

## Influence of the dipole interaction on the direction of the magnetization in thin ferromagnetic films

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(Received 12 November 1993; revised manuscript received 19 January 1994)

In this paper we investigate the influence of a surface anisotropy and the dipole interaction on the magnetization of thin films. The surface anisotropy favors a perpendicular orientation while the dipole interaction favors an in-plane magnetization. It is shown that these competing interactions for certain values of the parameters may result in a temperature-driven switching transition from an out-of-plane to an in-plane ordered state.

### I. INTRODUCTION

Recent experiments on thin ferromagnetic films<sup>1,2</sup> and multilayers<sup>3-6</sup> have shown that the magnetic behavior of these systems is strongly affected by anisotropy fields which act on the spins in the surface and at the interface. These experiments show that for many systems the direction of the magnetization vector is a very sensitive function of the temperature. At low temperatures the anisotropy fields force the magnetization vector perpendicular to the surface, but by increasing the temperature the magnetization vector switches to an in-plane direction. The switching temperature decreases by increasing the film thickness.<sup>1</sup>

To understand this effect one has to recall that there appears to be a competition between the surface anisotropy which may favor a perpendicular magnetization and the long-range dipole interaction which favors an in-plane magnetization. Theoretical work on this problem is mostly focused on semi-infinite systems. Mills<sup>7,8</sup> has shown that in a semi-infinite ferromagnet with an external field parallel to the surface in presence of a strong surface anisotropy a spin canting can occur. A similar spin canting at the surface of a semi-infinite Heisenberg ferromagnet is found by Endo<sup>9</sup> who considered this model in a molecular-field approximation. For different anisotropy parameters at the surface and in the bulk he obtained a canted, an in-plane, and a perpendicular spin state. Similar results are found by O'Hanley and Woods<sup>10</sup> for an exchange-coupled ferromagnet with dipole energy and a surface anisotropy in an external field within a classical continuum approach. In all these investigations the spin canting at the surface is not driven by the temperature but rather by different anisotropy constants acting at the surface and in the bulk.

Thin films have been investigated by Jensen and Bennemann<sup>11,12</sup> within a continuum approach. They

found a switching of the magnetization vector between a perpendicular and an in-plane ordering by increasing the temperature. They explained this interesting behavior as due to a larger entropy of the in-plane spin state as compared to a perpendicular spin state. This reorientation of the magnetization by increasing the temperature is also found for a two-dimensional Heisenberg monolayer by Pescia and Pokrovsky<sup>13-15</sup> with a renormalization-group approach and by Taylor and Gyorffy<sup>16</sup> who studied a two-dimensional classical Heisenberg system. The influence of a magnetic field on the magnetization of an ultrathin film is investigated by Erickson and Mills.<sup>17,18</sup>

The increasing interest in the behavior of thin films and multilayers especially experimentally<sup>1-6</sup> calls for theoretical work in which in particular the interplay between surface/interface anisotropies, long-range dipole interaction, and film thickness is elucidated. As a first step in this direction we study in this paper a three-layer film with surface anisotropies and dipole interaction. The calculations are done within a quantum-mechanical Heisenberg model which is treated within mean-field theory. The main result of our investigation is that for different values of the surface anisotropy and the dipole interaction a small temperature region where the magnetization vector switches from the out-of-plane to the in-plane direction is found. For films with a very strong surface anisotropy it is possible that the magnetization of the film is perpendicular for all temperatures up to  $T_c$ . On the other hand it is also possible that for films with a very strong dipole interaction the magnetization vector switches from the out-of-plane to the in-plane direction at rather low temperatures.

### II. THEORY

We consider a model consisting of three ferromagnetic layers with a square structure. The model is described by the Hamiltonian

$$\mathcal{H} = -\frac{J}{2} \sum_{l,m} \sum_{i,j} \tilde{\mathbf{S}}_{li} \cdot \tilde{\mathbf{S}}_{mj} - \sum_{l,i} D_l \left( \tilde{S}_{li}^z \right)^2 + \frac{w}{2} \sum_{l,m} \sum_{i,j} \frac{\left[ \tilde{\mathbf{S}}_{li} \cdot \tilde{\mathbf{S}}_{mj} - 3 \left( \tilde{\mathbf{S}}_{li} \cdot \mathbf{u}_{li,mj} \right) \left( \mathbf{u}_{li,mj} \cdot \tilde{\mathbf{S}}_{mj} \right) \right]}{r_{li,mj}^3}, \quad (1)$$

where the  $\tilde{S}_{li}$  are spin operators and the  $z$  axis is normal to the surface of the film. The indices  $l$  and  $m$  denote the number of the layer and  $i$  and  $j$  are the lattice points at the sites  $\mathbf{r}_i$  and  $\mathbf{r}_j$  in the  $x$ - $y$  plane.  $J$  is the ferromagnetic nearest-neighbor coupling constant. Due to the broken symmetry at the surfaces of the film a strong anisotropy in the surface layers is expected.<sup>19</sup> In the first and the third layers we assume a positive surface anisotropy  $D_l$  ( $D_1 = D_3 > 0$ ) which favors an orientation of the spins at the surface in the  $z$  direction and in the second layer the anisotropy constant ( $D_2$ ) is set to zero.  $\mathbf{u}_{li,mj} = \frac{\mathbf{r}_{li,mj}}{r_{li,mj}}$  is the relative position vector of the spins  $\tilde{S}_{li}$  and  $\tilde{S}_{mj}$  and  $w = \frac{\mu_0}{4\pi} \frac{(g \cdot \mu_B)^2}{a^3}$  is the strength of the long-range dipole interaction ( $a$  is the lattice constant).

In the following we assume translational invariance within the layers, i.e., the expectation values  $\langle \tilde{S}_{mj}^x \rangle$  and  $\langle \tilde{S}_{mj}^z \rangle$  are independent of  $j$ :

$$\begin{aligned} \langle \tilde{S}_{mj}^x \rangle &= S_m^{\parallel} , \\ \langle \tilde{S}_{mj}^z \rangle &= S_m^{\perp} . \end{aligned} \quad (2)$$

For the Hamiltonian (1) a molecular-field approximation is implemented resulting in the following effective Hamiltonian:

$$\begin{aligned} \mathcal{H} &= \sum_{l,i} \mathcal{H}_{li}^{\text{mol}} , \\ \mathcal{H}_{li}^{\text{mol}} &= \tilde{S}_{li}^x \Pi_l^x + \tilde{S}_{li}^z \Pi_l^z - D_l \left( \tilde{S}_{li}^z \right)^2 - T_l \end{aligned} \quad (3)$$

with

$$\begin{aligned} \Pi_l^x &= w \sum_{m,j} \left[ \frac{S_m^{\parallel}}{r_{li,mj}^3} - 3 \frac{u_{lj,mj}^x \cdot \{ \mathbf{u}_{li,mj} \cdot \langle \tilde{S}_{mj} \rangle \}}{r_{li,mj}^3} \right] \\ &\quad - K_l^x , \\ K_l^x &= 4 S_l^{\parallel} + S_{l-1}^{\parallel} + S_{l+1}^{\parallel} , \\ T_l &= \frac{1}{2} \left[ S_l^{\parallel} \Pi_l^x + S_l^{\perp} \Pi_l^z \right] . \end{aligned} \quad (4)$$

The equations for  $\Pi_l^z$  are obtained from  $\Pi_l^x$  by replacing  $x \rightarrow z$  and  $S_l^{\parallel} \rightarrow S_l^{\perp}$ . Note that  $\Pi_l^x$  and  $\Pi_l^z$  do not depend on  $i$  due to the translational invariance of the local magnetization within the layers. The energy eigenvalues  $\epsilon_{l\alpha}$  of this Hamiltonian in case of a spin 1 system are obtained from the equation

$$\lambda_{l\alpha}^3 - D_l \lambda_{l\alpha}^2 - \left( (\Pi_l^z)^2 + (\Pi_l^x)^2 \right) \lambda_{l\alpha} + D_l (\Pi_l^z)^2 = 0 \quad (5)$$

with  $\lambda_{l\alpha} = \epsilon_{l\alpha} + D_l + T_l$  and  $\alpha$  labels the three eigenvalues. For the expectation values  $S_l^{\parallel}$  and  $S_l^{\perp}$  we obtain

$$\begin{aligned} S_l^{\parallel} &= 2 \sum_{\alpha=1}^3 \frac{1}{N_{l\alpha}} \left( \lambda_{l\alpha}^2 - (\Pi_l^z)^2 \right) \lambda_{l\alpha} \Pi_l^x P_{l\alpha} , \\ S_l^{\perp} &= 2 \sum_{\alpha=1}^3 \frac{1}{N_{l\alpha}} (\Pi_l^z)^2 \Pi_l^z \lambda_{l\alpha} P_{l\alpha} \end{aligned} \quad (6)$$

with

$$N_{l\alpha} = (\Pi_l^x)^2 \left( (\Pi_l^z)^2 + \lambda_{l\alpha}^2 \right) + \left( (\Pi_l^z)^2 - \lambda_{l\alpha}^2 \right)^2 \quad (7)$$

and

$$P_{l\alpha} = \frac{\exp(-\beta \lambda_{l\alpha})}{\sum_{\alpha=1}^3 \exp(-\beta \lambda_{l\alpha})} . \quad (8)$$

Finally, the free energy is given by

$$F = N_{\parallel} \sum_{l=1}^N \left[ -D_l - T_l - \frac{1}{\beta} \ln \left\{ \sum_{\alpha=1}^3 \exp(-\beta \lambda_{l\alpha}) \right\} \right] , \quad (9)$$

where  $N_{\parallel}$  is the number of atoms in each layer,  $N$  is the number of layers of the system, and  $\beta = \frac{1}{k_B T}$ . In the case of an  $N$ -layer system we get  $N$  equations for  $S_l^{\parallel}$  and  $S_l^{\perp}$  which have to be solved in a self-consistent manner.

A self-consistent solution minimizes the free energy functional (9). In this paper we restricted ourselves to a three-layer system,  $N = 3$ . To obtain a self-consistent solution we start with a random orientation of the spin expectation values  $\langle S_l^{\parallel} \rangle$  and  $\langle S_l^{\perp} \rangle$  in the three layers and iterate Eq. (6). A self-consistent solution is obtained after a large number of iterations. The number of iterations depends very much on the parameters chosen, especially on temperature. The amount of computer time needed can be reduced considerably by starting the iteration with a spin configuration which is already close to the self-consistent one. This can be achieved by changing the temperature slightly and starting the iteration for this new temperature with the self-consistent spin configuration obtained before. To make sure that this procedure does not produce metastable solutions we checked for a reduced set of temperature values that the same self-consistent solution is also obtained by starting with a random spin configuration.

Due to the slow convergence of the dipole sums many terms are needed. In the numerical calculations the sums over the dipole fields are extended to a distance of 1500 (in units of the lattice constant). In addition, a large number of iterations are needed to achieve self-consistency. The numerical effort can be reduced considerably by first rearranging the dipole sums into rapidly converging series.<sup>20</sup> Such an approach is of particular importance when one wants to extend the present calculations to thicker films and to multilayers.

### III. RESULTS AND DISCUSSION

The complex behavior of the magnetization of thin films is due to a competition between a surface anisotropy which favors a perpendicular orientation of the magnetization and the dipole interaction which favors an in-plane magnetization. We found that for a rather strong surface anisotropy ( $D_1 = D_3 = 0.2$ ,  $D_2 = 0$ ; all quantities in units of  $J$ ) and a dipole interaction of  $w = 0.01$  the ground-state magnetization is perpendicular to the film. Increasing the temperature the magnetization switches continuously from the out-of-plane direction at a reduced

temperature  $t_{\perp} \approx 0.297$  to the in-plane direction at a reduced temperature of  $t_{\parallel} \approx 0.374$  (Fig. 1). The switching temperature  $t_s$  defined as  $t_s = \frac{1}{2}(t_{\perp} + t_{\parallel})$  which is well below the Curie temperature and is a very sensitive function of the ratio of the strength of the surface anisotropy and the dipole interaction. For a weaker dipole interaction ( $w = 0.0075$ ) we find a much higher switching temperature of  $t_s \approx 0.79$  (Fig. 2). Note that the temperature interval where the transition from the out-of-plane to the in-plane state occurs is very small in the case of a weak dipole interaction compared to the strong dipole interaction. From this result we may extrapolate to the behavior of thicker films: increasing the film thickness the influence of the surface anisotropy relative to the dipole interaction in the bulk is decreasing resulting also in a decreasing switching temperature.<sup>1</sup> At a critical value  $D_s$  of this surface anisotropy this switching temperature approaches  $T = 0$ , i.e., to have a perpendicular ground-state magnetization  $D$  has to be larger than  $D_s$ . This critical value  $D_s$  depends very sensitively on the strength of the dipole interaction (and on film thickness). For a three-layer film and  $w = 0.01$  its value is about  $D_s \approx 0.195$ .

The physical origin of the reorientation of the magnetization vector is a significant entropy increase of the system when going from a perpendicular magnetization to an in-plane ordered state, an idea formulated by Jensen and Bennemann.<sup>11,12</sup> This can be seen explicitly from the calculated free energy and the entropy obtained from it. A stable self-consistent solution of Eq. (6) is characterized by the minimum of the free energy Eq. (9). We have calculated the free energy and the corresponding entropy as function of the spin orientation and found that for temperatures in a broad interval around  $t_s$  the maximum of the entropy is always given for spin orientations parallel to the film. For small temperatures the entropy contribution to the free energy is small so that the spin orientation is primarily determined by the internal energy which favors perpendicular orientation. However, by increasing the temperature the entropy contribution

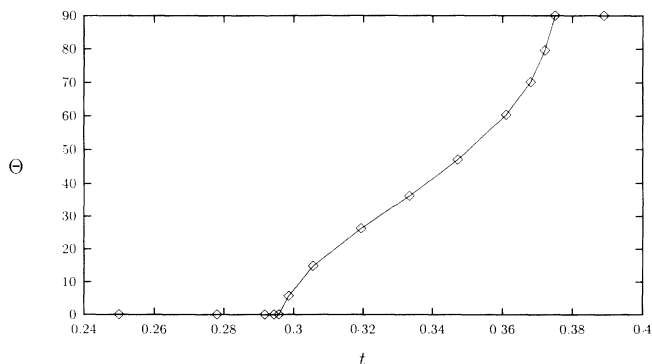


FIG. 1. Temperature dependence of the angle of the magnetization vector of the first layer for a thin ferromagnetic film ( $N = 3$ ) with  $D_1 = D_3 = 0.2$ ,  $D_2 = 0$ , and  $w = 0.01$ .  $\theta$  is the angle between the magnetization vector and the film normal and  $t$  is the reduced temperature  $T/T_c$ .

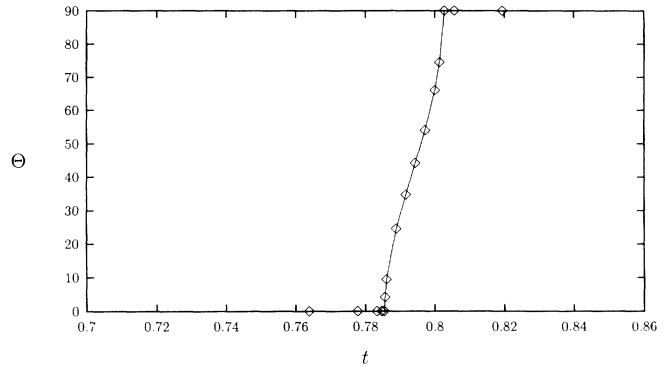


FIG. 2. The same system as in Fig. 1 but for  $w = 0.0075$ .

increases leading to a shift of the minimum of the free energy and to an in-plane state of the magnetization, i.e., resulting in a switching transition of the spins.

In all our calculations so far (cf. Figs. 1 and 2 and additional data not shown here) the switching transition is continuous. For very small dipole interactions a tendency to a rather sharp transition from the out-of-plane direction to the in-plane direction can be seen (cf. Fig. 2) but nowhere did we find a discontinuous transition. Note that experimentally both cases are hard to distinguish due to the rather small temperature interval in which the transition takes place. In a related work by Levinson, Luban, and Shtrikman<sup>21</sup> on the orientation of the magnetization of a three-dimensional bulk system both types of transitions were observed depending on system parameters. In this work the free energy  $F$  is obtained as a function of a single canting angle  $\theta$  which has to be chosen in such a way that  $F$  becomes minimal. In the present case the free energy depends on two angles  $\theta_i$ , the canting angles of the layer magnetizations, but both angles differ only slightly for the minimal  $F$ . Thus it is instructive to calculate  $F$  as a function of only one canting angle  $\theta$  neglecting the difference between  $\theta_1$  and  $\theta_2$  (Fig. 3). This free energy can be interpreted as an effective free energy within a Landau-Ginzburg framework with  $\theta$  as an order parameter and it is possible to calculate within this framework semiheuristicly a correlation length  $\xi(T)$  from which information about the quality of the mean-field approximation can be obtained similar to Ref. 21. The correlation length appears to be very large. Even far away from the critical temperatures  $t_{\perp}$  and  $t_{\parallel}$  it is larger than 1000 lattice constants [ $\xi(t_s) \approx 1200$  lattice constants for  $w = 0.01$ ]. This very large correlation length gives us the confidence that the mean-field approach used in the present paper is quite reliable except perhaps in tiny regions around the critical temperatures.

Our results explain the switching of the magnetization vector which is observed experimentally in Fe/Tb multilayers<sup>3-6</sup> qualitatively. Multilayers which consist of Tb layers with different thicknesses and which are prepared at different temperatures will have on the one hand different anisotropy values at the interface and on the other hand their dipole interaction depends on thickness. As was discussed above, the orientation of the magnetization depends very sensitive on these parameters. There-

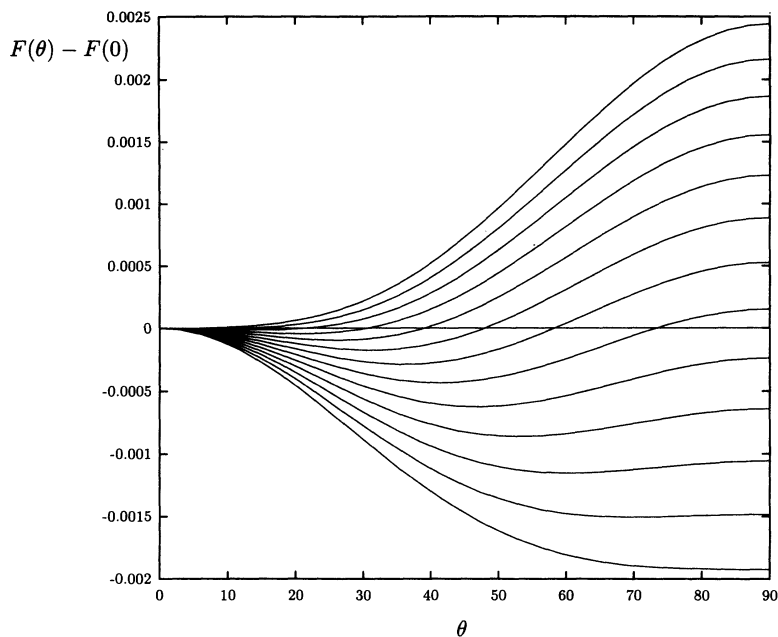


FIG. 3. The free energy as a function of canting angle for temperatures between  $t = 0.292$  (upper curve) –  $0.375$  (lower curve) for a dipole interaction of  $w = 0.01$ . Note that the canting angles in different layers are set equal as explained in the text.

fore, for these multilayers a complicated behavior of the magnetization vector at the interface as a function of film thickness and film preparation is expected to occur. The switching observed can be understood as the result of these competing interactions on which the magnetization depends very sensitively. For a qualitative comparison with experiments, however, more information about microscopic parameters which enter the theory is needed.

The detailed behavior of the magnetization vector at the interfaces of multilayers consisting of different magnetic materials and the influence of the dipole interaction in thicker films is left for future research.

#### ACKNOWLEDGMENTS

This work was supported by the Deutsche Forschungsgemeinschaft through Sonderforschungsbereich 166.

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