March 2003

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Ruihau Cheng
University of Nebraska-Lincoln

A. N. Caruso
University of Nebraska-Lincoln, carusoan@umkc.edu

Lu Yuan
University of Nebraska-Lincoln, lyuan2@bigred.unl.edu

Sy_Hwang Liou
University of Nebraska-Lincoln, sliou@unl.edu

Peter A. Dowben
University of Nebraska-Lincoln, pdowben@unl.edu

Magnetic coupling in $Co/Cr_2O_3/CrO_2$ "trilayer" films

Ruihua Cheng, A. N. Caruso, L. Yuan, S.-H. Liou, and P. A. Dowben[a)]
Department of Physics and Astronomy and the Center for Materials Research and Analysis (CMRA), Behlen Laboratory of Physics, University of Nebraska-Lincoln, Lincoln, Nebraska 68588-0111

(Received 23 October 2002; accepted 13 January 2003)

The ferromagnetic coupling between Co and CrO_2, through an insulator (Cr_2O_3) was characterized by *in situ* magneto-optic Kerr effect. By evaporating 20–60 Å Co thin films on top of epitaxial CrO_2 films, a $Co/Cr_2O_3/CrO_2$ trilayer system can be readily fabricated; this is possible because the native surface layer of CrO_2 is Cr_2O_3. *In situ* x-ray photoemission studies show that the Co is oxidized at the interface between Co and Cr_2O_3, so that the system more resembles $Co/CoO/Cr_2O_3/CrO_2$. The Co thickness and temperature dependence of the magnetic hysteresis loops indicate that magnetic coupling strength increases with increasing Co thickness and decreases with increasing temperature. The magnetic coupling through the insulator barrier may be related to defect states in the insulating barrier layer. [DOI: 10.1063/1.1558212]

Interlayer coupling between two ferromagnetic films, separated by a nonmagnetic layer, is now fairly well understood,[1] but exchange coupling of two ferromagnetic layers through a nonmagnetic and nonmetallic spacer layer also exists for amorphous semiconductor and insulator spacer materials.[2–5] This coupling sometimes appears to be distinct from the very low temperature tunneling phenomena between two ferromagnets, through a dielectric spacer layer,[6–9] as the coupling is sometimes oscillatory.[2–5] There is an increasing body of evidence that impurities, in the insulating layer, will "dope" the insulating layer and alter the net polarization of electrons injected into the insulating layer,[10–17] with strong temperature effects,[12,14] most recently demonstrated in Cr_2O_3.[14] Because of the interest in CrO_2-insulating-ferromagnetic FM/I/FM junctions,[14,16,18,19] including $CrO_2/Cr_2O_3/Co$,[19] we have examined the magnetic coupling properties between Co and CrO_2 ferromagnetic layers "through" the stable insulating Cr_2O_3 surface of CrO_2.

The CrO_2 epitaxial thin films were fabricated by chemical vapor deposition with 100 atm of oxygen, using CrO_3 as the molecular precursor, on single crystal rutile $TiO_2(100)$ substrates. The reaction at 390 °C, leads to growth of a stable CrO_2 phase. The x-ray diffraction data exhibit the sharp diffraction lines characteristic of high quality epitaxial CrO_2 thin films grown on $TiO_2(100)$.[20] The CrO_2 samples, of thickness 1–3 μm, were placed in UHV chambers equipped with x-ray photoemission spectrometer (XPS) and magneto-optical Kerr effect (MOKE). Prior to Co evaporation, the CrO_2 samples were cleaned by sputtering and annealing to remove surface contamination.[16,20–22] From the outset, i.e., from the initial stages of surface preparation, the core level binding energies indicated that the stable surfaces were Cr_2O_3. The native Cr_2O_3 surface on CrO_2[22] was used as a native insulating spacer layer between CrO_2 and Co, as has been undertaken elsewhere.[19]

Samples were fabricated with 20-, 40-, and 60-Å-thick Co layers. We have established, by XPS, that Co does oxidize at the Cr_2O_3 interface, leading to CoO formation at the Co/Cr_2O_3 interface,[21] and this will diminish the nominal thickness of the Co layer. Because the Co oxidizes at the interface between Co and Cr_2O_3/CrO_2, following Co evaporation, we have actually formed $Co/CoO/Cr_2O_3/CrO_2$ multilayers, and an insulating barrier layer, effectively, of greater thickness. Although CoO and Cr_2O_3 are nominally antiferromagnetic, with Néel temperatures around 297 and 307 K, respectively, by considering the fact that Néel temperature decreases in very thin films, the samples studied in this work appear to behave much like a ferromagnetic/paramagnetic insulator/ferromagnetic trilayer system in the temperature range of 240–400 K. We used *in situ* longitudinal MOKE to obtain the hysteresis loops, before and after cobalt deposition, as described elsewhere.[20] For all measurements, the applied field was in the plane of the film along the *c* axis of the CrO_2, the easy axis of CrO_2, and we note that the remnant magnetization is highest along the *c* axis.[20]

The hysteresis loops of the $Co/CoO/Cr_2O_3/CrO_2$ multilayer samples show step-like behavior, as shown in Fig. 1. These steps in the hysteresis loop indicate that the Co and CrO_2 layers flip at different fields. In order to get a clear picture of the magnetic coupling, if any, between these two ferromagnetic layers, we measured the minor loops. First, we applied a magnetic field at 200 Oe or −200 Oe to saturate the sample, then swept the field in a range smaller than the coercivity of the "complete" hysteresis loop. Figure 1 shows the complete hysteresis loop for this Co(40 Å)/$CoO/Cr_2O_3/CrO_2$ multilayer FM/I/FM "trilayer"-like system and the minor loops, taken at 338 K. The coercivity of the Co top layer is far larger than the coercivity of the CrO_2 layer for all samples, in spite of the considerable difference in thickness. The hysteresis loops for CrO_2 alone (inset to Fig. 1) are similar to the minor loops in this $Co/CoO/Cr_2O_3/CrO_2$ multilayer system, and it is clear that these minor loops are the consequence of reversal in the CrO_2 underlayer alone. There is a shift between the centers of two minor loops, denoted as $2H_{ex}$, which is the external field required to cancel out the magnetic interlayer coupling.

[a)]Author to whom correspondence should be addressed; electronic mail: pdowben@unl.edu

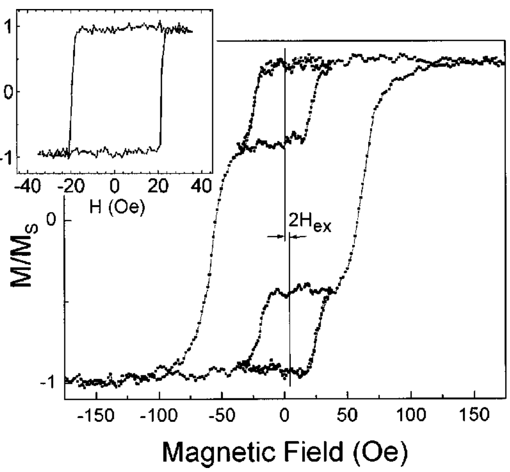

FIG. 1. The complete hysteresis loop and minor loops measured at 338 K for the sample Co(40 Å)/Cr_2O_3/CrO_2. The minor loops were obtained after saturation and $2H_{ex}$ is the shift between the centers of the two minor loops. The hysteresis loop for the CrO_2 thin film substrate (alone) is shown as the inset, also at 338 K.

The small value and positive sign of H_{ex} in our data, as illustrated by the data obtained Co(40 Å)/CoO/Cr_2O_3/CrO_2 in Fig. 1, compellingly suggests that the Co layers and the CrO_2 layer are weakly ferromagnetically coupled through the CoO/Cr_2O_3 barrier, with weak temperature dependence similar to that suggested by the model of Bruno.[23] Weak ferromagnetic coupling was also observed for thicker insulating barriers in Fe/MgO/Fe/Co multilayers.[5]

If the two ferromagnetic layers, with uniaxial anisotropy, are coupled ferromagnetically, the total energy of this system can be expressed[24,25] as

$$\begin{aligned} E = & -M_{CrO_2} t_{CrO_2} H \cos(\theta - \beta) \\ & + K_1^{CrO_2} \sin^2 \beta - M_{Co} t_{Co} H \cos(\theta - \alpha) \\ & + K_1^{Co} \sin^2 \alpha - J_1(T) \cos(\alpha - \beta), \end{aligned}$$

where M_{CrO_2}, t_{CrO_2}, $K_1^{CrO_2}$ are the magnetization, thickness and first order anisotropy constant of the CrO_2 layer, respectively, while M_{Co}, t_{Co}, K_1^{Co} are the magnetization, film thickness, and first anisotropy constant for the Co layer, respectively, and α, β, θ are the angle of M_{CO}, M_{CrO_2}, and H with respect to the easy axis respectively, H is the applied field and J_1 is the coupling constant between the two ferromagnetic layers. CrO_2 and Co are ferromagnetically coupled when $\alpha = \beta$. Both the magnetic state of the trilayer system and the coercivity can be determined from this energy expression. The value of H_{ex}, in Fig. 1, is related to the coupling constant J_1 by $J_1 = H_{ex} M_{CrO_2} t_{CrO_2}$.

From the hysteresis loops for the Co(40 Å)/CoO/Cr_2O_3/CrO_2 and Co(60 Å)/CoO/Cr_2O_3/CrO_2 multilayer FM/I/FM trilayer-like systems, we obtained H_{ex} as a function of temperature and this is plotted in Fig. 2. It is evident that for 40 Å Co and 60 Å Co Co/CoO/Cr_2O_3/CrO_2 samples, the measured H_{ex} increases slowly with increasing temperature. The magnetization of CrO_2 decreases very fast as the temperature approaches the Curie temperature of CrO_2 is 390 K.[20] As a result, the coupling constant J_1 must decrease with increasing temperature.

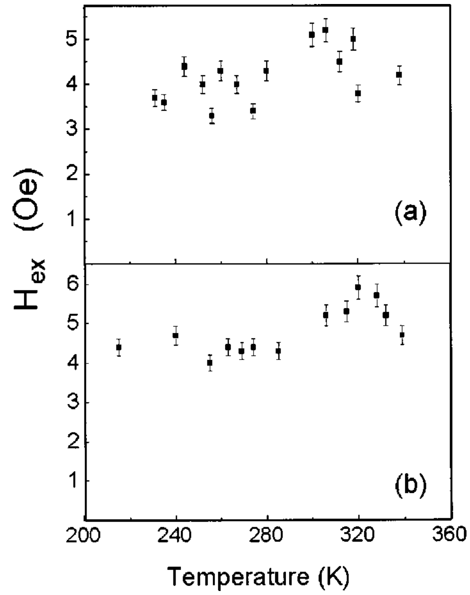

FIG. 2. H_{ex} as a function of temperature for Co/Cr_2O_3/CrO_2 multilayers of different Co thickness. Panel (a) shows the result for sample with 40 Å Co and panel (b) shows the results for 60 Å Co.

The picture of coupling between the Co and CrO_2 layers is clearer when we plot the coercive switching fields for the Co and CrO_2 layers in the Co/CoO/Cr_2O_3/CrO_2 multilayers, as a function of temperature, depicted in Fig. 3. We define the coercive switching field of each layer, according to the step-like complete hysteresis loops, which, although offset in magnetization, provide an indication of the individual layer coercive field. These "coercive" fields, as a function of temperature, differ for different Co thickness Co/CoO/Cr_2O_3/CrO_2 multilayer samples, as seen in Fig. 3. It is obvious that with a Co layer (forming the Co/CoO/Cr_2O_3/CrO_2 multilayers instead of the Cr_2O_3/CrO_2 bilayer), the coercive switching field of CrO_2 underlayer is larger than for the bare CrO_2 layer. The differences between coercive fields for bare CrO_2 layer alone, without any Co coverage (plotted in each panel of Fig. 3 for reference), and in the Co/CoO/Cr_2O_3/CrO_2 multilayers, increase with increasing Co coverages. This increase in the effective CrO_2

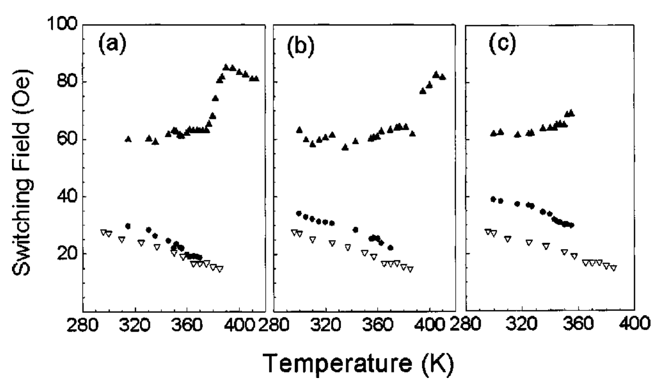

FIG. 3. Switching field as a function of temperature for a variety of Co/Cr_2O_3/CrO_2 multilayer samples with different Co thickness. (▲) shows the switching field of Co layer in the Co/CoO/Cr_2O_3/CrO_2 multilayers, (●) shows the switching field of CrO_2 underlayer in the Co/CoO/Cr_2O_3/CrO_2 multilayers, and (▽) shows the switching field of CrO_2 without any Co overlayer coverage, for reference. Panel (a) shows the data for the sample with 20 Å Co, panel (b) shows the data for 40 Å Co and panel (c) shows the data for 60 Å Co.

coercive field is the result of the ferromagnetic coupling between Co and CrO_2.

Strong ferromagnetic coupling would tend to make the Co and CrO_2 ferromagnetic layers switch together. While the coercive field of Co is larger than the CrO_2 layer by itself, in spite of the disparity in film thickness, these two layers still do not reverse magnetization ''together.'' Rather there is an increase in the coercive switching field of CrO_2 layer, in the Co/CoO/Cr_2O_3/CrO_2 multilayer, compared to the CrO_2 alone. This perturbation of the CrO_2 coercive field, by a cobalt layer, and the existence of a nonzero H_{ex} make it is clear that while there is coupling between the ferromagnetic layers, the coupling must be weak.

As temperature increases and approaches the CrO_2 Curie temperature, M_{CrO_2} and $K_1^{CrO_2}$ decrease and eventually CrO_2 becomes paramagnetic. From the energy equation for a trilayer system it is clear that Co becomes the driving layer near the T_c of CrO_2, but this does not mean that the ferromagnetic Co cannot continue to weakly polarize the CrO_2 layer above the CrO_2 T_c. There is evidence of both phenomena in our MOKE data.

As temperature increases toward the Curie temperature of CrO_2 (390–397 K), the coercive switching field of CrO_2, in the Co/CoO/Cr_2O_3/CrO_2 multilayer, decreases and approaches the coercive switching field of CrO_2 alone, while the coercive switching field of the Co top layer increases. This supports the contention that the CrO_2 layer tends to be the ''spectator,'' while the cobalt layer tends to be the ''actor'' or ''driver,'' but this spectator behavior of CrO_2 is more extreme near the CrO_2 Curie temperature. At temperatures above the Curie temperature of CrO_2 (390–397 K), there is a critical temperature, above which, the Co layer, in the Co/CoO/Cr_2O_3/CrO_2 multilayer, exhibits decreasing coercivity. This latter critical temperature in the cobalt layer behavior increases with increasing thickness of the Co layer in the Co/CoO/Cr_2O_3/CrO_2 multilayer from 390 K for the sample with 20 Å Co coverage to 405 K for 40 Å Co. Since the coupling between the Co and the CrO_2 layers is stronger with increasing Co thickness, it should not be too surprising that the induced polarization of ''paramagnetic CrO_2'' also increases with increasing Co layer thickness.

The mechanisms for the weak ferromagnetic coupling between Co and CrO_2, in the Co/CoO/Cr_2O_3/CrO_2 multilayer, have not been precisely identified. Simple Ruderman–Kittel–(Kasuya)–Yosida coupling,[26] perpendicular coupling,[26,27] or orange peel effect (correlated roughness) coupling[5] of the ferromagnetic layers isolated by a nonmagnetic, nonmetallic barrier layer, as has been suggested for other oxide barrier layers, are not completely appropriate models for the coupling of Co and CrO_2 through the CoO/Cr_2O_3 barrier, and do little to explain the polarization of the CrO_2 layer above the CrO_2 T_c. Models based on tunneling of the wave functions of each ferromagnet through the insulating barrier,[23,28] applied elsewhere to the Fe/MgO/Fe/Co system,[5] require unrealistically small barrier heights. We have observed weak coupling above the antiferromagnetic polytype Néel temperature of Cr_2O_3 through a dielectric barrier layer material with a band gap well above 2 eV and without an appreciable density of states at E_F.[14,16] The fact that both Cr_2O_3 and CoO are insulators does not alter that these barrier layers will weakly polarize, with increasing polarization with increasing temperature.[21] Polarization of defects within the barrier layer[16,21] could be one possible mechanism for coupling.

The support of the NSF (DMR 98-02126), the Office of Naval Research, the NSF MRSEC (DMR 0213808), the Center for Materials Research and Analysis (CMRA), and the Nebraska Research Initiative are gratefully acknowledged. The authors would like to acknowledge a number of helpful conversations with B. Doudin and E. Tsymbal.

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