Reorientation transition of ultrathin ferromagnetic films

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We demonstrate that the reorientation transition from out-of-plane to in-plane magnetization with decreasing temperature as observed experimentally in Ni films on Cu(001) can be explained on a microscopic basis. Using a combination of mean-field theory and perturbation theory, we derive an analytic expression for the temperature-dependent anisotropy. The reduced magnetization in the film surface at finite temperatures plays a crucial role for this transition as with increasing temperature the influence of the uniaxial anisotropies is reduced at the surface and is enhanced inside the film. [S0163-1829(97)01918-8]

The direction of the magnetization of thin ferromagnetic films depends on various anisotropic energy contributions like surface anisotropy fields which often favor an orientation[1] perpendicular to the film, dipole interaction which favors an in-plane magnetization, and eventually anisotropy fields in the inner layers. As a consequence of these competing effects, a temperature-driven reorientation transition from an out-of-plane ordered state at low temperatures to an in-plane ordered state at high temperatures may be observed at appropriate chosen film thicknesses. Experimentally, this transition has been studied in detail for various ultrathin magnetic films.[2–4] Recently, it was found by Schulz and Baberschke[5] that ultrathin Ni films grown on Cu(001) show an opposite behavior: the magnetization is oriented in-plane for low temperatures and perpendicular at high temperatures.

Phenomenological approaches for explaining the reorientation transition usually start from the energy (or the free energy at finite temperatures) which is expanded in terms of the orientation of the magnetization vector relative to the film introducing temperature-dependent anisotropy coefficients $K_i(T)$. The temperature dependence of these coefficients is then studied experimentally (for a recent review see Ref. 6).

To better understand the mechanism responsible for the temperature-driven transition, several investigations have been done in the framework of statistical spin models. The advantage of this approach is that only a few microscopic parameters enter: besides an exchange interaction the dipole interaction and an uniaxial anisotropy in the surface layers of the film. While Moschel and Usadel[7] showed that the temperature dependence of the reorientation transition is well described qualitatively within a quantum-mechanical mean-field approach, most other authors focused on classical spin models. Extended Monte Carlo simulations on monolayers[8,9] as well as mean-field calculations of both monolayers[10] and bilayers[11] agree in the sense that a temperature-driven reorientation transition is obtained. Nevertheless, there is still a controversy with respect to the order of this transition. While Chui[8] measured the expectation value of the components of the total magnetization and obtained a second-order transition for a monolayer we found, using an improved simulation algorithm and analyzing the Monte Carlo data with a histogram method, a transition of first order in agreement with the mean-field calculations for this system.[9] Furthermore, we could show that the order of the transition depends on the number of layers and on the distribution of the uniaxial anisotropies.[11]

In all of these theoretical investigations a temperature-driven reorientation transition from an out-of-plane state at low temperatures to an in-plane state at high temperature is found for appropriate sets of parameters, which is due to a competition of a positive surface anisotropy and the dipole interaction. The interesting result for ultrathin Ni films is argued[5] to have its origin in a stress-induced uniaxial anisotropy energy in the inner layers with its easy axis perpendicular to the film. This anisotropy is in competition with the dipole interaction and a negative surface anisotropy. While the thickness-dependent transition could be explained with these anisotropies,[5] the origin of the more interesting temperature-driven transition is not yet explained on a microscopic basis. Note that the reversed reorientation recently found by MacIsaac *et al.*[12] has a different origin as it only occurs at vanishing exchange interaction. It is the purpose of this paper to show that the dipole interaction together with uniaxial anisotropies in the film indeed may lead to a temperature-driven second-order reorientation transition from an in-plane magnetized film at low temperatures to a perpendicular magnetized film at high temperatures.

We recently became aware of a paper by Jensen and Bennemann[13] on the same topic. Starting from an expansion of the free energy in terms of uniaxial anisotropy and dipole interaction and employing then a mean-field approximation following earlier work[14] they calculated numerically the temperature of both types of reorientation transitions. In contrast to this calculation we develop in the present paper a fully self-consistent mean-field theory and analyze the reorientation transition within this approach since only within a nonlinear theory the canted phase and in particular its width can be analyzed. Additionally, a self-consistent calculation of the quantities $K_i(\tau)$ introduced below which are crucial for an understanding of the nature of the transition is not possible. A linearization of the free energy which is discussed in the last part of our paper agrees with the results of Ref. 13. Within such a linearized theory the approximate location of the transition can be obtained as a temperature somewhere within the canted phase but it is not possible to calculate the width of this phase. Thus, this calculation is only meaningful

for a situation where the canted phase occupies a rather small temperature interval. Note, however, that this width can be quite large, for instance, for strongly varying anisotropy energies or certain parameter configurations[11] in which case a nonlinear approach is necessary.

The calculations are done in the framework of a classical ferromagnetic Heisenberg model consisting of L two-dimensional layers on a simple cubic lattice. The Hamiltonian reads

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

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 $\vec{r}_{ij}=\vec{r}_i-\vec{r}_j$. J is the nearest-neighbor exchange coupling constant, D_λ is the uniaxial anisotropy which depends on the layer index $\lambda=1\cdots L$, 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 and μ is the effective magnetic moment of one spin). All energies and temperatures are measured in units of J ($k_B=1$) which is fixed to $J=1$ in this paper. Note that only second-order uniaxial anisotropies D_λ enter the Hamiltonian Eq. (1). In our calculations we will restrict ourself to the case that all anisotropies are the same except at one surface, as this scenario is sufficient for explaining the basic physics of the temperature-driven reorientation transition. Furthermore we will focus on the case of $L=4$ layers. A systematic investigation of the parameter and thickness dependence of the reorientation transition and in particular a calculation of the corresponding phase diagrams is under way.[15]

In the following we assume translational invariance within the layers and therefore we set $\langle\vec{s}_i\rangle=\vec{m}_\lambda$ if $\vec{s}_i$ is a spin in layer λ. For the Hamiltonian Eq. (1) a molecular-field approximation is implemented resulting in L effective one particle Hamiltonians from which the free-energy functional can be obtained: the mean field in layer λ is given by $\vec{h}_\lambda=\Sigma_\mu \mathbf{X}_{\lambda\mu}\vec{m}_\mu$ where $\mathbf{X}_{\lambda\mu}$ contains both exchange and dipole interaction. With the order parameter $\mathbf{M}=(\vec{m}_1,\ldots,\vec{m}_L)$ the Hamiltonian in layer λ becomes

$$\mathcal{H}_\lambda^{\mathrm{MF}}(\mathbf{M})=\vec{h}_\lambda\cdot\left(\frac{1}{2}\vec{m}_\lambda-\vec{s}_\lambda\right)-D_\lambda(s_\lambda^z)^2. \tag{2}$$

Integrating this mean-field Hamiltonian over the surface of the unit sphere in each layer yields the free energy per surface element,

$$\mathcal{F}(T,\mathbf{M})=-T\sum_{\lambda=1}^{L}\ln\oint d\vec{s}_\lambda e^{-\mathcal{H}_\lambda^{\mathrm{MF}}(\mathbf{M})/T}. \tag{3}$$

Due to the in-plane rotational invariance of Eq. (2) we can set $m_\lambda^x=0$ and thus the free energy in Eq. (3) depends on the $2L$ components m_λ^y and m_λ^z of $\mathbf{M}$ and is stationary with respect to variations of these quantities. This variation is done in two steps: First we minimize the free energy Eq. (3) with the constraint that the azimuth angle ϑ of the total magnetization $\vec{m}=L^{-1}\Sigma_\lambda\vec{m}_\lambda$ is fixed, and expand the resulting constrained free energy in powers of $\cos(\vartheta)$ to give the angle-dependent free energy

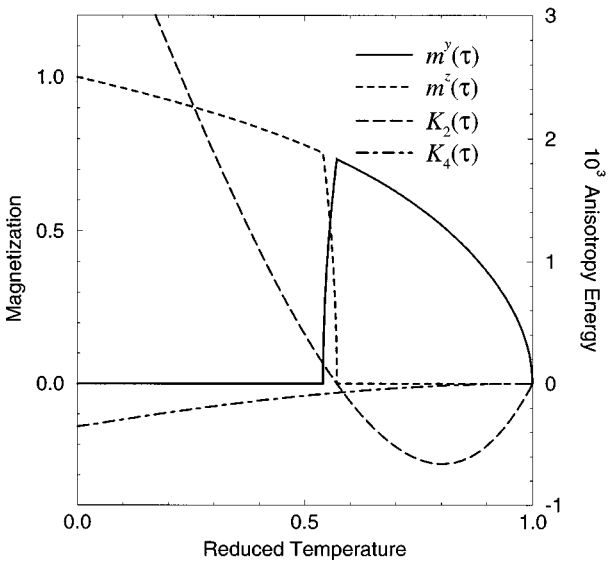

FIG. 1. Magnetization components and anisotropy energies for an Fe-type system with $L=4$ layers. $J=1$, $D_1/J=14\times10^{-3}$, $D_{\lambda>1}=0$, $\omega/J=38\times10^{-5}$. The parameters are based on iron.

$$\mathcal{F}(\tau,\vartheta)=\mathcal{F}_0(\tau)-K_2(\tau)\cos^2(\vartheta)-K_4(\tau)\cos^4(\vartheta)-\cdots, \tag{4}$$

with the reduced temperature $\tau=T/T_C$ (T_C is the Curie temperature of the film) and temperature-dependent expansion coefficients $K_i(\tau)$. These quantities are usually introduced phenomenologically. However in our approach we can calculate these coefficients $K_i(\tau)$ from the microscopic parameters of the system. The equilibrium free energy is then obtained as the minimum of Eq. (4) with respect to ϑ.

In this notation, the two reorientation transition temperatures τ_r^z, where $m^z\to0$, and τ_r^y, where $m^y\to0$, are given by the conditions

$$0=K_2(\tau_r^z), \tag{5a}$$

$$0=K_2(\tau_r^y)+2K_4(\tau_r^y). \tag{5b}$$

Figure 1 shows the temperature dependence of the components of the total magnetization $\vec{m}(\tau)$ and the anisotropy coefficients $K_2(\tau)$ and $K_4(\tau)$ for a situation where one of the layers (the surface layer) has a positive uniaxial anisotropy, $D_1>0$, and the others are set to $D_{\lambda>1}=0$. $K_6(\tau)$ and higher-order terms are nearly two magnitudes smaller than $K_2(\tau)$ and therefore are not depicted. This is the situation encountered, for instance, in ultrathin Fe films. The ground-state magnetization of the system is perpendicular to the film. Increasing the temperature, the magnetization switches continuously from the perpendicular direction at a temperature τ_r^y to the in-plane direction at τ_r^z.

For Ni films on Cu(001) there is a positive uniaxial volume anisotropy in the inner layers favoring perpendicular orientation, and eventually a negative anisotropy on the surface.[5] In competition to these energies is the dipole inter-

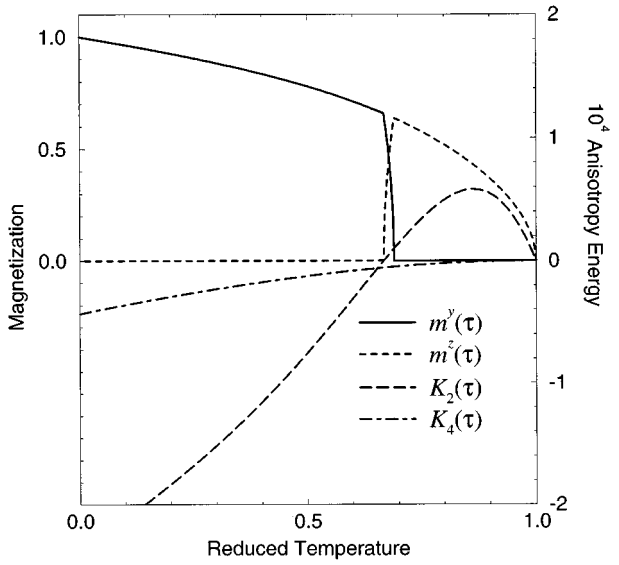

FIG. 2. Magnetization components and anisotropy energies for a Ni-type system with $L=4$ layers. $J=1$, $D_1/J=-3.5\times10^{-3}$, $D_{\lambda>1}/J=1.5\times10^{-3}$, $\omega/J=5\times10^{-5}$. The parameters are based on nickel.

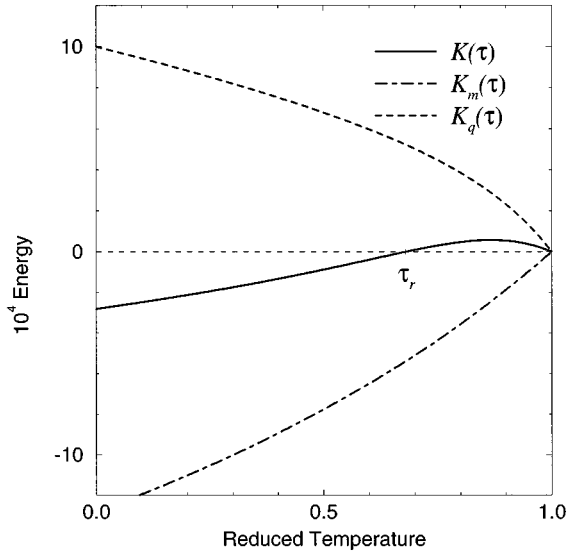

FIG. 3. Total anisotropy $K(\tau)$ and its two parts $K_q(\tau)$ and $K_m(\tau)$ from Eq. (7) for a Ni-type system. The model parameters are the same as in Fig. 2.

action which always favors in-plane magnetization. Figure 2 shows the temperature dependence of the components of the total magnetization vector and the anisotropy coefficients $K_i(\tau)$ for a Ni-type system with $L=4$ layers. The exchange interaction J is estimated from the Curie temperature of bulk nickel, the dipole constant ω is calculated from the ground-state magnetic moment and the lattice constant, and the anisotropy energies are taken from the experiment.[5] For these parameters with increasing temperature the magnetization starts to cant at a temperature τ_r^z and reaches the perpendicular state at τ_r^y as observed experimentally in Ni films. These results were obtained numerically by solving the corresponding mean-field equations.

In order to understand both the normal and the reversed reorientation transition we additionally applied a perturbation theory to the mean-field Hamiltonian Eq. (2) considering ω and D_λ as small perturbations of the pure isotropic Heisenberg Hamiltonian which is justified in view of the smallness of these parameters. We will only give the results of these calculations in this paper, the complete derivation will be reported in detail in a forthcoming paper.[15]

The total anisotropy $K(\tau)$ of the system is defined as the difference of the free energies of the in-plane state and the out-of-plane state

$$K(\tau)=\mathcal{F}(\tau,\pi/2)-\mathcal{F}(\tau,0)=K_2(\tau)+K_4(\tau)+\cdots. \quad (6)$$

When we neglect the narrow canted phase, the reorientation temperature τ_r is given by the condition $K(\tau_r)=0$. If the first derivative $\partial_\tau K(\tau_r)<0$, we have a normal transition from out-of-plane to in-plane magnetization direction, otherwise the transition is reversed.

In the framework of a perturbation theory we can derive an analytical expression for $K(\tau)$ involving the absolute value of the layer magnetizations $m_\lambda(\tau)$ and the fluctuations transversal to the magnetization direction $q_\lambda(\tau)=\langle(s_\lambda^\perp)^2\rangle(\tau)$, both calculated with the unperturbed Hamiltonian:

$$K(\tau)=K_q(\tau)+K_m(\tau)=\sum_{\lambda=1}^{L} D_\lambda[1-3q_\lambda(\tau)]$$

$$-\frac{3\omega}{4}\sum_{\lambda,\lambda'=1}^{L} m_\lambda(\tau)\Phi_{|\lambda-\lambda'|}m_{\lambda'}(\tau). \quad (7)$$

The constants Φ_δ contain the effective dipole interaction between the layers and can be calculated numerically to give $\Phi_0=9.0336$, $\Phi_1=-0.65493$, and $\Phi_{\delta>1}=\mathcal{O}(e^{-2\pi(\delta-1)})$.

At the critical temperature $K(\tau)$ vanishes and hence $K(\tau)$ must be curved in order to have another zero at a temperature $\tau_r<1$. Furthermore, a positive curvature is necessary for a normal reorientation transition, while a negative curvature of $K(\tau)$ is necessary for a reversed reorientation. Hence we will focus on the second derivatives of Eq. (7) and start with the dipole part $K_m(\tau)$. It turns out that $\partial_\tau^2 K_m(\tau)>0$ for all film thicknesses and temperatures since ω is positive and the main contribution of the sum is proportional to $\Sigma_\lambda m_\lambda^2(\tau)$ which always has a negative curvature. Thus the dipole interaction always favors the normal reorientation and can never lead to a reversed transition in an exchange-dominated system.

Now we will examine $K_q(\tau)$. First note that $q_\lambda(0)=0$ and $q_\lambda(1)=1/3$ in the unperturbated case. For $L=1$ and $L=2$ we have $q_\lambda(\tau)=\tau/3$ in the mean-field approximation, and then $K_q(\tau)=\Sigma_\lambda D_\lambda(1-\tau)$, i.e., the second derivative vanishes in this case. Consequently in systems with $L\leqslant2$ layers we only find normal reorientation transitions from the out-of-plane direction at low temperatures to an in-plane direction at higher temperatures.

This is not the case for $L>2$ layers since then, due to the reduced surface magnetization at finite temperatures, the transversal fluctuations in the surface layers $q_s(\tau)$ are en-

hanced [$q_s(\tau) \geqslant \tau/3$, $\partial_\tau^2 q_s(\tau) < 0$]. Combined with a negative surface anisotropy D_s this may lead to a negative curvature of $K_q(\tau)$. Furthermore, the transversal fluctuations in the inner layers are reduced by this effect [$q_b(\tau) \leqslant \tau/3$, $\partial_\tau^2 q_b(\tau) > 0$] and, when combined with a positive uniaxial anisotropy in the inner layers enhance the negative curvature of $K_q(\tau)$.

In Fig. 3 $K(\tau)$ is depicted together with the two competing parts $K_q(\tau)$ and $K_m(\tau)$ from Eq. (7) for the same parameters as in Fig. 2. A transition is obtained with increasing temperature because $K_q(\tau)$ tends slower to zero than $K_m(\tau)$.

In summary we have shown that the temperature-driven reorientation transitions seen in ultrathin ferromagnetic films are well described within a mean-field approximation if second-order uniaxial anisotropies and the dipole interaction are included in the Hamiltonian. In particular we can relate the unusual transition seen in Ni films to a microscopic model in which a positive uniaxial anisotropy energy is present in the inner layers. Additionally we can calculate the parameters $K_i(T)$ usually introduced phenomenologically from microscopic parameters of the system.

The $L=4$ layer film considered serves as a simple system showing the Ni-type transition, while the Fe-type transition is already observed in monolayers if the parameters are adjusted properly. This has a rather interesting physical origin: For systems with $L=1$ or $L=2$ layers the unperturbed system is homogeneous as every lattice site has the same environment. It turns out that in this case only a reorientation transition from out-of-plane to in-plane can occur, provided the exchange interaction is large with respect to the uniaxial anisotropies and the dipole interaction. When the film thickness $L>2$, the magnetization is not homogeneous through the film as the surface layers have a reduced magnetization at finite temperatures. This leads to an enhancement of the transversal fluctuations at the surface and to a reduction of these fluctuations in the inner of the film. Hence the influence of the uniaxial anisotropies is reduced at the surface and enhanced inside the film favoring a spin orientation parallel to the easy axis of the inner layer. This effect may lead to a temperature-driven reorientation transition of the type observed in nickel.

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