Finite-Size Effects and Uncompensated Magnetization in Thin Antiferromagnetic CoO Layers

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Finite-size effects in CoO, an antiferromagnetic insulator with localized moments, have been observed in CoO/SiO_2 multilayers. The Néel temperatures of the CoO layers, as determined by dc susceptibility measurements, show a finite-size scaling relation with a shift exponent $\lambda = 1.55 \pm 0.05$. Increased magnetization with an oscillation in magnitude, due to uncompensated antiferromagnetic layers, has also been observed.

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When one or more dimensions of a bulk solid is reduced to near or below a certain characteristic length scale, the associated properties are modified reflecting the lower dimensionality. Thin layers are ideal media for the studies of finite-size effects related to the approach to two dimensions. The correlation length in the lateral directions are unimpeded in thin layers, but terminated by the layer thickness in the third direction [1]. For systems with long correlation lengths, such as traditional superconductors having coherence lengths of the order of hundreds of Å, finite-size effects can be observed in comparatively thicker films [2]. The reduction of the superconducting transition temperature due to finite-size effects and, correspondingly, the dimensional crossover have been observed in numerous systems [2,3]. In strongly correlated systems with short correlation lengths, such as ferromagnets, an appreciable reduction of the Curie temperature (T_C) occurs only in ultrathin films [4,5]. Furthermore, the high values of T_C in most elemental ferromagnets (e.g., Co and Fe with $T_C > 1000$ K) effectively prevent the determination of T_C at such high temperatures without first degrading or destroying the ultrathin films. The few reports of the reduction of T_C in Co and Fe involve ultrathin films a few monolayers thick with $T_C < 400$ K [4,5]. These experiments, difficult as they are, preclude the finite-size scaling analyses, which are the results of critical behavior in the vicinity of bulk Curie temperature [$T_C(\infty)$]. Most detailed analyses of finite-size effects in ferromagnets have been made in Gd films with a convenient bulk $T_C(\infty)$ near room temperature [6]. Even in spin glasses, endowed with a longer correlation length, finite-size effects and dimensional crossover have already been observed [7,8].

Conspicuously lacking is the observation of finite-size effects in antiferromagnets with localized moments. Finite-size effects in antiferromagnets are intrinsically more interesting because the occurrence of antiferromagnetic (AF) order is made possible by the cancellation of two or more ferromagnetic sublattices resulting in zero net magnetization. In addition to the requirements of thin layers, the observation of finite-size effects in AF materials presents a further challenge because of the zero net magnetization. Under an external field, the response of an AF material is much weaker than that of a ferromagnet. The only and very recent report of finite-size effects in an AF material has been through the indirect measurement of resistivity in thin layers of Cr, which is an incommensurate spin-density-wave AF metal [9]. Since the results have been observed in Fe/Cr multilayers, the interlayer coupling through the Cr layers and the ferromagnetic ordering of the Fe layers complicate the analyses of the finite-size scaling of Cr layers.

In ultrafine AF particles, unusual magnetic properties may result from uncompensated moments, as first suggested by Néel [10], and weak ferromagnetic characteristics have been observed in ultrafine AF particles of about 22 Å [11]. For an AF ultrathin layer uncompensated moments may also be observable. The uncompensated magnetization can be particularly large for AF with alternating ferromagnetic planes. In this work, we report on the first observation of finite-size effects in a magnetically isolated AF insulator with localized moments via direct susceptibility measurements. We have also observed uncompensated net magnetization in very thin AF layers.

For the studies of finite-size effects, we have chosen CoO, a well-known AF insulator with a localized moment of $3.8\mu_B$ residing on the Co^{2+} sites [12]. It has a simple fcc NaCl structure with a lattice parameter of $a_0 = 4.26$ Å. Bulk CoO has an ideal Néel temperature (T_N) near room temperature. Any appreciable reduction of T_N can be readily measured without risking the integrity of the thin layers. Because of the intrinsically weak response to an external magnetic field, CoO samples in a multilayer form are imperative. To prevent possible interlayer coupling through metallic intervening layers, nonmetallic layers will be necessary. These demands have been satisfied by the successful fabrication of CoO/SiO_2 multilayers by RF sputtering.

Sputtering has been administered in an Ar atmosphere of 6 mTorr, at an RF power of 75 W, resulting in deposition rates of 2.8 and 1.5 Å/s for CoO and SiO_2, respectively. The (100) Si substrates have been mounted on a computer-controlled rotating platform, which sequentially moves the substrate to the deposition sources for

prescribed amounts of time. Each individual sample was cut from a multilayer consisting of 100 bilayers of wedged films of CoO varying in thickness from 5 to 125 Å, interleaved with a fixed SiO_2 layer of thickness either 50 or 75 Å. For all the samples measured, the thickness of the CoO is the *only* varying parameter. It turns out that both 50 and 75 Å of SiO_2 are more than sufficient to isolate the AF layers, and there is no difference between the samples with 50 or 75 Å of SiO_2.

In Fig. 1, low-angle x-ray diffraction results of representative CoO/SiO_2 multilayers are shown. The high quality of the multilayers are indicated by the large number of diffraction peaks. The bilayer thickness, determined from the peak separation, agrees very well with the designed value. High-angle x-ray diffraction shows that the CoO layers are predominantly (100) oriented, whereas the SiO_2 layers are amorphous. The excellent layer structure has also been confirmed by cross-sectional transmission electron microscopy. Susceptibility (χ) measurements have been performed on the samples by first cooling the samples in zero field to 5 K. A dc magnetic field (H) was then applied parallel to the film plane and the magnetization was measured in increasing temperatures. The response of AF layers to H is intrinsically small. Although a larger H would increase the signal and sharpen the peak in χ, it is at the expense of shifting the peak position to slightly lower temperatures for thicker CoO layers [13] and washing out the peak in χ for very thin CoO layers. Consequently, a measuring field of $H = 100$ Oe has been used. In Fig. 2, susceptibility measurement (χ vs T) of representative samples are shown with χ normalized to $\chi_{\max}(T)$ for clarity. As the thickness of the CoO layer is reduced, the value of T_N progressively decreases. Our results indicate that AF order ($T_N \geq 5$ K) persists down to a thickness of 10 Å.

Two methods have been used to determine the values of T_N: the peak in $\chi(T)$ [13] and the peak in $d(\chi T)/dT$ [14]. For each sample, the two values of T_N differ by only a few percent, with the value of the latter method always slightly lower. In Fig. 3(a), the values of the Néel temperature [$T_N(t)$], as determined from both methods described above, are shown as a function of the CoO layer thickness (t). These results have been analyzed in the context of finite-size scaling. Near the bulk Néel temperature [$T_N(\infty)$], the correlation length has a power law temperature dependence of

$$\xi(T) = \xi_0 \left[1 - \frac{T}{T_N(\infty)} \right]^{-\nu}, \qquad (1)$$

where ξ_0 is the extrapolated correlation length at $T = 0$ K, and ν is the critical exponent for the correlation length [1]. It follows then, the Néel temperature [$T_N(t)$] of a thin layer of thickness t varies as

$$\frac{T_N(\infty) - T_N(t)}{T_N(\infty)} = \left(\frac{\xi_0}{t} \right) \lambda, \qquad (2)$$

where λ is the shift exponent for the finite-size scaling, and $\lambda = 1/\nu$ [15,16]. Accordingly, there is a progressive reduction of T_N as experimentally observed. The solid curves in Fig. 3(a) are the calculated results of Eq. (2) obtained from data with $t \geq 22$ Å. If the values of $T_N(t)$ determined from the peak in $\chi(T)$ are used, the best fit values of $\lambda = 1.55 \pm 0.05$, $\xi_0 = 18 \pm 1$ Å, and $T_N(\infty) = 315 \pm 5$ K are obtained. If the values of $T_N(t)$ determined from the peak in $d(\chi T)/dT$ are used, the best fit values of $\lambda = 1.54 \pm 0.05$, $\xi_0 = 20 \pm 1$ Å, and $T_N(\infty) = 300 \pm 5$ K are obtained. Both analyses provide nearly identical values of λ and ξ_0. Although Eq. (2) is strictly valid only for $T_N(t)$ not far from $T_N(\infty)$, in reality, it adequately describes most of the data, and only data for $t < 20$ Å clearly deviate from the curves.

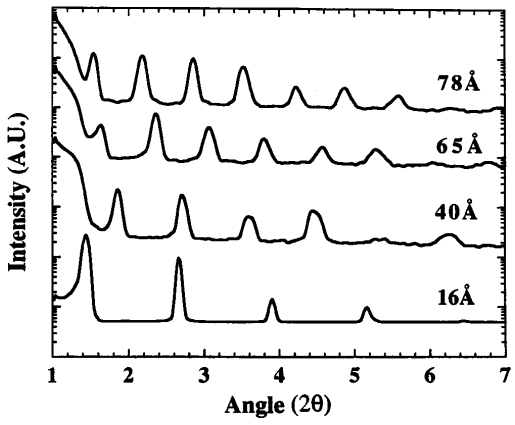

FIG. 1. Low-angle x-ray diffraction of multilayer films of CoO/SiO_2 with a fixed SiO_2 layer thickness of 50 Å. The CoO layer thickness of $t = 16$, 40, 65, and 78 Å are labeled.

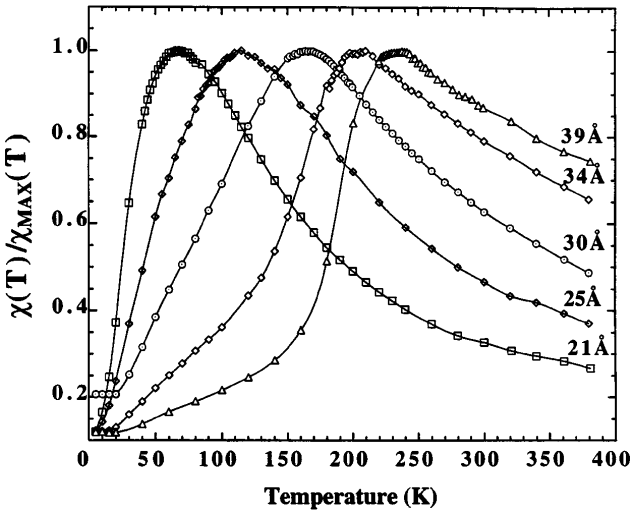

FIG. 2. Temperature dependence of dc susceptibility at $H = 100$ Oe of representative multilayer samples of CoO/SiO_2 with a fixed SiO_2 layer thickness of 50 Å and various CoO layer thicknesses of $t = 21$, 25, 30, 34, and 39 Å. For clarity, the results are normalized to the maximum susceptibility.

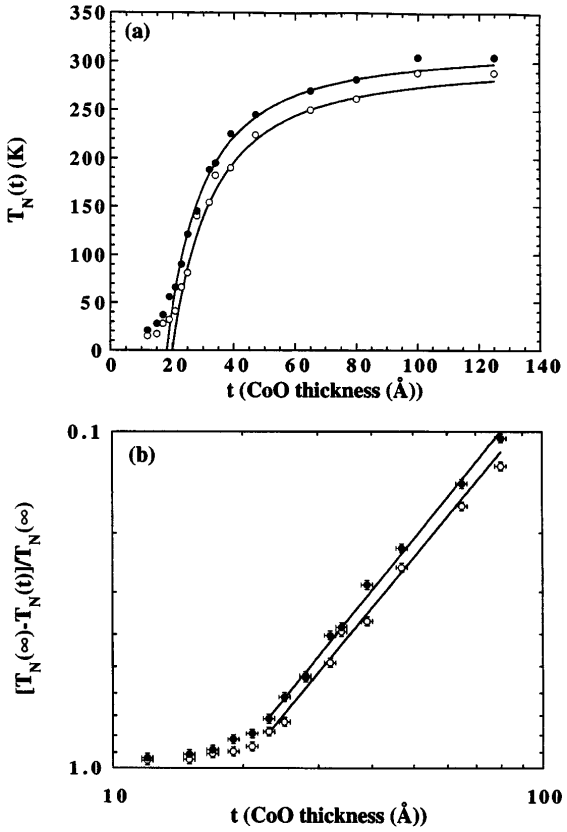

FIG. 3. (a) Néel temperature $T_N(t)$ of CoO/SiO_2 multilayers vs CoO thickness (t). The solid circles represent $T_N(t)$ determined from the peak in $\chi(T)$ and the open circles represent $T_N(t)$ determined from the peak in $d(\chi T)/dT$. The solid curves are the results of finite-size scaling relation with $\lambda = 1.55$, $\xi_0 = 18$ Å, and $T_N(\infty) = 315$ K (solid circles) and $\lambda = 1.54$, $\xi_0 = 20$ Å, and $T_N(\infty) = 300$ K (open circles). (b) Log-log plot of $[T_N(\infty) - T_N(t)]/T_N(\infty)$ vs CoO thickness t, where the straight lines have a slope of $\lambda = 1.55$ (solid circles) and $\lambda = 1.54$ (open circles).

To further illustrate the power law dependence, a log-log plot of $[T_N(\infty) - T_N(t)]/T_N(\infty)$ vs t is shown in Fig. 3(b), where the linear dependence with a slope of $\lambda = 1.55$ and 1.54, respectively, for $t \geq 22$ Å is evident. Theoretical studies of finite-size scaling indicate a shift exponent of $\lambda = 1.5584$ for the Ising systems [17], and $\lambda = 1.419$ for the Heisenberg systems [18]. Specific heat results indicate that CoO behaves more like an Ising than a Heisenberg system [19]. Our determined values of $\lambda = 1.55 \pm 0.05$ for CoO is also closer to the theoretical results for Ising systems than for the Heisenberg systems. As indicated in Fig. 3(a), the value of $T_N(t)$, extrapolated to $t \rightarrow 0$, is nearly zero. This is different from that in Fe/Cr multilayers, where the onset of antiferromagnetism occurs at $t_{Cr} = 42$ Å, below which no AF ordering has been found [9].

For a bulk AF material, the sublattice magnetizations are fully compensated resulting in zero net magnetization. The M vs H measurement of thick CoO layers at 5 K shows a linear behavior with no ferromagnetic characteristics as shown by the dashed line in Fig. 4. As first suggested by Néel [10], ferromagnetic characteristics may be observed in ultrafine AF particles [11], and by obvious extension, thin AF layers. For an AF ultrathin layer with alternating ferromagnetic planes uncompensated magnetization may be especially large. In the present case, ferromagnetic characteristics become progressively more evident for thinner CoO layers. An example for CoO layers with $t = 17$ Å is shown in Fig. 4, where ferromagnetic characteristics with a large remanence and a large coercivity ($H_C = 1.5$ kOe) are shown. Such strong ferromagnetic characteristics disappear at its $T_N = 36$ K. At T slightly higher than T_N, such as 50 K, remanence and coercivity have vanished and only the high susceptibility remains.

The antiferromagnetic ordering in CoO consists of alternating ferromagnetic planes. The susceptibility results of single crystals CoO indicate alternating ferromagnetic (100) planes along the [100] direction [20], whereas neutron diffraction shows ferromagnetic (111) planes alternating along the [111] direction [21,22]. The antiferromagnetic spin structure observed in a bulk specimen is likely to be altered in very thin CoO layers, especially due to the uncompensated magnetization, which is compelled to be in the film plane due to the shape isotropy. The magnetic results of our (100) oriented CoO/SiO_2 multilayers indicate (100) ferromagnetic planes, due to the observation of progressively larger magnetization for thinner CoO layers. The alternating ferromagnetic (100) planes are separated by $a_0/2 = 2.13$ Å. Within a simple model, in thin CoO layers, completely compensated moments would require an even number of (100) planes. Remnant magnetization (M_r) and spontaneous magnetization [$M_s(0)$] (see Fig. 4) may be expected for CoO layers with an *odd* number of (100) planes, hence at thicknesses of $t =$ (even number) $a_0/2$, i.e., $t = na_0$, where $n =$ interger. Given $3.8\mu_B$/Co and the lattice parameter of CoO, if all the Co moments were to order

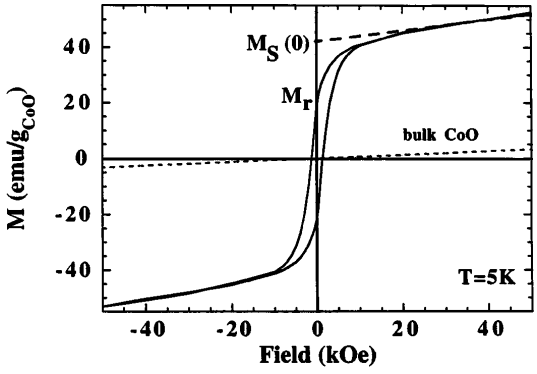

FIG. 4. Hysteresis loop at 5 K of a multilayer sample of [CoO(17 Å)/SiO_2(75 Å)] (solid curve), showing spontaneous magnetization [$(M_s(0)$] and remnant magnetization (M_r), and coercivity. The result for a single layer 500 Å CoO is shown as the dashed line.

ferromagnetically, a magnetization of $M_{CoO} = 238$ emu/g_{CoO} would be expected. The magnetization for CoO layers with an odd number of (100) planes would give a magnetization of $M_s(0) = [1/(2n + 1)M_{CoO}]$ for thickness $t = na_0$. Thus, as the CoO layer is reduced, both M_r and $M_s(0)$ should increase, and, furthermore, exhibit *oscillation* with a period of $a_0 = 4.26$ Å. The increased magnetization with oscillation cannot be a consequence of Co^{2+} vacancies [23] or other defect contributions [20], since the individual samples were cut from a wedge multilayer where the thickness of CoO is the only variable. Any contribution from defects would then be present in *all* samples.

The values of $M_s(0)$ and M_r (taken at $H = 0$) indeed increase with decreasing t as shown in Fig. 5. Equally significant, there are oscillations with increasing amplitude in both $M_s(0)$ and M_r as t is decreased. The measured $M_s(0)$ is close to $1/(2n + 1)M_{CoO}$. The oscillating nature of the magnetization is clear, although the limited number of data points may not have captured all the features. The available data suggest an oscillation period of about 5 Å, which is close to the expected 4.26 Å from the simple model. This slight discrepancy may be due to the less than ideal samples, or to the fact that the spacing between planes in ultrafine thin films could be different from that of bulk. Furthermore, the simple picture of alternating ferromagnetic planes outlined above may have to be modified in the thin layer limit where frustration effects are not negligible. Nevertheless, the oscillating M_s and M_r with increasing magnitude are the consequence of uncompensated moments of thin AF layers. The precise spin structure of thin CoO layer can best be ascertained by neutron diffraction, which is planned.

In summary, we have observed finite-size effects in CoO, an AF insulator with localized moments, by direct susceptibility measurements. The Néel temperature follows the finite-size scaling relation with a shift exponent of $\lambda = 1.55 \pm 0.05$. For very thin CoO layers, ferromagnetic characteristics with increasing remanence and magnetization have been observed. The uncompensated moments in the CoO layers exhibit oscillation in magnetization.

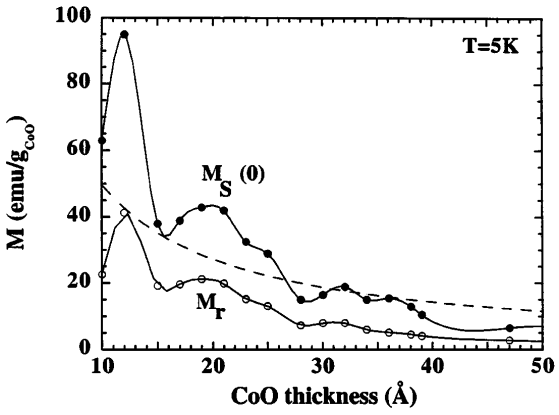

FIG. 5. Spontaneous magnetization $[M_s(0)]$ (solid circles) and remnant magnetization (M_r) (open circles) and at 5 K of CoO/SiO_2 multilayers as a function of CoO thickness t. The dashed curve is the expected magnetization for one uncompensated (100) CoO plane (see text).

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