On the reorientation transition of ultra–thin Ni/Cu(001) films

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Abstract

The reorientation transition of the magnetization of ferromagnetic films is studied on a microscopic basis within a Heisenberg spin model. Using a modified mean field formulation it is possible to calculate properties of magnetic thin films with non–integer thicknesses. This is especially important for the reorientation transition in Ni/Cu(001), as there the magnetic properties are a sensitive function of the film thickness. Detailed phase diagrams in the thickness–temperature plane are calculated using experimental parameters and are compared with experimental measurements by Baberschke and Farle (J. Appl. Phys. 81, 5038 (1997)).

The direction of the magnetization of thin ferromagnetic films depends on various anisotropic energy contributions like surface anisotropy fields, dipole interaction, and eventually anisotropy fields in the inner layers. These competing effects may lead to a film thickness and temperature driven reorientation transition (RT) from an out–of–plane ordered state at low temperatures to an in–plane ordered state at high temperatures at appropriate chosen film thicknesses. Experimentally, this transition has been studied in detail for various ultra–thin magnetic films [1]. Recently, it was found by Farle et al. [2] that ultra–thin Ni–films grown on Cu(001) show the inverse behavior: the magnetization is oriented in–plane for thin
films and at low temperatures and perpendicular for thick films and at high temperatures.

It has been shown by various authors [5–8] that the mechanism responsible for the temperature driven transition can be understood within the framework of statistical spin models. The RT from an out–of–plane state at low temperatures to an in–plane state at high temperature is found to be due to a competition of a positive surface anisotropy and the dipole interaction. It can occur because the surface anisotropy has a different temperature dependence that the dipole exchange anisotropy and vanishes more quickly when approaching the Curie temperature [7, 8]. The thickness driven reversed RT in ultra–thin Ni–films is argued to have its origin in a stress–induced positive uniaxial anisotropy energy in the inner layers with its easy axis perpendicular to the film [2]. This anisotropy is in competition with the dipole interaction and with a negative surface anisotropy. This scenario can indeed lead to a reversed temperature driven RT, but now the reduced surface magnetization plays a crucial role [7, 8]. A third type of RT has recently been found theoretically, where the magnetization switches from perpendicular to in–plane direction with increasing temperature, but with decreasing film thickness [9].

In this paper we will focus on the phase diagram of Ni/Cu(001) and on the influence of microscopic fourth–order anisotropy terms. The calculations are done in the framework of a classical ferromagnetic Heisenberg model consisting of \( L \) two–dimensional layers on a face centered cubic (001) lattice. The Hamiltonian reads

\[
\mathcal{H} = -J \sum_{\langle ij \rangle} \vec{s}_i \cdot \vec{s}_j - \sum_i D^i_2 (s^z_i)^2 + D^i_{4\perp} (s^z_i)^4 + \frac{\omega}{2} \sum_{ij} r^{-3}_{ij} \vec{s}_i \cdot \vec{s}_j - 3 r^{-5}_{ij} (\vec{s}_i \cdot \vec{r}_{ij})(\vec{r}_{ij} \cdot \vec{s}_j),
\]

where \( \vec{s}_i = (s^x_i, s^y_i, s^z_i) \) are spin vectors of unit length at position \( \vec{r}_i = (r^x_i, r^y_i, r^z_i) \), and \( \vec{r}_{ij} = \vec{r}_i - \vec{r}_j \). \( J \) is the nearest-neighbor exchange coupling constant. The uniaxial and fourth–order anisotropies \( D^i_n \) are position–dependent: \( D^i_n = D^S_n \) if spin \( i \) is in the top layer, \( D^i_n = D^{S'}_n \) if spin \( i \) is
in the bottom layer, and $D_n^i = D_n^V$ otherwise. Finally, $\omega = \mu_0 \mu^2 / 4 \pi a^3$ is the strength of the long range dipole interaction on a lattice with next-neighbor distance $a$ ($\mu_0$ is the magnetic permeability and $\mu$ is the effective magnetic moment of one spin).

The Hamiltonian Eq. (1) is handled in a molecular-field approximation [8]. In the following we assume translational invariance within the layers and furthermore that the magnetization is homogeneous inside the film and only deviates at the surfaces [9]. Therefore we can set $\langle \vec{s}_i \rangle = \langle \vec{s}_\lambda \rangle$ if $\vec{s}_i$ is a spin in a volume layer ($\lambda = V$) or in the top or bottom surface layer ($\lambda = S, S'$). We will focus on the case $L > 2$, i.e. both surfaces are complete. The resulting system with three mean field spins has the effective interactions $\tilde{x}_{\lambda \mu}$, where $x_\delta$ is either the number of next neighbors, $z_\delta$, or dipole sum in fcc(001) geometry, $\Phi_\delta = \sum_{i,j} \frac{r_{i,j}^2 + r_{i+j,j}^2 - \delta^2}{r_{i,j}^2 + r_{i+j,j}^2 + \delta^2} / 2$, respectively, between layers $\lambda$ and $\lambda + \delta$. We have $\tilde{x}_{SS'} = \tilde{x}_{SS} = 0$, $\tilde{x}_{SV} = x_0$, $\tilde{x}_{SV'} = x_1$, $\tilde{x}_{VV} = x_0 + 2 x_1 (1 - L_V^{-1})$, and $\tilde{x}_{VS} = \tilde{x}_{VS'} = x_1 L_V^{-1}$, with $L_S = L - 1$ and $L_V = L - 2$. The constants $z_\delta$ and $\Phi_\delta$ are $z_0 = z_1 = 4$, $\Phi_0 = 9.034$ and $\Phi_1 = 1.429$. $\Phi_{\delta>1}$ can be neglected in our approach. Note that the coupling between the volume spin and the surface spins is asymmetric. With this method we are not restricted to films with integer values of the thickness anymore. The effective interactions $\tilde{x}_{\lambda \mu}$ enter the mean field Hamiltonian via the mean fields in layer $\lambda$, $\tilde{h}_\lambda = \sum_{\mu=1}^3 J_{\lambda \mu} \tilde{m}_\mu + \tilde{\Phi}_{\lambda \mu} \vec{W} \tilde{m}_\mu$, with the dipole exchange anisotropy $\vec{W} = \text{diag}(1/2, 1/2, -1)$. In order to determine the phase diagram of this Hamiltonian in the thickness–temperature plane, we directly calculate the phase boundaries using a stability analysis of the mean field free energy. Defining the function $e_{\text{min}} = \text{evmin}(A)$ which returns the smallest eigenvalue $e_{\text{min}}$ of the matrix $A$, we can calculate the effective anisotropy $K_2$ from the Hessian of $F_{\text{MF}}$ with respect to the azimuth angle $\vartheta_\lambda$ of the magnetization $\vec{m}_\lambda$ [10].

$$K_2(L, T, \vartheta) = \frac{1}{2} \text{evmin} \left( \frac{\partial^2 F_{\text{MF}}(L, T)}{\partial \vartheta_\lambda \partial \vartheta_\mu} \bigg|_{\vartheta_\nu = \vartheta} \right).$$

(2)

Now consider the phase with magnetization parallel to the film normal $\vec{z}$, where $\vartheta_\lambda = 0$. At thicknesses $L$ and temperatures $T$ where this phase
is stable, we have $K_2(L, T, 0) > 0$. At the phase boundaries both to the canted phase and to the paramagnetic phase, $K_2(L, T, 0)$ becomes zero, while it is negative in the canted phase and in the phase with in–plane magnetization. On the other hand, $K_2(L, T, \pi/2)$ is positive when the in–plane phase is stable and becomes negative in the phases with perpendicular component of the magnetization. In the paramagnetic phase $K_2(L, T, \vartheta) = 0$ for all $\vartheta$. Hence the reorientation temperatures are given by $K_2(L, T, x^y (L), 0) = 0$ and $K_2(L, T, x^{z}(L), \pi/2) = 0$. Next we will examine the phase diagram of this model for parameters measured on Ni/Cu(001) [3]:

The exchange interaction is approximated via the Curie temperature $T_c = 631$ K of bulk Ni, using the mean field formula $3T_c = zJ$ with $z_{fc} = 12$ for the isotropic classical Heisenberg model to give $J \simeq 13.6$ meV. This is a rather rough estimate, but note that the exact value of $J$ has nearly no influence on the following results, as long as $J$ is large compared to the other energies in the model. With $\mu = 0.62\mu_B$ and $a = 2.5\text{Å}$ we get $\omega = 1.3 \mu\text{eV} \simeq 10^{-4} J$ for the dipole constant. The uniaxial surface anisotropy is $(D_S^2 + D_S')/2 = -60 \mu\text{eV} \simeq -47\omega$ and the uniaxial volume anisotropy is $D_Y^V = +40 \mu\text{eV} \simeq 31\omega$. Note that our surface anisotropies are the sum of the experimental surface and volume part, as our surface layers also count to the volume. For simplicity we assume that the bottom surface carries the volume anisotropies, hence we get $D_2^S \simeq -125\omega$.

To check the influence of $D_{4\perp}$, we show the phase diagram for the abovementioned parameter set with $D_{4\perp} = 0$. The phase boundaries are the thin lines in figure [4]. We obtain a reversed RT as expected for Ni/Cu(001), the reorientation thickness ranges from 5ML to 7ML which is slightly lower as in the experiment [4]. We find that the canted phase exist, but it is rather narrow at these parameters. This canted phase is stable because we allow non–collinear magnetizations in different layers. To reproduce the experimental finding of the rather broad canted phase, we set the fourth–order anisotropies $D_{4\perp} \simeq -1/4 D_2^\lambda$ [4]. The phase diagram of our model with these values and slightly modified $D_2^\lambda$ is depicted in figure [4] together with experimental data obtained by Baberschke and Farle [4]. As the mean field theory always overestimates $T_c$, especially for
low dimensional systems, the critical line $T_c(L)$ from [4] cannot be reproduced well within our theory. Thus we normalized the temperature axis not with $T_c^{\text{bulk}}$, but with the Curie temperature of a film with $L = 7$ ML.

In this paper we defined a modified mean field model to describe the RT in Ni/Cu(001). Using appropriate model parameters close to experimental findings, we find a rather nice agreement of theory and experiment. To reproduce the width of the phase with canted magnetization, we need to introduce microscopic fourth–order anisotropies $D_{4\perp}$.

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[10] $K_n(\tau)$ from [8] obey $K_2(\tau) = -LK_2(L, T/T_c(L), \pi/2)$ and $\sum_{n=1}^{\infty} nK_{2n}(\tau) = LK_2(L, T/T_c(L), 0)$. 
Figure 1: Calculated phase diagram for Ni/Cu(001). For the thick lines, the model parameters are $\omega = 10^{-4}J$, $D_S^S = -100\omega$, $D_{14}^Y = D_{22}^S = 24\omega$, $D_{44}^S = 24\omega$, $D_{14}^V = D_{44}^S = -4\omega$. The thick solid line represents the Curie temperature $T_c(L)$ of the film, the dotted and dashed lines are $T_r^z(L)$ and $T_r^{xy}(L)$, respectively. The symbols are experimental measurements of the reorientation transition in Ni/Cu(001) taken from [4]. The temperature axis is normalized to the Curie temperature of a film with thickness $L = 7$ ML. The parameters for the thin lines are described in the text.