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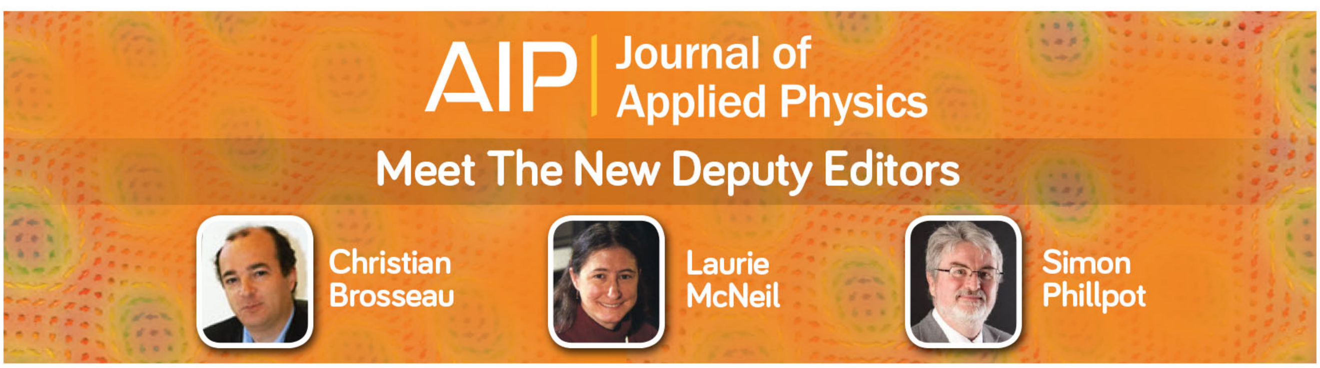

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Anomalous Hall effect behavior in (100) and (110) CrO_2 thin films

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First and second order magnetic anisotropy constants have been determined in (110) and (100) CrO_2 films using magnetometry and anomalous Hall effect measurements. Higher in-plane anisotropy is observed in strain-free (110) CrO_2 films as compared to strained (100) CrO_2 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. © *2011 American Institute of Physics*. [doi:10.1063/1.3583568]

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.[1] CrO_2 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,[2] and it has been experimentally verified to have nearly 100% spin polarization.[3–5] Moreover, the Curie temperature of CrO_2 is well above room temperature (around 395 K).[6] All of these characteristics make CrO_2 a promising candidate for spintronic applications, as well as a magnetic material of significant fundamental interests.

Recently, the epitaxial growth of CrO_2 thin films on TiO_2 substrates, as well as on Al_2O_3 substrates, has been demonstrated via chemical vapor deposition using a two zone furnace.[7,8] The crystallographic orientation of the substrate has a significant effect on the growth and the structural and magnetic properties of CrO_2.[7,9] CrO_2 films deposited on (100) TiO_2 substrates exhibit a large substrate-induced strain even up to 200 nm in thickness, whereas no significant strain is induced even on 25 nm CrO_2 films deposited on (110) TiO_2 substrates.[7] It has also been reported that this substrate-induced strain significantly affects the finite temperature saturation magnetization in (100) CrO_2 films.[9,10]

Although the temperature dependence of the magnetization in strained (100) CrO_2 films has been well investigated,[11] magnetic anisotropy has not been completely elucidated, especially for strain-free, low indexed (110) CrO_2 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 CrO_2 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.[12,13] In the present study, the temperature dependence of out-of-plane magnetic anisotropy (OPMA) values, K_{U1} and K_{U2}, are determined and reported based on the AHE observed in *strained* (100) and *strain-free* (110) CrO_2 films. AHE measurements also qualitatively demonstrate the temperature dependence of the magnetic anisotropy in the (100) and (110) CrO_2 films. The in-plane magnetic anisotropy (IPMA) in our films is determined using standard magnetometry techniques.

II. EXPERIMENT

All CrO_2 films of (100) and (110) orientations on respective TiO_2 substrates were fabricated using a previously reported method.[7,8] For our study, we prepared CrO_2 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) CrO_2 films at room temperature.[10] CrO_2 films of (110) and (100) orientationd have [110] and [100] axes along the film normal direction, respectively. The common easy axis of magnetization in both (100) and (110) CrO_2 is [001], i.e., the *c*-axis. The other in-plane directions are [010] and [− 110] for (100) and (110) CrO_2 films, respectively.

In all of our CrO_2 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 μA applied along the *c*-axes

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of the films at temperatures of 100 to 350 K. Data were analyzed using the generalized Sucksmith–Thompson (GST) method.[14,15] This method, originally derived by Sucksmith and Thompson,[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_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_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_2 film, which suggests that there is small IPMA in 25 nm (100) CrO_2 film. In comparison, (110) CrO_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_2 films is much larger than that of (100) CrO_2 films, and this is consistent with previously reported results.[7,11] However, the OPMA shows the opposite tendency—(100) CrO_2 films exhibit a higher magnetic anisotropy than that of (110) CrO_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 emu/cc at 10 K, which is in very good agreement with the previously reported low temperature magnetization of CrO_2.[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_2 films exhibited a more noticeable decrease in the magnetization due to substrate induced strain (details are published elsewhere[9,10]).

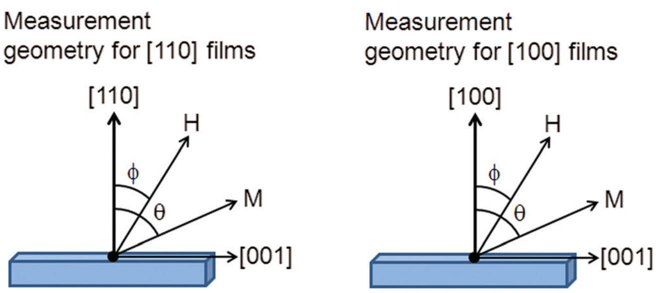

FIG. 1. (Color online) Sample geometry for AHE measurement on (100) CrO_2 films and (110) CrO_2 films.

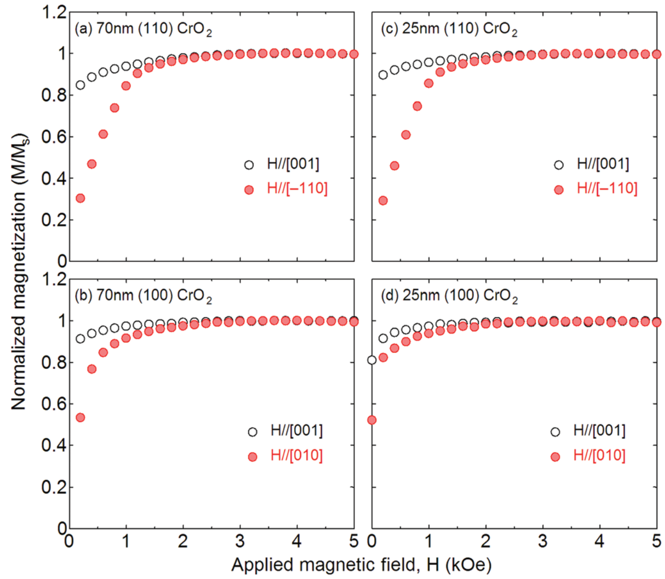

FIG. 2. (Color online) Room temperature magnetization curves (normalized) of (a) 70 nm thick (110) CrO_2 film, (b) 70 nm thick (100) CrO_2 film, (c) 25 nm thick (110) CrO_2 film, and (d) 25 nm thick (100) CrO_2 film, respectively.

Following the GST method, the equilibrium magnetization angle can be obtained as a condition of $de/d\theta = 0$ (*e* denotes the magnetic energy per unit volume) and expressed as follows:

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

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

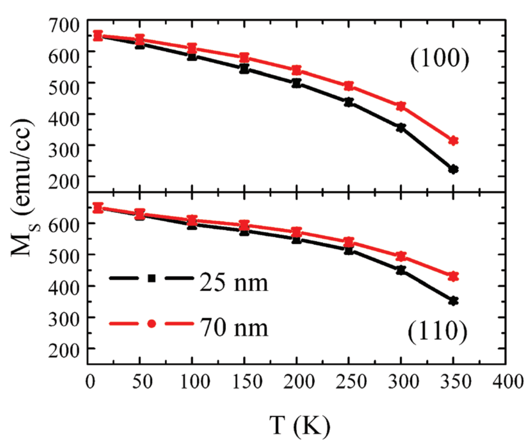

FIG. 3. (Color online) Saturation magnetization vs temperature of (100) and (110) CrO_2 films 25 and 70 nm in thickness measured via SQUID.

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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):

$$\frac{2K_{U1}^{eff}}{M_S} + \frac{4K_{U2}}{M_S}M_Z^2 = \alpha H, \tag{2}$$

$$M_Z = \cos\theta, \tag{3}$$

$$\alpha = \frac{\sqrt{1-M_Z^2}\cos\phi - M_Z\sin\phi}{M_Z\sqrt{1-M_Z^2}}, \tag{4}$$

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 αH versus M_Z^2. Equation (2) indicates that the αH 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 $2K_{U1}^{eff}/M_S$, 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 ($\phi = 0°$, $10°$, $20°$) 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, $K_{U2} = (1.8 \pm 0.46) \times 10^5$ erg/cm^3 for the (110) CrO_2 film and $K_{U1} = (8.9 \pm 0.42) \times 10^5$ 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}^{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,[17] as well as the strong temperature and thickness dependence of the AHE coefficient.[18]

The OPMA for (100) CrO_2 films shown in Fig. 5 is slightly higher than the anisotropy of bulk samples (2.6×10^5 erg/cm^3).[19] However, the OPMA values are much higher than the corresponding IPMA values reported in the literature,[7] 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.[11] 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

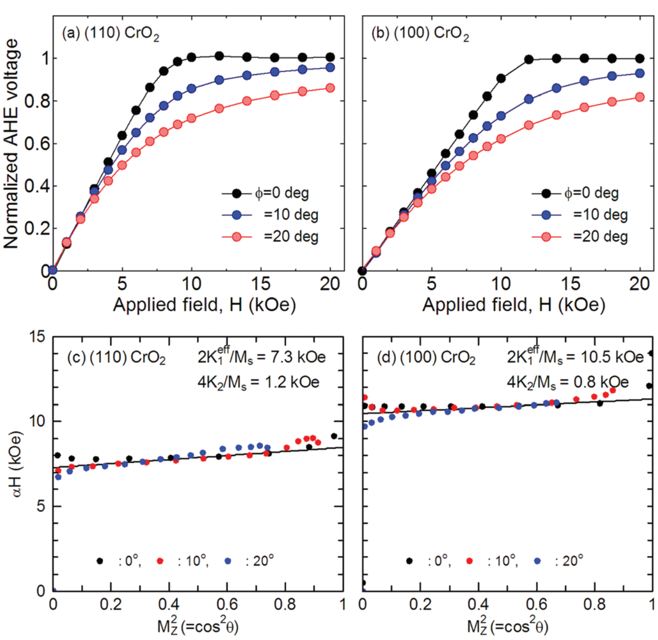

FIG. 4. (Color online) AHE magnetization curves for (a) 25 nm thick (110) CrO_2 film and (b) 25 nm thick (100) CrO_2 film, and corresponding GST curves [(c) and (d), respectively].

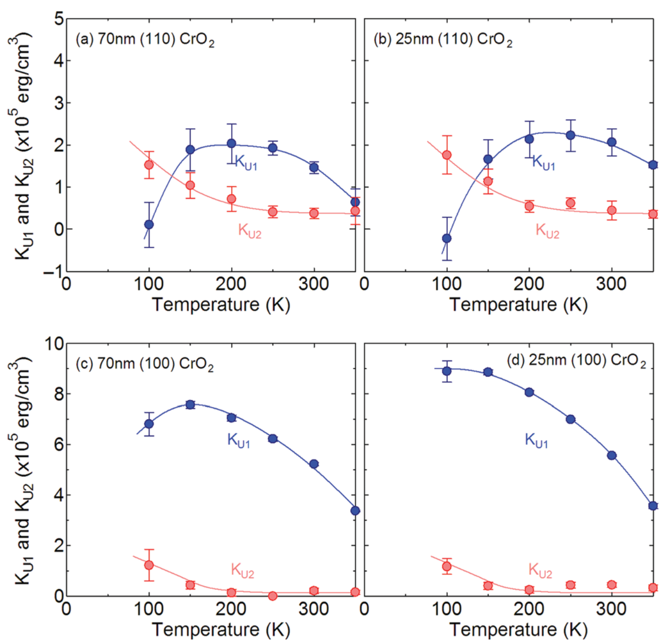

FIG. 5. (Color online) Temperature dependence of the out-of-plane magnetic anisotropy values for (a) 70 nm (110) CrO_2 film, (b) 25 nm (110) CrO_2 film, (c) 70 nm (100) CrO_2 film, and (d) 25 nm (100) CrO_2 film.

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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.[20] 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.[21] 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 magnetorestriction. A similar effect has been observed in the IPMA of (100) CrO_2 films.[11] 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 ($\sim$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*.[22] 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.[7,11] 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.[7] To briefly comment on the mechanism of the AHE in our (100) and (110) CrO_2 films, the intrinsic contribution is expected to dominate,[23] 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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