Critical behavior of epitaxial half-metallic ferromagnetic CrO_2 films

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Critical behavior of the epitaxial CrO_2 film, a half metallic ferromagnet, has been determined by magnetometry with the magnetic field along the uniaxial anisotropy axis. The critical temperature has been determined to be $T_C = 386.50 \pm 0.05$ K. The critical exponents of $\beta = 0.371 \pm 0.005$ and $\gamma = 1.43 \pm 0.01$ indicate that CrO_2 is a Heisenberg ferromagnet.

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I. INTRODUCTION

Chromium dioxide (CrO_2) has recently attracted a great deal of attention because it is a half-metallic ferromagnet with only one spin band at the Fermi level. Spin polarizations of 95 and 90%, the highest among all materials, have been reported using spin-polarized photoemission[1] and point-contact Andreev reflection[2] measurements, respectively. However, because CrO_2 is a metastable compound, much of its intrinsic properties remain not well known. Previously, only powder samples of CrO_2, synthesized under a high oxygen pressure, were available. Recently, epitaxial thin films of CrO_2 have been successfully grown by chemical vapor deposition at atmospheric oxygen pressure, the details of which have been described elsewhere.[3] The availability of epitaxially grown single-crystal CrO_2 films provides the necessary medium for the studies of its intrinsic properties. It is particularly interesting to determine the critical behavior of this unusual ferromagnet in view of its half metallic nature. In this work, we report the critical behavior of epitaxial CrO_2 films grown on single-crystal TiO_2 (100) substrate. The critical exponents of β, γ, as well as the critical temperature T_C have been determined. The results indicate that this half metallic ferromagnet is a Heisenberg ferromagnet.

II. EXPERIMENTAL RESULTS AND ANALYSIS

CrO_2 has a rutile crystal structure with a tetragonal unit cell of lattice parameters $a = b = 4.419$ Å and $c = 2.915$ Å. Epitaxial CrO_2 films can be grown onto isostructural TiO_2 single-crystal substrates. The CrO_2 samples used in this work are *a*-axis CrO_2 films of dimensions 5 mm × 5 mm × 5000 Å. The $\theta/2\theta$ scan of the CrO_2 film grown on TiO_2 (100) substrate shows only the (200) and (400) peaks of CrO_2 and TiO_2.[4] The rocking curve of CrO_2 (200) peak gives a full width at half maximum (FWHM) of only 0.066°, demonstrating the high quality of the CrO_2 film. As a comparison, the rocking curve of the TiO_2 (200) peak of the underlying single-crystal substrate shows a FWHM of 0.034°.

The single-crystal nature of the CrO_2 films has been revealed by the pole-figure measurements. The results establish that the sample is a single-crystal *a*-axis CrO_2 film epitaxially grown on an *a*-axis TiO_2 substrate.[4]

Magnetometry measurements have shown that there is an intrinsic in-plane uniaxial anisotropy in the CrO_2 film along the *c* axis, which is in the film plane for the *a*-axis film, with an anisotropy field in excess of 800 Oe at room temperature.[4] Switching occurs only along the *c* axis with a small switching field of 22 Oe. Furthermore, for an external field **H** applied at an angle θ with respect to the easy axis, only the component along the easy axis contributes to switching. It is therefore essential that the external magnetic field be applied along the *c* axis for the determination of the critical exponents using magnetometry.

We have measured the temperature dependence of magnetization from 370 to 400 K at different magnetic fields in the vicinity of the Curie temperature T_C in a superconducting quantum interference device (SQUID) magnetometry. During the measurements, the magnetic field **H** was applied parallel to the easy axis (*c* axis) in the film. The demagnetizing field is negligible because of the thin-film geometry.

Representative magnetization curves of M vs T curves at fields from 100 Oe to 10 kOe are shown in Fig. 1(a). At T close to T_C, the initial susceptibility χ_0 and the spontaneous magnetization M_s have the asymptotic relations of[6]

$$\lim_{H\to 0} \frac{H}{M} = \chi_0^{-1} \propto (T - T_C)^{\gamma}, \quad T > T_C, \tag{1}$$

$$\lim_{H\to 0} M = M_s \propto (T_C - T)^{\beta}, \quad T < T_C, \tag{2}$$

where γ and β are the critical exponents for χ_0 and M_s, respectively.

In magnetometry measurements, because of the requirement of an external field, M and not M_s is measured. The values of M_s and χ_0^{-1}, both are defined at $H = 0$, can be obtained by extrapolation to $H = 0$ with the use of the Arrott plots of $M^{1/\beta}$ vs $(H/M)^{1/\gamma}$, which, at T close to T_C, are

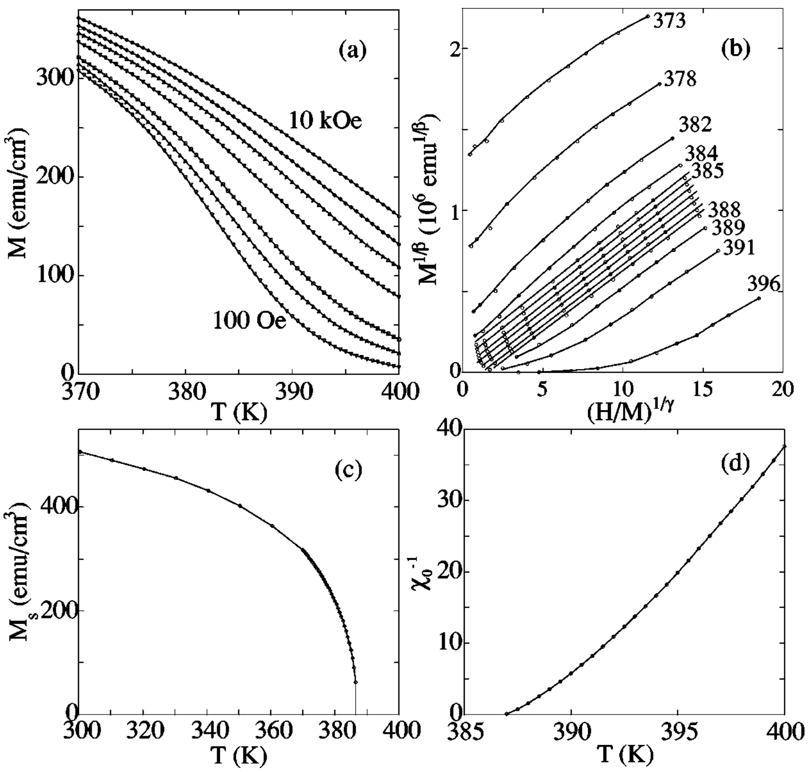

FIG. 1. (a) The M vs T curves at $H=100$, 500, 1000, 3000, 5000, 7000, and 10 000 Oe. (b) The Arrott plot, $M^{1/\beta}$ vs $(H/M)^{1/\gamma}$, for a CrO_2 film near T_C ($373<T<396$ K). (c) The spontaneous magnetization M_s and (d) the inverse initial susceptibility χ_0^{-1} deduced from (b).

straight lines.[7] As shown in Eqs. (1) and (2), the critical temperature T_C can be obtained from plots of $(d\ln\chi_0^{-1}/dT)^{-1}$ vs T and $(d\ln M_s/dT)^{-1}$ vs T, both of which are straight lines near T_C. The critical exponents β and γ can be determined from the plots of $\log M_s$ vs $\log[1-T/T_C]$ and $\log\chi_0^{-1}$ vs $\log[T/T_C-1]$, respectively. Since the values of T_C, β, and γ are not known *a priori*, we have determined these values by iterations. In each iteration, the starting values of β and γ in the $M^{1/\beta}$ vs $(H/M)^{1/\gamma}$ plot result in the slightly different final values of β and γ from the $\log M_s$ vs $\log[1-T/T_C]$ and $\log\chi_0^{-1}$ vs $\log[T/T_C-1]$ plots. The iteration continues until the differences between the starting and final values of β and γ are negligible.

In Fig. 1(b), we show the $M^{1/\beta}$ vs $(H/M)^{1/\gamma}$ plot for the CrO_2 film at constant temperatures near T_C with $\beta=0.371$ and $\gamma=1.43$. The value of M_s and χ_0^{-1}, obtained by linear extrapolation to $H=0$, are shown in Figs. 1(c) and 2(d). As shown in Figs. 2(a) and (b), the plots of $(d\ln\chi_0^{-1}/dT)^{-1}$ vs T and $(d\ln M_s/dT)^{-1}$ vs T, both are straight lines near T_C. We have determined $T_C=386.50\pm0.05$ K from Fig. 3(a) and $T_C=386.51\pm0.05$ K from Fig. 2(b). The critical exponents of $\beta=0.371\pm0.005$ and $\gamma=1.43\pm0.01$ have been obtained from the plots of $\log M_s$ versus $\log[1-T/T_C]$ and $\log\chi_0^{-1}$ versus $\log[T/T_C-1]$, as shown in Fig. 3. Using the scaling relation of $\beta+\gamma=\beta\delta$,[6] we have determined the value of the critical exponent $\delta=4.85$ for the critical isotherm. Previously, Kouvel and Rodbell,[8] using Arrott plots of M^2 vs H/M, have found larger values of the critical exponents of $\gamma\approx1.6$ and $\delta\approx5.8$ using CrO_2 powder. However, the value of $T_C=386.50\pm0.05$ K determined from this work using epitaxial CrO_2 films agrees excellently with the value of $T_C=386.5$ K determined from powder samples.[8]

The most stringent test of the validity of the critical exponents is the scaling equation.[6] For a second-order phase transition, the scaling equation is

$$M/|T-T_C|^\beta=f[H/|T-T_C|^{\beta\delta}]. \quad (3)$$

It follows that if the exponents are accurately determined, in the plots $\ln M/|T-T_C|^\beta$ vs $\ln H/|T-T_C|^{\beta\delta}$, all the data near T_C should fall onto two curves, one for $T>T_C$ and the other for $T<T_C$. As shown in Fig. 4, this is indeed the case for the results of the epitaxial CrO_2 film.

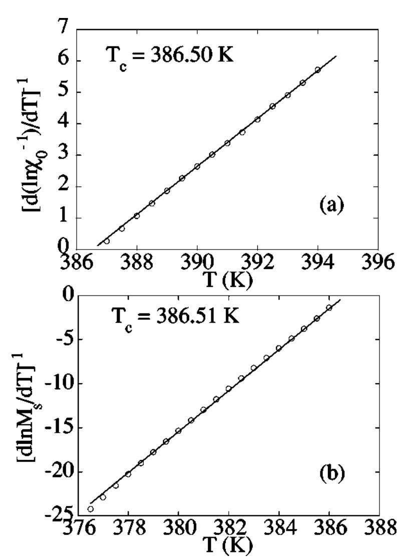

FIG. 2. Plots of (a) $(d\ln\chi_0^{-1}/dT)^{-1}$ vs T and (b) $(d\ln M_s/dT)^{-1}$ vs T of a CrO_2 film for the determination of T_C.

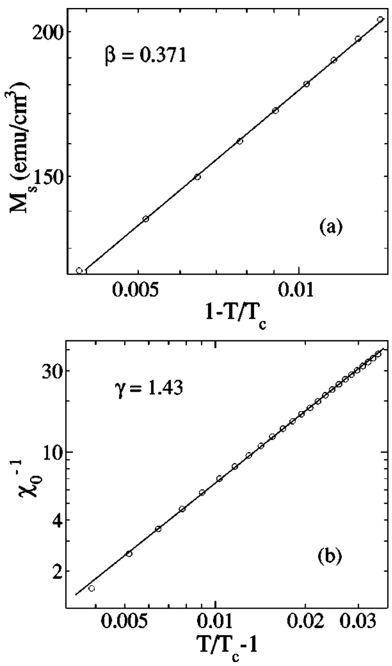

FIG. 3. Plots of (a) $\log M_s$ vs $\log[1-T/T_C]$ and (b) $\log \chi_0^{-1}$ vs $\log[T/T_C-1]$ for the determination of the critical exponents of $\beta = 0.371$ and $\gamma = 1.43$.

There are theoretical predictions of the values of the critical exponents, some of which are shown in Table I.[6] For three-dimensional Heisenberg model with short-range interactions, the values are $\beta = 0.365$ and $\gamma = 1.386$. For three-dimensional Ising model with short-range interactions, the values are $\beta = 0.325$ and $\gamma = 1.241$. For mean-field theory, the values are $\beta = 0.5$ and $\gamma = 1$. Our results of $\beta = 0.371$, $\gamma = 1.43$ are close to those of the three-dimensional (3D) Heisenberg model.

Since the epitaxial CrO_2 films have in-plane uniaxial magnetocrystalline anisotropy,[4] we comment on the influence of anisotropy on the critical exponents. The 3D Heisenberg exchange interactions are purely isotropic, i.e., the exchange integral $J(\mathbf{r})$ is the same in all directions. But most real magnetic materials have magnetic anisotropy, such as the uniaxial anisotropy in the epitaxial CrO_2 films, in which $|J_\parallel| > |J_\perp|$, where $|J_\parallel|$ and $|J_\perp|$ are the exchange integrals along and perpendicular to the anisotropy axis, respectively. The extreme anisotropic limit $J_\perp = 0$ corresponds to the Ising model.[6] In our recent work,[4] we have determined the anisotropy constant in CrO_2 to be $K_1 = 2.7 \times 10^5$ ergs/cm^3, which is comparable to the values of $K_1 = 4.8 \times 10^5$ ergs/cm^3 for Fe and $K_1 = -0.5 \times 10^5$ ergs/cm^3 for Ni. Since the anisotropy energy decreases very rapidly as the temperature is raised toward T_C, the measurements of critical exponents are not affected by the anisotropy in the critical region, which is known in Fe and Ni[5]. We conclude that the magnetocrystalline anisotropy is unlikely to affect measurably the critical exponents in CrO_2. However, since CrO_2 is a uniaxial ferromagnet, it is essential that the magnetic field be applied along the easy direction.

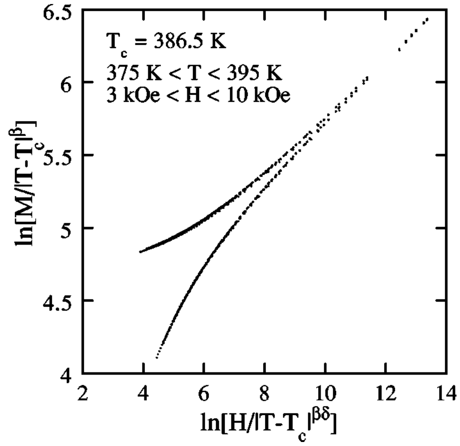

FIG. 4. Scaling plot of $\ln M/|T-T_C|^\beta$ vs $\ln H/|T-T_C|^{\beta\delta}$ shows that all the data near T_C fall onto two curves, one for $T > T_C$ and the other for $T < T_C$.

In addition to β and γ, there are other critical exponents associated with specific heat (α), critical isotherm (δ), and coherence length (ν). With the two exponents determined, the other exponents can be obtained by the scaling laws of $\beta + \gamma = \beta\delta, \alpha + 2\beta + \gamma = 2$, and $\nu d = 2 - \alpha$.[6] With the values of β and γ obtained for CrO_2, we have obtained $\delta = 4.85, \alpha = -0.09$, and $\nu = 0.70$. As shown in Table I, all the critical exponents for CrO_2 are close to those predicted for 3D Heisenberg interactions. Thus we concluded that, despite its unusual band structure, CrO_2 is a three-dimensional Heisenberg ferromagnet.

III. CONCLUSION

In summary, we have investigated the critical behavior in epitaxial CrO_2 films. Critical exponents were determined

TABLE I. Critical exponents for specific heat (α), order parameter (β), susceptibility (γ), critical isotherm (δ), and coherent length (ν) predicted by mean field, 3D Ising, and 3D Heisenberg models, and those measured from epitaxial CrO_2. The values of α, ν, and δ for CrO_2 have been calculated from the values of β and γ.

	Specific heat α	Order parameter β	Susceptibility γ	Critical isotherm δ	Coherence length ν
Mean field	0	0.5	1	3	0.5
3D Ising	0.11	0.32	1.24	4.82	0.63
3D Heisenberg	-0.12	0.36	1.39	4.80	0.71
CrO_2	-0.09	0.371	1.43	4.85	0.70

from the temperature and field dependence of magnetization with the values of $\beta = 0.371 \pm 0.005, \gamma = 1.43 \pm 0.01$. The critical temperature T_C has been determined to be 386.50 $\pm$ 0.05 K. These critical exponents indicate that the half-metallic CrO_2 is a 3D Heisenberg ferromagnet.

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