Anomalous spin-wave damping in exchange-biased films

S. M. Rezende, A. Azevedo, M. A. Lucena, and F. M. de Aguiar
Departamento de Física, Universidade Federal de Pernambuco, 50670-901 Recife, PE-Brazil
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Ferromagnetic resonance and Brillouin light-scattering techniques have been used to investigate the spin-wave damping in ferromagnetic (FM)/antiferromagnetic bilayers exhibiting exchange bias. The measurements were done in the prototype system NiFe/NiO sputtered on Si(100) as a function of the NiFe film thickness. The linewidths measured with both techniques are more than one order of magnitude larger than in similar NiFe films without exchange bias and increase dramatically with decreasing FM film thickness. The data are consistently explained by a relaxation mechanism based on two-magnon scattering processes due to the local fluctuation of the exchange coupling caused by interface roughness. The local interface energy necessary to account for the measured linewidths is on the same order of the atomic exchange coupling.

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I. INTRODUCTION

A thin ferromagnetic (FM) film can exhibit a hysteresis loop shifted along the field axis when in contact with a suitably prepared antiferromagnetic (AF) material. Discovered more than four decades ago,[1] the so-called exchange bias effect has been found to be of practical use in recent years, notably in high-density magnetic memory elements.[2] The technological applications naturally stimulated extensive experimental studies of FM/AF bilayers, leading to several interesting observations which are not fully understood. Well established is the fact that the characteristic field shift H_{eb} in the hysteresis loop results from a unidirectional anisotropy due to the exchange coupling at the interface between the FM and AF materials.[3] However, different mechanisms have been suggested to explain why the experimentally observed shifts are much smaller than the exchange field expected in atomically flat uncompensated interfaces.[3] In order to settle this and other issues, experimental techniques that probe the microscopic interactions governing the motion of the magnetization are welcome. Ferromagnetic resonance (FMR) and Brillouin light scattering (BLS) are among the most important ones for that purpose.

The linewidths measured in FMR and BLS spectra provide direct information on the spin-wave damping, or relaxation rate, in magnetic materials. Very sensitive to the details of the microscopic interactions, the damping also bears significance for switching applications of thin ferromagnetic films, since they depend on the way the magnetization relaxes towards equilibrium. In this respect, several authors have observed anomalous line broadening in exchange-biased systems.[4–7] The origin of this broadening has been attributed[5,6] to the local variation of the exchange field at the FM-AF interface, giving rise to relaxation via two-magnon scattering processes. However, no systematic linewidth measurements and quantitative interpretation with a theoretical model have been made to establish the proper relaxation mechanism. This paper aims at filling this gap.

Here results are reported of a detailed investigation of the FMR and BLS linewidths in exchange-biased NiFe/NiO bilayers sputtered on Si(100) substrates. The observed damping is more than an order of magnitude larger than in NiFe films deposited on a nonmagnetic substrate. As a function of the FM layer thickness t, the linewidths scale with t^{-2}, which, indeed, is shown to be consistent with a two-magnon scattering mechanism due to interface roughness. Perturbation theory is used to calculate the magnon relaxation rates, extending the linewidth mechanism proposed by Arias and Mills[8] for a single FM film, to magnons with wave number $k=0$ and with $k\neq 0$ in exchange-biased films. The model accounts quantitatively for the data, consistently with a local interface energy on the same order of the atomic exchange coupling.

II. EXPERIMENT

A series of FM/AF bilayer samples of $Ni_{50}Fe_{50}/NiO$ with varying thicknesses was prepared by dc magnetron sputtering on substrates of commercial electronic grade Si(001) wafers. We used a Balzers/Pfeiffer PLS500 system with base pressure 2.0×10^{-7} Torr in the sputter-up configuration, with the substrate at a distance of 9 cm from the target. No external magnetic field was applied during deposition, but a small stray field from the system magnet (8 Oe) was present in the position of the sample. Initially the antiferromagnetic NiO layer was deposited directly onto the heated Si substrate by reactive sputtering in an argon and oxygen atmosphere, using a deposition rate of 1.6 Å/s. The ferromagnetic NiFe layer was deposited on top, in a 3.4×10^{-3} Torr argon atmosphere, with deposition rate of 0.7 Å/s. The purity of the NiFe target is 99.9% and that of the argon gas is 99.999%. The substrate temperature was kept at 130 °C during the deposition of both layers. To induce a NiO(111) texture, high power (200 W) was used for the reactive sputtering. The as-deposited films were analyzed by a Siemens D5000 x-ray diffractometer with Cu $K\alpha$ radiation, indicating well texturized (111) films. The deposition rates were calibrated by measuring the frequencies of volume spin-wave modes in thicker films with BLS and confirmed by measurements in a surface profiler. Six samples were prepared with fixed AF layer thickness of 860 Å and varying FM layer thickness in the range 37–137 Å.

The FMR data were taken with a home-made X-band spectrometer using a YIG-tuned sweep oscillator as the

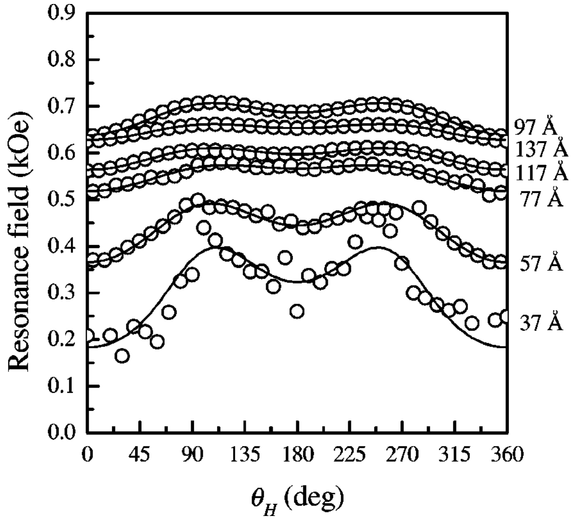

FIG. 1. FMR resonance field versus in-plane angle for the series NiFe($37<t<137$ Å)/NiO(860 Å) and the corresponding fits.

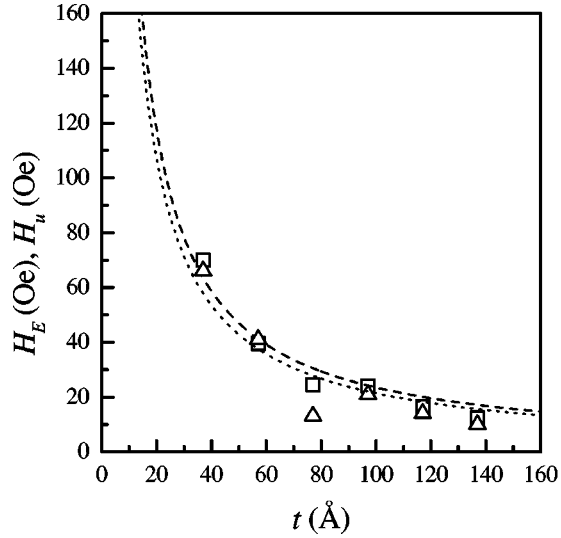

FIG. 2. Fields obtained from the fits to the FMR data. The symbols □ and △ mean, respectively, H_E and H_u, while the lines are fits with $1/t$.

microwave source. Measurements were done at several frequencies by employing various TE_{102} rectangular microwave cavities with Q factor in the range 2500–3000 and an appropriate oscillator-cavity frequency stabilization circuit. The sample was mounted on the tip of an external goniometer and introduced through a hole in the shorted end of the cavity so that it could be rotated in the plane to allow measurements of the in-plane resonance field and linewidth as a function of the angle. The dc magnetic field was provided by a 9-in electromagnet and was modulated with a 1.1 kHz ac component of a few oersteds using a pair of Helmholtz coils. The measurements reported here were obtained at room temperature and at a frequency of 8.53 GHz.

The Brillouin light scattering (BLS) measurements were carried out in the backscattering geometry. The sample was mounted between the poles of an electromagnet with the field H on the film plane. The light source was a single-mode-stabilized argon-ion laser operating at 5145 Å, with power 80 mW, and plane of incidence normal to both the film plane and the field. The scattered radiation was collected by a $f/1.7$ camera lens, frequency analyzed with a Sandercock tandem Fabry-Perot interferometer in a (2×3)-pass configuration, and then directed to a low-noise solid-state photodetector. The spectra were fit to Lorentzian line shapes to determine the frequency shifts and linewidths of the surface spin waves propagating at 90° with respect to the field.

The FMR data were obtained by measuring the field scans of absorption derivative as the sample is rotated in the plane so as to vary the angle θ_H of the applied field. The spectra were fit to the derivative of a Lorentzian line shape in order to determine the resonance field H_R and the linewidth ΔH, characterized by the peak-to-peak field spacing. The angular variation of the resonance field, shown in Fig. 1 for all samples, displays the double peaked shape observed in exchange biased films. Except for the sample with thickness $t=97$ Å, the resonance fields shift downward with decreasing thickness. This field shift has been interpreted with a model that assumes the FM magnetization interacting with independent antiferromagnetic grains in the AF layer.[9] The double peaked structure is a result of the combined effect of the uniaxial anisotropy field H_u along $\theta_H=0$ or π and the AF/FM interfacial exchange field H_E along $\theta_H=0$. Since H_E adds to the external field along $\theta_H=0$ and subtracts along $\theta_H=\pi$, the values of H_R at the two minima differ by $2\,H_E$. The solid curves in Fig. 1 represent fits with theoretical results obtained from the equations of motion for the magnetization with an energy model including Zeeman, demagnetizing, uniaxial anisotropy, and interfacial exchange coupling contributions,[3,5,10] from which we obtain all magnetic parameters. Figure 2 shows the exchange and anisotropy fields obtained from the $H_R\times\theta_H$ data, indicating that they fit well a t^{-1} dependence with sample thickness. The fact that both H_E and H_u increase with decreasing thickness is responsible for preserving the double peaked shape of the $H_R\times\theta_H$ curve in all samples. There are systems in which H_u does not vary much with thickness, and since H_E varies as t^{-1}, the curves become bell shaped as the film thickness decreases.[11] From the data in Fig. 2 we obtain for the macroscopically averaged interfacial exchange energy the value $J_E=H_EMt=0.025\ \mathrm{erg/cm^2}$. This is nearly 50% smaller than the value measured from the hysteresis loop shift in the same samples. The fact that the exchange field H_E measured by FMR is consistently smaller than the loop shift H_{eb} has been attributed to the different natures of the two techniques.[5,9] While FMR is a perturbative technique that probes a reversible response of the system, the hysteresis loop shift expresses an irreversible property. We believe that the difference arises from the fact in the FMR measurement the sample is saturated, while in the hysteresis loop it is not. In the latter case the formation of FM domains alters the macroscopically averaged coupling between the FM magnetization and the complex arrangement of uncompensated AF moments at the interface.[12] However, regardless of which technique measures the correct coupling, both yield interfacial energies more than two orders of magnitude lower than the local atomic exchange coupling.[3,12]

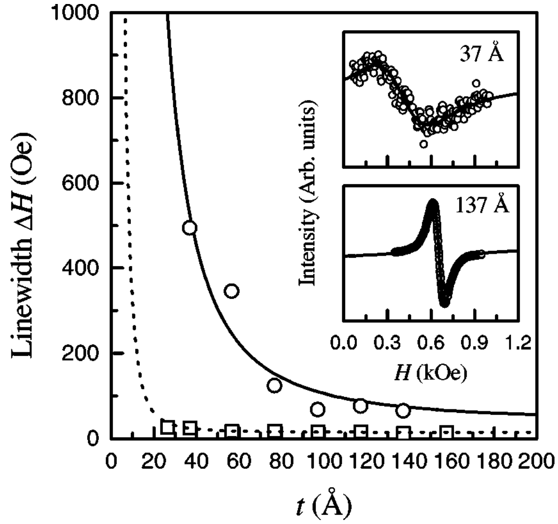

FIG. 3. Thickness dependence of the angular averages of the FMR linewidths measured at 8.53 GHz in NiFe(t)/NiO(860 Å) (circles) and in NiFe(t)/Si(100) (squares). The solid line is a fit with Eq. (8) plus a constant term of 40 Oe, as described in the text. Insets illustrate the fitting of the spectra with derivatives of Lorentzian lines and show the broadening with decreasing FM film thickness.

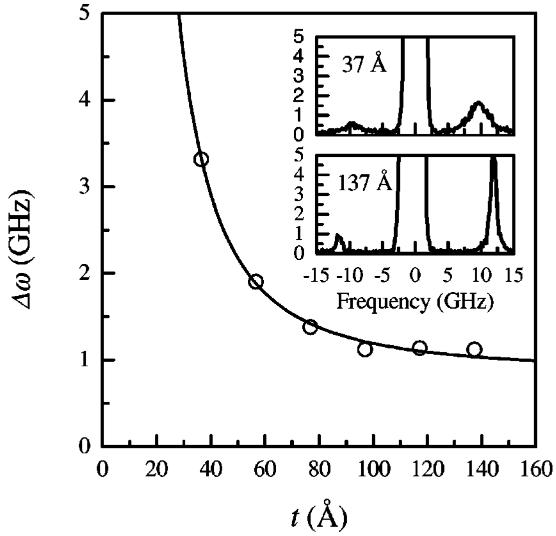

FIG. 4. Thickness dependence of the BLS linewidths in NiFe(t)/NiO(860 Å) and fit with Eq. (9) plus a constant term of 0.69 GHz, as described in the text. Insets illustrate the broadening with decreasing FM film thickness.

The FMR linewidth data obtained at 8.53 GHz are shown in Fig. 3. The circles represent angular averages of the linewidths measured in the NiFe/NiO samples, which fluctuate randomly by up to 20% as the sample is rotated. For comparison we also show by the square symbols the linewidths measured in NiFe films deposited directly on Si(100) substrates and by the dashed line the corresponding fit.[13] In both cases, the linewidths increase with decreasing sample thickness, but the films deposited on the AF layer show a dramatic 20-fold increase in the linewidth compared to those on Si. The insets in Fig. 3 illustrate the line broadening of the exchange-biased film with decreasing thickness. The solid curves are fits to a constant term plus a t^{-2} dependence predicted by the model presented in the next section.

The FMR linewidth is related to the relaxation rate of the magnon with zero wave number k. In order to obtain more information on the magnon relaxation, we have also investigated the behavior of magnons with $k \neq 0$, as measured with BLS. The insets in Fig. 4 show typical frequency shift spectra obtained with 1000 scans, with counting intervals of 1 ms per channel. The BLS lines show the pronounced broadening observed in NiFe/NiO with decreasing film thickness. The measurements were made with several values of the field applied along $\theta_H = 0$. Since the linewidth changes with the magnon frequency, the data in Fig. 4 correspond to the values interpolated to 10 GHz. The solid line is, again, a fit to a t^{-2} dependence predicted by the theory. As we show in the next section, the FMR and BLS linewidth data are consistently explained by a relaxation mechanism based on two-magnon scattering processes due to the local fluctuation of the exchange coupling caused by interface roughness.

III. THEORY FOR THE SPIN-WAVE DAMPING

In this section we present a theoretical model for the spin-wave relaxation in exchange-biased FM films that accounts for the experimental results. The relaxation process considered is two-magnon scattering off the rough FM/AF interface. The roughness gives rise to a large fluctuating field because the FM magnetization interacts alternatively with one or the other AF sublattice via the atomic exchange coupling. The two-magnon process is a well-known relaxation mechanism effective in bulk samples, both insulating[14,15] and metallic,[16] and which has been suggested to be present in exchange-biased thin films.[5] Recently Arias and Mills[8] have studied this process theoretically for a thin FM film without exchange bias. They have shown that the fluctuations in the surface anisotropy arising from surface roughness provides an important extrinsic contribution to the FMR linewidth in ultrathin films due to two-magnon scattering. Their predictions have been verified in NiFe films on Si substrates.[13] Here we extend the two-magnon scattering calculation to exchange-biased films, both for the $k=0$ magnon, appropriate for the FMR linewidth, and for $k \neq 0$ magnons, as probed in BLS experiments. However, instead of using the Green's-functions formalism employed by Arias and Mills, we use a much simpler perturbation-theory calculation.

The scattering of a magnon with wave vector $\vec{k}$ into another magnon $\vec{k}'$ by a process that does not conserve momentum is described by the Hamiltonian[14]

$$H = \sum_{k'} V(kk')(c_k c_{k'}^{\dagger} + c_k^{\dagger} c_{k'}), \tag{1}$$

where $c_k^{\dagger}$ and c_k are the creation and annihilation magnon operators and $V(kk')$ represents the scattering perturbation (from now on the vector sign is dropped for simplicity). In the case of a FM/AF bilayer, the main source of scattering is the fluctuation in the exchange coupling due to the interface roughness. Following the ideas of Arias and Mills[8] we assume a simple model for the roughness. The interface is considered to be atomically flat, with randomly distributed defects in the form of bumps and pits, with the shape of parallelepipeds with large faces of area A_d parallel to the film plane, and height, or depth b. Defects which have b equal to an odd multiple of the AF intersublattice distance result in a local perturbation of the exchange coupling with energy 2 $J_I \cos\theta$ per unit area, where J_I represents the local interfacial exchange energy and θ is the angle between the FM and AF moments. Then it can be shown that the strength of the scattering potential is

$$V(kk')=\frac{\gamma^2\hbar H_I\cos\theta A_d}{2A\omega_k}S_d(k-k')[H_x(k)+H_y(k)], \tag{2}$$

where $\gamma=g\mu_B/\hbar$ is the gyromagnetic ratio, $H_I=J_I/Mt$ is the local exchange coupling field, A is the film area, $S_d(k-k')=\Sigma\exp[-i(k-k')r_i]$ is a structure factor for the defects with positions r_i, and ω_k is the frequency of the k magnon, given by

$$\omega_k=\gamma[H_x(k)H_y(k)]^{1/2}, \tag{3}$$

$$H_x(k)=H\cos(\theta-\theta_H)+2\pi Mkt\sin^2\theta_k+Dk^2+H_E\cos\theta, \tag{4}$$

$$H_y(k)=H\cos(\theta-\theta_H)+4\pi M(1-kt/2)+Dk^2+H_E\cos\theta, \tag{5}$$

where H is the external field applied in the film plane at an angle θ_H with the direction of the unidirectional anisotropy field H_E, θ_k is the angle of the magnon wave vector in the plane, and D is the exchange stiffness of the FM film. A standard transition probability calculation yields for the magnon energy relaxation rate[14]

$$\eta_k=\frac{2\pi}{\hbar}\sum_{k'}|V(kk')|^2\delta(\hbar\omega_k-\hbar\omega_{k'}). \tag{6}$$

This result implies that the incoming magnon k relaxes by scattering into all degenerate modes with wave vector k'. Figure 5 shows the dispersion relations for magnons propagating in the film plane at an angle θ_k with the field, calculated for a NiFe film with $t=50$ Å, $H=0.8$ kOe, $4\pi M=12$ kG, $D=2\times10^{-9}$ Oe cm^2, and neglecting H_E. One can see that for small propagation angles the ω_k curves have negative slope for small k, bending upwards at larger k due to the effect of the exchange interaction. Actually, for $\theta_k<\theta_c$, where $\sin^2\theta_c=H/(H+4\pi M)$, the spin waves have characteristics of volume modes,[8,17] whereas for $\theta_k>\theta_c$ they are surface modes. As a result, the $k=0$ uniform mode detected in FMR experiments can decay only into degenerate volume modes with $\theta_k<\theta_c$, as illustrated in the inset of Fig. 5. On the other hand, magnons with $k>0$, as detected in BLS experiments can decay into volume as well into surface modes, with any value of θ_k. Thus the relaxation rate measured in BLS is expected to be different from the one measured by FMR.

The relaxation rate of the $k=0$ uniform mode can be evaluated as in Arias and Mills.[8] First the summation in k' in Eq. (6) is transformed into an integral in the plane, with

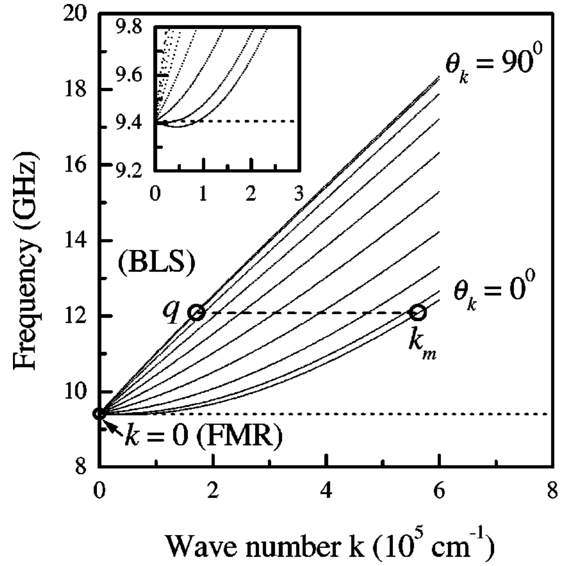

FIG. 5. Dispersion relations for magnons propagating in the film plane calculated with Eqs. (3)–(5) for a NiFe film with $t=50$ Å, $H=0.8$ kOe, $4\pi M=12$ kG, $D=2\times10^{-9}$ Oe cm^2, and neglecting H_E. The angle θ_k between the wave vector and the field varies in steps of 10°. The inset shows the region around $k=0$ expanded to illustrate the modes degenerate with the FMR mode.

$d^2k'=k'dk'd\theta$. Then the delta function in energy is replaced by one in k', so that the integrals are easily calculated. Assuming that the defects in the interface have random positions in the plane and a distribution of areas with mean value $\langle A_d\rangle$, one obtains

$$\eta_0=\frac{\gamma p\langle A_d\rangle\langle\cos^2\theta\rangle}{\pi D}H_I^2, \tag{7}$$

where p is the fraction of the surface covered by defects with height or depth corresponding to an odd multiple of AF sublattice spacings. In evaluating Eq. (7) we have also assumed that the AF film is polycrystalline, so that the angle between the FM and AF moments varies from grain to grain,[9] and that $H, H_E \ll 4\pi M$. The peak-to-peak field linewidth as measured by FMR, related to the relaxation rate by the factor $2/\sqrt{3}(d\omega_0/dH)^{-1}$, becomes

$$\triangle H=\frac{4p\langle A_d\rangle\langle\cos^2\theta\rangle H^{1/2}}{\sqrt{3}\pi D(H+4\pi M)^{1/2}}H_I^2. \tag{8}$$

In order to illustrate the calculation of the relaxation rate of magnons with $k>0$, we indicate in Fig. 5 a point corresponding to a magnon as observed in BLS. In the backscattering configuration with a laser incident at an angle α, the magnon wave number is $q=2k_L\sin\alpha$, where $k_L=1.2\times10^5$ cm^{-1} is the Ar laser wave number. For $\alpha=45°$ this gives $q=1.7\times10^5$ cm^{-1}. In evaluating Eq. (6) for this case, it is more convenient to replace the frequency by the angle θ_k in the delta function, since there are degenerate modes with all values of θ_k. Then the integral in k' runs from q to $k_m=(2\pi Mtq/D)^{1/2}$, the wave number of the degenerate mode with $\theta_k=0$. Considering that $H, H_E, Dq^2 \ll 4\pi M$, one can show that the full magnon linewidth becomes

$$\triangle \omega_q = \frac{\gamma^2 p \langle A_d \rangle \langle \cos^2\theta \rangle 4\pi M \xi}{2\pi D \omega_q} H_I^2, \qquad (9)$$

where ξ is a numerical factor given by

$$\xi = \int_{x_0}^{x_m} dx \left(\frac{x_0}{x} - x \right)^{-1/2} \left(1 + x - \frac{x_0}{x} \right)^{-1/2}, \qquad (10)$$

where $x = k'/k_0$, $x_0 = q/k_0$, $x_m = k_m/k_0$, and $k_0 = (2\pi M t/D)^{1/2}$. Numerical evaluation of Eq. (10) for NiFe gives $\xi = 0.75$ for $t = 20$ Å and $\xi = 0.6$ for 100 Å. So, ξ is of the order of unity and varies slowly with film thickness. Hence the thickness dependence of the BLS linewidth is essentially contained in H_I^2.

Equations (8) and (9) express quantitatively the contribution to the FMR and BLS linewidths arising from the fluctuation in the exchange coupling due to interface roughness treated in a simple model. Note that both linewidths vary with the interface energy as J_I^2 and with film thickness as t^{-2}. In addition, the BLS linewidth is inversely proportional to the magnon frequency, which explains, at least partially, some experimental results.[6,7]

IV. DISCUSSION

The main signature of the extrinsic contribution to the spin-wave damping arising from two-magnon scattering due to surface or interface roughness is the thickness dependence t^{-2} predicted by Eqs. (8) and (9). Indeed, the solid lines in Figs. 3 and 4 are fits to a t^{-2} function plus a constant term, this one due to Gilbert damping. However, in order confirm that the mechanism responsible for the magnon scattering is the fluctuating interfacial exchange, one must consider the magnitude of the quantities involved. First we analyze the FMR linewidth. In the case of NiFe films on a Si substrate, the origin of the scattering potential has been identified to be surface anisotropy.[8,13] The sample with $t = 37$ Å has an extrinsic linewidth of 10 Oe, which is accounted for by a scattering potential provided by a surface anisotropy field of 2.2 kOe.[13] However, the NiFe film with $t = 37$ Å deposited on the NiO AF layer has an extrinsic linewidth of 450 Oe, implying that the scattering potential is almost one order of magnitude larger than in the single film. Only the local interfacial exchange coupling can provide such a large field. Of course, in order to do an accurate calculation of the linewidth one needs to know in detail the structure of the interface. Since we have no means of measuring this, we assume realistic estimates for the geometry of the defects, $p = 0.3$, $\langle A_d \rangle = 20^2$ Å^2, and $\langle \cos^2\theta \rangle = 0.5$. The fit of Eq. (8) to data, shown in Fig. 3, gives for the local interface coupling energy $J_I = 11.6$ erg/cm^2. This value is close to the one expected for the atomic exchange coupling in uncompensated interfaces of NiFe/NiO.[12] Note that this energy corresponds to a field of $H_I = 31.5$ kOe for the sample with $t = 37$ Å, which is more than two orders of magnitude larger than the macroscopic field H_E.

The BLS linewidth data confirms the interpretation for the anomalously large spin-wave damping in NiFe/NiO. Assuming the same parameters for the roughness of the interface, $p = 0.3$, $\langle A_d \rangle = 20^2$ Å^2, $\langle \cos^2\theta \rangle = 0.5$, and $\xi = 0.65$, the fit of the data in Fig. 5 with Eq. (9) yields $J_I = 11.2$ erg/cm^2, in very close agreement with the value obtained from the FMR data.

V. CONCLUSIONS

In conclusion, we have shown that the pronounced linewidth broadening observed in FMR and BLS measurements in FM/AF bilayers is caused by fluctuations of the local exchange coupling energy arising from the roughness of the interface. A model for two-magnon scattering produced by defects on the interface yields simple expressions for the linewidths allowing quantitative calculations in terms of the coupling energy. Application to FMR and BLS measurements in NiFe/NiO yields for the local interface coupling an energy of 10 erg/cm^2. This is on the order of the expected atomic exchange coupling and more than two orders of magnitude larger than the macroscopic unidirectional anisotropy energy measured by several techniques.

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