Monte Carlo simulation of critical properties of ultrathin anisotropic Heisenberg films

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Abstract. The crossover from two-dimensional monolayer to three-dimensional system in ultrathin magnetic films is studied using a Monte Carlo technique. Ultrathin magnetic films is modeled as an anisotropic Heisenberg model. Finite-size scaling of the critical exponents as a function of film thickness is investigated. It was found that the magnetic behaviour changes from two-dimensional to three-dimensional universality class with increasing film thickness.

1. Introduction
The magnetic behaviour of ultrathin films has become of great technological importance due to the applications in magnetic storage devices [1]. Magnetic order in ultrathin ferromagnetic films is very complex due to a strong influence of the shape and the magnetocrystalline anisotropies of the sample. In the past 20 years, a considerable amount of experimental results on different aspects of magnetism in ultrathin films has appeared [2]. Nevertheless it is difficult to reach general conclusions even in seemingly basic things such as the kind of magnetic order at low temperatures. In view of this complexity, theoretical work on simplified models and computer simulations are essential for rationalizing and guiding new experimental work.

The dimensional crossover of magnetic properties from two-dimensional (2D) to three-dimensional (3D) character in magnetic multilayers has currently attracted much interest as a result of both technological and fundamental importance. Of particular interest is the critical behaviour of magnetic thin films for which the dimensionality d is not well established. It is interesting to consider how magnetic properties such as the magnetization m, magnetic susceptibility χ, and critical temperature Tc depend on the thickness of the film.

Critical temperature Tc in multilayered systems are known to change from 2D to 3D values with increasing numbers of layers. Magnetic films, however, should belong to a 2D universality class owing to the correlation length being constrained by the film thickness and allowed to expand only in the in-plane direction. This is not apparent from experimental studies of thin films of Ni [3] which provide evidence of a dimensional crossover of the critical exponents β from 2D to 3D.

For magnetic systems, the spin dimensionality as well as the spatial extension determines the universality class, giving rise to a myriad of ordering phenomena on different length scales. Furthermore, there are transition regions not represented by any universality class with corresponding critical exponents, but representing something in between. For example, the thickness dependence of the critical exponents of thin magnetic films exhibit such a transition, in which the exponents are continuously varying with the thickness of the layers, from typical
two-dimensional (2D) Ising (β = 0.125) to three-dimensional (3D) Heisenberg (β = 0.364(4) [4]) behaviour.

2. Models and methods

We have performed Monte Carlo simulations on the anisotropic Heisenberg model [5] with Hamiltonian for ultrathin films on a square lattice of side \( L \) as follows:

\[
H = -J \sum_{ij} [(1 - \Delta)(S_i^x S_j^x + S_i^y S_j^y) + S_i^z S_j^z],
\]

where \( S_i = (S_i^x, S_i^y, S_i^z) \) is a unit vector in the direction of the classical magnetic moment at lattice site \( i \), the sum is extended over nearest-neighbor pairs on the cubic lattice, \( J > 0 \) being the exchange constant, and \( \Delta \) characterizes the amount of anisotropy. \( \Delta = 0 \) is the isotropic Heisenberg case, \( \Delta = 1 \) the Ising case. In order to study the critical properties of anisotropic Heisenberg magnets we thus made Monte Carlo calculations, studying cubic lattice with periodic and free boundary conditions are used for the in-plane and out-plane directions, respectively. The anisotropy constant for different sizes of the film was chosen from experimental studies of thin films of Ni(111)/W(110) [3]. The thickness dependence of critical temperatures presented in figure 1. The anisotropy constant was chosen proportional to the critical temperature for different film thicknesses. The resulting dependence was shown in figure 2.

The simulation are carried out for simple cubic films of size \( N_s = L \times L \times N \) where \( L \times L \) represents the number of sites (spins) in each layer of the film and \( N \) is the number of layers. We consider temperature interval \( T = 0.01 \div 5.01 \) \( J/k_B \) with step \( T_{\text{step}} = 0.02 \). The spin configurations of the films are updated using the Swendsen-Wang cluster algorithm [6]. The spin system is divided into clusters. The bonds between sites in cluster are created with probability \( 1 - \exp \left[ -2J(S_i r)(S_j r)/T \right] \) only if the condition \( (S_i r)(S_j r) > 0 \) is true, where \( r \) is random unit vector. After that each cluster is flipped with probability 1/2. We always used a completely ordered ferromagnet as a starting configuration. We measured the total magnetization

\[
m = \left\langle \frac{1}{N_s} \left[ \left( \sum_i^{N_s} S_i^x \right)^2 + \left( \sum_i^{N_s} S_i^y \right)^2 + \left( \sum_i^{N_s} S_i^z \right)^2 \right]^{1/2} \right\rangle,
\]

out-plane magnetization

\[
m_z = \left\langle \frac{1}{N_s} \sum_i^{N_s} S_i^z \right\rangle,
\]
the in-plane magnetization 

\[ m_\parallel = \left\langle \frac{1}{N_h} \left[ \left( \sum_i^N S_i^x \right)^2 + \left( \sum_i^N S_i^y \right)^2 \right]^{1/2} \right\rangle, \tag{4} \]

where angle brackets denote the statistical averaging. Temperature dependence of magnetization and susceptibility for different groups presented on figure 3. These curves were obtained by averaging over 3000 samples for each size of films. Temperature dependence of the susceptibility \( \chi_m \sim [\langle m^2 \rangle] - [\langle m \rangle]^2 \) was calculated for different lattice size in order to estimate the critical temperature \( T_c \). The position of the susceptibility maximum allowed to estimate range of values of the critical temperature. Temperature dependencies of the forth order Binder cumulant were calculated in order to clarify the critical temperatures. The scaling dependence of the cumulant makes it possible to determine the critical temperature \( T_c \) from the coordinate of the intersection points of the curves specifying the temperature dependence \( U_4(L, T) \) for different \( L \).

In early experiments on Fe/Cu(100) films [7, 8], it was observed a spin reorientation transition (SRT) from a region with perpendicular magnetization to one with in-plane magnetization. SRT transitions in films has actively been studied both theoretically [9, 10] and experimentally [11, 12], recently.

We measured orientational order parameter \( O_2 \) \[13, 14\]

\[ O_\alpha = \left\langle \left| \frac{n_{h\alpha} - n_{v\alpha}}{n_{h\alpha} + n_{v\alpha}} \right| \right\rangle, \tag{5} \]

where angle brackets denote the statistical averaging, \( \alpha \in \{x, y, z\} \), \( n_h \) and \( n_v \) are the number of horizontal and vertical pairs of nearest neighbor spins with antialigned perpendicular component,

\[ n_{h\alpha} = \sum_r \left\{ 1 - \text{sgn} \left[ S_{\alpha}(r_x, r_y), S_{\alpha}(r_x + 1, r_y) \right] \right\}, \]

\[ n_{v\alpha} = \sum_r \left\{ 1 - \text{sgn} \left[ S_{\alpha}(r_x, r_y), S_{\alpha}(r_x, r_y + 1) \right] \right\}. \tag{6} \]
Figure 4. Temperature dependence of $m(T)$, $m_z(T)$, $m_{||}(T)$ and $\chi_m(T)$, $\chi_O(T)$ for films with size $N = 15$ $L = 64$

It was observed a transition from an out-of-plane ordering at low temperatures to an in-plane configuration as described by the magnetization behaviour shown in figure 4. We show $m(T)$, $m_{||}(T)$, $m_z(T)$ and susceptibility $\chi_m(T)$, $\chi_O(T) \sim \langle O_z^2 \rangle - \langle O_z \rangle^2$ in the same figure for comparison. The susceptibility $\chi_m(T)$ curve presents two peaks see figure 4. The peak at low temperature is pronounced and is centered in the temperature in which occurs the rapid decrease of the out-of-plane magnetization $T = 2.71$. The susceptibility $\chi_O(T)$ has a maximum at the same temperature, that is why first peak is corresponds to spin reorientation transition from ferromagnetic phase to planar ferromagnetic. The second peak appears at $T = 3.91$ and it corresponds to the second order phase transition from a ferromagnetic state to a paramagnetic. In the temperature $T = 1.75$ the out-of-plane magnetization starts to decrease and becomes zero.

Table 1. Dependence of critical temperature $T_c(N)$ from film thickness

| N   | SRT | FM – PM | N   | SRT | FM – PM |
|-----|-----|---------|-----|-----|---------|
| 2   | -   | 1.03    | 13  | 1.87| 1.87    |
| 3   | -   | 1.15    | 14  | 2.45| 3.41    |
| 4   | -   | 1.25    | 15  | 2.71| 3.91    |
| 5   | -   | 1.31    | 16  | 2.89| 4.15    |
| 6   | -   | 1.35    | 17  | 2.87| 4.09    |
| 7   | -   | 1.39    | 21  | 1.17| 1.61    |
| 8   | 0   | 1.45    | 22  | 0.77| 1.55    |
| 9   | 0.65| 1.49    | 23  | 0   | 1.46    |
| 10  | 0.91| 1.57    | 26  | -   | 1.43    |
| 11  | 1.13| 1.61    | 30  | -   | 1.43    |
| 12  | 1.43| 1.63    | 31  | -   | 1.43    |
at \( T = 3.89 \). In this temperature interval the state with the biased magnetization is possible too.

The thickness dependence of critical temperature is presented in table 1 for film with thickness from \( N = 2 \) to \( N = 31 \). The FM-PM – is the second order phase transition from ferromagnetic phase to paramagnetic, SRT – spin reorientation transition from ferromagnetic phase to planar ferromagnetic. In the experimental [15] and theoretical [9] studies devoted to the study of single-layer magnetic materials, it is predicted that the orientation of the spin transition is a weak first-order transition.

We consider finite size scaling form for film geometry [16] to find how \( m \) and \( \chi \) scale with the size \( L \) and thickness \( N \) of the systems and use this to extract the effective critical exponents from our results. The basic finite-size scaling ansatz [17] rests on an assumption that only a single correlation length \( \xi \) is needed to describe the critical properties of thin films. Hence the empirical scaling forms for \( m \) and \( \chi \) at some fixed \( N \)'s can be written as

\[
\langle m(T, N) \rangle = L^{-\beta/\nu} \tilde{m}(L^{1/\nu} \tau, N) \tag{7}
\]

\[
\chi(T, N) = L^{\gamma/\nu} \tilde{\chi}(L^{1/\nu} \tau, N)
\]

where \( \gamma, \beta, \) and \( \nu \) are the effective critical exponents associated with \( \chi, m, \) and \( \xi \), respectively. For monolayer system with \( N = 1 \), the effective exponents are the critical exponents for the 2D system. The functions \( \tilde{\chi} \) and \( \tilde{m} \) are scaling functions for a given \( N \) and \( \tau = T/T_c - 1 \) is the reduced temperature. We consider the \( f(m) = L^{\beta/\nu} m \) as a scaling function of \( L^{1/\nu}(T_c - T)/T_c \).

We calculate these scaling functions for thin film with thickness from 2 to 5 layers with \( L = 32, 48, 64 \). These scaling functions for a range of \( L \) collapsed onto a single curve with the correct critical temperature and effective critical exponents. The results are shown in figure 6.

The effective exponent \( 1/\nu \) can be extracted from the derivative of the cumulant \( U_4 \) with respect to \( L \) at \( T_c \) owing to its variation with system size as \( L^{1/\nu} \). The scaling dependence of the cumulant allowed to calculate critical exponents \( \nu \).

\[
\frac{dU_4}{dT} \sim L^{1/\nu} \tag{8}
\]

We calculated the critical exponent for different temperatures above critical temperature and for different linear sizes of lattice (figure 5). The effective value of \( \nu \) can be obtained only in

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure5.png}
\caption{Temperature dependence critical exponents \( \nu(T, L) \)}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure6.png}
\caption{Dependence \( f(m) = L^{\beta/\nu} m \) as a function of \( L^{1/\nu}(T_c - T)/T_c \)}
\end{figure}
Table 2. Critical exponents $\nu$, $\beta$, $\gamma$ and effective dimension $d_{\text{eff}}$

| $N$ | $\nu$     | $\beta$     | $\gamma$    | $d_{\text{eff}}$ |
|-----|-----------|-------------|-------------|------------------|
| 2   | 1.021(18) | 0.126(8)    | 1.816(69)   | 2.046(59)        |
| 3   | 1.011(27) | 0.128(8)    | 1.770(94)   | 2.007(125)       |
| 4   | 1.018(14) | 0.126(9)    | 1.713(112)  | 2.007(101)       |
| 5   | 0.972(59) | 0.129(9)    | 1.609(150)  | 1.992(98)        |

thermodynamic limit. These values were calculated using linear approximation of linear size $L$ to infinity and linear approximation of temperature to critical [18]. The exponents for films with thickness from $N = 2$ to $N = 6$ are $\nu(N = 2) = 1.021(18)$, $\nu(N = 3) = 1.011(27)$, $\nu(N = 4) = 1.018(14)$, $\nu(N = 5) = 0.972(59)$, $\nu(N = 6) = 0.974(62)$. These values are very close to the 2D Ising values $\nu(2\text{D Ising}) = 1$.

Note that if equation (7) correctly encapsulates the nature of magnetic critical behaviour in films, we can extract the effective exponents $\beta/\nu$ and $\gamma/\nu$ from the slopes of the log-log plots of $m$ or $\chi$ against $L$ at $T_c$. Based on the hyperscaling relation $\gamma/\nu + 2\beta/\nu = d$, it is possible to consider the effective dimensionality $d_{\text{eff}}$. For $N/L \ll 1$, 2D-like behaviour is expected; i.e., $d_{\text{eff}}$ should stay close to 2.

We perform the calculation of the effective dimension $d_{\text{eff}} = \gamma/\nu + 2\beta/\nu$. Using expressions

$$m \sim L^{-\beta/\nu}, \quad \chi \sim L^{\gamma/\nu},$$

we have found that, in case $N = 2 - 5$, $d_{\text{eff}}$ has a value of 2 within error bars as shown in table 2 as expected. This confirms the 2D universality in thin films and ensures the possibility of using equation (7) to describe the critical behaviour of the thin films.

The exponents $\beta$ can be obtained from the temperature dependence of the magnetization near the critical point $m \sim (T_c - T)^{\beta}$ for different thickness of the magnetic film (table 3). The crossover from two-dimensional Ising model to three-dimensional Heisenberg model with increasing film thickness was found from this dependence.

Anisotropy constant $\Delta$ is close to 1 for thin films with thickness from 15 to 17. This case corresponds to Ising model. For these films we can select two temperature intervals corresponding to the 2D and 3D Ising critical behaviour. In first temperature interval near the critical point exponents $\beta(L = 48) = 0.123(7)$, $\beta(L = 64) = 0.124(5)$ for $N = 15$ (figure

Table 3. Critical exponents $\beta$ of magnetization for different film thickness

| $N$ | $\beta$     | $N$ | $\beta$     | $N$ | $\beta$     |
|-----|-------------|-----|-------------|-----|-------------|
| 2   | 0.126(9)    | 9   | 0.294(14)   | 18  | 0.338(2)    |
| 3   | 0.128(8)    | 10  | 0.310(13)   | 21  | 0.344(2)    |
| 4   | 0.126(9)    | 12  | 0.299(11)   | 22  | 0.352(6)    |
| 5   | 0.129(9)    | 13  | 0.314(5)    | 25  | 0.343(4)    |
| 6   | 0.170(11)   | 15  | 0.324(7)    | 26  | 0.358(1)    |
| 7   | 0.184(5)    | 16  | 0.329(8)    | 30  | 0.370(2)    |
| 8   | 0.286(11)   | 17  | 0.343(10)   | 31  | 0.368(2)    |
Figure 7. Dependence of magnetization from reduced temperature $\tau = (T_c - T)/T_c$

7(a)) and $\beta(L = 48) = 0.126(8)$, $\beta(L = 64) = 0.127(7)$ for $N = 17$ (figure 7(b)) are close to 2D Ising value of exponent $\beta = 0.125$. Critical exponents are $\beta(L = 48) = 0.316(2)$, $\beta(L = 64) = 0.319(3)$ for $N = 15$ and $\beta(L = 48) = 0.336(2)$, $\beta(L = 64) = 0.337(3)$ for $N = 17$ in the second temperature interval. This corresponds to 3D Ising critical behaviour $\beta = 0.325[19]$. This is because the correlation length is increased in all directions when approaching the critical temperature and demonstrate the 3D critical behaviour, reaching a boundary film continues to increase only in-plane direction of the film and shows critical behaviour of 2D system.

3. Conclusion
We have studied the magnetic behaviour of Heisenberg thin films in simple cubic structures with periodic and free boundary conditions for the in-plane and out-plane directions, respectively using extensive Monte Carlo simulations.

For thin films with $N = 9..22$ spin-reorientation transition was discovered. For these films the temperature of SRT have been determine from peak of susceptibility $\chi_0(T)$. In these system we consider the SRT from out-plane to in-plane ferromagnetic phase and second order phase transition from in-plane ferromagnetic to paramagnetic phase.

We have found the dimensional crossover of $m$ from 2D to 3D like with increasing film thickness. We have examined the critical regime of these systems in detail and extracted effective critical exponents based on a finite-size scaling method for films. For thin films with $N \leq 5$ the exponents are essentially the same as 2D and from this it can be implied that thin films fall into the 2D class. For thickness $N \geq 15$ the exponent $\beta$ move to 3D values as expected. The thickness dependence of critical exponents demonstrate the crossover from 2D Ising to 3D Ising and then to 3D Heisenberg behaviour.

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