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Surface and volume anisotropy from dipole-dipole interactions in ultrathin ferromagnetic films

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At the boundary of a ferromagnetic material, the local change in the surroundings of the atomic magnetic moments induces an additional magnetic anisotropy. The dipole-dipole interaction, responsible for the form-dependent demagnetizing field inside the ferromagnet, differs for magnetic moments at the boundary and magnetic moments inside the bulk material. By calculation it is shown that the demagnetization factor for an ultrathin ferromagnetic film is thickness dependent. However, the anisotropy resulting from the dipole-dipole interaction can be interpreted as a surface and a volume anisotropy which depend on the crystalline structure and orientation of the film, but are independent of the thickness of the film.

INTRODUCTION

One of the interesting magnetic properties of thin films is the anisotropy, which determines the preferential orientation of the magnetization. Experimentally it has been found that when the thickness of ferromagnetic films is reduced to several atomic layers, the anisotropy differs considerably from its value in thick films. As the difference depends on the thickness t as $1/t$, it is attributed to the two surfaces of the film and consequently called surface anisotropy.¹ The influence of the surfaces (or interfaces, when in contact with another material) can be large enough to change the preferential orientation of the magnetization from in the plane of the film to perpendicular to the film.²

Despite early attempts to determine the surface anisotropy theoretically,³ present calculations seem to be restricted to monolayers⁴ and do not include the thickness t . Therefore, the dependence of the anisotropy on $1/t$, as quoted above, has not yet been confirmed from first principles.

In this paper we want to evaluate the influence of the dipolar interaction on the surface anisotropy. First we will consider the ferromagnetic film as a continuum and second as a set of discrete, atomic dipoles, neatly ordered in the film. Using this last approach, the result of the calculation will depend on the crystal structure of the film. Since we will only consider simple ferromagnets, it seems appropriate to concentrate on high-symmetric structures as body centered cubic (bcc), face centered cubic (fcc), hexagonal close packed (hcp), and tetragonal. The structure can have different orientations relative to the film and we will indicate this by the axis that is perpendicular to the plane of the film, e.g., bcc[110] means the film has the bcc structure with the [110] axis perpendicular to the film.

We will assume that the magnitude of the magnetic moment does not depend on its orientation, as is the case for most ferromagnets. The anisotropy can then be calculated as the energy difference between two magnetic saturated states, one with the magnetization parallel to the plane of the film and the other one with the magnetization perpendicular to the plane of the film. Further, it will be assumed that the magnitude of the magnetic moment does not depend on the thickness of the film or its position in the film.

Apart from saturated states, domain structures in these ultrathin films are of interest. We will not consider these, but refer to a recent paper by Yafet and Gyorgy,⁵ which theoretically treats them for a ferromagnetic monolayer, and to Draaisma and de Jonge⁶ for domain configurations in a multilayer.

CONTINUUM APPROACH

Neglecting the discrete nature of matter, magnetization can be treated as a field $\mathbf{M}(\mathbf{r})$, which obeys the currentless Maxwell equations. At interfaces between two regions with different magnetization we have the usual boundary conditions: the normal component of \mathbf{B} and the tangential component of \mathbf{H} should be continuous. The magnetostatic energy is the total energy difference between the situation in which the sample has a given magnetization distribution to the situation in which there is no sample at all.⁷ This involves both the magnetic field inside and outside the sample, but by taking the appropriate expressions for the energy, the volume of integration can be limited to the sample volume. When a field \mathbf{M} is given, the solutions for \mathbf{B} and \mathbf{H} are often formulated with the use of the magnetic potential Ψ , defined by $\mathbf{H} = -\nabla\Psi$. The function Ψ is a solution of Laplace's equation $\nabla^2\Psi = 0$. In this formulation the average magnetostatic energy density can be expressed as⁸

$$E = -\frac{\mu_0}{2V} \int dV \mathbf{M} \cdot \mathbf{H} = \frac{\mu_0}{2V} \int dV (-\nabla \cdot \mathbf{M}) \Psi, \quad (1)$$

with V for the volume of the sample. In the planar geometry of a thin film we find that, when the magnetization is saturated and θ is the angle between the axis normal to the plane of the film and \mathbf{M} , the average energy density becomes

$$E(\theta) = \frac{1}{2} \mu_0 M_s^2 \cos^2 \theta, \quad (2)$$

in which M_s is the saturation value of the magnetization. The anisotropy E_a is the difference between the magnetostatic energy for the parallel orientation ($\theta = \pi/2$) and the perpendicular orientation ($\theta = 0$), which yields

$$E_a = -\frac{1}{2} \mu_0 M_s^2. \quad (3)$$

In this approach the thickness of the film plays no role and

therefore no surface contributions proportional to $1/t$ are predicted.

DISCRETE DIPOLES

When the thickness of the ferromagnetic film is reduced to a few atomic layers, the assumption that the film can be treated as a magnetic continuum is no longer valid. In that case we treat the magnetic system as a collection of discrete magnetic dipoles, which are regularly arranged on a crystal-line lattice. The dipolar energy of a dipole i can then be expressed as

$$\mathcal{E}_i = -\frac{1}{2} \frac{\mu_0}{4\pi} \sum_{j \neq i} \left(-\frac{m^2}{r_{ij}^3} + \frac{3(\mathbf{m} \cdot \mathbf{r}_{ij})^2}{r_{ij}^5} \right). \quad (4)$$

\mathbf{m} is the magnetic moment of the dipoles, $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$ is the relative position of dipoles i and j and $r_{ij} = |\mathbf{r}_{ij}|$. This dipole-dipole interaction can be interpreted as the energy of the dipole in the field of all the other dipoles:

$$\mathcal{E}_i = -\frac{1}{2} \mathbf{m} \cdot \mathbf{B}_i, \quad \mathbf{B}_i = \frac{\mu_0}{4\pi} \sum_{j \neq i} \left(-\frac{\mathbf{m}}{r_{ij}^3} + \frac{3(\mathbf{m} \cdot \mathbf{r}_{ij}) \mathbf{r}_{ij}}{r_{ij}^5} \right). \quad (5)$$

The factor of $\frac{1}{2}$ results from the fact that every pair of dipoles should only be counted once. The field \mathbf{B}_i is called the dipole field and can be viewed upon as the sum of the demagnetizing field and the Lorentz field.⁹ \mathbf{B}_i can be written as

$$\mathbf{B}_i = \mu_0 \mathbf{d}_i \mathbf{m} = \mu_0 \mathbf{D}_i \mathbf{M}_s, \quad (6)$$

where we have used

$$\mathbf{M}_s = \mathbf{m} / V_{\text{dip}}, \quad (7)$$

with V_{dip} for the volume per dipole. For a given dipole i , \mathbf{D}_i is a second rank tensor which depends on the position of the dipole, relative to the other dipoles, but is independent of the dimensions of the sample and the unit cell of the crystallographic lattice. In Cartesian coordinates we can write its components as

$$(\mathbf{D}_i)_{kl} = \frac{V_{\text{dip}}}{4\pi} \sum_{j \neq i} \left(-\frac{\delta_{kl}}{r_{ij}^3} + \frac{3(r_{ij})_k (r_{ij})_l}{r_{ij}^5} \right), \quad (8)$$

with $k, l = x, y, z$. Note that \mathbf{D}_i is symmetric for all positions i and also $D_{xx} + D_{yy} + D_{zz} = 0$.

In our case, the dipoles are arranged in an infinitely large thin film. We choose the z axis perpendicular to the plane of the film. An additional property of \mathbf{D}_i is then $(\mathbf{D}_i)_{xz} = (\mathbf{D}_i)_{yz} = 0$ and \mathbf{D}_i is only dependent on the position of the atomic layer in the z direction. So \mathbf{D}_i is determined by three independent parameters: D_{xx} , D_{xy} , and D_{zz} for each atomic layer in the film. A further reduction occurs when the atomic layers have more than twofold rotational symmetry. In that case $D_{xy} = 0$ and $D_{xx} = D_{yy}$ and therefore $D_{xx} = -\frac{1}{2} D_{zz}$, so there is only one parameter left.

The ferromagnetic film consists of N atomic layers in the z direction. The anisotropy $E_a(n)$ contributed by the dipoles in atomic layer n is the difference in energy between the state in which all dipoles point in the x direction ($\theta = \pi/2$) and the state in which they all point in the z direction ($\theta = 0$). From Eqs. (5), (6), and (7) we can derive that

$$E_a(n) = [\mathcal{E}_i(\theta = \pi/2) - \mathcal{E}_i(\theta = 0)] / V_{\text{dip}} = -\frac{1}{2} \mu_0 M_s^2 (D_{xx} - D_{zz}), \quad (9)$$

when dipole i is in layer n . Introducing the reduced anisotropy $k(n)$, we can write in a high symmetric lattice

$$k(n) = E_a(n) / \frac{1}{2} \mu_0 M_s^2 = -(D_{xx} - D_{zz}) = \frac{3}{2} D_{zz}. \quad (10)$$

The actual calculation of D_{zz} has been performed for various films consisting of monolayers up to several atomic layers. The dipoles are divided into two sets: those within a cylinder of radius R with its rotation axis perpendicular to the film plane around a dipole in layer n and those outside that cylinder. The contribution to D_{zz} of dipoles from the first set is summed discretely, according to formula (8). The contribution of dipoles from the second set is included by integration. Figure 1 shows this graphically by indicating a dot for a discretely summed dipole and a line for an integrated area. Note that the integrals are absolutely convergent due to the two-dimensional configuration of the dipoles. R is chosen large enough to have no influence on the results, which is about 100 atomic distances in the present calculations.

As an example we show in Fig. 2 the value of $k(n)$ for a bcc[100] film consisting of $N = 1, \dots, 10$ atomic layers. The value at the outside layers is 23% smaller than at the inside layers, whereas at the second layer $k(n)$ is 2% larger than at the inside. This result is similar to an earlier calculation of the dipole field at the surface of small particles by Christensen and Mørup.¹⁰

The total reduced anisotropy k is the average of the anisotropy of the different layers:

$$k = \frac{1}{N} \sum_{n=1}^N k(n). \quad (11)$$

This reduced anisotropy k can partly be considered as an equivalent of the demagnetizing factor, but we will not dwell on this point of view. Note that the definition of k is such that a negative value indicates a preferential direction in the plane of the film and a positive value indicates a perpendicular preferential direction. In this formulation the continuum approach yields $k = -1$.

RESULTS

The calculated results of the reduced anisotropy k for various crystal structures and orientations is shown in Fig. 3(a). The value of k is independent of the lattice constant for one and the same structure, but in order to compare different structures, the lattice constants have to be relatively fixed. We have chosen to take the same V_{dip} for all structures

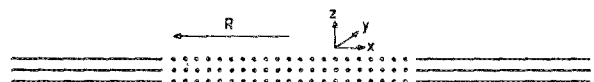


FIG. 1. Magnetic dipoles within a cylinder of radius R are summed in a discrete way, whereas those outside are integrated. R is taken large enough (about 100 times the interatomic distance) to have no influence on the results. The contributions of each atomic layer are summed to get the final result.

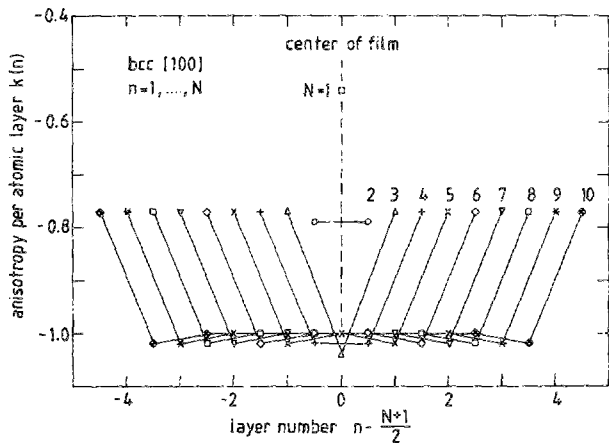


FIG. 2. Anisotropy of a dipole in a ferromagnetic film is dependent on the layer in which the dipole is located. In this figure the anisotropy of a dipole field in a bcc[100] film is shown as function of the layer number n ($n = 1, \dots, N$, where N is the number of atomic layers in the film). The horizontal axis is shifted for different N to keep the center of the film at the same position.

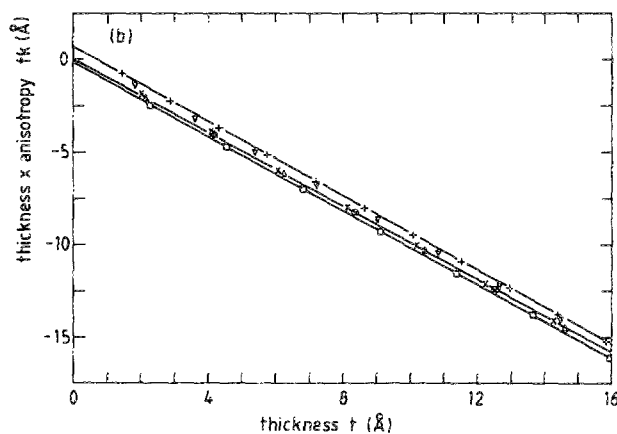
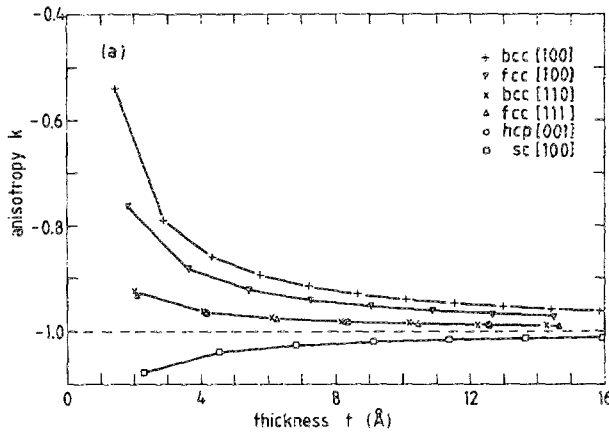


FIG. 3. (a) Calculated anisotropy k vs the thickness of the film t for films of various crystalline structures and orientations. Only for large thicknesses the anisotropy tends to its continuum value ($k = -1$). (b) The same data as shown in (a) are plotted as thickness times anisotropy vs thickness, showing that the anisotropy resulting from dipole-dipole interactions between the magnetic moments can be separated into a surface and a volume anisotropy, according to Eq. (12).

$[V_{\text{dip}} = (2.28 \text{ \AA})^3]$, so that the same magnetic moment in different structures leads to the same magnetization. For all high-symmetric structures k approaches the continuum limit $k = -1$ for thick layers. As the thickness is reduced, the total anisotropy deviates appreciably from the continuum value. For monolayers, represented by a thickness of approximately 2 \AA , it is of course impossible to define the crystal structure, but it is understood that the same crystallographic net can be taken as for an atomic layer in the complete crystal and the same value for V_{dip} .

As quoted in the introduction, the analysis of experimentally observed anisotropies seems to indicate that a volume and a surface contribution can be distinguished. This observation can be expressed as

$$kt = 2k_s d + k_v t, \quad (12)$$

with t for the thickness of the film and d for the distance between two successive atomic layers in the z direction. d is introduced only to make k_s dimensionless, just as k and k_v (note $t = Nd$). k_v is the contribution proportional to the volume of the film, whereas k_s represents the contribution proportional to the surface of the film. In the spirit of these analyses of experimental data we have treated our calculations accordingly. In Fig. 3(b) the product of the thickness t and the anisotropy k have been plotted as a function of t . For all cases calculated in Fig. 3(a) indeed a linear dependence is found. In Table I the resulting k_s and k_v as they follow from least-square fits for the points in Fig. 3(b) are tabulated. For the crystal structures considered here, we find $k_v = -1$ which means that there is no contribution from the dipole-dipole interaction to what is usually called the crystalline anisotropy. There is, however, a contribution from the dipole-dipole interaction to the anisotropy which can be designated as dipole-dipole surface anisotropy. It has to be noted that relation (12) does not exactly fit the calculated values. However, the differences are much smaller ($< 1\%$) than usual experimental errors, so that this relation can be used fruitfully.

From the reduced value of the anisotropy we can find the actual value via

$$K_s = k_s d \frac{1}{2} \mu_0 M_s^2. \quad (13)$$

To estimate the order of magnitude, let us take Co in fcc[100] orientation. As parameters we take the lattice constant $a = 3.55 \text{ \AA}$ and the magnetization $\mu_0 M_s = 1.76 \text{ T}$. The volume contribution to the total anisotropy resulting from dipole-dipole interactions is then $K_v = -1.23 \times 10^6 \text{ J/m}^3$

TABLE I. Least-square parameters for the volume and surface anisotropy in a thin film as a result of the dipole-dipole interaction between the magnetic moments. The values included to obtain this fit are those shown in Fig. 3.

Film	k_v	k_s
sc[100]	-0.9999	-0.0393
fcc[111]	-0.9999	0.0344
fcc[100]	-1.0003	0.1178
bcc[110]	-1.0001	0.0383
bcc[100]	-1.0016	0.2187
hcp[001]	-0.9994	0.0338

and the surface contribution $K_s = 25.7 \times 10^{-6} \text{ J/m}^2$. In the fcc [111] orientation K_v is the same, but $K_s = 8.67 \times 10^{-6} \text{ J/m}^2$. Experimentally, in Pd/Co multilayers it is found that $K_s = 550 \times 10^{-6} \text{ J/m}^2$,¹¹ which is more than an order of magnitude larger. This means that other sources for anisotropy, such as spin-orbit coupling, are more important for the Pd/Co interface.

Of course, this calculation can be performed for other structures as well. As an example to show the influence of a reduction of symmetry in the crystal structures relative to the high-symmetric structures, we have calculated the anisotropy for the tetragonal structure with different c/a ratios. In Fig. 4 and Table II the results show that again the anisotropy can be divided into an interface and a volume contribution, but now there is a substantial contribution from the dipole-dipole interaction to the crystalline anisotropy. The volume anisotropy k_v even becomes positive for $c/a = 0.6$, which means that the preferential direction for the magnetization becomes perpendicular to the film when the film is thick enough to overcome the surface anisotropy, which is negative in this case. The fact that k_v becomes more positive when the ratio c/a decreases can qualitatively be

TABLE II. Volume and surface anisotropy for tetragonal structures with different c/a ratios, when the dipole-dipole interaction is the only source for anisotropy.

c/a	k_v	k_s
1.0	-1.000	-0.0392
0.9	-0.834	-0.0685
0.8	-0.624	-0.1196
0.7	-0.337	-0.2102
0.6	0.0913	-0.3725
0.5	0.793	-0.6788
0.4	2.078	-1.2960

understood by observing that the magnetic dipole density in the direction of the c axis increases, which strongly favors the alignment in that direction.

When magnetic layers can be epitaxially grown on materials with larger lattice constants,¹² this causes an expansion of the lattice in the plane of the film and a contraction perpendicular to it. With the mechanism sketched above, this can have a large influence on the magnetic anisotropy energy.

CONCLUSION

We have shown that the variation of the anisotropy with the thickness of thin ferromagnetic films can be understood from first principles in the case that only dipole-dipole interactions are taken into account. The calculated anisotropy in this case can very well be described by a surface and a volume contribution. In a way this result is somewhat surprising in view of the long range of the dipole-dipole interaction, in contrast to local sources of anisotropy (primarily resulting from spin-orbit coupling). The results justify the analysis of the experimental anisotropy in ultrathin films in terms of a surface and volume contribution based on the thickness dependence of the anisotropy. Corrections for the demagnetization, based on the continuum approach, should be avoided, since they are not physically meaningful in this case.

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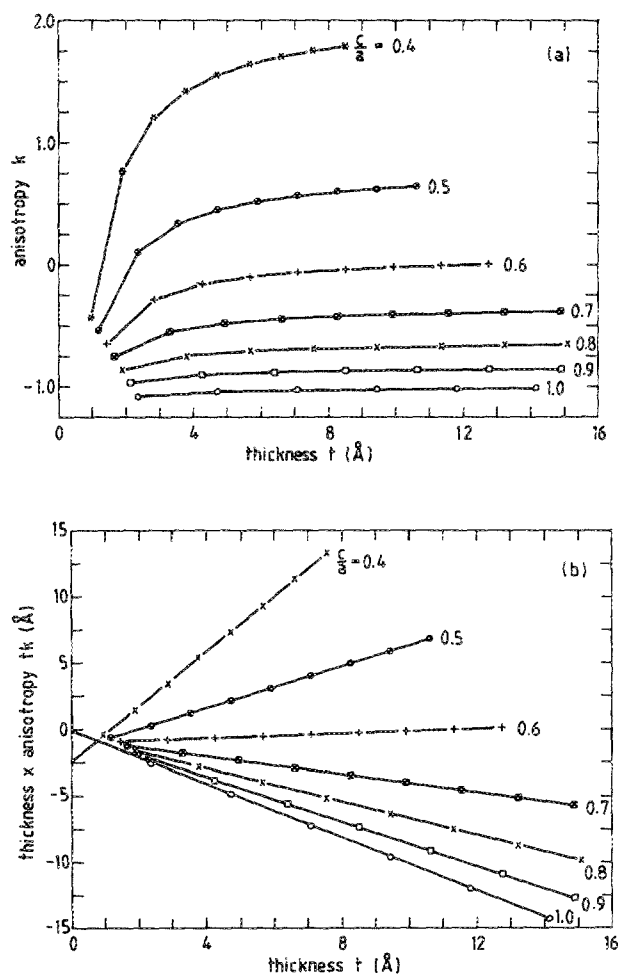


FIG. 4. (a) Anisotropy vs thickness for tetragonal structures with different c/a ratios. (b) Thickness times anisotropy vs thickness for tetragonal structures showing that also in this case a distinction between surface and volume anisotropy can be made.

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