Size-dependent spin reorientation transition in nanoplatelets

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We demonstrate that in nanometer-size magnets the superposition of the lattice dependence of the dipolar energy and the truncation of dipolar sums leads to size- and lattice-dependent effective perpendicular anisotropy. As a consequence, the spin reorientation transition in small platelets of identical shape on different lattices occurs at different sizes for identical anisotropy energy. In contrast to conventional results influences of size on the magnetic behavior can be found even at large aspect ratios of size to thickness.

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The patterning of a continuous magnetic film into an array of small magnetic particles can potentially provide a huge gain in information storage density.[1] The increased ratio of boundary to nonboundary atoms in such nanostructures will lead to changes of physical properties. Hence, the understanding of the influence of the finite size on magnetic behavior in small magnets is of high significance for the fundamental physics of magnetic materials as well as for technological applications.

Theoretically, magnetic materials can be successfully treated as an ensemble of classical magnetic moments S, which are regularly arranged on a crystalline lattice.[2] The configuration of these moments, i.e., the magnetization configuration in the absence of an external magnetic field depends on the balance between the exchange energy, the dipolar energy, and the magnetocrystalline anisotropy.[2] The contribution of the dipolar interaction to the anisotropy energy is called demagnetizing energy or shape anisotropy. In thin films the demagnetizing energy is often responsible for in-plane magnetization. It is usually determined as the difference between the dipolar energy of the up- and the in-plane single-domain states $E_D=\xi_{\uparrow}-\xi_{\rightarrow}$. The infinite continuous magnet has $E_D=\text{const}=2\pi M_S^2$, where M_S is the saturation magnetization. M_S is defined as magnetic moment S per atomic volume V, $M_S=S/V$. We take $V=a^3$, with a the nearest-neighbor distance, for a square lattice that corresponds to the simple cubic stacking and $V=a^3/\sqrt{2}$ for a triangular lattice that corresponds to the hcp(0001) or fcc(111) stacking. The magnetocrystalline anisotropy energy (ξ_A) may be responsible for a perpendicular magnetization. ξ_A depends only on the orientation of the moment with respect to the film normal and does not depend on the neighboring moments. For a uniaxial system with a perpendicular easy axis the angle dependence of the free energy can be written as $\xi_A=K_1\Sigma_i \sin^2\theta$, where K_1 is the first-order anisotropy constant and θ is the angle to the film normal.[2] The total anisotropy energy is defined as $E_A=\xi_{A\uparrow}-\xi_{A\rightarrow}$.

The competition between the demagnetizing and the perpendicular magnetic anisotropy energy determines the magnetization direction. If the relative strength between these quantities is reversed a change of the magnetization orientation will occur. One such phenomenon called the spin reorientation transition (SRT) has been studied for infinite ultrathin films[3–6] and observed experimentally. In Co/Au(111) thin films, for example, a transition from vertical (low thickness) to in-plane magnetization (high thickness) was found around 5 monolayers (ML).[7]

Sufficiently large and thin disc-shaped platelets ($L \gg t$) are usually considered to have the demagnetizing energy of an oblate spheroid (a special case of ellipsoid). The shape anisotropy of such spheroids is well known.[8,9] For an oblate spheroid the shape anisotropy depends only on the ratio $k=L/t$ and can be represented by a universal curve $E_D=f(k)$ (Fig. 1). For the sake of simplicity the shape anisotropy energy is normalized with respect to $2\pi M_S^2$ in Fig. 1. E_D deviates from unity only for structures where L and t are comparable.

The magnetic anisotropy is a local property and is constant for a given thickness. Thus, it can be represented by a straight line in Fig. 1. The intersection of E_D and E_A gives a critical length $L_C=k_C\cdot t$ where the magnetization orientation switches, i.e., reorientation appears. Since the shape anisotropy in ellipsoid approximation deviates from unity only at small k the reorientation can happen only at $L\approx t$ (Fig. 1). Thus, it is commonly assumed that the orientation of mag-

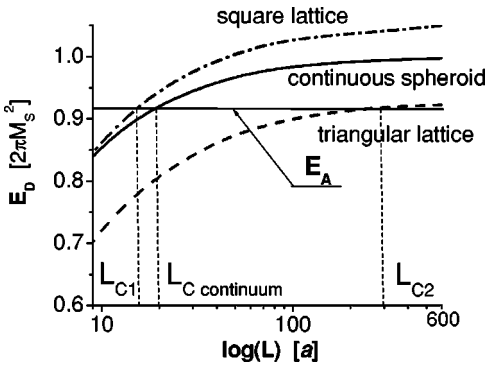

FIG. 1. Comparison of the analytically calculated magnetostatic energy density $E_D=\xi_{D\uparrow}-\xi_{D\rightarrow}$ of a continuum oblate spheroid and the numerically calculated shape anisotropy E_D of a disc on a triangular and a square lattice as a function of the diameter of a spheroid. The demagnetizing energy is normalized with respect to $2\pi M_S^2$. The straight horizontal line corresponds to the perpendicular magnetocrystalline anisotropy E_A. The vertical lines denote the critical sizes L_{C1}, L_{C2}, and $L_{Ccontin}$ of the reorientation for a triangular and square lattice, and an oblate spheroid.

netization in structures with $L \gg t$ depends only on the thickness and the temperature of the sample. If E_A is larger than the demagnetizing energy of the infinite film the reorientation of magnetization will not appear. However, the so-called effective anisotropy $E_{eff}=E_A-E_D$ will increase with shrinking size due to the truncation of the lattice sum.

On the other hand the demagnetizing energy of an infinite monolayer depends on the lattice structure.[3] The superposition of the lattice dependence and the shape dependence of the demagnetizing energy can lead to L_C different from that expected from continuum theory.

This study is devoted to analysis of the validity of the continuum ellipsoid approximation for ultrathin films on a discrete lattice. It turns out that the superposition of two effects—the lattice dependence of the demagnetizing energy and the truncation of dipolar sums—leads to a size- and a lattice-dependent change of the magnetization orientation and an apparent enhancement of the perpendicular magnetic anisotropy.

We have investigated discs of finite diameter L on a discrete lattice by means of Monte Carlo simulations. The Monte-Carlo procedure is the same as in Ref. 6. The Hamiltonian of the problem includes exchange, dipolar interactions, and perpendicular anisotropy: $H=\xi_{ex}+\xi_D+\xi_A$. The ratio of dipolar to exchange constant $D/J \approx 10^{-3}$ used in the calculations corresponds to real materials. Hence, we do not use any rescaling of the sample size. For the chosen D we expect to find a single-domain magnetization configuration in the samples. In that case the exchange energies of an in-plane and an out-of-plane configuration are identical for a collinear solution. First, we prove whether it is also true for the relaxed solution. Then we compare $E_D=f(L)$ and $E_A=f(L)$ for the relaxed and nonrelaxed solutions with the analytical ellipsoid approach.

For the computations we have taken a monolayer of three-dimensional classical magnetic moments S of a unit length on a triangular and a square lattice. We have investigated the low-temperature magnetic microstructure in samples of sizes $100a \leqslant L \leqslant 350a$ where a is the lattice parameter. Thus the lateral size of the platelets has been chosen to be much larger than the thickness t $(L>100t)$.

For $D/J=10^{-3}$ and $L \leqslant 300a$ the exchange energy increase with increasing temperature T is size independent and proportional to $M(T)^2$, with $M(T)$ the magnetization. For given L the exchange energy of the relaxed solution $\xi_{ex}(relax)$ is identical for the up- and the in-plane configurations. This means that the deviation from the collinearity is merely due to temperature fluctuations and not to changes in the magnetic microstructure and ξ_{ex} does not influence the value of L_C. For $D/J>10^{-3}$ or for $D/J=10^{-3}$ and $L \gg 300a$ the microstructure, of the relaxed configuration (especially in plane) deviates from that of the thermally agitated monodomain. Different magnetization patterns can be obtained for different sets of D, K_1, and J. $\xi_{ex}(relax)$ is very sensitive to the type of microconfiguration (vortex, flower, leaf, etc.) and should be taken into account.[10,11] However, that investigation goes beyond the scope of the present paper.

We have explored a wide range of the total anisotropy energy. Here we report on the case in which E_A is slightly smaller than $2\pi M_S^2$, i.e., $E_A \approx 0.9 \cdot 2\pi M_S^2$. In the continuous ellipsoid approximation the selected sizes and anisotropy allow any shape effects to become effective at $L_C \approx 30t$. Hence, in all calculated structures with $L>100t$ an in-plane magnetization configuration should be expected. We have not considered different anisotropies for edge atoms since this goes beyond the scope of our paper.

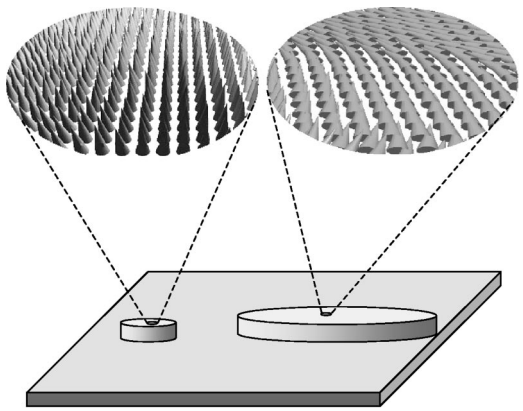

FIG. 2. The low-temperature magnetic microstructure of two discs on a triangular lattice with $L_1=100$ and $L_2=330$; $E_A=0.9(2\pi M_S^2)$. The exchange, the anisotropy, the dipolar energy constants, and the temperature are identical for both samples. For the sake of an appropriate representation a perspective view of an enlarged part of each sample is shown. For clarity, only one spin row out of two is drawn as cones. The smaller island has a vertical single-domain structure. The larger structure presents an in-plane single-domain magnetization configuration.

The results of the simulations for a triangular lattice are presented as magnetization configurations in Fig. 2. Above $L=300a$ the magnetization forms a single domain within the film plane in agreement with the ellipsoid approximation (Fig. 1). Surprisingly, we find a vertical monodomain below $L=230a$. For sizes between $L=230a$ and $L=300a$ intermediate-spin orientations are found. Thus, in contradiction to the analytical approximation the reorientation of the magnetization on a triangular lattice takes place far beyond the k range that is deduced from the ellipsoid approximation. Thus, the results of the Monte Carlo simulations demonstrate that the magnetization direction can change by shrinking the lateral size without changing parameters such as thickness or temperature.

For the square lattice the results are completely different. We find for all structures with $L>100t$ an in-plane single domain in accordance with the ellipsoid approximation. By comparison with the triangular lattice we see that the critical size of the reorientation L_C depends on the type of crystalline lattice.

To find an explanation we have calculated the anisotropy and the demagnetizing energy for a range of sizes L for collinear and relaxed magnetization orientations. In the collinear (nonrelaxed) case the anisotropy cannot be the reason for size-dependent transition since K_1 is a constant in the simulations. The functions $E_D(collinear)(L)$ are presented in Fig. 1. We obtain three different curves $E_D(L)$ for the spheroid and the platelets on the triangular or the square lattice.

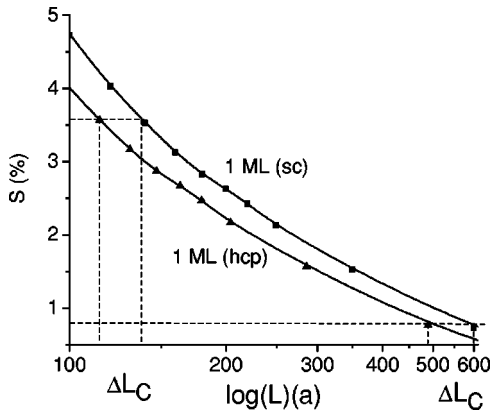

FIG. 3. The deviation of the demagnetizing energy from the saturation value $S=[E_D(L\rightarrow\infty)-E_D(L)]/[E_D(L\rightarrow\infty)]\cdot 100\%$ for a square and a triangular lattice as a function of size. Dashed vertical lines denote the hypothetical sizes at which S is identical in both lattices. ΔL_C denotes the shift of those sizes in different lattices.

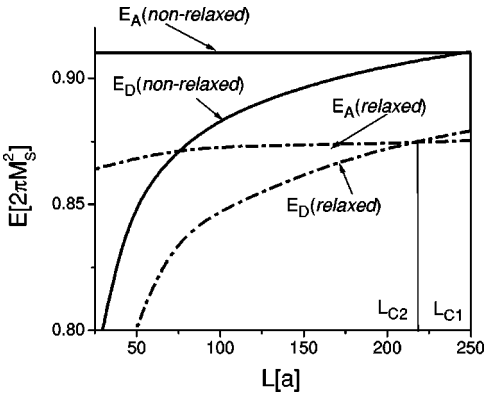

FIG. 4. Comparison of the demagnetizing E_D and the anisotropy E_A energy of a disc on a triangular lattice as a function of size for strictly collinear and relaxed solutions. All energetic parameters J, D, and K_1 are identical in both cases. The energy is normalized with respect to $2\pi M_S^2$, $kT/J=0.05$, and $D/J=10^{-3}$. The vertical lines denote the critical sizes L_{C1} and L_{C2} of the magnetization reorientation for collinear and noncollinear configurations.

For the triangular lattice $E_{D\Delta}(L)$ lies below $2\pi M_S^2$. For the square lattice $E_{D\square}(L)$ is larger than $2\pi M_S^2$ already for $L\approx 60a$. The critical size of the SRT can be derived from the data of Fig. 1 for a given value of E_A. For the case in which reorientation should happen, i.e., $E_A<2\pi M_S^2$ (horizontal line in Fig. 1) we might find one critical size $L_C=L_C(continuum)$ in all platelets. L_C of the triangular lattice, however, varies by more than a factor of 10 from that expected from the ellipsoid approximation and that for the square lattice ($L_{C\Delta}>10\cdot L_{C\square}$). For a different value of E_A one can get different values of critical size but $L_{C\Delta}$ will never equal $L_{C\square}$ as expected from the ellipsoid approximation. The slope of the $E_D(L)$ curves is different for square and triangular lattices. At large sizes the difference is less than 1% (see Fig. 3). However, even such small deviations lead to the remarkable shift of the critical size $\Delta L_C\approx 100a$ for large sizes while $\Delta L_C\approx 25a$ for smaller sizes despite the larger difference in curvature (Fig. 3).

E_D and E_A of the noncollinear solution due to thermal disorder are smaller than those of the collinear case. Figure 4 gives $E_D(L)$ and $E_A(L)$ of platelets on a triangular lattice for strictly collinear and noncollinear solutions. The dipolar and the anisotropy energies exhibit different temperature dependencies which is exactly the reason for the temperature-induced magnetic reorientation in a ferromagnetic monolayer. Interestingly, the anisotropy energy of the relaxed solution is no longer a constant but is size dependent. As a consequence L_C is shifted to smaller sizes comparably to the collinear case.

Thus the critical size of the reorientation is dependent on the lattice type and can be very large comparably to the thickness of the sample. This documents that the size dependence of the reorientation transition in discrete lattices is not due to the shape effect of the continuous model that depends on the ratio of the object dimensions. The effect found for the monolayer example is even more pronounced in thicker samples due to the thickness dependence of the demagnetizing energy of platelets on a discrete lattice.[13]

For $E_{D\Delta}(L\rightarrow\infty)<E_A<E_{D\square}(L\rightarrow\infty)$ the reorientation of magnetization will appear only in the platelet on a square lattice. The effective perpendicular anisotropy of a triangular lattice will increase due to the shape and the lattice dependence of E_D. This is sometimes erroneously interpreted as the increase of perpendicular magnetic anisotropy with shrinking size, since E_D is commonly assumed to be constant. Experimental findings pointing in this direction have been published recently.[14]

The size and lattice dependencies of the shape anisotropy arise from the inhomogeneity of the dipolar energy in ultrathin ferromagnets.[12] The dipole field in such magnets changes with depth and depends on the film thickness.[12] The dependence of the dipolar energy on the lateral position of an atom is just a consequence of the long-range character of the interaction. As the ratio of boundary to nonboundary atoms increases an influence of the inhomogeneous demagnetizing field on the shape effect appears.

In conclusion, we demonstrate that in laterally confined ultrathin magnetic structures the magnetic behavior depends on the type of the lattice and the sample size. As a consequence, the spin reorientation transition in small platelets of identical shape on different lattices occurs at different sizes for identical anisotropy energy. For $E_D<E_D(L\rightarrow\infty)$ the reorientation from an in-plane configuration for larger sizes to an out-of-plane configuration below a critical size L_C occurs. L_C can be very large compared to the film thickness. We have shown that an enhancement of the effective perpendicular anisotropy E_{eff} can occur with shrinking size.

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