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Magnetization and perpendicular anisotropy in Tb/Fe multilayer films

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A single-ion model approximated by point-charge interactions is proposed to account for the perpendicular anisotropy K_u in Tb/Fe multilayer films with variant modulation periods. In multilayers, K_u depends not only on the crystal field, but also on the orientation of the magnetic moments along the z direction in the period. A quasi-self-consistent method of determining the crystal-field coefficient D and the average orientation of the moments in the interface was developed during the fitting process of K_u. The crystal-field coefficient $D=2.4\times10^{-17}$ erg is determined. The best-fit values of K_u and M_s are in good agreement with experiment.

I. INTRODUCTION

Recently, the properties of the rare-earth–transition-metal (RE-TM) multilayer films have widely been investigated. The interest was mainly focused on the origin of the perpendicular anisotropy, the possibility of the enhancement of the Kerr rotation, and the effect of the interface.[1-3]

The origin of the perpendicular anisotropy is, unfortunately, not yet completely clear, although several authors[4,5] attempted to explain it. In this paper, we propose a single-ion model approximated by point-charge interactions.[6-8] It is responsible for the perpendicular anisotropy of the Tb/Fe multilayer films with a variant modulation period $L=d_{\mathrm{Fe}}+d_{\mathrm{Tb}}$, where d_{Fe} and d_{Tb} are the thicknesses of the single Fe and Tb layers, respectively. The effect of stress on the anisotropy exerted by the substrate onto the multilayer is probably negligible, since the perpendicular anisotropy constant K_u of samples on glass or Kapton substrates are very similar. Additional effects due to interlayer stress might well play a role, but are not considered here. Single-ion anisotropy resulting from crystal or electrostatic fields is generally most important for the $4f$ rare-earth atoms because of their large orbital moment. By the action of spin-orbit coupling, the magnetic moment of the atom is therefore confined to a certain easy direction. The single-ion model is reasonable to explain the anisotropy K_u for RE compounds or amorphous RE-TM alloys. When using this model in the case of multilayer films, K_u is dependent not only on the crystal field but also on the orientation of the magnetic moments with respect to the vertical direction. Furthermore, the orientation is not homogeneous along the vertical direction over the modulation period, especially for periods which are large compared with the atomic distances. In order to treat this problem, a quasi-self-consistent method is developed in this paper.

We consider the simultaneous action of both the crystal field, characterized by a random distribution around the z direction, and the mean field on the magnetic moment, similar to the method used by Harris, Plischke, and Zuckerman[9] and Callen, Liu, and Cullen[10] in their successful treatment of the magnetism and the remanence of amorphous systems. Then, within the framework of a self-consistent numerical calculation of the perpendicular anisotropy to be fitted to the experimental data, the crystal-field coefficient and a parameter representing the variable orientation of the magnetic moments along the direction of the modulation wave vector are determined.

In the process of calculating K_u, the distribution of the concentration of both Fe and Tb along the perpendicular direction in the period are calculated for multilayer films with different periods L. The fits to the measured saturation magnetizations are achieved by using the known dependence of the saturation magnetization on the composition of amorphous Tb-Fe films. The calculation is based on the model that a period of the multilayer film is subdivided into a series of thin sublayers with different atomic concentrations due to the diffusion during sample preparation. Then, by the help of the mean field model we can deduce the quantity of the magnetic moment and the molecular field for each individual sublayer. This will be, in turn, utilized to determine the crystal-field coefficient and the orientation of the magnetic moment in the period. We discuss this point in more detail in Sec. II B.

II. MODEL

A. The expression of the perpendicular anisotropy

By applying the point-charge model,[6] the anisotropy energy with respect to the z direction for the $4f$ electrons of a Tb atom at site i is expressed as[4]

$$W_c(i)=\tfrac{3}{4}|e|\langle r^2\rangle\alpha_J(J^2-J/2)[\cos^2\theta_M(i)-\tfrac{1}{3}]\times\sum_j q_j(3\cos^2\theta_{ij}-1)/R_{ij}^3\ , \tag{1}$$

where e is the electronic charge, $\langle r^2\rangle$ the size of the $4f$ orbit, and α_J the Stevens factor.[11] θ_M is the polar angle of the direction of the atomic magnetic moment μ_i (Fig. 1). R_{ij} and θ_{ij} are the polar coordinates of the nearest-neighbor ion j with the charge q_j and the sum is taken

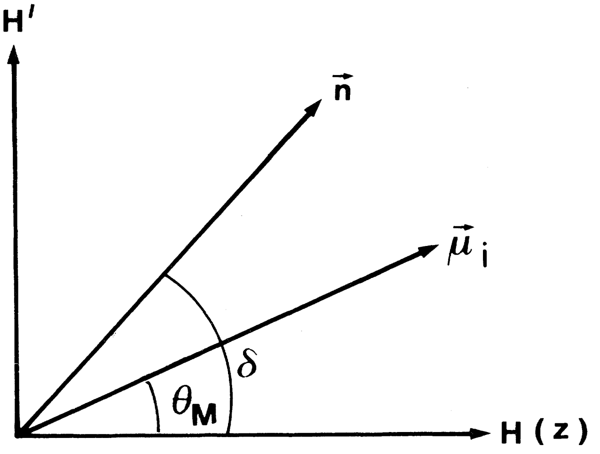

FIG. 1. Relative orientation of the atomic magnetic moment μ_i and the local easy axis n_i at the ith site. z is the direction of the magnetic field; H' is the in-plane field.

over the nearest-neighbor ions j. J is the quantum number of the total angular momentum of the $4f$ electrons and we let $J=1$ in the following numerical calculation as done by Harris *et al.*[9]

If we assume

$$\sum_j q_j/R_{ij}^3 = Z|e|a^3 \ , \tag{2}$$

we obtain

$$\begin{aligned} W_c(i) &= (3/4a^3)Ze^2\langle r^2\rangle\alpha_J(J^2-J/2)[\cos^2\theta_M(i)-\tfrac{1}{3}] \\ &\quad\times\sum_j(3\cos^2\theta_{ij}-1) \\ &= D(J^2-J/2)[\cos^2\theta_M(i)-\tfrac{1}{3}] \ , \end{aligned} \tag{3}$$

with

$$D=(3/4a^3)Ze^2\langle r^2\rangle\alpha_J\sum_j(3\cos^2\theta_{ij}-1) \ . \tag{4}$$

Here, a is the average distance between the nearest atoms, Z is a constant varying between 2 and 2.5,[7] and D is referred to as the crystal-field coefficient.

Usually, atomic diffusion between neighboring layers in multilayer films occurs. For this reason, as will be shown below, the direction of the atomic magnetic moment μ_i, θ_M varies with the z coordinate along the period L. Hence, when applying the expression (3) to the case of the multilayers, both the local concentration of the Tb atoms $G(z)$ and $\cos^2\theta_M(z)$ have to be averaged over the period L. This is done in the following expression for the total anisotropy energy:

$$K_u=\sum_i W_c(i)=D(J^2-J/2)\left[\frac{\int NG(z)\,dz}{\int dz}\right]\times\left[\left[\frac{\int\cos^2\theta_M(z)\,dz}{\int dz}\right]-\frac{1}{3}\right] \ , \tag{5}$$

where N is the number of Tb atoms per unit volume. Equation (5) can eventually be written in the following concise form:

$$K_u=DN\langle G(z)\rangle(J^2-J/2)[\langle\cos^2\theta_M(z)\rangle-\tfrac{1}{3}] \ . \tag{6}$$

It should be noticed that the last bracket in Eq. (6) simultaneously denotes a thermal average. It vanishes at large Tb concentration, when considering Tb/Fe alloys, which are paramagnetic at $x>0.37$ (Fig. 2).

B. The orientation of the magnetic moment

Now we describe how to determine the angle θ_M of the magnetic moment with the z direction. As illustrated in Fig. 1, for an amorphous system, there is an anisotropic exchange energy, a Zeeman energy, and a single-ion anisotropy energy. For multilayers, the energy induced by an in-plane field should be considered, if in-plane anisotropy exists within the period. Then the total energies for ith moment μ_i at the coordinate z is expressed as

$$E_i(z)=-\mu_i(H+\lambda M)\cos\theta-D\cos^2(\delta-\theta)-\mu_i H'\sin\theta \ , \tag{7}$$

where H is the applied field. λ is the mean-field coefficient,[12] and M is equal to $\mu_i nm$, with n the number of moments per unit volume and

$$m=\langle\cos\theta(=\theta_M)\rangle \ , \tag{8}$$

which accounts for the deviation of the local moment μ_i from the z direction. H' is the field in the x-y plane and we anticipate that it has to be accounted for owing to the existence of the in-plane magnetic vector in the pure Fe layers. δ is the polar angle of the easy direction $\mathbf{n}$ of the local single-ion field (Fig. 1).

Classically the equilibrium position θ of the moment is given by

$$\begin{aligned}\partial E_i(z)/\partial\theta &= \mu_i(H+\lambda M)\sin\theta-D\sin[2(\delta-\theta)] \\ &\quad-\mu_i H'\cos\theta=0 \ . \end{aligned} \tag{9}$$

Then we obtain

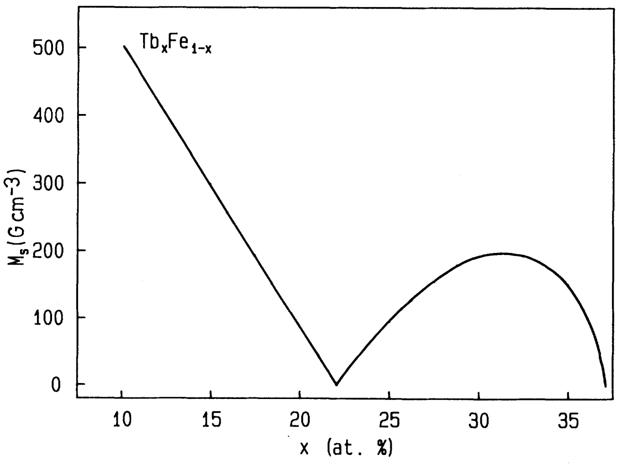

FIG. 2. Saturation magnetization M_s vs x in amorphous Tb_xFe_{1-x} films.

$$[\mu_i(H+\lambda M)/D]\sin\theta-(\mu_i H'/D)\cos\theta=\sin[2(\delta-\theta)]$$

or

$$[(h+m)/d]\sin\theta-(h'/d)\cos\theta=\sin[2(\delta-\theta)]\ . \qquad (10)$$

$d=D/n\lambda\mu_i^2$ is the ratio of the energy of the single-ion anisotropy to the exchange energy $E_{\text{ex}}=n\lambda\mu_i^2$, and $h'=\mu_i H'/n\lambda\mu_i^2$ is the ratio of the in-plane Zeeman energy to the exchange energy. We let $h'=0.1$ in the fitting procedure, because the in-plane field is much smaller than the exchange coupling and it appears reasonable from the fitting process. $h=\mu_i H/n\lambda\mu_i^2$ is a reduced applied field. Following the treatment of Ref. 10, where the reduced magnetization is taken as the spherical average,

$$m=\langle\cos\theta\rangle=\frac{\int\cos\theta\sin\delta\,d\delta}{\int\sin\delta\,d\delta} \qquad (11)$$

assuming $0\le\delta\le\pi/2$. We obtain the following expressions under the approximation of small and larger angle θ:

$$m\approx 1-(\tfrac{4}{15}+3h'/2)d^2,\quad \text{for small } \theta\ , \qquad (12)$$

$$m\approx 0.5+(1-h')/6d,\quad \text{for larger } \theta\ . \qquad (13)$$

From Eqs. (12) and (13) we can see that there is a corresponding value of $\langle\cos\theta\rangle$ for each value of d. Then, using Eq. (6) in connection with Eq. (12) or (13) we can determine the value of D if the exchange coupling energy E_{ex} and h' are known in advance. Clearly, Eqs. (12) and (13) do not apply to Tb-rich paramagnetic parts of the multilayer, $x>0.37$, where $\langle\cos^2\theta_M\rangle=\frac{1}{3}$ (Sec. II A).

After deducing the approximate expression of $\langle\cos\theta\rangle$ for small and large angles θ we should explain the difference of the definition for θ_M in Eqs. (6) and (8). In Eq. (6), θ_M is the angle between the magnetic moment of a Tb atom and the z direction. Now we consider the Tb/Fe system. Therefore, the moment μ_i in Eq. (7) should be replaced by an effective moment, which is equal to the moment difference of the two sublattices with ferrimagnetic exchange interaction. Accordingly, θ in Eq. (8) is the angle between the effective moment and the z direction.

C. The magnetization

As mentioned previously, the multilayers consist of many sublayers with different composition within the period L. The total saturation magnetization is, hence, an average value taken over these sublayers. In the following we assume that the distribution of the concentration of Fe and Tb is described by sinusoidal curves along the z axis within their individual periods $l_{\text{Fe}}=2d_{\text{Fe}}$ and $l_{\text{Tb}}=2d_{\text{Tb}}$. Then we can write the distribution of the concentration of Fe and Tb atoms along the z axis in real Tb/Fe multilayers within one period of length L as follows:

$$G_{\text{Fe}}(z)=k_0+k_l\sin(2\pi z/l_{\text{Fe}})\ ,$$
$$\text{with } G_{\text{Tb}}(z)=1-G_{\text{Fe}}(z) \text{ and } 0\le z\le d_{\text{Fe}}\ ; \qquad (14)$$

$$G_{\text{Tb}}(z)=(1-k_0)+k_2\sin[2\pi(z+d_{\text{Fe}})/l_{\text{Tb}}]\ ,$$
$$\text{with } G_{\text{Fe}}(z)=1-G_{\text{Tb}}(z) \text{ and } d_{\text{Fe}}\le z\le L\ ; \qquad (15)$$

and $0<G_{\text{Fe}},G_{\text{Tb}}<1$. k_0 is the concentration of Fe in an ideal interface layer, which includes a monolayer of Fe atoms and a monolayer of Tb atoms in neighboring position. In other words, no diffusion between the neighbor layers is taken into account for the ideal interface layer. k_1 and k_2 are fitting parameters. The total saturation magnetization M_s is equal to

$$M_s=\int_0^L M(z)\,dz/L\ , \qquad (16)$$

with

$$M(z)=\sum_i nm\mu_i\ . \qquad (17)$$

In order to determine the distributions given by Eqs. (14) and (15) we refer to M_s in Eq. (16) as the experimental result obtained on Tb/Fe multilayers with period L. The values $M(z)$ are assumed to correspond to the concentration-dependent saturation magnetization as obtained on amorphous Tb-Fe films (Fig. 2), $M(z)=M(G_{\text{Fe}}(z),G_{\text{Tb}}(z))$. Then, by inserting k_0 and systematically varying k_1 and k_2 the the concentration distributions of Tb and Fe atoms in the period can be determined.

III. EXPERIMENTAL

We have prepared the multilayer Tb/Fe films by dc magnetron sputtering using separate Tb and Fe targets. The diameter of the targets was about 12 cm. The glass substrates with thickness 0.17 mm were rotated and cooled by water during the sample preparation. The choice of the modulation periods comprised (1.25 Å Fe)/(1.75 Å Tb), (2.5 Å Fe)/(3.5 Å Tb), (5 Å Fe)/(7 Å Tb), (7.5 Å Fe)/(10.5 Å Tb), (10 Å Fe)/(14 Å Tb), (12.5 Å Fe)/(17.5 Å Tb), and (15 Å Fe)/(21 Å Tb), respectively. It was limited by both the speed of the rotating substrate and the power supply. Both small- and large-angle x-ray diffraction were used to check the structure. All samples show the character of multilayers except the sample (1.25 Å Fe)/(1.75 Å Tb), which lacks any peaks in the small-angle x-ray diffraction pattern. The saturation magnetization M_s was measured by a vibrating sample magnetometer, and the perpendicular anisotropy constant K_u was determined by a very sensitive torque meter.

IV. RESULTS AND DISCUSSION

Figure 3 shows the saturation magnetization M_s as a function of L for various Tb/Fe multilayer films (rectangle symbols). As indicated above, best-fit distribution function of Eqs. (14) and (15) were chosen such as to reproduce M_s by virtue of Eq. (16) in connection with the well-known curve M_s versus x of amorphous $\text{Tb}_x\text{Fe}_{1-x}$ films (Fig. 2). The resulting best-fit curve M_s versus L (solid line in Fig. 3) agrees well with the experimental data. The best-fit distribution functions $G_{\text{Tb}}(z)$ and $G_{\text{Fe}}(z)$ are shown in Fig. 4 for (2.5 Å Fe)/(3.5 Å Tb), (5 Å

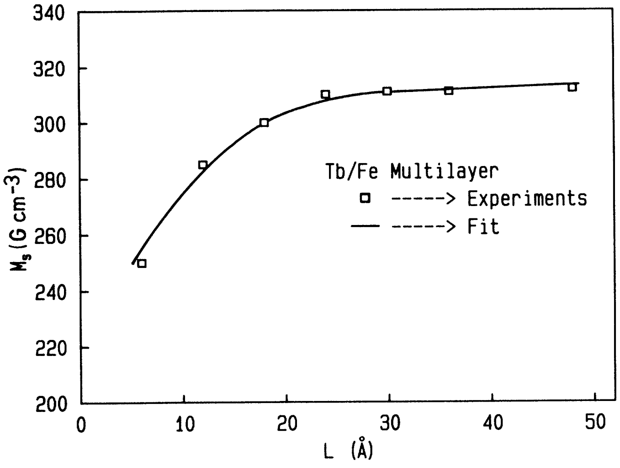

FIG. 3. Saturation magnetization M_s vs L for Tb/Fe multilayer films obtained by both experiment (open square) and by fitting (solid line).

Fe)/(7 Å Tb), (10 Å Fe)/(14 Å Tb), (12.5 Å Fe)/(17.5 Å Tb), and (15 Å Fe)/(21 Å Tb). It is seen that these curves have truncated sinusoidal shapes, the slopes of which characterize the interface alloying. In the case of (15 Å Fe)/(21 Å Tb) the interface width equals about the sum of the diameters of one Fe and one Tb atom. Pure Fe and Tb layers, where $G_{\mathrm{Fe}}=1$ and $G_{\mathrm{Tb}}=1$, respectively, become increasingly important as L increases. The sublayers with $G_{\mathrm{Tb}}=1$ are referred to as dead layers, since bulk Tb is paramagnetic at room temperature. In other words, within our model calculation the Tb atom that possesses indirect exchange interaction with Fe as the nearest neighbors of Tb atom, has a contribution to magnetization. Consequently, $G_{\mathrm{Tb}}=1$ is approximated to be the situation that the Tb's surrounding atoms are the same kind of atoms.

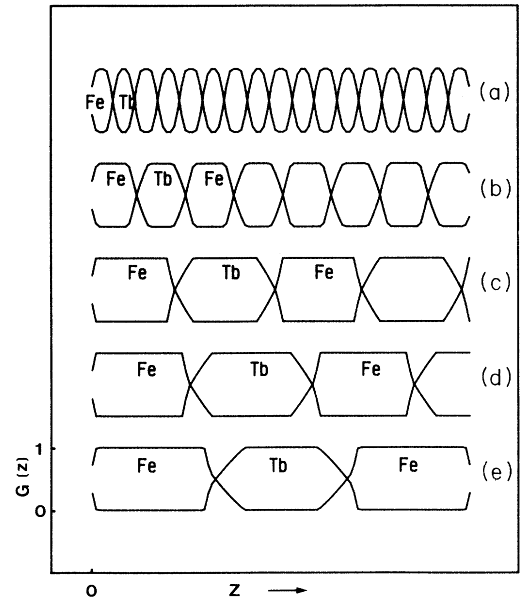

FIG. 4. Distribution of Fe and Tb concentrations along the z direction in multilayer films with A: (2.5 Å Fe)/(3.5 Å Tb), B: (5 Å Fe)/(7 Å Tb), C: (10 Å Fe)/(14 Å Tb), D: (12.5 Å Fe)/(17.5 Å Tb), and E: (15 Å Fe)/(21 Å Tb).

The distribution functions of the Tb and Fe atoms thus obtained are the starting point of calculating the perpendicular anisotropy constant K_u. According to Eq. (6) we additionally need the single-ion anisotropy constant D and the orientational average $\langle \cos^2\theta_M \rangle$. In the fitting process of K_u we first consider the multilayer system with the smallest period, $L=d_{\mathrm{Fe}}+d_{\mathrm{Tb}}=6$ Å, and assume that its exchange energy $E_{\mathrm{ex}}=n\lambda u_i^2$ at the coordinate z in the period is substituted by that obtained from amorphous alloy with the same composition. Within the framework of mean-field theory we calculated the molecular field constant λ from its Curie point T_c and the magnetic moment μ_i for each individual sublayer with the known concentration. Eventually, the total exchange energy is obtained by taking into account the average mixing ratio. We then obtain K_u as a function of d by inserting $D=dE_{\mathrm{ex}}$ and $\langle \cos^2\theta_M(z) \rangle \approx m^2(d)$ according to Eq. (12) into Eq. (6). This is solved numerically for d by using the experimental value of K_u. We eventually obtain the total average value, $D=2.1\times 10^{-17}$ erg and $\langle \cos\theta \rangle=0.95$ in the case of period $L=6$ Å. In order to fit the anisotropy data of the other systems with $L>6$ Å (Fig. 5), we assume that D remains constant, but E_{ex} varies because of the increasing inhomogeneity of the layered structures (Fig. 4). Again, the experimental values of K_u are fitted to Eq. (6), but this time by solving numerically for λ, which is implicitly contained in $\langle \cos^2\theta_M \rangle$. The resulting curve K_u versus L is shown to fit well with the experimental data in Fig. 5, including negative K_u value at $L>41$ Å. It should be noticed that this curve has been obtained by subsequently optimizing the initial value of K_u in order to minimize the mean-square deviation of all data points. The final best-fit parameter, $D=2.4\times 10^{-17}$ erg, is believed to account for slight deviations from complete homogeneity of the $L=6$ Å multilayer. By means of Eqs. (12) and (13)

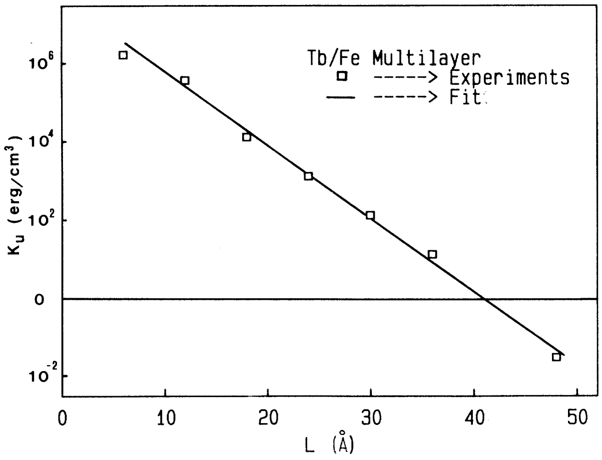

FIG. 5. Perpendicular anisotropy constant K_u vs L for Tb/Fe multilayer films obtained experimentally (open square) and by fitting (solid line).

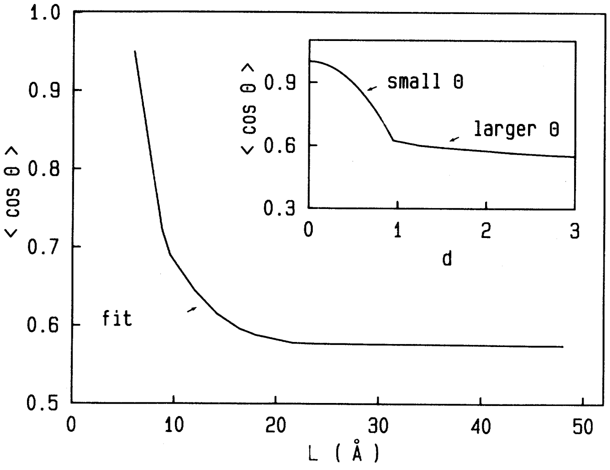

FIG. 6. ⟨cosθ⟩ vs L in Tb/Fe multilayer films obtained by fitting of experimental data (Figs. 3 and 5). Inset: ⟨cosθ⟩ vs d according to Eqs. (12) and (13).

we are now able to determine the spin texture, ⟨cosθ⟩. As shown in the insert of Fig. 6 we assume the crossover between the small- and larger-angle approximation to take place at $d=1$. The curve ⟨cosθ⟩ versus L thus obtained and plotted in Fig. 6 describes the remanence as a function of L. We remark that these results fit reasonably well with experimental data obtained previously[12,13] on the same set of Tb/Fe multilayers. It should be noticed that the statistical model[10] starts from the assumption that the magnetic moments are distributed on the upper hemisphere only. This is confirmed by our calculation revealing ⟨cosθ⟩>0.55 (Fig. 6). The excellent agreement between the experimental data of M_s (Fig. 3) and K_u versus L (Fig. 5) with the numerical calculation seems to confirm the relevance of the underlying model. The perpendicular anisotropy of Tb/Fe multilayer films is, hence, very probably due to the randomly distributed single-ion anisotropy, which is successfully described within a point-charge crystal-field model. In our model calculation the entire value of K_u is assumed to be due to the "effective" parts of the interfaces, where $0<G_{\text{Tb}}<0.37$. The volume fraction contributing to K_u becomes smaller as L increases for multilayers with large period lengths.

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