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Temperature dependence of spin waves in Co/CoO bilayers

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Brillouin light scattering measurements of spin-wave frequencies in an exchange coupled ferromagnet/antiferromagnet epitaxial Co/CoO bilayer are reported. A striking temperature dependence of the measured spin-wave frequencies in the cobalt layer in the range 77–300 K was observed which has been demonstrated to be due to exchange coupling to the ultrathin (7 Å) CoO layer as antiferromagnetic order develops. The temperature dependence of the spin-wave frequency demonstrates that interface exchange coupling occurs in the absence of the unidirectional anisotropy. A study of the mode line width shows a broadening with reducing temperature which indicates that locally ordered antiferromagnet regions persist above the Néel temperature and play a central role in determining the magnetic behavior of the bilayer system. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1359463]

Exchange biasing in strongly coupled ferromagnet (FM)/antiferromagnet (AF) structures is of considerable interest due to its relevance to magnetoresistive devices.[1] The effect has been attributed to the interfacial exchange interaction between the layers. There have been numerous studies[2] of the exchange-biasing phenomenon since the first observation by Meiklejohn and Bean.[3] Most studies have mainly concentrated on measurements of static properties. Using Brillouin light scattering (BLS) one can gain insight into the bulk and interface exchange coupling via the spin-wave frequencies. There have, however, been few measurements of spin-wave frequencies in FM/AF bilayers reported to date.[4,5] In this article, a detailed study of the temperature dependent spin-wave frequency in Co/CoO bilayers for the temperature range 77–300 K is presented using the BLS technique. The Néel temperature T_N for cobalt oxide is around 290 K for the bulk material, but T_N is thickness dependent in thin films, as has been demonstrated,[6] and so can be reduced in sufficiently thin films. A marked increase in the Co spin-wave frequencies with reducing temperature has been previously reported[7] for ultrathin Co/CoO bilayer structures. This study prompted the development by Stamps, Camley, and Hicken[8] of a possible theoretical description for the observed temperature dependent behavior in terms of an exchange coupled temperature dependent spin configuration from the AF layer. We choose to study structures in which the CoO layer is very thin (7 Å) in order to maintain an epitaxial structure throughout the Co/CoO bilayer. For such a thin CoO layer, the T_N should be well below the lowest studied temperature 77 K,[6] and the presence of such a thin layer therefore acts to perturb the Co spin-wave frequencies via the interfacial coupling. This permits a study of the influence of the AF layer spin at the interface, in the case where the interfacial coupling is too weak to give rise to a unidirectional anisotropy.

A Co/CoO bilayer was grown at room temperature on a Cu/Si(001) template by molecular beam epitaxy under ultrahigh vacuum conditions with a base pressure of order 10^{-10} mbar. Prior to loading into the growth chamber, the substrates were chemically cleaned and etched in diluted HF solution. After 30 Å of Co was grown, a shutter is placed to cover half of the substrate and a 30 Å Cu cap was deposited to form a control sample without an oxide layer. The sample was then oxidized for 30 min in 10^{-3} mbar of high purity oxygen, and the shutter repositioned to enable the oxidized half to be capped, again with 30 Å of Cu. Figure 1 shows the reflection high-energy electron diffraction (RHEED) images along the ⟨110⟩ azimuth of the (a) Si(001) substrate (b) 1000 Å Cu/Si(001), (c) 30 Å Co/Cu/Si(001), and (d) the Co layer after oxidation. After completion of the Cu layer, sharp streaks with low background are observed. No qualitative change is observed in the RHEED pattern after completion

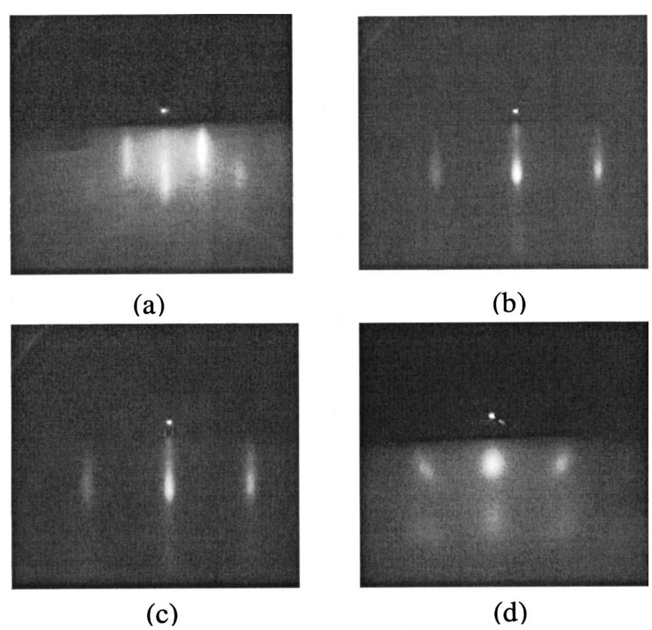

FIG. 1. RHEED patterns at various stages of the sample growth (a) Si(001) substrate, (b) Cu layer, (c) Co film, (d) oxidized Co film.

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TABLE I. The layer thicknesses and magnetic moments for the Cu/Co/Cu(001) and the Cu/CoO/Co/Cu(001) film as determined from PNR measurements.

	Thickness	Magnetic moment	Thickness	Magnetic moment
Cu	56±2 Å		52±2 Å	
CoO			7±2 Å	
Co	31±2 Å	1.27±0.08 μ_B	28±2 Å	1.27±0.08 μ_B
Cu	928±5 Å		928±5 Å	

of the Co layer. This agrees with the earlier finding that three-dimensional epitaxial growth occurs along the [001] direction with the Cu and Co axis rotated in plane by 45° with respect to the Si(001) principle axis. After oxidation, the RHEED pattern of the Co film becomes more spot like. This suggests that the films are still epitaxial, although some disorder is introduced.[9] The in-plane parameter for the Co film becomes smaller after oxidation.

Polarized neutron reflectivity (PNR) experiments were carried out on the CRISP time-of-flight neutron reflectometer at the Rutherford Appleton Laboratory.[10] The samples were held at room temperature with a 7000 Oe magnetic field applied in plane along the direction normal to the scattering plane using an electromagnet. For fitting the PNR data, the magnetic moments and layer thicknesses are adjusted. Several pronounced oscillations are seen in both the reflectivity and spin asymmetry data and excellent fits to the data are obtained throughout the entire q range studied. Similar quality fits are obtained to the Cu/Co/Cu sample. The magnetic moments, layer thicknesses, and interface roughness obtained from the fits are shown in Table I for both the Cu/Co/Cu and Cu/Co/CoO/Cu sample. The fit to the PNR data for the oxidized Co film yields a Co layer thickness of 28 Å and a CoO layer thickness of approximately 7 Å. *Ex situ* magneto-optical Kerr effect measurements show the samples to have fourfold in-plane anisotropy. The oxidized sample shows an increased coercivity (Fig. 2) over the control Co sample as is to be expected from the increased surface disorder shown by the RHEED measurements. Our BLS system

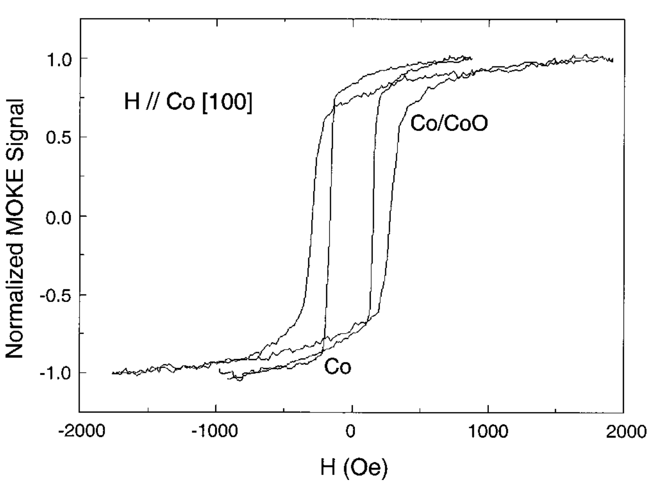

FIG. 2. In-plane MOKE measurements at room temperature showing increased coercivity in the Co/CoO sample along the Co[100] hard axis.

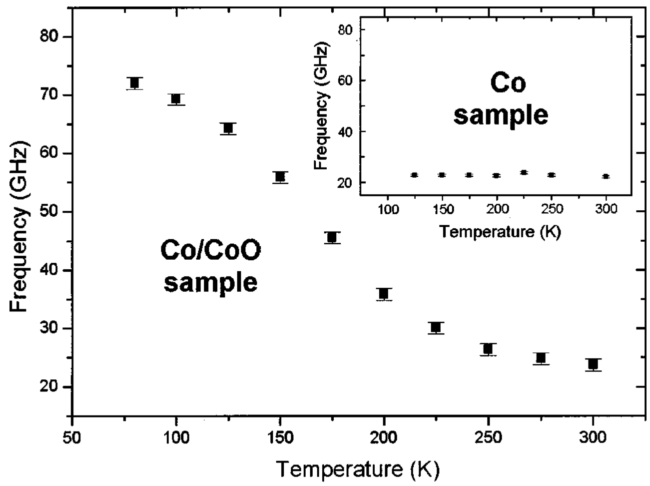

FIG. 3. Temperature-dependent spin-wave frequency of the Co/CoO sample for fields directed along the Co[100] axis. Inset show that of the control Co sample.

uses a 3+3 pass tandem Fabry-Pérot interferometer with a computer-based stabilization system.[11] For the low temperature measurements, the sample was mounted in a customized continuous flow cryostat.

In these epitaxial fcc Co/CoO bilayer structures, the Damon–Eshbach spin-wave modes of the Co layer are observed. Measurements of the spin-wave frequency as a function of in-plane applied field angle were carried out and the angular dependent results reveal that the predominant anisotropy in the bilayer system is of fourfold symmetry.[12] The data from the control Co sample could be well modeled by assuming a bulk Co moment, $K_1/M=-500$ Oe, and including a small uniaxial anisotropy field, $K_u/M=35$ Oe. From these results, the value of the surface anisotropy, K_s, can be deduced which is found to be -0.5 erg cm^{-1} favoring in-plane magnetization. The angle scan from the Co/CoO sample could be fitted using the layer thicknesses and Co layer magnetization from the PNR measurements. K_s is found to be reduced to -0.3 erg cm^{-1} due to the difference between the Co/CoO and Co/Cu interfaces. If it is assumed that the two Co/Cu interfaces in the control sample are equivalent, then one can infer a value of $K_s=-0.1$ erg cm^{-1} for the Co/CoO interface.

Figure 3 shows the temperature dependence of the spin-wave frequency as the sample is cooled in steps of 25 K with magnetic field applied along the hard crystallographic axis. As BLS probes thermally excited spin waves it follows that, at low temperatures, the intensity of the scattered signal is reduced. The frequency can be seen to increase as the temperature is reduced as expected. Similar measurements for the control Co sample show no detectable variation in frequency. Low temperature angle-dependent measurements were performed but were unable to detect any unidirectional anisotropy as expected for such thin CoO layers. Such a variation, if it exists, is likely to be within the errors of the experiment. Due to the finite size effect,[6] a completely ordered AF layer is not achieved in the CoO film at 290 K which greatly exceeds T_N at this temperature. Therefore, as the temperature is reduced and the AF layer begins to order

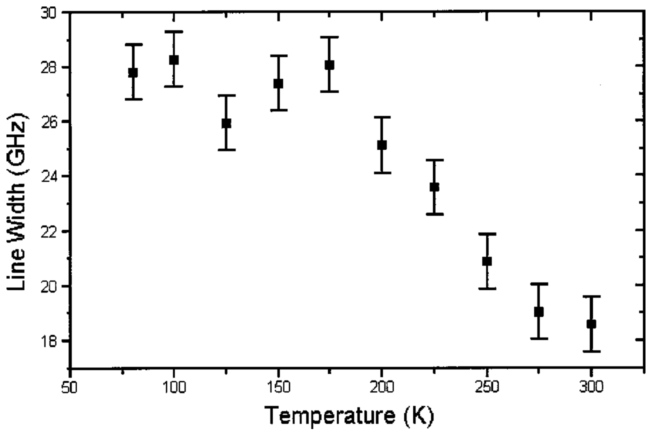

FIG. 4. Temperature-dependent spin-wave linewidth of the Co/CoO sample.

magnetically, the spin waves in the FM layer begin to experience the effect of spin disorder both due to structural imperfections and thermal fluctuations in the AF layer by virtue of the interface coupling. The spin wave frequency increases monotonically from its high temperature value as the temperature is reduced. If the observed low temperature increase in frequency is indeed due to the ordering of the AF to which the FM is coupled, then it follows that, as the system becomes completely ordered, the FM mode frequency must saturate. We therefore conclude that the spin-wave frequency will be at a maximum when the system is completely ordered.

The BLS linewidth (full width at half maximum) is determined from the spectra by fitting peaks with Gaussian functions. Figure 4 shows the mode linewidth as a function of temperature. The instrument broadening is approximately 0.5 GHz. It is clear that a significant broadening occurs as the temperature is reduced. Linewidth studies for the unoxidized sample showed no such behavior and, accordingly, the intensity variations are found to be much smaller. We conclude from the clear difference in the linewidth behavior of the two samples that spin disorder exists in the AF CoO layer at the interface. The microscopic origin of these effects is not yet clear but we may speculate that locally ordered AF spins in random arrangement regions in the AF layer give rise to the increase in linewidth.

In conclusion, we have shown that even in the absence of an exchange bias field associated with the fully AF order state in the ultrathin CoO layer, a spin-wave frequency increase and linewidth broadening can occur due to the onset of AF order in the CoO layer. This result demonstrates that interface exchange coupling occurs in the absence of the unidirectional anisotropy.[5]

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