# TEMPERATURE DEPENDENCE OF THE FMR LINEWIDTH AND LINE-SHIFT IN EXCHANGE-BIASED NIO/PERMALLOY FILMS

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Abstract: We report a detailed investigation of ferromagnetic resonance (FMR) in the exchange-biased NiFe/NiO ferromagnetic/antiferromagnetic (FM/AFM) bilayers obtained by sputtering. A striking temperature dependence of negative line-shift for both in-plane and perpendicular directions of the applied field is accompanied with a pronounced maximum in temperature dependence of FMR linewidth at ~150 K, well below the Néel temperature of NiO. The same structures without any exchange-bias exhibit no such behavior and can serve as reference samples of the "free" FM thin films. This enables us to distinguish spin dynamics phenomena related solely to FM/AFM interactions. Our results are compared with previous experimental FMR and Brillouin light scattering data on various FM/AFM structures and suggest that spin dynamics (spin wave damping and anomalous line-shift) in the FM/AFM structures can be described in a consistent way by a single mechanism of the so-called slow-relaxation.

## 1. INTRODUCTION

Exchange bias is referred to as pinning of the magnetization of a ferromagnetic (FM) layer in contact with an antiferromagnet (AFM) to a certain reference direction so that a hysteresis loop of the FM/AFM structure exhibits a horizontal shift and an enhanced coercivity. Since the exchange bias was discovered [1], it has been found in many FM/AFM structures [2] and now it is widely applied in spin valve structures for giant magnetoresistive heads in magnetic recording technology.

A remarkable fact is that the problem of interaction between FM and AFM at the interface has not been yet solved [2] and only very recently it has been clearly shown [3] by using X-ray magnetic circular dichroism that a tiny fraction of interfacial spins is responsible for the effect. With the rapid progress of nanotechnology and of high-density recording [4] there is a great interest in studying magnetization dynamics of the exchange-biased structures. Nevertheless, some essential issues including magnetization dynamics [5] are still not clear. In this context ferromagnetic resonance (FMR) [6, 7] and the Brillouin high scattering (BLS) [8-10] proved to be useful method for investigation of exchange bias.

The ongoing results include investigations of exchange anisotropy within the framework of phenomenological model [6] and other aspects concerning magnetization dynamics in the exchange biased structures. Specifically, it has been found that:

(i) FMR linewidth of the exchanged biased FM films shows a pronounced maximum at temperatures well below the Néel temperatures of AFM [11, 12],

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- (ii) there is an anomalous line-shift [8,13], which was found to be consistent with the existence of rotatable anisotropy,
- (iii) there is a large additional FMR linewidth, which follows the angular dependence of the number of nearly degenerate modes [7] in agreement with the predictions of a two magnon scattering model,
- (iv) there is a characteristic  $1/t^2$  dependence FMR linewidth on the thickness t of FM layer, which may be also interpreted in terms of two-magnon scattering [9].

In this paper we present the results of temperature FMR measurements of the magnetization dynamics of the exchange-biased NiO/NiFe films. It is shown that in the structures without exchange bias the magnetization dynamics is nearly temperature independent. Former experimental data on the temperature dependence of magnetization dynamics in exchange-biased films have been analysed in the framework of the slow relaxation mechanism [11] but have not been addressed to the influence of the interaction of a ferromagnetic "reservoir" to a system of a slow relaxing "impurities", which we believe are present at the FM/AFM interface. In this case both an anomalous spin-wave damping and an anomalous line-shift is expected, like in magnetic garnets containing magnetic impurities of transition and rare-earth ions [14].

# 2. EXPERIMENTAL

Two sets of NiO (50 nm)/NiFe (5 nm)/Ta (2 nm) and NiO (30 nm)/NiFe (4 nm)/Cu (40 nm) were prepared on glass substrates using rf diode sputtering. The NiO layers were deposited by a reactive sputtering from a Ni target in an Ar/O<sub>2</sub> gas mixture of 100 sccm Ar and 0.5 sccm O<sub>2</sub>. The resulting exchange bias field was of about 150 Oe. Structure characterisation, made by a cross section transmission electron microscopy, showed that the structures are polycrystalline with NiO crystallites of about 10 nm. External magnetic field of 40 Oe was applied to establish a unidirectional anisotropy. FMR spectra were taken with a home-made spectrometer at an X-band microwave frequency  $\omega/2\pi$  of 9.08 kHz with the field applied in the film plane and perpendicular to the film plane (in-plane and out-of-plane orientations). The FMR measurements were done in a temperature range from 78 K up to 500 K controlled by a nitrogen gas flow system. Some samples were annealed at 540 K and cooled down with no magnetic field. For the Cu capped structure, such a procedure resulted in disappearing of the exchange-bias. In the Ta capped structures annealing led to a slight modification of the exchange bias [15].

# 3. RESULTS

A striking behaviour of the FMR linewidth  $\Delta H$  in thin films coupled to NiO is its anomalous several times increase for the magnetization vector lying in the film plane [7]. To check if the same behaviour takes place in our NiO/NiFe structures, we performed the same measurements of the resonance field  $H_r$  and the resonance linewidth  $\Delta H$  as a function of the out-of-plane  $\Theta_H$  field angle. Using the same calculation procedure as described in Ref. [7, 16] the frequency-swept linewidth values  $\Delta \omega / \gamma$  were calculated from  $H_r$  and  $\Delta H$  values measured at various angles of  $\Theta_H$ . The resultant  $\Delta \omega / \gamma$  versus  $\Theta_H$  plots are shown in Fig. 1 for an exchange-biased NiO/NiFe film (continuous line) and for the same structure without the exchange bias (dashed line). It is seen that there is a smooth increase in  $\Delta \omega / \gamma$  in the exchange biased NiO/NiFe films as the field is

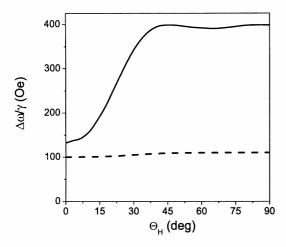
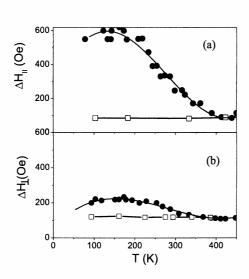


Fig. 1. Frequency-swept linewidth  $\Delta\omega/\gamma$  as a function of the angle  $\Theta_H$  between sample normal and the applied field for NiO/NiFe structure with (continuous line) and without (dashed line) exchange bias

rotated from the film normal (0°) to in-plane (90°). For the same films with no exchange bias  $\Delta\omega/\gamma$  practically does not depend on  $\Theta_H$ . Therefore, we can confidently conclude that the additional linewidth in the exchange biased NiO/NiFe film is solely due to coupling of ferromagnet spins to the antiferromagnetic NiO.



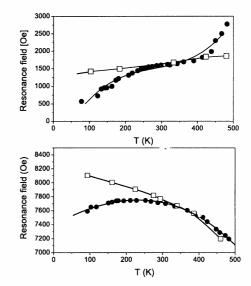


Fig. 2. FMR linewidth  $\Delta H$  as a function of temperature for the NiO/NiFe structures measured in the in-plane (a) and the out-of-plane (b) configurations. Full and open symbols indicate the data for the biased and unbiased structure, respectively

Fig. 3. Resonance field of the NiO/NiFe structures taken in the in-plane (upper panel) and the out-of plane (bottom panel) configurations. Full and open symbols represent the data for the biased and unbiased structure, respectively

The linewidth data for NiO/NiFe structures with and without the exchange bias are shown in Figure 2. Figure 2a displays the temperature dependence of  $\Delta H_{\parallel}$  for magnetization oriented in

the film plane. There is a clearly seen broad maximum in the linewidth at ~150 K. Similar measurements for the film without exchange bias show no variation in  $\Delta H$  with temperature. Figure 2b displays  $\Delta H_{\perp}$  vs. T for the magnetization oriented out-of the plane. Again, a shallow maximum is seen for the exchange biased structure while practically no variation of the linewidth is seen in for the film with no bias. We define the difference between  $\Delta H^b$  in the presence of the exchange bias and  $\Delta H^{nb}$  for the sample without bias:  $\delta(\Delta H) = \Delta H^b - \Delta H^{nb}$  as solely due to the presence of the exchange bias. A remarkable feature is that the maximum value of  $\delta(\Delta H_{\parallel}) \approx 500$  Oe at 150 K is several times larger higher than  $\delta(\Delta H_{\parallel}) \approx 100$  Oe.

The anomalous temperature behaviour of the FMR linewidth for the exchange-biased films is associated with a substantial resonance line-shift of the NiO/NiFe exchange biased structure with respect to that of the unbiased structure. It is seen in Fig. 3 that in the unbiased NiO/NiFe structure the resonance fields depend on temperature according to the "normal" behaviour due to a temperature dependence of the Permalloy magnetization. For the out-of-plane orientation (Fig. 3 bottom panel) there is a monotonic increase in  $H_r$  while for the in-plane configuration (upper panel) there is a slight decrease in  $H_r$  with decreasing temperature. In contrast, for the exchange-biased structure there is an anomalous decrease in the resonance field  $H_r$  at T < 200 K both for the in-plane and out-of-plane configurations. We define the line-shift due to the presence of exchange bias as  $\delta H_r = H_r^{\ \ b} - \Delta H_r^{\ \ nb}$ , where  $H_r^{\ \ b}$  and  $H_r^{\ \ nb}$  refer to as the resonance field of biased and unbiased structure, respectively. It is seen that  $\delta H_r$  (in-plane)  $\approx \delta H_r$  (out-of-plane)  $\approx 600-800$  Oe at 100K in contrast to the results presented in Ref. [13] where  $\delta H_r$  (out-of-plane)  $\approx 2 \ \delta H_r$  (in-plane).

# 4. DISCUSSION

Ferromagnetic resonance has been proved very useful for investigation of dissipation process in ferromagnets. The linewidth measured in FMR spectra provide direct information on spin-wave damping or relaxation rate and much work have been done to explain various possible paths from a spin system to the lattice [17].

The effects reported in this paper – anomalous broadening of the linewidth and the negative line-shift – seem to be a quite general feature in the exchange biased structures. [5, 11, 12]. However, the interpretation of the anomalous spin-wave damping in exchange-biased films reveals some ambiguity. On one hand, a strong dependence of the damping on the thickness of FMR layers was explained by a relaxation mechanism based on two-magnon scattering due to the local fluctuation of the exchange field. On the other hand, this approach (see Eq. (8) in Ref. [9]) does not explain a substantial temperature dependence of the damping, which has been observed both in FMR [11] and BLS [10] measurements.

The main results of our temperature measurements, the anomalous linewidth  $\delta(\Delta H)$  and the line-shift  $\delta H_r$ , are shown in Fig. 4 for the in-plane (solid symbols) and for the out-of-plane (open symbols) orientation, respectively. As was suggested for the first time by McMichael et al. [12], the enhanced relaxation of ferromagnetic moments by a thermally activated process may be regarded as a source of behaviour of  $\Delta H$  with the maximum well below  $T_N = 520$  K for NiO. Here however, we show for the first time that the anomalous broadening  $\delta(\Delta H)$  is accompanied with the anomalous negative line-shift  $\delta H_r$ , both for the magnetization lying the in-plane and out-of-the plane (Fig. 4 bottom panel).

Our results seem to be consistent with relaxation related to thermally driven impurity relaxation process which is known for nearly five decades as so-called "slow relaxing ion" mechanism [18]. This mechanism involves modulation of the impurity levels in the vicinity of thermal equilibrium by small magnetization oscillations. Since the energy modulation is absorbed by the lattice in externally fast rate in some cases ( rare-earth ions in YIG) both  $\delta(\Delta H)$  and  $\delta H_r$  may be as large as several hundreds Oe [14].

In a simplified form the theory (see Ref. [14] for details) predicts:

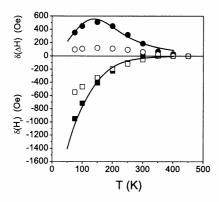
$$\delta(\Delta H) \propto \frac{c_{\text{imp}}}{T} \frac{\omega \tau}{1 + (\omega \tau)^2}$$
 (1)

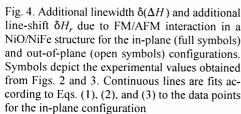
$$\delta H_r \propto \frac{c_{\text{imp}}}{2T} \frac{(\omega \tau)^2}{1 + (\omega \tau)^2}$$
 (2)

and thus

$$\frac{2\,\delta H_r}{\delta(\Delta H)} = -\,\omega\,\tau\,,\tag{3}$$

where  $\tau$  is a temperature dependent relaxation time of the populations of the ground state of impurity ions of concentration  $c_{\text{imp}}$ . By plotting  $\omega \tau$  versus T (Fig. 5) we can deduce the dependence of relaxation time on T. The experimental data is best fitted with  $\tau \propto T^{-2}$ . These relaxation times deduced from the experiment were used to calculate  $\delta(\Delta H)$  and  $\delta H_r$  and the results of calculations (shown as the continuous lines in Fig. 4) are in good agreement with the measured values for the in-plane configuration.





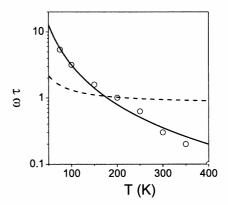


Fig. 5.  $\omega \tau$  versus temperature T. Symbols depict the experimental values obtained from Eq. (3). Continuous line is a dependence  $\omega \tau \propto T^{-2}$ . Dashed line shows a dependence  $\omega \tau \propto \exp(\Delta E/k_BT)$ 

A similar behaviour of the FMR linewidth [11] was interpreted in terms of the slow relaxation mechanism induced by the thermal reversals of small AFM grains. The relaxation time for such small grains is known to depend on temperature as  $\tau \propto \exp(\Delta E/k_B T)$  [19], where  $\Delta E$  is a height of energy barrier and  $k_B$  is the Boltzmann constant. However, it is clearly seen from Fig. 5 that this is not the case and  $\tau \propto T^{-2}$  much better describes the experimental situation than the Néel relaxation time with  $\Delta E \approx 0.7 \times 10^{-14}$  erg  $\approx 35$  cm<sup>-1</sup>. Therefore, we can argue that the relaxation mechanism in the NiO/NiFe structures is rather related to other entities than the small NiO grains. These might be, for example, paramagnetic Ni ions in various valence states which are located at the interface. In this case, due to repopulation of the energy levels upon magnetization precession, the energy of the ferromagnetic reservoir (i.e., NiFe) can be effectively transferred to the lattice (NiO).

In summary, we an have observed temperature dependent magnetization dynamics in thin NiFe layers in contact with an antiferromagnetic NiO. The characteristic increase in the spin-wave damping and the negative dynamic line-shift are consistently interpreted in terms of the slow relaxation mechanism via interaction of the magnetization precession with the "impurity" spins. The nature of the slow relaxing impurities is yet not clear, but we are arguing from  $T^2$  dependence of relaxation times that they might be the Ni ions.

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