Structural order and magnetic anisotropy transition in Co/Fe multilayers

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Co/Fe multilayers were electron-beam evaporated in ultrahigh vacuum onto quartz substrates keeping the Co layer thickness (10 nm) constant and changing that of Fe (10–30 nm). For Fe layer thicknesses up to 24 nm, the magnetization substantially lies in the film plane and shows a uniaxial magnetic anisotropy. The coercive field measured along the easy axis is ~10 Oe, and the x-ray reflectivity patterns show a superlattice behavior. For a Fe layer thickness equal to 30 nm, the in-plane texture strongly decreases, the coercive field increases up to ~100 Oe, the magnetization direction forms an out-of-plane angle of ~36° and a stripe magnetic domain structure takes place. The observed in-plane anisotropy and the changing in the magnetic order as a function of the iron layer thickness is discussed and justified, assuming that the growth of the first Co layer occurs by the nucleation of ordered zones, influencing the subsequent layer order via exchange interaction.
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I. INTRODUCTION

Magnetic multilayers have attracted growing interest from both a technological and fundamental point of view,[1–4] and a wide research effort has been devoted to correlate the magnetic properties with the growth conditions and the architecture of the structure. Indeed, the magnetic properties of these low-dimensional systems are strongly affected by the film microstructure and thickness, grain size, surface and interface morphology.[5] The nature of the constituting elements and the deposition order are also of great importance.[6] In some cases, it has been observed that multilayers, also non-epitaxially grown, can show an in-plane uniaxial magnetic anisotropy. The causes that determine such phenomena are not well understood, several hypotheses have been advanced and specific experimental approaches have been performed in order to clarify its origin.[7]

The Co/Fe multilayers are good candidates for this kind of investigation, showing a large in-plane uniaxial magnetic anisotropy.[8] The aim of the present work has been to investigate the roles played both by the external sources and intrinsic characteristics in determining the magnetic order of the system.

II. EXPERIMENTAL PROCEDURES

Co/Fe multilayers were electron-beam evaporated in ultrahigh vacuum onto amorphous quartz substrates, and washed in acetone-trichloroethilene solution followed by an ultrasound bath in isopropylic alcohol. The starting vacuum was $\sim 10^{-8}$ Pa and the operating one was $\sim 10^{-6}$ Pa. The deposition rate was ~0.5 nm/min. The film thickness was measured during deposition by a quartz microbalance. The multilayers consisted of five bilayers of Co (10 nm) on Fe (t nm) - with $t=10$, 16, 24, and 30 - covered by a 10-nm-thick Co layer, and were labeled in the following as 5[Co10/Fe t]. A Co10/Fe30/Co10 trilayer was also deposited. For interface analyses, 5[Co5/Fe0.5] and 5[Co5/Fe2] multilayers were also grown.

Grazing incidence x-ray reflectivity (GIXRR) measurements were performed on Station 9.4 at the Synchrotron Radiation Source at Daresbury Laboratory at 298 K. Because of the very similar refractive indices of Co and Fe, an x-ray wavelength was chosen (1.743 Å) just below the Fe K edge to enhance contrast and minimizing attenuation.

Conversion electron Mössbauer spectroscopy measurements were carried out by means of a 50 mCi ^{57}Co(Rh) source with the γ-ray incidence perpendicular to the multilayer surface. A least squares minimization routine with a combination of linear and nonlinear regressions was used to fit the spectra.

The in-plane hysteresis loops and initial magnetization curves were measured at room temperature by an alternating gradient force magnetometer (AGFM) applying the magnetic field along different directions in the film plane. The magnetic domain structure was studied by magnetic force microscopy (MFM), in tapping mode at room temperature.

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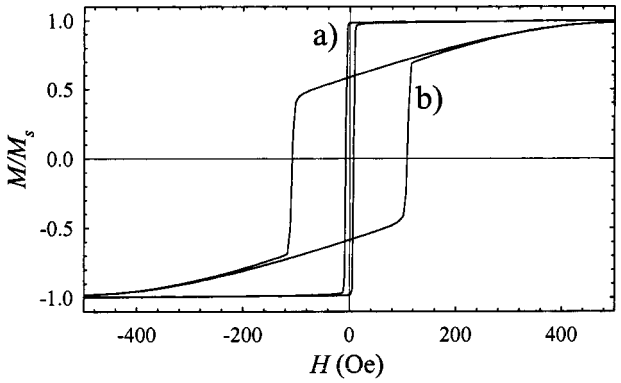

FIG. 1. Room-temperature, in-plane hysteresis loops for (a) 5[Co10/Fe24] and (b) 5[Co10/Fe30] multilayers, normalized to saturation magnetization M_s.

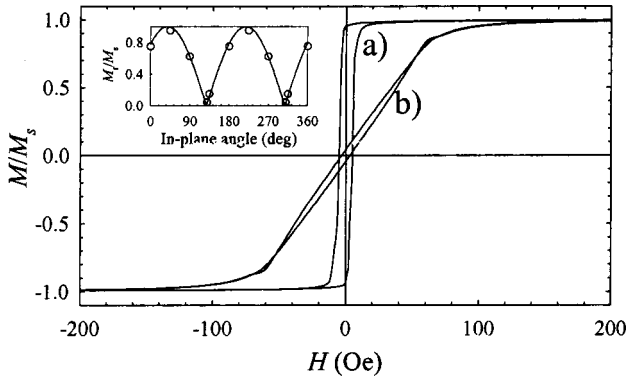

FIG. 2. Room-temperature, in-plane hysteresis loops for 5[Co10/Fe10] multilayer measured in (a) easy and (b) hard directions. Remanence ratio (M_r/M_s) dependence on the angle of the in-plane applied magnetic field direction (inset).

III. RESULTS AND DISCUSSION

The AGFM hysteresis loops for all samples show the shape expected for a single magnetic phase (Fig. 1). This suggests that a strong exchange interaction between the Co and Fe layers is present.[9]

From the line intensity ratios of the Mössbauer spectra, the angles between magnetization and film plane have been deduced. The magnetization substantially lies in the film plane for 5[Co10/Fe t], with $t \leqslant 24$ nm, forming an 8°–10° angle with respect to the film plane and rises to ~37° for 15 nm thickness. For $t \leqslant 24$ nm, the M_r/M_s values obtained applying the magnetic field parallel to the film plane at different angles, ϑ, show a 180° periodicity with a $|\sin \vartheta|$ behavior (see, for example, Fig. 2 inset), which is typical of a system having a uniaxial symmetry. The saturation magnetization M_s, and the anisotropy field H_a, deduced from the magnetization curves, allowed us to determine the anisotropy constant K_1 values ranging from 1.77 to 7.63×10^4 erg/cm^3.

On the origin of such a magnetic texture, a number of experiments have been carried out and several causes have been considered: stresses induced by sample clamping[10] and lattice mismatch,[11] magnetostatic interactions due to stepped substrates,[12,13] magnetostriction phenomena at film–substrate interface,[14] mechanical influence,[15] patterning,[16] growing incidence angle,[17,18] structural phase transition in growing layers,[19] and external magnetic field during growth.[6]

In previous works,[8,20] an in-plane uniaxial magnetic anisotropy was observed in Co/Fe multilayers grown under the same conditions, and any influence from the substrate material, the stresses induced by sample cutting, and the precipitation of metal oxides was excluded. In the present work, 5[Co10/Fe10] multilayers were simultaneously deposited on two separated substrates, in order to check the eventual influence of environmental causes such as earth and dispersed magnetic fields, or small deviations from the perpendicular incidence between the electron-beam source and substrate. Indeed, the effectiveness of these perturbations could influence in the same way the uniaxial symmetry of both films. The AGFM analysis performed on samples cut from the two multilayers, or for different positions in the same multilayer, shows that no correlation exists among the directions of their in-plane anisotropy axes [see, for example, Figs. 3(a) and 3(b)]. These results suggest that different regions (with size of the same order of the analyzed samples, i.e., a few mm^2) show different easy axis directions in the plane, probably due to different nucleation sites formed during the growth of the initial Co layer.

For Fe layer thickness from 10 to 24 nm, the coercive fields measured in the easy direction increase from ~6 to ~20 Oe. These low values can be explained on the basis of a high structural order with a very low defect density, which implies high domain wall mobility.

GIXRR measurements, show patterns typical of a superlattice structure (Fig. 4). Because of the low thickness of the samples, the signal intensities are rapidly vanishing by increasing the reflection angle. This hindered a careful determination of roughness and/or interdiffusion at the Co–Fe interfaces. In the case of 5[Co5/Fe0.5], a good quantitative fit to the data proved very difficult, presumably because of a nontrivial interface formed between Co and Fe layers. In particular, in order to reproduce the number and intensity of the fringes between the first and second superlattice reflections, it was necessary to model the Fe layers with an average roughness of the order of the layer thickness. The data show that in the limit of very thin films a well-defined layer cannot be assumed. The reflectivity values for 5[Co5/Fe0.5]

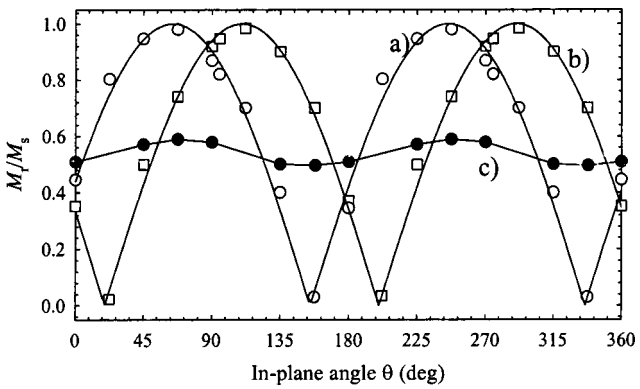

FIG. 3. Remanence ratio (M_r/M_s) vs the direction angle of the in-plane applied magnetic field for: (a) and (b) two samples cut from different positions of 5[Co10/Fe10] multilayer, and (c) 5[Co10/Fe30] multilayer.

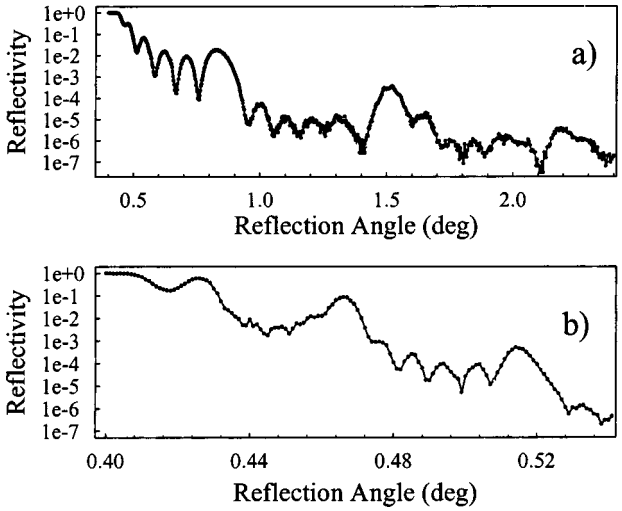

FIG. 4. Grazing incidence x-ray reflectivity patterns for (a) 5[Co5/Fe0.5] and(b) 5[Co10/Fe16] multilayers.

are a few orders of magnitude lower than those for the 5[Co10/Fe16] multilayer. In addition to the different thicknesses, this can be due to differences in interface roughness and/or interdiffusion. In agreement, the Mössbauer analysis carried out on samples grown under the same conditions[21] suggested that (i) for very thin iron layers, a Co diffusion occurs reasonably because of a high defectivity of the Fe layer, and (ii) for iron layer thickness greater than 2 nm, the Co diffusion is very low or absent thanks to the high compactness of iron.

For the 5[Co10/Fe30] multilayer, the hysteresis loop shows a single-phase magnetic behavior with a weak in-plane magnetic texture $\{M_r/M_s$ ranging from ~ 0.5 to ~ 0.6 [Fig. 3(c)]$\}$. The coercive field measured in the film plane increases up to $H_c \cong 115$ Oe. Integrating the initial magnetization curves measured along easy and hard axes, an anisotropy energy of 7.5×10^4 erg/cm^3 was calculated. The presence of an out-of-plane component determined by the Mössbauer analysis is consistent with the approach to saturation observed in the hysteresis loop reported in Fig. 1(b). In agreement, the MFM images show a well-defined stripe domain structure.

All these results are consistent with an increased morphological disorder, which determines a random arrangement of the magnetic moments, with a strong reduction of the in-plane texture, and the concomitant appearance of a significant perpendicular component of the magnetization. The increased microstructural disorder can also justify the increased H_c value.

On the other hand, the magnetization curve for the Co10/Fe30/Co10 trilayer shows a strong uniaxial magnetic texture ($M_r/M_s = 0.995$ and 0.064 for easy and hard directions, respectively), and in agreement, no evidence of stripe domains is observed. This fact suggests that, for thick Fe layers, the exchange anisotropy interaction exerted initially by Co is effective for the early layers. The addition of further layers gives rise to a loss of coherence, which determines the magnetic disorder. In fact the angular dependence of M_r/M_s for the 5[Co10/Fe30] multilayer [Fig. 3(c)] can be interpreted as the superposition of contributions from the early in-plane oriented and the subsequent isotropic layers.

IV. CONCLUSIONS

All multilayers show a single magnetic phase behavior because of the strong exchange interaction established between Co and Fe layers. Up to 24 nm Fe layer thickness, a superlattice behavior is observed. The magnetization lies substantially in the film plane and shows a uniaxial magnetic anisotropy. The origin of this in-plane anisotropy is attributed to nucleation, during the growth of the first Co layer, of various zones with different preferential orientation in the plane. These ordered Co regions influence the growth of the subsequent layers via exchange interaction. For Fe$=$30 nm, a significant out-of-plane magnetization component appears, giving rise to a stripe domain magnetic structure, and the in-plane uniaxial texture strongly decreases. These effects can be attributed to the weakening of exchange interaction as the Fe thickness and the number of layers increases.

[1] S. Yamashita, J. Yamasaki, M. Ikeda, and N. Iwabuchi, J. Appl. Phys. **70**, 6627 (1991).
[2] D. D. Awshalom and D. P. DiVincenzo, Phys. Today **48**, 43 (1995).
[3] H. Le Gall, R. Sbiaa, and S. Pogossian, J. Alloys Compd. **275–277**, 677 (1998).
[4] P. Grünenberg, Phys. Today **54**, 31 (2001).
[5] M. Carbucicchio, C. Grazzi, M. Rateo, G. Ruggiero, and G. Turilli, Nanostruct. Mater. **11**, 775 (1999).
[6] M. H. Park, Y. K. Hong, S. H. Gee, M. L. Mottern, and T. W. Jang, J. Appl. Phys. **91**, 7218 (2002).
[7] W. O. Golub, R. Gontarz, G. N. Kakazei, and N. A. Lesnik, J. Magn. Magn. Mater. **174**, 95 (1997).
[8] M. Carbucicchio, M. Rateo, G. Ruggiero, and G. Turilli, J. Magn. Magn. Mater. **242–245**, 601 (2002).
[9] G. Asti, M. Carbucicchio, M. Ghidini, M. Rateo, G. Ruggiero, M. Solzi, F. D'Orazio, and F. Lucari, J. Appl. Phys. **87**, 6689 (2000).
[10] H. Kockar and T. Meydan, Physica B **321**, 124 (2002).
[11] E. Jartych, M. Jałochowski, and M. Budzyński, Appl. Surf. Sci. **193**, 210 (2002).
[12] A. Encinas-Oropesa and F. Nguyen Van Dau, J. Magn. Magn. Mater. **256**, 301 (2003).
[13] M. C. Xu, T. Iimori, K. D. Lee, and K. Komori, Surf. Sci. **505**, 243 (2002).
[14] D. J. Twisselmann, P. G. Chambers, C. A. Ross, G. Khanna, and B. M. Clemens, J. Appl. Phys. **92**, 3223 (2002).
[15] R. Murao, C. Okuyama, K. Takahashi, A. Kikuchi, Y. Kitamoto, and S. Ishida, J. Magn. Magn. Mater. **242–245**, 352 (2002).
[16] R. Nakatani, M. Yamamoto, H. Yakame, Y. Kamada, and Y. Kawamura, J. Magn. Magn. Mater. **239**, 231 (2002).
[17] K. Itoh, F. Ichikawa, Y. Ishida, K. Okamoto, T. Uchiyama, and I. Iguchi, J. Magn. Magn. Mater. **248**, 112 (2002).
[18] A. Lisfi and J. C. Lodder, J. Magn. Magn. Mater. **242–245**, 370 (2002).
[19] S. Tacchi, F. Bruno, G. Parlotti, D. Cvetko, L. Floreano, G. Gubbiotti, M. Madami, A. Morgante, and A. Verdini, Surf. Sci. **507–510**, 324 (2002).
[20] L. Agazzi, S. Bennett, F. J. Berry, M. Carbucicchio, M. Rateo, G. Ruggiero, and G. Turilli, J. Appl. Phys. **92**, 3231 (2002).
[21] M. Carbucicchio, C. Grazzi, L. Lanotte, M. Rateo, G. Ruggiero, and G. Turilli, Hyperfine Interact. **139/140**, 553 (2002).