TITLE: Oscillatory Angular Dependence of Exchange Bias for Epitaxial NiO-Co [001] Bilayers

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OSCILLATORY ANGULAR DEPENDENCE OF EXCHANGE BIAS FOR EPITAXIAL NiO-Co (001) BILAYERS

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ABSTRACT

We have sputter-deposited NiO-Co bilayers on MgO (001) substrates. NiO grows epitaxially on MgO at 900°C and subsequently the room deposited 80 Å thick Co films have a fcc crystal structure in epitaxy with the oxide underlayer. These samples were warmed up to 300° C and then zero-field or field cooled through the NiO Néel temperature (a 300 Oe magnetic field was applied along the [100] or the [110] MgO axis). Magnetic hysteresis loops were obtained by magneto-optical Kerr effect, the magnetic field being oriented in the plane of the substrate for various angles α with respect to the [100] direction. The usually expected behavior for such experiments is a smooth angular α dependence of the exchange bias H_E close to a cosine with only one sign change over 180°. The high crystallographic coherence of our NiO/Co bilayers induces a very unusual oscillatory H_E (α) dependence with several sign changes according to the NiO axis field application. Despite of the Co magnetization switching mechanism which is not a pure coherent rotation, we propose a Stoner-Wohlfhart model including four fold anisotropy and unidirectional exchange anisotropy giving a realistic description of these typical magnetic properties.

INTRODUCTION

Exchange coupling between a ferromagnetic (F) and an antiferromagnetic (AF) was first observed more than 40 years ago by Meiklejohn and Bean [1]. It is presently a subject of intense investigations in thin magnetic layers community because of its applications in magnetoresistive (MR) devices such as spin-valves and magnetic tunnel junctions (one layer of the MR device is left « free » while the other is hardened by exchange coupling with an AF). The main effect induced by this coupling is a typical shift of the ferromagnet hysteresis loop called exchange bias H_E and an increase of its coercive field H_C. In most models, the AF-F system global anisotropy is considered as a sum of uniaxial and unidirectional anisotropies. The former inducing an increase of H_C and the latter the loop shift H_E. Since the coupling is assumed to be interfacial, H_E can be expressed as:

$$H_E = \frac{J_C}{M_F t_F} \quad (1)$$

Where J_C is the AF-F interfacial exchange coupling intensity, M_F and t_F are respectively the ferromagnet magnetization and thickness. Realistic values of H_E involve the formation of domain walls in the AF during the magnetization reversal of the F layer and relate J_C to the energy of such a domain wall [2,3] instead of pure exchange between the AF and F spins at the interface. They lead to the following expression: $J_C \approx \sqrt{A_{AF} K_{AF}}$. However, the presence of structural defects like interface roughness or grain boundaries is also supposed to have a significant effect on the AF-F behavior. One can also relate J_C to the limitation of the extent of domain walls in the AF or to the presence of loose spins at the AF-F interface. Therefore, a better understanding of

the mechanism of exchange anisotropy requires AF-F systems as ideal as possible. NiO-Co epitaxial bilayers are good candidates for such investigations since NiO has a relatively high Néel temperature (525 K) and a low magnetocrystalline anisotropy, such properties are expected to lead to exchange bias effects visible at room temperature and relatively sensitive to the structural quality of the samples (a small anisotropy leads to large domain structures which are mainly expected to show up for samples with excellent crystallographic coherence). Thus this system has been chosen by our group [4][5] and others like Lai et al. [6] for [001] or [111] NiO-Co bilayers. In the present study, we will focus to magnetic cycles measured on such samples for varying angles between the anneal field direction and the in-plane applied field direction. Such a procedure was first proposed by Ambrose et al. [7]. A cosine-like dependence of H_E is expected for standard AF-F systems, the exchange field varying smoothly from $-H_{E-Max}$ to $+H_{E-Max}$ over a 180° rotation, as expected from an uniaxial/unidirectional effect. We demonstrate in this paper that, combined with the magnetocrystalline anisotropies of the F and AF layers, exchange bias may have unexpected oscillations superposed to the smooth cosine effect observed for polycrystalline samples like in ref. [9]. This behavior can be interpreted with the simple Stoner-Wohlfarth (SW) approach as the sum of unidirectional and four-fold anisotropies.

EXPERIMENTAL DETAILS

Samples were grown in a Plassys MPU 600S ultra high vacuum sputter system on epi-polished MgO (001) substrates. Substrates were first annealed for 1 hour at 900°C with a base pressure better than 5×10^{-8}mB. NiO was rf-sputtered from a facing target magnetron with a deposition rate of ≈ 0.19 Å/s. The MgO substrates were heated at 900°C during the NiO growth. The plasma was obtained from a Ar(90%)-O_2(10%) gaz mixture to prevent oxygen understoichiometry. The total working pressure during the growth was set to $P=5.6\times10^{-3}$ mB. The NiO thicknesses of our series of samples ranged from 335 to 1000 Å. Just after NiO deposition, the sample was cooled down in zero magnetic field down to room temperature for Co deposition. Typical Co growth conditions were achieved with a standard magnetron operated in pure Ar plasma and with a deposition rate of ≈ 0.15 Å /s. In a last step, samples were capped by a 20 Å non magnetic (Al or Mo) layer before air exposure. In-situ Reflection High Energy Electron Diffraction (RHEED) was performed at all steps of the growth (bare MgO substrates, NiO layer, Co film) with a 20 kV STAIB set-up.

After the growth, the samples surfaces were investigated by Atomic Force Microscopy with a Nanoscope III microscope in tapping mode. TEM and HREM experiments, presented elsewhere [10] were also carried out on a Philips CM30/ST working at 300kV with a point resolution of 1.9 Å to evidence the crystalline quality of the bilayers.

Then the samples were annealed from 300°C to room temperature with a 300 Oe in-plane applied magnetic field to organize the NiO spin with respect to the cobalt layer. The angle between the anneal field and the in-plane reference direction [100] is referred as ω in Fig. 1. Electron microscopy was performed before and after their thermal treatments to make sure that no structural change was induced. Finally, the magnetic properties were studied at room

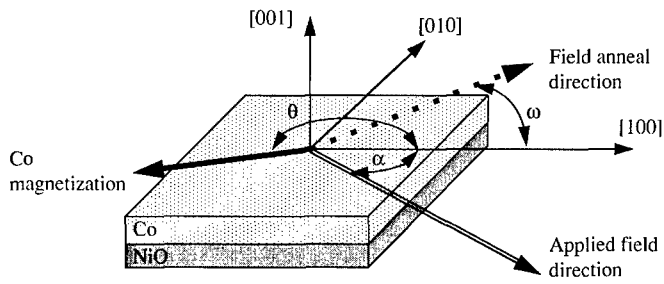

Figure 1: Configuration schematics of our orientation magnetic measurements. Angle θ represents the magnetization angle with respect to [100] axis used for Stoner-Wohlfarth modelling.

temperature by Magneto-Optic Kerr Effect (MOKE) with a polarized He:Ne laser (λ = 6328 Å). Accurate orientations of the sample in plane directions α with respect to the applied magnetic field were obtained with a goniometric sample holder. The schematic configuration of our measurements is presented in Fig. 1.

STRUCTURAL PROPERTIES

RHEED patterns, AFM scans and TEM and HREM experiments were carried out on these samples. Figure 2 shows RHEED patterns obtained for a MgO (001) bare substrate heated at 900°C prior to the NiO deposition along [100] and [110] azimuths. The same experiments are also presented after a 400 Å NiO film was deposited. The last two patterns are indicative of a smooth NiO growth with a cube-on-cube epitaxial relashionship NiO [100] (100) // MgO [100] (100). Not presented in Fig. 1, RHEED patterns on subsequently deposited Co layers reveal a similar symmetry but with a broadening of the RHEED lines due to the large misfit between NiO and Co. However, fcc cobalt structure with interface dislocations was evidenced by HREM in plane views or cross sectionnal specimens [4,5,9,10]. The surfaces are very smooth as evidenced by AFM with peak-to-peak features smaller than 15 Å.

MAGNETIC PROPERTIES

Figure 3 represents several magnetization loops obtained for a NiO(330 Å)-Co(60 Å) bilayer field-cooled along MgO [110]. Compared to a cobalt single layer, the mean coercive field increases from ≈ 40 Oe up to ≈ 500 Oe. Along the field anneal direction, the loop is shifted of $H_E \approx$ -100 Oe. Note that the maximum values of H_E are actually obtained ≈ 20-30° away from this direction. For increasing α, H_E absolute value decreases and gets a zero value close to α = 90-100°, then its sign gets positive from 100 to 130° and experiences a narrow negative domain between 135 and 170° followed by the expected positive values. All this behavior is summarized in Fig. 4. Note that H_C also fluctuates, evidencing four minima and maxima on 360°. This is indicative of the succession of four easy and hard axes. Thus we have simulated the experimental magnetization loops with the SW model. The bilayer energy can be decomposed as shown in Equation 2:

$$E = -HM_F \cos(\theta - \alpha) - \frac{J_C}{t_F} \cos(\theta - \omega) - K_{eff} \cos^2(2\theta) \qquad (2)$$

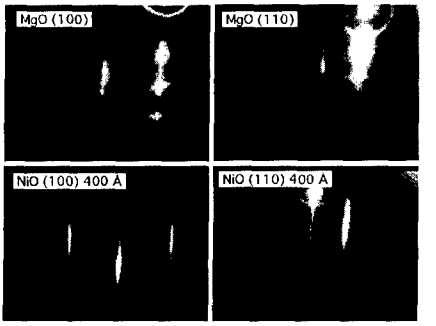

Figure 2: RHEED patterns of MgO (001) substrate prior to NiO 400 Å deposition. Pictures have been taken along both (100) and (110) azimuths.

Where the first term is the Zeeman energy of the Co layer, the second is the unidirectional exchange bias term and the third is a four-fold anisotropy term. θ is the angle between the magnetization of the Co layer and [100] axis. K_{eff} is the effective four-fold anisotropy constant. The fit was done with $\omega = 45°$ since the field anneal was done along [110]. Fit of the coercive field is not presented in Fig. 4. It yields to underestimated values. We attribute this discrepancy to the main limitation of SW model which assumes a magnetic switching by coherent rotation whereas our samples switch by a combination of coherent rotation, domain nucleation and domain wall propagation as it has been recently observed by Kerr microscopy [11]. The values used for this fit are $J_C = 7.8\times10^{-2}$ erg/cm^2 and $K_{eff} = 7.5\times10^4$ erg/cm^3. Statistics on our data collected for several thicknesses of NiO and Co yield values for J_C and K_{eff} respectively comprized between a 3×10^{-2} and 9×10^{-2} erg/cm^2 and 6 10^4 and 1×10^5 erg/cm^3. Indeed, Fig. 5 gives some insight of the validity of the SW model for the description of the oscillatory angular dependence of H_E. This figure is a grey levels image plot the bilayer magnetic energy obtained from Eq. (3) with $\alpha=105°$ (therefore at the exchange bias oscillation position) versus the magnetization angle θ and the applied field value. The solid white line corresponds to the decreasing field switching process. The magnetization angle progressively changes from 105° to ≈80°, then it switches rapidly to the opposite direction. The broken white line corresponds to the increasing field switching process. It is more vertical, signicative of an easier magnetic direction and the switching field is larger in absolute value (≈ 170 Oe). This behavior can be noticed in the marked asymmetry of the ninth magnetization loop of Fig. 3 marked with an asterisk. However, the experimental switching field values are far from the calculated ones. This discrepancy is due to the actual switching process wich involves domain nucleation and wall motion and that the basic SW approach does not take into account.

CONCLUSION

We have prepared exchange-biased NiO-Co bilayers by a combination of facing-target and standard magnetron sputtering. The epitaxial quality of our samples has allowed us to observe

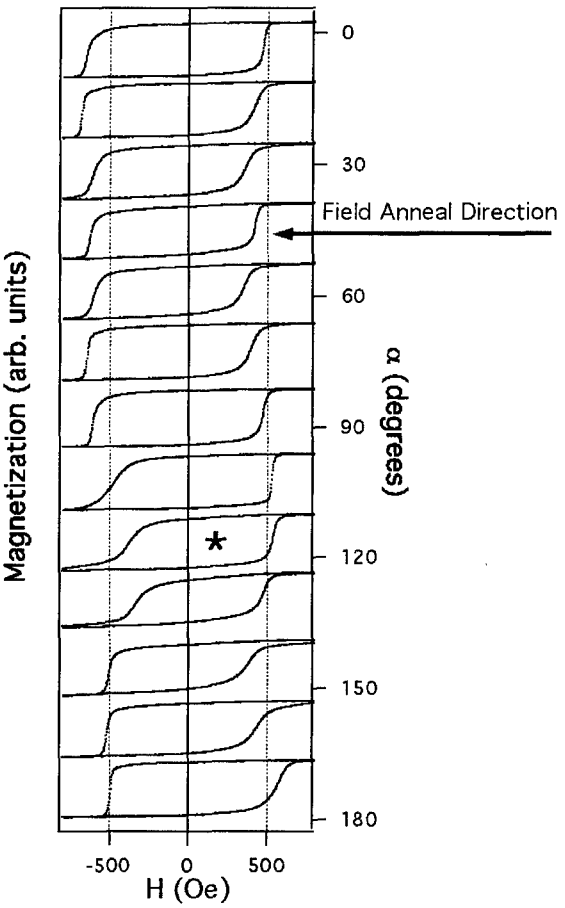

Figure 3: Magnetization loops of a NiO(330 Å)-Co(80 Å) bilayer for various angles between [100] axis and applied field direction. Note the original H_E sign changes.

original angular dependence of exchange bias. This behavior can be fully understood in terms of co-existence of uniaxial anisotropy and a four-fold extra term. The origin of the four-fold anisotropy is clearly due to the large crystallographic coherence of the samples. Such results emphasize the fundamental interest of high quality AF-F bilayers for understanding the mechanism of AF-F exchange coupling.

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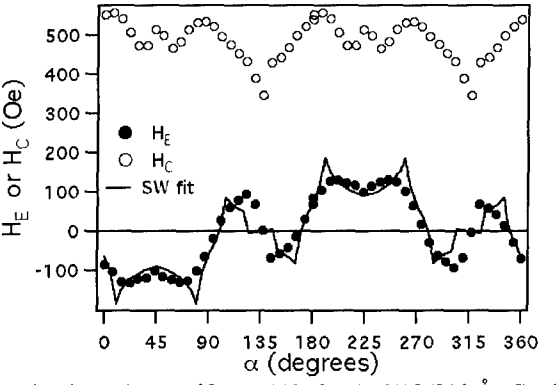

Figure 4: Angular dependence of H_C and H_E for the NiO(330 Å)-Co(60 Å) bilayer.

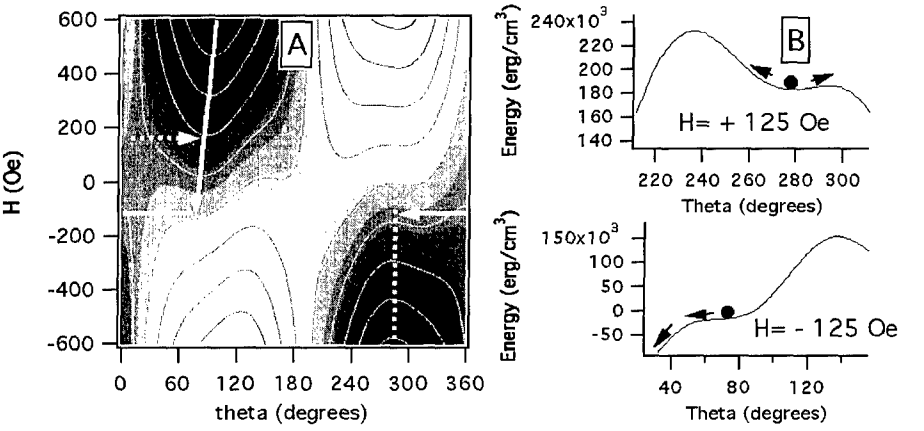

Figure 5: A: Two-dimensional grey level plot of the bilayer magnetic energy for α=105°.White lines correspond to the Co layer switching (broken line: increasing field). B: Profiles of the energy surface for H=±125 Oe evidencing the switching fields asymmetry.

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