Uniaxial anisotropy and switching behavior in epitaxial CrO_2 films

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Epitaxial *a*-axis oriented CrO_2 films with a strong in-plane uniaxial anisotropy exhibit easy-axis switching at small fields and coherent rotation of magnetization at larger fields. Unique angular dependence of $|\cos\phi|$ and $|\sec\phi|$ have been observed for remanence and switching field, respectively. © *2000 American Institute of Physics.* [S0003-6951(00)05228-1]

Metallic ferromagnets with substantial spin polarization are of importance in magnetoelectronic devices. Half-metallic ferromagnet, with only one spin band at the Fermi level, has the highest spin polarization.[1] Chromium dioxide (CrO_2) has the highest measured spin polarization (93%–100%) of all materials to date.[2–4] Together with a Curie temperature about 390 K, CrO_2 provides excellent prospects for magnetoelectronic devices at room temperature.

These attribute notwithstanding, CrO_2 is a metastable compound, which irreversibly decomposes at about 200 °C.[5] Hence, the investigation of the magnetic properties of CrO_2 has previously been limited largely to powders, with which essential magnetic properties, such as magnetic anisotropy and switching behavior, cannot be addressed. In this work, we report the observation of in-plane uniaxial magnetocrystalline anisotropy in *a*-axis oriented epitaxial CrO_2 thin films made by chemical vapor deposition. Due to the strong uniaxial anisotropy intrinsic to the crystal structure, we have observed very simple angular dependence of $|\cos\phi|$ for the remnance and $|\sec\phi|$ for the switching field. The hysteresis loops at all angles can be quantitatively reproduced using a simple model with uniaxial anisotropy.

The crystal structure of CrO_2 is tetragonal with lattice parameters of $a=b=4.419$ Å and $c=2.915$ Å. Epitaxial thin films of (100) CrO_2 have been fabricated by chemical vapor deposition at atmospheric oxygen pressure onto (100)TiO_2 single-crystal substrates, as described elsewhere.[6] The CrO_2 samples are 5×5 mm^2 in size and about 5000 Å in thickness. The $\theta/2\theta$ diffraction scan of a CrO_2 film shows only the (200) and (400) peaks indicating that it is an *a*-axis oriented film. The high quality of the sample is shown by the narrow width of 0.066° in the rocking curve of the (200) peak [Fig. 1(b)]. The epitaxial nature of the CrO_2 film has been established by the pole-figure scans [Figs. 1(c) and 1(d)], where the (220) and (202) spots observed at tilt angles of $\Psi=45°$ and 56.6° have the expected two-fold symmetry. X-ray diffraction conclusively demonstrates that the (100) CrO_2 film is epitaxial to the (100)TiO_2 substrate with the *b* and *c* axes in the film plane.

We have used vibrating sample magnetometry (VSM) to determine the easy axis of magnetization, the magnetocrystalline anisotropy energy, and the angular dependence of the magnetic characteristics at 300 K. The magnetic field **H** has been applied in the film plane at various angles ϕ with respect to the *c* axis, and the magnetization *M* has been measured along the **H** direction. The angular dependence of the hysteresis loops is shown in Fig. 2. As shown in Fig. 2(a), the easy axis of magnetization is the *c* axis with a square hysteresis loop and a saturation magnetization M_s of 655 emu/cm^3. Along the easy axis, the magnetization reverses sharply at a small switching field of $H_s(0)=22$ Oe. Along the *b* axis at $\phi=90°$, a hard-axis loop is observed as shown in Fig. 2(f). The uniaxial anisotropy along the *c* axis is the result of the in-plane two-fold crystal symmetry of the *a*-axis oriented CrO_2 films.

As ϕ is increased from 0° to 90°, the remnant magnetization M_r progressively decreases from M_s to zero. The angular dependence of squareness M_r/M_s is shown in Fig. 3(a)

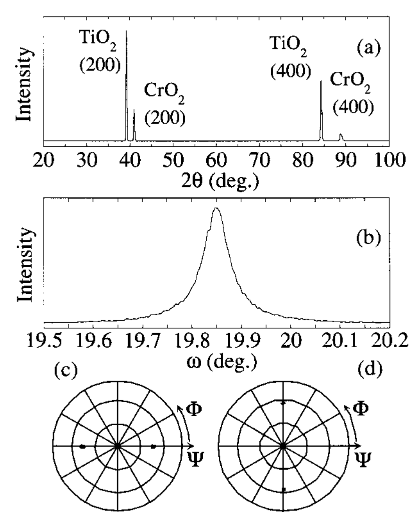

FIG. 1. X-ray diffraction patterns of a 5000-Å-thick *a*-axis CrO_2 film on a TiO_2(100) substrate: (a) $\theta/2\theta$ scan, (b) rocking curve of (200) peak with a full width at half maxima of 0.066°, pole-figure measurements of (c) (220) and (d) (202) peaks, showing two-fold symmetry at tilt angle $\Psi=45°$ and 56.6°, respectively.

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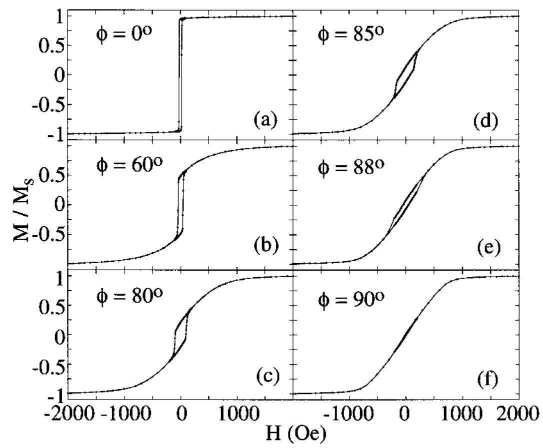

FIG. 2. Hysteresis loops at $T=300$ K of a 5000-Å-thick single-crystal *a*-axis CrO_2 film with magnetic field at different angles of ϕ of (a) 0°, (b) 60°, (c) 80°, (d) 85°, (e) 88°, and (f) 90° from the *c* axis.

as the open circles, which are very well described by $|\cos\phi|$ (solid curve). This simple angular dependence is due to the uniaxial magnetocrystalline anisotropy along the *c* axis. Regardless of ϕ, the magnetization $\mathbf{M}_s$ is always along the *c* axis at $H=0$. Since the VSM measures the magnetization component along **H** at an angle ϕ with respect to the *c* axis, the measured remnant magnetization is $M_r=M_s\cos\phi$. Thus, $M_r/M_s=|\cos\phi|$ is observed.

The free energy density of a ferromagnet with uniaxial anisotropy is

$$E=K_0+K_1\sin^2\theta+K_2\sin^4\theta+HM_s\cos(\phi-\theta), \qquad (1)$$

where θ is the angle between $\mathbf{M}_s$ and the easy axis, and K_0, K_1, and K_2 are anisotropy energy constants. The last term is the Zeeman term due to the magnetic field **H** applied at an angle of ϕ with respect to easy axis. Although usually only the K_1 term is included, we have found that both the K_1 and the K_2 terms are needed for CrO_2. The values of $K_0=1.5\times10^4$ erg/cm^3, $K_1=2.7\times10^5$ erg/cm^3, and $K_2=7.5\times10^4$ erg/cm^3 have been determined by the area method using the areas between the magnetization curves and the *M* axis at three different angles (e.g., $\phi=0°$, 45°, and 90°).[7] The strength of the uniaxial anisotropy field H_K is given by $2K_1/M_s$,[7] for which the value is 830 Oe. It is noted in CrO_2 that K_0 is an order of magnitude smaller than K_1.

Using the free energy in Eq. (1) the magnetizing curve (*M* vs *H*) can be obtained from two equations, one from the condition of $\partial E/\partial\theta=0$, which leads to

$$H=\frac{2K_1\sin\theta\cos\theta+4K_2\sin^3\theta\cos\theta}{M_s\sin(\phi-\theta)}, \qquad (2)$$

and the other is

$$M=M_s\cos(\phi-\theta), \qquad (3)$$

the magnetization measured along **H**. The magnetization curve given by Eqs. (2) and (3) describes coherent rotation of **M** as a single domain. Since the epitaxial CrO_2 film is macroscopic and not a single-domain particle of submicron size, coherent rotation of **M** will be taken over by other processes at the switching field.

For increasing ϕ, the switching field $H_s(\phi)$ of epitaxial CrO_2 *increases* monotonically from $H_s(0)=22$ Oe to very large values as shown in Fig. 2. We note the distinction between the switching field H_s, at which **M** switches, and the coercive field H_c, at which $M=0$. For $\phi\lesssim80°$, $H_s=H_c$. However, for $\phi\gtrsim80°$ [e.g., $\phi=85°$ and 88° shown in Figs. 2(d) and 2(e)], *M* crosses zero before switching occurs at H_s, and hence, $H_c<H_s$. The coercive field thus has the unusual apparent behavior of first increasing with ϕ($\phi\lesssim80°$), then decreasing with ϕ($\phi\gtrsim80°$), and vanishing at 90°. It should be noted that the angular dependence of the hysteresis loops shown in Fig. 2 is very different from those usually observed in ferromagnetic materials, for which the largest H_c occurs when **H** is along the easy axis.[7] In contrast, in epitaxial CrO_2 films, the *smallest* H_s is observed along the easy axis, and the value of H_s increases unabatedly for increasing ϕ. The observed angular dependence of the switching field $H_s(\phi)$ of CrO_2 is shown in Fig. 3(b). The solid curve, which is in excellent agreement with the data, is $H_s(\phi)=H_s(0)|\sec\phi|$ with $H_s(0)=22$ Oe. The unusual angular dependence of H_s is, first of all, due to the fact that $\mathbf{M}_s$ is essentially along the *c* axis at small fields, as manifested in the observation of $M_r=M_s\cos\phi$. The $|\sec\phi|$ dependence indicates that switching occurs when the projection of **H** onto the *c* axis reaches the critical value of $H_s(0)=22$ Oe, i.e., $H_s(\phi)\cos\phi=H_s(0)$.

The unusual angular dependence of $H_s(\phi)=H_s(0)|\sec\phi|$ shows that switching of magnetization at H_s consists of domain reversal along the *c* axis. Indeed, this type of domain reversal has been predicted to display the $|\sec\phi|$ angular dependence.[8] Experimental observation of $|\sec\phi|$ dependence of H_s requires single crystals with intrinsic uniaxial anisotropy. Previously, the angular dependence of $|\sec\phi|$ has been observed over a large range of ϕ in single crystals (e.g., orthoferrites).[9,10] In magnetic thin films, the angular dependence of H_s of $|\sec\phi|$ is generally not observed. In rare cases, it has been observed but only for small values of ϕ in the range of $1\leqslant H_s(\phi)/H_s(0)\leqslant1.4$.[11] In epitaxial CrO_2 film, it is remarkable that, $H_s(\phi)=H_s(0)|\sec\phi|$ has been observed for essentially all values of ϕ with $1\leqslant H_s(\phi)/H_s(0)\leqslant13$. This unique angular dependence further attests to the high quality of the thin films.

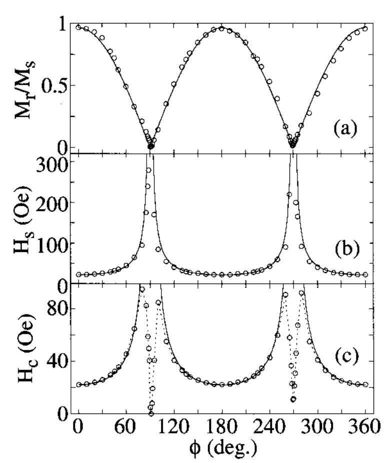

FIG. 3. Angular dependence of (a) squareness (M_r/M_s), (b) switching field H_s, and (c) coercivity H_c. The open circles are experimental data and the solid lines are $|\cos\phi|$ in (a), $|\sec\phi|$ in (b) and (c).

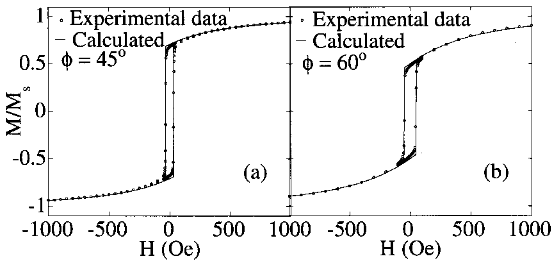

FIG. 4. Calculated hysteresis loops (solid lines) at (a) $\phi=45°$ and (b) $\phi=60°$ and the experimental data (open circles) of a 5000-Å-thick *a*-axis CrO_2 film.

The constant K_0 in Eq. (1) is $M_sH_s(0)$, which is 1/4 of the loop area in Fig. 2(a) for $\phi=0$, thus a measure of the energy loss of the hysteresis loop. As shown in Fig. 2, for $\phi\neq 0$, the loop is approximately parallelogram in shape with an area of $4M_rH_s$. Because of the unique angular dependence of M_r and H_s of $|\cos\phi|$ and $|\sec\phi|$, respectively, $M_rH_s(\phi)=M_sH_s(0)$, i.e., the loop area is independent of ϕ. Since the loop area is the result of the energy loss due to domain reversal, its constancy also indicates that switching is accomplished by the same process, namely, reversal of domains along the *c* axis.

In most ferromagnets, the magnetization-reversal mechanisms are much more complex and the hysteresis loops can not be easily reproduced theoretically. In the present epitaxial CrO_2 films with uniaxial anisotropy, together with the switching field of $H_s(\phi)=H_s(0)|\sec\phi|$, hysteresis loops at all angles can be numerically calculated using Eqs. (2) and (3). As illustrated in Fig. 4 for the hysteresis loops for $\phi=45°$ and 60°, the calculated results are in excellent agreement with the experimental data except near the switching fields, where slight deviations occur. The excellent quantitative agreement shows that the epitaxial CrO_2 film behaves as a macroscopic single domain that rotates coherently at all fields and angles except near the switching fields.

The unusual magnetic characteristics observed in the CrO_2 film stems from the strong in-plane uniaxial magnetocrystalline anisotropy, which is intrinsic to the in-plane crystal structure of the epitaxial film. As a result of $K_1 \gg K_0$ in the CrO_2 films, the ratio of $H_K/H_s(0)=38$ is much larger than those for other ferromagnetic films with small switching field, such as permalloy films, which generally show ratios of $H_K/H_s(0)$ of order 1.[8]

In summary, epitaxial *a*-axis oriented CrO_2 films exhibit in-plane uniaxial magnetocrystalline anisotropy intrinsic to the crystal structure. It displays coherent rotation of magnetization at all fields and angles except near the switching fields. The remanence M_r shows a $|\cos\phi|$ dependence due to the uniaxial anisotropy along the *c* axis. Switching occurs along the *c* axis and the switching field exhibits a classic $|\sec\phi|$ dependence due to domain reversal along the uniaxial axis. The calculated hysteresis loops with uniaxial anisotropy constants $K_1=2.7\times 10^5$ erg/cm^3 and $K_2=7.5\times 10^4$ erg/cm^3 are in good agreement with the experimental results.

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