Theory for the magnetic phase diagram of thin films: Role of domain formation

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The magnetic phase diagram of a thin film is determined at $T=0$. The exchange coupling, the magnetic dipole coupling, as well as the second- and fourth-order lattice anisotropies are taken into account. The long-range dipole coupling causes an instability of the uniform magnetization with perpendicular orientation and leads to a domain phase. Four different phases, in particular stripe domain structures with perpendicular and canted orientations of the domain magnetization, are found to be stable. The parameter region near the magnetic reorientation is studied in greater detail. It is shown that the higher-order lattice anisotropies may induce a continuous reorientation from a perpendicular to an in-plane magnetization, also for a domain phase. By carrying out lattice sums we conclude that a periodic array of straight domain walls should be stable against spatial dislocations. Our results suggest that the domain phases may alter the nature of the phase transitions at finite temperatures in thin films. Rather than phase transitions with critical phenomena a simple rotation of the magnetization might occur.

I. INTRODUCTION

Recently, the magnetic properties of ultrathin ferromagnetic films have been studied intensively both experimentally and theoretically.[1] A two-dimensional (2D) Heisenberg magnet with isotropic interactions only should not exhibit any long-range order at finite temperatures.[2] However, due to anisotropic interactions which are always present in magnetic systems an ordered state may occur with a Curie temperature T_C of the order of the strong exchange coupling.[3] Indeed, ferromagnetic thin films with a few atomic layers are observed at room temperatures.

Furthermore, the anisotropic interactions determine the direction of the magnetization relative to the atomic lattice. A rotation from a perpendicular to an in-plane magnetization with increasing temperature has been observed for a number of thin-film systems like Fe/Cu(100),[4–6] Fe/Ag(100),[7] Co/Au(111),[8] and others.[9] The reason for this magnetic reorientation is the larger entropy for the in-plane magnetization,[10,11] which becomes more important at higher temperatures. In addition, the reorientation is accompanied by a region of strongly reduced or even vanishing global thin-film magnetization. In a previous publication[12] we have investigated this loss of the global magnetization, which, in principle, could result from a mutual cancellation of anisotropic interactions near the magnetic reorientation. Then the long-range magnetic order may disappear due to the vanishing remanent magnetization. The width of this vanishing magnetization ΔT was estimated by a simple scaling law.[13] However, we have shown that the calculated size of ΔT is comparable to the measured one only if the reorientation temperature is close to T_C. Thus, in general different mechanisms have to be considered which cause the loss of the global magnetization. An alternative explanation for this behavior is due to the appearance of a magnetic domain structure. Such a phase exhibits a finite remanent but a vanishing global magnetization. Experiments with an improved spatial resolution have observed such a domain structure.[5] The theoretical investigation of domain phases in presence of long-range interactions have attracted much effort recently.[14–20]

It is not known exactly whether the magnetic reorientation at finite temperatures represents a phase transition or not.[21] Around a second-order phase transition the free energy exhibits a nonanalytical behavior leading to critical fluctuations of the order parameter. In a magnetic-field induced reorientation a strong increase of inelastically scattered light was observed.[22] However, it is not clear whether this property is caused by a magnetic domain structure or by critical phenomena. Calculations obtain a strong increase of transversal fluctuations, leading to a diminished magnetization.[23] However, we would like to emphasize that the consideration of additional phases like domain structures may alter the nature of the reorientation transition.

The main anisotropic interactions in thin magnetic films are the following. First, the long-range magnetic dipole interaction (shape anisotropy, demagnetizing field) is present which prefers always an in-plane magnetic orientation. The dipole energy per unit area of a thin film is almost proportional to the film thickness. Secondly, lattice anisotropies are present originating from the relativistic spin-orbit coupling. Due to the broken cubic symmetry for surface or interface layers a second-order anisotropy is induced which may be 100 times larger than in bulk.[24] Since such a strong anisotropy is confined only to the surface or interface layers, only for very thin films this anisotropy may be strong enough to overcome the dipole coupling, causing then a perpendicular orientation of the magnetization. Consequently, a magnetic reorientation from perpendicular to in-plane direction of the thin-film magnetization is observed for increasing film thickness.[5–8]

A peculiar feature of the thin-film magnetization is that the long-range property of the dipole interaction leads to an instability of the uniform perpendicular magnetization.[14–17] The ferromagnetic phase breaks up into a periodic structure

of striped shaped regions with uniform magnetic order and alternating orientation along the film normal (stripe domains). The periodicity and the wall width are found to depend sensitively on the involved coupling parameters. A recent calculation has shown that with increasing temperature the magnetic structure of a thin film may undergo a series of phase transitions corresponding to different domain structures.[18]

In previous calculations of the direction of the magnetization only the dipole coupling and the second-order surface anisotropy were considered. However, it is known that also the higher-order lattice anisotropies have an important influence on the magnetic phases and also on the shape of magnetic domain walls.[9] For example, an uniform magnetization with a canted orientation may exist for certain values of the second- and fourth-order anisotropies. Therefore, in order to determine the global magnetic phase diagram of thin films, we consider in this publication higher-order anisotropies as well as different domain phases. In particular, the phase boundaries are calculated near the magnetic reorientation, since we expect that in this region domain phases corresponding to a vanishing global magnetization are most likely to occur. We limit here our calculations to $T=0$, the case $T>0$ will be studied separately. In Sec. II we present the theory and the method of calculation. The results are outlined in Sec. III. A discussion is given in Sec. IV. In the Appendix we calculate the influence of fluctuations on uniform as well as on domain phases. By performing lattice summations we conclude that a domain phase with an array of straight domain walls is stable at $T=0$.

II. THEORY

We consider a (100) thin film with the film normal parallel to the z axis (uniaxial symmetry). For simplicity we assume a magnetic monolayer first, corresponding to a 2D square lattice with coordination number $q=4$. $\vec{S}_i$ is a three component classical vector spin on lattice site i, $|\vec{S}_i|=1$. The components of $\vec{S}_i$ are given by the polar and azimuthal angles θ_i and ϕ_i: $\vec{S}_i=(\sin\theta_i\cos\phi_i, \sin\theta_i\sin\phi_i, \cos\theta_i)$. We take the following Hamiltonian:

$$\mathcal{H}=-\frac{J}{2}\sum_{\langle ij\rangle}\vec{S}_i\vec{S}_j+\frac{(g\mu_B)^2}{2}\sum_{\substack{ij\\ i\neq j}}\left(\frac{\vec{S}_i\vec{S}_j}{r_{ij}^3}-\frac{3}{r_{ij}^5}(\vec{S}_i\vec{r}_{ij})(\vec{S}_j\vec{r}_{ij})\right)$$
$$-\sum_i\left(K_2\cos^2\theta_i+K_4\cos^4\theta_i\right.$$
$$\left.+\frac{K_s}{4}\sin^4\theta_i(\cos 4\phi_i+3)\right). \quad (1)$$

Here, J is the isotropic Heisenberg exchange coupling, $\langle ij\rangle$ denotes the sum over nearest-neighbor pairs. The last term is the (local) lattice anisotropy, with K_2 and K_4 are the second- and fourth-order uniaxial anisotropy constants. K_s is the quartic in-plane anisotropy of the (100) face. We consider here $K_s>0$ only, i.e., the x and y directions are the easy axes in the film plane ($\phi_i=n\pi/2$). At $T=0$ the in-plane anisotropy K_s produces only a shift of the magnetic phase boundaries in the phase diagram. However, for $T>0$ this term is very important, since it stablizes the in-plane magnetization with $\theta=\pi/2$. The second term represents the magnetic dipole coupling, g is the Landé factor, and μ_B is the Bohr magneton. The vector $\vec{r}_{ij}$ connects the lattice sites i and j. Specific care should be taken with the dipole sums, especially with oscillating ones which appear in the case of periodic (domain) phases. Since the energy of the different phases has to be calculated with high accuracy, the slowly converging lattice sums are replaced by rapidly converging ones via the Ewald summation.[25] Then the lattice sums are given by the following quantities:

$$\mathcal{S}(p,q)=\sum_{\substack{u,v=-\infty\\ u\neq v}}^{+\infty}\frac{u^2}{r^5}e^{ipu}e^{iqv}$$
$$=\frac{16}{3}\sum_{m=1}^{\infty}\sum_{l=-\infty}^{+\infty}(\pi l+q/2)^2\cos(pm)$$
$$\times\mathcal{K}_2(2m|\pi l+q/2|), \quad (2)$$

with $r^2=u^2+v^2$. $\mathcal{K}_2(x)$ is the modified Bessel function of the order 2. For latter purpose we define the demagnetizing energy for a square lattice $E_0=3\mathcal{S}(0,0)(g\mu_B)^2/(2a_0^3)=2\pi c(g\mu_B)^2/a_0^3$, which is the difference of dipole energy between uniform perpendicular and in-plane magnetization, a_0 the lattice constant, and $c=1.078\,309$. A Zeeman term $-\vec{H}\Sigma_i\vec{S}_i$ may also be included into the Hamiltonian, Eq. (1).

For the calculation of the magnetic phase diagram we take into account different magnetic phases. The uniform phases with different orientations are characterized by the angle $\theta_i=\theta_u$ between film normal and magnetization: $\theta_u=0$ denotes the perpendicular, $\theta_u=\pi/2$ the in-plane, and $0<\theta_u<\pi/2$ the canted uniform magnetization. In the case of an uniform phase the dipole contribution represents merely a local field with strength E_0 and may be absorbed into the second-order lattice anisotropy K_2. In addition periodic stripe domain phases with site-dependent polar angles θ_i are considered. For this purpose we adopt the domain profile as used by Yafet and Gyorgy.[15] According to this model the direction of magnetization varies periodically along the y direction with periodicity $2L$, and is uniform along the x direction. Inside the domains the magnetization is directed along the $\pm z$ direction ($\theta_d=0$). This phase we call "perpendicular domain phase." In addition, the direction of the domain magnetization may be tilted away from the z axis by the angle θ_d, this phase will be denoted by "canted domain phase." The ordered magnetic domains are separated by 180°-Bloch-type walls, i.e., the magnetization inside the walls is confined to the xz plane and exhibits no component along the wall normal, which is the y direction ($\phi_i=0$): $\vec{S}_i=(\sin\theta_i,0,\cos\theta_i)$. For the domain-wall shape with width b we assume for simplicity a cos function.[15] Then the profile of the periodic domain structure with $\cos\theta_i=\cos\theta(y)$ is given by

$$\cos\theta(y)=\begin{cases}\cos\theta_d, & 0\leq y\leq (L-b)/2\\ \cos(\theta_d+\pi(2y+b-L)/2b), & (L-b)/2\leq y\leq (L+b)/2\\ \cos(\theta_d+\pi), & (L+b)/2\leq y\leq L\end{cases}$$

and symmetrically for $-L\leq y\leq 0$. The restrictions resulting from the assumption of this special shape will be discussed later. The domain phases are characterized by the canting angle θ_d ($0\leq\theta_d\leq\pi/2$), the periodicity $2L$, and the wall width b. The energy of the domain phase is minimized according to these three parameters. The minimization procedure is simple, since the energy difference between domain and uniform magnetic phases shows a smooth (parabolic) dependence on all three parameters L,b,θ_d, exhibiting a single (global) minimum. We consider only the case of strong exchange coupling and comparable weak anisotropy (Heisenberg spins), which is appropriate to $3d$ transition metals. Then the periodicity L and the wall width b are much larger than the lattice constant a_0, and a continuum model is suitable. Since the domain phase is periodic, we apply a Fourier transformation for the calculation. Typical numbers of Fourier coefficients [n sum in Eq. (4)] as well as of the terms of the l and m sums for the quantities $\mathscr{S}(p,q)$, Eq. (2), are $l\simeq 10$–30 and $m,n\simeq 100$–300. These numbers are limited by the accuracy to which the Bessel function $\mathscr{K}_2(x)$ is known to us, i.e., $\epsilon\approx 10^{-7}$.[26] Special care has to be taken for small values of p for which considerably larger numbers of terms have to be considered. For special parameter values L,b,θ_d we have checked our results by performing also the direct lattice sums.

As suggested by the calculations in the Appendix with the help of discrete lattice summations, the stripe domain phase with straight Bloch walls is stable against infinitesimal fluctuations at $T=0$, since the long-range dipole interaction and, of course, the exchange coupling favors straight domain walls. Note that this result was also obtained by Abanov *et al.*,[18] but is in contrast to the findings of Chui.[19] Anyway, in the following we will assume straight domain walls for our calculations, since the main goal of this paper is to study the effect of the long-range dipole coupling, the higher-order lattice anisotropy terms, and the resulting domain formation on the magnetic phase diagram of a thin film at $T=0$. As was already pointed out in Ref. 19, straight domain walls would result also from impurity pinning.

III. RESULTS

We now determine the different magnetic phases of the monolayer in the (K_2,K_4) plane for constant $K_s>0$ at $T=0$. The energies are given in units of the demagnetizing (dipole) energy E_0. In all our calculations the exchange coupling is assumed to be $J/E_0=25$, appropriate for $3d$ transition metals.[15] In addition we set $K_s/E_0=0.01$. First, we review briefly the uniform magnetic phases. The energy per spin of these phases is given by

$$E_{\rm uni}(\theta_u)=-\frac{q}{2}J-K_2\cos^2\theta_u-K_4\cos^4\theta_u-K_s\sin^4\theta_u+E_0\left(\cos^2\theta_u-\frac{1}{3}\right). \tag{3}$$

It follows that the perpendicular ($\theta_u=0$) and the in-plane magnetization ($\theta_u=\pi/2$) exist everywhere in the (K_2,K_4) plane, the latter being more stable for $K_2<E_0+K_s-K_4$ [lower left part of the (K_2,K_4) plane, see Fig. 1]. At $K_2=E_0+K_s-K_4$ a discontinuous (first-order) transition from perpendicular to in-plane magnetization occurs. In addition a canted phase exists in a limited part of the (K_2,K_4) plane determined by $|E_0+2K_s|\leq|K_2|\leq|E_0-2K_4|$. For $K_4<-K_s$ this phase is more stable than the perpendicular and the in-plane magnetization. The canting angle is given by $\cos^2\theta_u=(E_0+2K_s-K_2)/(2K_s+2K_4)$. Continuous (second-order) transitions occur at $K_2=E_0-2K_4$ from perpendicular to canted magnetization, and at $K_2=E_0+2K_s$ from canted to in-plane magnetization. At finite temperatures these correspond to second-order phase transitions, where critical fluctuations are expected to occur.

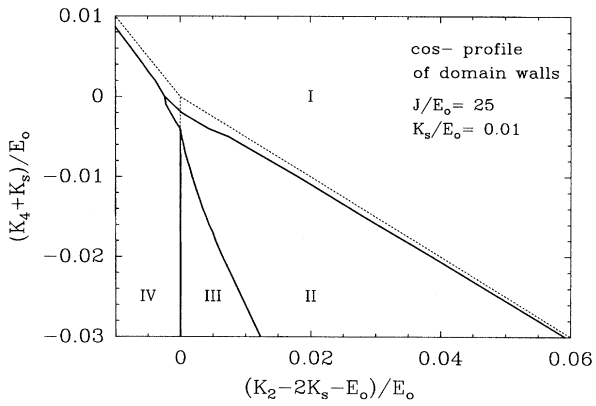

FIG. 1. Magnetic phase diagram of a thin film at $T=0$ in the (K_2,K_4) plane. J is the exchange coupling, E_0 is the demagnetizing energy. K_2 and K_4 denote the second- and fourth-order uniaxial, K_s is the quartic in-plane anisotropy constants. Assuming a periodic domain structure with a cos profile of the domain walls, four different magnetic phases are obtained to be stable (bold lines): domain phases with a perpendicular (I) and a canted (II) orientation of the domain magnetization, and uniform phases with a canted (III) and an in-plane (IV) magnetization. For comparison the transition lines between magnetic phases are shown if only uniform phases are considered (dotted). The energies are given in units of E_0. We have assumed $J/E_0=25$, appropriate for $3d$ transition metals (Ref. 15), and $K_s/E_0=0.01$.

A metastable canted magnetization does not exist beyond the phase boundaries. Note that the canted phase removes the discontinuous transition from perpendicular to in-plane orientation for $K_4<-K_s$. At $T=0$ a canted phase exists only for a nonvanishing fourth-order anisotropy $K_4+K_s<0$. In the case of uniform phases the dipole coupling causes solely a shift of the phase boundary lines parallel to the K_2-axis, see Fig. 1.

Now we consider in addition to the uniform magnetic phases also stripe domain structures as described in Sec. II. The energy per spin of the domain phases with periodicity $2L$ and wall width b is given by

$$E_{\mathrm{dom}}(\theta_d)=-\frac{J}{2}\left(q-\frac{\pi^2}{bL}\right)-K_2\left[\left(1-\frac{b}{L}\right)\cos^2\theta_d+\frac{b}{2L}\right]-K_4\left[\left(1-\frac{b}{L}\right)\cos^4\theta_d+\frac{3b}{8L}\right]-K_s\left[\left(1-\frac{b}{L}\right)\sin^4\theta_d+\frac{3b}{8L}\right]$$
$$+\frac{E_0}{6\mathscr{S}(0,0)}\sum_{n=1,3,\ldots}^{\infty}c_n^2\left[(1+\alpha_n^2)\mathscr{S}\left(\frac{\pi n}{L},0\right)+((1-3\sin^2\theta_d)+\alpha_n^2(1-3\cos^2\theta_d))\mathscr{S}\left(0,\frac{\pi n}{L}\right)\right], \quad (4)$$

where

$$c_n=(-1)^{\frac{n-1}{2}}\frac{4}{n\pi(1-\alpha_n^2)}\cos\left(\frac{n\pi b}{2L}\right),\ (n\ \text{odd}) \quad (5)$$

are the Fourier coefficients of the domain profile, and $\alpha_n=nb/L$. As was already shown by Yafet and Gyorgy for $K_4=K_s=0$, a stripe domain phase with a perpendicular orientation of the domains is more favorable than the uniform perpendicular magnetization for large values of $K_2>E_0$.[15] This is also true for finite values of K_4 and K_s. For $K_4>-K_s$ a discontinuous transition occurs from the perpendicular domain phase to the uniform in-plane magnetization with decreasing K_2. On the other hand for $K_4<-K_s$ the perpendicular domain phase changes to a canted domain structure for decreasing K_2. The canting angle θ_d raises and finally reaches the value θ_u of the canted uniform magnetization. The resulting magnetic phase diagram in the (K_2,K_4) plane is shown in Fig. 1. The transition lines in presence of domain phases are shifted slightly to smaller values of K_2 compared to the case of uniform phases only. However, a new transition line is introduced which separates the canted domain from the canted uniform phase. We have shown in Fig. 1 a comparably small but interesting region of the (K_2,K_4) plane near the magnetic reorientation. Here the second-order lattice anisotropy and the dipole coupling nearly cancel, since in this region a domain phase exhibiting a vanishing global magnetization is most likely to occur. In addition, the small shift of the phase boundaries due to domain structures is visible.

The behavior of the periodicity L and the wall width b as a function of K_2 is different for the regions $K_4>-K_s$ and $K_4<-K_s$. Within the perpendicular domain phase the periodicity L raises almost exponentially with increasing K_2, whereas the wall width b decreases slightly. For decreasing K_2 the domain phase reaches the special case $L=b$, i.e., a pure cosine-like modulated phase. However, before this special domain phase is reached, for $K_4>-K_s$ the uniform in-plane magnetization becomes energetically more favorable, and a discontinuous transition occurs. In contrast, the dependence of L and b on K_2 for $K_4=-0.02E_0<-K_s$ is shown in Fig. 2. When the canted domain phase is reached, L does not decrease any longer with decreasing K_2 but stays almost

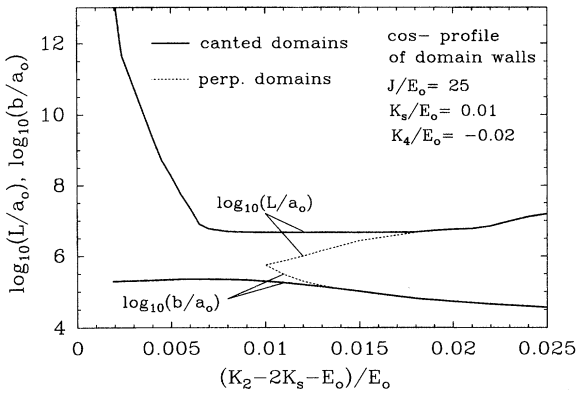

FIG. 2. Resulting periodicity L and wall width b of a periodic domain structure as a function of K_2 for $K_4/E_0=-0.02$. The dotted lines denote L and b of a domain phase with a perpendicularly oriented domain magnetization. Note that for $(K_2-2K_s-E_0)/E_0<0.002$ the (canted) domain phase vanishes. For the other notations we refer to Fig. 1.

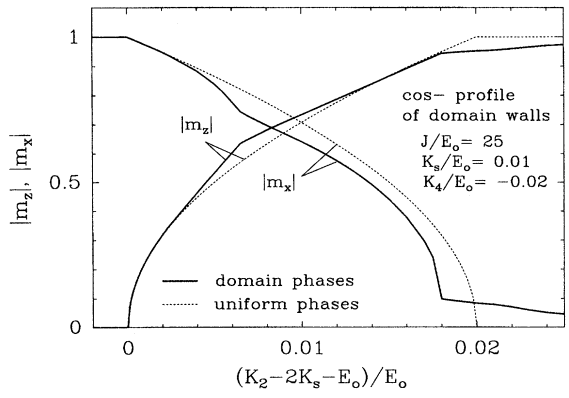

FIG. 3. Absolute values of the perpendicular ($|m_z|$) and in-plane ($|m_x|$) components of the magnetization as a function of K_2 for $K_4/E_0=-0.02$. For comparison the dotted lines show m_z and m_x with consideration of uniform phases only. For the other notations we refer to Fig. 1.

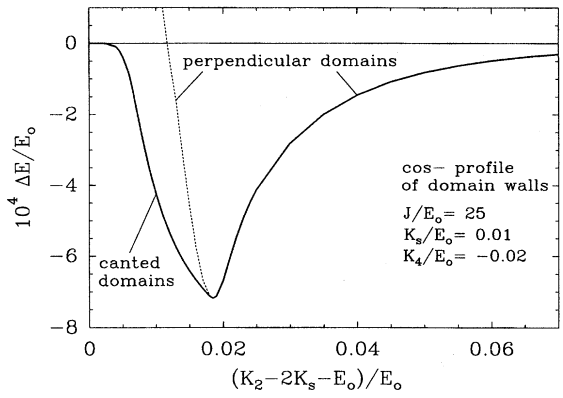

FIG. 4. Energy difference ΔE in units of $10^{-4}E_0$ between periodic domain phases and the uniform magnetization as a function of K_2 for $K_4/E_0=-0.02$. The domain phase with perpendicularly oriented domains, which is stable for $(K_2-2K_s-E_0)/E_0>0.02$, becomes metastable for $(K_2-2K_s-E_0)/E_0<0.02$ (dotted line). In this region the canted domain phase is most stable, which finally merges into the uniform in-plane magnetization, characterized by $\Delta E=0$. For the other notations we refer to Fig. 1.

constant. Then L raises strongly again and diverges at the value of K_2 where the canted uniform magnetization emerges. Thus the canted domain phase merges smoothly into the canted uniform phase. In Fig. 3 we show the resulting absolute values of the perpendicular and in-plane magnetization components $|m_z|$ and $|m_x|$, together with m_z and m_x of the uniform phase. We emphasize that with consideration of the domain phases an in-plane component $|m_x|$ is always present, whereas the perpendicular component $|m_z|$ appears only for $K_2>E_0+2K_s$. Thus, we expect a different phase transition behavior at finite temperatures for the transitions from in-plane to canted magnetization compared with canted to perpendicular. In Fig. 4 the energy difference ΔE between the uniform magnetization and the domain phase is shown for $K_4/E_0=-0.02$. This binding energy of the domain phase is quite small, so that already small applied magnetic fields will recover the uniform phases.[13] Around the transition of the perpendicular domain phase to the in-plane uniform magnetization ($K_4>-K_s$) or to the canted domain phase ($K_4<-K_s$) the value of ΔE is maximal, consequently in these regions domain phases are most likely to occur, as observed experimentally.[5]

IV. DISCUSSION

We have investigated the magnetic phase diagram of a monolayer, which is expected to reflect also the magnetic properties of a thin film with several atomic layers. The exchange coupling, the magnetic dipole interaction, and the second- and fourth-order uniaxial anisotropies are taken into account. The perpendicular uniform magnetic phase vanishes completely in favor of a domain structure with a perpendicularly oriented domain magnetization. As was already shown for $K_4=K_s=0$,[15] this is caused by the long-range property of the dipole coupling leading to an instability of the perpendicular uniform magnetization. A periodic stripe domain phase emerges with varying periodicity and wall width. Since the authors of Ref. 15 used an approximate evaluation of the dipole sums, we have recalculated their model and found almost the same results. In the presence of a fourth-order uniaxial anisotropy $K_4<0$ also a domain phase with a canted orientation of the domains is stable in a certain region of the phase diagram. A domain phase with in-plane oriented domains ($\theta_d=\pi/2$) is found to be unstable. The existence of a canted domain phase can explain the experimental observation of a stripe domain structure in both the perpendicular and in-plane components of the magnetization.[5] The domain phases are observed preferentially near the magnetic reorientation, since here the domain binding energy ΔE is largest, see Fig. 4. Weak magnetic fields may stabilize the uniform phases outside the reorientation region, leading to an asymmetric shape of the global magnetization near the magnetic reorientation,[13] as observed experimentally.[7]

The long-range dipole interaction stabilizes the magnetic domain walls in a thin film. As can be seen in Fig. 2, the wall width b of the periodic domain structure stays finite near the magnetic reorientation. In contrast, the width b_0 of a single Bloch wall will diverge at $K_2=E_0+2K_s$, since b_0 is given approximately by $b_0\propto\sqrt{J/(K_2-2K_s-E_0)}$.[27] For $K_2\approx E_0+2K_s$ the energy minimum of the domain phase $E_{\rm dom}$ is characterized by $L\approx b$. The case of a single wall requires $L\to\infty$, thus in turn also $b=b_0\to\infty$.

On the basis of lattice summations outlined in the Appendix, we conclude that a periodic stripe domain structure with straight domain walls is stable against a transversal dislocation at $T=0$. This is in accordance with Ref. 18, but disagrees with the result obtained by Chui.[19] We are presently not able to solve this discrepancy. Possibly, this disagreement may be caused by rewriting sums as integrals as was done in Ref. 19. Also the leading term of the dipole energy difference for small wave vectors $|\delta E_{\rm dip}|\propto k^2|\ln k|$ as obtained in Ref. 19 seems to be a property of the continuum approximation. This term was derived for the edge of a semi-infinite dipole monolayer, corresponding to an infinitesimal domain-wall width. We were not able to derive such a behavior of $\delta E_{\rm dip}$ from the discrete lattice summation. However, we expect that the much stronger difference in exchange coupling $\delta E_{\rm ex}>0$, see the Appendix, will maintain straight domain walls, which are observed experimentally.[5] In the case of deformed domain walls we expect a slight shift of the phase boundaries of the magnetic phase diagram, see Fig. 1, and larger changes of the periodicity L and the wall width b. At finite temperatures a magnetic domain phase with deformed walls may be the most stable phase, since then fluctuating domain walls may be induced on entropical grounds.[18] Our result for $T=0$ is also in agreement with the results of McConnell,[16] who concluded that an array of narrow rectangular stripes of electric dipoles should be stable against spatial fluctuations. It is quite interesting to remark that in an electric dipole monolayer, in a 2D ferroelectric system, or in a 2D ferro-fluid, the long-range dipole interaction should be much more important for the ordering properties in these systems, since here a strong exchange coupling is, in general, not present. However, the line tension at the edges of these systems act like an exchange coupling or an anisotropy. Long-range dipole interactions may cause distorted uniform phases as stated in Ref. 16, and as also indicated by us in the Appendix.

We have calculated the magnetic phases of a monolayer only, since we expect that the general features emerge already for a single layer. For thin films with a finite number of atomic layers the dipole energy as well as the anisotropic contributions have to be summed over the layers. A strong surface anisotropy K_2 is confined only to the surface or interface layers, whereas the dipole energy is almost the same for each layer. Therefore, the second-order lattice anisotropy energy per unit area is $2K_2 \approx$ const, whereas the dipole energy per unit area is $\approx E_0 d$ for increasing film thickness d, resulting in a shift of the phase boundaries in the (K_2, K_4) plane of the phase diagram parallel to the K_2 axis, see Fig. 1. For thicker films the dipole coupling dominates, resulting in an uniform in-plane magnetization. However, in the case of a strained film a strong second-order strain anisotropy comparable to the strength of the dipole coupling may exist also in the inner layers. This may lead to a reversed magnetic reorientation behavior, namely from an in-plane to a perpendicular magnetic reorientation with increasing film thickness. This unusual feature is observed in Ni/Cu(100) films.[28]

In addition we have considered domain structures with different wall profiles. For example, we have calculated a profile described by a tanh function, which is the correct profile for a single Bloch wall in bulk.[27] The energy calculated for such a structure is lower than for the cos-type domain wall, as expected physically, whereas the overall features are the same. Thus, we expect that our results obtained for the easily calculated cos profile resembles correctly the magnetic phase diagram of a thin film. The general profile of the periodic stripe domain structure may be obtained by minimizing the energy, Eq. (4), with respect to the quantities c_n and α_n. A square instead of a stripe domain pattern was calculated to be stable only at unphysically large dipole couplings and anisotropies.[17] We have also considered magnetic domain phases with walls exhibiting a magnetization component along the wall normal (Néel walls, $\phi_i \neq 0, \pi$). The energy of these domain structures are obtained to be always higher than for Bloch-type domain walls. We emphasize that we have considered only a limited number of magnetic phases. The consideration of additional structures may lead to further changes in the magnetic phase diagram.

The calculated magnetic phase diagram of Fig. 1 should also be valid at finite temperatures, if the free energy is given by Eq. (1) with temperature-dependent coupling coefficients. Then a variation of the temperature refers to a certain path in the parameter space of the phase diagram presented in Fig. 1, determined by the actual temperature dependence of the coupling coefficients $E_0(T), K_2(T), \ldots$. In particular, this will lead to a temperature dependent periodicity $L(T)$ and wall width $b(T)$ of the domain structures, and may also cause a temperature-induced magnetic reorientation. The temperature dependence of the second-order lattice anisotropy and the dipole interaction are expected to differ slightly, thus their difference should be a slowly varying function of temperature.[11] Therefore, the range of vanishing magnetization ΔT due to the appearance of domain phases may be considerably large, especially much larger than in the case where the loss of the global magnetization results from a vanishing remanent magnetization due to mutual cancellation of anisotropies, as calculated in our previous study.[12] The quantity ΔT will depend sensitively on the temperature dependence of the coupling coefficients. These functions $E_0(T), K_2(T), \ldots$ were calculated for bulk systems,[29] whereas for thin films the temperature dependence has been calculated only approximately.[11] Thus, we have not attempted to calculate ΔT around the reorientation temperature, since a precise theory for the temperature dependence of the coupling coefficients is presently not known to us, and since this is not the main goal of this paper.

Finally, we like to discuss possible implications of the domain structures on the magnetic phase transitions in thin films at finite temperatures. It is known that at a second-order phase transition the dispersion relation $\varepsilon(\vec{k})$ vanishes at $\vec{k}=0$, $\vec{k}$ the 2D wave vector. Then critical fluctuations are expected to occur, and the magnetization vanishes. Second-order phase transitions exist between in-plane and canted orientations and between canted and perpendicular magnetization, if only uniform phases are considered. We expect that the existence of additional phases may alter the nature of the phase transitions. For instance, within the domain phases the in-plane component of the magnetization m_x does not vanish suddenly with increasing K_2, but maintains a small value for $K_2 > E_0 - 2K_4$, see Fig. 3. Therefore, critical phenomena at the phase transition line $K_2 \lesssim E_0 - 2K_4$, $K_4 < -K_s$, between the canted and the perpendicular phases might vanish. On the other hand, the perpendicular component m_z vanishes at $K_2 = E_0 + 2K_s$, $K_4 < -K_s$. Thus, the nature of this phase transition between canted and in-plane magnetization is expected not to be affected by domain phases, and critical phenomena occur as in the case of uniform magnetic phases. The nature of the first-order phase transition line for $K_4 > -K_s$ between perpendicular and in-plane magnetization seems also to be preserved. In the case of such a discontinuous phase transition critical phenomena are often masked by hysteresis effects. In addition, at finite temperatures the appearance of the domain structure may change. For example, straight domain walls may be dislocated, or additional domain walls may be created.[18] Also a canted magnetization may occur for a vanishing fourth-order anisotropy.[20]

In summary, domain structures will influence the magnetic phase diagram of thin films at $T=0$ markedly. In accordance with previous work our results suggest that the vanishing global magnetization observed in Refs. 4–7 are caused by a magnetic domain formation. Such a domain phase is most likely to occur near the magnetic reorientation transition, since its binding energy ΔE is largest in the reorientation region or for a canted magnetization, see Fig. 4. At finite temperatures the determination of the reorientation temperature and the temperature range of vanishing magnetization requires the precise knowledge of the temperature-dependent coupling coefficients. The consideration of higher-order lattice anisotropies may induce a continuous transition from perpendicular to in-plane magnetization, mediated by a canted domain phase. Such a canted domain phase can explain the simultaneous occurrence of stripe domain structures of both the perpendicular and the in-plane magnetization, as observed experimentally.[5] Presently, we calculate the domain structure of thin films at finite temperatures.

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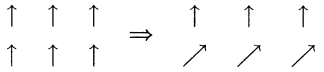
APPENDIX

In this appendix we investigate the effect of fluctuations on the long-range dipole interaction energy of a (100) monolayer at $T=0$ on the basis of lattice summations. First we consider an uniform (ferromagnetic) in-plane magnetization directed along the x axis. We distinguish between two different kinds of fluctuations. A sinusoidal distortion of the azimuthal angle $\phi(x_i)=\phi_0 \sin(kx_i)$ in the xy plane at lattice site i is considered as depicted:



ϕ_0 is the amplitude, k the wave vector along the x direction. After averaging over the distortion period $2\pi/k$ the dipole energy per spin is calculated to be

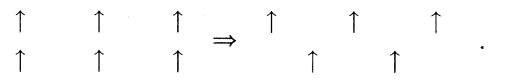
$$E_{\rm dip}^{(1)}(k)=-\frac{E_0}{6\mathscr{S}(0,0)}\sum_{\substack{u,v=-\infty\\ u\neq v}}^{\infty}\frac{1}{r^5}\left[(u^2+v^2)J_0\left(2\phi_0\sin\frac{ku}{2}\right)+3(u^2-v^2)J_0\left(2\phi_0\cos\frac{ku}{2}\right)\right]. \tag{A1}$$

$J_0(x)$ is the Bessel function of order 0, E_0 the demagnetizing energy, and $r^2=u^2+v^2$. The quantities $\mathscr{S}(p,q)$ are defined in Eq. (2). For small amplitudes ϕ_0 the difference of dipole energy between the distorted phase and the uniform magnetization is given by

$$\delta E_{\rm dip}^{(1)}(k)=\frac{E_0}{6\mathscr{S}(0,0)}\phi_0^2(\mathscr{S}(0,0)+\mathscr{S}(k,0)-2\mathscr{S}(0,k))+\mathscr{O}(\phi_0^4), \tag{A2}$$

where we have used $J_0(x)\simeq 1-x^2/4$ for small x. Our calculations show that $\delta E_{\rm dip}^{(1)}(k)$ is negative, i.e., the dipole interaction favors a sinusoidal distortion of the azimuthal angle for an in-plane magnetization.

A spatial transversal dislocation of the lattice sites perpendicular to the x axis has to be studied separately:



Such a sinusoidal dislocation is introduced by $\vec{r}_i=[x_i, y_i+y_0\cos(kx_i)]$. After averaging over the distortion period $2\pi/k$, the dipole energy difference per spin caused by this fluctuation for small amplitudes y_0 is given by

$$\delta E_{\rm dip}^{(2)}(k)=\frac{E_0}{6\mathscr{S}(0,0)}y_0^2\sum_{\substack{u,v=-\infty\\ u\neq v}}^{\infty}\left(\frac{12}{r^5}-\frac{105u^2v^2}{r^9}\right)\times[1-\cos(ku)]+\mathscr{O}(y_0^4). \tag{A3}$$

These slowly converging direct lattice sums may be converted to rapidly converging ones similar as applied to Eq. (2). Our calculations show that $\delta E_{\rm dip}^{(2)}(k)$ is positive, thus this kind of distortion of the uniform magnetic phase is unfavorable. However, this increase of dipole energy may be balanced by an appropriate variation of the azimuthal angle, see Eq. (A2). Note that in a magnetic monolayer these fluctuations cause an unfavorable energy increase of the in-plane lattice anisotropy and/or the much stronger exchange coupling. These energy contributions will overcome a gain of dipole energy, thus maintaining the uniform magnetization. In contrast, for an electric dipole monolayer or a 2D ferroelectric system a distorted phase may be stable, as shown in Ref. 16 for the edges of a Langmuir layer, since in such systems a strong nearest-neighbor exchange coupling is not present.

Secondly, we calculate the stability of the stripe domain structure with an array of straight domain walls. We apply the same domain-wall profile with periodicity $2L$ and wall width b as described in Sec. II, assuming $\theta_d=0$. A sinusoidal transversal dislocation is assumed, corresponding to a shift $\bar{y}(x)$ of the domain-wall center with wave vector k along the x direction: $\cos(\pi y/b)\rightarrow\cos\{\pi[y-\bar{y}(x)]/b\}=\cos[\pi y/b-\bar{\theta}(x)]$, with $\bar{\theta}(x)=\theta_0\cos(kx)=\pi y_0\cos(kx)/b$, assuming $y_0\ll b<L$. For such a fluctuation the values of L and b remain unchanged. After averaging over the distortion period $2\pi/k$ along the x axis, and using the identity $\int_0^{2\pi}dx\,\cos[a\cos(x)+b\sin(x)]=2\pi J_0(\sqrt{a^2+b^2})$, the energy per spin of the dipole interaction due to this periodic distortion of the domain walls is given by

$$E_{\rm dip}(k)=\frac{E_0}{12\mathscr{S}(0,0)}\sum_{\substack{n=-\infty\\ n\ \text{odd}}}^{\infty}\sum_{\substack{u,v=-\infty\\ u\neq v}}^{\infty}\frac{c_n^2}{r^5}\left[\left(v^2-\frac{u^2}{2}\right)(1+\alpha_n^2)J_0\left(2\theta_0\sin\frac{ku}{2}\right)+\frac{3}{2}u^2(1-\alpha_n^2)J_0\left(2\theta_0\cos\frac{ku}{2}\right)\right]e^{i\pi nv/L}, \tag{A4}$$

with $\alpha_n=nb/L$. The Fourier coefficients c_n are defined in Eq. (5). The dipole energy difference for small amplitudes θ_0 is calculated to be

$$\delta E_{\rm dip}(k)=\frac{E_0}{24\mathscr{S}(0,0)}\theta_0^2\sum_{n=1,3,\ldots}^{\infty}c_n^2\left\{(1+\alpha_n^2)\left[\mathscr{S}\left(\frac{\pi n}{L},k\right)-\mathscr{S}\left(\frac{\pi n}{L},0\right)\right]-(2-\alpha_n^2)\left[\mathscr{S}\left(k,\frac{\pi n}{L}\right)-\mathscr{S}\left(0,\frac{\pi n}{L}\right)\right]\right\}+\mathscr{O}(\theta_0^4). \tag{A5}$$

Our calculations show that $\delta E_{\rm dip}(k)>0$ for $k>0$, thus a periodic array of straight Bloch domain walls with $k=0$ will be stable in accordance with Refs. 16,18. Since we have performed a discrete lattice summation, we are not able to derive the dependence $\delta E_{\rm dip}(k)\propto k^2|\ln k|$ for small wave vectors k as obtained by a continuum approximation.[16,19] Different domain profiles may also be considered, resulting in different quantities c_n and α_n.

In the previous calculations we have considered only a shift $\bar{y}(x)$ of the domain-wall center, whereas the spins are still confined to the xz plane. In addition, we have also considered an additional variation of the azimuthal angle ϕ_i of the spins, i.e., inside the domain walls the magnetization exhibits a y component. In the case of an uniform magnetic phase such a fluctuation was obtained to exhibit a favorable change of dipole energy, see Eq. (A2). To accomplish an alignment of the spins parallel to the domain-wall edges we put $\phi_0=-k\theta_0$. Our calculations show that this additional azimuthal distortion does not lower considerably the increase in dipole energy as given by Eq. (A5).

Finally, we calculate the difference of exchange energy due to a sinusoidal dislocation of a periodic array of straight domain walls. After averaging over the period $2L$ along the y direction of the domain structure and over the distortion period $2\pi/k$ along the x direction, the exchange energy per spin is given approximately by

$$E_{\rm ex}(k)\approx-\frac{J}{2}\left(q-\frac{\pi^2}{bL}-\frac{b}{2L}(\theta_0 k)^2\right), \tag{A6}$$

with $q=4$ the coordination number. We have used $\cos(\theta_{i+1}-\theta_i)\simeq 1-(\partial\theta/\partial x)^2/2$, etc. The difference of the exchange coupling caused by the sinusoidal domain-wall dislocation for small amplitudes θ_0 and/or small wave vectors k is calculated to be

$$\delta E_{\rm ex}(k)\approx\frac{Jb}{4L}(\theta_0 k)^2=\frac{J}{4}\frac{\pi^2}{bL}(y_0 k)^2>0. \tag{A7}$$

$\delta E_{\rm ex}(k)$ scales with b/L, since a distorted spin structure is present only in the domain walls, whereas the domain interior remains uniformly ordered. The lattice anisotropy energy is not affected by this kind of domain-wall distortion.

In summary, we have shown that the dipole interaction as well as the exchange coupling prefer straight walls for a periodic domain structure. Also an impurity pinning will support straight domain walls.[19] Quite interestingly, an uniform in-plane phase should be unstable against azimuthal fluctuations already at $T=0$ for a vanishing exchange coupling, as is the case of an electric dipole layer.

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