} = K_{U1} + 2\pi M_S^2)\). \(K_{U1}\) and \(K_{U2}\) include the magnetic crystalline anisotropy and additional magnetic anisotropy derived from the induced strain, which will be discussed below. \(\theta\) is the magnetization angle with respect to the film normal direction, \(M_S\) is the saturation magnetization, \(H\) is the applied magnetic field, and \(\phi\)

![FIG. 1. (Color online) Sample geometry for AHE measurement on (100) CrO\textsubscript{2} films and (110) CrO\textsubscript{2} films.](Image 52x71 to 296x175)

![FIG. 2. (Color online) Room temperature magnetization curves (normalized) of (a) 70 nm thick (110) CrO\textsubscript{2} film, (b) 70 nm thick (100) CrO\textsubscript{2} film, (c) 25 nm thick (110) CrO\textsubscript{2} film, and (d) 25 nm thick (100) CrO\textsubscript{2} film, respectively.](Image 316x533 to 560x746)

![FIG. 3. (Color online) Saturation magnetization vs temperature of (100) and (110) CrO\textsubscript{2} films 25 and 70 nm in thickness measured via SQUID.](Image 342x71 to 534x229)
is the magnetic field angle with respect to the film normal direction. Equation (1) can be simplified further into the following equations ($M_Z$ is the normalized vertical magnetization component):

$$\frac{2K_{U1}^\text{eff}}{M_s} + \frac{4K_{U2}}{M_s} M_Z^2 = xH,$$

$$M_Z = \cos \theta,$$

$$x = \frac{\sqrt{1 - M_Z^2} \cos \phi - M_Z \sin \phi}{M_Z \sqrt{1 - M_Z^2}},$$

where $\cos \theta$ is the normalized vertical component of the magnetization, which is equal to the normalized AHE voltage.

According to Eqs. (2)–(4), the results are plotted as $xH$ versus $M_Z^2$. Equation (2) indicates that the $xH$ versus $M_Z^2$ plot can be fitted by a linear function that is independent of the magnetic field angle. The intercept of the fitted curve yields $2K_{U1}^\text{eff}/M_s$, and the slope gives $4K_{U2}/M_s$. Figure 4 shows typical normalized AHE voltage curves for (a) (100) CrO$_2$ film and (b) (110) CrO$_2$ film, respectively, both 25 nm in thickness and measured at 100 K. For more accurate measurements of the magnetic anisotropy values, the AHE magnetization curves have been measured at three different magnetic field angles ($\phi = 0^\circ$, 10°, 20°) for all of the films as a function of temperature. The perpendicular magnetization of (110) CrO$_2$ film saturates at a lower field than does that of the (100) CrO$_2$ film, which indicates that (110) CrO$_2$ films have a smaller OPMA. Figures 4(c) and 4(d) show the GST curves corresponding to Figs. 4(a) and 4(b), respectively. Both GST curves are fitted well by the specific linear function, which yields $K_{U1} = (2.2 \pm 0.51) \times 10^4$ erg/cm$^3$, $K_{U2} = (1.8 \pm 0.46) \times 10^3$ erg/cm$^3$ for the (110) CrO$_2$ film and $K_{U1} = (8.9 \pm 0.42) \times 10^3$ erg/cm$^3$, $K_{U2} = (1.2 \pm 0.31) \times 10^5$ erg/cm$^3$ for the (100) CrO$_2$ film, respectively, at 100 K. The values of $K_{U1}$ are obtained by subtracting the shape anisotropy term $2\pi M_s^2$ from the $K_{U1}^\text{eff}$ values.

In the present study, we also made temperature dependent measurements on the CrO$_2$ films in the temperature range between 100 and 350 K in order to alleviate the substantial difficulty in subtracting the contribution of the normal Hall effect voltage at low temperatures, as well as the strong temperature and thickness dependence of the AHE coefficient.

The OPMA for (100) CrO$_2$ films shown in Fig. 5 is slightly higher than the anisotropy of bulk samples (2.6 $\times$ 10$^5$ erg/cm$^3$). However, the OPMA values are much higher than the corresponding IPMA values reported in the literature, primarily because of the competing influence of substrate-induced strain in the CrO$_2$ films grown on (100) TiO$_2$ substrates. Therefore, for (100) CrO$_2$ films, we conclude that the $a$-axis is the hard axis and the $c$–$b$ plane contains the easy plane of the magnetization. Further, it was observed that the easy magnetization axis changes from the in-plane $c$ to the in-plane $b$ direction with decreasing temperature. The temperature dependence of the OPMA for (100) films shows the expected trend: the anisotropy value increases with decreasing temperature. In contrast, the (110) CrO$_2$ films in Fig. 5 show an unexpected variation of $K_{U1}$ in that the OPMA increases with decreasing temperature up to 200 K and then decreases at lower temperatures. It is expected that the sign of $K_{U1}$ will change from positive to negative in the temperature region below 100 K, which indicates an out-of-plane tendency of the magnetization in these films. In contrast, the in-plane $K_{U1}$ values obtained from SQUID measurements increased monotonically with decreasing temperature (not shown). Based on the magnetic anisotropy behavior of these films, we suggest that the magnetorestriction effect that originates from the difference in the thermal...
expansion coefficients of CrO$_2$ film and TiO$_2$ is quite significant. It is known that the lattice constant of TiO$_2$ decreases with decreasing temperature. The corresponding data are not available for CrO$_2$. However, the $c$ lattice constant of CrO$_2$ has been reported to increase with decreasing temperature in the high temperature region of 0–400 °C. Therefore, it is expected that, due to the cube-on-cube epitaxy in this system, changes in the TiO$_2$ substrate will influence the CrO$_2$ lattice constants significantly, giving rise to additional magnetic anisotropy due to magnetostriction. A similar effect has been observed in the IPMA of (100) CrO$_2$ films. This effect becomes weaker with increasing film thickness, because it is dominated by the substrate–film interface region between the CrO$_2$ film and the TiO$_2$ substrate. Because our (100) CrO$_2$ films are significantly strained with thicknesses much higher than the monolayer thickness of CrO$_2$ (~0.441 nm), we did not observe any strong inverse thickness dependence of the anisotropy at any temperatures, as was observed in the ferromagnetic resonance study of ultrathin epitaxial Co (001) films by Kowalewski et al. The substrate strain effect is usually extremely weak in nonoxide metal films and relaxes within a few monolayers, in stark contrast to oxide films, in which it can remain strained to well over 100 monolayers, depending on the lattice mismatch. In the particular example of (100) CrO$_2$, as previously investigated, the room temperature IPMA has a more complicated dependence on the film thickness and saturates at film thicknesses higher than 200 nm, which coincides with complete strain relaxation in the film. In contrast, the strain free (110) CrO$_2$ films did not show any significant thickness dependence of the IPMA at room temperature for relevant thicknesses in this work, as reported in a previous study. To briefly comment on the mechanism of the AHE in our (100) and (110) CrO$_2$ films, the intrinsic contribution is expected to dominate, as the studied films are significantly impurity free and epitaxial. Moreover, impurities, if any, are mainly nonmagnetic in nature (Ti$^{4+}$ ions from the substrate) with a small spin–orbit interaction. However, a detailed quantification would require further study on, say, impurity doped CrO$_2$ samples, which is beyond the scope of this work. The primary objective of this study is to estimate the temperature dependent OPMA in strained CrO$_2$ through the AHE measurement technique, which has been presented thoroughly.

**IV. SUMMARY**

In summary, we have studied the temperature dependence of magnetic anisotropy in epitaxial CrO$_2$ films grown on (100) and (110) TiO$_2$ substrates with AHE measurements. The in-plane anisotropy is larger in (110) films than in (100) films. The magnetic anisotropy values are strongly influenced by the substrate-induced strain because of the lattice mismatch between the substrate and the film, and as a result the magnetization prefers the $c$–$b$ plane in (100) films. The smaller anisotropy values in (110) CrO$_2$ films below 200 K indicate an out-of-plane tendency of the magnetization in these films.

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