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Magnetic coupling in Co/Cr$_2$O$_3$/Co$_2$ O$_2$ “trilayer” films

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The ferromagnetic coupling between Co and Cr$_2$O$_3$ through an insulator (Cr$_2$O$_3$) was characterized by in situ magneto-optic Kerr effect. By evaporating 20–60 Å Co thin films on top of epitaxial Cr$_2$O$_2$ films, a Co/Cr$_2$O$_3$/Co$_2$O$_2$ trilayer system can be readily fabricated: this is possible because the native surface layer of Cr$_2$O$_2$ is Cr$_2$O$_3$. In situ x-ray photoemission studies show that the Co is oxidized at the interface between Co and Cr$_2$O$_3$, so that the system more resembles Co/CrO$_2$/Cr$_2$O$_3$/Cr$_2$O$_2$. 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.

Interlayer coupling between two ferromagnetic films, separated by a nonmagnetic layer, is now fairly well understood, but exchange coupling of two ferromagnetic layers through a nonmagnetic and nonmetallic spacer layer also exists for amorphous semiconductor and insulator spacer materials. This coupling sometimes appears to be distinct from the very low temperature tunneling phenomena between two ferromagnets, through a dielectric spacer layer, as the coupling is sometimes oscillatory. 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, with strong temperature effects, most recently demonstrated in Cr$_2$O$_3$. Because of the interest in CrO$_2$-insulating-ferromagnetic FM/I/FM junctions, including CrO$_2$/Cr$_2$O$_3$/Co$_2$, we have examined the magnetic coupling properties between Co and CrO$_2$ ferromagnetic layers “through” the stable insulating Cr$_2$O$_3$ 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). 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.

Samples were fabricated with 20-, 40-, and 60-Å-thick Co layers. We have established, by XPS, that Co does oxidize at the Cr$_2$O$_3$ interface, leading to CoO formation at the Co/Cr$_2$O$_3$ interface, and this will diminish the nominal thickness of the Co layer. Because the Co oxidizes at the interface between Co and Cr$_2$O$_3$/Cr$_2$O$_2$, following Co evaporation, we have actually formed Co/CoO/Cr$_2$O$_3$/Cr$_2$O$_2$ multilayers, and an insulating barrier layer, effectively, of greater thickness. Although CoO and Cr$_2$O$_3$ 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. 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 Co, and we note that the remnant magnetization is highest along the c axis.

The hysteresis loops of the Co/CoO/Cr$_2$O$_3$/Cr$_2$O$_2$ multilayer samples show step-like behavior, as shown in Fig. 1. These steps in the hysteresis loop indicate that the Co and Cr$_2$O$_3$ 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$_2$O$_3$/Cr$_2$O$_2$ 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 Cr$_2$O$_3$ layer for all samples, in spite of the considerable difference in thickness. The hysteresis loops for Cr$_2$O$_2$ alone (inset to Fig. 1) are similar to the minor loops in this Co/CoO/Cr$_2$O$_3$/Cr$_2$O$_2$ multilayer system, and it is clear that these minor loops are the consequence of reversal in the Cr$_2$O$_2$ underlayer alone. There is a shift between the centers of two minor loops, denoted as $2H_c$, which is the external field required to cancel out the magnetic interlayer coupling.
The small value and positive sign of $H_{ex}$ in our data, as illustrated by the data obtained Co(40 Å)/CoO/CoO/CrO 2 multilayer samples, as seen in Fig. 3. It is obvious that with a Co layer (forming the Co/CoO/CoO/CrO 2 multilayers instead of the Co/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/CoO/CrO 2 multilayers, increase with increasing Co coverages. This increase in the effective CrO 2

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/CoO/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/CoO/CrO 2 multilayer samples, as seen in Fig. 3. It is obvious that with a Co layer (forming the Co/CoO/CoO/CrO 2 multilayers instead of the Co/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/CoO/CrO 2 multilayers, increase with increasing Co coverages. This increase in the effective CrO 2
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 CoO$_2$ layer, in the Co/CoO/Co$_2$O$_3$/CrO$_2$ multilayer, compared to the CrO$_2$ alone. This perturbation of the Co$_2$O$_3$ coercive field, by a cobalt layer, and the existence of a nonzero $H_{ex}$, make it is clear that there is coupling between the ferromagnetic layers, the coupling must be weak.

As temperature increases and approaches the CrO$_2$ Curie temperature, $M_{CoO}$, and $K_{1CrO_2}$ 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 CoO$_2$, in the Co/CoO/Co$_2$O$_3$/CrO$_2$ multilayer, decreases and approaches the coercive switching field of CoO$_2$ 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/Co$_2$O$_3$/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/Co$_2$O$_3$/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/Co$_2$O$_3$/CrO$_2$ multilayer, have not been precisely identified. Simple Ruderman– Kittel–(Kasuya)–Yosida coupling, perpendicular coupling, or orange peel effect (correlated roughness) coupling 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 Co/Co$_2$O$_3$ 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, applied elsewhere to the Fe/MgO/Fe/Co system, require unrealistically small barrier heights. We have observed weak coupling above the antiferromagnetic polytype Néel temperature of CrO$_3$ through a dielectric barrier material with a band gap well above 2 eV and without an appreciable density of states at $E_f$. The fact that both CrO$_3$ and CoO are insulators does not alter that these barrier layers will weakly polarize, with increasing polarization with increasing temperature. Polarization of defects within the barrier layer could be one possible mechanism for coupling.

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