Magnetism of Rare-Earth–Transition-Metal Nanoscale Multilayers

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Systematic studies have been performed on the layer-thickness dependence of magnetization and anisotropy for amorphous rare-earth–transition-metal compositionally modulated films with characteristic bilayer thicknesses (λ) in the range 0.6–3 nm (6–30 Å). The results permit for the first time the development of a detailed model for the magnetization and perpendicular magnetic anisotropy as a function of λ. Single-ion anisotropy is shown to be the major contributor to the perpendicular anisotropy for Dy/Co and similar systems.

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A major challenge at present in condensed matter physics is that of understanding the magnetism of ultrathin layers[1] and multilayers[2-4] of magnetic elements. Of particular interest is the question of the existence and origins of perpendicular magnetic anisotropy (PMA) when the thickness of the layers becomes less than about one nanometer (several monolayers). In the cases of several multilayer systems such as Co/Au [3] and Ni/Mo,[2] it has been possible to separate the various contributions to the anisotropy (K_u), and to determine the interface anisotropy by studying the thickness dependence of K_u.

A related problem with important technological implications is that of the origins of PMA in certain nominally amorphous rare-earth–transition-metal (RE-TM) films. Although this effect was discovered more than a decade ago,[5] the basic physical and structural mechanisms responsible for it have remained controversial. Often it is claimed that the origin is an excess of TM pairs in the plane, or RE-TM pairs perpendicular to the film plane. In an attempt to probe these latter effects we have performed systematic studies on several RE/TM multilayered systems with characteristic layer thicknesses in the nanometer region.[6] This paper reports the results of these studies and develops a physical model in terms of which the PMA of such systems can be understood. We exploit the extra degree of freedom in compositionally modulated films (CMF) to control the anisotropic pair correlations, and thus the PMA. Since PMA and a large magneto-optic Kerr effect (MOKE) are essential for the new high-density MO data-storage technology, there is great interest in understanding and optimizing these properties in multilayer thin films. Initial MOKE studies in multilayered systems such as Fe/Cu [7] and RE/TM [8] have been reported. In this paper we shall focus on the magnetism and PMA of Dy/Co and similar CMF, and will point out similarities to TM multilayers such as Co/Au.

The samples were grown on Mylar substrates in a multiple-gun sputtering system and the magnetic properties were measured with a vibrating-sample magnetometer. The preparation conditions and the procedure of determining the magnetic characteristics, e.g., the anisotropy, are the same as those mentioned in our previous papers.[6,9] Large- and small-angle x-ray diffraction measurements have been performed on selected samples and some examples are shown in Ref. 6. Briefly, the structure is polycrystalline for individual layer thickness greater than about 15 Å, and for thinner layers the structure is amorphous. For these amorphous multilayers, the small-angle x-ray diffraction patterns only show the first-order peaks which implies the compositionally modulated structure has a sinusoidal form.[6,10]

In order to study systematically the layer-thickness dependence of the properties, a total of sixty samples of the form Y-Å-Dy/X-Å-Co have been made. The intervals of X and Y are about one atomic diameter, namely 2.5 Å for Co and 3.5 Å for Dy, but in some regions the intervals are only about one-half atomic diameter, which enables us to investigate more precisely the layer-thickness dependence of magnetic properties.

One example of the layer-thickness dependence of magnetic properties for Y-Å-Dy/X-Å-Co is shown in Fig. 1. The films possess perpendicular anisotropy for intermediate values of layer thicknesses. Note that all five samples in Fig. 1 have the same chemical ratios of Co and Dy. This means that the behavior is clearly controlled by the layer thickness rather than the chemical composition.

The measured uniaxial anisotropy, K_u', is determined by the area between the parallel and perpendicular $\sigma(H)$ curves. Figure 2 shows a plot of K_u' vs X for 8-Å-Dy/X-Å-Co, which is a straight line for $X \geqq 15$ Å. This behavior, which has been observed in numerous magnetic multilayers,[2,3,6,9] can be understood in terms of the following expression:

$$\lambda K_u' = 2K_i + (K_v - 2\pi M_s^2)X , \quad (1)$$

where K_i, K_v, and $2\pi M_s^2$ are the interface, volume, and demagnetization anisotropy energies, respectively. The K_i value for this series is ~ 0.7 erg/cm^2. However, we

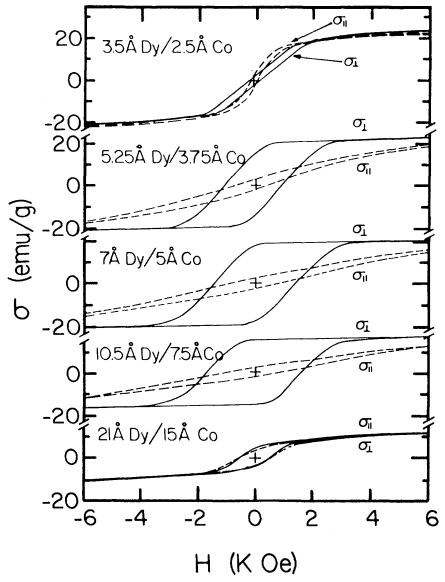

FIG. 1. Layer-thickness dependence of hysteresis loops for several Y-Å-Dy/X-Å-Co multilayers. Data in this and subsequent figures were obtained at room temperature.

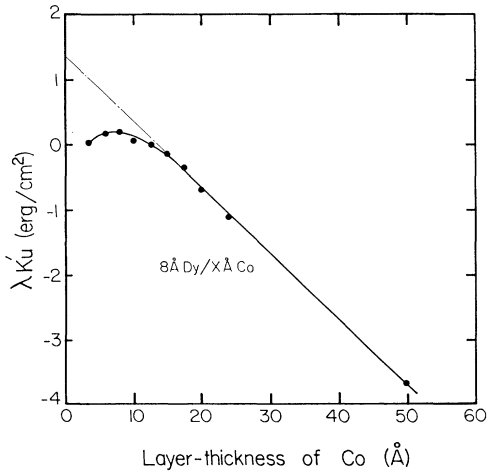

FIG. 2. The anisotropy energy K_u' multiplied by the bilayer thickness vs the Co layer thickness for 8-Å-Dy/X-Å-Co samples.

see from Fig. 2 that Eq. (1) breaks down as a description of the data for $X \leqq 15$ Å, just where the structure changes from compositionally modulated crystalline to compositionally modulated amorphous. In the rest of this paper we focus on the small-X and -Y region where PMA occurs.

Figure 3 illustrates the systematics of the layer-thickness dependence of the magnetization. The valley between the two peaks of magnetization, where $\sigma_\perp = 0$, originates from the antiferromagnetic coupling of Co and Dy moments; thus this valley traces the compensation points of the films of different compositions. As X and Y approach zero, $|\sigma_\perp|$ decreases because the structure tends toward that of a homogeneous amorphous alloy with a magnetic structure either spin-glass-like or paramagnetic as is shown in Ref. 10. A three-dimensional plot, similar to Fig. 3, has been made for the *intrinsic* anisotropy, $K_u = K_u' + 2\pi M_s^2$. It shows a positive peak (PMA) centered around $X \approx 6$ Å, $Y \approx 6$ Å. As X and $Y \rightarrow 0$, $K_u \rightarrow 0$ and for X larger than about 16 Å, $K_u < 0$.

We analyze the magnetization and anisotropy data by means of models adapted for the CMF microstructure. A brief discussion of the procedure is given below with the details to be published elsewhere.[10]

First, a mean-field model[11] is fitted to experimental data on *homogeneous* a-DyCo alloys to obtain the sublattice magnetizations, M_r and M_t, as a function of composition. Then the amorphous CMF with sinusoidal compositional modulation is modeled by

$$\eta_t(z) = \eta_{t0} + (A/2)\cos(2\pi z/\lambda) , \qquad (2)$$

where η_t (η_r) is defined to be the atomic fraction of the TM (RE). (For the purposes of this derivation we assume $X = Y$ for simplicity.) η_{t0} is a constant and A is the peak-to-peak compositional modulation of the TM. The RE/TM multilayer is divided into thin slices parallel to the interfaces with the z axis normal to them. The CMF magnetization can be expressed as the average over the slices:

$$M = \frac{1}{\lambda}\sum_i [M_t(\eta_t(z_i)) - M_r(\eta_r(z_i))]\Delta z_i . \qquad (3)$$

Within the constraints of the CMF composition, the parameters η_{t0} and A are determined by fitting the calculated magnetization to experimental data.

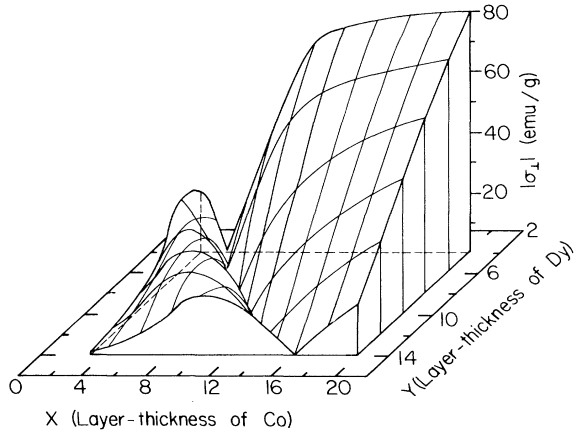

FIG. 3. Layer-thickness dependence of magnetization for Y-Å-Dy/X-Å-Co at $H = 8$ kOe. $\sigma_\perp$ means the magnetization was measured as the applied field was perpendicular to the film plane.

As an example the calculated results for 6-Å-Dy/X-Å-Co samples are shown in Fig. 4. It is seen that the calculated and experimental magnetization data agree very well. The average magnetization of Co and Dy subnetworks are also drawn in this figure. Figure 4 also shows the Co-layer-thickness dependence of the Co-atomic-fraction modulation, A. The A value is only about 0.1 for the thinnest Co layer thickness of 3.5 Å and its value increases as the Co layer becomes thicker. This feature is understandable in terms of a larger mixing effect for the thinner-layer samples, which results from some combination of diffusion and roughness introduced at interfaces during fabrication.

The main sources of magnetic anisotropy in RE-TM systems are dipolar interactions and the single-ion anisotropy due to spin-orbit interactions.[12,13] In order to assess the relative importance of the two interactions in the system under study, our experimental data for K_u for several RE/TM systems are plotted in Fig. 5. They show that the anisotropy K_u for Dy/Fe, Dy/Co, and Tb/Fe CMF is roughly an order of magnitude larger than that of Gd/Fe and Gd/Co. Since Gd has no spin-orbit interactions, it is reasonable to attribute the main origin of the perpendicular anisotropy of the Dy and Tb multilayers to single-ion anisotropy of the rare-earth ion. This is further supported by the very good agreement between the experiment and the following model based on single-ion anisotropy.

The single-ion anisotropy energy of the ith rare-earth ion in a disordered system with respect to the easy direction z can be written as[14]

$$E_i = 2\alpha_J \langle r^2 \rangle A_2^0 J_z^2 , \tag{4}$$

where α_J is the Steven's factor, $\langle r^2 \rangle$ is the quantum-mechanical average of the square of the radius of the $4f$ orbit, J_z is the z component of the angular momentum, and A_2^0 is a crystal-field term. A_2^0 is given by

$$A_2^0 \propto \sum_j q_j (3\cos^2\theta_j - 1) r_j^{-3} , \tag{5}$$

where summation is over the neighboring ions with charge q_j and distance r_j from the rare-earth ion and θ_j is the angle with respect to the easy direction. For a compositionally modulated amorphous structure the sum over j is replaced with an integral weighted with an anisotropic pair distribution function[12]

$$P_{ij}(\mathbf{r}) = \eta_j(z) R_{ij}(r)[1 + \beta_j \cos\theta_j + \cdots] , \tag{6}$$

where $\eta_j(z)$ is the atomic fraction of the jth neighboring atom, $R_{ij}(r)$ is the *isotropic* part of the distribution, and β_j is the lowest-order anisotropic contribution. The anisotropy in the pair distribution function can arise from the composition modulation of multilayers and inverse magnetostriction due to stresses at the various interfaces. We have found that the anisotropy is the same for mica, Ta, Cu, and Mylar substrates ruling out any significant magnetostrictive effect at the sample-substrate interface. This effect is also not expected to be significant at the Dy/Co interfaces due to their diffuse nature. Thus the compositional modulation appears to be the main source of the anisotropic pair distributions and magnetic anisotropy.

The lowest-order contribution to A_2^0 comes from the first-order term in the expansion of η from Eq. (2) about the rare-earth ion and it is proportional to A/λ. Also, the first-order term in η has the maximum value at the interface and the charges q_i are expected to be most significant in the interface region. Thus, as expected, most of the anisotropy comes from the interface region.

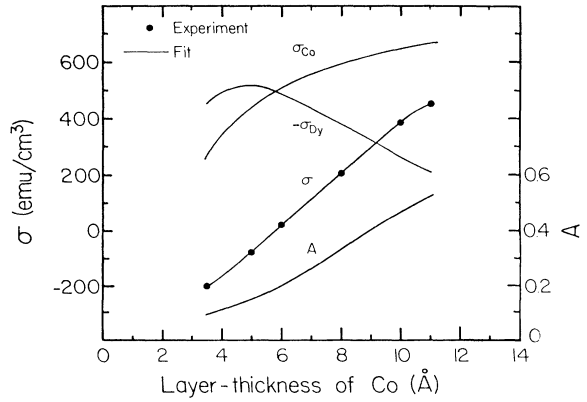

FIG. 4. The comparison of the calculated magnetization (emu/cm^3 of film) with the experimental data for 6-Å-Dy/X-Å-Co (X=3.5, 5, 6, 8, 10, and 11) and the Co-layer-thickness dependence of the Co-atomic-fraction modulation.

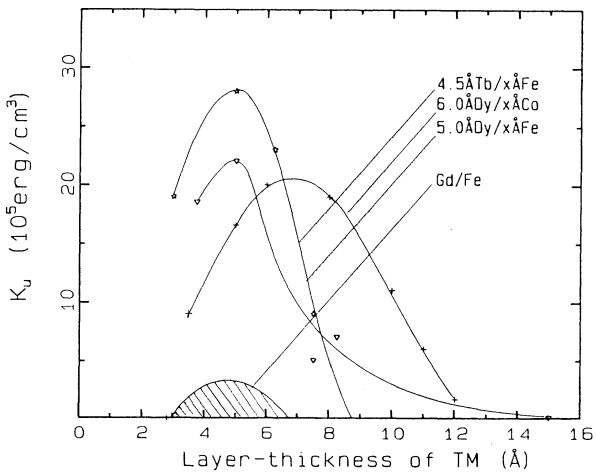

FIG. 5. The anisotropy characteristics for 4.5-Å-Tb/X-Å-Fe, 5-Å-Dy/X-Å-Fe, 6-Å-Dy/X-Å-Co, and Gd/Fe CMF. The anisotropy data of 2.3-Å-Gd/3-Å-Fe, 3-Å-Gd/3-Å-Fe, 3-Å-Gd/5-Å-Fe, etc., are in the shaded area. Data for several Gd/Co samples are negligibly small or negative on this figure.

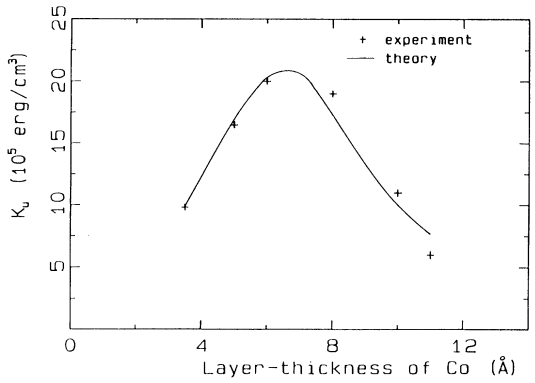

FIG. 6. A comparison between the calculated and experimental anisotropy for 6-Å-Dy/X-Å-Co (X=3.5, 5, 6, 8, 10, and 11).

Combining the results for A_2^0 with E_i in Eq. (4), the anisotropy energy K_u for a CMF can be written as

$$K_u = \xi \frac{A}{\lambda} \langle M_r^2 \rangle , \tag{7}$$

where ξ is a constant and $\langle M_r^2 \rangle$ is the average of the square of the saturation magnetization of the rare-earth subnetwork in the easy direction.[10] With ξ as an adjustable parameter, Eq. (7) is fitted to the K_u data for 6-Å-Dy/X-Å-Co samples and the results are shown in Fig. 6. In view of the single parameter, the agreement between the experimental data and calculated results is remarkable. The fitted value of $\xi = 5.26 \times 10^{-6}$ cm leads to an average value of the single-ion anisotropy parameter $D = 2 \times 10^{-17}$ erg, which is reasonable in terms of the typical value of the single-ion random-anisotropy parameter in amorphous RE-TM alloys. Finally, this model gives similar fits for other RE/TM CMF which will be published elsewhere.

As noted earlier, there are similarities in the $K_u(X)$ data for RE/TM CMF and TM multilayers such as Co/Au. Namely, when X becomes less than about 10 Å, Eq. (1) breaks down because of a high degree of disorder in the multilayer. Presumably an analysis similar to that given above will be applicable to systems such as Co/Au and related multilayers as well.

An additional point is that our analysis has not *proved* that K_u should be positive corresponding to PMA rather than in-plane anisotropy. To do this requires a detailed structural model and assumptions on the magnitude and *sign* of the charge transfers on atoms in the vicinity of the interface. This is a challenging problem for the future.

In summary, we have systematically investigated the layer-thickness dependence of magnetization and anisotropy for Dy/Co CMF. For RE/TM nanoscale multilayers, in those cases where the RE atom possesses orbital angular momentum, the main source of anisotropy is found to be the single-ion anisotropy associated with the RE. We have given a new model, based on single-ion anisotropy of the rare-earth ions, for understanding the thickness dependence of the anisotropy in the important region between crystalline individual layers (relatively thick) and zero-thickness layers (which correspond to a homogeneous glass). The model is in excellent agreement with the experimental data for the perpendicular anisotropy of RE/TM multilayers. The control of the microstructure, and thus the magnetic properties, may find important applications in various magneto-optic or other data-storage technologies.

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