Eur. Phys. J. B **45**, 175–179 (2005)
DOI: 10.1140/epjb/e2005-00180-9

THE EUROPEAN PHYSICAL JOURNAL B

Complex angular dependence of exchange bias on (001) epitaxial NiO-Co bilayers

S. Dubourg[1,a], J.F. Bobo[1,b], B. Warot[2], E. Snoeck[2], and J.C. Ousset[2]

[1] LNMH-CNRS, ONERA, 2 avenue Edouard Belin, 31400 Toulouse, France
[2] CEMES CNRS, 29 rue Jeanne Marvig 31055 Toulouse, France

Received 15 September 2004 / Received in final form 19 April 2005
Published online 28 June 2005 – © EDP Sciences, Società Italiana di Fisica, Springer-Verlag 2005

Abstract. We have sputter-deposited NiO-Co bilayers on MgO(001) substrates. NiO and Co grow epitaxially on MgO and reproduce its fcc structure. The high quality of our samples, in terms of flatness and crystallographic coherence of the interface, allows the observation of an additional fourfold magnetic anisotropy term by standard magnetometry. This term is induced by interfacial interaction assigned to the same origin as exchange bias. Additional measurements of exchange bias azimuthal dependence versus the crystallographic axes of the film plane reveal unusual behaviors with several sign changes related to this fourfold anisotropy.

PACS. 75.50.Ee Antiferromagnetics – 75.60.-d Magnetic domain effects, magnetization curves, and hysteresis – 75.70.-i Magnetic properties of thin films, surfaces, and interfaces

1 Introduction

Almost fifty years ago, Meiklejohn and Bean evidenced exchange coupling between a ferromagnet F and an antiferromagnet AF in partially oxidized cobalt particles [1]. It is characterized by a shift of the ferromagnet hysteresis loop called exchange bias (H_E). Recently, lots of efforts were made to understand its origin since the effect has important applications in magnetoresistive sensors based on multilayer stacks where one layer is magnetically pinned by exchange bias with an AF layer. Most theoretical models were based on the Stoner-Wohlfarth representation of magnetization switching. Exchange bias adds an unidirectional term to the energy leading to the following H_E expression:

$$H_E = \frac{J_C}{M_F \cdot t_F}$$

where M_F and t_F are respectively the F magnetization per volume unit and the F layer thickness. J_C is the interfacial strength of the unidirectional anisotropy induced by the exchange coupling. Some authors link the J_C value to the presence of AF uncompensated spins at the AF/F interface [2,3] while others suspect the spreading of a domain wall in the AF layer during the magnetization switching [4–6]. All these models predict a realistic value of J_C and H_E. They all make assumptions about the samples structure, especially their crystallinity and interface roughness. Preparation of samples with a high crystallographic coherence is expected to allow a better understanding of the real origin of the interfacial coupling [7–9]. For example, Noguès et al. [10] evidenced an antiferromagnetic coupling between AF and F layers in FeF_2-Fe samples. The sign and the value of the exchange bias field could be modulated according to intensity of the applied field during cooling through the AF Néel temperature, but this property is absent in rough interface samples [11]. Here, we have grown NiO-Co bilayers with a very high crystalline coherence and very smooth interface on MgO(001). We evidence an additional anneal-induced interfacial four-fold anisotropy term. Combined with field cooling, it leads to an unusual angular dependence.

2 Samples preparation and structural characterization

NiO(x Å)-Co(80 Å) (x = 0, 50, 160, 335, 600) and NiO(335 Å)-Co(y Å) (y = 20, 40, 80, 160, 320) samples were grown in a Plassys MPU 600S ultrahigh vacuum sputter system on epi-polished MgO(001) (these substrates have an initial RMS roughness lower than 1 nm). Substrates were first annealed 1 hour at 900 °C with a base pressure better than 5×10^{-8} mbar. NiO was rf-sputtered from a facing target magnetron with a deposition rate ∼0.19 Å/s. Reactive plasma with 10% oxygen partial pressure was used in order to ensure the correct Ni:O stoichiometry. During NiO growth, the MgO was heated at 900 °C. Cobalt layers were deposited at room temperature with a standard magnetron and capped with a 30 Å aluminum layer. High angle X-Ray diffraction

[a] *Present address*: CEA Le Ripault, 37260 Monts, France.
[b] e-mail: jfbobo@onecert.fr

and HREM (High Resolution Electron Microscopy) experiments were carried out on samples before and after field cooling. They both emphasize the high epitaxial quality of the samples. Due to their very close respective lattice parameters (4.18 Å and 4.22 Å), NiO reproduces the MgO fcc structure. Cobalt also adopts a fcc structure but relaxes with misfit dislocations. Then, low NiO deposition rate causes a very flat surface. Low angle X-Ray diffraction patterns indicate for a 1000 Å thick NiO sample a RMS NiO/Co interface roughness smaller than 4 Å. HREM pictures reveal a sharp NiO/Co interface. The detailed samples structure is presented elsewhere [12]. To study their magnetic properties, each sample was cut in four pieces. The first piece was zero-field cooled through the NiO Néel temperature ($T_N = 250$ °C) from 300 °C to room temperature. The second and third ones were field cooled in a $H_a = 300$ Oe magnetic field either along the [100] MgO axis or along the [110] MgO axis. The last one was left at room temperature as a reference sample. No structural modification was found after the thermal treatments.

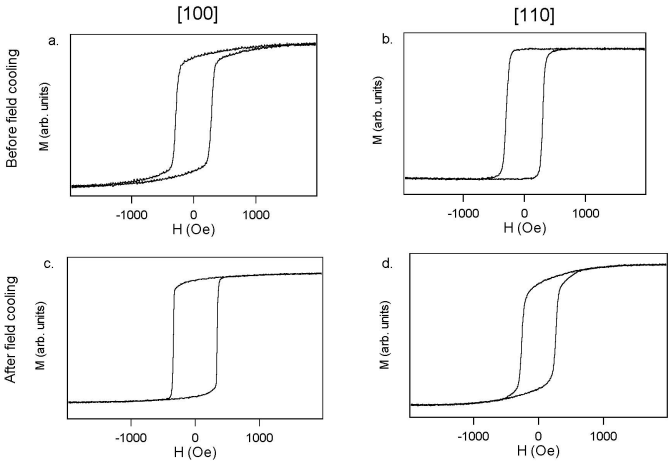

Fig. 1. Hysteresis loops of MgO/NiO(335 Å)/Co(80 Å)/Al at room temperature along the [100] MgO direction (a and c) or along the [110] (b and d) before (a and b) and after (c and d) a zero field cooling.

3 Zero field cooled samples

Magnetization loops of the samples were measured by Kerr effect with a He:Ne laser ($\lambda = 633$ nm) with s-polarization in the longitudinal configuration at room temperature. For all NiO/Co bilayers, the Co magnetization is in-plane, exhibiting a four-fold magnetic anisotropy. The easy axis direction depends on layer thicknesses and on thermal treatments.

In Figure 1 are presented hysteresis loops at room temperature of the MgO(001)/NiO(335 Å)/Co(40 Å)/Al sample along the [100] (id. [010]) and the [110] (id. [1$\overline{1}$0]) directions before and after zero field cooling. Note that the loop squareness M_R/M_S (remnant magnetization over saturation magnetization) decreases from 100% to 72% along the [110] axis after cooling through T_N and at the same time increases from 72% to 95 % along the [100] axis. Easy axis is assigned to the in-plane azimuth with the largest squareness. Before field cooling, all samples exhibit easy axes along the [110] and [1$\overline{1}$0] MgO directions with a remnant magnetization equal to the saturation magnetization. After cooling (with or without magnetic field), easy axes of some samples have switched to the [100] and [010] MgO directions.

In Figure 2a are plotted the NiO(xÅ)/Co(80 Å) loop squareness versus nickel oxide thickness after zero field cooling. Thin NiO layers (<200 Å) do not significantly induce easy axes changes since the [110] M_R/M_S ratio is still equal to 100% after cooling through T_N. However, we note a squareness increase along the [100] directions. In thicker samples, the squareness along the [110] axis drops to 72% while it reaches more than 90% along [100]. So a critical NiO thickness close to 250 Å is required to obtain easy axes switching. Figure 2b represents the influence of the cobalt layer thickness on the hysteresis loop squareness for a constant NiO thickness equal to 335 Å. No easy axis rotation is evidenced for cobalt layers thicker than 160 Å. The M_R/M_S ratio remains constant along the [110] directions. The squareness change appears for cobalt layers thinner than 120 Å. Along the [100] axis, the squareness raises to 95% and it drops down to 84% along the [110] axis. These results indicate that the observed effect has an interfacial origin. The thicker cobalt layers keep their easy axes along the [110] and [1$\overline{1}$0] axes while, in the thinner, the interfacial contribution overcomes the bulk anisotropy and forces the [100] and [010] as the new easy axes.

One would attribute this phenomenon to a relaxation of the induced strain by annealing. This necessitates a lattice mismatch change. High Resolution Electron Microscopy (HREM) micrographs were carried out before and after field cooling on the NiO(335 Å)/Co(80 Å) sample since it presents a strong anisotropy switch. They reveal no crystal structure changes. We can rule out, as well, the hypothesis of an interfacial oxidation of the cobalt layer leading to the appearance of a thin and magnetically hard CoO_x layer between NiO and Co which cannot be revealed by HREM. We exclude this oxidation as the basic mechanism of the easy axis switching since the effect depends on the NiO layer thickness (it appears beyond a critical thickness (250 Å)). On the contrary, interfacial oxidation would affect the anisotropy for any NiO thickness. Let us point out that this effect has great similarities with exchange bias field: it is an interfacial coupling which requires a minimal NiO thickness and appears after a cooling through T_N. These features are strong arguments in favor that the exchange coupling induces these easy axes switchings. It is known that AF reorganize their domain structures during a field cooling to minimize their energy. We think that a reorganization can be driven by the remnant state. Once combined with an epitaxial structure and a highly flat AF-F interface, these new domains would lead to the appearance of the additional four-fold anisotropy. The thickness dependence shows that a minimum NiO thickness is required to induce this new

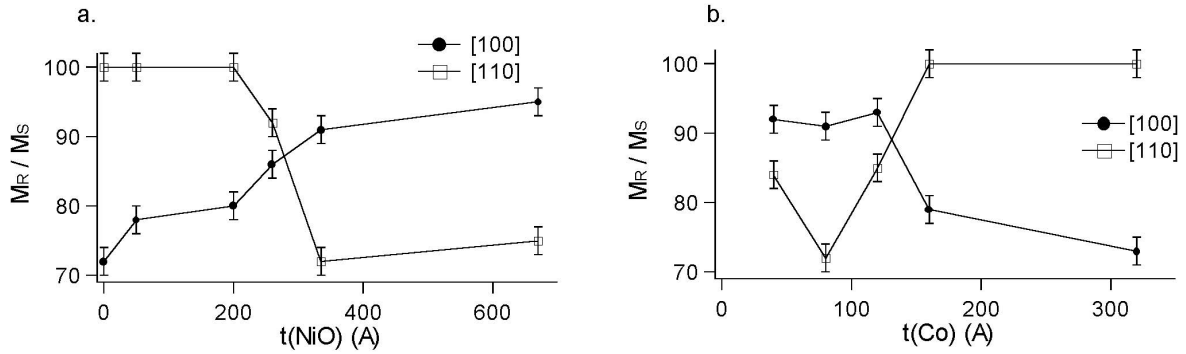

Fig. 2. M_R/M_S ratio versus nickel oxide and cobalt thicknesses at room temperature. a. $t(\mathrm{Co}) = 80$ Å. b. $t(\mathrm{NiO}) = 335$ Å after a zero field cooling from 300 °C to room temperature. Solid lines are guides for the eyes.

anisotropy. It overcomes the intrinsic cobalt anisotropy of the thinnest ferromagnetic layers and reorients the easy axes towards new directions.

4 Field cooled samples

After field cooling along the [100] MgO axis or the [110] MgO axis, the NiO thick samples ($t_{\mathrm{NiO}} > 200$ Å) exhibit exchange bias as previously reported by Laï et al. [13]. As proposed by Ambrose et al. [16], we have studied the H_E angular dependence of the field cooled sample MgO/NiO(335 Å)/Co(80 Å)/Al. For that purpose, we have measured hysteresis loops of cobalt layers with the magnetic field applied in the plane of the substrate at several azimuthal angles α. The [100] MgO axis is referred to as $\alpha = 0$. Field cooling was performed along two different axes [100] and [110]. The angular dependences are shown in Figures 3a and b. The cooling field direction and the crystalline structure impose the following H_E angular symmetries:

$$H_E(\alpha) = H_E(-\alpha), H_E(\alpha) = -H_E\left(\alpha + 180^\circ\right)$$

for [100] field cooling and

$$H_E\left(\alpha + 45^\circ\right) = H_E\left(45^\circ - \alpha\right),$$
$$H_E\left(\alpha + 45^\circ\right) = -H_E\left(\alpha + 225^\circ\right)$$

for [110] field cooling.

This behavior is common to field annealed AF/F systems as previously reported for example by Xi et al. [17]. They have studied polycrystalline samples and have reported a smooth H_E variation very close to a cosine law with only two sign changes over the [0,360°] α range. Our epitaxial samples follow the same symmetries. In addition the H_E angular dependences reveal more sign changes. In particular, when the cooling field is applied along the [110] axis, H_E exhibits the oscillatory dependence of Figure 3b. Maximum exchange bias value close to 20 Oe is obtained at 20° and 70°. It becomes negative between 45° and 90° away from the cooling field direction and its sign changes again 45° further.

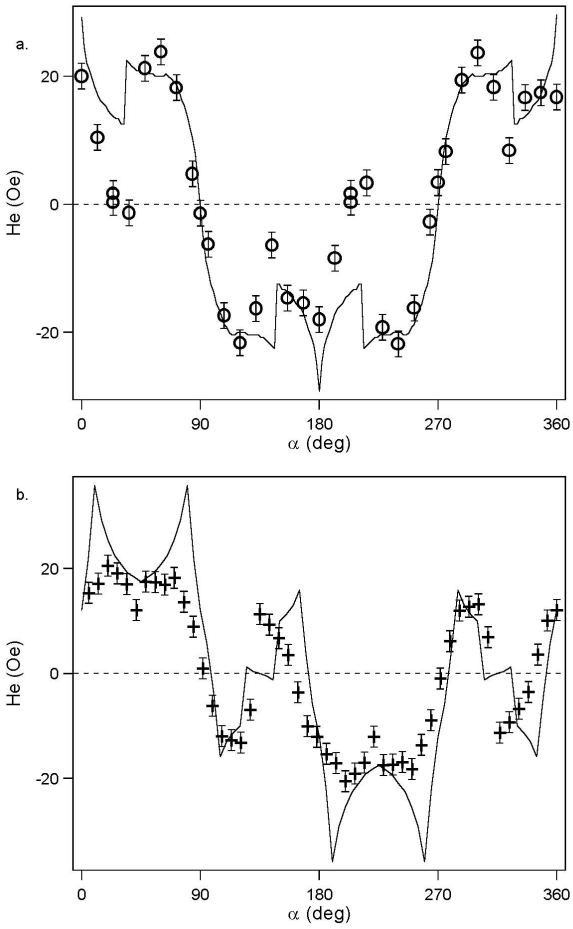

Fig. 3. Exchange bias angular dependence of MgO/NiO(335 Å)/Co(80 Å)/Al field cooled with $H_a = 300$ Oe along the [100] MgO axis (a) or along the [110] MgO axis (b) at room temperature. Solid lines are numerical simulations of the experimental data obtained by equation (1).

This behavior can be roughly modeled using the Stoner-Wohlfarth coherent rotation model [19]. Note that such modeling was recently done by Hoffmann et al. [18]. As a natural extension of the observed anisotropy symmetry, an interfacial fourfold anisotropy term is added to the

intrinsic fourfold anisotropy of the bulk cobalt layer. This term includes bulk and interfacial contribution which were evidenced in the zero field cooling section. The expression of the NiO-Co bilayer energy is given by equation (1).

$$E = -HM_F\cos(\theta-\alpha) - \frac{J_C}{t_F}\cos(\theta-\omega) - \frac{K_{eff}}{4}\cos^2(2\theta). \quad (1)$$

The first term is the Zeeman energy, θ is the angle between the magnetization and the [100] axis and α is the angle between the applied field and the [100] direction. M_F is the cobalt saturation magnetization equal to 1400 emu/cm^3. J_C and K_{eff} respectively represent the unidirectional exchange anisotropy and the effective fourfold anisotropy, ω is the angle between the field cooling direction and the [100] MgO axis, so $\omega = 0$ if $H_a \parallel$ [100] and $\omega = 45°$ if $H_a \parallel$ [110]. Note that we have modeled the angular dependence for the two configurations with almost the same value of K_{eff}.

The numerical procedure used for computing the angular dependence is based on the perfect delay convention (this means the system remains in the local energy minimum until this minimum disappears, this convention neglects the formation of reversed domains or thermal activation for magnetic switching). Least square fitting to experimental points $H_E(\alpha)$ was performed for the 0–360 degrees angular range.

The set of parameters for our calculation is $J_C = 3.36 \times 10^{-2}$ erg/cm^2 and $K_{eff} = 3.2 \times 10^5$ erg/cm^3 when $H_a \parallel$ [100] and $J_C = 2 \times 10^{-2}$ erg/cm^2 and $K_{eff} = 3 \times 10^5$ erg/cm^3 when $H_a \parallel$ [110].

One can note that our data points show some scattering or non symmetry (see Fig. 3a for example at $\alpha = 135$ and 225° where a zero-crossing is expected for H_E. We assume these features are related, either to some slight misalignment of the anneal magnetic field with respect to the sample $\alpha = 0$ or 45° reference directions, or to some possible training effects [14,15].

Addition of higher order terms in the free energy of a pinned layer has been previously proposed by Ambrose et al. [16] for polycrystalline samples to explain angular dependence of exchange coupling which is more complicated than a cosine. Adding an uniaxial easy axis parallel to the unidirectional axis is in good agreement with experiments in polycrystalline samples. It explains both the coercive field increase and the angular dependence of the exchange bias in a very soft magnetic layer such as NiFe. Induced fourfold anisotropy in NiO-NiFe has also been proposed to explain the hysteresis loops shapes [21,22]. All the results clearly demonstrate that exchange coupling cannot be described by a simple cosine term in the free energy. The oscillatory angular exchange bias dependence after [110] field cooling is indicative of the existence of this new contribution to exchange coupling. Even if recent studies made by Kerr microscopy indicate that pinned layer switching occurs by nucleation and propagation of domain walls or by incoherent rotation, the simple Stoner-Wohlfarth model provides a broad outline of the angular dependence for both cooling field directions. However, we reach the limitations of the Stoner-Wohlfarth model with agreement not very good between the model and experiments in the direction [100]. The calculated effective fourfold anisotropies are the sum of bulk and interfacial terms leading to a good description of the H_E oscillatory behavior. The competition between the unidirectional and the fourfold anisotropies lead to sharp changes in H_E which falls down from its maximum value (20 Oe) to its almost lowest value (−17 Oe) 45° further. This unexpected sign change for H_E is shown in Figure 4 for a NiO(330 Å)-Co(60 Å) bilayer field annealed along [110] [23]. While the loop shift should smoothly vary from negative at $\alpha = 45°$ to positive at $\alpha = 45 + 180 = 225°$, we observe a sudden sign change for H_E around $\alpha \sim 120°$ which is the signature for the extra four fold anisotropy generated by exchange coupling of Co layer with the epitaxial NiO underlayer.

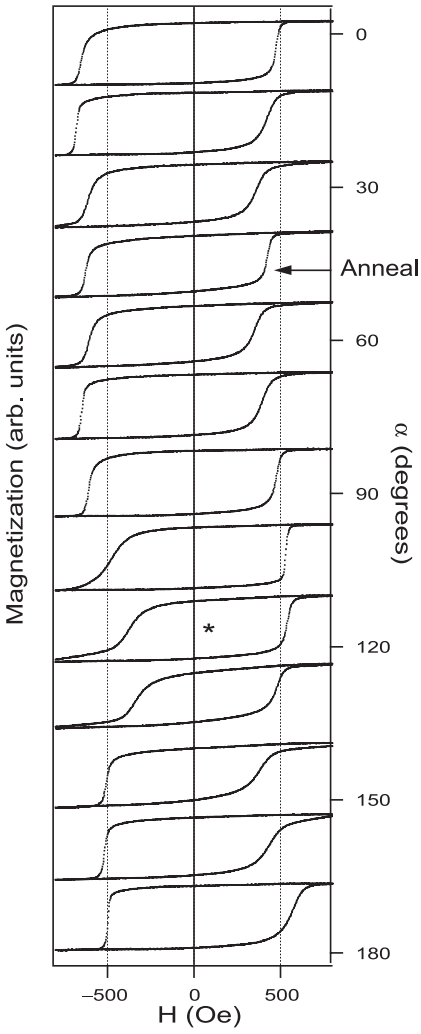

Fig. 4. Set of hysteresis loops recorded for a NiO(330 Å)-Co(60 Å) bilayer. The initial field anneal was performed along [110] axis ($\alpha = 45°$). The * label indicates the anomalous positive H_E values observed for $100° < \alpha < 140°$.

Such an evidence for an extra four-fold anisotropy induced by the antiferromagnet has been reported by other authors, in each case on epitaxial systems, either by FMR studies in FeMn-Fe system [24] or by standard magnetometry in NiFe-FeMn bilayers [25]. Our study shows strong evidence of the presence of an extra fourfold anisotropy in a metal-oxide system, with field annealings performed along two crystallographic axes.

5 Conclusion

In conclusion, we have prepared NiO-Co bilayers with a high crystallographic coherence and a very low interface roughness on MgO(001). Exchange coupling was evidenced by the usual unidirectional anisotropy plus an additional interfacial fourfold anisotropy. An oscillatory exchange bias angular dependence is generated by the addition of all the energy contributions.

We would like to thank J. Noguès for his fruitful comments about this work. Region Midi Pyrenées Council is also acknowledged for financial support through CTP 00004252 grant.

References

1. W.H. Meiklejohn, C.P. Bean, Phys. Rev. **105**, 904 (1957)
2. K. Takano, R.H. Kodama, A.E. Berkowitz, W. Cao, G. Thomas, Phys. Rev. Lett. **79**, 1130 (1997)
3. A.P. Malozemoff, Phys. Rev. B **35**, 3679 (1987)
4. N.C. Koon, Phys. Rev. Lett. **78**, 4865 (1997)
5. D. Mauri, H.C. Siegmann, P.S. Bagus, E. Kay, J. Appl. Phys. **62**, 3047 (1987)
6. M.D. Stiles, R.D. McMichael, Phys. Rev. B **59**, 3722 (1999)
7. J.F. Bobo, S. Dubourg, E. Snoeck, B. Warot, P. Baules, J.C. Ousset, J. Magn. Magn. Mater. **206**, 118 (1999)
8. S. Dubourg, N. Negre, B. Warot, E. Snoeck, M. Goiran, J.C. Ousset, J.F. Bobo, J. Appl. Phys. **87**, 4936 (2000)
9. S. Dubourg, J.F. Bobo, B. Warot, E. Snoeck, J.C. Ousset, Phys. Rev. B **64**, 054416 (2001)
10. J. Nogues, D. Lederman, T.J. Moran, I.K. Schuller, Phys. Rev. Lett. **76**, 4624 (1996)
11. C. Leigthon, L. Noguès, H. Suhl, I. Schuller, Phys. Rev. B **60**, 12837 (1999)
12. B. Warot, E. Snoeck, P. Baulès, J.C. Ousset, M.J. Casanove, S. Dubourg, J.F. Bobo, J. Appl. Phys. **89**, 5414 (2001)
13. C. Laï, H. Matsuyama, R. White, T. Anthony, G. Bush, J. Appl. Phys. **79**, 6389 (1996)
14. J. Nogués, I.K. Schuller, J. Magn. Magn. Mater. **192**, 203 (1999)
15. S.G.E. te Velthuis, A. Berger, G.P. Felcher, B.K. Hill, E.D. Dahlberg, J. Appl. Phys. **87**, 5046 (2000)
16. T. Ambrose, R. Sommer, C. Chien, Phys. Rev. B **56**, 83 (1997)
17. H. Xi, R. White, J. Appl. Phys. **86**, 5169 (1999)
18. A. Hoffmann, M. Grimsditch, J.E. Pearson, J. Nogués, W.A.A. Macedo, I.K. Schuller, Phys. Rev. B **67**, 220406(R) (2003)
19. E. Stoner, E. Wohlfarth, Philos. Trans. R. Soc. London Ser. A **240**, 74 (1948)
20. Z. Qian, J.M. Sivertsen, J.H. Judy, J. Appl. Phys. **83**, 6825 (1998)
21. Y. Tang, X. Zhou, X. Chen, B. Liang, W. Zhan, J. Appl. Phys. **88**, 2054 (2000)
22. R.P. Michel, A. Chaiken, C.T. Wang, L.E. Johnson, Phys. Rev. B **58**, 8566 (1998)
23. S. Dubourg, J.F. Bobo, B. Warot, E. Snoeck, J.C. Ousset, Mat. Res. Symp. Proc. **674**, T1-6 (2001)
24. M.J. Pechan, D. Bennet, N. Teng, C. Leighton, J. Nogués, I.K. Schuller, Phys. Rev. B **65**, 064410 (2002)
25. T. Mewes, H. Nembach, M. Ricart, S.O. Demokritov, J. Fassbender, B. Hillebrands, Phys. Rev. B **65**, 224423 (2002)