Exchange bias relaxation in CoO-Biased Films

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Because the memory of the bias direction is carried by the antiferromagnetic order in exchange biased films, the stability of the antiferromagnetic order is critical to the existence of the exchange bias field. Ferromagnetic resonance was used to measure the relaxation behavior of polycrystalline CoO films coupled to films of $Ni_{80}Fe_{20}$, probing the system on the time scale of the experiment, $\approx 10^3$ s, and the time scale of the magnetic precession, 10^{-10} s. Unidirectional anisotropy (exchange biasing) and isotropic resonance field shifts are observed at the lowest temperatures. Above the apparent exchange bias blocking temperature, isotropic resonance field shifts persist. At still higher temperatures, diminishing resonance field shifts are accompanied by peaks in the FMR linewidth. The results highlight the effects of varying relaxation rates in the CoO relative to the two experimental time scales.

I. INTRODUCTION

Ferromagnetic resonance (FMR) has been shown to be a very powerful tool for investigation of reversible and irreversible behavior in the antiferromagnetic layer in exchange biased bilayers. In addition to measuring unidirectional anisotropy, which in many cases is roughly equivalent to the exchange bias field measured in hysteresis loops, FMR provides a way to measure the effects of the fraction of the antiferromagnetic layer that is not stable, i.e., the fraction that "forgets" the cooling field direction during the experiment[1,2].

The separation of stable and unstable fractions of the antiferromagnet is based on comparison between the relaxation/fluctuation time scale of the antiferromagnet[11,9], τ, with the two time scales of the FMR experiment: the measurement time scale, $\tau_{\rm exp}$, and the period of the microwave excitation, $\tau_{\rm res}$. The relaxation time τ is the typical time required for the AF order to switch from one easy axis to another, or to reverse along the same easy axis[11,9] A measurement of exchange bias by FMR involves sweeping the applied field magnitude to locate the resonance field, $H_{\rm res}$, for each of a number of field directions, usually in the plane of the sample. In the experiments described here, this series of field sweeps requires approximately 15 min ($\tau_{\rm exp} \approx 10^3$ s) to span 360° in the plane of the sample. The microwave excitation period is equal to the precession period of the magnetization at resonance, $\tau_{\rm res} \approx 10^{-10}$ s in these experiments.

In general, the *angular dependence* of the measured resonance fields is due to anisotropy fields which are stable on the experimental time scale. Specifically, the unidirectional exchange biasing is due to interaction only with those CoO grains for which $\tau > \tau_{\rm exp}$.

However, because the resonance field for a particular applied field direction is determined by perturbing the magnetization at microwave frequencies (9.4 GHz here), individual resonance fields are determined by effective anisotropy fields that need only be stable on a much shorter time scale, the period of the perturbation, $\tau_{\rm res} \approx 10^{-10}$ s. The CoO grains with intermediate relaxation times, $\tau_{\rm exp} > \tau > \tau_{\rm res}$, having "forgotten" the initial conditions, will not contribute to exchange bias, but they will contribute to an isotropic resonance field shift, or rotatable anisotropy[1,2].

One might expect only very long relaxation times at low temperatures. However, because the ferromagnetic order can exert a torque on the surface layer of antiferromagnetic spins, the energy of the barrier to reversal may go to zero, and the antiferromagnet may be switched for some orientations of the ferromagnetic magnetization. In this situation, in contrast to the model in ref. 9, exchange bias will not freeze in, and rotatable anisotropy will persist to low temperatures.

Very rapidly fluctuating CoO grains, $\tau < \tau_{\rm res}$ would not affect the resonance field significantly since any effect they might have would tend to average out over the period of a precession.

A summary of the effects and their relationship to the various time scales is given in Table I.

The linewidth of exchange biased films also depends on the relaxation time of the antiferromagnetic order. Linewidth in exchange biased films has been interpreted in a number of different ways. The out-of-plane angular dependence of the linewidth in NiO-biased $Ni_{80}Fe_{20}$ has been favorably compared to the two-magnon model[3–5] where the energy of the uniform precession is coupled to spin-wave modes through nonuniformity of the exchange bias field[6]. The in-plane variations of linewidth in $Ni_{80}Fe_{20}$ biased by FeMn have been interpreted in terms of a large-scale dispersion in the exchange bias field that causes separate regions of the film to resonate at different applied fields[7,8]. These models of linewidth may be expected to apply when the antiferromagnetic order is stable on the precession time scale.

In addition to these linewidth mechanisms, a third mechanism will be required to explain some of the data shown below. The "slow relaxer" linewidth mechanism[12,13,5] involves coupling of the magnetization to entities that have a relaxation time, τ, comparable to the excitation angular frequency, ω. The slow relaxer model predicts a contribution to the linewidth of the form

$$\Delta H \propto \frac{\omega\tau}{1+(\omega\tau)^2}. \qquad (1)$$

For thermally driven relaxation, τ is expected to be strongly temperature dependent, so the slow relaxer linewidth mechanism is expected to produce a peak in the temperature dependence of the linewidth.

The original derivation of the slow relaxer linewidth model was applied to garnet materials where the relaxation involved charge transfer between Fe^{3+} and Fe^{4+} ions. For the purposes of this paper, however, the relaxing entities are thermally excited antiferromagnetic grains, rather than mobile charges. It is interesting to note that this model also predicts a resonance field shift similar to the rotatable anisotropy described above[13].

II. EXPERIMENT

The films measured in this study were deposited by dc magnetron sputtering in 260 mPa (2 mTorr) of Ar on thermal oxide coated Si substrates. The base pressure before depositing was typically 10^{-6} Pa (10^{-8} Torr). Cobalt oxide was deposited by sputtering cobalt with a partial pressure of 2.6 mPa (2×10^{-5} Torr) of oxygen. Permalloy based films were selected after a number of experiments on Co films yielded low quality results due to the large temperature dependence of magnetocrystalline anisotropy in Co. The $Ni_{80}Fe_{20}$ based samples used in the experiments described below are listed in Table II. The major ferromagnetic part of the films consists of $Ni_{80}Fe_{20}$ and the Co metal layer is introduced to prevent the formation of NiFe oxide during CoO deposition or during the formation of native oxide in air. The native oxide thickness was determined from low angle x-ray reflectivity measurements to be 3 to 4 nm thick.

After cooling each sample in a 0.1 T field, the angular dependence of H_{res} was measured at a series of increasing temperatures. At each temperature, resonance fields were measured at 20° field direction intervals in the plane of the sample. Values of angle-averaged resonance fields, H_{ave}, and unidirectional anisotropy, H_{ex} were obtained by fitting H_{res} values to a free energy model[1]. Values of the effective shape anisotropy are determined from out-of-plane FMR measurements at room temperature.

III. RESULTS

The results to be discussed below include measurements of resonance field (fig. 1), linewidth (fig. 2) and exchange bias field (fig. 3).

Angle-averaged in-plane resonance fields are plotted in fig. 1. At room temperature, the resonance fields differ primarily because of film-to-film variations in surface properties. Following the data down from room temperature, there is a change in slope of $H_{\mathrm{res}}(T)$ and a shift toward lower H_{res} values, which is interpreted as the onset of a rotatable anisotropy[1,2].

Linewidth values, measured during cooling and shown in fig. 2, show peaks as a function of temperature, especially for sample B, the sample with the thin native CoO layer. The linewidth *peak* temperatures are slightly lower than the onset of rotatable anisotropy discussed above, but the increased linewidths extend to temperatures above the corresponding rotatable anisotropy onsets in fig. 1. The linewidth increase due to interactions with CoO is quite modest, on the order of 2 mT. For comparison, the linewidth increase observed in NiO-biased films, appropriately scaled is approximately 10 mT[1].

Finally, the exchange bias field, determined from fits to the angular dependence of the in-plane resonance field, is plotted in fig. 3. The temperature of exchange bias onset, at the "blocking temperature," is lower than or similar to the rotatable anisotropy onset temperature. The exchange bias fields, on the order of 3 mT are much smaller than the 20 to 30 mT field shifts caused by rotatable anisotropy.

IV. DISCUSSION

The behavior of the film with the thinnest layer of native cobalt oxide (sample B) is consistent with the following scenario[9]. Between 150 K and 300 K, antiferromagnetic order is established in the 3 to 4 nm thick layer of native cobalt oxide, although this order fluctuates rapidly in a manner analogous to superparamagnetic grains of ferromagnetic material. For comparison, the ordering temperature in CoO/SiO_2 multilayers in this thickness range is 150 to 200 K[14]. At 100 K, a significant fraction of these grains fluctuate at a rate $\tau \approx \tau_{\rm res}$ producing an increased linewidth through the slow relaxer mechanism[12,13]. Below 100 K, these grains fluctuate at a slower, intermediate rate, $\tau_{\rm exp} > \tau > \tau_{\rm res}$, where they produce a rotatable anisotropy which reduces the resonance field. At temperatures below the capabilities of our FMR apparatus, some grains are stable for the duration of an experiment, $\tau > \tau_{\rm exp}$ and stable exchange bias effects are observable.

The films with thicker CoO follow a different scenario. There are only weak linewidth peaks near the onset of rotatable anisotropy, suggesting that the majority of the larger CoO grains become ordered and couple to the $Ni_{80}Fe_{20}$ film with a relaxation time longer than τ_{res}. At lower temperatures, the isotropic resonance field shift does not appear to freeze out below the exchange bias blocking temperature[9], suggesting that even at low temperatures, the instability of the CoO grains which produce the resonance field shift is driven my rotation of the magnetization.

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TABLES

TABLE I. Summary of FMR effects due to interactions with antiferromagnetic order having relaxation time τ relative to experimental duration $\tau_{\rm exp}$ and excitation period $\tau_{\rm res}$.

Time scale	Resonance field	Linewidth
$\tau > \tau_{\rm exp}$	Exchange bias, Anisotropy	Dispersion Two-magnon
$\tau_{\rm exp} < \tau < \tau_{\rm res}$	Rotatable anisotropy	Dispersion Two-magnon
$\tau \approx \tau_{\rm res}$	Rotatable anisotropy	Slow relaxer
$\tau >> \tau_{\rm res}$	none	none

TABLE II. Structure of the polycrystalline films used in these experiments. Layer thicknesses are in nm.

A	30 $Ni_{80}Fe_{20}$ \ 5 Co \ 5 Ta	Control
B	30 $Ni_{80}Fe_{20}$ \ 5 Co	Native oxide
C	30 $Ni_{80}Fe_{20}$ \ 5 Co \ 30 CoO	Top oxide
D	30 CoO \ 5 Co \ 30 $Ni_{80}Fe_{20}$ \ 5 Ta	Bottom oxide

FIGURES

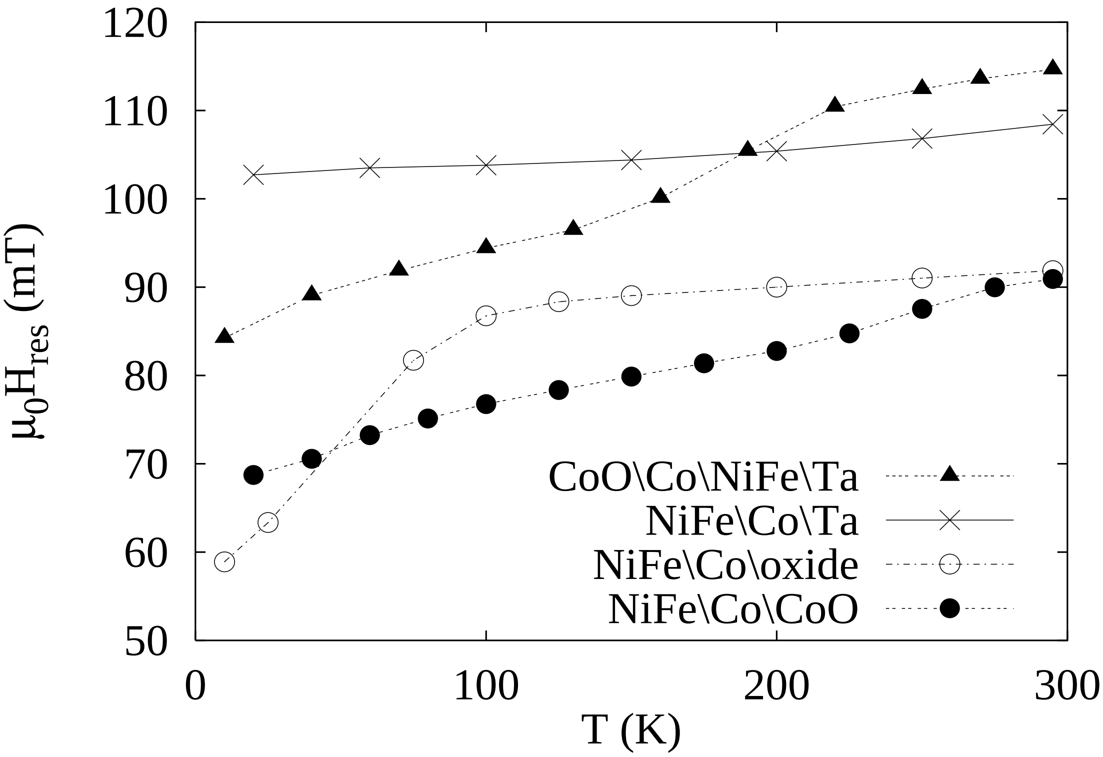

FIG. 1. Average in-plane ferromagnetic resonance fields as a function of temperature. For films coupled to CoO or allowed to oxidize, changes in slope relative to the Ta-capped control sample (NiFe\Co\Ta) indicate the onset of rotatable anisotropy.

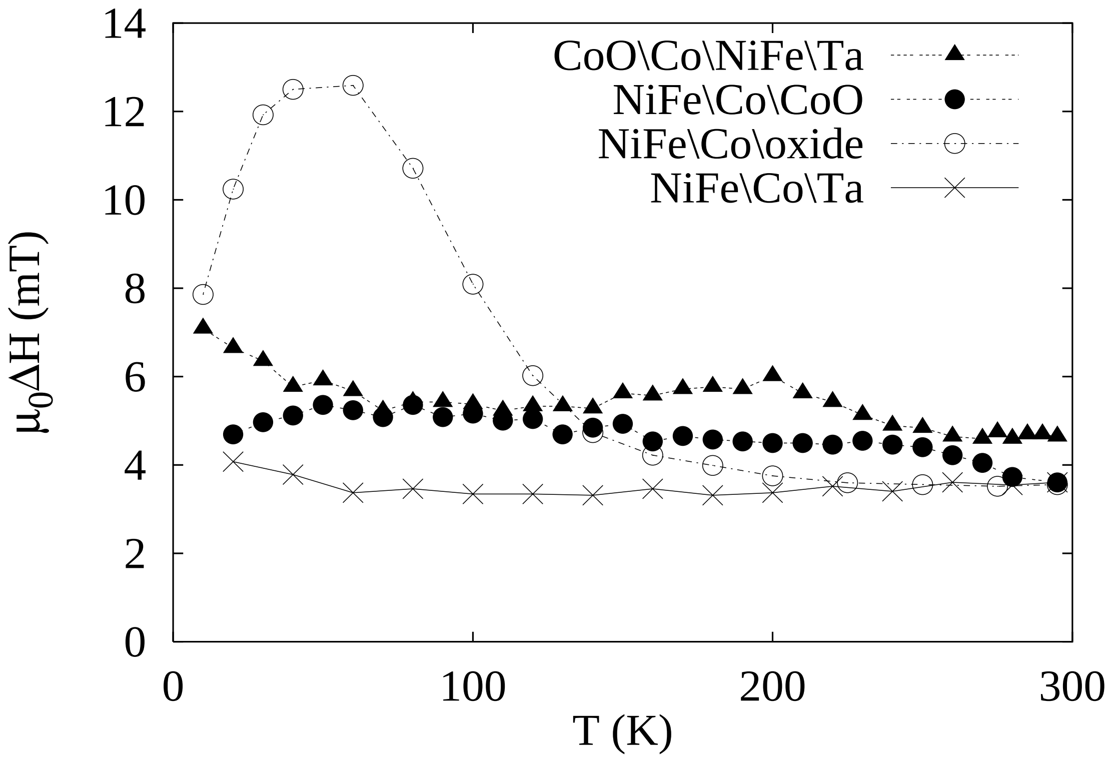

FIG. 2. FMR linewidth as a function of temperature in thin magnetic films. Peaks in the linewidth are characteristic of relaxation processes with rates comparable to the inverse precession time.

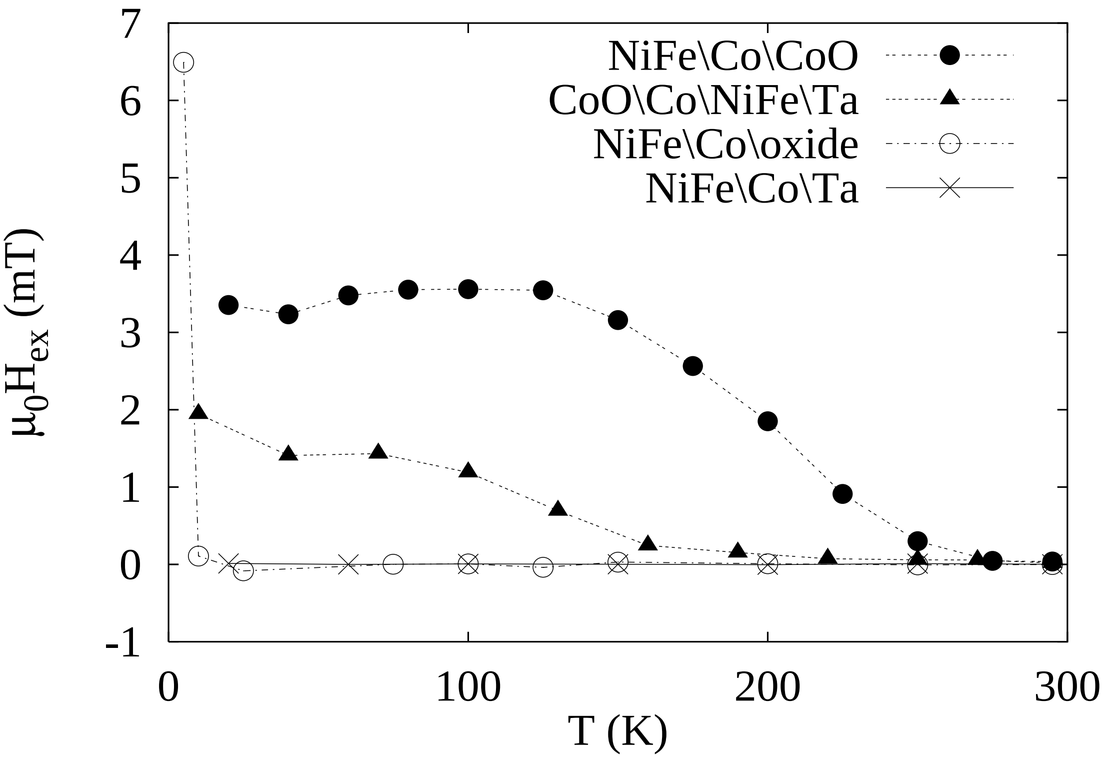

FIG. 3. Unidirectional exchange bias field measured by FMR as a function of temperature in thin magnetic films coupled to CoO. The lowest temperature data point for the NiFe\Co\oxide sample is measured by SQUID magnetometry.