Measurements of the ferromagnetic/antiferromagnetic interfacial exchange energy in CO/CoO and Fe/FeF_2 layers (invited)

E. Dan Dahlberg,^{a)} Brad Miller, and Bradford Hill

Physics Department and Magnetic Microscopy Center, University of Minnesota, Minneapolis, Minnesota 55455

B. J. Jonsson, Valter Strom, and K. V. Rao

Department of Condensed Matter Physics, Royal Institute of Stockholm, Stockholm, Sweden

Josep Nogues and Ivan K. Schuller

Department of Physics, University of California, San Diego, La Jolla, California

Two measurement techniques, both relying on reversible rotations of the magnetization, have been used to determine the magnitude of the interfacial exchange energy (IEE) between ferromagnetic and antiferromagnetic (F/AF) layers. One technique is to use the anisotropic magnetoresistance to determine rotations of the magnetization away from the unidirectional easy axis, where the rotation is accomplished by applying external magnetic fields less than the effective F/AF exchange field. The second technique uses measurements of the ac susceptibility as a function of the angle between the ac field and the unidirectional exchange field. Both of the reversible process techniques result in values of the IEE larger (by as much as a factor of 10 in Co/CoO bilayers) than the traditional irreversible technique of measuring a shift in the hysteresis loop. The ac susceptibility technique was also used to measure one Fe/FeF₂ bilayer. For this sample, the IEE values obtained by reversible and irreversible methods are equivalent. © 1998 American Institute of Physics. [S0021-8979(98)53611-0]

Meikeljohn and Bean^{1,2} discovered an interfacial exchange energy between ferromagnets and antiferromagnets which could generate a unidirectional easy axis for the ferromagnet. This unidirectional anisotropy is generated by field cooling the system from above the Néel temperature to below. Although this discovery occurred almost 40 years ago, we have not yet developed a fundamental understanding of this phenomena. Briefly, the first model to attempt to describe the observed behavior assumed that the ferromagnet was uniformly coupled to one sublattice of the antiferromagnet. However simple calculations with this model gave interfacial exchange energies (IEE) approximately a factor of 100 times larger than the values reported by experiments. Due to this discrepancy alternate models of the IEE were developed, however, none of these appear to completely explain the observed data.³ Our lack of understanding of the IEE may arise, not from a lack of theoretical understanding but instead from an inherent error in the experimental determination of the IEE.

Historically the experimental determinations of the IEE phenomenon have been almost exclusively from measurements of the shift or offset in the hysteresis loops of direct exchange coupled ferromagnet/antiferromagnet systems. This measurement technique is intrinsically irreversible, and is actually a measure of the nucleation and propagation of domain walls during the reversal process. As such its relation to the exchange bias energy is not necessarily simple. Another difficulty with this technique is the common observation that cycling through multiple hysteresis loops results in decreasing values of the shift in the loops.

We have recently used two different experimental techniques which involve only reversible rotations of the magnetization to determine the interfacial exchange energy in Co/CoO bilayers.^{4,5} These measurements produce values of the energy which are consistently larger than those determined by hysteresis loop measures on the same films. Although these reversible measures are larger than the irreversible measures, they are still less than the original predictions based on a direct coupling between the ferromagnet surface and a single sublattice of the antiferromagnet.

In what follows, we will briefly summarize both the techniques and results from the above mentioned two works. In both, the samples consisted of sputtered Co films (thicknesses from 2 to 30 nm) with a native oxide coating formed upon removal from the sputtering system (thus forming bilayers of Co/CoO). As the CoO has a Néel temperature slightly below room temperature, the exchange bias direction was easily controlled by field cooling the bilayers from room temperature to low temperature. In addition to the Co/CoO work, we will mention one study of another system, Fe exchange coupled to epitaxially grown antiferromagnetic FeF₂. At the end are a series of conclusions which can be drawn from this work.

Our first reversible measurement of the IEE relied on the anisotropic magnetoresistance (AMR) to determine the direction of the magnetization in thin films.⁴ In this work, the resistance of Co/CoO bilayers were measured as a function of the angle between an in-plane applied magnetic field and the exchange bias direction. The applied magnetic fields

0021-8979/98/83(11)/6893/3/\$15.00

6893

^{a)}Electronic mail: dand@physics.spa.umn.edu

were as large as 40% of the effective field of the IEE. Using the relationship for the resistance of a ferromagnet with an angle between the current and the magnetization,⁶ we were able to fit the resistance of the films as a function of the applied field direction with one adjustable parameter, the average exchange biasing energy. As shown in this work, the applied magnetic field could be rotated through a full 2π twice obtaining the same angle dependent resistance data indicating the reversible nature of the measurement technique.

In order to prove a difference between the reversible and irreversible measurements, the maximum possible value for the hysteresis loop measured exchange anisotropy was used, the value of the magnetic field when the magnetization reversed. Thus, this determination included half the width of the hysteresis loop in the shift, i.e., the reported values for the hysteresis loop determination of the exchange bias were the exchange bias field plus the coercivity (this was not clearly stated in this work). This is certainly an overestimate of the exchange bias but even so, it was still smaller than that determined by the reversible AMR technique. In general, when correcting for the coercivity, the AMR determined IEE was about a factor of 4 times larger than the hysteresis loop measurement for films on the order of 4 nm thick.

Another interesting feature found in this work was that the magnetization rotation through the ferromagnetic film was not uniform in the AMR or reversible measurements. Although this was not observed in the hysteresis measurement, it is rather obvious it should be present as shown in the following. The exchange pinning of the ferromagnet occurs at the interface between the Co and the CoO. In a thick Co film, say on the order of 10 nm or more, the spins at the free surface, opposite the interface, are only weakly pinned (the bias must propagate through the film via the direct exchange of the Co from one layer to the next). In this case if a magnetic field is applied at a large angle to the biasing direction, the spins on the free surface rotate towards the field more than those at the pinning interface. Thus, in progressing through the film thickness, the rotation varies from a maximum amount (on the free surface) to a minimum (at the pinning interface). This situation is not dissimilar to having a domain wall or a partial wall form through the thickness of the film.

The second reversible technique we used to measure the IEE was the ac susceptibility. In this case the ac susceptibility was measured as a function of the angle between the exchange bias direction and the ac magnetic field with ac magnetic fields as small as 0.1% of the effective exchange bias field. A simple understanding of how this was used to determine the IEE is to consider the analogy with the susceptibility of an antiferromagnet.⁷ The measured susceptibility of an antiferromagnet depends upon the orientation of the magnetic field to the spins in the sublattices, with the collinear susceptibility smaller than the perpendicular susceptibility. The difference between these susceptibilities is related to the exchange energy between the two sublattices.

The ac susceptibility measurements were performed as a function of temperature revealing a linear temperature dependence of the exchange biasing magnitude (the AMR study was performed only at 4 K). The results from this second study were in agreement with the AMR study, in that this reversible measurement of the IEE was larger than that determined by the irreversible hysteresis loop technique. However, it found energies larger by as much as a factor of 10 than the hysteresis loop measurements (recall a factor of 4 was observed in the AMR studies). It is important to note that the two reversible studies were made on different sets of samples so it is not clear if this difference in the factor is due to the smaller field used in the ac susceptibility study or sample differences.

An interesting feature of note in the susceptibility study came from two Co samples of the same thickness. One was partially capped with Ag prior to oxidation to prevent the formation of the CoO over some fraction of the surface while the other did not have the oxide inhibiting Ag coating. Surprisingly, the hysteresis loop measurement of the sample with the partial Ag overcoat indicated an IEE larger than the sample with the full oxide. This was contradicted by the ac susceptibility measurements on the same two samples which indicated a reduction in the IEE for the sample with the smaller ferromagnetic antiferromagnetic interface area as one would expect. Although this comparison was between only these two samples, it does suggest that there may be difficulties in trying to determine even the systematics of the interfacial exchange energy by hysteresis loop measurements.

Another system we have recently investigated by the ac susceptibility method is Fe deposited on epitaxially grown FeF₂.⁸ This system is rather unique for two reasons; it does not exhibit a decay or alteration of the ferromagnetic antiferromagnetic IEE upon repeated hysteresis loop cycles at low temperatures and the determination of the exchange energy from the loop shifts agree with that determined by the reversible techniques. That the loops are preserved upon repeated cycles is probably the result of two things. The crystalline anisotropy energy of the FeF2 is high and the epitaxial growth with twinning results in the FeF₂ film being large single crystals. Thus the FeF₂ sublattice orientations are well fixed and the applied magnetic field and the exchange from the iron film is insufficient to reorient the sublattice orientations. This might be sufficient to assume the two techniques would give the same value for the IEE, however this means that either the energy and dynamics for domain wall formation (which is localized to the wall width) is identical to the uniform rotation of the magnetization at the interface, or the hysteresis loop reversal mechanism is not by wall formation but instead of coherent rotation.

In ending, a number of conclusions and questions can be drawn from these studies.

First, in general, reversible measurements of the interfacial exchange coupling energy are more accurate than measures relying on irreversible processes. A cautionary note however arises upon considering the anisotropy energy of the antiferromagnet. If the crystalline anisotropy or the crystallite size of the antiferromagnet is small compared to exchange bias energy, then pinning of the ferromagnet is not only by the interfacial exchange coupling but instead will be a mixture of the exchange energy and the anisotropy energy. Thus in this case, even reversible measurements do not provide an accurate determination of the interfacial exchange energy.

Next, there is the question of what the hysteresis loop measures. The irreversible techniques are almost certainly measurements of some combination of domain wall nucleation energies in the ferromagnetic film where the lowest interfacial energy occurs and domain wall pinning. For this reason, even systematic studies of the IEE where grain size or some other parameter is varied is suspect unless their effect on the wall nucleation and pinning are well known.

It is interesting that the Fe/FeF₂ system reveals equivalent results for the IEE using both reversible and irreversible measures. This preliminary result is interesting for the following reasons. The robust nature of the loop shift upon repeated magnetic reversals may be an indicator when the hysteresis loop technique is accurate for determining the IEE. However, when the details are considered, it is surprising that the energetics of wall formation and propagation are the same as the as small rotations of the magnetization. However, as stated, only one sample was investigated and certainly more must be studied before any conclusions are drawn.

Finally there are the apparent differences in the measured values of the energy for the AMR (factor of 4 times larger than the hysteresis determined value) and ac susceptibility (as much as a factor of 10 times larger) techniques. One possibility is this may be due to the differences in the field of measurement as the minimum ac susceptibility magnetic field is approximately 0.01 that of the AMR technique. However, it is important to note that these measurements were not performed on the same set of samples. This clearly warrants a study of the exchange bias energies determined by the ac susceptibility, AMR, and hysteresis loop techniques on the same samples.

Research was supported by ONR Grant No. N/N00014-95-1-0799 and DOE.

- ¹W. H. Meiklejohn and C. P. Bean, Phys. Rev. 102, 1413 (1956).
- ²W. H. Meiklejohn and C. P. Bean, Phys. Rev. **105**, 904 (1957).
- ³A. P. Malozemoff, J. Appl. Phys. **63**, 3874 (1988).
- ⁴B. H. Miller and E. D. Dahlberg, Appl. Phys. Lett. **69**, 3932 (1996).
- ⁵V. Strom, B. J. Jonsson, K. V. Rao, and D. Dahlberg, J. Appl. Phys. 81, 5003 (1997).
- ⁶T. R. McGuire and R. I. Potter, IEEE Trans. Magn. MAG-11, 1018 (1975).
- ⁷C. Kittel, *Introduction to Solid State Physics*, 7th ed. (Wiley, New York, 1996).
- ⁸J. Nogues, D. Lederman, T. J. Moran, I. K. Schuller, and K. V. Rao, Appl. Phys. Lett. **68**, 3186 (1996); J. Nogues, D. Lederman, T. J. Moran, and I. K. Schuller, Phys. Rev. Lett. **76**, 4624 (1996).