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Dynamical effect in measurement of the exchange-bias field: A consequence of the slow-relaxer mechanism

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We report on the temperature dependence of ferromagnetic resonance (FMR) measurements on NiFe/FeMn bilayer. This includes investigations about the angular distribution of the resonance, H_R, and that of the linewidth, ΔH. By considering the domain-wall formation model, the exchange-bias field, $H_{EB,FMR}$, is derived from the FMR measurements. The disagreements observed at low temperature between these values and the ones obtained from superconducting quantum interference device measurements, $H_{EB,SQUID}$, are discussed. We demonstrate that the negative line shift and the maximum of the linewidth of the FMR line are both perfectly interpreted through the slow-relaxer mechanism. This enabled us to elucidate the intriguing question about the yielding to different values of the exchange-bias anisotropy by both techniques.

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The exchange-bias phenomenon that arises from the coupling between ferromagnetic (F) and antiferromagnetic (AF) layers is known to manifest itself by a shift in the hysteresis loop of the ferromagnet. This shift is termed the exchange-bias field, H_{EB}. Although this effect was discovered more than half a century ago by Meiklejohn and Bean[1] in partially oxidized Co particles, it is still the subject of great interest for both technological and fundamental points of view. In particular, there is some controversy about the fact that reversible and irreversible techniques such as the ferromagnetic resonance and the dc magnetometry may yield different values of H_{EB}. In dc-magnetometry measurements, H_{EB} value is unambiguously extracted from the shift of the hysteresis loop, whereas in ferromagnetic resonance (FMR) experiments it is derived from the resonant field (H_R) angular distribution. This latter is known to depend strongly on the magnetic anisotropies of the system and to, thus, lead to a large variety of shapes. A correct determination of H_{EB} is then only feasible through comparison of the experimental H_R angular distribution with a theoretical model that includes, for example, the uniaxial H_U anisotropy, the direct coupling strength, H_E, and the domain-wall formation H_W anisotropy, as done in the domain-wall formation (DWF) model.[2] At low temperature, the likely occurrence of additional dynamical effects makes more complex H_{EB} determination. Indeed, an isotropic negative field shift is often observed. It is explained by some grains, which have forgotten the initial conditions because of irreversible transitions in the AF. This effect was predicted by Néel[3] and then applied theoretically to the FMR-field shift by Stiles and McMichael through the use of the rotatable anisotropy, H_{RA}.[4,5]

At low temperature, other effects are observed in FMR measurements. Among them, the resonant line is broadened, and the linewidth, ΔH, displays a maximum below the Néel temperature of the AF. To explain such a behavior, the "slow impurities relaxing mechanism" was proposed in[6] and demonstrated in the case of rare-earth (RE)-doped yttrium iron garnets (YIGs). This mechanism is based on the occurrence of two anisotropic levels of the paramagnetic ions split by exchange interaction between the spin of the RE ions and that of the iron ions. In such a mechanism, the impurity energy splitting is modulated by the magnetization motion. Thus, the damping parameter temperature dependence is induced by the delay in the establishment of the thermal equilibrium of the populations of the two levels. The slow-relaxer model has already been applied to NiFe/NiO and NiFe/CoO exchange-biased bilayers.[7–9] The observation of a peak in the thermal dependence of the linewidth by McMichael *et al.*[7] and Lubitz *et al.*[8] drove them to analyze the slow relaxation mechanism as the consequence of the thermal reversal of the antiferromagnetic grains and to consider the antiferromagnetic grains as the impurities coupled to the ferromagnet by exchange. This scenario was rejected by Dubowik *et al.*[9] who considered paramagnetic Ni^{2+} and Fe^{2+} ions at the NiFe/NiO interface as impurities responsible for the slow-relaxer process.

Here, we report on the temperature dependence of FMR resonance on NiFe/FeMn bilayer. This system was chosen because of the observation of the exchange-bias anisotropy at room temperature (RT). The angular distribution of H_R and that of ΔH were both measured. By considering the DWF model, which takes into account the magnetic anisotropies mentioned above, the $H_{EB,FMR}$ field is derived from the FMR measurements. The disagreements observed at low temperature between these values and the ones issued from superconducting quantum interference device (SQUID) measurements are discussed, here, in terms of rotatable anisotropy and slow-relaxer mechanism. Moreover, the investigations about entities responsible for the relaxation process are reported.

The Ta(5 nm)/NiFe(20 nm)/FeMn(20 nm)/Ta(5 nm)/Al(5 nm) sample was grown by dc magnetron sputtering onto a silicon substrate. A 100 Oe magnetic field was applied during the deposition to determine the unidirectional and uniaxial axis. The (111) texture of the NiFe and FeMn was checked by x-ray diffraction in order to get the (111) γ phase of FeMn required to display an exchange-bias coupling. The FMR measurements were made on an ESR Bruker Elexsys 500

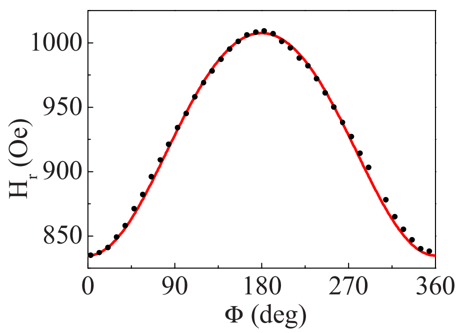

FIG. 1. (Color online) The resonant field value H_r as a function of the azimuthal angle Φ. DWF calculations are represented by full lines.

spectrometer at 9.40 GHz and from RT to 4 K.

Let us consider, at first, the temperature dependence of the FMR resonant field H_R. At every temperature, the resonant field angular distributions were fitted by a cosinlike function with the DWF model, where H_U, H_W, H_E, and H_{RA} are included. As an example, we show in Fig. 1 the result of the fitting at room temperature. The observed shapes were very well reproduced and interpreted by the following case: $H_W \gg H_E \gg H_U$. The values obtained at room temperature were: $H_W=1400$ Oe, $H_E=86$ Oe, and $H_U=10$ Oe. Then, $H_{EB,FMR}$ field corresponds to the amplitude of the sinusoid and is equal to H_E. One should note that the case where $H_E \gg H_W \gg H_U$ could be theoretically used since identical DWF curves are obtained when the values of H_E and H_W are interchanged in these two limit cases. However, according to the recent study by Geshev *et al.*,[10] it is not physically acceptable since it leads to a direct coupling constant, $J_E=1.9$ erg/cm^2, definitely higher than the values reported in the literature. Figure 2 illustrates the temperature dependence of the resonant field, H_R, for two in-plane directions of the magnetic field, namely the unidirectional (Φ=0°) direction and the opposite one (Φ=180°). As indicated, $H_{EB,FMR}x2$ corresponds to the difference between the fields measured in each direction. The negative line shift observed in both directions when decreasing the temperature is explained in the DWF model through the rotatable anisotropy, H_{RA}. This anisotropy assumes an isotropic line shift, *S*, and can be therefore extracted as indicated in Fig. 2. It is worth

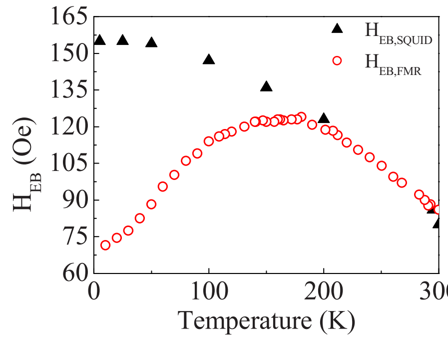

FIG. 3. (Color online) Comparison of the exchange-bias anisotropies measured by SQUID (triangle) and FMR (open circles).

noting that, at 300 K, the magnetization and gyromagnetic factor values are consistent with the typical values observed for permalloy, which leads to $H_{RA}=0$ Oe. Since the origin of H_{RA} is founded on the instability of the antiferromagnet near its Néel temperature,[4] one expect this anisotropy to increase at room temperature instead at low temperature. Such behavior remains unclear at present.

A comparison of the H_{EB} values derived from SQUID experiments with those from FMR measurements is presented on Fig. 3. One should note the increase of $H_{EB,SQUID}$ displayed by SQUID measurements when the temperature is decreasing. This behavior, which has been frequently observed, can be understood as either the temperature dependence of the anisotropies or an increase of the number of grains implicated in the exchange bias at low temperature.[11] At high temperatures, the agreement between both techniques is good in contrast to the low-temperature region, where an unusual decrease of $H_{EB,FMR}$ is visible. This behavior can be explained from the theoretical study by Stiles and McMichael,[4] but only when $H_E \gg H_W$.

To gain more insight into this discrepancy, we investigated the second major effect: the linewidth broadening displayed at low temperature by FMR measurements. Figure 4 illustrates the temperature dependence of the resonance linewidth, ΔH, and shows clearly a broad peak at about 80 K in both directions: Φ=0° and Φ=180°. Such an enhancement

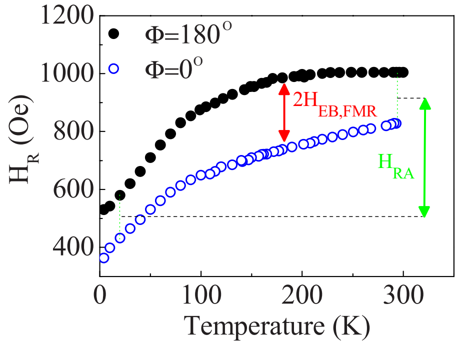

FIG. 2. (Color online) Thermal dependence of the resonant field, H_R, along the unidirectional axis (Φ=0°) and at 180° with respect to this axis.

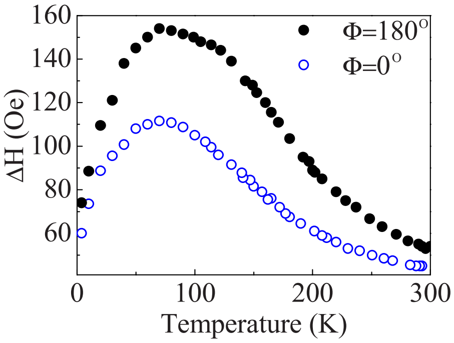

FIG. 4. (Color online) Thermal dependence of the linewidth, ΔH, along the unidirectional axis (Φ=0°) and at 180° with respect to this axis.

of ΔH at low temperature is explained by the slow relaxation impurity mechanism, which is based on the presence of impurities coupled to magnetic ions. The relaxation process leads to the following expression of the FMR linewidth:[12]

$$\Delta H \propto \frac{\omega\tau_n}{1+(\omega\tau_n)^2} \quad (1)$$

where τ_n is the relaxation time of the two energy levels of the impurities, and ω is the microwave frequency. The maximum found for $\omega\tau_n=1$ leads, thus, to the maximum observed for ΔH in our measurements. In addition to its contribution to ΔH, the slow-relaxer mechanism predicts also a line shift, S, of the FMR resonance, which is given by the following relation:

$$\frac{-2S}{\Delta H} = \omega\tau_n \quad (2)$$

From Eq. (2), one can expect first a negative shift, S, and second a constant $S/\Delta H$ ratio. Figure 2 shows clearly this negative shift. Concerning $S/\Delta H$ ratio, the differences observed in Fig. 4 about ΔH measurements at 0 and 180° suggest that S should be also different at these two different angles. This leads to the conclusion that, within the validity of the slow-relaxer process, the line shift, S, is not isotropic in contradiction with the rotatable anisotropy effect in the DWF model. This result may explain the discrepancy observed at low temperature on Fig. 3 since the amplitude of the sinusoid does not reflect anymore the H_{EB} value. To elucidate this intriguing question, we focused on the $2S/\Delta H$ ratio. As suggested by Clarke *et al.* on YIG,[13] the lineshift S was experimentally measured by taking the exchange anisotropy from the SQUID measurements. Figure 5 illustrates the thermal dependence of the $-2S/\Delta H$ ratio at $\phi=0°$ and $\Phi=180$. The good agreement between our experimental results at the two different angles evidences the ability of the slow-relaxer mechanism to describe the maximum of the linewidth as well as the line shift and enables us to explain the disagreement observed, here, between both techniques.

The ratio, $-2S/\Delta H$, is also proportional to the relaxation time of impurities in the slow-relaxer model. Our results demonstrate that the exponential function used by Néel[14] to describe the relaxation of antiferromagnets is unsuited to reproduce the experimental data. Then, considering the thermally driven reversal of AF grains as made by Lubitz *et al.*[8]

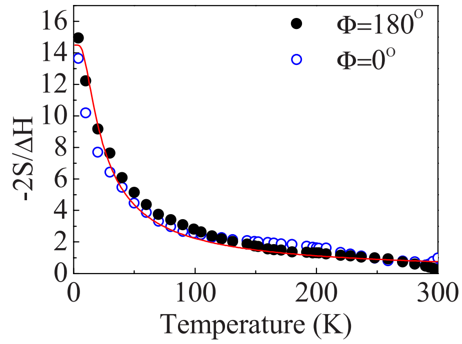

FIG. 5. (Color online) Thermal dependence of the $-2S/\Delta H$ ratio at $\Phi=0°$ and $\Phi=180°$. The full curve corresponds to the relaxing time of the direct process.

may be questionable. The relaxation time is, here, very well described by the $\tau=\tau_0 \tanh(\delta/2T)$ function, which corresponds to the direct-process spin-lattice relaxation time calculated by Orbach[15] and used successfully for the paramagnetic doping of RE-doped YIG in Ref. 13. The parameters were set as follows: $\tau_0=1.5\times10^{-9}$ s and $\delta=31$ K. The impurities responsible for the slow-relaxer process are, therefore, well described in our study by paramagnetic atoms at the interface. One should note that it is in agreement with the results of Dubowik *et al.*[9] obtained in a more restricted temperature region. As evoked by Safonov and Bertram,[16] their appearance could be explained by imperfections in fine grains consisting in either paramagnetic ions such as Mn^{3+}, or surface magnetic atoms that behave like impurities.

In summary, the present study evidenced the validity of the slow-relaxer model for the description of FMR experiments on exchange-biased bilayers. It allowed the identification of the difference observed between the evaluations of the exchange-bias field by reversible and irreversible techniques, directly correlated with a measured non isotropic line shift in FMR experiments at low temperature. Slow relaxation of paramagnetic impurities is responsible of these effects. Indeed, the thermal dependence of the relaxation time highlighted the presence of paramagnetic species at the interfaces due certainly to the presence of fine grains. Such case is not restricted to exchange-biased bilayers and can be found more generally when dealing with interfaces composed with one ferromagnet.

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