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Effect of dipolar interaction in molecular crystals

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Abstract

In this paper we investigate the ground state and the nature of the transition from an orientational ordered phase at low temperature to the disordered state at high temperature in a molecular crystal. Our model is a Potts model which takes into account the exchange interaction J between nearest-neighbor molecules and a dipolar interaction between molecular axes in three dimensions. The dipolar interaction is characterized by two parameters: its amplitude D and the cutoff distance r_c. If the molecular axis at a lattice site has three orientations, say the x, y or z axes, then when $D = 0$, the system is equivalent to the 3-state Potts model: the transition to the disordered phase is known to be of first order. When $D \neq 0$, the ground-state configuration is shown to be composed of two independent interpenetrating layered subsystems which form a sandwich whose periodicity depends on D and r_c. We show by extensive Monte Carlo simulation with a histogram method that the phase transition remains of first order at relatively large values of r_c.

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

1. Introduction

The nature of the transition from one phase to another is one of the most important problems in statistical physics and in various areas of materials science not limited to physics. Since the introduction of the renormalization group [1, 2] with new concepts based on the system symmetry and scale, the understanding of the nature of the phase transition in many systems becomes clear. However, there are complicated systems where that method encounters much difficulty in application. One can mention frustrated systems where competing interactions cause highly degenerate ground states with instabilities [3].

In this paper, we are interested in the phase transition in molecular crystals, which has been a subject of intensive investigations for 40 years. This spectacular development was due to numerous applications of liquid crystals in daily life [4, 5]. Liquid crystals are somewhere between solid and liquid states where molecules have some spatial orientations which, under some conditions, can order themselves into some structures such as nematic and smectic phases. Liquid crystal display uses properties of these ordered phases [6, 7]. In spite of the large number of applications using experimental findings, theoretical understanding in many points is still desirable. One of the most important aspects of the problem is the origin of layered structures observed in smectic ordering: in smectic phases the molecules are ordered layer by layer with periodicity. Experiments have discovered, for example, smectic phases of 2-layer, 3-layer, 4-layer, or 6-layer periodicity [8–13]. Different theories have been suggested to interpret these observations [14–21]. One of the unanswered questions is what is the origin of the long-period layered structure in observed smectic phases? Hamaneh *et al* [22] suggested that the effective long-range interaction is due to bend fluctuations of the smectic layers which may stabilize commensurate structures, in particular the six-layer phase. This suggestion is too qualitative to allow a clear understanding of the long-period layered structure in smectic phases. In our opinion, a long-range correlation does not imply necessarily a particular long-period configuration [2].

To look for a physical origin of long-period structures, we concentrate in the present paper on the effect of a dipolar

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interaction in a Potts model, in addition to an exchange interaction between nearest neighbors (NN). We suppose that each molecule has a molecular axis which can lie on one of the three principal directions. An example of such a molecule is the ammoniac molecule NH_3. Without the dipolar interaction, the interaction between neighboring molecules gives rise to an orientational order of molecular axes at low temperature. In this situation, the system can be described by a 3-state Potts model in three dimensions (3D). The Potts model is very important in statistical physics. It is in fact a class of models, each of which is defined by the number of states q that an individual particle or molecule can have. The interaction between two neighboring molecules is negative if they are in the same state, it is zero otherwise. Exact solutions are found for many Potts models in two dimensions (2D) [23]. In 2D, the phase transition is of second order for $q \leq 4$ and of first order for higher q. In 3D, a number of points are well understood. For example, the phase transition is of first order for $q > 2$. The molecular crystal described above undergoes therefore a first-order transition in the absence of a dipolar interaction.

The purpose of this paper is to investigate the effect of the dipolar interaction on the ground-state (GS) structure and on the nature of the phase transition. The dipolar interaction is a long-range interaction which yields different GS structures depending on the shape of the sample, as will be discussed in section 2. To carry out our purpose, we use a steepest-descent method for the GS determination and the Monte Carlo (MC) simulation combined with the histogram technique to distinguish first- and second-order characters.

In section 2, we show our model and analyze the GS. Results of MC simulations are shown and discussed in section 3. Concluding remarks are given in section 4.

2. Model and ground-state analysis

We consider a simple cubic lattice where each site is occupied by an axial molecule. The molecular axis can be in the x, y or z direction. Let us denote the orientation of the molecule at the lattice site i by a unit segment, not a vector, which can lie in the x, y or z direction. We attribute the Potts variable $\sigma = 1, 2$ or 3 when it lies in the x, y or z axes, respectively, in the calculation. By the very nature of the model shown below, the results do not depend on these numbers as in the standard q-state Potts model.

We suppose that the interaction energy between two nearest molecules is $-J$ ($J > 0$) if their axes are parallel, zero otherwise. With this hypothesis, the Hamiltonian is given by the following 3-state Potts model:

$$\mathcal{H} = -J \sum_{(i,j)} \delta(\sigma_i, \sigma_j) \tag{1}$$

where σ_i is the 3-state Potts variable at the lattice site i and $\sum_{(i,j)}$ is made over the nearest sites σ_i and σ_j. The Potts model describes the strongest limit of possible repulsion between molecules. This is due to the Kronecker symbol in the Hamiltonian: minimum energy if molecular moments are similar, zero energy otherwise. In reality, if molecular moments are modeled by continuous spins such as the Heisenberg model, the repulsion is less strong because the interaction energy between Heisenberg spins varies continuously. The Potts model used here is thus intended to ‘amplify’ the repulsive effect.

The dipolar interaction between Potts variables is written as

$$\mathcal{H}_{\rm d} = D \sum_{(i,j)} \left\{ \frac{\mathbf{S}(\sigma_i) \cdot \mathbf{S}(\sigma_j)}{r_{ij}^3} - 3\frac{[\mathbf{S}(\sigma_i) \cdot \mathbf{u}_{ij}][\mathbf{S}(\sigma_j) \cdot \mathbf{u}_{ij}]}{r_{ij}^3} \right\} \delta(\sigma_i, \sigma_j) \tag{2}$$

where $\mathbf{u}_{ij}$ is the vector of unit length connecting sites i and j, D a positive constant depending on the material, $\mathbf{S}(\sigma_i)$ is defined as the unit vector lying on the axis corresponding to the value of σ_i. The sum is limited at some cutoff distance $r_{\rm c}$. Without the Kronecker condition and in the case of classical XY or Heisenberg spins, the dipolar interaction gives rise to spin configurations which depend on the sample shape. For example, in 2D or in rectangular slabs spins lie in the plane to minimize the system energy.

The origin of the frustration comes from the competition between the first term and the second term in the brackets of the dipolar interaction given by equation (2). The first term is positive or zero, while the second term can be positive, zero or negative, depending on the orientations of the molecular dipoles $\mathbf{S}(\sigma_i)$ and $\mathbf{S}(\sigma_j)$ with respect to the vector $\mathbf{u}_{ij}$ which connects these dipoles. For examples, if $\mathbf{u}_{ij}$ is on the x axis, the second term is: (i) equal to zero if the dipoles $\mathbf{S}(\sigma_i)$ and $\mathbf{S}(\sigma_j)$ are on y or z axes since they are perpendicular to $\mathbf{u}_{ij}$, the scalar products are then zero, (ii) equal to $-3/r_{ij}^3$ if $\mathbf{S}(\sigma_i)$ and $\mathbf{S}(\sigma_j)$ lie on the x axis. Note that the calculation of the minimum of the dipolar interaction is very complicated, and cannot be done by hand because of the very large number of neighbors. However, it can be done numerically by the steepest-descent method used in this paper.

Let us first discuss the GS in the Potts model introduced above. When $D = 0$ the GS is uniform with one orientation value, namely it is 3-fold degenerate. However, for a nonzero D, the ground state changes with varying $r_{\rm c}$. To determine the GS, we use the steepest-descent method, which works very well in systems with uniformly distributed interactions. This method is very simple [24, 25] (i) we generate an initial configuration at random, (ii) we calculate the local field created at a site by its neighbors using (1) and (2), (iii) we take the Potts variable to minimize its energy (i.e. align the ‘Potts spin’ in its local field), (iv) we go to another site and repeat until all sites are visited: we say we make one sweep, (v) we do a large number of sweeps per site until a good convergence is reached.

We show some examples in figure 1. For very small D, the GS is uniform for any $r_{\rm c}$, as shown in figure 1(a). For increasing D, the GS has layered structures with period $p = 1$ (alternate single layers), $p = 2$ (double layers), $p = 3$ (triple layers), ... depending on $r_{\rm c}$ and D. Note that this work was motivated by the observation that in most experimental systems very long-ranged interaction can be neglected. The concept that the interaction range between particles can go to

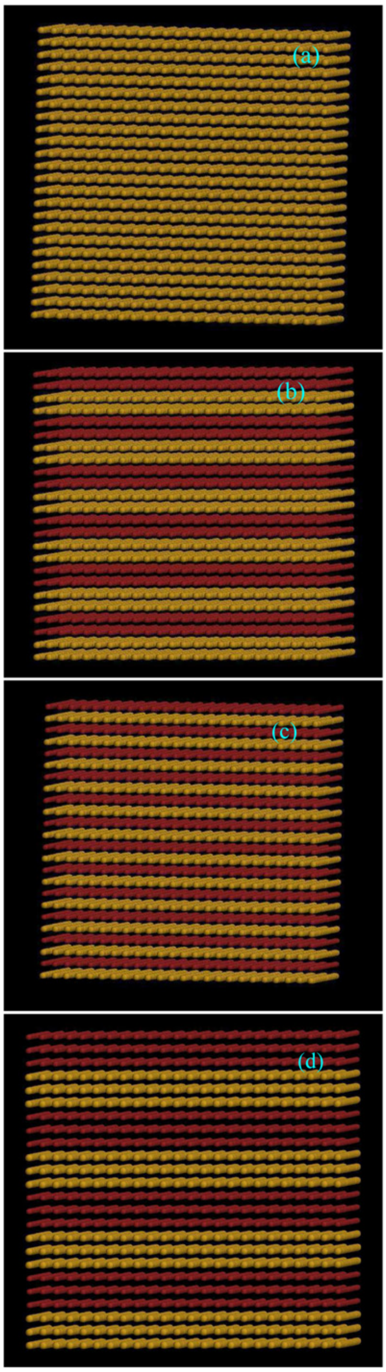

Figure 1. Ground state in the case where (a) $D/J = 0.4$, $r_c = 2.3$: uniform configuration; (b) $D/J = 1$, $r_c = 2.3$: double-layer structure; (c) $D/J = 2$, $r_c = 2.3$: single-layer structure and (d) $D/J = 0.4$, $r_c = \sqrt{10} \simeq 3.16$: triple-layer structure. See text for comments.

infinity is a theoretical concept. Models in statistical physics limited to interaction between nearest neighbors are known to interpret with experiments successfully [2]. Therefore, we wish to test how physical results depend on r_c in the dipolar interaction. If we know for sure that in a system the interaction is dipolar and that a triple-layer structure is observed, we can suggest the interaction range beyond which interaction can be neglected.

We note that in figure 1 there are only two kinds of molecular orientations represented by two colors (on line) in spite of the fact that we have three possible orientations. Let us discuss the single-layer structure shown in figure 1(c). It is very important to note that each layer has no coupling with two nearest layers whose molecules lie on another axis. However, it is coupled to two next-nearest layers of the same color, namely the same molecular axis, lying within r_c. In other words, the GS is composed of two interpenetrating 'independent' subsystems. The same is true for other layered structures: in a double-layer structure molecules of the same orientation (same color on line) interact with each other but they are separated by a double layer of molecules of another orientation. To our knowledge, this kind of GS has never been found before. It may have important applications at macroscopic levels.

Note also that if the molecules of the single-layer structure are successively in x-oriented and y-oriented planes, then the stacking direction of these planes is the z direction. The molecules can choose the x and z orientations or the y and z directions. In those cases the stacking directions are respectively the y and x directions. These three possibilities are equivalent if the sample is cubic. In rectangular shapes, the stacking direction is along the smallest thickness.

We emphasize that the steepest-descent method corresponds in most cases we have studied to the minimization of the energy of the system given by the Hamiltonian [24, 25]. Until now, we have encountered metastable states while using this method only in some frustrated and simultaneously disordered systems such as spin glasses. To check if one has a real GS or a metastable state, one can perform MC simulations at very low T to see if the energy converges to the minimum energy obtained by the steepest-descent method. The lowest energy for a given set (D, r_c) shown in figure 2 corresponds indeed to the extrapolation of the MC energy (not shown) from very low T ($T = 0.05$ in our unit) to $T = 0$.

In order to understand how such GS configurations found by the steepest-descent method depend on D and r_c, we have considered the structures 121212... (single-layer structure), 11221122... (2-layer structure), 111222111222... (3-layer structure) and carried out the calculations of the energy of a molecule σ_i interacting with its neighbors σ_j. The case where the configuration is uniform, i.e. there is only one kind of molecular orientation, say axis x, the dipolar energy of σ_i is

$$E_i = D \sum_j \left[\frac{1}{r_{ij}^3} - 3\frac{(u_{ij}^x)^2}{r_{ij}^3} \right]. \tag{3}$$

Note that with the use of the Potts model for the dipolar term in equation (2), the energy depends only on the axis, not on its direction as seen by the square term $(u_{ij}^x)^2$. In this sense, the model is suitable to describe axial, but non-directed, interacting molecules.

If we transform the sum in integral, the sum in the first term gives $4\pi \ln r_c$ (integrating from 1 to r_c), while the second term gives $-4\pi \ln r_c$, which cancels the first term. This is valid for r_c larger than 1. Thus the dipolar energy $E_d = D[4\pi \ln r_c - 4\pi \ln r_c]$ is zero for the uniform configuration in 3D space in

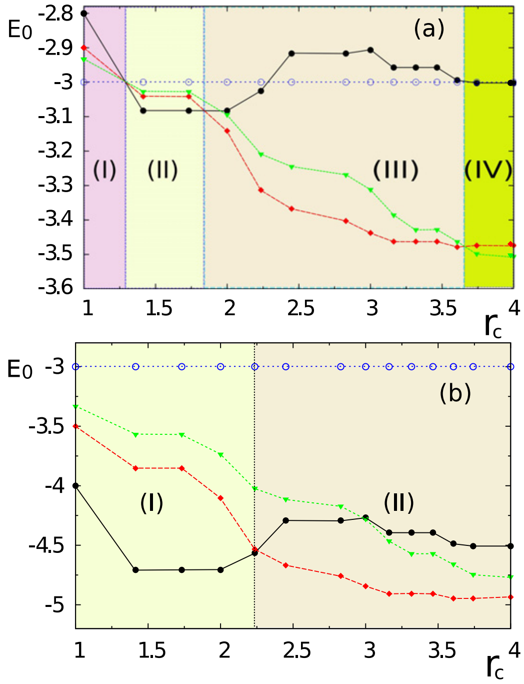

Figure 2. Ground-state energy versus r_c in the case where (a) $D/J = 0.8$, (b) $D/J = 2$. Blue, black, red and green lines represent the GS energy of the uniform, single-layer, double-layer and triple-layer structures, respectively. Zones I, II, II, IV indicate these respective different GS. See text for comments.

the framework of our model. The energy of the system comes from the short-range exchange term, equation (1).

The energy per site of other layered structures with periodicity $p = 2$ and 3 is numerically calculated and shown in figure 2 for $D/J = 0.8$ and 2. In each figure, the GS is the structure which corresponds to the lowest energy. One sees in figure 2(a) the following GS configurations with varying r_c:

- uniform GS: for $1 \leq r_c \leq 1.3$ (zone I);
- single-layer structure: for $1.3 \leq r_c \leq 1.8$ (zone II);
- double-layer structure: for $1.8 \leq r_c \leq 3.65$ (zone III);
- triple-layer structure: for $r_c \geq 3.65$ (zone IV).

For $D/J = 2$ there are only two possible GS with varying r_c as shown in figure 2(b).

We summarize in figure 3 the different GS in the space (r_c, D) with $J = 1$. We note that for a given D, for example $D = 0.8$, the GS configuration starts with uniform configuration then with period $1, 2, 3, \ldots$ for increasing r_c. At large D, uniform configuration is not possible at any r_c. Long-period configurations are on the other hand favored at small D and large r_c.

3. Phase transition: results

In this section, we study the phase transition at a finite temperature T where the above ordered GS structures become disordered.

We consider a sample size of $N \times N \times N_z$, where N and N_z vary from 24 to 48 but N_z can be different from N in order to

r_c \ D	0.2	0.4	0.6	0.8	1.2	1.5	2.0	5.0
1.0 (6)	0	0	0	0	1	1	1	1
$\sqrt{2}$ (18)	0	0	0	1	1	1	1	1
$\sqrt{3}$ (26)	0	0	0	1	1	1	1	1
2.0 (32)	0	0	0	2	1	1	1	1
$\sqrt{5}$ (56)	0	0	2	2	2	2	1	1
$\sqrt{6}$ (80)	0	0	2	2	2	2	2	2
$\sqrt{8}$ (92)	0	0	2	2	2	2	2	2
3.0 (122)	0	0	2	2	2	2	2	2
$\sqrt{10}$ (146)	0	3	2	2	2	2	2	2
$\sqrt{11}$ (170)	0	3	3	2	2	2	2	2
$\sqrt{12}$ (178)	0	3	3	2	2	2	2	2
$\sqrt{13}$ (202)	0	3	3	2	2	2	2	2
$\sqrt{14}$ (250)	0	3	3	3	2	2	2	2
4.0 (256)	0	3	3	3	2	2	2	2

Figure 3. Ground state in space (r_c, D): the numbers 0, 1, 2 and 3 denote the uniform, single-layer, double-layer and triple-layer structures, respectively. The first column displays the values of r_c with the number of neighbors indicated in the parentheses. $J = 1$ has been used.

detect the shape dependence of the ground state. The 3-state Potts model with NN exchange interaction $J = 1$ is used to describe the three molecular orientations. For the dipolar term, a cutoff distance r_c is taken up to $\sqrt{10} \simeq 3.16$ lattice distance. At this value, each molecule has a dipolar interaction with 146 neighbors. Periodic boundary conditions in all directions are employed.

We have used the standard MC method [26] with the system size from 24^3 to 48^3. The equilibrating time N_1 is about 10^6 MC steps per site, and the averaging time N_2 is between 10^6 and 10^7 MC steps per site. The averages of the internal energy $\langle U\rangle$ and the specific heat C_V are defined by

$$\langle U\rangle = \langle \mathcal{H} + \mathcal{H}_d\rangle \tag{4}$$

$$C_V = \frac{\langle U^2\rangle - \langle U\rangle^2}{k_B T^2} \tag{5}$$

where $\langle\cdots\rangle$ indicates the thermal average taken over N_2 microscopic states at T.

We define the Potts order parameter Q by

$$Q = [q\max(Q_1, Q_2, \ldots, Q_q) - 1]/(q-1) \tag{6}$$

where Q_n is the spatial average defined by

$$Q_n = \sum_j \delta(\sigma_j - n)/(N \times N \times N_z) \tag{7}$$

n ($n = 1, \ldots, q$) being the value of the Potts variable at the site j. For $q = 3$, one has $n = 1, 2, 3$ representing respectively the molecular axis in the x, y and z directions. The susceptibility is defined by

$$\chi = \frac{\langle Q^2\rangle - \langle Q\rangle^2}{k_B T}. \tag{8}$$

In the following, we shall express the energy in units of J, and the temperature in units of J/k_B. In practice, we will set $J = 1$ and $k_B = 1$ in the presentation.

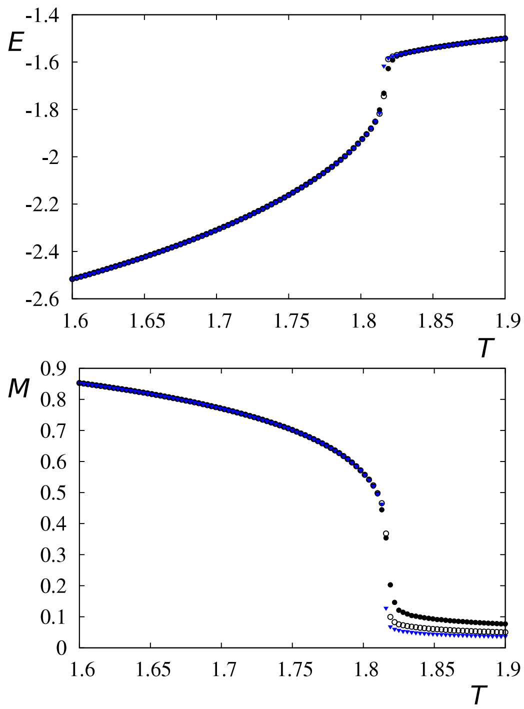

Figure 4. Internal energy per spin E (upper) and order parameter $M = \langle Q \rangle$ (lower) versus temperature T (in units of J/k_B) in the case without dipolar interaction, namely $D = 0$, for several sizes $N_z = N = 24$ (black circles), 36 (void circles), 48 (blue triangles, color on line), $J = 1$. Note that $r_c = 1$ for this pure Potts model.

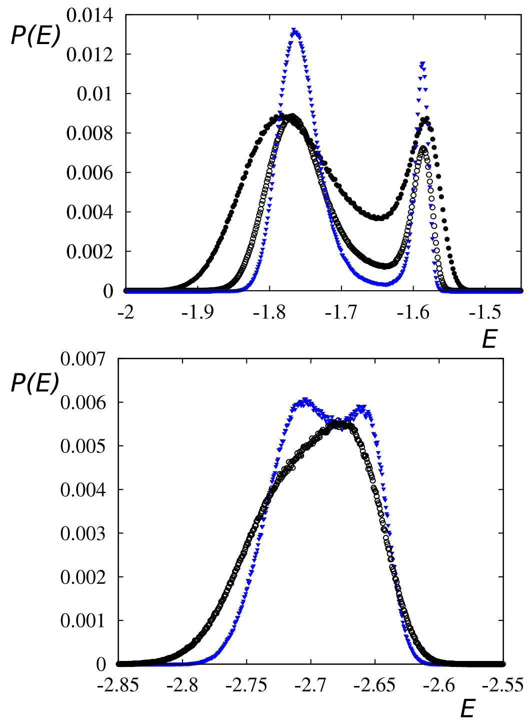

Figure 5. Energy histogram at the transition temperature without ($D = 0$, upper) and with ($D = 2$, lower) dipolar interaction, for several sizes $N_z = N = 24$ (black circles), 36 (void circles), 48 (blue triangles, color on line), $r_c = \sqrt{10}$, $J = 1$. For $D = 2$, only the two sizes 36^3 and 48^3 are shown because the smaller size 24^3 does not show a double peak due to its very weak first-order transition. See text for comments.

In MC simulations, we work at finite sizes, so for each size we have to determine the 'pseudo' transition which corresponds in general to the maximum of the specific heat or of the susceptibility. The maxima of these quantities need not to be at the same temperature. Only at infinite size should they coincide. The theory of finite-size scaling [27–29] permits one to deduce the properties of a system at its thermodynamic limit. We have used in this work a size large enough to be close to the extrapolated bulk transition temperature.

We show first in figure 4 the energy per site $E \equiv \langle U \rangle/(N \times N \times N_z)$ and the order parameter $M = \langle Q \rangle$ versus T, for several lattice sizes in the absence of the dipolar interaction, i.e. $D = 0$. As said earlier this case corresponds to the 3-state Potts model. We find a very sharp transition. Note that the finite-size effect is clearly seen in the transition region. For each size, there is a transition temperature T_0 which corresponds to the change of the curvature of E in the case of continuous transition, or to the discontinuity of E in the case of strong first order. In order to check the first-order nature of this transition, we used the histogram technique, which is very efficient in detecting weak first-order transitions and in calculating critical exponents of second-order transitions [28, 29]. The main idea of this technique is to make an energy histogram at a temperature T_0 as close as possible to the transition temperature. Often, one has to try at several temperatures in the transition region. Using this histogram in the formulas of statistical physics for canonical distribution, one obtains energy histograms in a range of temperatures around T_0. In second-order transitions, these histograms are Gaussian. They allow us to calculate averages of physical quantities as well as critical exponents using the finite-size scaling. In first-order transitions, the energy histogram shows a double-peak structure at large enough lattice sizes. Using this method, we have calculated the histogram shown in figure 5 for several lattice sizes. With increasing size, the two peaks are well separated, with the dip going down to zero, indicating a tendency toward an energy discontinuity at infinite size. The distance between the two peaks is the latent heat ΔE. Note that for $D = 2$ in figure 5, we have shown only results of the two sizes 36^3 and 48^3, because the smaller size 24^3 does not show a double peak due to its very weak first-order transition. The reason is for large values of D, the first order is weakened, and this can be seen only from a large lattice size, for instance from 48^3 up.

We take into account now the dipolar interaction. In order to see its progressive effect, we perform simulations with r_c varying between $r_c = \sqrt{6}$ and $\sqrt{10}$ and follow the change of the characteristics of the phase transition.

We show in figure 6 the energy and the order parameter versus T for $D = 2$, $r_c = \sqrt{10}$. The transition is still very sharp but the strong first-order character diminishes.

We show in figure 7 the energy histogram for several values of r_c with $D = 2$. As seen the latent heat becomes small at $r_c = \sqrt{10}$.

We observe that the latent heat ΔE diminishes with increasing r_c. However, we cannot conclude that the

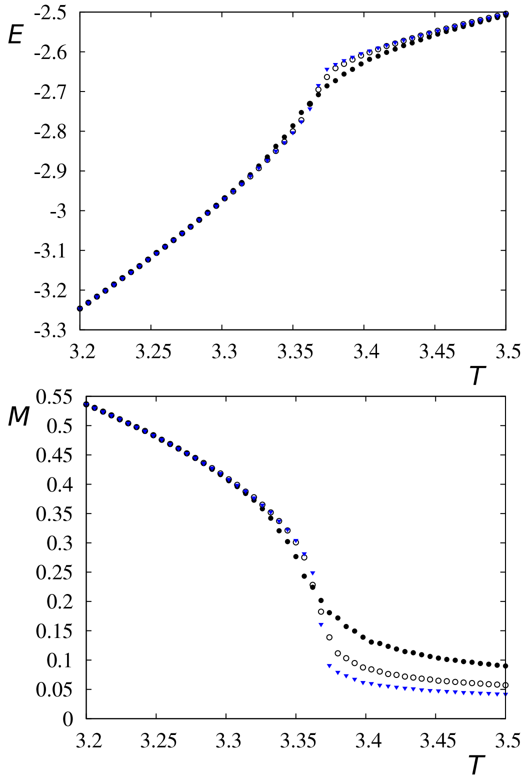

Figure 6. Internal energy per spin E and order parameter $M = \langle Q \rangle$ versus T (in unit of J/k_B) for $D = 2$, $r_c = \sqrt{10}$ with several lattice sizes $N_z = N = 24$ (black circles), 36 (void circles), 48 (blue triangles, color on line), and $J = 1$.

first-order disappears at large r_c. To check that point we need to go to larger r_c, which will take a huge CPU time because of the increasing number of neighbors (we recall that for $r_c = \sqrt{10}$, we have 146 neighbors for each molecules). We observe from figure 7 that the latent heat does not change significantly with r_c up to $r_c \simeq 2.85$, meaning that the first-order transition is dominated by the short-range 3-state Potts interaction. The latent heat decreases rather strongly afterward, but we do not know if it tends to zero or not.

As said, even numerically we cannot prove that the first order will disappear for large D and large r_c; we conjecture that the first order should disappear in this limit. The physical argument for this conjecture is based on the observation of two limiting cases (i) when $J = 1$ and $D = 0$ (no dipolar interaction), the transition is a strong first-order 3-state Potts model, (ii) when $J = 0$, and D is not zero with infinite r_c, the transition is of dipolar origin. In this limit, spin models such as Ising, XY and Heisenberg are known to yield a continuous (second-order) transition in 3D. We think that for the 3-state Potts model it should be of continuous type. If this is true, the nature of the transition should depend on the ratio $a = D/J$: when $a \ll 1$, the transition is of first order and when $a \gg 1$, the transition should become of a continuous type. This explains why at $D/J = 2$, the first-order transition is 'weakened', as shown in figures 5 and 6. It is noted that the weakening is stronger for larger r_c, as seen in figure 7, because one tends to the limiting case (ii) mentioned above.

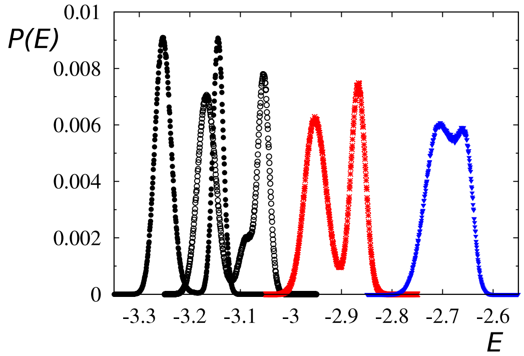

Figure 7. Energy histogram showing double-peak structure for $D = 2$ at several values of r_c: from left to right $r_c = \sqrt{6}$ (black circles), $\sqrt{8}$ (void circles), 3 (red stars, color on line) and $\sqrt{10}$ (blue triangles, color on line). $N_z = N = 48$, $J = 1$.

4. Conclusion

We have studied in this paper a molecular crystal characterized by three possible orientations of the molecular axes. The model is described by a short-range 3-state Potts model and a dipolar Potts interaction. We have analyzed the ground state as functions of the dipolar interaction strength D and its cutoff distance r_c. The ground state shows remarkable properties: it is composed of two interpenetrating independent subsystems with a layered structure whose periodicity depends on D and r_c. To our knowledge, such a ground state has not been seen in uniformly interacting molecules. We have used the Monte Carlo histogram method to study the phase transition in this system. In the absence of the dipolar interaction, the model which is a 3-state Potts model is known to undergo a first-order phase transition from the orientational ordered phase to the disordered state. Upon the introduction of a dipolar interaction of sufficient strength into the Potts model, the ground state is broken into layers as described above. We have shown that the transition remains of first order as the cutoff distance is increased at least up to $r_c = \sqrt{10}$.

The stacking of independent ordered layers is reminiscent of a smectic ordering. The coupling between adjacent different ordered planes is here strictly zero at $T = 0$, due to the Potts condition in the model. Note however that the model cannot describe smectic phases where molecular axes make tilted angles from the perpendicular (stacking) axis. In order to describe those phases in a realistic manner, it should be necessary to introduce continuous degrees of freedom for the molecular axis instead of the discrete Potts model studied here. For instance, we can consider spin models in the same way as in helimagnets, where helical structures result from an anisotropic Heisenberg spin model with competing interactions between nearest and next-nearest neighbors in the stacking direction, in addition to the dipolar interaction which favors layered structures as found here. Work in this direction is under way.

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