Switching Behavior and Its Strain Dependence in Epitaxial CrO_2 Thin Films

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***Abstract*—Chromium dioxide thin films grown epitaxially on TiO_2 substrates exhibited a strong in-plane uniaxial anisotropy. Field dependence of high frequency (10^6 Hz) transverse susceptibility (χ_T) measurements were used to probe the magnetic anisotropy and switching in CrO_2 films for varying temperature (10 K to 300 K) and different orientations of the applied field with respect to the magnetic easy axis. Singular peaks in χ_T were observed and are associated with the anisotropy and switching fields. The χ_T data at low temperatures reveal an unusual variation of the singular peaks. This can be described by including magnetoelastic contributions resulting from the in-plane tensile strain in the film due to lattice mismatch with the substrate.**

***Index Terms*—Magnetic anisotropy, magnetoelastic energy, transverse susceptibility.**

I. Introduction

FERROMAGNET chromium dioxide belongs to a technologically important class of magnetic oxides. It is considered to be an ideal material for spin-polarized magnetic tunnel junctions, as it is a half-metallic ferromagnet [1], i.e., it is a metal for majority electrons (spin-up), but exhibits a semiconductor type gap for minority electrons (spin-down).

While structural, transport and magnetic properties of chromium dioxide have been investigated by several groups [2]–[6], the high frequency switching characteristics have not been explored in detail. It is vital to obtain a good understanding of the dynamic magnetic response in CrO_2 for RF electromagnetic fields.

In this paper, we present RF transverse susceptibility (χ_T) measurements on high quality epitaxially grown thin films of CrO_2. Singular peaks are observed in χ_T at the characteristic anisotropy fields that differ by an order of magnitude for static magnetic field (H) applied parallel and perpendicular to the magnetic easy axis. We observe also evidence for the temperature dependent influence of strain in the films due to lattice mismatch between the film and substrate. All the observed data are well described by a generalized theoretical model, based on coherent rotation of the magnetization.

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II. Experimental

The studied CrO_2 films were 5×5 mm^2 in size and typically 2000 Å thick grown on single crystal (100) TiO_2 substrates by chemical vapor deposition (CVD) at atmospheric pressure [3]. Both, CrO_2 and TiO_2 have a rutile structure but the lattice mismatch between the film and the substrate has been noted to be significant causing an anisotropic in-plane tensile strain in the film. The structural and magnetic properties of these films have been extensively characterized [3]–[5]. An important property of the epitaxial grown CrO_2 (100) thin films is the very large in-plane uniaxial anisotropy, with well-defined magnetic easy and hard axis along the [001] (c-axis) and [010], (b-axis), respectively, that is reflected in the magnetic measurements [4], [7]. As we show in this paper the anisotropy can be probed sensitively and explored in great detail using precise transverse susceptibility experiments.

The transverse susceptibility χ_T is measured by applying a small alternating magnetic field, h_{ac}, perpendicular to the main bias field H and detecting the component of the magnetization variation along the h_{ac} direction. To probe the dynamic transverse susceptibility in the radio-frequency range we employed a very sensitive method, based on a novel tunnel-diode oscillator (TDO) technique [8], which was developed by us and used for exploring the anisotropy in various nanostructured magnetic materials [9], [10]. Thin films are placed in the core of a coil that forms part of the self-resonant ultrastable tunnel diode oscillator with a resonant frequency around 5 MHz. The change in the resonant frequency is measured as the bias field, H, is ramped from negative to positive saturation and vice versa. This quantity is proportional to the change in inductance that in turn is governed by the transverse susceptibility (χ_T) of the films. The field dependence of χ_T is plotted as a dimensionless ratio $\Delta\chi_T/\chi_T(\%) = [\chi_T(H) - \chi_T^{sat}] \times 100/\chi_T^{sat}$, where χ_T^{sat} is the transverse susceptibility at the saturating field $H = 5$ kOe.

III. Results and Discussion

In Fig. 1, we present the transverse susceptibility $(\Delta\chi_T/\chi_T(\%) \equiv \chi_T)$ obtained for three different orientations (ψ) of the bias magnetic field, with the respect to the easy axis (c-axis) when the magnetic field is ramped from positive saturation to negative one (full symbols) and vice versa (open symbols).

For $\psi = 90°$ [Fig. 1(a)] we observe the presence of identical peaks in χ_T, symmetrically located around $H = 0$ with the peak

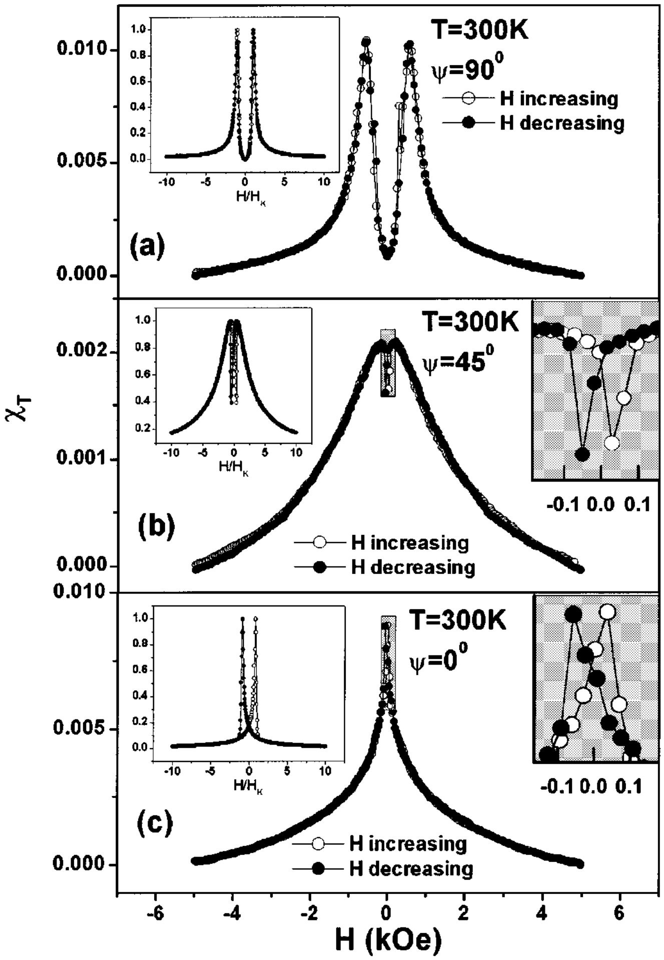

Fig. 1. Measured χ_T at room temperature for three different orientations, ψ, of the bias magnetic field with respect to the easy axis: (a) $\psi = 90°$; Inset: Calculated χ_T for the same geometry. (b) $\psi = 45°$; Inset (right): Exploded view of the data at low fields; Inset (left): Calculated χ_T for the same geometry. (c) $\psi = 0°$. Inset (right): Exploded view of the data at low fields; Inset (left): Calculated χ_T for the same geometry.

positions at ± 600 Oe, followed by a gradual approach to saturation at higher fields. The calculated transverse susceptibility based on the coherent rotation model [11], [12] is plotted in the inset of Fig. 1(a). We observe a very good qualitative agreement between the experimental data and the calculated curve. In particularly, the fields at which the singular χ_T peaks are observed in the data can be identified with the anisotropy fields $(\pm H_K)$ for this system. This is in good agreement with the values obtained from magnetic hysteresis loop measurements for this geometry [7].

Fig. 1(b) and (c) shows the measured χ_T data for the cases when H is applied at $\psi = 45°$ and parallel to the easy axis (i.e., $\psi = 0°$), respectively. The right insets are a zoom view of the low field data, and the left insets show the numerically calculated transverse susceptibility for the same bias field direction. In these two cases, unlike that for $\psi = 90°$, the experimentally obtained χ_T curves are not reversible for bipolar sweeps of the bias field and this is also reproduced by the model.

For $\psi = 45°$ [Fig. 1(b)], the experimental χ_T curves do not exhibit singular peaks, like for the other two cases of $\psi = 90°$ and $\psi = 0$, but a rounded peak followed by a singular dip at very low field, around ± 50 Oe. Note the difference in the vertical scale for the three graphs, that is consistent with the more singular character of the peaks at $\psi = 90°$ and $\psi = 0°$.

The peaks for $\psi = 0°$ [Fig. 1(c)] also occur at very low fields around ± 60 Oe, as predicted by the theoretical model. So, unlike the case in Fig. 1(a), where two peaks are observed for a unipolar field sweep, only one peak occurs for this geometry. This variation of χ_T is representative of magnetization switching and the measured peak fields are associated with the switching fields $(\pm H_s)$. Our results are consistent with the M–H hysteresis loop [7] indicating that epitaxial CrO_2 is a soft ferromagnet in this direction showing almost ideal switching characteristics. A significant property of CrO_2 that clearly emerges from our experimental and theoretical χ_T results presented in Fig. 1 is that the sample behaves like single domain particle where coherent rotation dominates the magnetization processes.

The estimated value of the anisotropy constant, using the determined 600 Oe value for the anisotropy field $(H_K = 2K_1/M_s)$ with $M_s = 650$ emu/cm^3, is $K_1 = 1.9 \times 10^5$ erg/cm^3. This value is in good agreement with values reported in other studies [3], [13]. However, we would like to present a note of caution that the large variation of H_K with ψ raises the issue whether a simple picture for the magnetocrystalline anisotropy, with only the single term K_1 is sufficient to describe the behavior in CrO_2. In fact higher order anisotropy constants need to be taken into account for a complete quantitative description of the magnetic anisotropy.

The evolution of the transverse susceptibility peaks shows an interesting trend as the temperature is lowered below 300 K, and reveals yet another aspect of the magnetic anisotropy in these films. Fig. 2(a) shows the field dependent χ_T data, while the bias field is swept from negative to positive saturation, for four different temperatures in the geometry where H is perpendicular to the easy axis. The normalized curves are relatively shifted by a constant value along the vertical axis for clarity. While the peaks at $\pm H_K$ remain relatively unchanged, a very sharp peak just before $+H_K$ shows up and rapidly increases in height at lower temperatures.

Since the experimental configuration remains the same at lower temperatures as it is at 300 K, the origin of this peak must lie in the magnetic response intrinsic to the thin film samples. We can reproduce the evolution of the peak structure using the coherent rotation model by introducing a misalignment in angle between H and the easy axis in the perpendicular geometry. This trend is clearly illustrated in Fig. 2(b) that shows a series of calculated transverse susceptibility curves for several angles deviating from $\psi = 90°$ case considered earlier.

The qualitatively similarity between the measured and calculated χ_T is quite striking and suggests that as the temperature is lowered a gradual angular misalignment develops between the applied static field and the magnetic easy axis in the sample. This effect can be accounted for by considering the influence of strain on the thin films. As noted earlier, the lattice mismatch between the CrO_2 films and TiO_2 substrates introduces an in-plane tensile strain. The angular misalignment as the sample temperature is lowered could be associated with changing strain. Introducing this angular deviation between H and the easy axis is equivalent to including a net magnetoelastic energy term in the

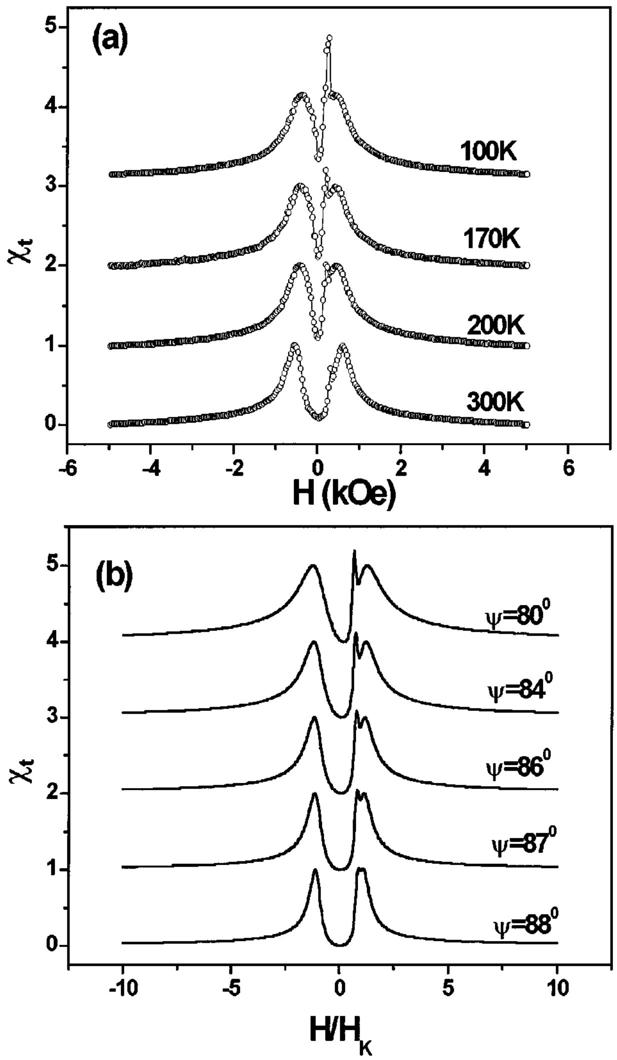

Fig. 2. (a) Measured χ_T, in perpendicular geometry ($\psi = 90°$) at different temperatures for unidirectional field ramp, negative to positive saturation. (b) Calculated χ_T for small angular deviations between H and the easy axis. Curves in (a) and (b) are normalized, and relatively shifted for clarity.

total anisotropy energy as is shown in [7]. Consequently, the result of combining two uniaxial anisotropies (for example a magnetocrystalline anisotropy described by the anisotropy field H_K and a magnetoelastic anisotropy described by the stress field $H_\sigma = 3\lambda_s\sigma/M_s$, where λ_s is the magnetostriction coefficient and σ is the applied stress) acting along two directions with an angle δ between them is also uniaxial in nature. The direction of the resultant uniaxial anisotropy makes an angle θ^* with the direction of the crystalline uniaxial anisotropy, which depends on H_K, H_σ and δ, as follows:

$$\tan 2\theta^* = H_\sigma \sin 2\delta/(H_K + H_\sigma \cos 2\delta). \qquad (1)$$

Assuming a magnetostriction coefficient, $\lambda_s = 5 \times 10^{-6}$ from [3] and including the values of M_s and H_K from our measurements, we can estimate the stress required to cause the angular deviation of $\theta^* = 10°$ shown in Fig. 2(b) using equation (1). For example, for $\delta = 45°$, this gives us a value of $\sigma = 0.9 \times 10^{10}$ dynes/cm^2, which is smaller than the estimated in-plane stress values along the c and b axis due to lattice mismatch. It has been reported that as the film thickness is reduced to <700 Å, the in-plane strain causes switching of the easy axis from the c direction to the b direction. Our transverse susceptibility results and analyses indicate that this tendency for the easy axis to gradually deviate by a small angular orientation from c axis toward the b axis in plane is evident even for 2000-Å-thick films.

It must be noted that temperature dependence of χ_T for the other orientations of the static field ($\psi = 0°$ and $45°$ also contains information about the influence of stress on the switching behavior. However, this effect is not as prominent as in perpendicular geometry, which is the most sensitive to H orientation.

In conclusion, our resonant RF experiments identify several important features in the magnetic anisotropy and switching behavior of epitaxially grown CrO_2 thin films.

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