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Magnetic coupling in Co/Cr₂O₃/CrO₂ “trilayer” films

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The ferromagnetic coupling between Co and CrO₂, through an insulator (Cr₂O₃) was characterized by *in situ* magneto-optic Kerr effect. By evaporating 20–60 Å Co thin films on top of epitaxial CrO₂ films, a Co/Cr₂O₃/CrO₂ trilayer system can be readily fabricated; this is possible because the native surface layer of CrO₂ is Cr₂O₃. *In situ* x-ray photoemission studies show that the Co is oxidized at the interface between Co and Cr₂O₃, so that the system more resembles Co/CoO/Cr₂O₃/CrO₂. 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. © 2003 American Institute of Physics. [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 Cr₂O₃.¹⁴ Because of the interest in CrO₂-insulating-ferromagnetic FM/I/FM junctions,^{14,16,18,19} including CrO₂/Cr₂O₃/Co,¹⁹ we have examined the magnetic coupling properties between Co and CrO₂ ferromagnetic layers “through” the stable insulating Cr₂O₃ surface of CrO₂.

The CrO₂ epitaxial thin films were fabricated by chemical vapor deposition with 100 atm of oxygen, using CrO₃ as the molecular precursor, on single crystal rutile TiO₂(100) substrates. The reaction at 390 °C, leads to growth of a stable CrO₂ phase. The x-ray diffraction data exhibit the sharp diffraction lines characteristic of high quality epitaxial CrO₂ thin films grown on TiO₂(100).²⁰ The CrO₂ 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 CrO₂ 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 Cr₂O₃. The native Cr₂O₃ surface on CrO₂²² was used as a native insulating spacer layer between CrO₂ 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 Cr₂O₃ interface, leading to CoO formation at the Co/Cr₂O₃ interface,²¹ and this will diminish the nominal thickness of the Co layer. Because the Co oxidizes at the interface between Co and Cr₂O₃/CrO₂, following Co evaporation, we have actually formed Co/CoO/Cr₂O₃/CrO₂ multilayers, and an insulating barrier layer, effectively, of greater thickness. Although CoO and Cr₂O₃ 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 CrO₂, the easy axis of CrO₂, and we note that the remnant magnetization is highest along the *c* axis.²⁰

The hysteresis loops of the Co/CoO/Cr₂O₃/CrO₂ multilayer samples show step-like behavior, as shown in Fig. 1. These steps in the hysteresis loop indicate that the Co and CrO₂ 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/Cr₂O₃/CrO₂ 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 CrO₂ layer for all samples, in spite of the considerable difference in thickness. The hysteresis loops for CrO₂ alone (inset to Fig. 1) are similar to the minor loops in this Co/CoO/Cr₂O₃/CrO₂ multilayer system, and it is clear that these minor loops are the consequence of reversal in the CrO₂ 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.

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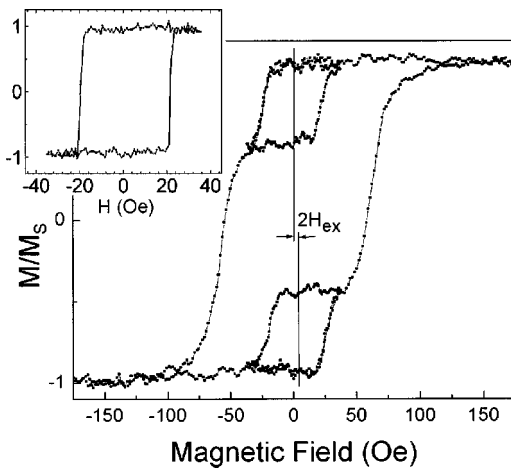


FIG. 1. The complete hysteresis loop and minor loops measured at 338 K for the sample Co(40 Å)/Cr₂O₃/CrO₂. The minor loops were obtained after saturation and $2H_{\text{ex}}$ is the shift between the centers of the two minor loops. The hysteresis loop for the CrO₂ thin film substrate (alone) is shown 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/Cr₂O₃/CrO₂ in Fig. 1, compellingly suggests that the Co layers and the CrO₂ layer are weakly ferromagnetically coupled through the CoO/Cr₂O₃ 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

$$E = -M_{\text{CrO}_2} t_{\text{CrO}_2} H \cos(\theta - \beta) + K_1^{\text{CrO}_2} \sin^2 \beta - M_{\text{Co}} t_{\text{Co}} H \cos(\theta - \alpha) + K_1^{\text{Co}} \sin^2 \alpha - J_1(T) \cos(\alpha - \beta),$$

where M_{CrO_2} , t_{CrO_2} , $K_1^{\text{CrO}_2}$ are the magnetization, thickness and first order anisotropy constant of the CrO₂ 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. CrO₂ 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 $J_1 = H_{\text{ex}} M_{\text{CrO}_2} t_{\text{CrO}_2}$.

From the hysteresis loops for the Co(40 Å)/CoO/Cr₂O₃/CrO₂ and Co(60 Å)/CoO/Cr₂O₃/CrO₂ 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/Cr₂O₃/CrO₂ samples, the measured H_{ex} increases slowly with increasing temperature. The magnetization of CrO₂ decreases very fast as the temperature approaches the Curie temperature of CrO₂ is 390 K.²⁰ As a result, the coupling constant J_1 must decrease with increasing temperature.

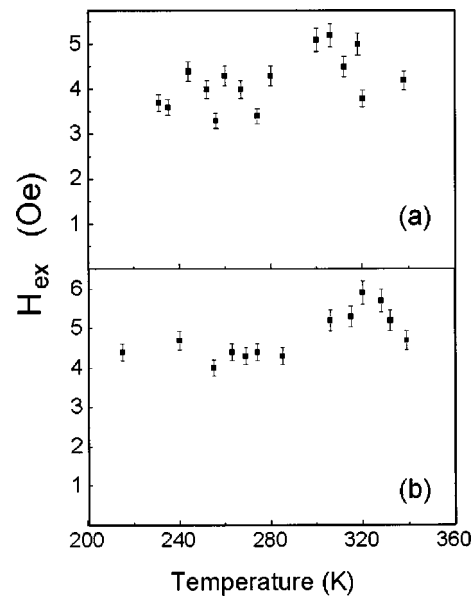


FIG. 2. H_{ex} as a function of temperature for Co/Cr₂O₃/CrO₂ multilayers of different Co thickness. Panel (a) shows the result for sample with 40 Å Co and panel (b) shows the results for 60 Å Co.

The picture of coupling between the Co and CrO₂ layers is clearer when we plot the coercive switching fields for the Co and CrO₂ layers in the Co/CoO/Cr₂O₃/CrO₂ 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/Cr₂O₃/CrO₂ multilayer samples, as seen in Fig. 3. It is obvious that with a Co layer (forming the Co/CoO/Cr₂O₃/CrO₂ multilayers instead of the Cr₂O₃/CrO₂ bilayer), the coercive switching field of CrO₂ underlayer is larger than for the bare CrO₂ layer. The differences between coercive fields for bare CrO₂ layer alone, without any Co coverage (plotted in each panel of Fig. 3 for reference), and in the Co/CoO/Cr₂O₃/CrO₂ multilayers, increase with increasing Co coverages. This increase in the effective CrO₂

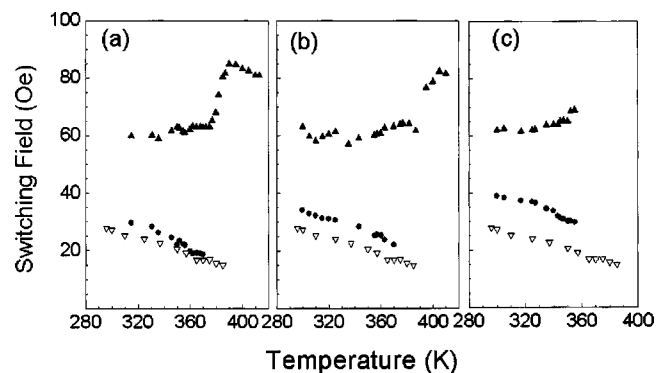


FIG. 3. Switching field as a function of temperature for a variety of Co/Cr₂O₃/CrO₂ multilayer samples with different Co thickness. (▲) shows the switching field of Co layer in the Co/CoO/Cr₂O₃/CrO₂ multilayers, (●) shows the switching field of CrO₂ underlayer in the Co/CoO/Cr₂O₃/CrO₂ multilayers, and (▽) shows the switching field of CrO₂ 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 CrO₂.

Strong ferromagnetic coupling would tend to make the Co and CrO₂ ferromagnetic layers switch together. While the coercive field of Co is larger than the CrO₂ 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 CrO₂ layer, in the Co/CoO/Cr₂O₃/CrO₂ multilayer, compared to the CrO₂ alone. This perturbation of the CrO₂ 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 CrO₂ Curie temperature, M_{CrO_2} and $K_1^{\text{CrO}_2}$ decrease and eventually CrO₂ becomes paramagnetic. From the energy equation for a trilayer system it is clear that Co becomes the driving layer near the T_c of CrO₂, but this does not mean that the ferromagnetic Co cannot continue to weakly polarize the CrO₂ layer above the CrO₂ T_c . There is evidence of both phenomena in our MOKE data.

As temperature increases toward the Curie temperature of CrO₂ (390–397 K), the coercive switching field of CrO₂, in the Co/CoO/Cr₂O₃/CrO₂ multilayer, decreases and approaches the coercive switching field of CrO₂ alone, while the coercive switching field of the Co top layer increases. This supports the contention that the CrO₂ layer tends to be the “spectator,” while the cobalt layer tends to be the “actor” or “driver,” but this spectator behavior of CrO₂ is more extreme near the CrO₂ Curie temperature. At temperatures above the Curie temperature of CrO₂ (390–397 K), there is a critical temperature, above which, the Co layer, in the Co/CoO/Cr₂O₃/CrO₂ 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/Cr₂O₃/CrO₂ 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 CrO₂ layers is stronger with increasing Co thickness, it should not be too surprising that the induced polarization of “paramagnetic CrO₂” also increases with increasing Co layer thickness.

The mechanisms for the weak ferromagnetic coupling between Co and CrO₂, in the Co/CoO/Cr₂O₃/CrO₂ multilayer, have not been precisely identified. Simple Ruderman–Kittel–(Kasuya)–Yosida coupling,²⁶ perpendicular coupling,^{26,27} or orange peel effect (correlated roughness) coupling⁵ 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 CrO₂ through the CoO/Cr₂O₃ barrier, and do little to explain the polarization of the CrO₂ layer above the CrO₂ 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 Cr₂O₃ 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 Cr₂O₃ 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.

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- ¹F. J. Himpsel, J. E. Ortega, G. J. Mankey, and R. F. Willis, *Adv. Phys.* **47**, 511 (1998).
- ²B. Briner and M. Landolt, *Phys. Rev. Lett.* **73**, 340 (1994); B. Briner and M. Landolt, *Z. Phys. B: Condens. Matter* **97**, 459 (1995).
- ³M. Hunziker and M. Landolt, *Phys. Rev. Lett.* **84**, 4713 (2000).
- ⁴G. J. Strikers, J. T. Kohlepp, H. J. M. Swagten, and W. J. M. de Jonge, *Phys. Rev. Lett.* **84**, 1812 (2000).
- ⁵J. Faure-Voncent, C. Tiusan, C. Bellouard, E. Popova, M. Hehn, F. Montaigne, and A. Schuhl, *Phys. Rev. Lett.* **89**, 107206 (2002).
- ⁶J. S. Moodera, L. R. Kinder, T. M. Wong, and R. Meservey, *Phys. Rev. Lett.* **74**, 3273 (1996).
- ⁷T. Miyazaki and N. Tezuda, *J. Magn. Magn. Mater.* **139**, L231 (1995).
- ⁸J. M. de Teresa, A. Barthelemy, A. Fert, J. P. Contour, F. Montaigne, and P. Seneor, *Science* **286**, 507 (1999).
- ⁹M. Ziese, *Rep. Prog. Phys.* **65**, 143 (2002).
- ¹⁰M. E. Flatte and J. M. Beyers, *Phys. Rev. Lett.* **84**, 4220 (2000).
- ¹¹C. L. C. Ochi and N. Majlis, *Phys. Rev. B* **51**, 14221 (1995).
- ¹²Y. Endo, O. Kitakami, and Y. Shimada, *IEEE Trans. Magn.* **34**, 906 (1998).
- ¹³H. Itoh and J. Inoue, *Surf. Sci.* **493**, 748 (2001).
- ¹⁴R. Cheng, T. Komesu, H.-K. Jeong, L. Yuan, S.-H. Liou, B. Doudin, P. A. Dowben, and Ya. B. Losovyj, *Phys. Lett. A* **302**, 211 (2002).
- ¹⁵J. S. Parker, S. M. Watts, P. G. Inanov, and P. Xiong, *Phys. Rev. Lett.* **88**, 196601 (2002).
- ¹⁶A. Sokolov, C.-S. Yang, L. Yuan, S.-H. Liou, R. Cheng, H.-K. Jeong, T. Komesu, B. Xu, C. N. Borca, P. A. Dowben, and B. Doudin, *Europhys. Lett.* **58**, 448 (2002).
- ¹⁷A. M. Bratkovsky, *Phys. Rev. B* **56**, 2344 (1997).
- ¹⁸J. D. M. Coey, A. E. Berkowitz, L. Balcells, F. F. Putris, and A. Barry, *Phys. Rev. Lett.* **80**, 3815 (1998).
- ¹⁹A. Barry, J. D. M. Coey, and M. Viret, *J. Phys.: Condens. Matter* **12**, L173 (2001).
- ²⁰R. Cheng, Z. Y. Liu, X. Bo, S. Adenwalla, L. Yuan, S.-H. Liou, and P. A. Dowben, *Mater. Lett.* **56**, 295 (2002).
- ²¹R. Cheng, C. N. Borca, N. Pilet, B. Xu, L. Yuan, B. Doudin, S. H. Liou, and P. A. Dowben, *Appl. Phys. Lett.* **81**, 2109 (2002).
- ²²R. Cheng, B. Xu, C. N. Borca, A. Sokolov, C.-S. Yang, L. Yuan, S.-H. Liou, B. Doudin, and P. A. Dowben, *Appl. Phys. Lett.* **79**, 3122 (2001).
- ²³P. Bruno, *Phys. Rev. B* **52**, 411 (1995).
- ²⁴T. Yoroazu and X. Hu, *J. Appl. Phys.* **86**, 1581 (1999).
- ²⁵J. P. Wang, Z. S. Shan, S. N. Poramanayagam, and T. C. Chong, *IEEE Trans. Magn.* **37**, 1445 (2001).
- ²⁶Y. Ijiri, *J. Phys.: Condens. Matter* **14**, R947 (2002).
- ²⁷Y. Ijiri, J. A. Borchers, R. W. Erwin, S.-H. Lee, P. J. van der Zaag, and R. M. Wolf, *Phys. Rev. Lett.* **80**, 608 (1998).
- ²⁸J. C. Slonczewski, *Phys. Rev. B* **39**, 6995 (1989).