

Temperature dependence of ferromagnetic resonance in permalloy/NiO exchange-biased films

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Received 2 September 2004 / Received in final form 9 March 2005

Published online 28 June 2005 – © EDP Sciences, Società Italiana di Fisica, Springer-Verlag 2005

Abstract. The temperature dependencies of the ferromagnetic resonance (FMR) linewidth and the resonance field-shift have been investigated for NiO/NiFe exchange-biased bilayers from 78 K to 450 K. A broad maximum in the linewidth of 500 Oe, solely due to the exchange-bias, is observed at ≈ 150 K when the magnetic field is applied along the film plane. When the magnetic field is applied perpendicular to the film plane, the maximum in the linewidth is less pronounced and amounts to 100 Oe at the same temperature. Such a behavior of the FMR linewidth is accompanied with a monotonic increase in the negative resonance field-shift with decreasing temperature. Our results are compared with the previous experimental FMR and Brillouin light scattering data for various ferromagnetic/antiferromagnetic (FM/AF) structures, and suggest that spin dynamics (spin-wave damping and anomalous resonance field-shift) in the FM/AF structures can be described in a consistent way by a single mechanism of the so-called slow-relaxation.

PACS. 75.70.-i Magnetic films and multilayers – 76.50.+g Ferromagnetic, antiferromagnetic, and ferrimagnetic resonances; spin-wave resonance – 75.30.Et Exchange and superexchange interactions

1 Introduction

With the rapid progress of nanotechnology and of high-density recording [1] there is a great interest in studying magnetization dynamics in magnetic nanostructures including the exchange-biased structures. Exchange-bias (EB) gives rise to a horizontal shift of a hysteresis loop of ferromagnetic (FM) film in contact with antiferromagnet (AFM) due to interaction at FM/AFM interface. This effect is extensively applied in magnetoresistive devices, because EB pins a soft ferromagnetic layer used as a reference layer in spin-valves [2]. However, some essential issues, including magnetization dynamics [3] are still not clear. In this paper we are interested in a problem of magnetization dynamics of EB structures: in an extended Introduction we present state-of-the-art of magnetization dynamics in these structures. The rest of the paper deals with our experimental results concerning enhanced ferromagnetic resonance (FMR) damping in NiO/NiFe EB bilayers and an alternative model in the virtue of slow-relaxing impurity model proposed by Clogston [4]. Recently a detailed evaluation of relaxation rates for slow-relaxing impurity process was given in reference [5].

Both FMR [6–14] and Brillouin light scattering (BLS) [13,15–18] have been extensively applied to study magnetization dynamics in EB systems. From these measurements one can gather a number of fairly well documented facts concerning spin-wave relaxation in EB structures. Specifically, it has been found that: (i) FMR and BLS lines are significantly broadened [6,8,14,15] and the linewidth often shows a pronounced maximum at temperatures well below the Néel temperatures of AFM [8,11,12,17], (ii) there is an anomalous FMR field-shift [7,9] and a significant shift in BLS frequency [15–17], (iii) the additional broadening of the FMR line in EB films was found to depend strongly on orientation of the magnetization [8], and (iv) there is a characteristic $1/t^2$ dependence of the FMR and the BLS linewidth on the thickness t of FM layer [13,18]. Most of FMR experiments on EB structures (see Ref. [19] and references therein) suggest, that magnetization of FM layer is not homogeneous throughout its thickness. Alternatively, the enhanced FMR linewidth can be understood in terms of a mosaic of antiferromagnetic domains and, hence, fluctuations of a local field each domain exerts on the ferromagnetic layer [6]. The linewidth is assumed to be equal to the root-mean-square fluctuations of the local field. Later this approach was extended to a more realistic [7,13] one

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that assumes relaxation mechanism based on two-magnon scattering in thin FM films caused by interface roughness [20]. It was shown [13] that the FMR and BLS linewidths vary with film thickness as $1/t^2$. A microscopic mean-field-theory for spin-waves in EB structures was proposed by Stamps and Camley [21]. In this model spin motion of FM layer with minimal anisotropy is exchange coupled to an AFM layer with a significant anisotropy. Moreover, the frequency of spin-wave mode of FM film can be substantially changed due to the dynamic coupling. The above approaches assume either a static distribution of local field due to the presence of AFM domains, static local variations of the exchange field that gives rise to relaxation via two-magnon scattering or static anisotropies in exchange coupled layered system. Hence, they can be regarded as the “static” approaches. It is worth noticing that the static approaches do not give any explicit temperature dependence of the linewidth (see Eq. (11) in Ref. [6] or Eq. (8) in Ref. [13]) or assume temperature dependence of frequency of spin-wave modes due to temperature dependence of magnetic moments [21].

To explain temperature dependence of the FMR (BLS) linewidth and the field (frequency)-shift another approach was proposed that can be seen as an extension of Fulcomer and Charap model [22] of thermal fluctuation aftereffect in EB systems. This approach [10,23] can be regarded as a “dynamic” approach since it assumes that the enhanced linewidth in a temperature region below the Néel temperature results from thermal instabilities of the AFM layer which is believed to be composed of fine antiferromagnetic grains with superparamagnetic behavior. Thus, spin-dynamics of the FM film exchange-biased to the AFM layer may be described in terms of a slow-relaxation impurity model [4,5,24] in which relaxing entities are thermally excited AFM grains rather than impurity atoms (e.g., rare-earth (RE) ions in YIG). Assuming there are stable and unstable fractions of the AFM grains with superparamagnetic relaxation times τ short or long in comparison to the two time scales of the FMR experiment (the measurement time $\tau_{\text{exp}} \approx 10^3$ s and the precession period $\tau_{\text{res}} \approx 10^{-10}$ s), either line broadening or isotropic resonance field shift is expected.

Our data has been analyzed in terms of the interaction between a ferromagnetic “reservoir” and a system of slowly relaxing “impurities”, which we believe from our experiment and from a discussion of other experimental results, is present at the FM/AFM interface. We argue that both an anomalous spin-wave damping and an anomalous field-shift are similar to those observed in magnetic garnets containing magnetic impurities of transition and RE ions [24].

2 Experimental details

Two sets of NiO (50 nm)/NiFe (5 nm)/Ta (2 nm) and NiO (30 nm)/NiFe (4 nm)/Cu (40 nm) thin film samples 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/O₂ gas mixture of 100 sccm

Ar and 0.5 sccm O₂. External magnetic field of 40 Oe was applied to establish a unidirectional anisotropy. The resulting exchange-bias field (determined from hysteresis loops) was of about 150 Oe and was higher than the unidirectional anisotropy field $H_{\text{ex}} = 80$ Oe determined from the in-plane resonance fields using a fitting procedure similar to that applied in reference [7]. The fit also yielded a uniaxial anisotropy field of 10 Oe. Structure characterization, made by a cross section transmission electron microscopy, showed that the structures are polycrystalline with NiO crystallites of about 10 nm. FMR spectra were taken with a home-made spectrometer at an X-band microwave frequency $\omega/2\pi$ of 9.08 GHz 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 carried out in a temperature range from 78 K up to 450 K controlled by a nitrogen gas flow system. The temperature dependence of the in-plane resonance fields and linewidths were measured with the magnetic field applied in a direction in between the directions opposite and along H_{ex} . Some samples were annealed at 540 K and cooled down without presence of magnetic field. For the Cu capped structure, such a procedure resulted in disappearing of the exchange-bias, probably due to fast grain boundary diffusion of Cu to the NiO/NiFe interface. In the Ta capped structures annealing led only to a slight modification of the exchange-bias [25].

3 Results

A striking behavior of the FMR linewidth ΔH (for the magnetization vector lying in the film plane) in thin FM films coupled to NiO is its anomalous several times increase [8]. To check if the same behavior takes place in our NiO/NiFe structures, we performed the same measurements of the resonance field H_r and the resonance linewidth ΔH as a function of the out-of-plane Θ_H field angle. Using the same calculation procedure as described in references [10,26], the frequency-swept linewidth values $\Delta\omega/\gamma$ were calculated from H_r and ΔH values measured at various angles of Θ_H . The resultant $\Delta\omega/\gamma$ versus Θ_H plots are shown in Figure 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 rotated from the film normal (0°) to the in-plane (90°) direction. For the same films with no exchange-bias $\Delta\omega/\gamma$ practically does not depend on Θ_H . Therefore, we can confidently conclude that the additional linewidth in the exchange-biased NiO/NiFe film is solely due to coupling of ferromagnetic spins to the antiferromagnetic NiO. The field-swept linewidth measured at temperatures from 78 K to 450 K for NiO/NiFe structures with and without the exchange-bias are shown in Figure 2. Figure 2a displays the temperature dependence of ΔH_{\parallel} for magnetization oriented in the film plane ($\Theta_H = 90^\circ$). A clearly seen broad maximum in the linewidth is placed at ~ 150 K. For the film without exchange-bias there is almost no variation in ΔH

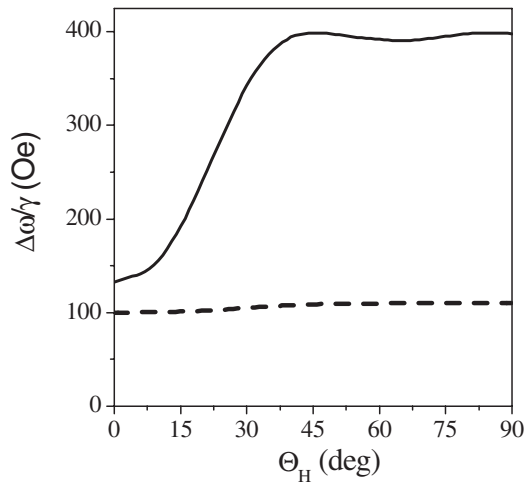


Fig. 1. Frequency-swept linewidth $\Delta\omega/\gamma$ as a function of the angle Θ_H between sample normal and the applied field for NiO/NiFe structure with (continuous line) and without (dashed line) exchange-bias.

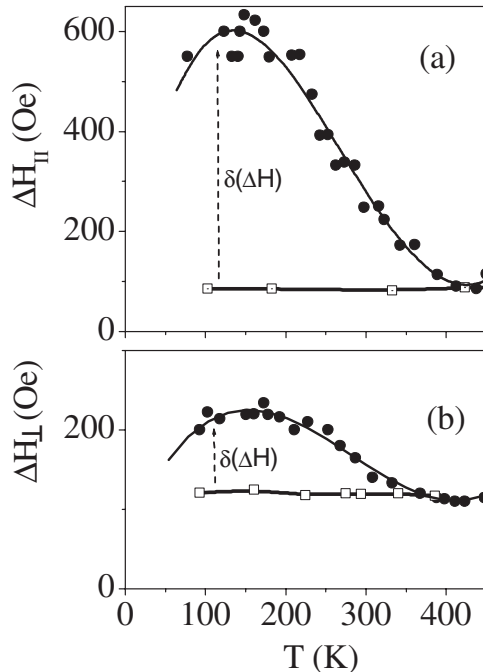


Fig. 2. FMR linewidth ΔH as a function of temperature for the NiO/NiFe structures measured in the in-plane (a) and the out-of-plane (b) configurations. Full and open symbols indicate the data for the biased and unbiased structure, respectively.

with temperature. Figure 2b displays ΔH_{\perp} vs. T for the magnetization oriented out-of the plane. Again, a shallow maximum is seen for the exchange-biased structure while practically there is no variation in the linewidth for the film with no bias. We define the difference between ΔH^b in the presence of the exchange-bias and ΔH^{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 $\delta(\Delta H_{\parallel}) \simeq 500$ Oe at 150 K is several times higher than $\delta(\Delta H_{\perp}) \simeq 100$ Oe, however

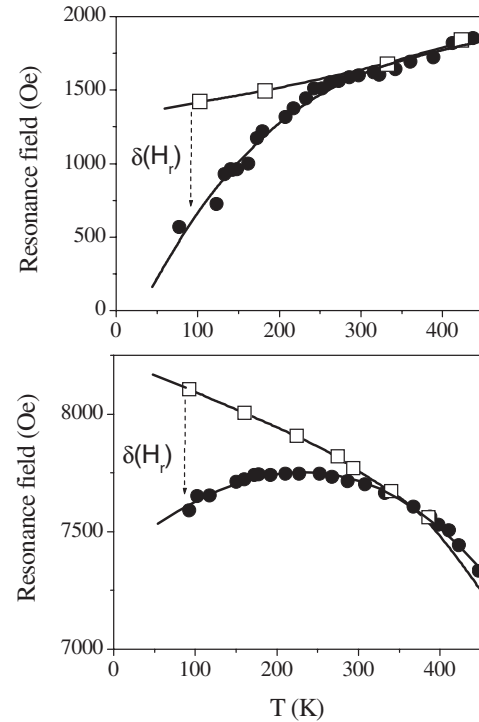


Fig. 3. Resonance field of the NiO/NiFe structures taken in the in-plane (upper panel) and the out-of plane (bottom panel) configurations. Full and open symbols represent the data for the biased and unbiased structure, respectively.

both are observed. The anomalous temperature behavior of the FMR linewidth for the exchange-biased films is associated with a substantial resonance field-shift of the NiO/NiFe exchange-biased structure with respect to that of the unbiased structure. It is seen in Figure 3 that in the unbiased NiO/NiFe structure the resonance fields depend on the temperature according to the “normal” behavior 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. On the contrary, 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 resonance field-shift due to the presence of exchange-bias as $\delta(H_r) = H_r^b - H_r^{nb}$, where H_r^b and H_r^{nb} refer to as the resonance field of biased and unbiased structure, respectively. For example, it is seen that $\delta(H_r)$ (in-plane) $\simeq \delta(H_r)$ (out-of-plane) $\simeq 600$ – 800 Oe at 100 K, in contrast with the results presented in reference [9] where $\delta(H_r)$ (out-of-plane) $\simeq 2 \delta(H_r)$ (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. Much

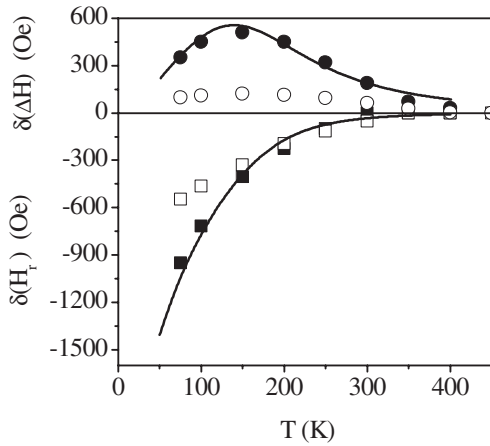


Fig. 4. Additional linewidth $\delta(\Delta H)$ and additional field-shift $\delta(H_r)$ due to FM/AF interaction in a NiO/NiFe structure for the in-plane (full symbols) and out-of-plane (open symbols) configurations. Symbols depict the experimental values obtained from Figures 2 and 3. Continuous lines are fits according to equations (1–4) to the data points for the in-plane configuration.

work has been done to explain various possible paths of transferring energy from a spin system to the lattice [27]. The effects reported in this paper, anomalous broadening of the linewidth and the negative field-shift, seem to be the main features in the exchange-biased structures [10–12,17]. 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 FM 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. [13]) does not explain a substantial temperature dependence of the damping, which has been observed both in FMR [12] and BLS [17] measurements. Temperature dependence of the FMR linewidth has been interpreted in terms of the mechanism of slow-relaxation [12]. We should emphasize here that there is no compatibility between these two mechanisms.

The main results of our temperature measurements, the anomalous linewidth $\delta(\Delta H)$ and the resonance field-shift $\delta(H_r)$, are shown in Figure 4 for the in-plane (solid symbols) and for the out-of-plane orientation, respectively. As it was suggested for the first time by McMichael et al. [10], the enhanced relaxation of AF grains by a thermally activated process may be regarded as a source of behavior of $\delta(\Delta H)$ 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 field-shift $\delta(H_r)$, both with the magnetization lying in-the plane and out-of-the plane. Our results seem to be consistent with relaxation related to thermally driven impurity relaxation process which is known for nearly five decades as the “slow relaxing ion” mechanism [4]. This mechanism involves modulation of the impurity levels in the vicinity of

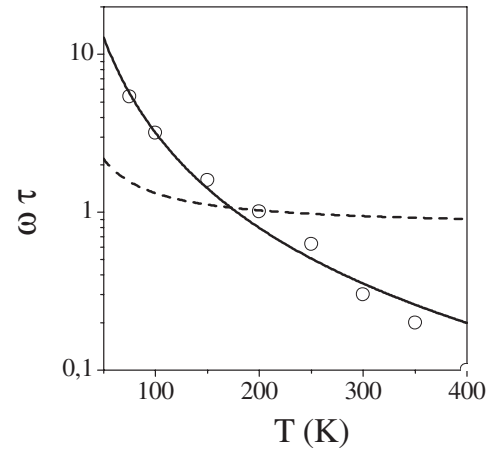


Fig. 5. $\omega\tau$ versus temperature T . Symbols depict the experimental values obtained from equation (3). Continuous line is a dependence $\tau \propto T^{-2}$ (Eq. (4)). Dashed line shows a dependence $\tau \propto \exp(\Delta E/k_B T)$.

thermal equilibrium by small magnetization oscillations observed in RE doped yttrium-iron garnets (YIG) [4, 24]. In a simplified form the theory (see reference [24] for details) predicts:

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

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

and thus

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

where τ is a temperature dependent relaxation time of the populations of the paramagnetic “impurity” ground state with a concentration c_{imp} . Since the energy modulation is absorbed by the lattice in extremely fast way in YIG doped with the RE ions, both $\delta(\Delta H)$ and $\delta(H_r)$ may be as large as several hundreds Oe [24].

By plotting $2\delta(H_r)/\delta(\Delta H)$ versus T (from Fig. 4) we can deduce the dependence of relaxation time on T (Fig. 5). The experimental data is best fitted with

$$\tau(T) = \frac{1}{BT^2}. \quad (4)$$

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. For the in-plane configuration data the fitting parameters were $c_{\text{imp}} = 1.7 \times 10^5$ and $B = 1.6 \times 10^{-3}$ GHz/K², which is of the same order as that obtained in reference [5]. For the out-of-plane resonance data the fit yields $c_{\text{imp}} = 0.6 \times 10^5$ and $B = 1.5 \times 10^{-3}$ GHz/K². However, the fit (not shown in Fig. 4) to the out-of-plane resonance data is somewhat worse than that for the in-plane resonance data. The value of the “impurity” concentration c_{imp} plays a role of a scaling factor while

the impurity interactions with conduction electrons has been suggested [5] to be responsible for $\tau \propto T^{-2}$ temperature dependence. The difference between the in-plane and out-of-plane resonance data shown in Figure 4 (and described by different c_{imp} values) suggests that FMR taken in the out-of-plane configuration is less sensitive to defects (impurities). Generally, the slow relaxer model for the RE-impurities in YIG monocrystals predicts a strong anisotropy in $\delta(\Delta H)$ and $\delta(H_r)$ values [24], but in thin polycrystalline Permalloy films in contact with NiO this difference remains to be explained.

A similar behavior of the FMR linewidth [10, 12] was interpreted in terms of the slow relaxation mechanism induced by the thermal reversals of small AF grains. The relaxation time for such small grains is known to depend on temperature as $\tau \propto \exp(\Delta E/k_B T)$ [28], where ΔE is a height of energy barrier and k_B is the Boltzmann constant. However, it is clearly seen from Figure 5 that this is not the case and $\tau \propto T^{-2}$ much better describes the experimental situation than the Néel relaxation time with $\Delta E \approx 0.7 \times 10^{-14} \text{ erg} \approx 35 \text{ cm}^{-1}$. Interestingly, the $\tau \propto T^{-2}$ dependence nicely agrees with that suggested by Safonov and Bertram [5] who have recently analyzed the microscopic relaxation mechanisms in thin films.

If the NiO grains in NiFe/NiO structures were superparamagnetic, they would have had two contradictory features: they should be somehow coupled to NiFe layer and they should have a short relaxation time of the order of $2\pi/\omega$. Thermal fluctuations of the magnetization in fine magnetic particles are strongly diminished by the magnetic interactions. For example, the magnetic dipolar interactions may result in an ordering of the magnetic particles at temperatures at which they would have been superparamagnetic if they were noninteracting [29]. Calculations of relaxation times in exchange-biased nanostructures [30] showed that exchange coupling increases superparamagnetic blocking temperature (i.e., increases relaxation times). On the other hand, it is not known how the nominally weakly-coupled fine NiO particles could effectively transfer energy flow from uniform precession to the lattice.

Alternative, and more physically appropriate approach would be an approach which takes into account fast energy transfer from the spin system to the lattice via a system of paramagnetic impurities – the slow-relaxing impurity mechanism. This mechanism is relevant if the spin system is tightly coupled to the system of impurities like in YIG doped with RE ions [24]. Using X-ray absorption spectroscopy Regan [31] showed that oxidation/reduction reactions occur at metal/oxide interfaces. The oxide regions, few angstroms in thickness, should provide such a tight coupling between Ni^{2+} and Fe^{2+} ions and NiFe layer. After Safonov and Bertram [32] the use the word “impurities” applies strictly to the lowest energy splittings of ions whose angular momentum is not quenched by the crystal field. Recent experimental [33] as well as theoretical calculations [34] have shown that the orbital moments of Ni^{2+} and Co^{2+} are not quenched and amount to 19 and 34% of the total moment in NiO and CoO, respectively. There-

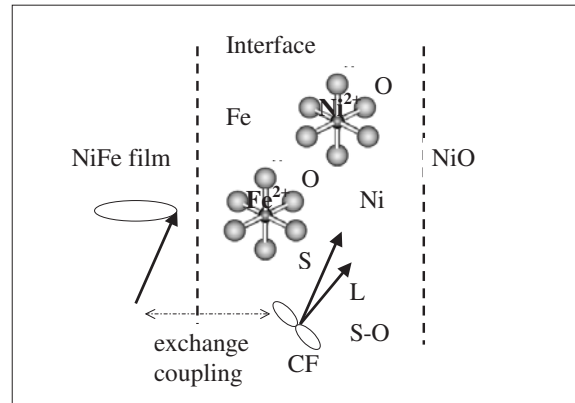


Fig. 6. Schematic view of a possible NiFe/NiO interface. Ni^{2+} and Fe^{2+} ions are inside oxygen octahedra and exchange coupled to NiFe layer. S-O and CF denote spin-orbit coupling and crystal field, respectively. Interface region is about 0.5 nm thick according to reference [31].

fore, both Ni^{2+} , Fe^{2+} and Co^{2+} ions at NiFe/NiO and NiFe/CoO interfaces can be seen as entities which fulfil the basic requirements of the slow-relaxing impurities mechanism [24, 35]: exchange coupling between spin system (NiFe layer) and impurities (Ni^{2+} and Fe^{2+} ions), substantial orbital moment of impurities, and the existence of anisotropic states of impurities which markedly depend upon orientation of magnetization with respect to the crystallographic axes of NiO [24]. The latter feature seems to be important in crystalline FM/AFM structures. Most of FMR experiments have been performed on polycrystalline samples but recent experiments [11] in quasiepitaxial Fe/MnFe₂ exchange-biased structure have revealed substantial divergence of FMR linewidth in the hard directions with no clues on the origin of the effect. However, the similar sharply peaked variations in the linewidth in YIG doped with Ho^{3+} were interpreted [36] as resulting from a strong anisotropy in energy splittings of Ho^{3+} ion in the crystal field of YIG.

In most papers concerning FMR in exchange-biased FM/AFM structures the exchange anisotropy is regarded to be responsible for the enhanced spin-wave damping and isotropic field-shift. However, Ercole et al. [16, 17] showed a strong enhancement in spin-wave damping in Co/CoO bilayers even in the absence of the unidirectional anisotropy. They explained the effect in terms of locally ordered AF regions which persist even above the Néel temperature. In view of the slow-relaxing impurity mechanism their results can be plausibly explained as resulting from exchange coupling between Co spin system and interfacial Co^{2+} impurities. Co^{2+} ion is known to be the most anisotropic 3d ion [37] and its orbital moment amounts to more than 30% of the total moment [34].

Keeping in mind the above discussion concerning results in various FM/AFM structures, we conclude that the paramagnetic Ni^{2+} and Fe^{2+} ions which are located at the NiFe/NiO interface may be responsible for the observed effects in our exchange-biased NiFe/NiO films. Such a scenario is schematically shown in Figure 6.

5 Conclusions

In summary, we have observed the temperature dependent magnetization dynamics in thin NiFe layers in contact with an antiferromagnetic NiO. The characteristic maximum in the spin-wave damping and the large negative dynamic resonance field-shift at low temperatures are consistently interpreted in terms of the slow-relaxation mechanism via interaction of the magnetization precession with the slowly-relaxing impurities. The nature of the slowly-relaxing impurities is not clear yet, but we suppose from the T^{-2} dependence of relaxation time and from discussion of other FMR/BLS experiments that they might be the Ni^{2+} and Fe^{2+} ions.

This work was supported by the Centre of Excellence for Magnetic and Molecular Materials for Future Electronics within the European Commission Contract No. G 5MA-CT-2002-04049 and KBN project PBZ/KBN-044/P03-2001 and partially by KOSEF through Quantum Photonic Science Research Center (q-Psi).

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