Anisotropy of ultrathin ferromagnetic films and the spin reorientation transition

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The influence of uniaxial anisotropy and the dipole interaction on the direction of the magnetization of ultrathin ferromagnetic films in the ground state is studied. The ground-state energy can be expressed in terms of anisotropy constants which are calculated in detail as a function of the system parameters and the film thickness. In particular noncollinear spin arrangements are taken into account. Conditions for the appearance of a spin reorientation transition are given and analytic results for the width of the canted phase and its shift in applied magnetic fields associated with this transition are derived.

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I. INTRODUCTION

Experimentally it became possible in recent years to grow epitaxial thin films of ferromagnetic materials on nonmagnetic substrates with a very high quality. This offers the possibility to stabilize crystallographic structures which are not present in nature, and which may exhibit new properties of high technological impact. To understand the magnetic structure of these systems is a challenging problem both experimentally and theoretically.

Generally speaking, for not too thin films the magnetization is in-plane due to the dipole interaction (shape anisotropy). However, in very thin films this may change due to the increasing importance of surface effects. Indeed, at surfaces due to the broken symmetry uniaxial anisotropy energies arise which in generally are much higher than in the bulk. These anisotropy energies may favor a perpendicular orientation of magnetization.[1] Additionally in the inner layers of the film due to strain-induced distortion bulk anisotropy energies may appear absent or very small in the ideal crystal. As a consequence in these films a reorientation of the spontaneous magnetization is observed either as a function of film thickness or as a function of temperature. This spin reorientation transition has been discussed extensively in the past.[2–5]

Phenomenologically in order to describe the magnetic properties, anisotropy coefficients K_n compatible with the underlying symmetry of the film are introduced which are supposed to arise from an expansion of the energy (or the free energy at finite temperatures) in terms of the orientation of the magnetization vector relative to the film. These coefficients are then studied experimentally (for a review see Ref. 6). In ferromagnetic resonance (FMR) experiments, for instance, these coefficients directly enter the resonance frequency (for references see for instance Refs. 7 and 8).

Theoretically, it has been shown that the anisotropy coefficients $K_n(T)$, which are in general temperature dependent, can be calculated numerically at finite temperatures within mean field theory, starting from a Hamiltonian with microscopic constant anisotropy parameters.[9] Furthermore, the temperature dependence of the lowest order anisotropy $K(T)$ was determined analytically using a combination of mean field theory and first-order perturbation theory.[9,10] In other approaches the magnetization of the film is calculated directly within mean field and spin wave theory[11–16] or with full numerical calculations like Monte Carlo simulations.[17,18]

In the present paper we describe the ferromagnetic film within a classical local spin model with dipolar interaction and uniaxial anisotropy. We will concentrate on ground-state properties of thin films in order to clarify the discussion and to eliminate all uncertainties connected with finite temperature calculations. A major goal of the present study is the calculation of the anisotropy coefficients at zero temperature from the parameters of an underlying Hamiltonian. The important point is that even in this situation the dependence of these coefficients on the microscopic parameters is far from being trivial due to non-collinear magnetic states in the thin film. It is the purpose of this paper to elucidate this behavior.

II. THE MODEL

The calculations of the ground-state properties of ultrathin ferromagnetic films are done within the framework of a classical ferromagnetic Heisenberg model consisting of L two-dimensional layers with the $\vec{z}$ direction normal to the film. The Hamiltonian reads

$$\mathcal{H}=-\frac{J}{2}\sum_{\langle ij\rangle}\vec{s}_i\cdot\vec{s}_j+\frac{\omega}{2}\sum_{i\neq j}\frac{\vec{s}_i\cdot\vec{s}_j}{r_{ij}^3}-\frac{3(\vec{s}_i\cdot\vec{r}_{ij})(\vec{r}_{ij}\cdot\vec{s}_j)}{r_{ij}^5}$$
$$-\sum_i D^{(2)}_{\lambda_i}(s_i^z)^2-\sum_i D^{(4)}_{\lambda_i}(s_i^z)^4-\sum_i \vec{B}\cdot\vec{s}_i, \qquad (1)$$

where $\vec{s}_i=(s_i^x,s_i^y,s_i^z)$ are spin vectors of unit length at position $\vec{r}_i=(r_i^x,r_i^y,r_i^z)$ in layer λ_i and $\vec{r}_{ij}=\vec{r}_i-\vec{r}_j$. The positions $\vec{r}_i$ are normalized such that nearest neighbors obey $r_{\langle ij\rangle}=1$. J is the nearest-neighbor exchange coupling constant, $D^{(2)}_{\lambda_i}$ and $D^{(4)}_{\lambda_i}$ are the local uniaxial anisotropies of second and fourth order, respectively, $\vec{B}$ denotes the external magnetic field with the effective magnetic moment μ of the spins incorporated, and $\omega=\mu_0\mu^2/4\pi a^3$ is the strength of the long range dipole interaction on a lattice with lattice constant a (μ_0 is the magnetic permeability).

To calculate the ground-state energy per spin we assume translational invariance of the spin structure parallel to the film. This assumption is not correct rigorously since it can be

TABLE I. Number of nearest neighbors z_δ and dipole sums Φ_δ for different lattice types. δ is the distance between layers.

Lattice	z_0	z_1	$z_{\delta>1}$	Φ_0	Φ_1	$\Phi_{\delta>1}$
sc(001)	4	1	0	9.0336	-0.3275	$\sim -16\pi^2 e^{-2\pi\delta}$
fcc(001)	4	4	0	9.0336	1.4294	$\sim \mp 16\pi^2 e^{-\sqrt{2}\pi\delta}$
bcc(001)	0	4	0	5.8675	2.7126	$\sim \mp 6\sqrt{3}\pi^2 e^{-\pi\delta}$

shown that for a perpendicular oriented magnetization, for instance, a state with striped domains is energetically slightly more favorable. However the corresponding energy difference for ultrathin films is of order $e^{-J/2\omega}$ and therefore negligible for realistic parameters of Fe or Ni films showing spin reorientation transitions.[18]

Assuming translational invariance in the xy plane the summation over all spins within a plane can be done exactly resulting in the energy per surface spin

$$E(\vec{s}) = -\frac{J}{2}\sum_{\mu,\nu=1}^{L} z_{|\mu-\nu|}\vec{s}_\mu\cdot\vec{s}_\nu - \frac{\omega}{2}\sum_{\mu,\nu=1}^{L}\Phi_{|\mu-\nu|}\vec{s}_\mu\cdot\begin{pmatrix}\frac{1}{2} & 0 & 0\\ 0 & \frac{1}{2} & 0\\ 0 & 0 & -1\end{pmatrix}\cdot\vec{s}_\nu - \sum_{\mu=1}^{L}\left[D_\mu^{(2)}(s_\mu^z)^2 + D_\mu^{(4)}(s_\mu^z)^4 + \vec{B}\cdot\vec{s}_\mu\right] \tag{2}$$

with $\vec{s}=(\vec{s}_1,\ldots,\vec{s}_L)$. The quantities μ and ν denote layer indices, $z_{|\mu-\nu|}$ is the number of nearest neighbors between layer μ and ν, and $\Phi_{|\mu-\nu|}$ are constants arising from a partial summation of the dipole interaction. The quantities Φ_δ have been calculated previously[19,20] and they are listed together with z_δ in Table I.

With an external magnetic field $\vec{B}=(0,B_\parallel,B_\perp)$ in the yz plane, all spins $\vec{s}_\mu$ are confined to this plane. They therefore can be expressed by their azimuthal angle ϑ_μ, $\vec{s}_\mu=(0,\sin\vartheta_\mu,\cos\vartheta_\mu)$. Equation (2) thus can be rewritten as

$$E(\vec{\vartheta}) = -\frac{1}{2}\sum_{\mu,\nu=1}^{L}\left[\left(Jz_{|\mu-\nu|}-\frac{\omega}{4}\Phi_{|\mu-\nu|}\right)\cos(\vartheta_\mu-\vartheta_\nu) - \frac{3\omega}{4}\Phi_{|\mu-\nu|}\cos(\vartheta_\mu+\vartheta_\nu)\right] - \sum_{\mu=1}^{L}\left[D_\mu^{(2)}\cos^2\vartheta_\mu + D_\mu^{(4)}\cos^4\vartheta_\mu + B_\parallel\sin\vartheta_\mu + B_\perp\cos\vartheta_\mu\right] \tag{3}$$

with $\vec{\vartheta}=(\vartheta_1,\ldots,\vartheta_L)$. The ground state is obtained by minimizing the energy $E(\vec{\vartheta})$ with respect to $\vec{\vartheta}$. In zero external field two stationary points of the energy given in Eq. (3) are easily identified to be given by $\vec{\vartheta}^\perp=(0,\ldots,0)$ and $\vec{\vartheta}^\parallel=(\pi/2,\ldots,\pi/2)$, respectively. We define a total anisotropy per surface spin in zero field K by the corresponding energy difference, $K=E(\vec{\vartheta}^\parallel)-E(\vec{\vartheta}^\perp)$. This quantity is given by

$$K=\sum_{\mu=1}^{L}(D_\mu^{(2)}+D_\mu^{(4)}) - \frac{3\omega}{4}\sum_{\mu,\nu=1}^{L}\Phi_{|\mu-\nu|}. \tag{4}$$

The first term is the sum of the anisotropy constants of second and fourth order while the second term is due to the dipole interaction. Note that this dipole term is identical to the dipole anisotropy per unit area $(L/2)\mu_0 m^2$ calculated within continuum theory, but with additional surface correction, as

$$\frac{3\omega}{4}\sum_{\mu,\nu=1}^{L}\Phi_{|\mu-\nu|} = \frac{L}{2}\mu_0 m^2 - \frac{3\omega}{2}\Phi_1 + \mathcal{O}(\Phi_2). \tag{5}$$

For $K>0$ a perpendicular magnetization is more favorable than an in-plane magnetization and vice versa. However, in certain parameter intervals additional stationary points appear which may lead to an even lower energy resulting in a canted spin structure. This will be discussed in detail in Sec. V.

In general the minimization of Eq. (3) has to be done numerically. For realistic parameters appearing for instance for Fe or Ni films, however, the exchange interaction is by far the largest term in the Hamiltonian leading to a nearly collinear spin structure. In this situation the anisotropy terms can be treated as small perturbation and as a consequence the minimization can be done to a large extent analytically.

III. PERTURBATION CALCULATION

We define an averaged angle, $\theta=1/L\,\Sigma_{\nu=1}^{L}\ \vartheta_\nu$ and deviations from it, ϵ_ν, so that $\vartheta_\nu=\theta+\epsilon_\nu$ and $\Sigma_{\nu=1}^{L}\ \epsilon_\nu=0$. Finite ϵ_ν appear due to the various anisotropy terms and they are therefore small for anisotropy terms (including the external magnetic field) which are small compared to the exchange energy. This will be assumed in the following. Under these circumstances a perturbative treatment is possible. We decompose the energy Eq. (3) into two parts,

$$E(\vec{\vartheta}) = E^{(0)}(\theta) + \delta E(\theta,\vec{\epsilon}), \tag{6}$$

with $\delta E(\theta,\vec{0})=0$ and

$$E^{(0)}(\theta) = -\frac{J}{2}\sum_{\mu,\nu=1}^{L} z_{|\mu-\nu|} + \frac{3\omega}{8}\cos(2\theta)\sum_{\mu,\nu=1}^{L}\Phi_{|\mu-\nu|} - \cos^2\theta\sum_{\mu=1}^{L}D_\mu^{(2)} - \cos^4\theta\sum_{\mu=1}^{L}D_\mu^{(4)} - L(B_\parallel\sin\theta + B_\perp\cos\theta). \tag{7}$$

An expansion of $\delta E(\theta,\vec{\epsilon})$ in terms of $\vec{\epsilon}$ then gives

$$\delta E(\theta,\vec{\epsilon}) = \vec{a}(\theta)\cdot\vec{\epsilon} + \frac{1}{2}\vec{\epsilon}\cdot\mathbf{C}\cdot\vec{\epsilon} + \mathcal{O}(\vec{\epsilon})^3, \tag{8}$$

where we have introduced an obvious matrix notation. The gradient

$$\vec{a}(\theta)=\frac{\partial}{\partial\vec{\epsilon}}\delta E(\theta,\vec{\epsilon})\big|_{\vec{\epsilon}=\vec{0}} \tag{9}$$

is given by

$$\vec{a}(\theta)=\vec{A}(\theta)\sin(2\theta) \tag{10}$$

with

$$A_\lambda(\theta)=D_\lambda^{(2)}+2D_\lambda^{(4)}\cos^2\theta-\frac{3\omega}{4}\sum_{\mu=1}^{L}\Phi_{|\lambda-\mu|}. \tag{11}$$

Thus, to lowest order the anisotropy terms are linear in $\vec{\epsilon}$ while the exchange term expressed in Eq. (8) by the matrix **C** with matrix elements

$$C_{\mu\nu}=-Jz_{|\mu-\nu|}+\delta_{\mu\nu}\sum_{\lambda=1}^{L}Jz_{|\mu-\lambda|} \tag{12}$$

is quadratic in $\vec{\epsilon}$.

The minimum of $\delta E(\theta,\vec{\epsilon})$ appears for ϵ_ν of the order of the anisotropy terms showing that the truncated Eq. (8) gives the correct energy up to second order in $\vec{\epsilon}$. Note that up to this order the Zeeman term enters only Eq. (7). Therefore, at this level of truncation θ agrees with the azimuthal angle of the averaged magnetization.

It can be easily seen from the definition Eq. (12) that $\vec{e}_0=(1,\ldots,1)$ is an eigenvector of **C** with eigenvalue zero. With this vector it is convenient to rewrite the constraint $\Sigma_{\nu=1}^{L}\epsilon_\nu=0$ as a scalar product, $\vec{e}_0\cdot\vec{\epsilon}=0$. This notation will be used in the following.

The minimalization of the energy is done in two steps. First we keep θ fixed and minimize with respect to ϵ_ν under the constraint $\vec{e}_0\cdot\vec{\epsilon}=0$. The corresponding energy at the minimum, $E(\theta)$, is accessible for instance by varying the external magnetic field and it is precisely this quantity which for instance is needed to calculate the FMR signal. Finally the ground state energy is obtained by minimizing $E(\theta)$ with respect to θ.

The variation with respect to ϵ_ν is achieved by introducing the function

$$\Psi(\theta,\vec{\epsilon})=\vec{a}(\theta)\cdot\vec{\epsilon}+\frac{1}{2}\vec{\epsilon}\cdot\mathbf{C}\cdot\vec{\epsilon}+\lambda\vec{e}_0\cdot\vec{\epsilon}, \tag{13}$$

where λ denotes a Lagrangian multiplier. Stationarity of $\Psi(\theta,\vec{\epsilon})$ gives

$$\mathbf{C}\cdot\vec{\epsilon}+\vec{a}(\theta)+\lambda\vec{e}_0=\vec{0}. \tag{14}$$

Taking the scalar product with $\vec{e}_0$ and noting that $\vec{e}_0\cdot\mathbf{C}=\vec{0}$ the multiplier λ is obtained as

$$\lambda=-\frac{1}{L}\vec{e}_0\cdot\vec{a}(\theta). \tag{15}$$

Thus $\vec{\epsilon}$ is determined from

$$\mathbf{C}\cdot\vec{\epsilon}+\left(\mathbf{1}-\frac{1}{L}\mathbf{E}\right)\cdot\vec{a}(\theta)=\vec{0} \tag{16}$$

with identity matrix **1** and a matrix **E** with $E_{\mu\nu}=1$ for all matrix elements. To solve this equation for $\vec{\epsilon}$ we introduce the pseudoinverse $\mathbf{C}^\dagger$ of the matrix **C**, which in our case fulfills

$$\mathbf{C}\cdot\mathbf{C}^\dagger=\mathbf{1}-\frac{1}{L}\mathbf{E}. \tag{17}$$

The matrix $\mathbf{C}^\dagger$ is uniquely defined if one requires that it is a symmetric matrix with eigenvector $\vec{e}_0$ and corresponding eigenvalue zero. The matrix elements of $\mathbf{C}^\dagger$ are explicitly given by[20]

$$C^\dagger_{\mu\nu}=\frac{1}{2LJz_1}\left[\frac{L^2-1}{6}-L|\mu-\nu|+\left(\mu-\frac{L+1}{2}\right)^2+\left(\nu-\frac{L+1}{2}\right)^2\right]. \tag{18}$$

It is easy to see that with the help of this matrix Eq. (16) can be rewritten as

$$\mathbf{C}\cdot(\vec{\epsilon}+\mathbf{C}^\dagger\cdot\vec{a}(\theta))=\vec{0}. \tag{19}$$

Since $\vec{e}_0$ is the only eigenvector of **C** with eigenvalue zero the term in brackets has to be parallel to $\vec{e}_0$. Multiplying this term by $\vec{e}_0$ and using $\vec{e}_0\cdot\vec{\epsilon}=0$ and $\vec{e}_0\cdot\mathbf{C}^\dagger=\vec{0}$ it follows

$$\vec{\epsilon}=-\mathbf{C}^\dagger\cdot\vec{a}(\theta). \tag{20}$$

Inserting into Eq. (13) we get the final result

$$E(\theta)=E^{(0)}(\theta)+\delta E(\theta), \tag{21a}$$

$$\delta E(\theta)=-\frac{1}{2}\vec{a}(\theta)\cdot\mathbf{C}^\dagger\cdot\vec{a}(\theta)+\mathcal{O}(\vec{\epsilon})^3, \tag{21b}$$

where we used the general property $\mathbf{C}^\dagger=\mathbf{C}^\dagger\cdot\mathbf{C}\cdot\mathbf{C}^\dagger$ of the pseudoinverse. The ground state energy is obtained by minimizing $E(\theta)$ with respect to θ.

Equation (21) is the main result of this work, giving a general expression for the ground state energy of a thin magnetic film in second-order perturbation theory. The influence of a noncollinear spin structure on the ground state energy will be discussed in the following.

IV. RESULTS

In the following we drop terms of order $\mathcal{O}(\vec{\epsilon})^3$ in $E(\theta)$ and we specialize to a special case in order to obtain analytic results. We neglect the exponentially small effective dipole interactions between layers with distance larger than one, i.e., $\Phi_{\delta>1}=0$, and we assume that the anisotropies $D_\lambda^{(n)}$ which enter the Hamiltonian Eq. (1) are constant within the thin film but may deviate from its constant value at the surface ($\lambda=1$) and at the interface to the substrate ($\lambda=L$), i.e.,

$$D_\lambda^{(n)} = D_v^{(n)} + \delta_{\lambda,1} D_s^{(n)} + \delta_{\lambda,L} D_i^{(n)}, \tag{22}$$

$$A_\lambda(\theta) = A_v(\theta) + \delta_{\lambda,1} A_s(\theta) + \delta_{\lambda,L} A_i(\theta) \tag{23}$$

with

$$A_{s,i}(\theta) = D_{s,i}^{(2)} + 2D_{s,i}^{(4)} \cos^2\theta + \frac{3\omega}{4}\Phi_1. \tag{24}$$

It is easy to see that

$$\vec{A}(\theta)\cdot\mathbf{C}^\dagger\cdot\vec{A}(\theta) = C_{1,1}^\dagger (A_s^2(\theta) + A_i^2(\theta)) + 2C_{1,L}^\dagger A_s(\theta) A_i(\theta) \tag{25}$$

since $\mathbf{C}^\dagger \cdot \vec{e}_0 = \vec{0}$ and $C_{1,1}^\dagger = C_{L,L}^\dagger$. Then the second-order correction calculated in the previous section [Eq. (21b)] can be written as

$$\delta E(\theta) = \Delta(\theta, L)\sin^2(2\theta) \tag{26}$$

with

$$\Delta(\theta,L) = -\frac{L-1}{8Jz_1}\left[\frac{L-2}{3L}(A_s(\theta) + A_i(\theta))^2 + (A_s(\theta) - A_i(\theta))^2\right]. \tag{27}$$

Note that from now on L can be considered as a continuous parameter and all quantities are explicitly L-dependent. Inserting $\vec{a}(\theta)$ and $E^{(0)}(\theta)$ into Eq. (21) and introducing the quantities

$$K_0(L) = \frac{J}{2}(zL - 2z_1) + \frac{\omega}{2}\left(2\pi L - \frac{3}{2}\Phi_1\right), \tag{28a}$$

$$K_2(L) = LD_v^{(2)} + D_s^{(2)} + D_i^{(2)} - \omega\left(2\pi L - \frac{3}{2}\Phi_1\right), \tag{28b}$$

$$K_4(L) = LD_v^{(4)} + D_s^{(4)} + D_i^{(4)}, \tag{28c}$$

we can finally write for the energy per surface spin

$$E(\theta,L) = -K_0(L) + \Delta(\theta,L)\sin^2(2\theta) - K_2(L)\cos^2\theta - K_4(L)\cos^4\theta - L(B_\parallel \sin\theta + B_\perp \cos\theta). \tag{29}$$

Note that the total anisotropy energy K introduced in Eq. (4) fulfills

$$K(L) = K_2(L) + K_4(L), \tag{30}$$

as $\delta E(\theta)$ vanishes at the collinear stationary points $\vec{\vartheta}^\parallel$ and $\vec{\vartheta}^\perp$, respectively. $K_2(L)$ and $K_4(L)$ contain the microscopic anisotropy parameters and the dipole terms of the film averaged over the different layers.

It is easy to see that an equation for $E(\theta,L)$ in the form given by Eq. (29) often introduced phenomenologically,[8] but without the Δ term, is obtained if one assumes that all spins in the film are strictly parallel. The important point to note here, however, is the fact that an additional anisotropy energy $\Delta(\theta,L)$ enters Eq. (29) which is connected to noncollinear spin structures originated by inhomogeneities in the magnetic film. Indeed, this quantity only vanishes in the homogeneous case $A_s = A_i = 0$. However, for a magnetic thin film the amplitudes A_λ in general are not constant. Even if the microscopic anisotropy constants $D_\lambda^{(n)}$ are homogeneous (which is unlikely to occur for a realistic film) this is not the case for the dipole term.

To discuss the implications of this additional anisotropy term $\Delta(\theta,L)$ we first consider the case that there is no microscopic uniaxial anisotropy of fourth order, $D_\lambda^{(4)} = 0$. In this case $\Delta(\theta,L) = \Delta(L)$ is independent of θ. Thus for an inhomogeneous distribution of amplitudes A_λ, an effective anisotropy term of fourth order in $\cos\theta$ is generated although there is no corresponding anisotropy term of this order in the Hamiltonian.

If there exists a microscopic anisotropy term of fourth order the situation is more complicated: Δ becomes θ-dependent meaning that higher order anisotropy term of up to eighth order are generated in $E(\theta,L)$.

Finally we mention that the quantity $\Delta(\theta,L)$ can be further simplified in two common special cases: In the case of a symmetric film $D_i^{(n)} = D_s^{(n)}$ we get $A_s(\theta) = A_i(\theta)$ and therefore

$$\Delta_{i=s}(\theta,L) = -\frac{(L-1)(L-2)}{6LJz_1}A_s^2(\theta), \tag{31a}$$

while for the case $D_i^{(n)} = 0$ and $D_s^{(2)} + D_s^{(4)} \gg (3\omega/2)\Phi_1$ we have $A_s(\theta) \gg A_i(\theta)$ and

$$\Delta_{i=0}(\theta,L) = -\frac{(L-1)\left(L-\frac{1}{2}\right)}{6LJz_1}A_s^2(\theta). \tag{31b}$$

As an important application of these results we will study spin reorientation transitions in the next section.

V. SPIN REORIENTATION TRANSITION

The direction of the magnetization in the ground state for a given thickness L is obtained by minimizing $E(\theta,L)$ [Eq. (29)]. If the total anisotropy energy $K(L)$ [Eq. (32)] changes sign as function of L, a spin reorientation transition takes place in which the direction of the magnetization changes either continuously or discontinuously depending on the specific form of $E(\theta,L)$. In the first case a so-called canted phase appears. Analytic results for the width and the position of this phase will be derived in this chapter.

We decompose $K(L)$ in volume and surface part the usual way[21] to get

$$K(L) = LK_v + K_s + K_i \tag{32}$$

with

$$K_v = D_v^{(2)} + D_v^{(4)} - 2\pi\omega, \tag{33a}$$

$$K_{s,i} = D_{s,i}^{(2)} + D_{s,i}^{(4)} + \frac{3\omega}{4}\Phi_1. \tag{33b}$$

Note that $A_{s,i}(\theta)$ from Eq. (24) can be written as

$$A_{s,i}(\theta)=K_{s,i}+D_{s,i}^{(4)}\cos(2\theta). \tag{34}$$

A spin reorientation transition occurs if the total anisotropy energy $K(L)$ passes through zero as function of L. If $K_s+K_i>0$ this happens for sufficiently large dipole interaction with increasing L, as then $K_v<0$. The corresponding transition is from perpendicular magnetization at small L to an in-plane magnetization for large L possibly with a canted magnetization in between. This type of transition occurs for Fe films. The opposite scenario can occur for negative K_s+K_i if a positive volume anisotropy $K_v>0$ is present as observed in Ni films. Thus, to lowest order the critical thickness is explicitly given by $K(L_r)=0$, leading to

$$L_r=-\frac{K_s+K_i}{K_v}. \tag{35}$$

For Fe/Ag(100) films $D_s^{(2)}+D_i^{(2)}\approx 37\omega$. In this case the other quantities $D_v^{(4)}$, $D_v^{(2)}$, and Φ_1 are negligible and we get $L_r\approx 5.8$ in good agreement with numerical calculations.[20]

For L in the vicinity of L_r the minimum of $E(\theta)$ may occur at a finite θ, i.e., a canted phase occurs. To deduce the limits of stability of the two phases for which $\theta=0$ and $\theta=\pi/2$, respectively, we expand Eq. (29) around these angles. From the sign of the corresponding expansion coefficient it follows that in general there are two transitions of second order at thicknesses $L_r^{\parallel}$ and $L_r^{\perp}$, respectively. The phase with $\theta=0$ becomes unstable at $L_r^{\parallel}$ where

$$K(L_r^{\parallel})+K_4(L_r^{\parallel})+4\Delta(0,L_r^{\parallel})=0 \tag{36a}$$

at this point. With increasing thickness the parallel phase with $\theta=\pi/2$ becomes stable at $L_r^{\perp}$ where

$$K(L_r^{\perp})-K_4(L_r^{\perp})-4\Delta\left(\frac{\pi}{2},L_r^{\perp}\right)=0. \tag{36b}$$

For $K_4(L_r)+4\Delta(L_r)=0$ both transitions coincide resulting in a jump from $\theta=0$ to $\theta=\pi/2$ at L_r. This is always the case for $L=1$ and in the symmetric case also for $L=2$ provided $D_\lambda^{(4)}$ vanishes. Otherwise a canted phase ($K_4+4\Delta>0$) or a region with hysteresis ($K_4+4\Delta<0$) appears as described in the following. Note that in the phases $\theta=0$ and $\theta=\pi/2$, respectively, $\vec{\epsilon}$ vanishes according to Eqs. (10) and (20) showing that in these phases all spins are strictly parallel. This is not the case in the canted phase. Note also that for finite magnetic fields which are neither perpendicular nor parallel to the film minimalization of Eq. (29) leads to a θ between zero and $\pi/2$ and therefore to a noncollinear spin structure.

The difference of the thicknesses at which the two collinear phases become unstable defines the width $\delta L_r=L_r^{\perp}-L_r^{\parallel}$ of the canted region which can be expressed as

$$\frac{\delta L_r}{L_r}=-\frac{2K_4(L_r)+4\left(\Delta(0,L_r)+\Delta\left(\frac{\pi}{2},L_r\right)\right)}{K_s+K_i}. \tag{37}$$

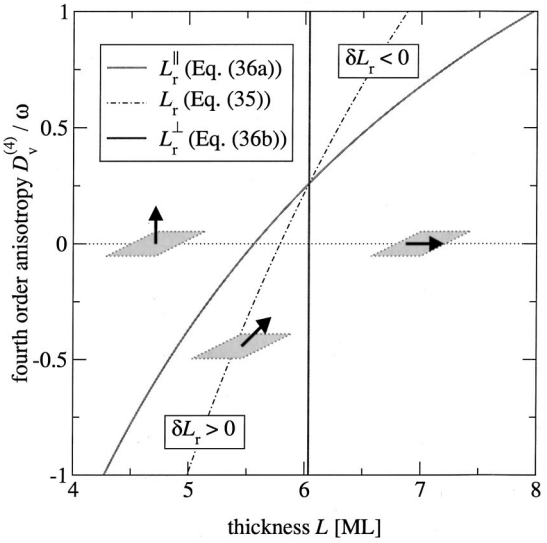

FIG. 1. Phase diagram of a *sc* Fe-like system in the $D_v^{(4)}$–L plane. The parameters are (Refs. 9 and 20) $J/\omega=2.6\times10^3$, $D_s^{(2)}/\omega=37$, and $D_{v,i}^{(2)}=D_{s,i}^{(4)}=0$.

Thus a negative fourth-order anisotropy energy $D_\lambda^{(4)}$ increases the width of the canted phase but even without such a term a canted region can be observed due to the effective anisotropy $\Delta(\theta,L_r)$. If the numerator of the right-hand side of Eq. (37) is positive, a canted phase occur, while for negative numerator we find a discontinuous transition with hysteresis. To illustrate these results, in Figs. 1 and 2 we show the phase diagram in the $D_v^{(4)}$–L plane of a sc Fe-like system and a fcc Ni-like system, respectively. The canted phase at $D_v^{(4)}=0$ is stabilized solely by a noncollinear spin structure. Note that the phase boundary $L_r^{\perp}$ is a vertical line, as $D_v^{(4)}$ cancels in Eq. (36b).

A similar calculation can be done in finite magnetic fields. If the field is orientated perpendicular to the film the thickness at which the phase $\theta=0$ becomes unstable is shifted by

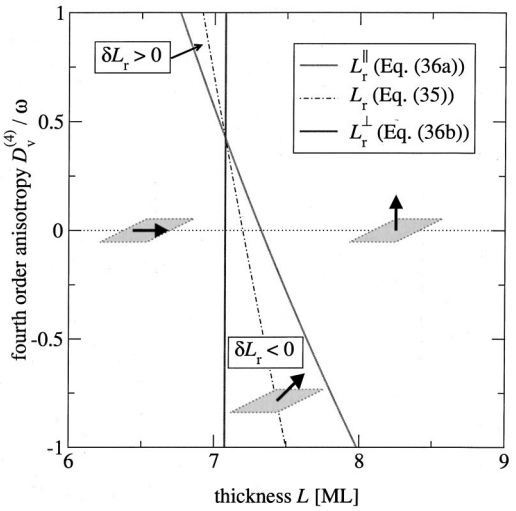

FIG. 2. Phase diagram of a fcc Ni-like system in the $D_v^{(4)}$–L plane. The parameters are (Refs. 9 and 20) $J/\omega=10^4$, $D_v^{(2)}/\omega=31$, $D_s^{(2)}/\omega=-180$, and $D_i^{(2)}=D_{s,i}^{(4)}=0$.

$$\frac{\delta L_r^{\parallel}}{L_r} = -\frac{B_{\perp}}{2K_v}, \tag{38a}$$

while for fields parallel to the film the corresponding shift is given by

$$\frac{\delta L_r^{\perp}}{L_r} = \frac{B_{\parallel}}{2K_v}. \tag{38b}$$

A phase diagram for finite temperatures and field has been obtained within mean field theory previously.[22] For small external fields the shifts of the phase boundaries obtained are linear in the field similar to the present situation.

VI. CONCLUSION

Starting from a microscopic model the ground state energy of a thin ferromagnetic film as a function of the direction of the magnetization is calculated. Explicit expressions for this energy are obtained which contain important anisotropy contributions due to noncollinear spin structures in certain parameter intervals. The microscopic parameters entering the Hamiltonian are not in a simple way related to the ground state energy. This is important for a comparison of measured and calculated anisotropy parameters. Our investigation shows that in general a canted phase is obtained and that the corresponding transitions into this phase are of second order. Analytic expressions are obtained for the width of the canted phase and its shift in external magnetic fields.

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