Model for pattern formation of charged particles

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We construct and investigate a model to study the pattern formation in a two dimensional system of vacancies and charged particles interacting with isotropic forces. By using Monte Carlo simulations, we derive the temperature dependence of correlation functions and order parameters. We show that a different pattern emerges if the interactions are restricted to nearest neighbors and if, additionally, particles can interact with next nearest neighbors. The analytic results for the nearest neighboring correlation function agree well with the simulations. Phase diagrams for the two types of interactions are also constructed. We demonstrate that in sufficiently low temperatures, the asymptotic state is a single cluster.

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I. INTRODUCTION

Studies of motion and the resulting pattern formation of charged particles on a surface have been carried out for a long time.[1–3] The problem is important in understanding the kinetics of adsorption and spatial ordering in surfaces. Such ordering is a cooperative phenomenon that results from competition between interactions among particles (and possibly with the substrate) and thermal excitations. The pattern formation of charged particles could also play a role in high-temperature superconductors.[4] Theoretical descriptions are based either on kinetic equations (see, e.g., a review by Gouyet *et al.*[5]), Kawasaki dynamics,[6] or Monte Carlo simulations.[7–9] Different problems have been studied, such as the estimation of diffusion parameters[5] and pattern formation.[6,7,9] A very good introduction to diffusion of particles and vacancies is in the paper of Sadiq and Binder,[10] wherein, however, only one type of particles is considered. A more general model that describes the kinetics of two types of particles (A and B) and vacancies, the so-called ABV model, has been studied by He and Pandey.[7] They considered a system of positively and negatively charged particles on a square lattice with a coverage equal to 0.5. Particles interacted with Coulomb-type forces depending on the distance (up to fourth nearest neighbors) and were also subject to a linear gradient external field. He and Pandey[7] observed the growth of antiferromagnetic domains with a lowering of the temperature. They also estimated the temperature at which a single, "infinite" cluster appears and called it a percolation-like transition. In a recent paper, Mertelj *et al.*[9] studied a system of charged particles, which is also on a square lattice. The particles interact with anisotropic Jahn–Teller forces for nearest neighbors (NNs) and next nearest neighbors (NNNs) and with isotropic long-range Coulomb forces. By using analytic (mean field) and simulation methods, Mertelj *et al.*[9] found that the system exhibits, depending on the forces acting, two types of phase transitions from a disordered to an ordered state. The distribution of cluster sizes as a function of temperature and coverage shows that, unlike in Ref. 7, there are mostly small clusters, and no tendency to form a single cluster has been observed.

In this paper, we shall study an ABV model with two types of oppositely charged particles moving on a square lattice. This is equivalent to the Ising spins with Kawasaki dynamics, which conserves the net magnetic moment, which corresponds here to the total charge. We would like to check whether charged particles have a tendency to group in a single cluster, as reported by He and Pandey,[7] or rather remain in many small clusters, as observed by Mertelj *et al.*[9] To keep the model simple, we restrict the interactions to isotropic ones acting between NNs and then also NNNs. Obviously, charged particles interact in general with long-range Coulomb forces. If, however, the local charge is screened, then we have a reduced range of the potential between sites i and j,[11,12]

$$V_{ij} \approx e^{-kr_{ij}}/r_{ij}, \tag{1}$$

where k is a constant. By choosing sufficiently large k, the interaction range could be diminished. Such a form of restricted effective interactions between charged particles has been used in analytical calculations and Monte Carlo (MC) simulations for a long time.[11–14] For the sake of brevity, whenever we shall refer to NNN interactions, we will mean interactions to NNs and up to NNNs. There will be no external field and the coverage will be equal in general to 0.2, although other coverage values will be also discussed. Our aim is to also find out what are the possible patterns in stationary states and whether such a simple system will self-organize into a single cluster. To have a measure of the ordering of the system, we will construct order parameters and corresponding phase diagrams.

II. MODEL

We investigate a system of a fixed number N of particles located at sites of a square lattice of linear dimensions $L \times L$ with periodic boundary conditions. The particles have positive and negative charges of value 1. The numbers of positively and negatively charged particles are equal. A particle located at a site i interacts with a particle j that is its NN with a force J_1 or with a NNN particle with a force $J_2 = J_1/\sqrt{2}$. Hence, the Hamiltonian for the model is

$$\mathcal{H} = -J_1 \sum_{i=1} \sum_{j=1}^{\delta} n_i n_j - J_2 \sum_{i=1} \sum_{j=1}^{\Delta} n_i n_j, \tag{2}$$

where $n_i = 0, \pm 1$, depending on whether the site i is empty or occupied by a positively or negatively charged particle, and where δ indicates summation over NNs and Δ indicates summation over NNNs. The number of particles, N, is smaller than the number of available sites, V. This is therefore analogous to the Hamiltonian for the ABV model,[5] with the interactions between likely charged particles equal. It is also a stochastic lattice gas with internal degrees of freedom. The system is in contact with a heat reservoir, which keeps the temperature T constant. During one Monte Carlo step (MCS), which is our time unit, each particle (on average) could move to an empty NN site. The dynamics is governed by the Metropolis algorithm.[15] A jump is realized with a probability of 1 if the new location is energetically favorable; otherwise, it is realized with a probability

$$p = e^{-\beta \Delta E}, \tag{3}$$

where $\beta = (k_B T)^{-1}$ is the inverse temperature and ΔE is the difference between the energies after and before the jump. In the following, we shall use the reduced temperature $T = k_B T / J_1$. Most of the analysis is done for the concentration of the particles $c = 0.2$; however, we will discuss also the case of other concentrations. It should be noted that there is always the same number of positively and negatively charges particles; hence, the total charge is zero. The dynamics we use is therefore the Kawasaki dynamics.[16] We have the following external parameters: linear size of the lattice, concentration, and temperature T. In simulations, our system will be described by an order parameter, which is defined below, and the correlation function G_d, which measures the correlation between particles separated by d lattice constants as follows:

$$G_d \equiv G(\vec{r}, \vec{r} + \vec{d}) = \frac{1}{2N} \sum_{i=1}^{N} (n_i n_{i+dx} + n_i n_{i+dy}). \tag{4}$$

We defined two, closely related order parameters, which are analogous to the ones introduced in Ref. 10. One is for interactions to NNs only,

$$\varphi_1 = \frac{1}{8N} \sum_{i=1}^{N} \left| \sum_{j=1}^{\delta} n_i n_{i+j} - \sum_{j=1}^{\Delta} n_i n_{i+j} \right|. \tag{5}$$

Similarly, for interactions up to NNNs, the order parameter φ_2 is defined as

$$\varphi_2 = 2\varphi_1. \tag{6}$$

If we restrict ourselves to interactions to NNs only, then correlation function (4) becomes

$$G_1 = \frac{1}{4N} \sum_{i=1}^{N} \sum_{j=1}^{\delta} n_i n_{i+j}. \tag{7}$$

III. MEAN FIELD

Here, we shall investigate interactions restricted to NNs and use the Oguchi approximation,[17] which is one of the simplest variations of the mean field approaches, yet it permits the calculation of a correlation function. This choice is motivated by the fact that the net charge (magnetization) is zero at all temperatures; hence, the only information we may get about the system when the temperature is changed comes from the correlation function. In the Oguchi method, the interaction between a pair of spins (particles) is explicitly calculated, while the interactions with other particles is replaced by an effective field, which, as usual, is proportional to the net magnetic moment. By denoting two possible charges of a particle by $s_i = \pm 1$, and basing upon the fact that a site could be either vacant or occupied by an occupation variable $t_i = 0, 1$ the Hamiltonian of our system within the Oguchi approximation will have the form

$$\mathcal{H} = -J t_1 t_2 s_i s_2 - H_{ef}(t_1 s_1 + t_2 s_2). \tag{8}$$

However, since H_{ef} is proportional to the net charge, and this is always zero, the partition function is

$$Z = \mathrm{Tr}_t \, \mathrm{Tr}_s \, e^{-\beta \mathcal{H} + \beta \mu} = \mathrm{Tr}_t \, \mathrm{Tr}_s \exp[K s_1 s_2 t_1 t_2 + \nu(t_1 + t_2)]. \tag{9}$$

Here, $\nu = \beta\mu$, μ is the chemical potential, and $K = \beta J$. Taking the traces, we get

$$Z = 8(1 + 2x + x^2 \cosh K), \tag{10}$$

where $x = e^{\nu}$. Now, we have to replace the chemical potential by the control parameter c, which is the density of particles,

$$c = \frac{x \, \partial \, \ln Z}{\partial x} = \frac{x + x^2 \cosh k}{1 + 2x + x^2 \cosh K}. \tag{11}$$

Since x must be non-negative and we are interested in small densities, hence, $2c < 1$, and we get

$$x = \frac{1 - 2c}{2(1 - c)\cosh K} \left[\sqrt{1 + \frac{4c(1 - c)\cosh K}{(1 - 2c)^2}} - 1 \right]. \tag{12}$$

From the partition function Z, we may also get the two-point (to NNs) correlation function as follows:

$$G_1 = \langle s_1 s_2 t_1 t_2 \rangle = \frac{\partial \ln Z}{\partial K} = \frac{x^2 \sinh K}{1 + 2x + x^2 \cosh K}. \tag{13}$$

Inserting x from Eq. (12) into Eq. (10), we get the partition function as a function of the concentration c and the (reduced) inverse temperature K, similarly for correlation function (13). The set of Eqs. (11) and (13), together with Eq. (12), corresponds to the equations for magnetization and density derived within the Bragg–Williams approximation in Ref. 18 and used later in Ref. 19.

IV. SIMULATION RESULTS

Most of the figures presented below have been obtained from simulations on $L = 100$ lattices; however, for better readability spatial patterns ("snapshots") are shown for $L = 30$ lattices. We have found out that averaging only over 10 independent runs provided errors bars on the order of the thickness of the lines on the figures. Periodic boundary con-

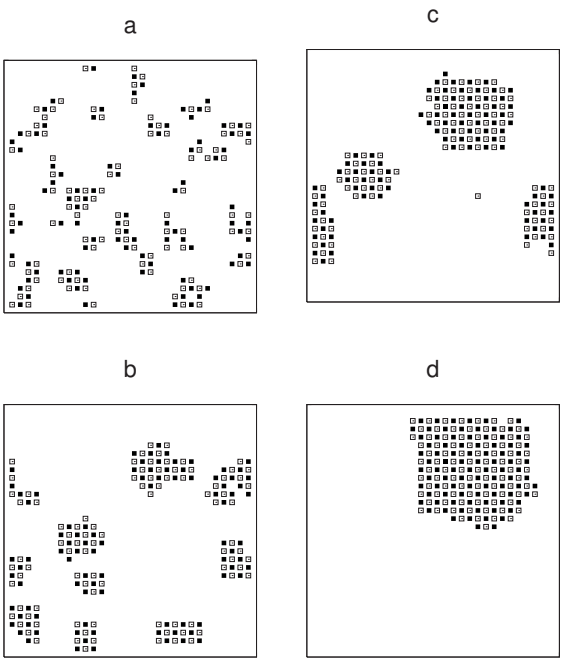

FIG. 1. Spatial position of two types of particles on a $L=30$ lattice: (a) $t=100$, (b) 10^4, (c) 5×10^5, and (d) 5×10^6 MCSs. Interactions to NNs only. The particles are marked by open and full squares. Periodic boundary conditions. $c=0.2$.

ditions have been adopted. We have used standard MC simulations, which for intermediate and high temperatures are satisfactory, and this is the region on which we focus our attention. It should, however, be noted that for low temperatures, when the mobility is small, there exists another, method which yields faster convergence—the multicanonical MC technique, which was introduced in Ref. 20 and is also discussed in Ref. 15.

A. Interactions to NNs

Let us start with interactions restricted to NNs. The time evolution of spatial arrangement at medium temperatures is shown in Fig. 1. As could be seen, there is a tendency to form clusters of strictly antiferromagnetic arrangement,

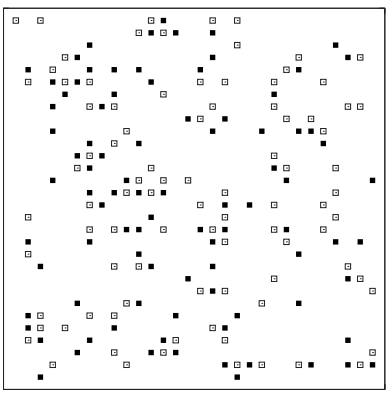

FIG. 2. Asymptotic state (after 2×10^6 MCSs) for $T=0.7$. Interactions to NNs only. $c=0.2$.

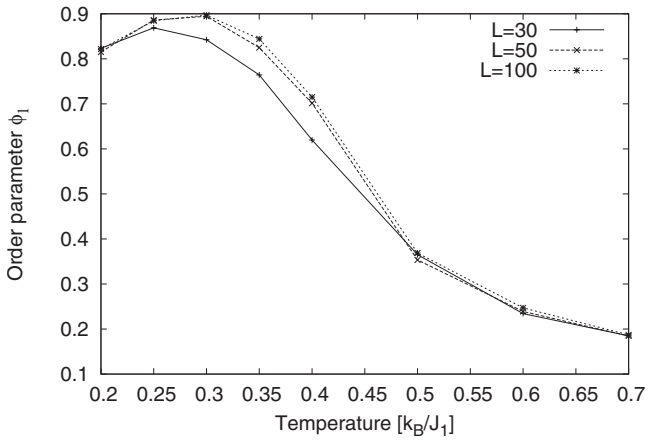

FIG. 3. Phase diagram for NN interactions and $c=0.2$.

which leads to an ordered phase. This agrees with the structure found in Ref. 7. The ordering is the same in small clusters of just a few particles formed after 10^4 MCSs and large ones after 5×10^6 MCSs. At lower temperatures, the process is considerably slowed down and the time scale of the clusters movements is quite different from our MC dynamics. By extrapolating the obtained results, we estimate that a single cluster will be formed at $T=0.2$ and for $L=50$, after about 1.5×10^8 MCS. At high temperatures, the system could not organize itself. This situation is presented in Fig. 2 for an asymptotic state (after 2×10^6 MCSs) and the temperature $T=0.7$. This should be compared to Fig. 1(d). This confirms the observation of He and Pandey[7] that lowering the temperature leads to a kind of "percolation transition," which in our case happens around $T_{\mathrm{per}}=0.3$. This value could be deduced either from the phase diagram (Fig. 3) or from the sequence of asymptotic states, part of which is reproduced in Fig. 1.

Correlation function versus distance between the particles is plotted in Fig. 4 for three values of the temperature. At high temperatures only a short order (to NNs, i.e., the range of interactions) persists, while at lower temperatures correlations are visible at nearly ten times the interaction range.

The two-point correlation function to NNs could be also calculated from Eq. (13). Figure 5 shows the comparison of

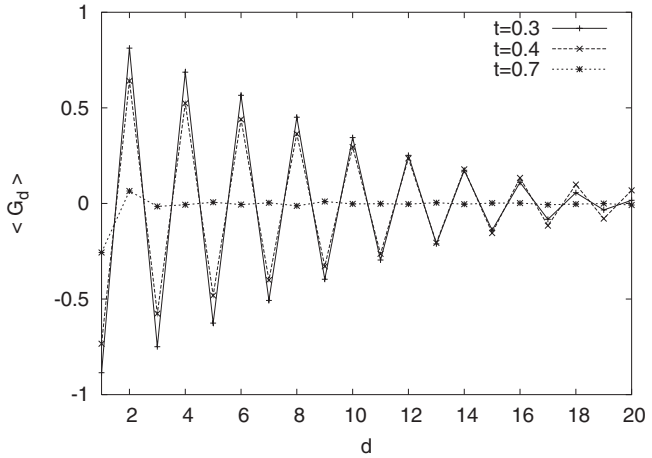

FIG. 4. Average correlation function $\langle G_d \rangle$ versus distance d for interactions to NNs and $c=0.2$.

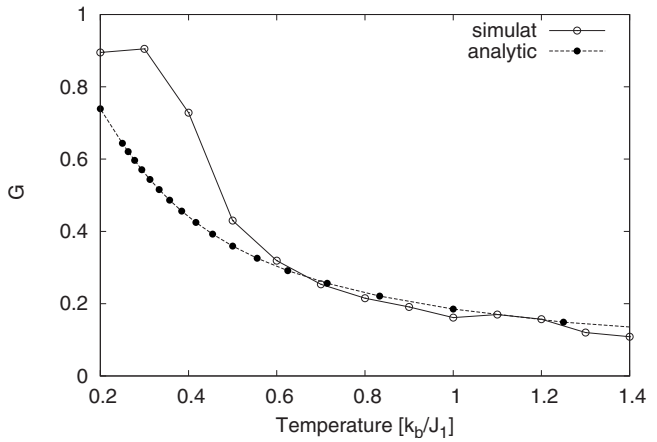

FIG. 5. Comparison of simulation and analytic dependences of correlation function on temperature for $c=0.2$.

the simulation and analytic formula for $c=0.2$. The agreement is good except for lower temperatures, i.e., below $T=0.3$, where the simulation shows much weaker correlations. This is due to the fact that at low temperatures, as we have mentioned above, the mobility of clusters is extremely slow; hence, during accessible simulation time, the system is not yet fully organized. This is also visible in the phase diagram shown in Fig. 3, wherein order parameter (5) is plotted against the reduced temperature. At very low temperatures, there is a drop in the value of the order parameter, showing the nonperfect ordering of the system.

B. Interactions to NNs and up to NNNs

Now, we add interactions up to NNNs, which have the same character, although they are weaker, as the interactions to NNs. As could be expected, the problem becomes more complex. The transition from a disordered state to an ordered one, provided the temperature is appropriate, is faster than before. This is illustrated in Fig. 6, which shows snapshots of spatial configurations at several time moments. One may notice that the essentially antiferromagnetic structure of NN interactions has been replaced by a "diluted" structure, wherein each particle has vacancies as its nearest neighbors. Also, the clustering is much faster and already after 5×10^5 MCSs we observe one big cluster and very few "free" particles.

For a system on a $L=50$ lattice and at $T=0.2$, we have also calculated the number of clusters in the system as a function of time for both cases. The results are plotted in Fig. 7. For interactions to NNs only, apart from the initial data for $t\leq10^4$ MCSs, we observe a rather good fit with a power law (over 3 decades) with the exponent $\alpha_{\mathrm{NN}}=-0.45$. For interactions up to NNNs, the initial data better fit a power-type dependence, but with a different exponent $\alpha_{\mathrm{NNN}}=-0.35$. However, at larger times, when only a few clusters remain in the system, the process of coalescence is much faster. If one would want to also see a power-type dependence here, an appropriate exponent would be about −1.4. The larger scatter observed for NNN than for NN interactions is due to more processes of consolidation and breaking apart of large clusters, especially at later stages, when only a few clusters exist in the system. These processes are virtually absent for NN interactions. The "percolation temperature" could be also estimated here and, as could be expected, it has a lower value than for interactions to NNs only, $T_{\mathrm{per}}\approx0.2$.

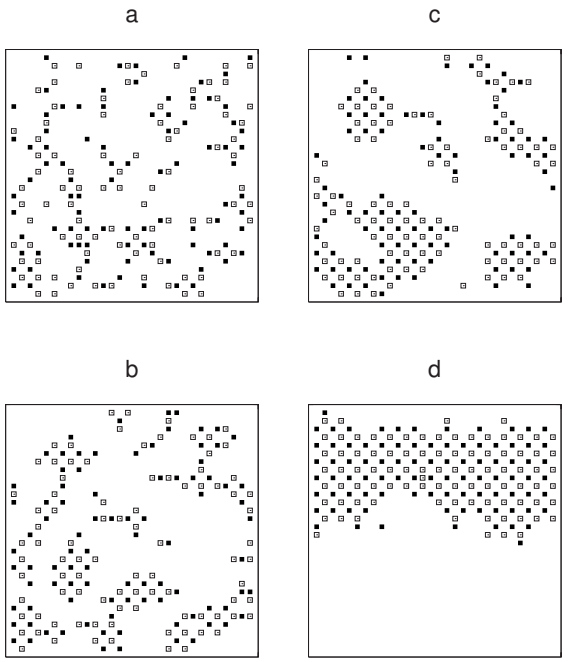

FIG. 6. Spatial position of two types of particles on a $L=30$ lattice for interactions up to NNNs and $c=0.2$. (a) $t=100$, (b) 2×10^3, (c) 10^5, and (d) 5×10^5 MCSs.

Figure 8 shows correlation function (4) in the asymptotic state (after 10^6 MCSs) for lower ($T=0.2$), medium ($T=0.3$), and high ($T=0.5$) reduced temperatures. A comparison to the analogous plot for interactions to NNs only (see Fig. 4) shows that the ordering is much better for interactions up to NNNs. For intermediate temperatures, we observe quite strong correlations, even at distances more than ten times larger than the range of interactions. For low temperatures, particles are correlated at much shorter distances but better

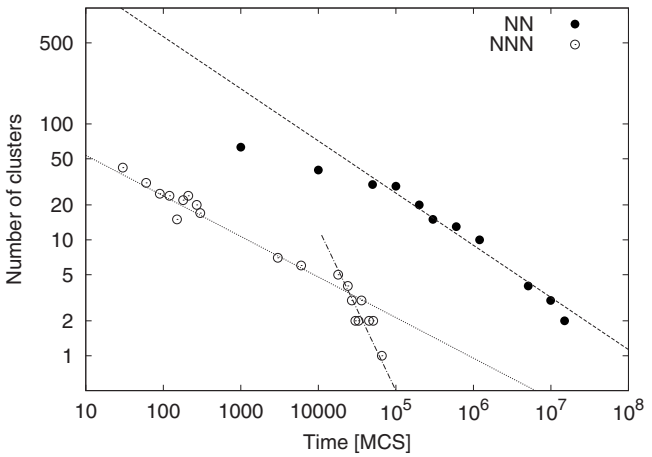

FIG. 7. Time dependence of the number of clusters in a system on 50×50 lattice and at $T=0.2$ for interactions to NNs and up to NNNs. $c=0.2$.

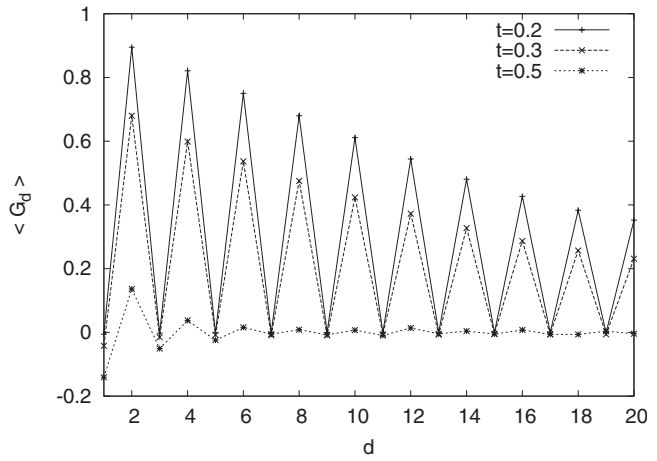

FIG. 8. Average correlation function $\langle G_d \rangle$ versus distance d in lattice constants for interactions up to NNNs and $c=0.2$ and $T=0.2$.

than for interactions to NNs only, which is a proof that in this case, the system is not well organized spatially because the mobility is limited. On the contrary, for high temperatures, we discover only a trace of nearest correlations, such as for NN interactions. One may notice that for $T=0.2$, the correlations linearly fall off with the distance. As could be seen from Fig. 9, order parameter (6) is quasi-independent of the size of the system. Only for very small lattices, such as $L=30$, we observe a small deviation from the general pattern. A similar independence of the results on the size of the system was also reported in Ref. 9. A comparison to the phase diagram for interactions to NNs only (Fig. 3) shows not only a similar general pattern but also a much smaller dip for very low temperatures, which is the result of better ordering when two types of interactions are present.

V. COVERAGE DEPENDENCE

When the concentration of particles is smaller than 0.5 and the temperature is not too high, we have observed the formation of a single cluster, which covers a part of the lattice. At a coverage equal to 0.5 and interactions to NNs, the

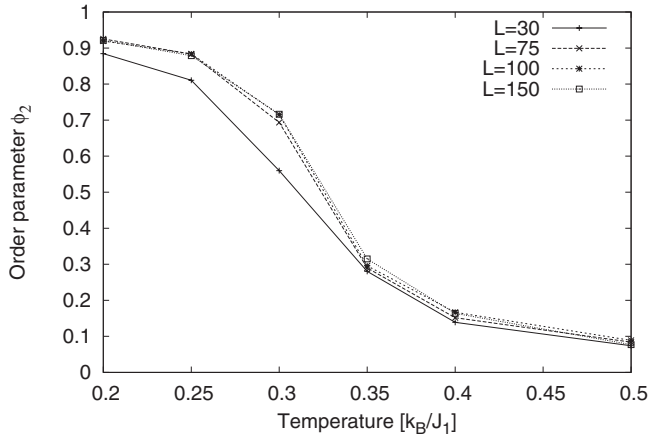

FIG. 9. Phase diagram for NNN interactions; $c=0.2$.

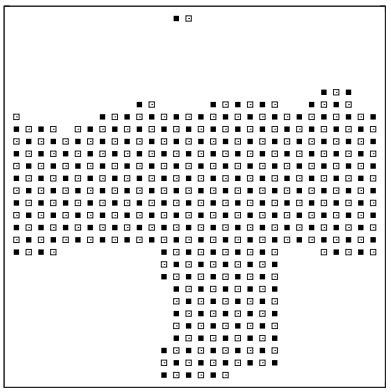

FIG. 10. Asymptotic state for NN interactions; $c=0.5$ and $T=0.2$.

particles form a single "antiferromagnetic" cluster spanning across the lattice (see Fig. 10). With increasing concentration, the cluster is simply growing. More interesting is the case of interactions up to NNNs. Then, at $c=0.5$ and a not too high temperature, the whole lattice is filled by a perfectly ordered structure [Fig. 11(a)]. At higher temperatures, the ordering is destroyed—in some places the particles are at NN sites, while at others larger voids are produced. Evidently, the most interesting region is at intermediate temperatures, where a stable pattern could be formed. For $c>0.5$ [Fig. 11(b)], we observe the formation of local, more compact, clusters wherein particles of the same type are organized into strips or lines. A comparison of the plots of the correlation functions for lower concentrations, $c=0.2$, and higher concentrations and interactions up to NNNs (Figs. 8 and 12) shows that the general features do not change with increasing concentration and, as should be expected, correlations with interactions up to NNNs are stronger. We may use again order parameter (6) to get the phase diagram (Fig. 13). We see that the slope of the division line is decreasing with increasing coverage, meaning, that the ordered phase is more stable for higher coverages. One interesting feature should be noticed. Only in the case of $c=0.5$ is the maximum value of the order parameter is equal to 1. Otherwise, it is always less than 1. This is so because only for $c=0.5$ the whole lattice could be perfectly organized; in other cases, there are always clusters and the existence of the clusters borders reduces the value of the order parameter. If we agree that the transition temperature (T_c) between the ordered and the disordered

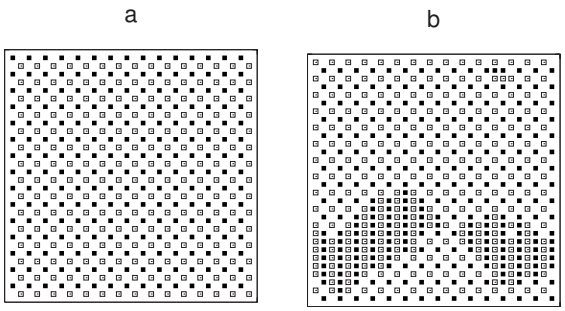

FIG. 11. Asymptotic state for NNN interactions and $T=0.2$: (a) $c=0.5$ and (b) $c=0.6$.

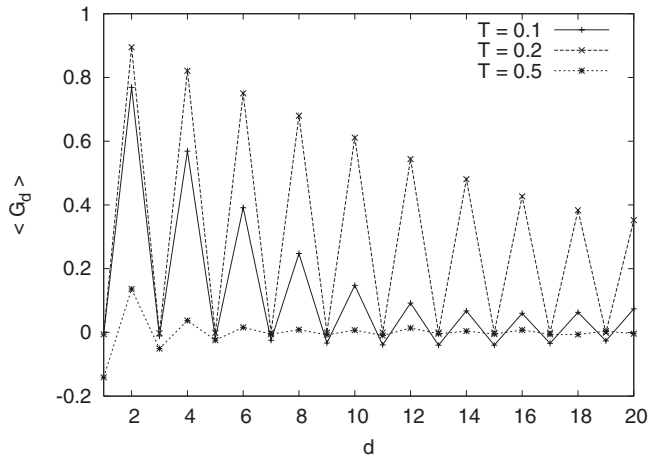

FIG. 12. Average correlation functions $\langle G_d \rangle$ versus distance d for interactions to NNs and up to NNNs and $c=0.5$.

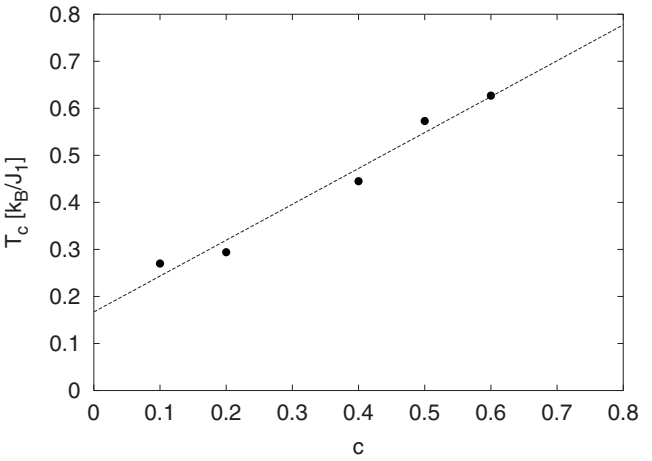

FIG. 14. Phase diagram in the (c,T_c) plane for NNN interactions.

states could be determined as the inflection point of the lines on the phase diagram in the (ϕ_2,T) plane (Fig. 13), then we may construct another phase diagram, this time on the (c,T_c) plane (see Fig. 14). As could be expected, with increasing concentration, the ordered phase becomes more stable and could exist at higher temperatures. The dependence is linear with the slope equal to 0.763.

VI. CONCLUSIONS

We have presented and discussed a simulation model that describes a system of the same number of particles of two types—positively and negatively charged. In all other aspects, the particles are identical. The particles interact via isotropic forces and are free to move to an empty nearest neighboring site. We have considered interactions only to NNs and to NNs and NNNs. We have assumed, in general, that the concentration of the particles is rather low, i.e., $c=0.2$. We have shown that the system self-organizes if the temperature is not too high. For very low temperatures, the mobility of the particles and even more so that of the clusters of particles is very restricted. There are many states with

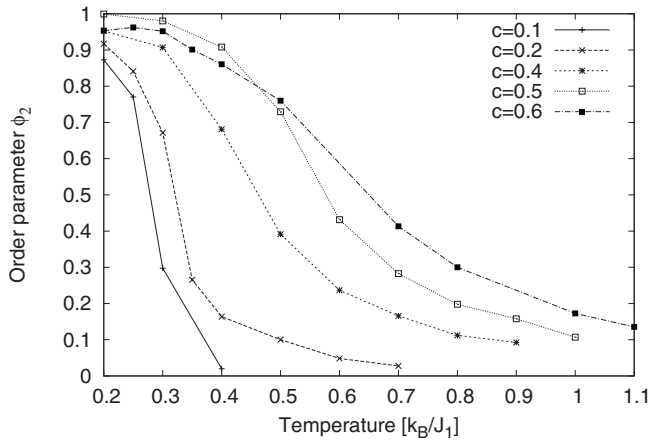

FIG. 13. Phase diagram in the (ϕ,T) plane for NNN interactions.

local minima and the particles do not have enough energy to overcome local energy barriers. This is similar in effect, although for different reasons, to the situation in the spin-glass models.[21] In high temperatures, the order, such as that in a paramagnet, is destroyed by chaotic jumps to all, not necessarily energetically favorable, positions. At high temperatures, we get many small and uncorrelated clusters, which, as the temperature goes down, coalesce into a single cluster antiferromagnetically ordered. We have thus obtained the same results as those of He and Pandey[7] but without the external field. Growing clusters retain their initial structure. Many small clusters reported in Ref. 9 we have observed only in the high temperature phase. One should, however, bear in mind that interactions in our model were isotropic, whereas in their model the short-range ones were anisotropic. We have shown that the process of formation, at a given temperature, of a single cluster, i.e., the number of clusters in the system versus time, has a power-type character. For interactions up to NNNs, the process of formation of a single cluster is faster and the resulting structure is more complex. Now, each positively charged particle has vacancies as its NN and negatively charged particles as its NNN. Again, this agrees with what was reported in Ref. 7. We have also constructed two, closely related, order parameters—for NN and NNN interactions—and we have determined their dependence on the temperature. For the NN interactions, we have calculated, within the Oguchi approximation, the correlation function to NNs. The dependence of this function on the temperature agrees well with simulation results and the one given in Ref. 9. From simulations, we have also calculated temperature dependence of the correlation function for up to farther neighbors. We have shown that at very low temperatures, due to very restricted movements of the particles, correlations are effective only at a short range. At slightly higher temperatures, the distance over which particles are correlated extends to ten times the range of the interactions. It is a little less than ten times for NN interactions and more than ten times for NNN interactions. At still higher temperatures, the correlations are broken and we have a disordered ("paramagnetic") state. We have also shown that below the temperature at which ordering is destroyed, single,

ordered cluster grows with increasing coverage. At $c=0.5$ and for NNN interactions, it spans the whole system, producing a perfect order. A further increase in the coverage leads to the formation of more compact clusters in the form of lines or stripes of particles of the same type, over a more sparsely ordered background. The phase diagram in the (φ,T) plane has basically the same character for all concentrations, although the curve that separates the ordered and the disordered phases is less steep for a higher coverage. We have demonstrated that at a higher coverage, the system is more stable against the temperature.

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