Re-orientation transition in molecular thin films: Potts model with dipolar interaction

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Abstract
We study the low-temperature behavior and the phase transition of a thin film by Monte Carlo simulation. The thin film has a simple cubic lattice structure where each site is occupied by a Potts parameter which indicates the molecular orientation of the site. We take only three molecular orientations in this paper, which correspond to the three-state Potts model. The Hamiltonian of the system includes (i) the exchange interaction $J_{ij}$ between nearest-neighbor sites $i$ and $j$, (ii) the long-range dipolar interaction of amplitude $D$ truncated at a cutoff distance $r_c$, and (iii) a single-ion perpendicular anisotropy of amplitude $A$. We allow $J_{ij} = J_s$ between surface spins, and $J_{ij} = J$ otherwise. We show that the ground state depends on the ratio $D/A$ and $r_c$. For a single layer, for a given $A$, there is a critical value $D_c$ below (above) which the ground-state (GS) configuration of molecular axes is perpendicular (parallel) to the film surface. When the temperature $T$ is increased, a re-orientation transition occurs near $D_c$: the low-$T$ in-plane ordering undergoes a transition to the perpendicular ordering at a finite $T$, below the transition to the paramagnetic phase. The same phenomenon is observed in the case of a film with a thickness. Comparison with the Fe/Gd experiment is given. We show that the surface phase transition can occur below or above the bulk transition depending on the ratio $J_s/J$. Surface and bulk order parameters as well as other physical quantities are shown and discussed.

(Some figures may appear in colour only in the online journal)

1. Introduction

Surface physics has been intensively developed during the last 30 years. Among the main reasons for this rapid and successful development we can mention the interest in understanding the physics of low-dimensional systems and an immense potential of industrial applications of thin films [1–3]. In particular, theoretically it has been shown that systems of continuous spins (XY and Heisenberg) in two dimensions (2D) with short-range interaction cannot have long-range order at finite temperature [4]. In the case of thin films, it has been shown that low-lying localized spin waves can be found at the film surface [5] and effects of these localized modes on the surface magnetization at finite temperature ($T$) and on the critical temperature have been investigated by the Green’s function technique [6, 7]. Experimentally, objects of nanometric size such as ultrathin films and nanoparticles have also been intensively studied because of numerous and important applications in industry. An example is the so-called giant magnetoresistance used in

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data storage devices, magnetic sensors, etc [8–11]. Recently, much interest has been attracted towards practical problems such as spin transport, spin valves and spin-torque transfer, due to numerous applications in spintronics.

In this paper, we are interested in the phase transition of the Potts model [12] in thin films taking into account a dipolar interaction and a perpendicular anisotropy. The q-state Potts model is very popular in statistical physics and much is known for models with short-range ferromagnetic interactions in 2D and 3D [12]. The Potts model with an algebraically decaying long-range interaction has been investigated in 1D [13, 14]. Such a monotonous long-range interaction can induce an ordering at finite T in one-dimensional systems. We focus in the present work on the re-orientation transition which occurs in very thin films. Experimentally, it has been observed in a thin Fe film deposited on a Cu(100) substrate that the perpendicular spin configuration at low temperatures undergoes a transition to a planar spin configuration as the temperature (T) is increased [15–18]. This re-orientation is called ‘re-orientation of type I’ hereafter. Several mechanisms have been proposed to explain this re-orientation transition. For instance, Pappas et al [15] proposed that the transition is possible because of the temperature-dependence of the anisotropy. Other authors have suggested that it is due to a dipolar interaction between Heisenberg spins of the film [19–23]. In the case of 1.5 monolayer of Fe deposited on a Gd(0001) substrate, the order of the re-orientation is inverted: it has been observed [24] that the low-T planar ordering undergoes a transition to a perpendicular configuration when T is increased, in contrast to the Fe/Cu case discussed above. Let us call this case ‘re-orientation of type II’. It is obvious that the order of the re-orientation, type I or type II, depends on the material and the mechanism lying behind the transition. To account for the re-orientation transition of type II observed in Fe/Gd, we consider the dipolar interaction in this paper with the Potts model. Note that the dipolar interaction with the Heisenberg model gives rise to the re-orientation of type I as discussed above. The dipolar interaction is very special because it contains two competing terms, which yield complicated orderings that depend on the sample shape [25]. For example, the dipolar interaction favors an in-plane ordering in films and slabs with infinite lateral dimensions.

We consider a thin film made of a molecular crystal where molecular spins can point along the x, y or z axis. The interactions between molecular spins include a dipolar interaction truncated at a distance r_c and an exchange interaction between nearest neighbors (NN). We also take into account a single-ion perpendicular anisotropy which is known to exist in ultrathin films [1]. The method we employ is Monte Carlo (MC) simulations. Phase transition in systems of interacting particles is a major domain in statistical physics. Much is now understood with the analysis provided by the fundamental concepts of the renormalization group [26] and with the use of the field theory [27]. But these methods encountered some difficulties in dealing with frustrated spin systems [28, 29]. MC simulations are therefore very useful to complete theories and to interpret experiments. They serve as testing means for new theoretical developments. Over the years, the standard MC method [30] has been improved by the finite-size scaling theory [31] and by other high-performance techniques such as histogram techniques [32, 33], cluster updating algorithms [34–36] and the Wang–Landau flat-histogram method [37]. We have now at hand these efficient techniques to deal with complex systems. We can mention our recent investigations by MC techniques on multilayers [38], on frustrated surfaces [39, 40] or on surface criticality [41, 42].

In section 2, we describe our model and the method we employ. Results of MC simulations are shown and discussed in section 3 for several cases: 2D, homogeneous films and effects of surface interaction. Concluding remarks are given in section 4.

2. Model and method

We consider a thin film of simple cubic lattice. The film is infinite in the xy plane and has a thickness L_z in the z direction. The Hamiltonian is given by the following three-state Potts model:

$$\mathcal{H} = - \sum_{i,j} J_{ij} \delta(\sigma_i, \sigma_j)$$  \hspace{1cm} (1)

where $\sigma_i$ is a variable associated with the lattice site $i$, $\sigma_i$ is equal to 1, 2 or 3 if the spin at that site lies along the x, y or z axis, respectively. $J_{ij}$ is the exchange interaction between NN at i and j. We will assume that (i) $J_{ij} = J_0$ if i and j are on the same film surface and (ii) $J_{ij} = J$ otherwise.

The dipolar Hamiltonian is written as

$$\mathcal{H}_d = D \sum_{i,j} \left\{ \frac{S(\sigma_i) \cdot S(\sigma_j)}{r_{ij}^3} - \frac{3 [S(\sigma_i) \cdot r_{ij}] [S(\sigma_j) \cdot r_{ij}]}{r_{ij}^5} \right\}$$  \hspace{1cm} (2)

where $r_{ij}$ is the vector of modulus $r_{ij}$ connecting site i to site j. One has $r_{ij} = r_j - r_i$. In equation (2), D is a positive constant depending on the material, the sum $\sum_{i,j}$ is limited at pairs of spins within a cutoff distance $r_c$, and $S(\sigma_i)$ is given by the following three-component pseudo-vector representing the spin state:

$$S(\sigma_i) = (s_x(i), 0, 0) \quad \text{if } \sigma_i = 1$$

$$S(\sigma_i) = (0, s_y(i), 0) \quad \text{if } \sigma_i = 2$$

$$S(\sigma_i) = (0, 0, s_z(i)) \quad \text{if } \sigma_i = 3$$

where $s_\alpha (\alpha = x, y, z)$ is the $\alpha$ component with values ±1.

The perpendicular anisotropy is introduced by the following term

$$\mathcal{H}_a = -A \sum_i s_z(i)^2$$  \hspace{1cm} (6)

where A is a constant.

Note that the dipolar interaction as applied in our Potts model is not similar to that used in the vector spin model where $S(\sigma_i)$ is a true vector. In our model, each spin can
only choose to lie on one of three axes, pointing in positive or negative direction.

We use \( J = 1 \) as the unit of energy. The temperature \( T \) is expressed in the unit of \( J/k_B \), where \( k_B \) is the Boltzmann constant.

In the absence of \( D \), the GS configuration is perpendicular to the film surface due to the term \( \mathcal{H}_d \). In the absence of \( A \), the GS is an in-plane configuration due to \( D \). When both \( A \) and \( D \) are present, the GS depends on the ratio \( D/A \). An analytical determination of the GS is impossible due to the long-range interaction. We therefore determine the GS by the numerical steepest-descent method, which works very well in systems with uniformly distributed interactions. This method is very simple [39, 40]: (i) we generate a random initial spin configuration, (ii) we calculate the local field created at a given spin by its neighbors using equations (1) and (2), (iii) we change the spin axis to minimize its energy (i.e. we align the spin in its local field), (iv) we go to another spin and repeat until all spins are visited—we say we make one sweep—and (v) we make a large number of sweeps until a good convergence to the lowest energy is reached.

We shall use MC simulation to calculate properties of the system at finite \( T \). Periodic boundary conditions are used in the \( xy \) planes for sample sizes of \( L \times L \times L_z \), where \( L_z \) is the film thickness. Free symmetric surfaces are supposed for simplicity. The standard MC method [30] is used to get general features of the phase transition. Systematic finite-size scaling to obtain critical exponents is not the purpose of the present work. In general, we discard several million MC steps per spin to equilibrate the system before averaging physical quantities over several million MC steps. The averaged energy and the specific heat are defined by

\[
\langle U \rangle = \langle \mathcal{H} + \mathcal{H}_d + \mathcal{H}_a \rangle \\
C_V = \frac{\langle U^2 \rangle - \langle U \rangle^2}{k_B T^2}
\]

where \( \langle \ldots \rangle \) indicates the thermal average taken over several million microscopic states at \( T \).

We define the order parameter \( Q \) for the \( q \)-state Potts model by

\[
Q = \frac{q \max(Q_1, Q_2, Q_3) - 1}{q - 1}
\]

where \( Q_n \) is the spatial average defined by

\[
Q_n = \sum_i \delta(\sigma_i - n)/(L \times L \times L_z)
\]

\( n (n = 1, 2, 3) \) being the value attributed to denote the axis of the spin \( \sigma_i \) at site \( i \). The susceptibility is defined by

\[
\chi = \frac{\langle Q^2 \rangle - \langle Q \rangle^2}{k_B T}.
\]

We did not use the theory of finite-size scaling [31–33] because the calculation of critical exponents is not the purpose of the present work. However, in order to appreciate finite-size effects, we carried out simulations in the 2D case for sizes from \( L \times L = 24 \times 24 \) to 60 \( \times \) 60 and in the case of thin films from \( L \times L \times L_z = 12 \times 12 \times 4 \) to 48 \( \times \) 48 \( \times \) 6.

\[ \text{Fig. 1. Energy } E \text{ and specific heat } C_V \text{ versus } T \text{ for } D = 0.09 \text{ (black solid circles) and 0.11 (blue open circles),} \]

\( L = 60, A = 0.5, J = 1, r_c = \sqrt{6} \).

Results for the largest size are not significantly different from those of smaller sizes, except for the thickness. We will show therefore, in the following, results for lateral lattice size \( L = 60 \) for the 2D case, and \( L = 24 \) for thin films with thicknesses \( L_z = 4 \) and 6. In order to check the first-order nature of a weak first-order transition, the histogram technique is very efficient [32, 33]. But in our case, as will be seen below, the re-orientation is a very strong first-order transition. The discontinuity of energy and magnetization is clearly seen at the transition. We just use the histogram technique to check the 2D case for a demonstration.

3. Ground state and phase transition

3.1. Two dimensions

In the case of 2D, for a given \( A \), the steepest-descent method gives the ‘critical value’ \( D_c \) of \( D \) above (below) which the GS is the in-plane (perpendicular) configuration. \( D_c \) depends on \( r_c \). Let us take \( A = 0.5 \) and vary \( D \) and \( r_c \) in the following. The GS numerically obtained is shown in table 1 for several sets of \( (D, r_c) \). For instance, when \( r_c = \sqrt{6} \approx 2.449 \), we have \( D_c = 0.100 \).

We show in figure 1 the energy per site \( E \equiv \langle U \rangle/(L \times L \times L_z) \) and the specific heat, and in figure 2 the order parameter \( M \equiv \langle Q \rangle \) as well as the susceptibility \( \chi \), as functions of \( T \) in the case of \( r_c = \sqrt{6} \), for \( D = 0.09 \) and \( D = 0.11 \) on two sides of \( D_c = 0.100 \). We observe one transition of second order
Table 1. Ground states as functions of \((D, r_c)\), with A = 0.5, J = 1: I stands for the perpendicular configuration and II for the in-plane configuration (spins pointing along x or y axis).

| \(r_c\)  | 0.080 | 0.090 | 0.093 | 0.096 | 0.101 | 0.113 | 0.124 | 0.167 |
|---------|-------|-------|-------|-------|-------|-------|-------|-------|
| 1.0     | I I I I | I I I I | I I I I | I | I | I | I | II |
| \(\sqrt{3}\) | I I I I | I I I I | I | I | I | I | II | II |
| 2.0     | I I I I | I I I I | I | I | I | I | II | II |
| \(\sqrt{6}\) | I I I I | I I I I | II | II | II | II | II | II |
| 3.0     | I I I II | II | II | II | II | II | II | II |
| \(\sqrt{12}\) | I II | II | II | II | II | II | II | II |
| 4.0     | I II | II | II | II | II | II | II | II |

Figure 2. \(M\) and \(\chi\) versus \(T\) for \(D = 0.09\) (black solid circles) and 0.11 (blue open circles). Note that \(M\) for \(D = 0.09\) is the perpendicular magnetization while \(M\) for \(D = 0.11\) is the in-plane magnetization, \(L = 60, A = 0.5, J = 1, r_c = \sqrt{6}\).

It is interesting to examine the region very close to \(D_c\), namely close to the frontier of two different GSs. We have seen in the past that many interesting phenomena occur at the boundaries of different phases: we can mention the reentrance phenomenon in frustrated spin systems [28, 29] and the re-orientation transition in the Heisenberg film with a dipolar interaction similar to the present model [20]. We have carried out simulation for values close to \(D_c\). We find indeed a transition from the in-plane ordering to the perpendicular one when \(T\) increases in the region \(D \in [0.100, 0.104]\). We show an example at \(D = 0.101\) in figure 3, where we observe that in the low-\(T\) phase \((0 \leq T < 0.93)\) the spins align parallel to the \(x\) axis and in the intermediate-\(T\) phase \((0.93 < T < 1.05)\) the spins point along the \(z\) axis perpendicular to the film. The system becomes disordered at \(T > 1.05\). Note that, in
the disordered phase, each ‘state’ of the Potts spin (along one of the three axes) has one-third of the total number of spins. This explains why $M_x$ and $M_z$ tend to one-third at high $T$ in figure 3. The transition from the in-plane to the perpendicular configuration is of first order, as seen in figure 3 by the discontinuity of $M_x, M_z$, the energy and the magnetization at the transition point. The first-order character has been confirmed by the double-peaked energy histogram at the re-orientation transition temperature as shown in figure 4.

We show in figure 5 (top) the phase diagram in the space $(D, T)$ for $r_c = \sqrt 6$ where the line of re-orientation transition near $D_c$ is a line of first order. Let us discuss the effect of changing $r_c$. Increasing $r_c$ will increase the dipolar energy at each site. Therefore, a smaller value of $D$ suffices to ‘neutralize’ the effect of perpendicular anisotropy energy. The critical value of $D_c$ is thus reduced, as seen in the phase diagram established with $r_c = 4$ shown in figure 5 (bottom), where $D_c = 0.090$ compared to $D_c = 0.100$ when $r_c = \sqrt 6$ (top).

It is interesting to compare the present system using the three-state Potts model with the same system using the Heisenberg spins [20]. In that work, the re-orientation transition line is also of first order but it tilts on the left of $D_c$, namely the re-orientation transition of type I occurring in a small region below $D_c$, unlike the re-orientation of type II found above $D_c$ for the present Potts model. To explain the ‘left tilting’ of the Heisenberg case, we have used the following entropy argument: the Heisenberg in-plane configuration has a spin-wave entropy larger than that of the perpendicular configuration at finite $T$, so the re-orientation occurs in ‘favor’ of the in-plane configuration; it goes from perpendicular to in-plane ordering with increasing $T$. Obviously, this argument for the Heisenberg case does not apply to the Potts model because we have here the inverse re-orientation transition. We think that, due to the discrete nature of the Potts spins, spin waves cannot be excited, so there is no spin-wave entropy as in the Heisenberg case. The perpendicular anisotropy $A$ is thus dominant at finite $T$ for $D$ slightly larger than $D_c$.

Experimentally, the ‘right tilting’ of the re-orientation line from $D_c$ in figure 5 has been observed in Fe/Gd [24].

So we emphasize that re-orientation depends on the system; it can be of type I or type II. Our simple model shows this possibility. Of course, to compare quantitatively our results with experimental data of Fe/Gd we need to take into account details of the real system such as Fe and Gd lattice structures and magnetic interactions. This is left for future investigations.

There is another important point which is worth mentioning: the scenario of the re-orientation transition. Arnold et al [24] have suggested a two-step transition with an intermediate phase. Theoretically, this scenario is possible: we have found in several exactly solved models [28, 29] that the transition between two ordered phases can be assisted by a very small disordered phase between them, which is called the ‘reentrant phase’. In this extremely small region, one can have a so-called ‘disorder line’ with dimension reduction to help the system go from one symmetry to another one. Numerically and experimentally the reentrance region is hardly detected due to its smallness. If the size of the reentrance region is large enough, we can have two second-order lines departing from $D_c$ (see discussion on several exactly solved models in [28]). If the size is very small, these two lines look like a single line within numerical and/or experimental resolution, then the re-orientation on the ‘single’ line has a first-order nature, which is necessary to allow a transition between two different symmetries [27]. We think that this latter hypothesis
Figure 6. Film with thickness $L_z = 4$ ($L = 24$). Top, $M_x$ (black solid circles) and $M_z$ (blue open circles); middle, total $M$; bottom, $E$ versus $T$ for $D = 0.31$ in the re-orientation transition region. See text for comments.

3.2. Thin films

The case of thin films with a thickness $L_z$, where $L_z$ goes from a few to a dozen atomic layers, has a very similar re-orientation transition to that shown above for the 2D case.

Let us show results for $J_s = J$ in figures 6–8 below. The effect of surface exchange integral $J_s$ will be shown in section 3.3.

Let us show in table 2 the GS obtained by the steepest-descent method with $A = 0.5$ and $J = 1$ as before, for two thicknesses $L_z = 4$ and $L_z = 6$. Changing the film thickness results in changing the dipolar energy at each lattice site. Therefore, the critical value $D_c$ will change accordingly.

We note the periodic-layered structures at large $D$ and $r_c$ for both cases. In the case $L_z = 4$, for $r_c = \sqrt{6}$ the critical value $D_c$ above which the GS changes from the perpendicular to the in-plane configuration is $D_c = 0.305$.

As in the 2D case, we expect interesting behaviors near the critical value $D_c$. For example, when $L_z = 4$, $r_c = \sqrt{6}$ and $A = 0.5$, we find indeed a re-orientation transition, which is shown in figure 6. The upper curves show clearly a first-order transition from in-plane $x$ ordering to perpendicular ordering at $T \simeq 1.41$. The total magnetization (middle curve) and the energy (bottom curve) show a discontinuity at that temperature. The whole phase diagram is shown in figure 7.

![Figure 6](image6.png)

![Figure 7](image7.png)
Note that the line separating the uniform in-plane phase (II) and the periodic single-layered phase (I) is vertical.

Here again, the line separating the perpendicular configuration (I) and the in-plane one (II) is a first-order line. However, we would like to emphasize that the same remark as given at the end of section 3.1 is applied to the thin film case: we cannot exclude the possibility of a very small reentrance phase between the two phases I and II.

To close this section, let us show in figure 8 the transition at values of $D$ far from the critical values of $D$. There is only one transition from the ordered phase to the paramagnetic phase. As seen, the transition from the in-plane ordering (phases II and I) to the paramagnetic phase is sharper than that from the perpendicular one (phase I), as in the 2D case.

### 3.3. Effect of surface exchange interaction

We have calculated the effect of $J_s$ by taking its values far from the bulk value ($J = 1$) for several values of $D$. In general, when $J_s$ is smaller than $J$ the surface spins become disordered at a temperature $T$ below the temperature where the interior layers become disordered. This case corresponds to the soft surface (or magnetically ‘dead’ surface layer) [7]. On the other hand, when $J_s > J$, we have the inverse situation: the interior spins become disordered at a temperature lower that of the surface disordering. We have here the case of a magnetically hard surface. We show in figure 9 an example of a hard surface in the case where $J_s = 3$ for $D = 0.6$ with $L_c = 4$. The same feature is observed for $D = 0.4$. Note that the surface and bulk transitions are seen by the respective peaks in the specific heat and the susceptibility. In the re-orientation region, the situation is very complicated as expected because the surface transition occurs in the re-orientation zone.

Let us discuss experimental data of Fe/Gd. It has been found that the overall transition to the paramagnetic phase in the experiment on Fe/Gd, driven by the Gd substrate [24], is a first-order transition which occurs at room temperature. To simulate the Fe/Gd system, one can proceed by changing $J_s$ and the nature of the surface spins to represent Fe atoms, one then can perform simulations in order to compare quantitatively with the experiment. This is left for future studies.

### 3.4. Discussion

Note that for a given $D$, the effect of the cutoff distance $r_c$ is to move the critical value of $D_c$ as seen in tables 1 and 2. At $r_c = \sqrt{10} \approx 3.16$ one has 146 neighbors for each interior spin (not near the surface). This huge number makes MC simulations CPU-time consuming. We therefore performed simulations at finite $T$ only with two values of $r_c$ in the 2D case. As seen in figure 5, the change of $r_c$ does not alter our conclusion on the re-orientation transition. We think that the cutoff is more than a technical necessity: it also involves physical reality. We have in mind the observation that in most experimental systems interaction between faraway neighbors can be neglected. The concept that the interaction range between particles can go to infinity is a theoretical concept. Models in statistical physics limited to interaction between NN are known to interpret experiments with success [1, 27]. Rarely do we have to go farther than third NN. For example, in our recent paper on the spin resistivity in semiconducting MnTe, we took interactions up to third neighbors to get an excellent agreement with experiments [43]. Therefore, we wanted to test in the present paper how physical results depend on $r_c$ in the dipolar interaction. If we know for sure that in a thin film the interaction is dipolar and that a double-layered structure for example is observed, from what is found above we can suggest the interaction range between spins in the system. Finally, we note that if we change $A$, the value of $D_c$ will change. The choice of $A = 0.5$, which is one-half of $J$, is a reasonable choice to make the re-orientation happen.
A smaller $A$ will induce a smaller $D_c$, but again the physics found above will not change.

4. Concluding remarks

We have shown in this paper MC results on the phase transition in thin magnetic films using the Potts model including a short-range exchange interaction $J$ and a long-range dipolar interaction of strength $D$, truncated at a distance $r_c$. We have also included a perpendicular anisotropy $A$ which is known to exist in very thin films.

Among the striking results, let us mention the re-orientation transition which occurs in 2D and in thin films at a finite temperature below the overall disordering. The planar spin configuration at low temperatures undergoes a phase transition to the perpendicular configuration as temperature increases. This re-orientation is a very strong first-order transition as seen by the discontinuity of the energy and the magnetization. We emphasize that the re-orientation is possible only because we have two competing interactions: the perpendicular anisotropy and the dipolar interaction. Experiments performed on 1.5 monolayers of Fe deposited on Gd(0001) \cite{24} show this re-orientation, in agreement with our results using a Potts model. In the light of what is known from exact results in similar situations \cite{28}, we cannot exclude the possibility that the first-order re-orientation line in the phase diagram is in fact a merging of two second-order lines separated by an extremely reentrant phase that we cannot distinguish with our numerical resolution.

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