Influence of magnetic fields on the spin reorientation transition in ultra-thin films

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Abstract

The dependence of the spin reorientation transition in ultra-thin ferromagnetic films on external magnetic fields is studied. For different orientations of the applied field with respect to the film, phase diagrams are calculated within a mean field theory for the classical Heisenberg model. In particular we find that the spin reorientation transition present in this model is not suppressed completely by an applied field, as the magnetization component perpendicular to the field may show spontaneous order in a certain temperature interval.

1 Introduction

Experimentally it became possible in recent years to grow epitaxial thin films of ferromagnetic materials on non-magnetic 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.

One of the very interesting effects one observes in ultra-thin ferromagnetic films is a reorientation of the spontaneous magnetization by varying
either the film thickness or the temperature. For not too thin films the magnetization generally is in-plane due to the dipole interaction (shape anisotropy). On the other hand in very thin films this may change due to various competing anisotropy energies in these materials. At the surfaces of the film due to the broken symmetry uniaxial anisotropy energies which may favor a perpendicular magnetization may develop [1] or in the inner layers of the film due to strain induced distortion bulk anisotropy energies may occur absent or very small in the ideal crystal.

A perpendicular anisotropy has been observed for instance for ultra-thin Fe-films grown on Ag(100) where in the ground state the magnetization is perpendicular to the film [2–4]. Increasing the temperature or the film thickness the magnetization stays perpendicular until at a certain temperature it starts canting reaching finally an in-plane orientation for temperatures well below the ordering temperature $T_c$.

A different type of spin reorientation transition (SRT) occurs in Ni grown on Cu(001). Very thin films then show a tetragonal distortion resulting in a stress-induced uniaxial anisotropy energy in the inner layers with its easy axis perpendicular to the film. If this is strong enough the magnetization is in-plane in the ground state but switches to an orientation perpendicular to the film for higher temperature or film thickness [5].

Phenomenologically in order to describe the SRT the energy (or the free energy at finite temperatures) is expanded in terms of the orientation of the magnetization vector relative to the film introducing temperature dependent anisotropy coefficients $K_i(T)$ compatible with the underlying symmetry of the film. The temperature dependence of these coefficients is then studied experimentally (for a review see [6]). Theoretically, it has been shown by various groups that the physics of the SRT can be understood quite well within the framework of statistical spin models [7–10]. Note that both types of SRTs observed experimentally can be explained within the same approach [11, 12] and that the anisotropy coefficients $K_i(T)$ can be calculated [12].
2 The model

In the present paper we focus on the dependence of the SRT on external magnetic fields. The calculations are done in the framework of a classical ferromagnetic Heisenberg model on a simple cubic lattice consisting of \( L \) two-dimensional layers with the \( z \)-direction normal to the film. The Hamiltonian reads

\[
\mathcal{H} = -\frac{J}{2} \sum_{\langle ij \rangle} \mathbf{s}_i \cdot \mathbf{s}_j - \sum_i D_\lambda (s^z_i)^2 - \sum_i \mathbf{B} \cdot \mathbf{s}_i + \frac{\omega}{2} \sum_{ij} r^{-3}_{ij} \mathbf{s}_i \cdot \mathbf{s}_j - 3 r^{-5}_{ij} (\mathbf{s}_i \cdot \mathbf{r}_{ij})(\mathbf{r}_{ij} \cdot \mathbf{s}_j),
\]

where \( \mathbf{s}_i = (s^x_i, s^y_i, s^z_i) \) are spin vectors of unit length at position \( \mathbf{r}_i = (r^x_i, r^y_i, r^z_i) \) in layer \( \lambda \), and \( \mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j \). \( J \) is the nearest-neighbor exchange coupling constant, \( D_\lambda \) is the local uniaxial anisotropy which depend on the layer index \( \lambda = 1 \ldots L \), \( \mathbf{B} \) denotes the external magnetic field with the effective magnetic moment \( \mu \) 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 \( a \) (\( \mu_0 \) is the magnetic permeability). All energies and temperatures are measured in units of \( \omega \), and we set \( k_B = 1 \). Assuming translational invariance of the Hamiltonian the local magnetization in mean field theory only depends on the layer index, i.e. \( \langle \mathbf{s}_i \rangle = \mathbf{m}_\lambda \) if \( \mathbf{s}_i \) is a spin in layer \( \lambda \). A molecular field approximation for the Hamiltonian (1) results in \( L \) effective one particle Hamiltonians from which the free energy functional can be obtained. For more details the reader is referred to Ref. [12].

For appropriate sets of parameters a SRT in zero magnetic field from perpendicular orientation at low temperatures to in-plane orientation at high temperatures is found [10]. In this contribution we will investigate the influence of an applied field on this transition, which has only been addressed in the framework of a phenomenological approach [13] until now.
Figure 1: Magnetization components $m^{xy}$ and $m^z$ of a bilayer in a small perpendicular magnetic field $B/\omega = 0.6\hat{z}$ versus reduced temperature $\tau$. The thin lines are calculated for zero magnetic field.

3 Results

Figure 1 shows the magnetization components of the total magnetization $m = (m_1 + m_2)/2$ as function of reduced temperature for an applied field $B/\omega = 0.6\hat{z}$ ($\hat{z}$ is the unit vector normal to the film). A bilayer is considered with an exchange interaction $J/\omega = 60$ and uniaxial anisotropies $D_1/\omega = 12$ in the first layer and $D_2/\omega = 3$ in the other layer. This system has already been studied in zero magnetic field [10] and it is sufficient to describe the continuous transition. With this set of parameters a SRT from perpendicular orientation at low temperature to in-plane orientation at high temperatures is obtained in zero magnetic field (thin lines in figure 1). In a perpendicular magnetic field $B \parallel \hat{z}$ the component of the magnetization $m^z$ parallel to the field is finite for all temperatures. Nevertheless, at a temperature $\tau^{xy}(B) > \tau^{xy}(0)$ an in-plane spontaneous magnetization $m^{xy}$ appears as in the field free case. This in-plane phase FM$_{xy}$ is not sup-
pressed by the perpendicular external magnetic field. We observe a quite interesting reentrant phase transition: a spontaneous in-plane magnetization appears when increasing the temperature, and it vanishes at a higher temperature \( \tau_c(B) \) slightly below the Curie temperature of the field free case \( \tau_c(0) = 1 \).

The corresponding phase diagram is depicted in figure 2. Within the area encircled by the broken line there exists an ordered phase \( FM_{xy} \) with in-plane component of the magnetization \( m^{xy} > 0 \). The sketches of the magnetization with respect to the film show how the magnetization rotates from \( m = -m^z \hat{z} \) inside the paramagnetic regime PM via remanence at \( m = m^{xy} \hat{y} \) to \( m = m^z \hat{z} \) when the applied field is increased from negative to positive values at a fixed temperature \( \tau > \tau^z_r(0) \).

The phase transition at the boundary is of second order. It is important to note that this phase boundary continuously connects the reorientation temperature \( \tau^{xy}_r(B) \) and the Curie temperature \( \tau_c(B) \) for all perpendicular
magnetic fields with $|B^z| < B^z_c$ ($B^z_c/\omega \approx 1.81$ for our set of parameters). As we do not find any multi-critical points on this boundary at least in mean field theory, the critical behavior should be the same at the whole boundary and should belong to the universality class of the two dimensional dipolar Heisenberg model.

The ordered phase FM$_x$ in which the perpendicular magnetization $m^z$ shows spontaneous order is only stable at $B^z = 0$ for temperatures below the second order critical point $\tau^z_r(0)$. A first order transition is obtained when crossing this phase in a perpendicular magnetic field. This phase is similar to the ordered phase in the two-dimensional ferromagnetic Ising model. Due to the uniaxial anisotropy $D_\lambda$ it very likely will belong to the same universality class.

In figure 3 we depict the field dependence of the magnetization for several temperatures below $\tau_c(0)$. Note that this diagram is symmetric with respect to the origin. For $\tau < \tau_{xy}^r(0)$ we find a jump in $m^z$ at zero field as we
cross the phase FMz. This jump is still present for $\tau_{xy}^z(0) < \tau < \tau_{xy}^z(0)$, but now the zero-field state is canted, and with increasing field we get a transition into the paramagnetic phase where $m \parallel B$. At $\tau_{xy}^z(0)$ the $zz$-component of the static zero-field susceptibility tensor $\chi_{0}^{\mu\nu} = dm^{\mu}/dB^{\nu}|_{B=0}$ diverges (note that the same holds at $\tau_{xy}^z(0)$ for the in-plane components $\chi_{0}^{xx}$ and $\chi_{0}^{yy}$). For $\tau > \tau_{xy}^z(0)$ the perpendicular remanence is zero, nevertheless the magnetization curve has a kink when we leave the ordered phase FM$_{xy}$.

Finally, for $\tau > \tau_{c}(0)$ we find the usual paramagnetic behavior (not displayed).

A different scenario is obtained in a parallel magnetic field $B = B^y \hat{y}$. Figure 4 shows the corresponding magnetization components. Due to the in-plane field the in-plane ordered phase FM$_{xy}$ is completely suppressed, the critical points $\tau_{xy}^z(B)$ and $\tau_{c}(B)$ are not defined anymore. Nevertheless, the $z$-component of the magnetization $m^z$ shows spontaneous order at temperatures below $\tau_{xy}^z(B) < \tau_{xy}^z(0)$, where it vanishes continuously with
Figure 5: Phase diagram in a parallel magnetic field $B = B^y \hat{y}$. The curves in figure 4 are calculated along the line $B^y/\omega = 0.6$. The arrows indicate the direction of the magnetization with respect to the film.

a second order phase transition. Above this temperature no spontaneous order is present in the system.

The corresponding phase diagram for this situation is shown in figure 5. To the left of the solid line we find an ordered phase $\text{FM}_z$ with order parameter $m^z > 0$. The phase boundary is of second order and ends at $\tau = 0$ and $\mathbf{B} = \pm B^y_c \hat{y}$ with $B^y_c/\omega \approx 2.24$ for our set of parameters. The in-plane phase $\text{FM}_{xy}$ is only stable at $B^y = 0$, it starts at $\tau^{xy}(0)$ and ends at $\tau_c(0)$ in analogy to figure 2. Again we sketched the orientation of the magnetization relative to the film.

In general we find that the behavior with in-plane magnetic field is similar to the case with perpendicular magnetic field if one exchanges both in-plane and perpendicular directions and reverses the temperature. To make this symmetry even clearer, in figure 6 we depict a three dimensional phase diagram of the system. The two phases with spontaneous magnetization intersect along the line $\mathbf{B} = 0, \tau^{xy}(0) < \tau < \tau^z_c(0)$. In this
range we find spontaneous canted magnetization.

As shown in Ref. [10], a bilayer and thicker films may show both second order and first order SRTs at zero field depending on the distribution of uniaxial anisotropies $D_\lambda$: If in the bilayer the deviation $\Delta = |D_1 - D_2|$ approaches the critical value $\Delta_c \approx \omega$, the critical points $\tau_{xy}^c(0)$ and $\tau_z^c(0)$ merge into one multi-critical point where the order of the SRT in zero field changes from second to first order. If we apply these former results to this work, we conclude that the intersection width of the phases FM$_z$ and FM$_{xy}$ is finite for $\Delta > \Delta_c$ and zero for $\Delta \leq \Delta_c$. 

Figure 6: Three dimensional phase diagram in the $(\tau, B^y/\omega, B^z/\omega)$ parameter space. The arrows sketch the spontaneous magnetization inside the phases FM$_z$ and FM$_{xy}$, outside these phases we have $m \parallel B$. 
4 Summary

For the Heisenberg model with dipole interaction and uniaxial anisotropy in an external magnetic field, we calculated magnetization curves as function of both temperature and magnetic fields in the mean field approximation and determined phase diagrams in the temperature-field parameter space. We find that the spin reorientation transition present in this model is not suppressed completely by an external magnetic field, as the magnetization component perpendicular to the field may show spontaneous order. The phase diagrams reveal that the field dependent transition point $T_{xy}(B)$, where a canted magnetization occurs with increasing temperature, and the field dependent Curie temperature $T_c(B)$ are connected by a continuous phase boundary. Hence both transitions should be in the same universality class.

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