Magnetic behavior of sputtered Gd/Co multilayers

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We have experimentally and theoretically explored the magnetic behavior at low field of Gd/Co sputtered multilayers. Magnetization measurements and simulations qualitatively agree only when the Gd layer is substituted by an alloyed GdCo layer close to the eutectic composition. The formation of this GdCo alloy, previously evidenced by experiments of composition depth profiles, is consistent with the observed compensation temperatures. Likewise, a certain reduction of the effective Co layer thickness due to alloy formation is taken into account and improves the agreement between theoretical calculations and measurements of magnetization versus temperature for various multilayers.

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I. INTRODUCTION

Magnetic layered systems exhibit fascinating properties which have attracted significant attention. This is not only due to their fundamental interest, but also because of their applications in magnetic storage devices and sensors. Phenomena such as giant magnetoresistance and interlayer magnetic coupling have aroused a great theoretical interest and opened new ways for the design of novel devices. In particular, magnetic multilayers with antiferromagnetic (AF) coupling are attractive systems to study interface coupling, magnetic transitions induced by an applied magnetic field (spin flop), and present a nontrivial behavior of magnetization versus temperature.

The periodic stacking of two distinct ferromagnetic materials with antiparallel or AF interlayer coupling gives rise to a variety of magnetic configurations.[1] Multilayers composed of a rare-earth (RE) element, such as Gd, and a transition metal like Fe or Co are an interesting example of such systems. Due to their very different ordering temperatures, complex magnetic configurations depending on the structural parameters, temperature, and magnetic field may occur.[2–8]

Gd/Fe multilayers have been more extensively investigated. The first experimental study of this system was presented by Morishita *et al.*[3] The possibility of having a compensation point for certain layer thicknesses stimulated much experimental[9–11] as well as theoretical work in this system: Camley and Tilley[2,12] developed a model to study ideal Gd/Fe multilayers, showing the existence of different magnetic phases as a function of the magnetic field, temperature, and structural parameters. If a magnetic field is applied parallel to the layers, they found that below the Curie temperature of Gd three main phases appear: the Gd-aligned phase, where Gd magnetic moments are parallel to the magnetic field and Fe moments are antiparallel; the aligned-Fe phase, with Fe moments in the field direction and Gd moments antiparallel to it; and the twisted or canted phase, where Fe and Gd moments are away from the field direction. Comparison of the magnetization dependence with the temperature of real samples and theoretical calculations was first reported by Motokawa[5]; later works comparing experiments and simulations[4,6,13–15] confirmed the suitability of the mean-field approach to account for the magnetic behavior of such systems.

Gd/Co multilayers have not been so thoroughly studied.[7,8,16,17] They are far from ideal samples with abrupt interfaces: in a recent work[18] the existence was shown of strong asymmetric interdiffusion that consequently leads to severe Co alloying throughout the Gd layer up to a composition near the eutectic point ($Gd_{0.63}Co_{0.37}$). Interestingly, it seems that when the eutectic composition is reached, further alloying is prevented, and the resulting composition profile turns out to be rather squared, composed of alternating Co and $Gd_{0.63}Co_{0.37}$ alloy layers. The alloy layer is clearly amorphous as a consequence of the strong amorphization reaction previously noticed by Hufnagel *et al.*[19] As to the magnetic behavior of the alloy layer, it was also shown[18] that the eutectic alloy layer is a $4f$ ferromagnet with a spontaneous magnetization at 4 K of about 1660 emu/cm^3 and a Curie temperature of $T_C = 180$ K.

In this article, we compare magnetization measurements with theoretical calculations which support nominal Gd/Co multilayers being actually composed of a thinned pure Co layer and an alloy layer of composition close to that of the eutectic alloy ($Gd_{0.63}Co_{0.37}$). This alloy is ferromagnetic with a Curie temperature around 180 K, i.e., 100 K lower than that of pure Gd, thus lowering the multilayer compensation temperature with respect to the expected value for an ideal Gd/Co system. To attain a qualitative agreement of the overall temperature dependence of the magnetization, we had to assume a reduced effective thickness of the Co layers with respect to the nominal value.[20] Such thinning of Co layers was verified with x-ray reflectivity measurements of the multilayers.

II. EXPERIMENT

Samples were grown in a multitarget sputtering system equipped with pure Co (99.99%) and Gd (99.9%) targets on unheated glass substrates. The background pressure was

around 4×10^{-7} torr. rf sputtering was carried out in pure argon (2.5 mtorr). Growth rates were 5 Å/s and 1.4 Å/s for Gd and Co, respectively; these values were obtained from thickness calibration of single films. Conventional x-ray diffraction $\theta/2\theta$ scans using Cu K_α radiation were made for structural determination. The magnetization was measured using a vibrating sample magnetometer (VSM) with applied fields up to 15 kOe in the film plane from 80 K to 300 K. Hysteresis curves of the samples were obtained using the transverse magneto-optic Kerr effect (T-MOKE), with ac magnetic fields in the film plane (up to 130 Oe) from 10 K to 300 K. Low-angle x-ray reflectivity demonstrated a chemical modulation perpendicular to the film plane in all multilayers. Auger electron spectroscopy (AES) under 5 keV electron irradiation combined with ion bombardment (Ar+ $\sim$80 Å/h) was used for depth analysis of the film composition. It indicated a strong asymmetric Co diffusion into the Gd deposit to build an eutectic alloy layer and, consequently, a nearly squared profile. Bulk composition measurements were done in a scanning electron microscope with energy-dispersive x-ray (EDX) microanalysis.

III. MODEL AND CALCULATIONS

We use a modified version of the molecular field model proposed by Camley and Tilley[2,12] with appropriate parameters for Co, Gd, and the GdCo alloy. To set such parameters, we suppose the multilayer is made of alternating hcp slabs of material A (Gd or GdCo alloy) and material B (Co) with abrupt interfaces. Each slab of material X is composed of N_X atomic layers. We model one atomic layer i by its magnetic moment M_i which can rotate freely in the plane parallel to the interfaces. This means that we neglect the out-of-plane magnetization in the slabs composing the bilayer, which is a reasonable assumption due to the strong shape anisotropy. To mimic the periodicity in the growth direction, we consider a slab of N_A+N_B two-dimensional (2D) magnetic moments with periodic boundary conditions. The 2D magnetic moment M_i is given by

$$M_i=\mu_B g n_i\langle S_i\rangle, \tag{1}$$

where μ_B is the Bohr magneton, g is the Landé factor, n_i is the atomic areal density of layer i, and $\langle S_i\rangle$ is the thermal-averaged value of the spin in layer i. This latter quantity is calculated within the mean-field approximation, i.e., by employing the Brillouin function $B_S(x)$:

$$\langle S_i\rangle=S_i B_S(x), \tag{2}$$

where $x=g\mu_B S_i H_i/k_B T$. Here H_i is the effective magnetic field in layer i, k_B is the Boltzmann constant, and T is the temperature. We only consider exchange interactions between nearest-neighbor layers; thus, the effective magnetic field in layer i is

$$H_i=(J_{i,i+1}\langle S_{i+1}\rangle+J_{i,i-1}\langle S_{i-1}\rangle)/g\mu_B+H_{ext}, \tag{3}$$

where J_{ij} is the exchange coupling interaction between layers i and j and H_{ext} is the external applied magnetic field. Therefore Eqs. (2) and (3) are to be solved self-consistently. For the cases considered in this work the external field is much smaller than the molecular field, so it can be neglected for the calculation of $\langle S_i\rangle$.

When a finite external field H_{ext} is applied, the relative orientation of the magnetic field and the spins in each layer is obtained by minimizing the sum of the exchange energy E_{ex} and the Zeeman energy E_Z:

$$E_{ex}+E_Z=-\frac{1}{2}\sum_i (n_i J_{i,i+1}+n_{i+1}J_{i+1,i})\langle S_i\rangle\langle S_{i+1}\rangle\cos(\theta_i - \theta_{i+1})-g\mu_B n_i\langle S_i\rangle H_{ext}\cos\theta_i, \tag{4}$$

where θ_i is the angle between the orientation of the external manetic field and the averaged value of the spin $\langle S_i\rangle$ in the ith layer.

The minimization procedure is as described by Camley and Tilley[2,12]: (i) A random spin configuration is generated; (ii) a spin i of the configuration is chosen randomly and rotated into the direction of the effective field H_i; (iii) the last step is repeated until a self-consistent spin configuration is obtained. As this procedure only minimizes locally the energy, a large number of initial configurations generated randomly is explored to ensure that the ground state is found. Then the magnetization in the field direction is calculated as

$$M(T)=\sum_{i=1}^{N_A+N_B} g\mu_B n_i\langle S_i\rangle\cos\theta_i/(N_A d_A+N_B d_B), \tag{5}$$

which we compare to the experimental values.

To set the parameters appearing in the former equations, we first discuss the case of pure-Gd and- Co multilayers. We assume the slabs are made of hcp-Co and hcp-Gd bulk metals grown in the (001) direction. This assumption is based on the following fact: we have grown Gd and Co thin films using the same sputtering conditions employed in multilayers and we have observed that both Co and Gd films are highly textured with their c axis perpendicular to the substrate. In the model we employ this is just a simple means of setting the structural parameters. Thus, the atomic layers are separated a distance $d=c/2$, i.e., $d_{Gd}=2.8925$ Å and $d_{Co}=2.035$ Å, and the areal densities are $n_{Gd}=0.874\,57\times10^{15}$ atom/cm^2 and $n_{Co}=1.832\,83\times10^{15}$ atom/cm^2.

As for the magnetic parameters, we take the atomic values of the spins, $S_{Gd}=7/2$ and $S_{Co}=2$. The interaction exchange constants employed in the model are given by $J_{Gd}=1.2\times10^{-15}$ erg, $J_{Co}=4.8\times10^{-14}$ erg, and for the interface coupling we have chosen a value between the exchange couplings J_{Gd} and J_{Co}, $J_{\rm int}=-2.1\times10^{-15}$ erg. The bulk exchange couplings are set to reproduce the bulk Curie temperatures of Gd and Co when modeling a multilayer with both slabs of the same material.

With respect to the eutectic alloy, we note that for that composition the Co atoms are not magnetic. Therefore we chose to model it as if it were Gd with lower density, $N_{eut}=0.85\,n_{Gd}$. The factor 0.85 is the volume fraction of Gd atoms in the eutectic alloy. The coupling for the eutectic alloy is $J_{Gd_{0.67}Co_{0.33}}=5.5\times10^{-16}$ erg, which as before, was chosen to reproduce the Curie temperature $T_C=180$ K.

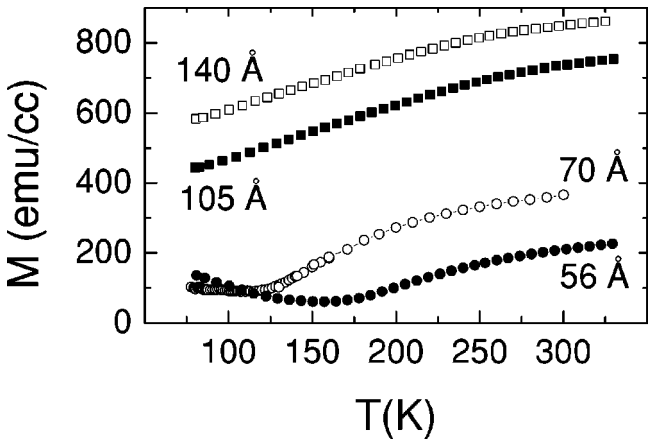

FIG. 1. Magnetization at 300 Oe measured as a function of temperature in a series of Gd(50 Å)/Co(X) multilayers of 20 repeats for different nominal thicknesses of the Co layer (X). Nominal cobalt thicknesses are $X=56$ Å (●), 70 Å (○), 105 Å (■), and 140 Å (□).

IV. MAGNETIZATION MEASUREMENTS AND ANALYSIS OF Gd (50 Å)/Co(X) MULTILAYERS

Figure 1 presents a plot of the measured magnetization as a function of temperature in a set of Gd(50 Å)/Co(X) for different nominal Co layer thicknesses (t_{Co}). Some curves show the characteristic compensation temperature (T_{comp})—a signature of antiferromagnetic coupling between the layers—below which the magnetization of the Gd layer dominates over that of the Co layer. As already observed by other authors,[14,2] for Gd thicknesses (t_{Gd}) smaller than a certain t_{Co} value the compensation point disappears and Co magnetization is dominant for all temperatures. We have chosen the present range of values to study the crossover between these two behaviors, where a small variation in a structural parameter may produce a qualitative change in the magnetic properties of the sample, thus being an interesting system to assess the validity of our suppositions. Such a sensitive behavior is also observed in the simulations; however, calculated curves with pure Gd layers (Fig. 2) show marked differences with respect to the measured curves. In particular, the compensation point, when present, appears at much higher temperatures.

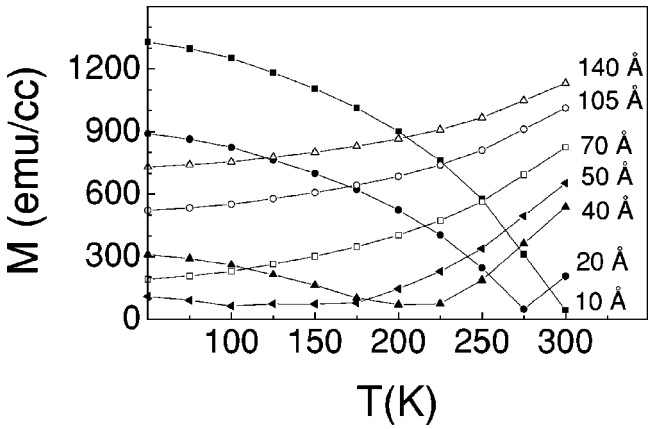

FIG. 2. Low-field magnetization calculated as a function of temperature in a series of Gd(50 Å)/Co(X) multilayers modelled with pure Gd and Co layers. Co thicknesses are displayed over the curves.

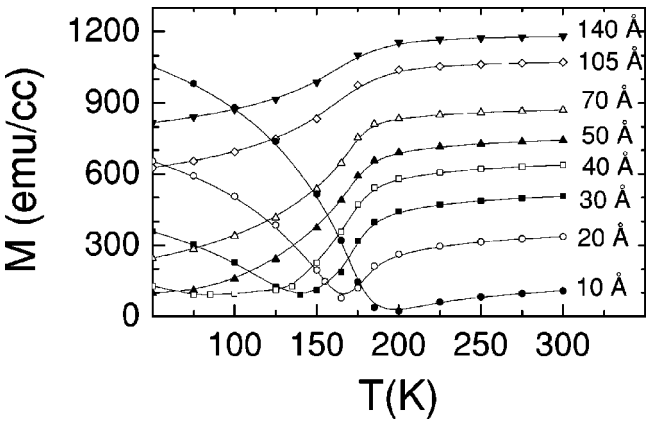

FIG. 3. Low-field magnetization calculated as a function of temperature in a series of $Gd_{0.63}Co_{0.37}$ (50 Å)/$Co(X)$ multilayers with eutectic Gd alloy (Curie temperature $T_C=180$ K). Co thicknesses are displayed over the curves.

But as we mentioned in Sec. II, elemental depth profiles obtained by AES show that our Gd/Co samples are actually comprised of Co and GdCo alloy layers with composition close to the eutectic point ($Gd_{0.63}Co_{0.37}$). As this alloy has lower Curie temperature, a substantial reduction in the compensation temperatures of GdCo/Co multilayers with respect to those of pure Gd/Co is to be expected, and we have explored this theoretically. In Fig. 3 we show the $M(T)$ calculated curves in a series of multilayers with the same thickness of GdCo eutectic alloy layer.

A straightforward comparison of compensation temperatures in measurements and simulations is not satisfactory for the same nominal thickness of Co. However, strong evidence indicates that a reduction of the effective Co thickness is necessary: we plot in Fig. 4 the measured layer magnetiza-

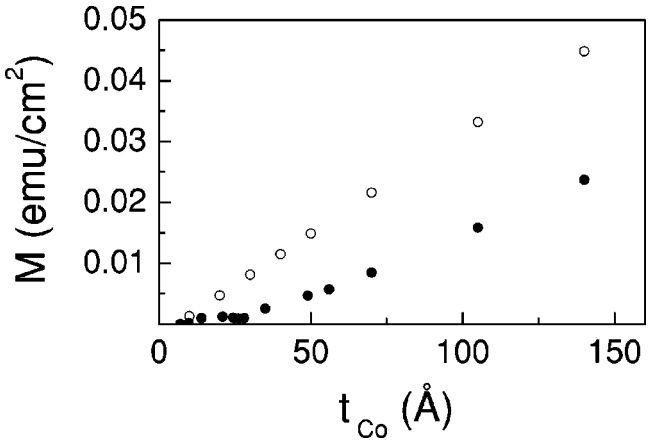

FIG. 4. Magnetization (300 Oe) at room temperature as a function of the Co layer thickness (solid circles). Open symbols (○) indicate the calculated magnetization with $Gd_{0.63}Co_{0.37}$ multilayers. Note that a linear slope appears above a certain thickness (25–30 Å); see text.

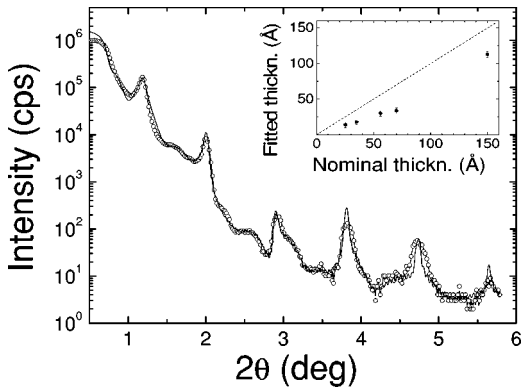

FIG. 5. Grazing x-ray reflectivity scan as a function of 2θ angle for a multilayer nominally Gd(50 Å)/Co(56 Å) (dots) and the corresponding simulation (solid lines). The inset displays the Co thickness inferred from the fits for the series Gd(50 Å)/Co(X).

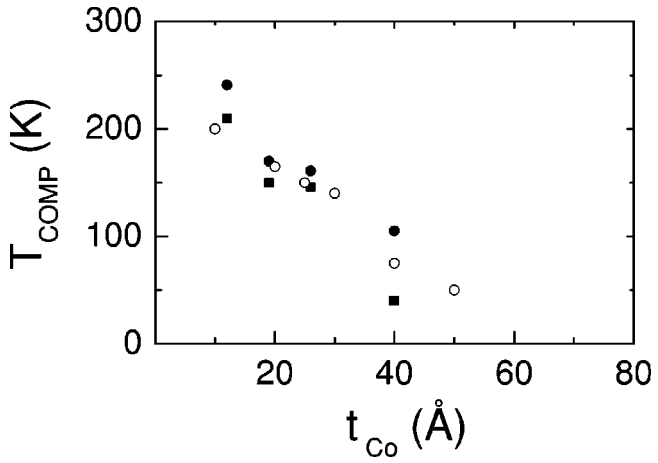

FIG. 6. Compensation temperatures from calculations of $Gd_{0.63}Co_{0.37}$ (50 Å)/Co(X) multilayers with eutectic Gd alloy (○) and experimental values from low-field magnetization of Gd(50 Å)/Co(X) (●) and from the magneto-optic Kerr effect of the same samples (■).

tion of the same series of multilayers at room temperature (300 K) together with the calculated values for saturated Co layers of the nominal thickness. We have chosen 300 K because at this temperature, higher than the compensation point, the total magnetization is expected to be only that of the Co layers. Clearly the theoretical points fall along a straight line, but the experimental points begin to show a linear behavior above 25–30 Å. This can be considered as a reduction of the Co thickness from the magnetic point of view, so in our calculations we should consider a reduction of the Co magnetic layer thickness to find an agreement with experiment. If we now take such a reduction into account and compare the experimental magnetization of samples Gd(50 Å)/Co(X) (Fig. 1) to calculated magnetization curves of multilayers $Gd_{0.63}Co_{0.37}$(50 Å)/Co($X-30$ Å) (Fig. 3), very good overall qualitative agreement is found, which is even quantitative as to the compensation temperatures.

In order to investigate such thinning effects in the Co layers, we have analyzed the x-ray reflectivity scans to infer the actual Co layer thickness. This has been accomplished with simulations using a standard code[21] and a model structure consistent with AES profiles. Figure 5 contains typical reflectivity scans and the simulations. Note the existence of Bragg peaks up to sixth order. The inset displays the fitted values of the Co thickness inferred from simulations as a function of nominal values. Interestingly, every multilayer is characterized by Co layers whose thickness is reduced a substantial amount (15–28 Å) compared to nominal values.

As to an explanation for the reduced Co thickness, our data point to a twofold origin. First, alloying of the Gd layers must have reduced the actual Co thickness from the nominal values to a certain extent.[20] However, from density data,[22] we estimate that in a Gd(50 Å)/Co(X) multilayer the decrease in the nominal Co thickness is not more than 10 Å. This value compares too low with the measured reduction of 15–28 Å. Second, interdiffusion may have formed a wide interface. In fact, elemental AES profiles provided an upper estimate of about 2 nm. A detailed study of the magnetic interface is underway. However, the present combined effects can explain the effective Co layer thickness.

With respect to the observed quantitative discrepancies between simulations and experiments, some comments are due. First, experimental values of M_S(300 K) of Gd/Co nibikater (ML) samples are much below the calculated ones. Besides, we have grown by sputtering pure Co single films and measured $M_S(300\ \mathrm{K}) = 1130$ emu/cm^3, 22% lower than the expected value. This has been observed by other groups, and it is supposedly related to a reduced mass density and/or an incomplete saturation arising from the sample growth. Similar values are obtained in multilayers in which a thin Cr layer was inserted at the interface in order to avoid interdiffusion.[18] As our purpose with the simulations was to corroborate the key role played by the change of composition and the reduced effective Co thickness, we chose not to modify the rest of parameters in the calculations, in order not to mask the importance of the ones studied here. Obviously the introduction of more adjustable parameters will improve numerical agreement, but this is not our main focus in the present work.

Second, the observed rounding of experimental $M(T)$ around T_{comp} is not reproduced in the simulation. We think this must be related to the details of the wide interface region. As stated in Sec. III, we have modeled an abrupt interface profile, having in mind that the main role of this region is to produce an AF coupling between Gd and Co slabs. We expect as well that modeling an interface with a smooth profile, when more detailed data of the interface region are available, may account for the smooth shape of the experimental curves.

Finally, we find that the $M(T)$ curves are fairly reproduced with this model except for samples with very thin Co layers. For these samples large quantitative discrepancies occur at temperatures much lower than the T_{comp}, where experimental values are far lower than that of the simulation ''Gd-aligned'' phase. This difference increases with reducing Co layer thickness. From Auger spectroscopy we know that very thin Co layers (nominally 10 Å) are oxidized; this probably reduces the spontaneous magnetization at low temperature (GdO_2 is a paramagnet).

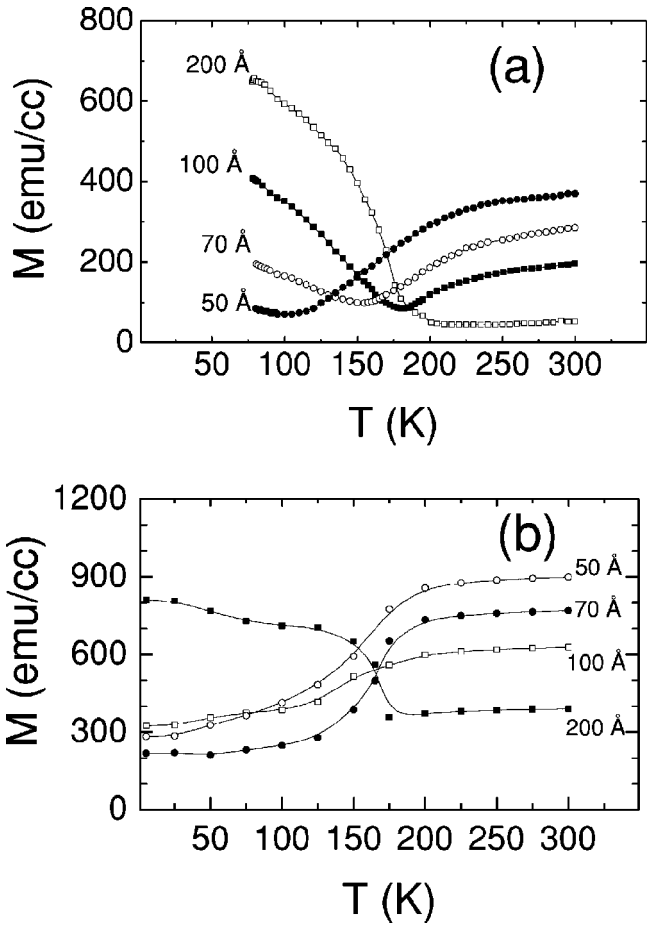

FIG. 7. (a) Experimental low-field magnetization of a series of Gd(X)/Co(70 Å) multilayers as a function of temperature. The Gd layer thickness ranges from 50 to 200 Å. (b) Calculated magnetization as a function of temperature in a series of Gd(X)/Co(70 Å) multilayers with pure Gd layers of the same thickness values.

As a summary, in Fig. 6 we show the experimental values of T_{comp} from VSM measurements and from the magneto-optic Kerr effect, as well as calculated values. It can be seen how this simple model accurately predicts the compensation temperatures once the reduced Co thickness and formation of the GdCo eutectic alloy are considered.

V. MAGNETIZATION MEASUREMENTS AND ANALYSIS OF Gd(X)/Co(70 Å) MULTILAYERS

In this section we study the magnetization of a series of multilayers with fixed Gd thickness in a range of Co thickness where compensation behavior is observed. Figure 7(a) shows the experimental results for a set of multilayers with a nominal Co thickness of 70 Å. In three of the four cases displayed there is a clear compensation point, all below 200 K. This can be explained by assuming the existence of GdCo alloy instead of pure Gd in the samples, like for the results presented in the previous section. To confirm this last assumption, we have modeled the magnetization versus temperature for pure Gd/Co multilayers with the same nominal thicknesses of the Gd layer; the curves are shown in Fig. 7(b). Only one of the simulated samples shows a clear compensation below room temperature. Another important disagreement is the behavior for the sample with the thickest Gd layer: the experimental magnetization curve of Gd(200 Å)/Co(70 Å) [Fig. 7(a)] is rather flat from 200 to 300 K, whereas the corresponding theoretical curve in Fig. 7(b) still has a strong temperature dependence in the same range. This indicates that the nominal Gd layer has a Curie temperature (180 K) much lower than that of pure Gd and in agreement with the Curie temperature of the eutectic alloy $Gd_{0.63}Co_{0.37}$. Thus the Curie point of multilayers with large Gd thicknesses can serve as a check of the alloy formation by means of Co interdiffusion. A substantial reduction of the Curie point within the Gd layers of Gd/Fe multilayers has also been reported,[23] without indicating a possible origin; in this work we give an explanation of such behavior in a similar system. On the other hand, at room temperature we have also observed the same quantitative discrepancy for the saturation magnetization as in the series with fixed Gd layer thickness (Sec. IV).

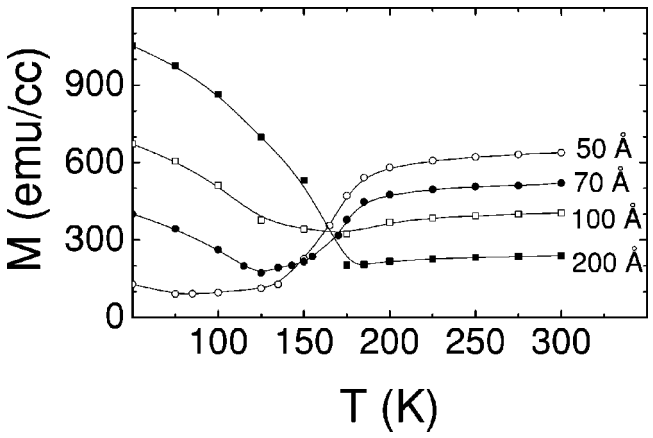

FIG. 8. Calculated magnetization as a function of temperature in a series of $Gd_{0.63}Co_{0.37}$ (X)/Co(40 Å) multilayers with Gd alloy layers of the same thicknesses (50 to 200 Å).

Following the procedure of the previous Section, we have calculated the corresponding $M(T)$ curves for a series of GdCo(X)/Co(40 Å) multilayers (Fig. 8). The compensation points experimentally observed are not reproduced if we only consider the change in composition of the RE layer. As for the series with fixed Gd thicknesses, a reduction in the Co layer is necessary to achieve an agreement. More interestingly, this is reached for the same Co reduction employed in the previous section, which points to the validity of our explanation. In summary, the actual compensation point is again quantitatively reproduced only if the eutectic alloy and the reduced effective thickness are both considered.

Again, discrepancies in the detailed shape of the magnetization curve may arise from the effect of a wide interface and/or significant magnetocrystalline fields. More detailed explanations will require knowledge of the interface structure (elemental and magnetic profiles) and including in the model magnetocrystalline effects. To this respect, the Gd/Co system seems more complex than Gd/Fe multilayers where simple model calculations are in fine agreement with magne-

tization data.[23] However, we would like to point out that the compensation point, when present, is given correctly within our approach. This indicates that the key parameters governing its behavior are the net magnetic moments in the atomic layers and the exchange coupling constants, rather than the precise structure of the interfaces.

VI. CONCLUSIONS

Calculations of the low-field magnetization $M(T)$ in multilayers with antiparallel interface coupling are consistent with the behavior of real sputtered Gd/Co samples. Following previous composition experiments, the use of an eutectic Gd alloy in the layering with Co is proved to successfully account for the compensation temperatures of real samples. Further experimental evidence that was necessary to explain the compensation point is a thinned magnetic Co layer, presumably as a result of the strong interdiffusion. Semiquantitative agreement is achieved in the case of thick Co layers, where both pure Co and $Gd_{0.63}Co_{0.37}$ layers are well formed, but is not good for very thin Co layers. The picture is consistent for series of multilayers made either with a fixed Gd layer thickness or with a fixed Co thickness. The crucial features of the system—namely, the overall dependence of magnetization with temperature and the appearance of compensation points—can be explained by a model that correctly accounts for the net magnetic moment in the atomic layers—reduced because of the alloy formation—and the exchange coupling, modified for the same reason.

ACKNOWLEDGMENTS

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