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Magnetic properties of Nd/Fe double layers grown on Si(111) by electron beam evaporation

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In-plane ferromagnetic resonance (FMR) has been used to study the room-temperature magnetic and crystalline properties of Nd/Fe bilayers. Several samples were grown by electron beam evaporation onto Si(111) substrates, for various Fe and Nd thicknesses. The symmetry of the resonance field as a function of the azimuthal angle departs from the sixfold one expected for a (111)Fe film. The FMR data were analyzed in the framework of a phenomenological model, which takes into account first- and second-order magnetocrystalline, uniaxial and perpendicular anisotropies. Our analysis reveals that the deviation from the sixfold symmetry is due to a combined effect of an induced uniaxial in-plane anisotropy with a small misorientation of the substrate surface from the (111) plane. We suggest that the in-plane uniaxial anisotropy may originate from an arrangement of (111) terraces separated by well oriented atomic steps formed during the small miscut. The effect of the Nd overlayer is to induce a perpendicular anisotropy which is not sufficiently large to overcome the demagnetizing field and push the magnetization entirely out-of-plane. © *1998 American Institute of Physics.* [S0021-8979(98)06108-8]

I. INTRODUCTION

In the last few years, magnetic multilayered films have been the subject of much attention because of the possibility of obtaining new magnetic properties, such as controlled interdiffusion at the interfaces, giant magnetoresistance, surface anisotropies, exchange interactions between neighboring magnetic layers, etc. The study and understanding of these properties offer the possibility of their application to new devices.[1]

Double layers of rare-earth and transition metal (RE/TM) thin films represent one of these new classes of materials, which exhibit interesting magnetic properties. The RE magnetic moments interact through an indirect exchange, according to the RKKY theory.[2] This interaction is generally one order of magnitude smaller than the exchange interaction RE-TM and two orders of magnitude smaller than the interaction TM-TM. By the other hand, many of these films exhibit strong out-of-plane anisotropy and therefore, some of these materials are potential candidate for perpendicular magnetic recording media with extremely high recording density. Due to the great lattice mismatch and the interdiffusion, the RE/TM forms a diffuse or amorphous interface in which the RE spins are randomly distributed, inducing a local anisotropy or random magnetic anisotropy (RMA).[3] Besides this, the different ordered TM-TM, TM-RE, and RE-RE pairs establish a competition, resulting in more complex and interesting magnetic phases. Although many attempts have been made to explain the magnetic anisotropies in RE/TM layers,[4–6] its origin is still a matter of controversy. Several work have been done in studying the magnetic properties of Nd/Fe, Dy/Fe, Tb/Fe, thin films grown onto glass or another kind of amorphous substrates.[7–9] These films, when grown in the form of an amorphous or polycrystalline matrix, show RMA and exhibit magnetic properties related to it.

The purpose of this work is to investigate the magnetic properties of monocrystalline Nd/Fe bilayers deposited by electron beam evaporation onto Si(111) substrates. For this, we have employed ferromagnetic resonance (FMR), which is a powerful tool to study the crystallinity and anisotropies of magnetic materials. As is well known, the resonance field value is very sensitive to properties of magnetic multilayer structures such as interlayer exchange coupling, surface induced anisotropies, as well as magnetic interactions between different elements at the interfaces. Our aim is to investigate the magnetic properties as a function of the Fe and Nd layer thicknesses. To interpret the experimental results we used a phenomenological model[10] which takes into account all relevant contributions to the free energy. From the best numerical fits to the experimental data we have extracted parameters such as in-plane and out-of-plane anisotropies and saturation magnetization.

This paper is organized as follows: in Sec. II we provide a brief description of the sample preparation and characterization methods. Section III has an outline of the theoretical model used to explain the results. In Sec. IV we present the experimental results, their interpretation and the analysis of the material parameters obtained from the theoretical fits. In Sec. V we summarize the results.

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II. EXPERIMENT

Fe/Nd bilayers were grown by electron beam evaporation onto Si(111) substrates at room temperature. The base pressure was $\sim 3\times 10^{-9}$ Torr and the deposition rate was 1 Å/s. The distance between the target and the substrate was approximately 50 cm, with the evaporated particles ejected normally onto the substrate. Two series of films were prepared. One set of Nd($t_{\rm Nd}$)/Fe(60 Å), with $0<t_{\rm Nd}<50$ Å, and another set of Nd(10 Å)/Fe($t_{\rm Fe}$), with 10 Å$<t_{\rm Fe}$ <100 Å. All samples were covered with a 25 Å Cr protective layer.

In-plane FMR measurements have been used to obtain the resonance field and linewidth as a function of the azimuthal angle. The microwave power was provided by a sweep oscillator with its frequency stabilized at the resonance of a TE_{102} rectangular cavity ($f=9.4$ GHz, $Q=2500$). The sample was mounted inside the cavity on the tip of an external goniometer that allowed us to rotate the plane of the film with respect to the field, which was provided by a 9″ VARIAN electromagnet. The in-plane external field was modulated by Helmholtz coils at 1 kHz. The detected signal corresponds to the derivative of the absorption lines with approximately Lorentzian line shapes. All measurements were carried out at room temperature.

III. THEORETICAL MODEL

The phenomenological model used to interpret the FMR measurements is based on theoretical considerations presented previously.[10] Here we summarize the main aspects of the model. The ferromagnetic resonance frequency is expressed in terms of the second derivatives of the free-energy density as[11]

$$\left(\frac{\omega}{\gamma}\right)^2=\frac{1}{M^2\sin^2\theta}\left[\frac{\partial^2E}{\partial\theta^2}\frac{\partial^2E}{\partial\varphi^2}-\left(\frac{\partial^2E}{\partial\theta\partial\varphi}\right)^2\right]_{\theta_0,\varphi_0}, \quad (1)$$

where ω is the angular frequency, γ is the gyromagnetic ratio, M is the saturation magnetization, and θ and φ are the polar and azimuthal angles of the magnetization vector, respectively. The expression (1) must be taken at the equilibrium positions of $\mathbf{M}$ determined by the condition $\partial E/\partial\theta=\partial E/\partial\phi=0$. The model takes into account the following contributions for the free-energy density

$$E=-\mathbf{H}\cdot\mathbf{M}+K_1(\alpha_1^2\alpha_2^2+\alpha_2^2\alpha_3^2+\alpha_1^2\alpha_3^2)+K_2\alpha_1^2\alpha_2^2\alpha_3^2 +2\pi(\mathbf{M}\cdot\mathbf{n})^2+K_u\sin^2(\varphi-\varphi_u)-K_n(\mathbf{M}\cdot\mathbf{n}/M)^2, \quad (2)$$

where the six terms are, respectively: the Zeeman energy; first- and second-order magnetocrystalline cubic anisotropy terms, where K_1 and K_2 are the anisotropy constants, α_1, α_2, and α_3 are the director cosines of the magnetization with respect to the principal cubic axes [100], [010], and [001]; the fourth term is the demagnetizing energy; and the last two terms are the uniaxial in-plane and perpendicular anisotropy energies, where K_u and K_n are the corresponding anisotropy constants, φ is the direction of the magnetization in the film plane, φ_u is the angle of the in-plane uniaxial anisotropy field, and $\mathbf{n}$ is the normal to the film plane. We also define an effective magnetization $4\pi M_{\rm eff}=4\pi M-2K_n/M$.

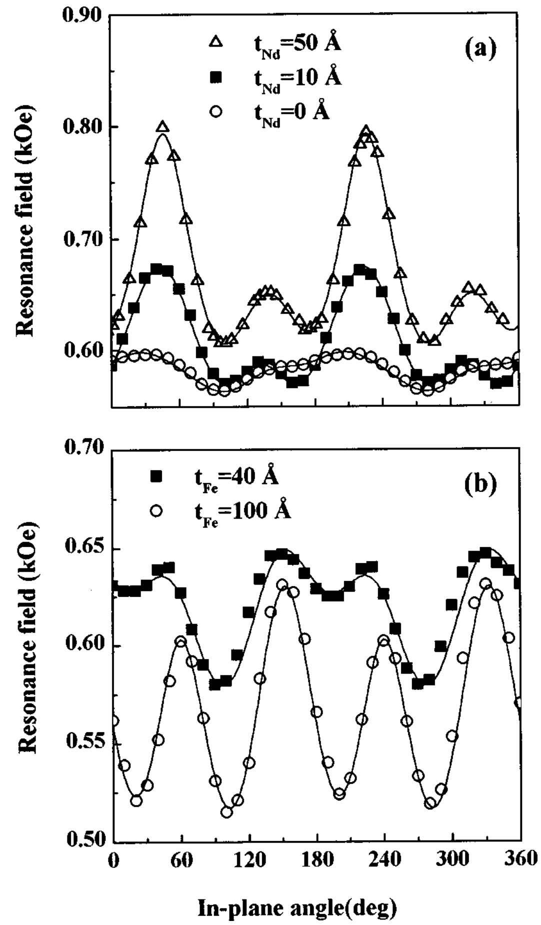

FIG. 1. In-plane angle dependence of the resonance field for (a) Nd($t_{\rm Nd}$)/Fe(60 Å)/Si(111) and (b) Nd(10 Å)/Fe($t_{\rm Fe}$)/Si(111). The solid lines are theoretical fits with the parameters shown in Table I.

The calculation is based on minimization of the free-energy density (2) to determine the equilibrium direction of the magnetization for each orientation of the applied field. A least-squares fitting of the FMR data with (1) is done in order to obtain the sample parameters. According to Ref. 10, the effect of small misorientations of the substrate surface with respect to the crystal plane can destroy the sixfold in-plane symmetry of the pattern of a (111)Fe film. In order to take into account this misorientation, we introduced in the calculation a small angle β between the [111] film orientation and the substrate normal.

IV. RESULTS AND DISCUSSION

A. Resonance field and linewidth

The symbols in Fig. 1(a) show the in-plane angle dependence of the resonance field for the films of the series Nd($t_{\rm Nd}$)/Fe(60 Å)/Si(111). One can clearly see that the sixfold symmetry expected for a single-crystal (111)Fe film is absent. As previously shown,[10] the sixfold symmetry is completely overwhelmed by a small misorientation, β, between the normal to the plane of the film and the [111] direction. This misorientation results from the current practice in mi-

TABLE I. Parameter values obtained from the best fits for the experimental data of the series Nd(t_{Nd})/Fe(60 Å)/Si(111) and Nd(10 Å)/Fe(t_{Fe})/Si(111).

Plane	t_{Fe}(Å)	t_{Nd}(Å)	$4\pi M_{eff}$ (kOe)	$2K_1/M$ (kOe)	$2K_u/M$ (kOe)	K_2/K_1	β(deg)
		0	17.48	0.300	0.013	0.0	0.9
(111)	60	10	17.0	0.50	0.050	0.05	1.8
		50	15.1	0.55	0.085	0.05	3.0
(111)	10	40	16.45	0.38	0.021	0.05	−1.6
		100	18.20	0.54	0.025	0.06	2.9

croelectronics of cutting the Si wafer in planes slightly tilted from (111). This procedure is used to stabilize the reconstruction of the Si(111) surface and so the epitaxial growth.[12,13] Figure 1(a) also shows that the thicker the Nd overlayer, the larger are the variations in the resonance field with the field direction. This is associated with an induced in-plane hard axis for the magnetization. A similar behavior is seen when the Nd layer is kept fixed and the Fe layer thickness is increased, as shown in Fig. 1(b) for the samples Nd(10 Å)/Fe(40 Å)/Si(111) and Nd(10 Å)/Fe(100 Å)/Si(111). The solid lines in Fig. 1 are theoretical fits to the experimental data. The values of the parameters used in these fits are listed in Table I. Excellent agreement with the FMR data could be found for realistic sets of the sample parameters, under the condition $\beta \neq 0$. This demonstrates that the films are crystalline, with the [111] direction slightly tilted with respect to the normal of the film. In particular, the value $2K_1/M = 0.30$ kOe obtained for the uncovered Fe(60 Å) layer is very close to the value 0.34 kOe measured for Fe(50 Å)/Si(111).[10]

The observed linewidth for the Fe(60 Å) film was approximately 37 Oe, close to the best values observed for Fe and other transition metal films,[14,15] indicating the good crystallinity of the samples. Figure 2 shows the variation of the linewidth versus in-plane angle for Nd(10 Å)/Fe(100 Å)/Si(111). The linewidth behaves similarly to the resonance field, being maximum at the hard axis and minimum at the easy magnetization axis. Heinrich *et al.*[16,17] reported a similar behavior in Ni(100)/Fe(100)/Ag(100) thin films.

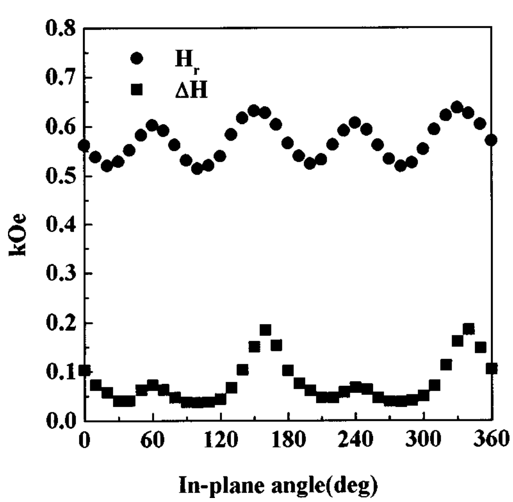

FIG. 2. Linewidth and resonance field vs in-plane angle for Nd(10 Å)/Fe(100 Å)/Si(111). The maximum of ΔH occurs at the axes of hard magnetization and the minimum occurs at the easy axes.

B. Anisotropy constants

Figures 3(a) and 3(b) show the thickness dependence of K_1 and K_u for the series Nd(10 Å)/Fe(t_{Fe})/S(111) and Nd(t_{Nd})/Fe(60 Å)/Si(111), respectively. For the first series, in which the Nd layer thickness is kept constant and $0 < t_{Fe} \leqslant 100$ Å, the first-order cubic anisotropy field ($2K_1/M_s$), increases with the Fe layer thickness until reaches the bulk value for iron, as expected. The continuous line in Fig. 3(a)

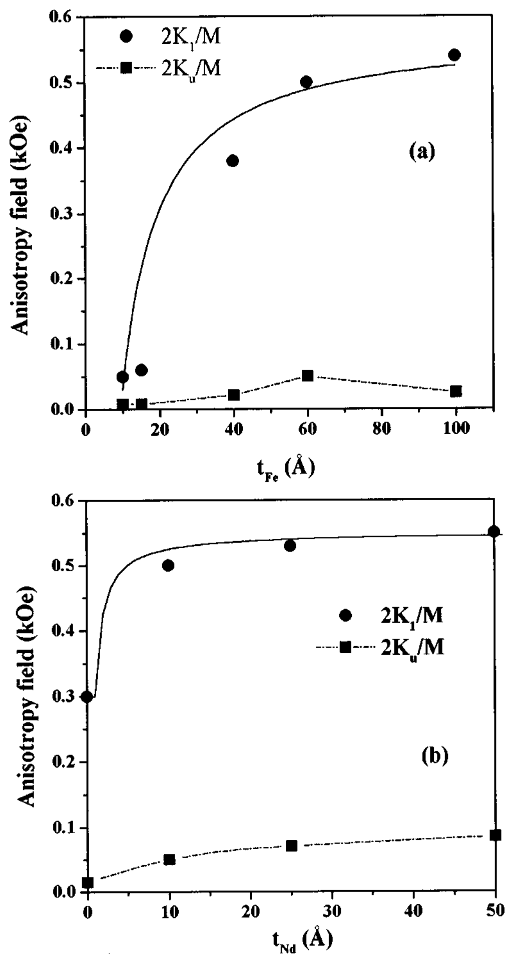

FIG. 3. Thickness dependence of the anisotropy constants K_1 and K_u for (a) Nd(10 Å)/Fe(t_{Fe})/Si(111) and (b) Nd(t_{Nd})/Fe(60 Å)/Si(111). The solid lines are fits using a constant plus a term proportional to $1/t$. The dashed lines are guides to the eyes.

is a fit according to the expression $2K_1/M=(0.58–5.5\ \text{Å}/t_{Fe})$ kOe,[17] where the first term is the bulk value of the magnetocrystalline constant and the term proportional to $1/t_{Fe}$ is the surface contribution to this anisotropy. For the second series of samples, in which the Fe layer thickness is kept constant and $0<t_{Nd}\leqslant 50\ \text{Å}$, the magnetocrystalline anisotropy constant shows qualitatively the same behavior with respect to t_{Nd}, increasing up to a saturation value of about 550 Oe. Here again the dependence of $2K_1/M$ on the Nd thickness can be described by the bulk magnetocrystalline constant plus a term proportional to $1/t_{Nd}$. The continuous line in Fig. 3(b) shows a fit according to $2K_1/M=(0.55–0.25\ \text{Å}/t_{Fe})$ kOe. The uniaxial anisotropy, $2K_u/M$, exhibits a nonlinear behavior with respect to the Fe layer thickness, characterized by a bell-shaped curve having a maximum at $t_{Fe}\approx 60\ \text{Å}$. It also presents a nonlinear behavior with respect to t_{Nd}, increasing by about 40 Oe as t_{Nd} increases from 0 to 10 Å, and remains approximately constant for $t_{Nd}>10\ \text{Å}$. This suggests that the Nd overlayer acts mainly in the interface region. It is known that a vicinal cut in a (111) surface leads to an arrangement of oriented terraces separated by stepped regions[18,19] and these can induce an uniaxial anisotropy in cubic or tetragonal films.[20] So we suggest that this in-plane uniaxial anisotropy can be originated from an arrangement of (111) terraces separated by atomic steps, formed during surface reconstruction due to the small miscut of angle β. As a consequence of our data-fitting analysis, a second-order magnetocrystalline anisotropy constant, K_2, of the order of 5% of the value of K_1, must be taken into account for all samples except for the uncovered Fe (60 Å) film.

C. Effective magnetization

The effective magnetization exhibits the expected dependence as a function of the Fe layer thickness, as shown in Fig. 4(a). It increases gradually with the Fe layer thickness reaching a value of about 18 kOe at $t_{Fe}=100\ \text{Å}$ still below the saturation value of 21.5 kOe. Quite different behavior is observed in the measured $4\pi M_{eff}$ vs t_{Nd} as shown in Fig. 4(b). The effective magnetization is reduced as the Nd layer thickness increases. Based on the previous definition of $4\pi M_{eff}$, this means that the Nd layer induces a perpendicular anisotropy, $2K_n/M$, which increases linearly with t_{Nd}. Although it reaches a value of $\sim$2.0 kOe, it is not sufficiently large to overcome the demagnetizing field and thus to push the magnetization entirely out-of-plane.

V. CONCLUSIONS

We have presented and discussed the magnetic properties of Nd/Fe bilayers grown by e-beam evaporation on Si(111). The FMR data reveal that the magnetic properties are due to an interface effect and a small misorientation of the substrate plane with respect to the [111] direction. These combined effects completely destroy the sixfold symmetry of the films, leading to a twofold symmetry, as shown by a theoretical analysis. The in-plane uniaxial anisotropy is induced by an arrangement of well ordered terraces formed during the miscut. The Nd layer induces a perpendicular anisotropy, which is not sufficiently large to overcome the demagnetizing field and push the magnetization entirely out-of-plane.

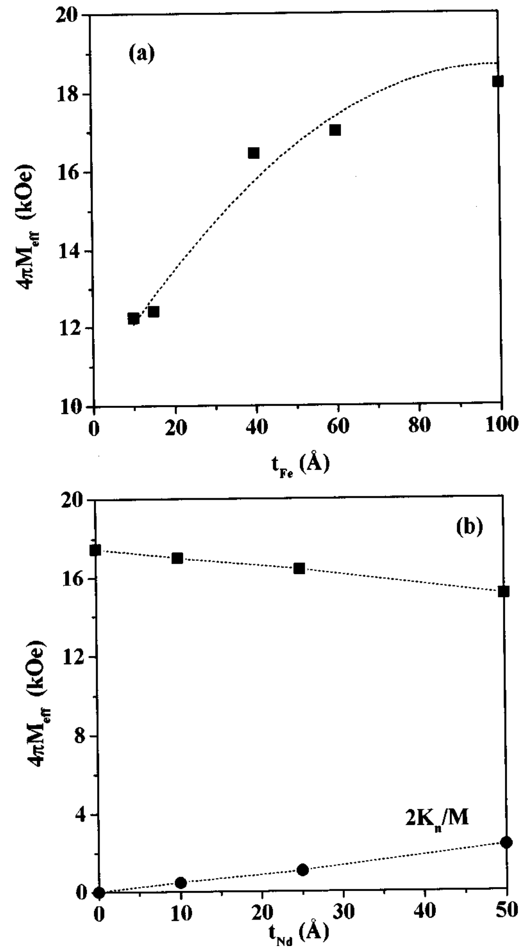

FIG. 4. Thickness dependence of $4\pi M_{eff}$ defined in the text, for (a) Nd(10 Å)/Fe(t_{Fe})/Si(111) and (b) Nd(t_{Nd})/Fe(60 Å)/Si(111). The dashed lines are to guide the eye. The solid circles represent the perpendicular anisotropy constant, $2K_n/M_s$.

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