## Spin waves in exchange-biased Fe/FeF<sub>2</sub>

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The unidirectional anisotropy in exchange biased Fe/FeF<sub>2</sub> bilayers is measured by Brillouin light scattering (BLS) from spin waves in the Fe layer. This technique, where the ferromagnetic layer is always saturated, allows for a straightforward, accurate determination of the exchange bias field. The value obtained from BLS is 25% larger than the one obtained from conventional magnetometry. Moreover, a large broadening of the spin-wave mode due to the presence of the antiferromagnet is observed. From the field dependence of this broadening we conclude that the mode broadening is unidirectional. [S0163-1829(99)00506-8]

Ferromagnetic (FM)/antiferromagnetic (AFM) interfaces are known to exhibit exchange bias (EB) giving rise to a shift of the hysteresis loop along the magnetic-field axis.<sup>1</sup> This effect was first found for oxidized cobalt particles.<sup>2</sup> Due to the technological importance of exchange bias for spin-valve systems<sup>3,4</sup> there is a renewed interest in this phenomenon,<sup>5-10</sup> however its microscopic origin still remains unclear.<sup>11–13</sup> The shift of the hysteresis loop,  $B_{EB}$ , is commonly used to quantify the strength of the exchange bias effect.<sup>1</sup> Here we use Brillouin light scattering (BLS) from thermally excited spin waves in the ferromagnetic layer to investigate exchange bias. In BLS the surface "Damon-Eshbach'' mode acts as a probe of the magnetic anisotropies present in the ferromagnetic layer.<sup>14,15</sup> From the shift of the spin-wave frequency induced by the unidirectional anisotropy due to the exchange bias effect, the exchange bias field  $B_{EB}$  can be easily and accurately determined. We have found that  $B_{EB}$  obtained from BLS is larger than the one obtained from superconducting quantum interference device (SQUID) measurements.

Furthermore, the spin-wave modes in the ferromagnetic layer show a strong enhancement of the linewidth at low temperatures. This broadening is strongly reduced by an external magnetic field, most efficiently in the direction of the exchange bias. This asymmetric reduction indicates a unidirectional character of the linewidth enhancement.

The Fe/FeF<sub>2</sub> films were grown on MgO(100) substrates. The substrates were heated to 450° C for 15 minutes prior to deposition and then cooled to the FeF<sub>2</sub> growth temperature  $(T_S = 200^{\circ} \text{ C})$ , at which 90 nm of FeF<sub>2</sub> were deposited at a rate of 0.2 nm/s by using an electron gun. Following the FeF<sub>2</sub> deposition, a 12-nm Fe layer was electron-beam evaporated, at a rate of 0.1 nm/s at  $T_S = 150^{\circ}$  C. In order to protect the Fe/FeF<sub>2</sub> bilayer, a 3-nm capping layer of Al was electron-beam deposited, at a rate 0.1 nm/s at  $T_S = 150^{\circ}$  C. The base pressure of the chamber was better than  $1 \times 10^{-7}$  Torr and the pressure during deposition was lower than  $2 \times 10^{-6}$  Torr. The thickness of the different layers was controlled by a calibrated quartz oscillator.<sup>16</sup> From x-ray studies the Fe and Al layers were found to be polycrystalline, where the Fe layer exhibits a roughness of about 1 nm.<sup>16</sup> However, the FeF<sub>2</sub> layer grows in the (110) direction, with typical rocking curves of  $\Delta \theta \approx 2^{\circ}$ . In-plane x-ray studies reveal that the FeF<sub>2</sub> layer is twinned, with [001]-[110] twins along the MgO[110] direction.<sup>5</sup> For the following discussion we will refer to the [001]-[110] twins as the [001] direction. The exchange bias system Fe/FeF<sub>2</sub> (FM/AFM) was chosen for its large exchange bias<sup>16</sup> and for its convenient Néel temperature ( $T_N$ =78 K).

The experimental setup of the Brillouin light scattering involves a computer controlled tandem Fabry-Perot interferometer<sup>17,18</sup> combined with a He cryostat that allows optical access to the Fe/FeF<sub>2</sub> sample. The cryostat is set up in an external magnet in order to apply fields up to  $\pm 400$  mT. Hysteresis loop measurements were carried out using a SQUID magnetometer.

To study the Fe layer anisotropies, the sample is exposed at room temperature to an external magnetic field of 200 mT. Figure 1 shows the spin-wave frequency as a function of in-plane angle  $\phi_B$  of the external magnetic field with respect to the [001] direction. The spin-wave frequency oscillates between 21 and 23.5 GHz due to the anisotropy fields present in the Fe film, clearly showing a fourfold symmetry.

These oscillations as a function of the angle  $\phi_B$  are well described by the relation<sup>14</sup>

$$\left(\frac{\omega}{\gamma}\right)^{2} = \left(B\cos(\phi_{B}-\phi) + \mu_{0}M_{s}\left(1-\frac{1}{2}q_{\parallel}d\right) + B_{\alpha}\right)$$
$$\times \left(B\cos(\phi_{B}-\phi) + \frac{\mu_{0}M_{s}}{2}q_{\parallel}d\sin^{2}(\phi-\phi_{q}) + B_{\beta}\right),$$
(1)

where  $\omega$  is the spin-wave frequency,  $\gamma$  the gyromagnetic ratio, *B* the external magnetic field,  $\phi$  the angle between the

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FIG. 1. Spin-wave frequency of Fe/FeF<sub>2</sub> at T=300 K as a function of the in-plane angle  $\phi_B$  between the external field B = 200 mT and the [001] direction for  $q_{\parallel} = 1.7 \times 10^5$  cm<sup>-1</sup>. The solid line is a fit using Eqs. (1) and (2). The resulting anisotropy constants are given in the figure.

direction of *M* and the [001] direction,  $M_s$  the saturation magnetization,  $q_{\parallel}$  the wave vector of the spin wave parallel to the surface, *d* the thickness of the magnetic layer,  $B_{\alpha} = (1/M_s)(\partial^2 F_{ani}/\partial \theta^2)$ ,  $B_{\beta} = (1/M_s)(\partial^2 F_{ani}/\partial \phi^2)$  where  $F_{ani}$  is the free anisotropy energy density,  $\theta$  the out-of-plane angle of the magnetization with respect to the surface normal, and  $\phi_q$  the angle between the wave vector  $q_{\parallel}$  and the [001] direction.

The measured spin-wave frequencies are fitted by Eq. (1) using a free anisotropy energy density<sup>10</sup>

$$F_{ani}(\phi,\theta) = K_p^4 * \sin^2 \phi \cos^2 \phi \sin^4 \theta + K_s^2 * \cos^2 \theta, \quad (2)$$

yielding the fourfold in-plane  $(K_p^4)$  and the twofold out-ofplane  $(K_s^2)$  anisotropy constants as given in Fig. 1. For these fits the parameters  $M_s = 1.707 \times 10^6$  A/m,<sup>19</sup> and  $\gamma = 1.84$  $\times 10^{11}$  T<sup>-1</sup> s<sup>-1</sup> (Ref. 20) were used. During the fitting process the direction of the magnetization  $\phi$  is calculated by minimzing the anisotropy energy and the magnetostatic energy with respect to  $\phi$ . The maximum deviation of the direction of the magnetization  $\phi$  from the direction of the external field  $\phi_B$  was found to be 3°. Therefore, for the sake of simplicity, we assume  $\cos(\phi_B - \phi) = 1$ .

The positions of the spin-wave frequency maxima along the  $\langle 001 \rangle$  directions in Fig. 1 show that at T=300 K there are four equivalent easy axes directions of the magnetization.

To induce exchange bias a field of +400 mT (or -400 mT) is applied along the hard axis direction (i.e.,  $\phi_B$  at 45° to the [001] direction) at 120 K and the sample is field cooled to 50 K (i.e., below  $T_N = 78$  K). To avoid sample heating due to the laser, spin-wave frequencies were measured at different laser powers. Since  $B_{EB}$  is temperature dependent, local heating reduces its magnitude, inducing changes of the observed spin-wave frequency. Above 50 mW laser power slight shifts of the frequency were observed, hence the remaining experiments were all carried out at 25 mW. At T=50 K the scattering intensities are rather weak, so that the spectra were recorded over a time of typically 8–12 h. The accumulation time in each channel [free spectral range (FSR)=2.4, 200 channels/FSR] amounts to about 50–75 s. The maximum number of counts in the peak is



FIG. 2. Spin-wave frequency of Fe/FeF<sub>2</sub> as a function of the applied magnetic field for  $q_{\parallel} = 7.5 \times 10^4 \text{ cm}^{-1}$ , for T = 100 K (crosses), T = 50 K cooling in +400 mT (full squares), and T = 50 K cooling in -400 mT (open circles). *B* is applied at 45° to [001]. The solid lines are a fit using Eq. (4).

usually 100-200 counts above the background noise. The spin-wave frequency at 50 K is depicted in Fig. 2 as a function of applied field B for two different cooling fields of  $\pm 400$  mT (upper and lower curve). Additionally, the field dependence of the spin-wave frequency is plotted at T= 100 K (above  $T_N$ ), where no EB occurs (middle curve). The cooling field orientation clearly shows a large effect on the spin-wave frequency. While a positive cooling field increases the spin-wave frequencies, a negative one reduces it. Other anisotropies influencing the spin waves are the same for both measurements except for the anisotropy induced by the cooling field. Hence, this frequency shift is attributed purely to the exchange bias. Therefore, the spin-wave frequency at T = 100 K which should not show any shift due to EB, falls exactly in the middle between the two 50-K exchange biased curves.

The exchange bias anisotropy can be described by adding the term

$$F_{ani}^{uni}(\phi) = K_p^1 \cos(\phi_{uni} - \phi) * \sin\theta$$
(3)

to Eq. (2), with  $K_p^1$  the first-order unidirectional anisotropy constant and  $\phi_{uni}$  the direction favored by exchange bias. Calculating the anisotropy fields  $(1/M_s)(\partial^2 F_{ani}/\partial \theta^2)$  and  $(1/M_s)(\partial^2 F_{ani}/\partial \phi^2)$  for fixed angles  $\theta = \pi/2$  (i.e., in-plane magnetization) and  $\phi = \phi_{uni}$  (i.e., magnetization along cooling field direction) and taking into account Eqs. (2) and (3) separately, Eq. (1) takes the form

$$\left(\frac{\omega}{\gamma}\right)^{2} = \left(B - \frac{K_{p}^{1}}{M_{s}} + \operatorname{const} + B_{\alpha}(K_{p}^{4}, K_{s}^{2})\right) \\ \times \left(B - \frac{K_{p}^{1}}{M_{s}} + \operatorname{const} + B_{\beta}(K_{p}^{4}, K_{s}^{2})\right), \qquad (4)$$

where  $B_{\alpha}$  and  $B_{\beta}$  are independent of  $K_p^1$ . Thus effectively the unidirectional anisotropy shifts the spin-wave frequency just like an external magnetic field  $B_{EB} = -K_p^1/M_s$ . To ob-



FIG. 3. Hysteresis loop of Fe/FeF<sub>2</sub> at 50 K after field cooling in +400 mT using a SQUID magnetometer. *B* is applied at 45° to [001]. The solid line is a guide to the eye.

tain the strength of the unidirectional anisotropy the spinwave frequencies for positive and negative cooling fields in Fig. 2 are fitted simultaneously using Eq. (4). The fitting parameters are  $K_p^4$ ,  $K_s^2$ , and  $K_p^1$ , where  $K_p^1$  is taken positive (negative) for negative (positive) cooling fields. This procedure enables us to directly and accurately obtain the value of  $K_p^1$  from which  $B_{EB} = -K_p^1/M_s$  is calculated. However,  $B_{EB}$ can also easily be obtained from the separation along the field axis between the fitted frequencies for positive and negative cooling fields in Fig. 2. Hence we obtain  $B_{EB}$ = 39.5±1.2 mT.

The shift of the hysteresis loop obtained from the SQUID measurement in Fig. 3 yields  $B_{EB}$ =30 mT. Thus the exchange bias field measured by magnetometry is about 25% smaller than the one obtained by BLS. Similar differences in the value of  $B_{EB}$  have been observed in Co/CoO between hysteresis loop measurements and techniques where the FM layer was kept in a remanent state.<sup>21,22</sup>

This difference can be explained if higher order terms of the unidirectional anisotropy are taken into account. Such higher-order terms have been suggested in exchange bias<sup>23</sup> and other systems.<sup>24</sup> In the BLS experiment the sample is always saturated, i.e.,  $M_s$  is either at  $\phi = 0^\circ$  or  $\phi = 180^\circ$ , thus all the orders of the unidirectional anisotropy  $[K_p^1 \cos(\phi) + K_p^3 \cos^3(\phi) + \cdots]$  are probed at their extremal values. However, in SQUID measurements the magnetization is turned continuously by the external field. As can be seen in Fig. 3 the shape of the hysteresis curve is not square looplike and reveals a slowly turning magnetization rather than an abrupt change of the magnetic moment. Here the exchange bias field is determined from the mean value of the two intersections of the loop with the field axis at M=0. Hence the magnetization is at  $\phi = 90^{\circ}$  with respect to the magnetic-field direction or, assuming domain formation, it is locally at some arbitrary value ( $0^{\circ} < \phi < 180^{\circ}$ ). Therefore the anisotropy energy is probed at the angle different from  $\phi = 0^{\circ}$  and  $\phi = 180^{\circ}$  where M = 0 and consequently the higher-order anisotropy terms contribute to  $B_{EB}$  not at their maximum values. Therefore the effect of the unidirectional anisotropy can be studied much more precisely from the shift of the spin-wave frequencies.

Furthermore, the measurements carried out at 50 K exhibit a strong increase in the spin-wave mode width. While at room temperature the peak width [full width at half maximum (FWHM)] is about 1.5 GHz (approximately the instrumental resolution), at 50 K the peak width increases up to 5 GHz. Similar mode broadening was observed in BLS measurements of FeNi/FeMn exchange bias layers,<sup>10</sup> but only its directional dependence was studied and no evidence for a unidirectional character of the mode broadening was found. Also, FMR studies of exchange bias systems, which probe spin waves with zero *k* vector, show similar mode width enhancements.<sup>25,26</sup>

The dependence of the mode broadening on the direction of the measuring field with respect to the cooling field is shown in Fig. 4. For parallel alignment of the external and cooling fields [Fig. 4(a)] the mode width decreases as the external field increases. At B = 400 mT the value found at room temperature (dotted line) is reached. For antiparallel alignment [Fig. 4(b)] the decrease of the peak width with the field is much weaker. Note, that well above  $T_N$  there is no broadening (dotted line), thus we attribute it to the presence of the antiferromagnetic FeF<sub>2</sub> layer. One of the reasons for



FIG. 4. Spin-wave mode width (FWHM) of Fe/FeF<sub>2</sub> as a function of the external field, for (a) parallel cooling and applied fields and (b) antiparallel cooling and applied fields. *B* is applied at 45° to [001]. The solid lines are least-squares fittings to straight lines. The dashed lines show the linewidth at T = 300 K, which corresponds to the spectrometer resolution.

this linewidth enhancement could be a spatially fluctuating magnetic structure formed at the interface. This fluctuating field could arise from local magnetization ripples at the interface or inhomogeneous exchange coupling across the interface. It is noteworthy that inhomogeneous FM domains have been observed in some systems even in fields large enough to saturate the sample.<sup>27</sup> This enhancement could also arise from an increased damping of the spin waves due to the presence of the ferromagnetic/antiferromagnetic interface. Such damping could be caused by the excited FM spins "dragging" some AFM spins. The different mechanisms for the observed linewidth enhancement may be distinguished by the resulting shape of the broadened mode. The spatially fluctuating interface would result in a Gaussian line shape while a damping would result in a Lorenzian line shape.<sup>28,29</sup> However, no clear evidence for one or the other line shape is found in our data.

There is a substantial difference in the broadening for parallel or antiparallel alignment of the cooling and the applied fields (see Fig. 4). The reduction of the linewidth for the parallel case that cannot be seen in the antiparallel case clearly proves the unidirectional character of the mode broadening.

This unidirectionality is probably related to the unidirectionality of the coupling. Parallel alignment of the cooling field and the applied field favors a low-energy configuration of the spins, thus broadening is reduced. Antiparallel alignment creates a high-energy spin configuration which leads to frustrated interface spins which should enhance the mode width. Although both linewidth enhancement and exchange bias result from the ferromagnetic-antiferromagnetic coupling they do not imply each other. This can be seen in Fig. 4(a) where at 400 mT no linewidth enhancement is observable for the parallel field alignment, whereas the shift of the spinwave frequency persists at 400 mT (Fig. 2). This means that an external magnetic field is able to reduce the origin of the linewidth broadening but the origin of exchange bias is unaffected.

In conclusion, we have shown the effect of exchange bias on the spin waves in a ferromagnetic layer which is coupled to an antiferromagnet. The frequency shift of the spin waves due to exchange anisotropy enables us to accurately quantify the unidirectional anisotropy and the exchange bias field by Brillouin light scattering measurements. A quantitative difference of 25% between conventional magnetometry and BLS is found and attributed to higher-order terms of the anisotropy. An enhanced spin-wave mode width is found due to the ferromagnetic/antiferromagnetic interface, revealing a spatially fluctuating magnetic state at the interface or increased damping. It is shown that the mode broadening is sensitive to an external magnetic field while the strength of the unidirectional anisotropy shows no significant change. The mode broadening is also shown to be unidirectional.

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