Generic Source of Perpendicular Anisotropy in Amorphous Rare-Earth–Transition-Metal Films

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We show that the magnetic dipolar energy of antiparallel dipoles in the surface layers contributes to the intrinsic uniaxial perpendicular anisotropy in amorphous rare-earth–transition-metal films. The volume anisotropy energy density K_u of this contribution depends inversely on the thickness of the film. For an amorphous GdCo film with a thickness of 1000 Å the calculated K_u is on the order of 10^3-10^4 ergs/cm^3. Dependence of K_u on temperature and atomic concentration is also calculated.

It is well known that amorphous rare-earth–transition-metal (RE-TM) thin films have a strong perpendicular anisotropy. This property makes the RE-TM films particularly useful in magneto-optic data storage. However, since the striking experimental discovery of the perpendicular anisotropy in sputtered GdCo films by Chaudhari, Cuomo, and Gambino in 1973,[1] there has not been a satisfactory theory on the intrinsic perpendicular anisotropy in amorphous RE-TM films. The origin of the anisotropy has been qualitatively attributed to many different mechanisms, e.g., pair ordering,[2] columnar microstructures,[3] single-ion anisotropy,[4,5] exchange anisotropy,[6] and bond-orientational anisotropy.[7] These mechanisms are common in assuming that there must be certain prior anisotropic structure (either atomic or microscopic) in order to explain the perpendicular anisotropy.

In this Letter we show that the magnetic dipolar energy of antiparallel dipoles in the surface layers of amorphous RE-TM films gives a positive contribution to the intrinsic perpendicular anisotropy energy density K_u. In contrast to previous mechanisms, this K_u does not relate to any anisotropic atomic structures or microstructures. It is thus a generic source of perpendicular anisotropy for RE-TM amorphous films. Since it is a surface effect, K_u is inversely proportional to the thickness of the film. For an amorphous GdCo film with a thickness of 1000 Å, the calculated value of K_u is on the order of 10^3-10^4 ergs/cm^3. This surface effect gives a straightforward explanation to the experiments showing that K_u decreases with increasing thickness.[8] As we shall show later, the observed dependence of K_u on temperature[9,10] and atomic concentration[11] may also be understood based on this mechanism.

Though we shall concentrate on RE-TM pairs at the surface layers, the mechanism to be described in this Letter will also lead to the bulk anisotropy as well, provided that there are more in-plane bulk RE-TM pairs than perpendicular ones. Many experiments have shown *how* the anisotropy is closely related to the anisotropic distribution of RE-TM pairs. Our model provides both a qualitative and a quantitative explanation as to *why*, so far as the magnetic dipolar energy is concerned.

An amorphous RE-TM film essentially contains two different types of magnetic ions: RE and TM. The exchange integrals are positive for RE-RE and TM-TM pairs and negative for RE-TM pairs. This leads to a parallel alignment for the same type of dipoles and an antiparallel alignment for the different types of dipoles. We shall show that in such a ferrimagnetic arrangement the magnetic dipolar interaction of the surface antiparallel dipoles produces the perpendicular anisotropy.

Let us first consider the magnetic dipolar energy of the antiparallel pairs shown in Fig. 1. The main difference between these arrangements is that in Fig. 1(a) the dipoles are perpendicular to the bond connecting the atomic centers, while in the cases of Fig. 1(b) the dipoles are parallel or antiparallel to the bond. The dipolar interaction energy of two magnetic dipoles $\mathbf{m}_i$ and $\mathbf{m}_j$ separated by $\mathbf{r}_{ij}$ is given by

$$U_{ij} = (1/r_{ij}^3)[\mathbf{m}_i \cdot \mathbf{m}_j - (3/r_{ij}^2)(\mathbf{m}_i \cdot \mathbf{r}_{ij})(\mathbf{m}_j \cdot \mathbf{r}_{ij})] . \quad (1)$$

Thus we find the dipolar energy $U_\perp = -m_{RE}m_{TM}/(r_{RE}+r_{TM})^3$ for the case of Fig. 1(a) and $U_\parallel = 2m_{RE}m_{TM}/(r_{RE}+r_{TM})^3$ for the cases of Fig. 1(b), where $m_{RE} = |\mathbf{m}_{RE}|$, $m_{TM} = |\mathbf{m}_{TM}|$, and r_{RE} and r_{TM} are the radii of RE and TM atoms, respectively. The fact that $U_\perp < U_\parallel$ indicates that antiparallel dipoles prefer the orientation perpendicular to the bond. Since there are more in-plane bonds at the surface, the antiparallel pairs there tend to orient perpendicularly to the film plane, which is the common direction perpendicular to all the randomly dis-
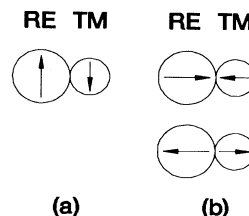

FIG. 1. Typical magnetic arrangements of antiparallel dipoles.

tributed in-plane bonds. Of course, based on the same reason, the TM-TM and RE-RE dipolar interactions will give negative contributions to the perpendicular anisotropy. However, since in most amorphous RE-TM films with perpendicular anisotropy the dipolar energy of a TM-TM pair is much smaller than a RE-TM pair and the number of RE-RE pairs is much fewer than that of RE-TM pairs, the contribution of the RE-TM pairs plays a dominant role. We shall show this point later in this Letter by considering the dipolar energy of all kinds of dipole pairs.

Before quantifying the above argument, let us show that, for a random distribution of atoms, the dipolar energy density U has the following dependence on the orientation of the dipoles:

$$U = U_0 + K_{tot} \sin^2 \Theta , \qquad (2)$$

where K_{tot} is the total anisotropy energy density and Θ is the angle between the dipole moment and the direction normal to the film. That U does not depend on the dipole's azimuthal angle Φ in the film plane means that the anisotropy is uniaxial. To prove (2), we consider the sum of U_{ij} in Eq. (1) over all dipoles for randomly distributed atoms. Since all the dipoles are either parallel or antiparallel, the first term in Eq. (1) does not depend on (Θ, Φ). The sum over the second term is also independent of Φ, because there is no preferential direction in the film plane. Therefore, without loss of generality, we can choose $\Phi = \pi/2$. In this coordinate system the sum over the second term of Eq. (1) is proportional to

$$\sum_i \sum_j (\sin^2 \Theta \sin^2 \theta_{ij} \cos^2 \phi_{ij} + \cos^2 \Theta \cos^2 \theta_{ij}$$
$$+ \tfrac{1}{2} \sin 2\Theta \sin 2\theta_{ij} \cos \phi_{ij}) , \qquad (3)$$

where (θ_{ij}, ϕ_{ij}) are the spherical angles of $\mathbf{r}_{ij}$. Obviously, the third term in (3) vanishes since ϕ_{ij} is randomly distributed in the interval $(0, 2\pi)$. The first two terms can be written in the form of Eq. (2). The proof is thus complete.

It is well known that in a homogeneous magnetic film the magnetic dipolar interaction produces a shape anisotropy density equal to $-2\pi M_s^2$, where M_s is the saturation magnetization. The minus sign indicates that the in-plane orientat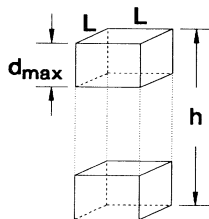ion of magnetization is preferred. In the literature the total perpendicular anisotropy density K_{tot} is usually written as $K_{tot} = K_u - 2\pi M_s^2$, where K_u is called the intrinsic perpendicular anisotropy density.

The magnitude of K_u contributed by dipolar interaction of antiparallel pairs at the surface can be estimated as follows. For a RE-TM pair in the surface of the film the energy difference between the orientations $\Theta = \pi/2$ and $\Theta = 0$ is equal to $\Delta U = U_\parallel - U_\perp = 3 m_{RE} m_{TM}/(r_{RE} + r_{TM})^3$. Suppose that the pair occupies a surface area $\pi(r_{RE}^2 + r_{TM}^2)$, and the film thickness is h, then the corresponding K_u is equal to

$$\frac{\Delta U}{\pi h (r_{RE}^2 + r_{TM}^2)} = \frac{3 m_{RE} m_{TM}}{\pi h (r_{RE} + r_{TM})^3 (r_{RE}^2 + r_{TM}^2)} .$$

For RE = Gd and TM = Co, one has $r_{RE} = 1.82$ Å, $r_{TM} = 1.25$ Å,[12] $m_{RE} = 7\mu_B$, $m_{TM} = 1.7\mu_B$ at $T = 0$ K,[13] where $\mu_B = 9.27 \times 10^{-21}$ emu is the Bohr magneton. Thus, for $h = 1000$ Å, a pair of antiparallel dipoles in the surface makes a contribution of 7×10^3 ergs/cm^3 to K_u.

To calculate the exact value of K_u, we should take into account the dipolar energy between all kinds of dipoles and for all possible interacting ranges. We shall use the following two steps. First, we calculate the dipolar energy for a surface area $L \times L$ in the film plane, i.e., we consider those dipolar interactions of which at least one dipole is located in the $L \times L \times h$ volume as illustrated in Fig. 2. The other dipole may be located inside or outside (even far apart from) that volume. Fortunately, as we shall show in Fig. 3, we need not calculate those dipole pairs which are too far apart from one another, since for long-range interactions only the macroscopic magnetization plays a role, which leads to the already known shape anisotropy. Therefore, we introduce a maximum interaction range d_{max} and calculate only the dipole pairs satisfying $r_{ij} \leq d_{max}$, where r_{ij} is the distance between the considered dipoles. Since d_{max} is finite, only dipoles in the two $L \times L \times d_{max}$ volumes adjacent to the surfaces as illustrated in Fig. 2 contribute to K_u. Other dipoles do not *feel* the existence of the surfaces. Second, we
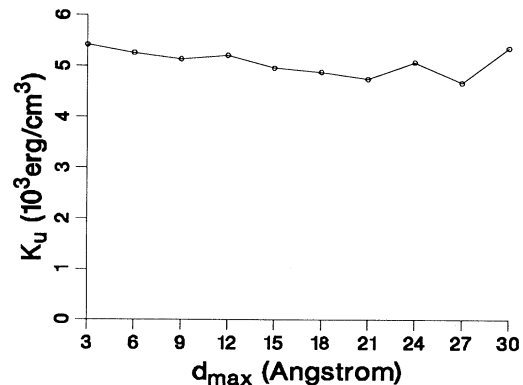

FIG. 2. Only atoms in the upper and lower boxes contribute to K_u.

FIG. 3. The calculated K_u as a function of the truncation length d_{max} for $\chi_{RE} = 40\%$ and $T = 300$ K.

subtract a term, which has already been included in the shape anisotropy, $-2\pi M_s^2$, from the dipolar energy calculated in the first step. This term should be equal to the dipolar energy calculated in a similar way described in the first step, except that the magnetic medium should be considered continuous and homogeneous. Given the maximum interaction range d_{max} and the film thickness h it can be shown that this term leads to a contribution of $-3\pi d_{max} M_s^2/4h$ to the perpendicular anisotropy density if $d_{max} \leq h$. Since the calculated K_u converges very fast with increasing d_{max}, we need not consider the case $d_{max} > h$.

Based on these two steps, the intrinsic anisotropy energy density K_u averaged over the column shown in Fig. 2 (and hence in the whole film) is given by

$$K_u = \frac{1}{L^2 h} \sum_{i,j} \left[U_{ij}\left(\Theta = \frac{\pi}{2}\right) - U_{ij}(\theta = 0) \right] + \frac{3\pi d_{max} M_s^2}{4h},$$

(4)

where U_{ij} is defined in Eq. (1), i labels any atom located inside the upper box shown in Fig. 2, and j any atom satisfying $r_{ij} \leq d_{max}$ which may be inside or outside the box. Since the two boxes shown in Fig. 2 are identical, we have considered only one of them and then multiplied the result by a factor of 2. This factor of 2 is canceled by another factor of $\frac{1}{2}$ arising from the fact that each U_{ij} is shared by two atoms.

We have calculated K_u from Eq. (4) based on a simple cubic lattice in which a certain percentage of sites are randomly occupied by RE atoms and the other sites are occupied by TM atoms. We assume that all first neighbors of RE are TM atoms. This is believed to be the case in amorphous $Tb_{20}Fe_{80}$ thin films as reported in recent EXAFS (extended x-ray-absorption fine-structure) studies.[14] This assumption ensures that there are enough antiparallel pairs. Obviously, there exist inaccuracies with reality in this model; e.g., in an amorphous material each atom has about nine nearest neighbors[14] (instead of

six in the present model) and the distances between atoms depend on the radii of the atoms. However, for the purpose of demonstrating the surface effects these discrepancies are of secondary importance. We mention in passing that for this simple cubic lattice Eq. (4) gives $K_u = 0$ on the average in the absence of surfaces.

We have calculated Eq. (4) for RE=Gd and TM =Co. In the curves illustrated in Figs. 3-5, the lattice constant is chosen to be 2.9 Å, which is the distance between two nearest antiparallel dipoles observed in amorphous GdCo films.[15] The lattice has a size $116 \times 116 \times 58$ Å^3. The square area defined in Eq. (4) and Fig. 2 is 58×58 Å^2 (i.e., $L = 58$ Å), which contains 400 surface atoms. For $d_{max} \leq 30$ Å, the only boundary for the atoms in the upper box shown in Fig. 2 is the surface of the lattice. The temperature T, the relative concentration of RE atoms χ_{RE}, and the truncation distance d_{max} are variable in these calculations.

Figure 3 shows the calculated K_u as a function of the truncation distance d_{max}. We see that K_u is approximately the same for d_{max} varying from 3 to 30 Å. This means that K_u is mainly contributed by the nearest-neighbor interactions and hence only by the two surface layers of the film.

K_u depends on temperature T through m_{RE} and m_{TM}. To calculate m_{RE} and m_{TM} as functions of T, we have applied the mean-field-theory results[13] for RE=Gd and TM=Co, where we have used the exchange energies $J_{RE-RE} = 0.5 \times 10^{-15}$, $J_{TM-TM} = 28 \times 10^{-15}$, and $J_{RE-TM} = -2.2 \times 10^{-15}$ ergs. The atomic radii and moments have been given before. The calculated Curie temperature T_C for these parameters is about 1000 K. The calculated function $K_u(T)$ is shown in Fig. 4. A similar curve has already been observed for $Gd_{0.15}Co_{0.74}Mo_{0.11}$ (Ref. 9) and $Gd_{0.10}Co_{0.67}Mo_{0.14}Ar_{0.09}$ (Ref. 10) films. K_u as a function of the relative concentration of RE atoms is shown in Fig. 5. A similar behavior has been observed in $Tb_x Fe_{1-x}$ thin films.[11] In this case we see that K_u drops if there are too few or too many RE

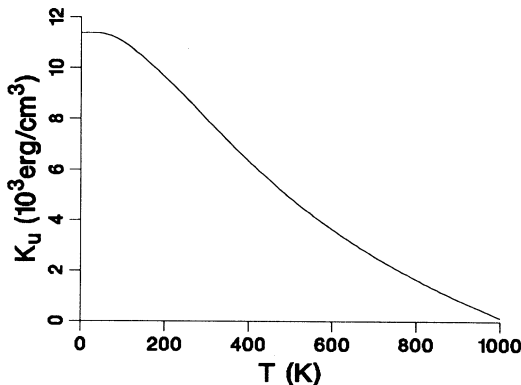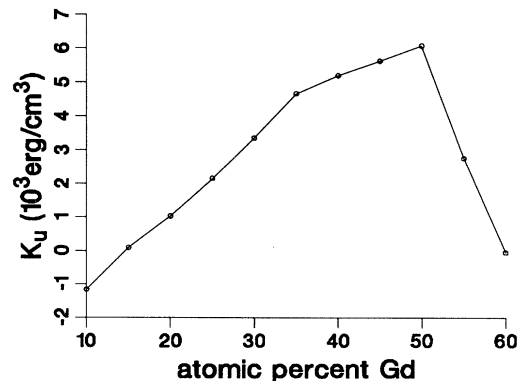FIG. 4. The calculated temperature dependence of K_u for $\chi_{RE} = 30\%$ and $d_{max} = 10$ Å.

FIG. 5. The calculated K_u as a function of χ_{RE} for $T = 300$ K and $d_{max} = 10$ Å.

atoms. The reason is that in these two limits there are too few antiparallel pairs.

In Eq. (4) we see that K_u is proportional to $1/h$, where h is the thickness of the film. This is due to the fact that only the surface antiparallel dipoles contribute to the anisotropy, since the atomic distribution is assumed to be isotropic in the film. In a real amorphous RE-TM film the atomic distribution may be anisotropic and both surface and bulk antiparallel dipoles may contribute to K_u. In this case, the dependence of K_u on h will become complicated. To compare our results with experimental data, GdCo and GdFe would be the best candidates, since for these materials the dipolar energy is possibly the dominant factor for the perpendicular anisotropy. However, to our knowledge, there is no systematic experimental data on the thickness dependence of K_u at the present time.

It should be noted that the radii r_{RE} and r_{TM} used in our calculations are deduced from the respective crystal constants. In practice, this is suitable for the like atoms, but may not be suitable for calculating the distance r_{RE-TM}, since the RE-TM pairs may be chemically combined. The length of the chemical bond of the antiparallel pair should be equal to the sum of the ionic radii of the two atoms, which are much smaller than the radii in the metallic state; e.g., the ionic radii of Co^{++}, Co^{+++}, and Gd^{+++} are equal to 0.74, 0.63, and 1.02 Å, respectively.[16] If this is the case, the anisotropy produced by the surface antiparallel dipoles could be about 10 times stronger than what we have calculated.

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