

Frequency-dependent exchange bias in NiFe/NiO filmsJ. Geshev, L. G. Pereira, J. E. Schmidt, and L. C. C. M. Nagamine
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Ferromagnetic (FM) resonance and magnetization curve measurements were performed at room temperature for a polycrystalline $\text{Ni}_{81}\text{Fe}_{19}$ coupled to NiO. It was observed that the shape of the angular variation of the resonance field is frequency dependent, with the curve at 9.65 GHz typical for a strongly exchange-coupled bilayer, while the 34.0 GHz curve is characteristic for relatively weak interactions. Numerical simulations of the resonance field and of the hysteresis loop shift, carried out through the domain wall formation model, as well as the resonance linewidth data indicated that there must be two fractions in the antiferromagnetic part of the interface, with stable and unstable grains. Only the stable grains contribute to the exchange bias. In our sample, whether an interfacial antiferromagnetic grain is stable or not is predominantly determined by the strength of the exchange coupling between this grain and the adjacent FM domain. The stable antiferromagnetic grains, whose contribution is sensed by the resonance experiment, are the *smaller* ones, which are more strongly coupled to the ferromagnet than the larger grains.

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The exchange-bias phenomenon,¹ which results from the interfacial coupling between ferromagnetic (FM) and antiferromagnetic (AF) materials, has been extensively studied in the last decade, motivated by fundamental and practical interests. One of the reasons for the continued interest is the fact that different experimental techniques may yield different exchange anisotropy values.^{2–9} In some cases, this was assigned to the fact that the measurements are performed on different sets of samples.⁴ Another source for the discrepancy could be the reliability of the model used in the experimental data interpretation.⁸ It has also been shown⁹ that the exchange-bias field values derived from magnetization and ferromagnetic resonance (FMR) measurements, must, in general, give different values in the framework of the domain-wall formation (DWF) model which assumes formation of a planar domain wall at the AF side of the interface with the reversal of the FM orientation.^{8,10}

Recent FMR studies on exchange-coupled NiFe films showed the importance of the magnetic state of the AF part of the interface. McMichael *et al.*¹¹ used FMR to study the relaxation behavior of polycrystalline AF layer and found that the stability of the AF order is critical to the existence of the exchange bias. Relaxation related to thermally driven reversal of the AF grains in such systems has also been observed.^{12,13}

The aim of the present work was to apply the phenomenological approach for the DWF model recently developed by our group⁹ on a real exchange coupled system. The paper describes FMR and magnetization measurements of a bilayer of Permalloy (Py) (here, $\text{Ni}_{81}\text{Fe}_{19}$) exchange-coupled to NiO. The striking frequency dependence of the FMR angular variation has been related to the relaxation behavior of the interfacial AF grains.

The sample under investigation was deposited by rf magnetron sputtering onto Si(100) substrate at room temperature in 2.0 mTorr Ar atmosphere with base pressure before depositing better than 5×10^{-8} Torr. The film consists of 300 Å $\text{Ni}_{81}\text{Fe}_{19}$ deposited on 500 Å NiO and capped with 50 Å Cu in order to prevent oxidation in air. External magnetic field has not been applied purposely; however, as a significant stray field from the magnetron may exist in the sputtering chamber during deposition, an in-plane uniaxial anisotropy was induced. The structural characterization, made via conventional x-ray diffractometry performed on a Philips X'Pert MRD machine employing Cu $K\alpha$ radiation, showed that both Py and NiO layers were polycrystalline, with no indications for dominant texture structure.

Room-temperature FMR spectra were taken with a commercial Bruker ESP-300 spectrometer operating at an X-band microwave excitation frequency ω of 9.65 GHz (in-plane and out-of-plane measurements) or 34.0-GHz Q -band frequency (in-plane measurements only), using standard phase-sensitive detection techniques. The sample was mounted on the tip of an external goniometer to allow measurements of the resonance field H_{res} as a function of the in-plane (ϕ_{H}) or out-of-plane (θ_{H}) field angles. Here ϕ_{H} and θ_{H} are equal to zero for magnetic field H applied along the exchange-bias and the normal-to-the-plane directions, respectively.

The symbols in Fig. 1 show the measured in-plane angular variations of H_{res} for both frequencies. As the spectra were taken on the same piece of sample, one should expect these variations to be almost identical in shape if the system's behavior can be explained in the framework of the DWF model.^{9,10} This is a consequence of the fact that although H_{res} depends on ω , small differences (in shape) between resonance field dependences at different frequencies

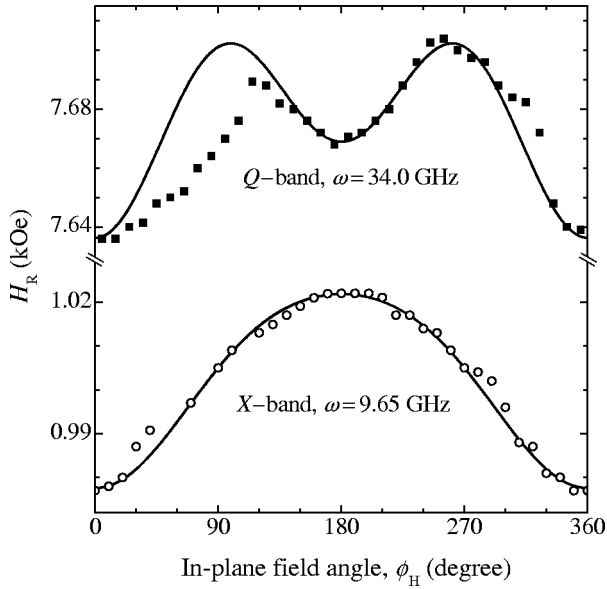


FIG. 1. In-plane resonance fields for a 300 Å Ni₈₁Fe₁₉ layer on 500 Å film of NiO capped by a 50 Å Cu layer at 9.65 and 34.0 GHz. The lines are fits to the experimental data; the parameters used are given in Table I.

should only be visible when the exchange coupling field H_E and AF domain-wall anisotropy field H_W have approximately the same value.⁹ [Here $H_E = J_E / (t_{FM} M_{FM})$ and $H_W = \sigma_W / (t_{FM} M_{FM})$, where σ_W ($\sim \sqrt{K_{AF}}$) is the energy per unit surface of a 90° domain wall at the AF side of the interface, K_{AF} is the AF anisotropy constant, J_E is the interfacial exchange coupling constant (here taken to be positive, which corresponds to ferromagnetic coupling), and t_{FM} and M_{FM} are the FM thickness and spontaneous magnetization, respectively.^{9,10}] One notes, however, that the shapes of the measured angular variations are very different. It seems that the measurement system perceives distinct parts of the sample at different frequencies. Here, we argue that this striking result can be explained taking into consideration the relaxation behavior of the interfacial AF magnetic moments.

The shape of the X-band $H_{res}(\phi_H)$ curve is characteristic for an exchange-coupled bilayer with $H_E \gg H_W$, i.e., strong interactions.⁹ The Q-band curve, however, is typical for the case of relatively weak interactions, when $H_E < H_W$. One notes that there is an asymmetry in the initial part of this plot; a very similar curve has been observed in the H_{res} angular variation for a NiFe/ α -Fe₂O₃ exchange-bias bilayer.¹⁴ There, the authors assigned this asymmetry to hysteresis memory, i.e., training effects, due to successive changes of the uncompensated interfacial AF magnetizations. We also believe that the effect observed here is a related phenomenon.

The solid lines in Fig. 1 are fits to the experimental data through the DWF model and assuming that the FM easy magnetization axis coincides with the AF one; the numerical procedure used is described in our previous papers.^{9,15} The values of the effective fields thus obtained are shown in Table I, where $H_U = (2K_{FM}/M_{FM})$ is the uniaxial anisotropy field of the ferromagnet with anisotropy constant K_{FM} , H_{ra}

TABLE I. Results of the fitting to the resonance field data in Figs. 1 and 2.

ω (GHz)	H_W (Oe)	H_E (Oe)	H_U (Oe)	H_{ra} (Oe)
9.65 (X band)	22	300	4.5	0
34.0 (Q band)	110	16	31	7

[$= 2K_{ra} / (t_{FM} M_{FM})$] is the rotatable anisotropy field, and K_{ra} is the corresponding anisotropy constant. H_{ra} is a field that rotates to be parallel to the equilibrium direction of the FM magnetization and in FMR measurements is responsible for the frequently observed resonance field shift.³ It is produced by the magnetizations of the smaller (unstable) AF grains which can be oriented along the applied field; this anisotropy can change substantially the shape of the magnetization curves and their characteristics.¹⁵

The Py spontaneous magnetization M_s^{Py} was estimated here to be 780 emu/cm³ from the fit to the out-of-plane resonance field variation at 9.65 GHz, shown in Fig. 2. The resulting from both in-plane and out-of-plane fits gyromagnetic ratio $\gamma/2\pi$ is 2.93 GHz/kOe. These values are typical for bulk Py. Stocklein *et al.*¹⁶ have shown that $\gamma/2\pi$ and M_s^{Py} of exchange-coupled Py thicker than 50 Å are practically independent on its thickness. Our values also agree with those obtained in other works on this system.^{3,14,17}

It can be seen that the two sets of estimated anisotropy parameters are quite different. The comparison between the effective fields (Table I) used in the fit of the angular variations in Fig. 1 points out that the 34.0 GHz curve corresponds to a fraction of FM moments weakly coupled to AF grains with rather high domain-wall anisotropy, while the parameters for $\omega = 9.65$ GHz are characteristic for a strongly interacting exchange-bias system with low-anisotropy AF grains. In order to reconcile these data, we adopt the approach used for exchange-coupled bilayers with polycrystal-

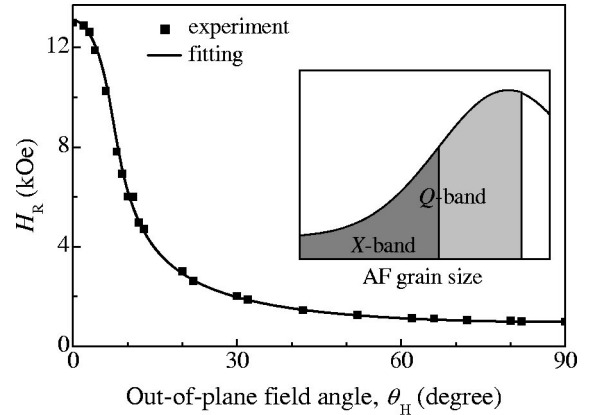


FIG. 2. Out-of-plane resonance fields for the exchange-coupled bilayer measured at 9.65 GHz. The line is a fit to the data, using the parameters given in Table I. The inset shows one possible AF grain size distribution, where the dark gray area corresponds to grains sensed at 9.65 GHz; the light gray area (which includes the dark one) is for $\omega = 34.0$ GHz.

line AF layer,^{18,19} where the basic assumption is that there must be two fractions, of stable and unstable interfacial AF grains. In such a case, one can accept that the FM film is also divided into domains, each coupled to the neighboring AF grain. This is supported by the results of Nolting *et al.*²⁰ which showed that the spin order in the FM domains close to the interface is determined, domain by domain, by the spin directions in the adjacent AF grains. In FMR experiments, whether a magnetic moment is stable or not¹¹ depends on its relaxation time τ (i.e., the time required for the moment to switch from one stable configuration to another), relative to the period of the microwave excitation, τ_{res} (i.e., the precession period of the magnetization at resonance). Only the grains for which $\tau > \tau_{\text{res}}$ contribute to the exchange bias. The AF grains with intermediate relaxation times $\tau \approx \tau_{\text{res}}$ do not contribute to exchange bias but they contribute to the rotatable anisotropy.^{3,11,12,19} Finally, the grains with $\tau < \tau_{\text{res}}$ do not affect significantly H_{res} as they fluctuate rapidly in a manner analogously to superparamagnetic particles.

Based on the above considerations, we argue that the behavior of our sample is consistent with the following scenario. The interfacial AF grains are distributed in size, which also implies distributions of their interfacial exchange coupling and anisotropy.^{18,19,21} These distributions depend on the area of each AF grain, its thickness, and the contact fraction with the ferromagnet. As has been theoretically explained,²² the largest grains have the highest H_{W} values. Nishioka *et al.*²³ observed that K_{AF} increases with the increase of the particle size; as $\sigma_{\text{W}} \sim \sqrt{K_{\text{AF}}}$, the same holds for H_{W} . Thermal activation can also reduce H_{W} when the grain size is decreased.²⁴ In contrast, the FM/AF coupling field H_{E} is lower for larger grains as shown both theoretically and experimentally by Takano *et al.*²⁵; such a dependence has also been used in the work of Stiles and McMichael.¹⁹ Thus, the fitting results indicate that the Q -band experiment senses predominantly the anisotropy of the *largest* grains, and exchange-bias effects characteristic for such grains (weak interactions) are observed. It was also necessary to account for a weak rotatable anisotropy due to the grains with intermediate relaxation time.^{11,15}

This result somehow contradicts the relaxation behavior normally expected for fine (uncoupled) grains in FMR experiments. Taking into consideration that $\tau_{\text{res}} < \tau$ is necessary for a moment to contribute to the anisotropic resonance field and also that an increase of ω implies a decrease of τ_{res} , it is easy to realize that if a particle has an intermediate relaxation time $\tau_{\text{res}}^{\text{Q}} < \tau < \tau_{\text{res}}^{\text{X}}$, then a Q -band experiment will detect this particle's contribution and an X -band experiment will not. In other words, the higher-frequency H_{res} will reflect the anisotropy of this particle; such a particle, however, will not contribute to the lower-frequency H_{res} . Thus, the stability of noninteracting moments in a FMR experiment is determined by the size of the corresponding grain: due to thermal activation, the smaller the particle size, the more unstable its moment and the smaller its τ value.

In our exchange-coupled system, whether an AF magnetic moment is stable or not at a fixed temperature is determined not by its size but mainly by the *strength of the exchange coupling* between this grain and adjacent FM domain. The

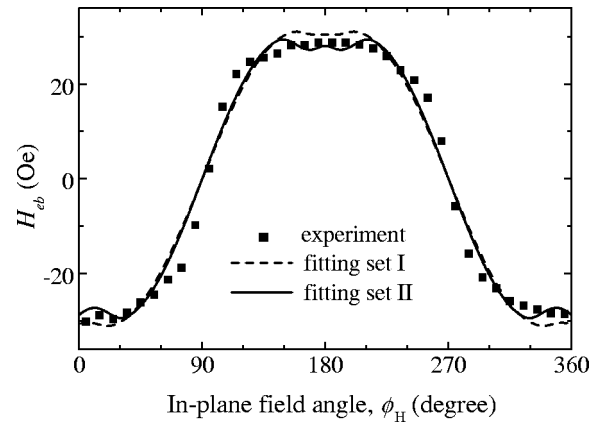


FIG. 3. In-plane angular variation of the hysteresis loop shift H_{eb} . The lines are fits to the experimental data (symbols), where two different parameter sets are used. These parameters are given in Table II.

stronger the coupling, the more stable is the AF grain. The excellent fit to the experimental H_{res} vs ϕ_{H} curve at the lower excitation frequency indicates that the X -band experiment senses the anisotropy of an AF grain fraction where the grains have low domain-wall anisotropy (i.e., small in size) but are strongly coupled to the adjacent FM domains. This fraction is a part of the one which contributes to the Q -band measurement. The results of the FMR (derivative peak-to-peak) linewidth ΔH reinforce this statement as well.

The in-plane ΔH is practically angular independent for our sample, with values of 80 and 220 Oe for the X - and Q -band measurements, respectively. Linear behavior of ΔH with ω is expected in thick metallic films where the relaxation processes are mainly determined by intrinsic damping. Our $\Delta H_{\text{Q band}}/\Delta H_{\text{X band}}$ ratio is smaller than the corresponding frequency ratio. This indicates that the X -band measurement gives information about a more dispersed AF grain system than a Q -band experiment: in polycrystalline materials, the linewidth is additionally increased because the dispersion in the anisotropy parameters causes separate regions of the film (which are stable on the precession time scale) to resonate at different applied fields.¹¹ This leads to a broadening and a loss of symmetry of the absorption curve. Such asymmetrical curves have been observed here at 9.65 GHz; the absorption curves at 34.0 GHz, however, were all symmetrical. One feasible AF grain size distribution is shown in the inset of Fig. 2; the dark gray area corresponds to the particles sensed at 9.65 GHz, and the light gray (which includes the dark one) area is for $\omega = 34.0$ GHz. Note that the dispersion relative to the corresponding mean grain size of the dark gray area is larger than that of the light one (where the majority of the grains are of size close to the mean size), in accordance with the statement above.

These deductions are supported by the magnetization measurements as well. In-plane hysteresis loops were obtained at room temperature by using an alternating gradient force magnetometer. No training effect (i.e., change of the switching fields for the descending and ascending parts of a hysteresis loop with the number of cycles) has been observed. Figure 3 shows the angular variation of the hysteresis

TABLE II. Parameter sets for two representative fittings to the in-plane magnetization data in Fig. 3.

	H_W (Oe)	H_E (Oe)	H_U (Oe)	H_{ra} (Oe)
Set I (small AF grains)	30	200	15	0
Set II (large AF grains)	110	27	22	0

loop field shift H_{eb} for our Py/NiO bilayer. Numerical fits in the framework of the DWF model (the procedure used is described elsewhere^{8,9,15}) to the experimental data for two different parameter sets, named set I and set II, are shown in Table II. As was done for the H_{res} angular variations, one can consider set I (i.e., the set with low H_W and high H_E values) as a parameter set that corresponds to small AF grains and the other as a set of effective fields for a fraction of large AF grains.

It can be seen that neither of the curves fits well the data points. Actually, there is no single parameter set which corresponds to the experimental data. It can be seen, however, that even the simple average of the above-cited $H_{eb}(\phi_H)$ curves represents a better fit. It is clear that for certain AF grain size distribution (its existence was suggested by the FMR data treatment), one could find a weighted average curve which actually fits the experiment. Such a distribution implies also a distribution of H_W and H_E (and, probably, distributed AF easy axis directions), the exact form of which is unknown from the available data. Thus, we show that it is important to consider a distributed in size AF grains. Although without knowledge of the actual grain size and an-

isotropy parameter distribution it was not possible to obtain a better fitting curve for the H_{eb} data, our results indicate that the behavior of the exchange-bias film can be explained well through the DWF model.

In summary, we have observed that the shape of the angular variation of the ferromagnetic resonance field is frequency dependent for our polycrystalline NiFe/NiO film. The curve at 9.65 GHz is characteristic for a strongly exchange-coupled bilayer and the 34.0 GHz curve for the case of relatively weak interactions. Our numerical simulations indicated that the sample's behavior is determined by the stability of the antiferromagnetic order at the interface and that there must coexist stable and unstable antiferromagnetic grains; only the stable grains contribute to the exchange bias. Whether an antiferromagnetic grain is stable or not is predominantly determined by the strength of the exchange coupling between this grain and the adjacent ferromagnetic domain. The stronger the coupling is, the more stable the grain is. In our sample, the smaller antiferromagnetic grains are more strongly coupled to the ferromagnet than the larger ones; as a consequence, the stable grains, which are sensed at resonance, are the smaller antiferromagnetic grains. This scenario is also supported by the resonance linewidth and magnetization curve measurements.

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