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Size-dependent magnetic properties in nanoplatelets

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Abstract

We demonstrate that the discrete dipolar sums can be separated into two contributions: thickness- and geometry-dependent parts. The geometry-dependent part is analogous to the shape dependence of the continuum approach. The correct normalization of the dipolar energy eliminates the apparent discrepancies of the discrete summation with the experimental results and continuum Maxwell theory. The superposition of the two contributions explains a new phenomenon, i.e. the size-dependent spin reorientation transition and/or enhancement of the effective perpendicular anisotropy.

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1. Introduction

Magnetism at small length scales has lately attracted considerable scientific attention (see for review Ref. [1]). Interesting physical phenomena occur in magnets with all three dimensions on the nanometer scale. We call such structures ultra-low dimensional as they have small but finite dimensions. An array of such ultra-low-dimensional magnetic particles can potentially provide a huge gain in information storage density (see for review Ref. [1]). Hence, the understanding of the micromagnetic ordering in ultra-low-dimensional objects is of high significance for the fundamental physics of magnetic materials as well as for technological applications. The increased ratio of boundary to non-boundary atoms in such structures can lead to unusual physical phenomena.

The orientation of magnetization in a magnet is determined by the balance between the exchange energy, the magneto-crystalline anisotropy and the dipolar energy. The strong exchange interaction tends to line up the magnetic moments in the same direction but does not prefer any orientation in space. In ultra-thin platelets with lateral size L and thickness t, magnetization configurations are mainly determined by the competition between the anisotropy and the dipolar energy. In ultra-thin objects the surface(interface) contribution of the magneto-crystalline anisotropy is often responsible for perpendicular magnetization. The angle dependence of the free energy of such an uniaxial system can be written as $E_{\rm A} =$

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$K_1 \sum_i \sin^2 \theta$, where K_1 is the first-order anisotropy constant and θ is the angle to the film normal [2].

The dipolar interaction is smallest when all magnetic moments compensate each other and the total magnetic charge is equal to zero. The dipolar energy E_D increases whenever magnetic poles are created in a material or at the boundaries. In magnets with $L \gg t$, E_D prefers an in-plane orientation of the moments. The contribution of E_D to the total anisotropy energy is called shape anisotropy. The shape anisotropy of a finite body (ΔE_D) is described by the demagnetizing tensor N: $\Delta E_D = N \cdot 2\pi M_S^2$, where M_S is the saturation magnetization and $2\pi M_S^2$ the shape anisotropy of the infinite continuous magnet. Neglecting the discrete nature of matter, N can be analytically calculated for uniformly magnetized bodies like ellipsoids.

2. Demagnetizing factors in continuum and discrete approach

Sufficiently large and thin, disk-shaped platelets ($L \gg t$) are usually considered to have the demagnetizing factors of an oblate spheroid (special case of ellipsoid).

The demagnetizing factors of such spheroids are well known [3,4]. For an oblate spheroid the shape anisotropy depends only on the ratio $k = L/t$ and can be represented by an universal curve $\Delta E_D = f(k)$ (Fig. 1). For the sake of simplicity, the shape anisotropy energy is normalized with respect to $2\pi M_S^2$ in Fig. 1. ΔE_D deviates from unity only for structures where L and t are comparable.

In literature [5–9], it is argued that in the limit of a few atomic layers the approximation of the film system by a magnetic continuum fails. The ultrathin magnet must be regarded as an assembly of discrete magnetic dipoles on a crystalline lattice. For a laterally infinite discrete lattice of magnetic point-dipoles calculations of the dipolar interactions have shown that the dipolar (demagnetizing) field is not uniform as in the continuous ellipsoid approximation. The dipole field changes with depth and depends on the film thickness. The shape anisotropy of every atomic plane for different lattices has been calculated [7–11]. The

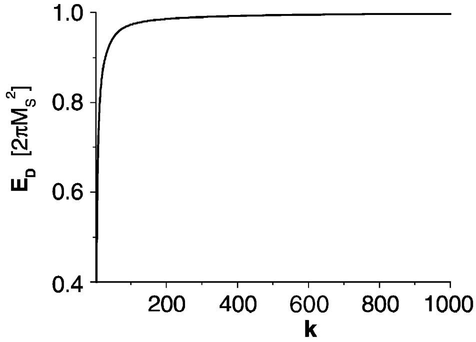

Fig. 1. Analytically calculated magneto-static energy density $\Delta E_D = (N_\perp - N_\parallel) \cdot 2\pi M_S^2$ as a function of the dimensional aspect ratio $k = L/t$ for the oblate spheroid in continuum approximation. The demagnetizing energy is normalized with respect to $2\pi M_S^2$.

average $\Delta\tilde{E}_D$ for a film containing N_{ML} atomic layers has been defined as $\Delta\tilde{E}_D = \sum_{i=1}^{N_{ML}} \Delta E_{D_i}/N_{ML} = c(N_{ML}) \cdot \Delta E_D$. Here $N_{ML} = t/d$ with d for the distance between two successive atomic layers. $\Delta\tilde{E}_D$ of an infinite film can deviate from ΔE_D of a continuum film by more than 10%. The deviation has been attributed to the change of the demagnetizing tensor $\tilde{N} = c(t) \cdot N$. The corrected demagnetizing factors for vertically magnetized infinite ultra-thin films $\tilde{N}_\perp = N_{ZZ}$ are listed in Refs. [7–9]. The in-plane demagnetizing factors are calculated as $\tilde{N}_\parallel = N_{XX} = N_{YY} = (1 - \tilde{N}_\perp)/2$ [8,9] as the sum rule for the demagnetizing field states that the diagonal sum of the demagnetizing tensor is unity inside the sample, i.e. $N_{XX} + N_{YY} + N_{ZZ} = 1$. From data given in Refs. [8,9] one can deduce that the in-plane demagnetizing field (and demagnetizing factors) of an infinite ultra-thin film is no longer zero, which is in disagreement with Maxwell's equations. In Ref. [7] this problem is avoided by assigning the change of the demagnetizing energy as an anisotropy contribution. Nevertheless, the authors claim that the $\tilde{N}$-tensor is thickness dependent. For the simple cubic lattice $\tilde{N}_\parallel$ is even negative as $\tilde{N}_\perp > 1$. All these statements are in contradiction with the continuum theory where the demagnetizing factor is introduced as a geometric parameter. The discrepancies of the continuum theory and the discrete calculations have lead to the opinion that the discrete summation of point-dipole fields can give questionable values of the demagnetization factors [9]. In this paper, we will show the connection between the classical continuous ellipsoid approach and the discrete dipolar model and solve the apparent controversy.

3. Results

We have investigated the size- and thickness dependence of the shape anisotropy in the ultra-thin platelets numerically. The platelets are disks of finite diameter L and thickness $t = N_{ML} \cdot d$ on a discrete lattice. We have considered the samples with dimensional ratio $k \geqslant 40$, i.e. with $L \gg t$. The shape anisotropy has been calculated as the difference between the dipolar energy of the vertical and the in-plane single domain state: $\Delta E_D = E_D(\perp) - E_D(\|)$. The dipolar energy of 1–6 monolayer (ML) thick disks has been calculated by direct lattice summation. Note that the discrete lattice sums are absolutely convergent due to the two-dimensional configuration of dipoles and finite sample dimensions.

The results of the calculations for a triangular lattice with HCP stacking are shown in Fig. 2 as a function of $k = L/t$ for 1–4 ML thick films. The calculated energies are normalized with respect to $2\pi M_S^2$. For other lattices similar results were obtained.

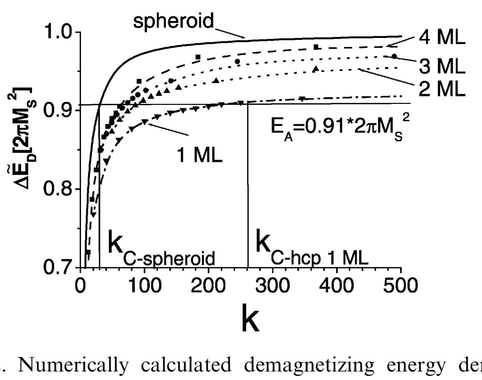

Fig. 2. Numerically calculated demagnetizing energy density $\Delta\tilde{E}_D$ as a function of the dimensional aspect ratio $k = L/d$ for 1–4 ML films on a triangular lattice with HCP stacking. $\Delta\tilde{E}_D$ is normalized with respect to the demagnetizing energy in the continuum limit $2\pi M_S^2$. The straight horizontal line corresponds to the perpendicular magneto-crystalline anisotropy E_A. The dashed vertical lines denote the critical size k_C of the reorientation.

The exact calculation of the dipolar sums deviates strongly from the magneto-static energy obtained from the continuum ellipsoid-ansatz (Fig. 2). The energy depends on thickness and size ($\Delta\tilde{E}_D = f(L,t)$) and not solely on the ratio k. The demagnetizing energy of a platelet of 100×1, for example, is 1.2 times smaller than that of a platelet of 300×3 although $k = 100$ for both and the demagnetization factors should be the same (Fig. 2). $\Delta\tilde{E}_D$ is changing significantly up to high k-ratios ($k \approx 500$) while in the continuous ellipsoid model ΔE_D is almost constant and equal to $2\pi M_S^2$ for large k. With increasing thickness the differences between the individual $\Delta\tilde{E}_D(k,t)$ curves vanish. For $t > 5$ ML $\tilde{E}_D(k,t)$ merges into $\Delta E_D = f(k)$. The function $\Delta\tilde{E}_D = f(k)$ for $t > 5$ ML is close to that of the continuous spheroid. The interpolation of $\Delta\tilde{E}_D = f(L,t)$ to infinite L gives values which are in good agreement with the data given in Refs. [7,8] for infinite expansion. Thus, the rigorous calculation of the dipolar sums reveals that the shape anisotropy is size dependent. The size dependence of $\Delta\tilde{E}_D$ is in disagreement not only with the conventional shape effect $\Delta E_D = f(k)$ but also with the common assumption that the dimensions are only important for $L \approx t$. It has been never considered before that in the range $L \gg t$ the size of the sample can define the magnetic behavior. Next we want to solve the puzzle that emerges from the exact calculation.

4. Discussion

Generally, the total demagnetizing energy ΔE_D is normalized to $2\pi M_S^2 = \text{const}$. The deviation of the total demagnetizing energy from $2\pi M_S^2$ is then attributed to the demagnetizing tensor N: $\Delta E_D = N \cdot 2\pi M_S^2$. Following this assumption, however, means that one has to postulate that N depends not solely on geometry (i.e. ratio k) but also on t, as claimed in Ref. [7], and on L. This assumption is in contradiction to the concept of the demagnetizing factors based on Maxwell theory, as already mentioned.

On the other hand, the dipolar energy of the atoms in the top most layers of the film deviates from that of the bulk atoms. The ratio of boundary to non-boundary atoms in such structures is increased. Consequently, the average demagnetizing energy of an ultra-thin film can deviate from $2\pi M_S^2$ even if N is unchanged. Hence, we may conclude that the discrepancy between the continuous ellipsoid ΔE_D and the discrete $\Delta\tilde{E}_D$ approximation may have different reasons, i.e. $\tilde{N}$ (geometry effect), $(2\tilde{\pi}M_S^2)$ or the combination of both. In Refs. [7–9] it was not possible to distinguish between the three cases as those calculations are related to objects of the same shape, i.e. infinite ultra-thin films ($k=\infty$). In our calculations, the geometry of the sample can be varied from $k=1$ to ∞. Thus, we can analyze all possible situations explicitly.

Taking the assumption $\Delta\tilde{E}_D = \tilde{N}\cdot\Delta E_D$ as in Refs. [7–9] we find $N_{XX}+N_{YY}+N_{ZZ}\neq 1$ for $k<\infty$. This means that the sum rule for the demagnetizing field fails. Assuming $\Delta\tilde{E}_D = (2\tilde{\pi}M_S^2) = X\cdot\Delta E_D$ we find that $(2\tilde{\pi}M_S^2)$ should strongly decrease with decreasing size for a given sample thickness. This outcome makes no sense as for $t=\text{const}$ the dipolar sum differs only for magnetic moments at the sample edge [6] and X should be a constant as long as $L\gg t$. Besides, it is unreasonable to neglect completely the geometry effects. The only remaining explanation of the inequality $\Delta\tilde{E}_D\neq\Delta E_D$ is the superposition of the thickness dependence of the dipolar sums and the geometry effect $\Delta\tilde{E}_D = X\cdot\tilde{N}\cdot\Delta E_D$. In order to decide whether this statement is true or not it is necessary to separate both effects, to compare $\tilde{N}, (2\tilde{\pi}M_S^2)$ with $N, 2\pi M_S^2$, and to check the sum rule for $\tilde{N}$.

In the ellipsoid approximation $N_\perp = 1$ and $N_\parallel = 0$ for laterally infinite ultra-thin films of any thickness. Assuming $\tilde{N}_\perp(\infty) = N_\perp(\infty) = 1$ the ratio $\Delta\tilde{E}_D(L=\infty)/\Delta E_D(L=\infty)$ is nothing else but the factor X. As discussed before, X is a constant for a constant thickness $t\ll L$. Hence, dividing $\Delta\tilde{E}_D$ by X for finite samples of different L but equal t, the pure geometry effect $\Delta\tilde{E}_D/X = \tilde{N}\cdot\Delta E_D$ can be separated.

The normalized curves $\Delta\tilde{E}_D/X = f(k)$ are given in Fig. 3. The functions are identical for all thickness and represent $\tilde{N}(k)$. Thus, an universal curve is found which one must expect from the classical continuum approximation as shape effect. More than that, the demagnetizing factors extracted from $\tilde{E}_D(\perp)/X = \tilde{N}_\perp\cdot 2\pi M_S^2$ and $\tilde{E}_D(\parallel)/X = \tilde{N}_\parallel\cdot 2\pi M_S^2$ give $\tilde{N}_\perp + 2\cdot\tilde{N}_\parallel = 1$ for all L and t, i.e. the sum rule for $\tilde{N}$ is confirmed. The demagnetizing factors are reasonable and close to those of the spheroid. The discrete model, however, gives a geometry dependence that saturates at higher k-values compared to the spheroid model. The reason for the minor difference between $\tilde{N}$ and N is the fact that the discrete model describes precisely the geometry which deviates from that of an ideal ellipsoid. Thus, the rigorous calculation of the dipolar sums for finite ultra-thin magnets are in accordance with the continuum approach.

The discrete summation, however, is more precise as it includes the thickness-dependent inhomogeneity of the dipolar energy while in the ellipsoid approximation $2\pi M_S^2$ is introduced as a constant. The values calculated in Refs. [7–9] are not the demagnetizing factors but the coefficients X as those calculations have been performed for infinite extended films with equal N. We have demonstrated that in contrast to the continuum ellipsoid approximation the demagnetizing energy in finite ultra-thin magnets is a two-variable function $\Delta\tilde{E}_D = f(L,t)$. The discrete dipolar sums can be separated into two contributions:

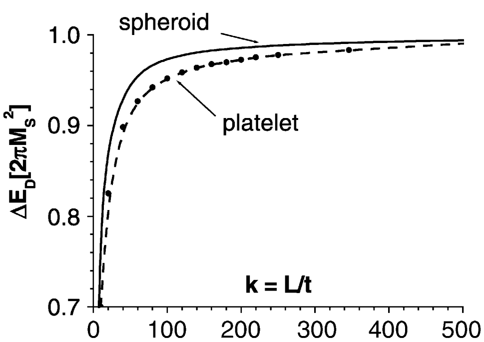

Fig. 3. Comparison of the shape effect of a continuous oblate spheroid and the geometry-dependence extracted from the numerically calculated $\Delta\tilde{E}_D$ of disks with thickness of 1–6 ML on a triangular lattice with the HCP stacking. The demagnetizing energy is normalized with respect to $2\pi M_S^2$.

thickness- and geometry-dependent parts. The geometry-dependent part is analogous to the shape dependence of the continuum approach.

We have checked the geometry effect and t-dependence of the demagnetizing energy for other lattice types. The results are presented in Tables 1 and 2. In Table 1, the demagnetizing factors of the platelets for $40 \leqslant k \leqslant 1000$ are listed. N depends only on the ratio k. The geometry effect does not depend on a lattice type. N is always lower than unity and identical for all lattices. The coefficients X found numerically for $N_{\mathrm{ML}} \leqslant 6$ are given in the Table 2. They depend on the type of the lattice and on thickness. The calculated X-values are in good agreement with values of the "reduced anisotropy" given in Ref. [7]. In contrast to N, the coefficients X can be lower or larger than unity. Thus, neither N nor X are size dependent. However, the superposition of the geometry effect and the thickness dependence of $2\pi M_S^2$ leads to the new behavior, i.e. L and t-dependence of the demagnetizing energy. The L, t-dependence is different for different lattices.

Tables 1 and 2 are universal to find the L, t-dependence of the demagnetizing energy for disks with $40 \leqslant k \leqslant 1000$ and thickness $N_{\mathrm{ML}} = t/d \leqslant 6$. As example, we find the demagnetizing energy of a platelet of diameter $L = 750a$ and thickness $N_{\mathrm{ML}} = 3$ on FCC[1 0 0] lattice. The distance between two subsequent layers is $d = a/\sqrt{2}$ (see Table 2). Hence, $t = N_{\mathrm{ML}} \cdot d = 3a/\sqrt{2}$ and $k = L/t \approx 350$. We find the coefficient $X = 0.922$ from Table 2 and $N_\perp - N_\parallel = 0.984$ from Table 1. Thus, the demagnetizing energy $\Delta\tilde{E}_D = (N_\perp - N_\parallel) \cdot X \cdot 2\pi M_S^2 \approx 0.9 \cdot 2\pi M_S^2$, i.e. 10% less than expected from the continuum theory. Coefficients X for platelets with $N_{\mathrm{ML}} > 6$ can be derived from Ref. [7]. However, the values of X for thicker films have only minor deviations ($<0.2\%$) from the values given for $N_{\mathrm{ML}} = 6$. $X \approx 1$ for $N_{\mathrm{ML}} > 6$. The demagnetizing factors for platelets with $k > 1000$ coincide with those of an oblate spheroid, i.e. are also close to unity [3,4]. Hence, the demagnetizing energy merges into $2\pi M_S^2$ for $N_{\mathrm{ML}} > 6$ and $k \geqslant 1000$.

Table 1
The demagnetizing factors calculated numerically for ultra-thin disks

$k = L/t$	$N_\parallel$	$N_\perp$	$N_\perp - N_\parallel$
1000	0.001	0.998	0.997
600	0.003	0.995	0.992
350	0.005	0.989	0.984
250	0.007	0.985	0.978
220	0.008	0.983	0.975
200	0.009	0.982	0.973
180	0.010	0.981	0.971
160	0.011	0.979	0.968
140	0.012	0.976	0.964
120	0.014	0.973	0.959
100	0.016	0.968	0.952
80	0.019	0.961	0.942
60	0.024	0.951	0.927
40	0.034	0.932	0.898

5. Size-dependent spin reorientation transition

A manifestation of the above model is the size-dependent spin reorientation transition and the apparent enhancement of the perpendicular

Table 2
The thickness-dependent coefficients X calculated numerically for the ultra-thin platelets with thickness $N_{\mathrm{ML}} \leqslant 6$ and the distances d between two successive layers. The apparent differences between the coefficients X for the structures having square lattice at 1 ML (SC(1 0 0), BCC(1 0 0), FCC(1 0 0)) are due to the different lattice constants a

Lattice	$N_{\mathrm{ML}} = 1$	$N_{\mathrm{ML}} = 2$	$N_{\mathrm{ML}} = 3$	$N_{\mathrm{ML}} = 4$	$N_{\mathrm{ML}} = 5$	$N_{\mathrm{ML}} = 6$	$d = \frac{t}{N_{\mathrm{ML}}}$
SC[1 0 0]	1.079	1.039	1.026	1.020	1.016	1.013	a
BCC[1 1 0]	0.924	0.962	0.975	0.981	0.985	0.987	$a\sqrt{2/3}$
BCC[1 0 0]	0.564	0.783	0.856	0.892	0.914	0.929	$a/\sqrt{3}$
FCC[1 1 1]	0.931	0.966	0.977	0.983	0.986	0.988	$a/\sqrt{2/3}$
FCC[1 0 0]	0.765	0.883	0.922	0.941	0.953	0.961	$a/\sqrt{2}$
HCP[0 0 0 1]	0.932	0.966	0.976	0.982	0.986	0.988	$a\sqrt{2/3}$

anisotropy in ultra-low-dimensional objects as reported recently [12]. In ultra-thin objects, the surface/interface anisotropy E_A which often favors out-of-plane magnetization is competing with the dipolar energy E_D which prefers an in-plane orientation. The magnetic anisotropy is a local property and constant for a given thickness. Thus, it can be represented by a straight line in Fig. 2. The intersection of $\Delta\tilde{E}_D$ and E_A gives a critical length $L_C = k_C \cdot t$, where the magnetization orientation switches, i.e. reorientation appears. As the dipolar energy is size dependent the reorientation of the magnetization can take place far beyond the L_C range deduced from the ellipsoid approximation (see Fig. 2).This fact has been confirmed by means of Monte-Carlo simulations [12]. Thus, in contradiction to the analytical ellipsoid assumption the spin reorientation transition in finite ultra-thin platelets is size- and lattice dependent.

6. Conclusions

In conclusion, we demonstrate that the dipolar sum can be separated into two contributions: thickness- and geometry-dependent parts. The geometry-dependent demagnetizing factors found by means of the discrete summation are identical to those found in continuum ellipsoid approximation. The demagnetizing energy of the ultra-thin magnets is size- and lattice dependent. The size- and lattice dependence of $\Delta\tilde{E}_D$ is due to the superposition of the geometry effect and the thickness dependence of the demagnetizing energy. The combination of these two effects leads to a new phenomenon: size-dependent spin reorientation transition and/or an enhancement of the effective perpendicular anisotropy E_{eff} with shrinking size. Critical size L_C of the reorientation can be very large compared to the film thickness.

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