Surface diffusion of charged particles: Monte Carlo study

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We investigate a model of a two-dimensional system of charged particles and vacancies. The particles interact with isotropic forces, attractive or repulsive, to nearest and next-nearest neighbors, and could move through the lattice. Using Monte Carlo simulations we determine mean-square displacement as a function of time, temperature, coverage, and range of interactions. We estimate also the tracer diffusion coefficient and the activation energy. We show that some characteristics are rather insensitive to the range of interactions while others change markedly. In particular formation of a single domain along one direction is possible for rather high coverages in the case of interactions to nearest neighbors but it could happen at much lower coverages when the interactions are also to next-nearest neighbors. Such a situation could be temporarily unstable—single cluster stretching along the lattice in one direction could break apart and then consolidate again, sometimes in the direction orthogonal to the previous one. We have shown that, depending on the temperature, coverage, and range of interactions, the model exhibits subdiffusion, normal diffusion, or superdiffusion, or the movements of the particles could not be written as a power-type function. We have also demonstrated that the type of pattern formed and type of diffusion depends on whether the lattice has odd or even number of sites.

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I. INTRODUCTION

Studies of diffusing particles on planar surfaces are very interesting from many points of view and have been carried out for a long time. For reviews see, e.g., Gomer[1] or Gouyet *et al.*[2] In nonequilibrium systems, where the concentration is not homogeneous, diffusion is complicated by existence of ordered domains (clusters) appearing for some interactions, certain range of coverage, and temperature. Modeling of such systems could be regarded as approximation of descriptions of transport properties of superionic conductors or diffusion of adsorbed ions on surfaces. The problem of ion diffusion on a surface received much attention (see, e.g., Refs. 3–6). Diffusion takes place in a periodic potential coming from the substrate, which could be conveniently modeled by a periodic lattice. Recently, fast developing microtechnologies and nanotechnologies require good understanding of processes happening during pattern formation on a surface.[7] Controlling the formation of patterns with a given symmetry is possible if at least basic mechanisms are known. Interactions among the constituents of the processes could have different origins, ranges, and characters. Capturing in a model all or even the most important features of such interactions may be difficult. It is therefore desirable to approach the problem from many viewpoints and using different models. Simple ones, such as the one presented by us, could also be helpful in understanding pattern formation and diffusion processes in the case of partially screened charged particles, moving on a surface with square lattice symmetry.

Diffusion of ions or atoms on a surface has many aspects and allows in some cases for analytical treatment although mostly within the mean-field-type approach.[2,8] The master-equation technique[9] or computer simulations[10] have been also used. The features commonly investigated are pattern formation[11–14] or dynamic parameters.[2,9,15] In some of these papers just one type of particles is considered, whereas in others[2,9,12,15] particles are of two types, A and B. Sometimes models allow for empty sites (vacancies) and such models are called the ABV models.[2] Forces acting between particles could have various character and range of interactions—from simple nearest-neighbor (NN) interactions[8,15] to next-nearest-neighbor (NNN) interactions (Refs. 9 and 15) and farther.[12,13] The interactions could be of one type (attractive or repulsive) or there could be a combination of both.[15] As has been proven before,[16] tracer movements of a free particle are different from movements of a particle with hard-core interactions.[17] In particular, the mean-square displacement, hence the diffusion coefficient, decreases when the interactions are switched on. Successive jumps are not independent but some correlations appear. Since exclusion principle is certainly an oversimplification of real interactions, it is important to check the possible effect of adding physically more realistic interactions to the system. In a series of papers Uebing and Gomer (UG) (Ref. 15) investigated a system in which particle interactions were determined by the distance between them. The square lattice was filled with a given coverage of one type of particles. Interactions were restricted to NN or NNN and the range defined the sign of the interactions, such that NN were repulsive but NNN were attractive, or vice versa. Using Monte Carlo simulations UG have derived the dependence of the chemical and tracer diffusion coefficients on the temperature and coverage of the diffusing atoms, as well as the activation energy. They have shown that switching on and off different interactions may lead to different behavior of the temperature dependence of the tracer diffusion coefficient and activation energy as functions of the surface coverage. Interactions which had different sign when restricted to NN and when extended to NNN have been also considered in Ref. 18. Sadiq and Binder[9] studied, via Monte Carlo simulations, a model of atoms moving on a square lattice with repulsive interactions of the same strength between NN and NNN. They have shown that this simple and isotropic model exhibits in the ordered-phase anisotropic diffusion.

In a recent paper[14] we have proposed a model, subject to dynamics of the Kawasaki type, of two types of particles on a two-dimensional (2D) square lattice. We have shown, using Monte Carlo simulations, how the resulting spatial organization of particles depends on the coverage, range of interactions (NN or NNN), and the temperature. In our model the sign of the interactions depends on the type of the interacting particles. It is repulsive between particles of the same type and attractive when the particles are of different type. The sign of the interaction depends on the types of interacting particles and not on their distance like in the previously studied models.[9,15]

In this paper we concentrate on the dynamic parameters (diffusion coefficient, average displacement, and activation energy) for the model introduced by us earlier.[14] We want to find out how allowing for NN interactions which could have both repulsive or attractive character, depending on the type of interacting particles, will influence the diffusion process. In particular, what will be the effect of these interactions on the dependence of the tracer diffusion coefficient and activation energy on coverage and temperature? Next, could such, in principle, symmetric interactions lead to anisotropic diffusion, as reported earlier?[9] We have demonstrated[14] that at sufficiently low temperatures a single cluster (domain) is formed without, however, discussing the dynamics of cluster formation and the effect such structures have on the diffusion processes. In this paper we investigate time dependence of the average cluster size at different temperatures. Such plots could help in understanding pattern formation mechanisms and the range of parameters (interactions, coverage, and temperature) for which the clusters remain relatively stable. We shall show how the above characteristics will change if we extend the interactions to NNN, which of the features will be affected by additional interactions, and which will remain only slightly changed. We shall also show how the patterns formed and the diffusion parameters depend on the size of the lattice, in particular whether it has even or odd number of nodes.

II. MODEL

A set of a fixed number of particles, located at sites of a square lattice with periodic boundary conditions, is considered. The lattice has $L \times L$ size and each site could be occupied by either a positively charged particle (A), negatively charged particle (B), or it could be empty. Double occupancy is forbidden. The total number of particles (A and B) divided by the number of available sites defines the coverage c. The number of A particles is equal to that of B particles and it remains constant. A particle at a site j interacts with a particle i at one of its NNs with force $J_1 > 0$, or with a NNN with force $J_2 = J_1/\sqrt{2}$. Therefore the Hamiltonian for the model has the form

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

where $n_i = 0, \pm 1$, depending on whether the site i is empty or is occupied by an A or B particle. δ indicates summation over NNs and Δ over NNNs. The model is equivalent to stochastic lattice gas with additional (charge) degrees of freedom. During one time unit [Monte Carlo step (MCS)] each particle could move, following the standard Metropolis algorithm,[19] to an empty NN site. Constant temperature is maintained through contact with a heat reservoir. In the following we shall use reduced dimensionless temperature, defined as

$$T = \frac{k_B T}{J_1}. \tag{2}$$

Since the number of A particles is always equal to that of B particles, the net charge of the system is conserved and is equal to zero; hence the Kawasaki dynamics[20] is used. In the simulations we shall determine the following basic quantities. Average distance $\langle r^2(t) \rangle$ is traveled by a particle in time t, as well as the distances along the OX and OY axes—$\langle x^2(t) \rangle, \langle y^2(t) \rangle$. Using these quantities other important functions could be determined.[1] The tracer diffusion coefficient D^*

$$D^*(t) = \frac{1}{4Nt} \sum_{i=1}^{N} \langle |\vec{r}_i(t) - \vec{r}_i(0)|^2 \rangle, \tag{3}$$

where N is the total number of particles while $\vec{r}_i(t)$ and $\vec{r}_i(0)$ are the positions of the particle i at time t and time 0, respectively. From Eq. (3) we may get the activation energy, E_a, as[15]

$$D^* \sim e^{-\beta E_a} \Rightarrow E_a = -k_B T \ln(D^*). \tag{4}$$

Useful information about the spatial arrangement of the system could be obtained from the time dependence of the average cluster size. Following Binder and Landau[19] we have calculated only clusters larger than ten particles. Information about cluster dynamics is essential in estimating the range of parameters for which the observed pattern is stable and where it is changing rapidly.

Most of the simulations are carried out for not so large systems, i.e., 50 by 50. The reason is that, as it is well known on larger systems, processes are happening at lower rates. To see the formation of a single domain for a 50×50 lattice, simulations had to be run until 3×10^7 MCS. The same effect will appear on a 100×100 lattice at least one order of magnitude later. We have performed checks for 50×50 and 100×100 lattices for different coverages and interaction ranges. For lower coverages (below 0.5), where there is normal diffusion, there are no differences between larger and smaller systems (see Fig. 1). For $c = 0.6$ and $T = 0.4$ subdiffusion appears much (about one order of magnitude) later on the 100×100 lattice. Most of the curves showing time dependence come from single runs since the observed effects could be shifted in time in different runs. Therefore the effects will be lost after averaging. We have performed several runs for each presented case to check if the effect is not particular to this realization.

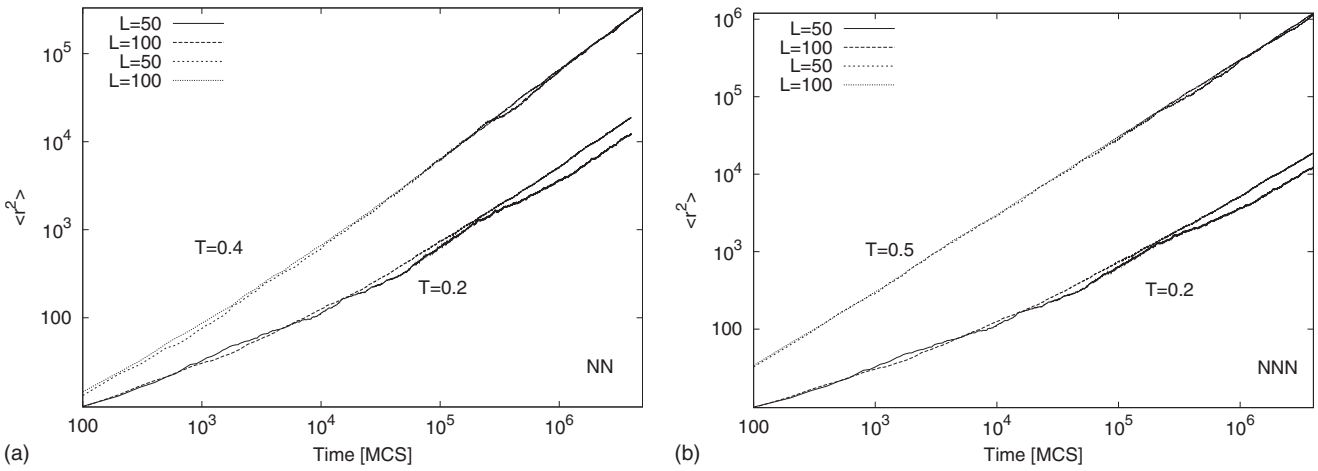

FIG. 1. Mean-square displacement for $L=50$ and $L=100$ lattices. Left panel: interactions to NN; right panel: interactions to NN and NNN. $c=0.2$.

III. RESULTS

A. Interactions to NN

We start with interactions restricted to NN only. Later on we shall consider interactions to NN and to NNN. In Fig. 2 time dependence of the mean-square displacement (MSD) $\langle r^2(t)\rangle$ is presented on a log-log plot for three values of the coverage c. To estimate the character of diffusion we have checked whether the data could be fit into a power-type function

$$\langle r^2(t)\rangle \sim t^{\alpha}. \tag{5}$$

Averaging here and later is over all particles in the systems. At low coverages ($c=0.2$) and $L=50$, this corresponds to an average of over 500 trajectories. As could be seen from Fig. 1 averaging over 2000 trajectories yields essentially the same result. For $c=0.2$ (left panel of Fig. 2) and low temperature ($T=0.2$), we have found that until $t \leq 10^5$ MCS the dependence of $\langle r^2\rangle$ on time does not have a power-type character. Afterward, it could be fitted into such a behavior with $\alpha \approx 0.8$, indicating subdiffusion since the interactions restricting movements of particles are stronger than thermal motion. Increasing the temperature to $T=0.3$ results in larger mobility and therefore α also grows to the value of about 0.92. At $T=0.4$ and higher, we observe normal diffusion with $\alpha=1$. In all cases the diffusion is the same along the X and Y directions, i.e., $\langle x^2(t)\rangle \approx \langle y^2(t)\rangle$.

Another situation is found when $c=0.5$ (right panel of Fig. 2). Also here at low temperature ($T \leq 0.3$) the time dependence of $\langle r^2\rangle$ has no power-type character and the diffusion becomes normal at $T=0.4$. For coverage exceeding 0.5, like $c=0.6$ (shown in the bottom panel of Fig. 2; Fig. 3), the low-temperature ($T=0.2$) diffusion is even slower than in lower coverage and also has no power-type character, which appears only after about 10^7 MCS. At high T (≥ 0.7) normal diffusion is observed. In between ($T=0.4$) a complex behavior is seen; after about 10^4 MCS normal diffusion begins and it lasts until about 7×10^5 MCS, then it slows down considerably only to burst into superdiffusion after some 2×10^6 MCS. Figure 4 shows the mean-square displacements, $\langle x^2\rangle, \langle y^2\rangle, \langle r^2\rangle$ versus time for $c=0.6$, $T=0.4$, and much longer, until 3×10^7 MCS simulations. We see that after some 5×10^5 MCS diffusion is slowed down in both X and Y directions. Most of the particles are grouped in a single cluster stretching, depending on the situation (simulation run) either along the OX or OY direction. In the presented case the cluster is along the OX axis; hence it blocks diffusion along the Y direction. Since the system is not in a stationary state, such clusters eventually break down into smaller ones which do not span the whole system, allowing for diffusion in both directions. Particles which do not belong to the clusters could "glide" along its borders at zero cost; hence such movements are indeed ballistic and they lead to the observed superdiffusion. Afterward, the situation changes again—a percolating cluster is formed but this time along the OY axis; hence the diffusion in the X direction is blocked. Snapshots taken at different times and presented in Fig. 4 illustrate the process. One should also notice that the value of $\langle r^2\rangle$ at the end of simulations is much lower when the coverage is high ($c \geq 0.5$) than when it is low. At high concentrations movement of particles is often blocked by occupied sites. Presented figures come from a single run. Using a different seed for the random number generator but for the same coverage may lead to formation of an initial cluster along the OY axis, and then the role of $\langle x^2\rangle$ and $\langle y^2\rangle$ would be reversed. This is why we present results coming from single runs without averaging. It should be noted that He and Pandey[12] also reported a (symmetric) drop in the mean-square displacement for low temperatures.

Flipping of the domain orientation (horizontal to vertical or v.v.) has some resemblance to magnetization flips in the Glauber dynamics of Ising spins reported in Ref. 21 for extremely small (six by six) system. To see whether the effect of domain flip is in our model due to relatively small size of the investigated system, we have performed simulations on smaller and larger lattices ($L=20$, 30, 40, 50, and 100). It turned out that in smaller lattices the time of formation of a single domain, blocking diffusion in one direction, occurs earlier (see Fig. 5). Also a change in the orientation of the domain happens earlier. We could not find any functional dependence giving either the time of the first appearance of a

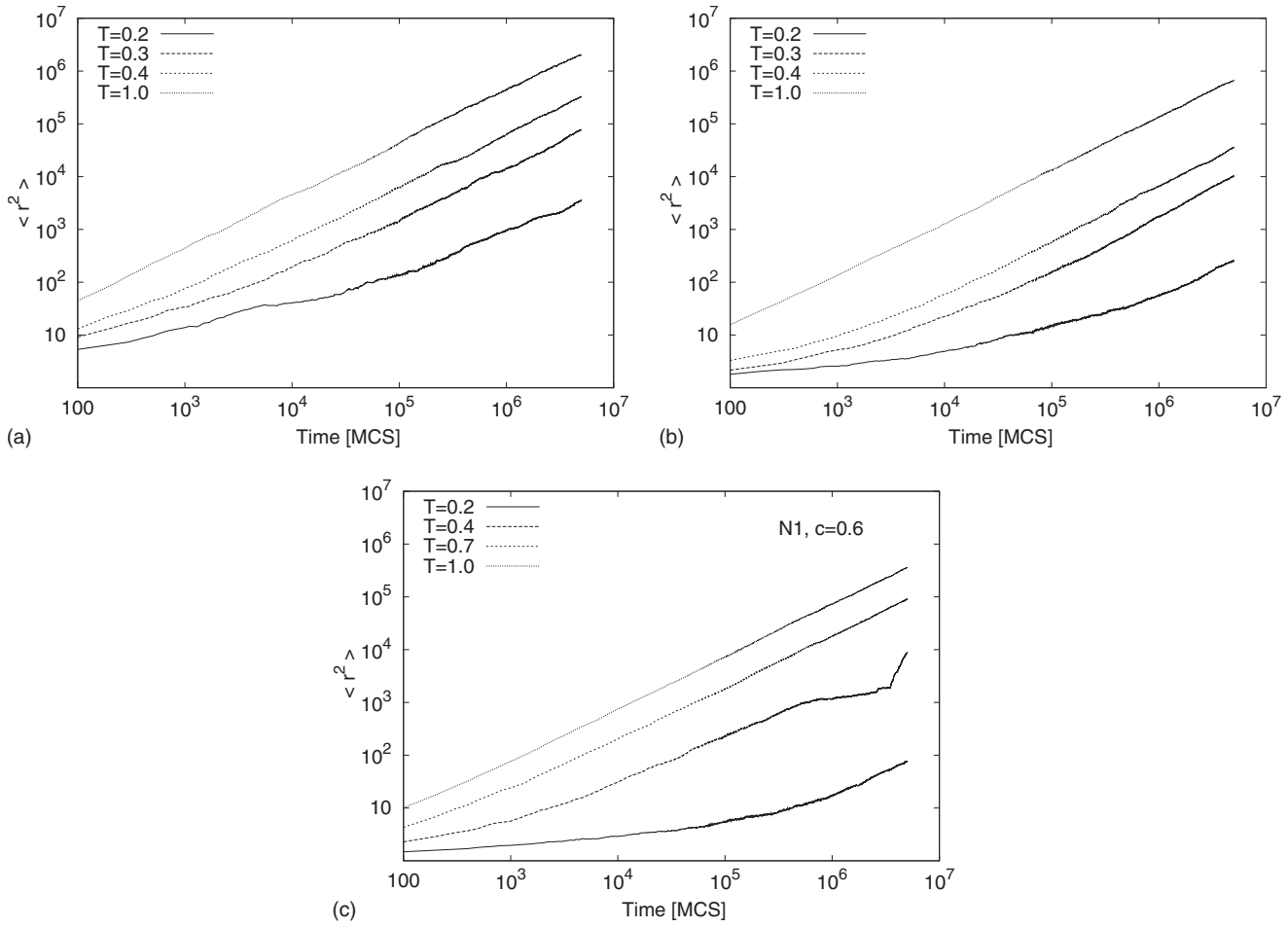

FIG. 2. Time dependence of the average distance (in lattice spacing) covered in time t for interactions to NN and $c=0.2$ (left panel), $c=0.5$ (right panel), and $c=0.6$ (bottom panel).

stripe or the time of its flip as a function of the system's size. Moreover not in all trials were such effects (formation of a blocking cluster or flips) observed, even for small systems. The outcome depends on spatial organization of atoms, which could be different even with the same values of the control parameters. In each case, even on small lattices where there are several flips, we have observed eventually setting of normal diffusion. Therefore our general conclusion is that formation and eventual flipping of domains blocking diffusion in one direction happen only in a transition period before the state of normal diffusion is reached. When this state will be reached depends on the size of the system. For a larger system one could expect the effect of cluster formation and eventual flip to happen later. Therefore in very large systems, such as 500×500, such effects will not be observed because they may happen at very large times, if at all. This is different from the magnetization flips, in the Glauber dynamics, which do not disappear in time. The fact that intermediate temperatures around $T=0.4$ are rather special could be also seen from Fig. 6, which shows the time dependence of the average size of cluster for the case of $c=0.2$, defined as

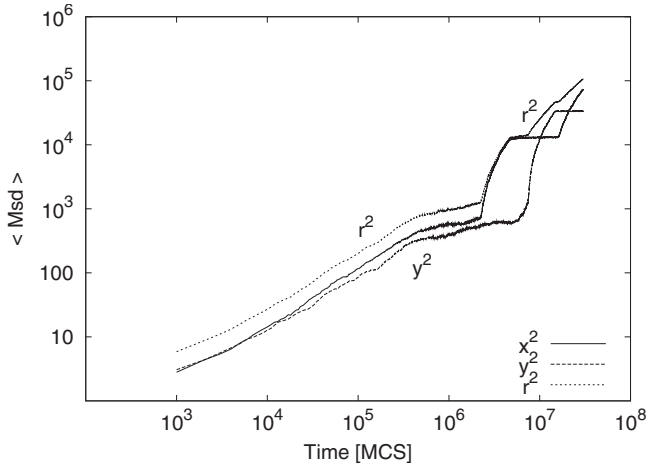

FIG. 3. Time dependence of the MSD along the OX and OY axes (in lattice spacing) covered in time t at $T=0.4$, for $c=0.6$, and interactions to NN. Time of simulations is ten times longer than in bottom panel of Fig. 2.

$$\langle l(t)\rangle = \frac{\sum_{l>10} l n_l(t)}{\sum_{l>10} n_l(t)}, \tag{6}$$

where $n_l(t)$ is the number of clusters of size l at time t. Two particles belong to the same cluster if they are NN or NNN. Following Ref. 19 we registered only clusters of size greater

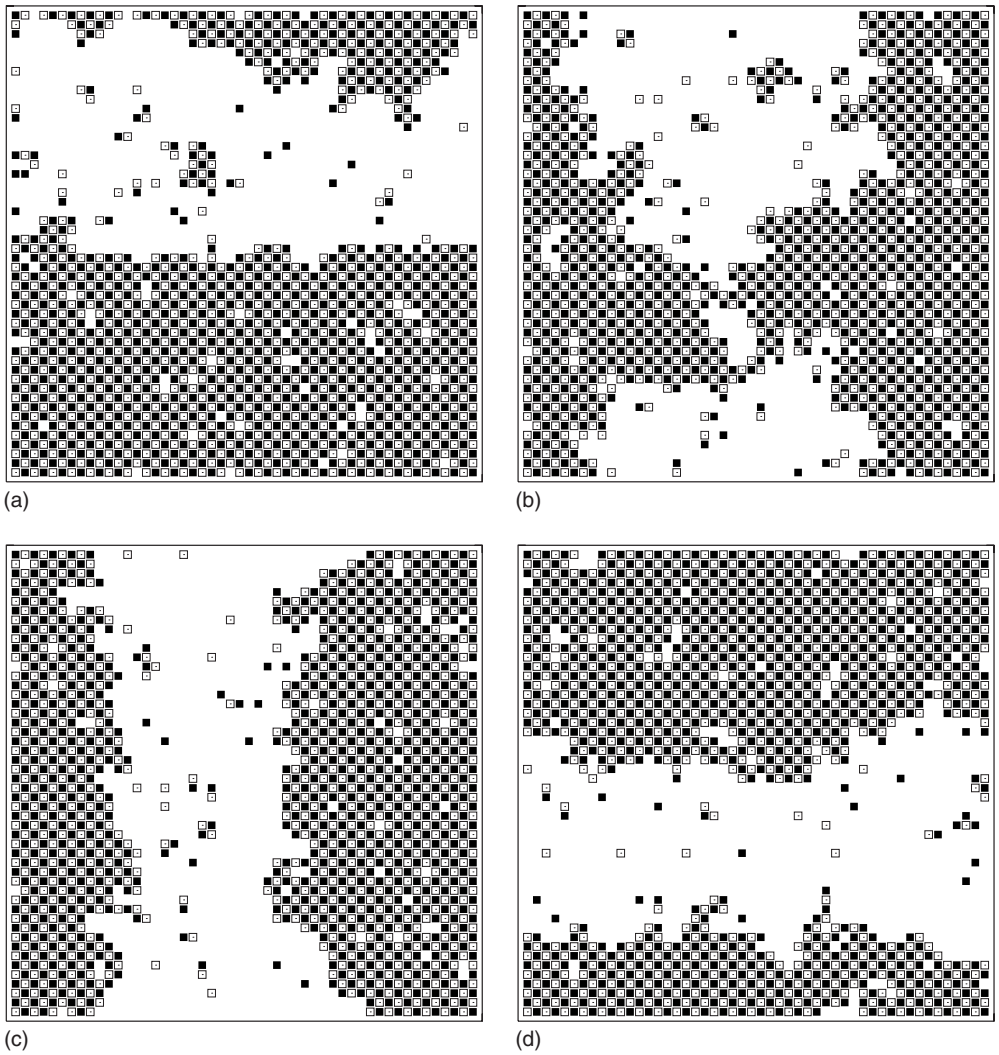

FIG. 4. Spatial arrangement for $c=0.6$ and NN interactions. Time $t=$ (a) 3×10^6, (b) 6×10^6, (c) 8×10^6, and (d) 2.3×10^7 MCS. Particles A are marked by open squares, and particles B by full squares.

than ten. As explained above, for $c\geq0.5$ one big cluster spanning the whole lattice is formed. Hence, plots showing dynamics of cluster formation would be useless in these cases. An interesting feature could be noticed in Fig. 6. At low temperatures small clusters prevail and the average size steadily grows, remaining, however, rather restricted. Also for $T\geq0.7$ we have only very small clusters. In both extremes there are no large scale fluctuations, which are observed at $T=0.4$. Here large clusters form and dissolve at a high rate, and the system is in a very dynamical mode. This property does not manifest itself in the $\langle r^2(t)\rangle$ dependence.

Equation (3) could be used to find out the dependence of the tracer diffusion coefficient D^* on the inverse temperature. The plots in Fig. 7 show that, at high coverage exceeding $c=0.5$, values of D^* are smaller and the data are more scattered. It is the effect of formation of a "percolating" cluster and asymmetric diffusion.

It is rather difficult to compare directly our plots with the ones obtained by UG (Ref. 15) since the models are different in many aspects. However, the basic features of Fig. 7 resemble those obtained by UG for attractive NN interactions.

B. Interactions to NN and NNN

Now we keep the interactions to NN as before but we add also weaker (by a factor of $1/\sqrt{2}$) interactions to NNN. We have presented in our previous paper[14] spatial patterns appearing in this case. Here we shall investigate the diffusion properties of the system. Figure 8 presents time dependence of the mean-square displacement for three values of the coverage: $c=0.2$, 0.5, and 0.6, respectively. At low coverage ($c=0.2$) and low temperatures ($T=0.2$) (left panel of Fig. 8), we observe that, after about 10^4 MCS, $\langle r^2(t)\rangle$ could be fitted into a power-law-type dependence [Eq. (5)] with the exponent $\alpha\simeq0.85$, which corresponds to subdiffusion caused by formation of a single domain. For higher temperatures the power-type behavior sets earlier and the values of α grow but still remain below one. Finally, for $T=0.5$ diffusion has a normal character and $\alpha=1$ from the beginning. In all cases diffusion is isotropic, $\langle x^2\rangle=\langle y^2\rangle$. For $c=0.5$ (right panel of Fig. 8) there are enough particles in the system to create a pattern covering the whole lattice. If the temperature is low ($T=0.2$) once the pattern is formed, at about 2×10^5 MCS, the particles do not have enough energy to break it; hence

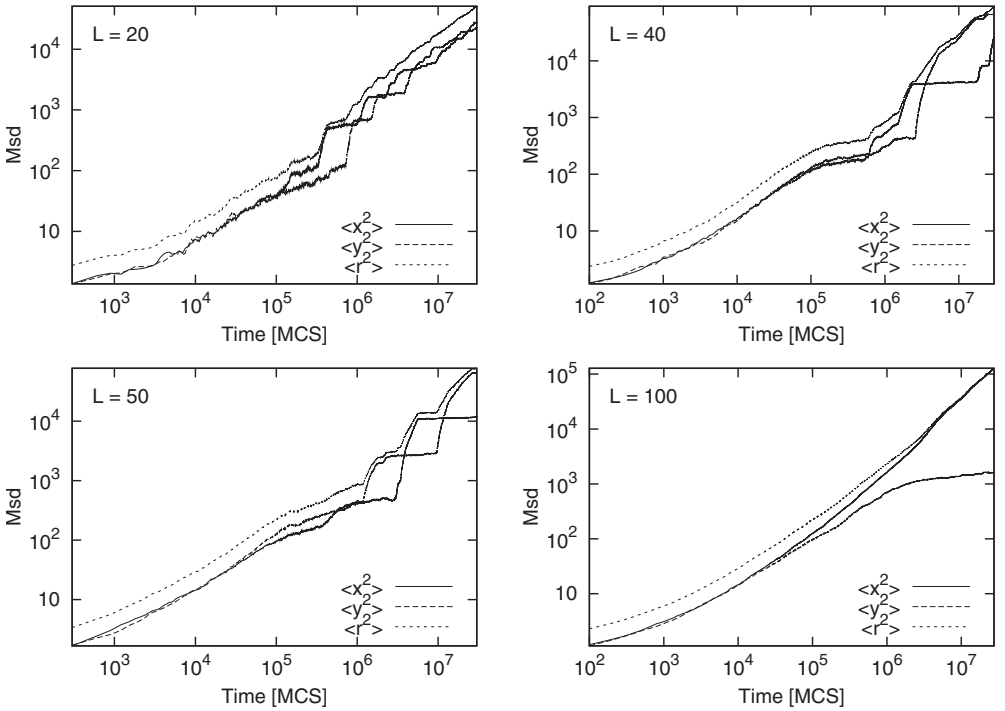

FIG. 5. MSD as a function of time in MCS for different sizes of the system. Changes of the slope of x^2 and y^2 mark formation and change in orientation of single cluster. $T=0.4$, $c=0.6$.

diffusion stops and the value of $\langle r^2 \rangle$ remains quasiconstant. Raising the temperature to $T=0.4$ gives them a chance to get away from fixed positions but $\langle r^2(t) \rangle$ could not be reasonably well fitted into a power-type dependence. Only at high temperatures, $T \geq 0.7$, normal diffusion is observed. When the coverage exceeds 0.5, such as $c=0.6$ shown in the bottom panel of Fig. 8, apart from a stable pattern there are also "free" particles for which there is no room in the pattern observed at $c=0.5$. Therefore, even at $T=0.2$ we see a slow (sub)diffusion. For higher temperatures, as before, normal diffusion is recovered. For $T=0.2$ and $c=0.3$ a single cluster (strip) analogous to the one presented in Fig. 4 is formed. By blocking long trajectories in one dimension, it introduces anisotropic diffusion (see Fig. 9). For higher coverages a more symmetric structure is formed and the anisotropy disappears like for higher temperatures.

In Fig. 10 we present the Arrhenius plot for interactions up to NNN and three values of the coverage. General features are quite similar to the case of NN interactions (see Fig. 7); however, the curves are more smooth since the diffusion is symmetric with respect to the X and Y directions. These plots show similar character as the curves derived by UG (Ref. 15) for two types of interactions.

Dependence of the average cluster size on time for $c=0.2$ is shown in Fig. 11. The main difference with respect to the case of NN interactions is that, at low temperatures ($T=0.2$) and for NNN interactions, the average cluster size is about 500 and it remains stable, while for NN interactions it

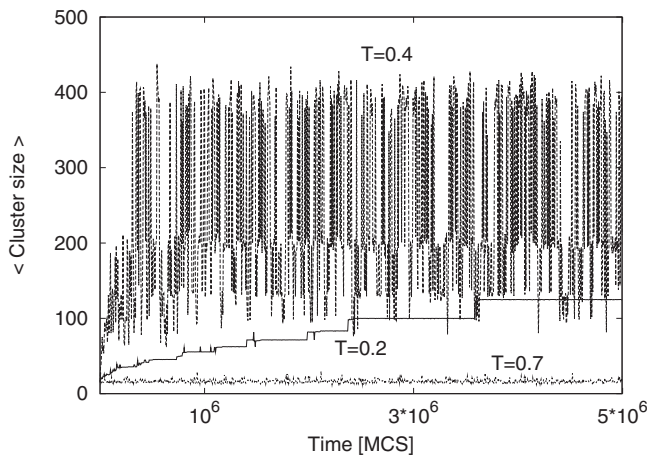

FIG. 6. Average cluster size versus time for $c=0.2$ and NN interactions.

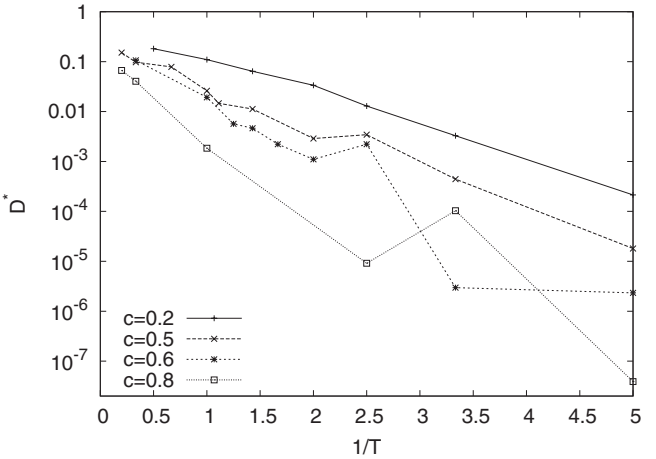

FIG. 7. Tracer diffusion coefficient D^* versus inverse temperature.

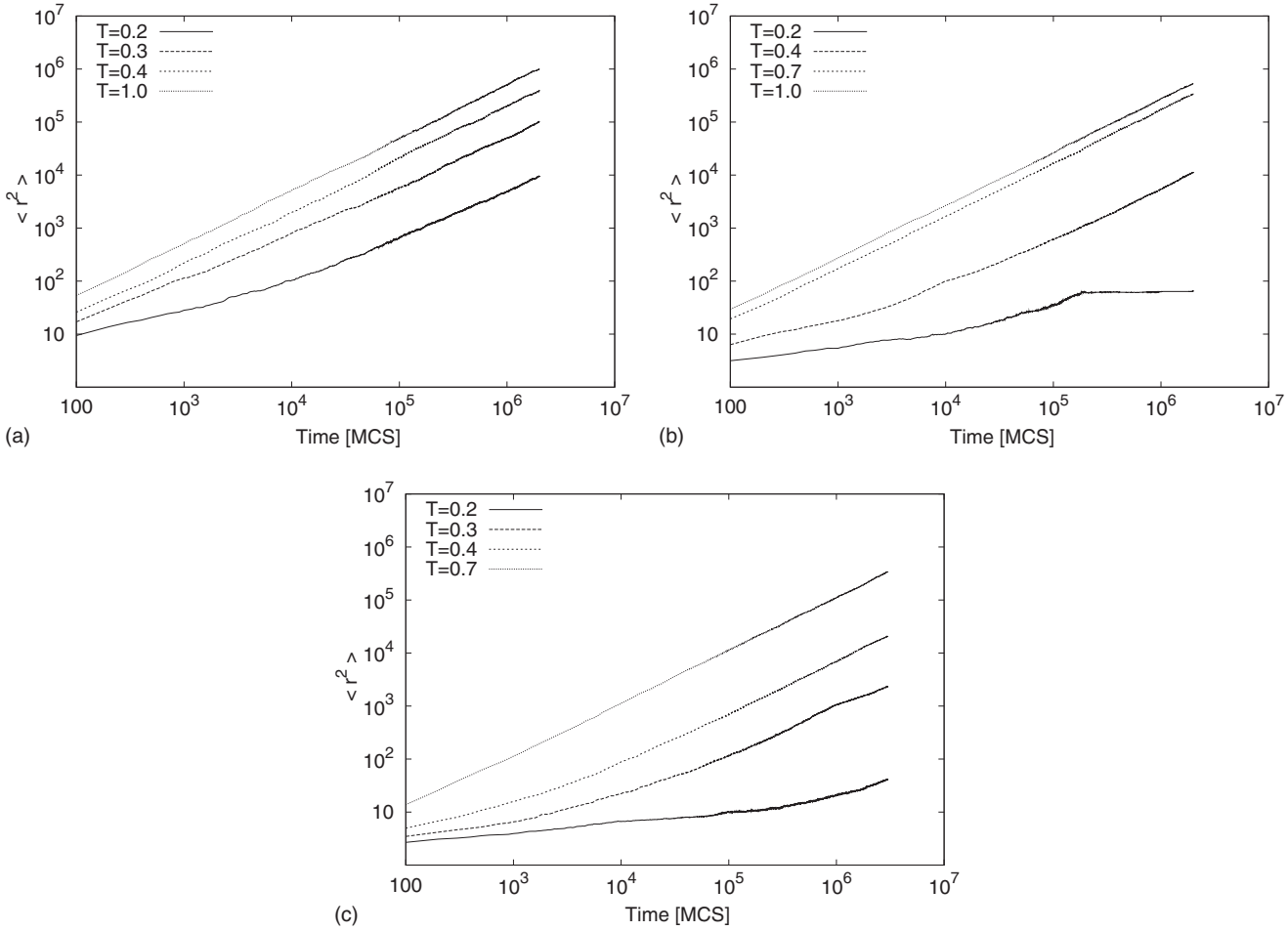

FIG. 8. Time dependence of the average distance covered in time t and interactions to NNN for $c=0.2$ (left panel), for $c=0.5$ (right panel), and for $c=0.6$ (bottom panel).

grows in time and at the end of simulations (5×10^5 MCS) has the value of 120. This is natural since additional interactions should, at least in low temperatures, lead to a faster spatial organization of particles. Fluctuations of patterns are more effectively suppressed by interactions of longer range. At intermediate and high temperatures the range of interactions does not play an essential role.

From the Arrhenius plots it is possible to estimate the activation energy[15] as a function of coverage, using Eq. (4). The results for the two considered cases—NN and NNN interactions—are shown in Fig. 12. The values of E_a are

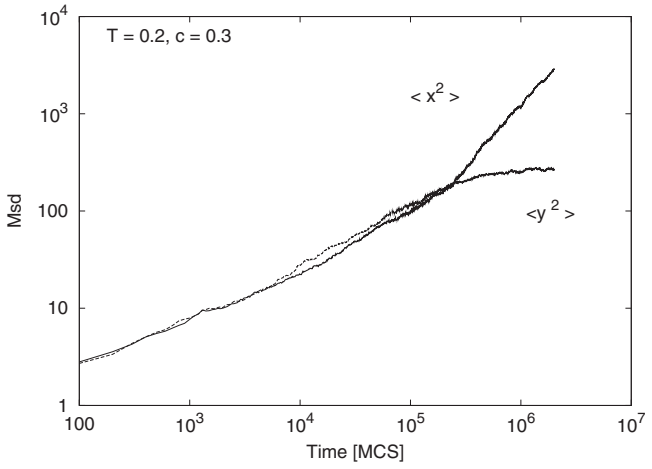

FIG. 9. Mean-square displacement along the X and Y axes for the case of interactions to NN and NNN, at $T=0.2$ and $c=0.3$. For higher and lower coverages, the anisotropy disappears.

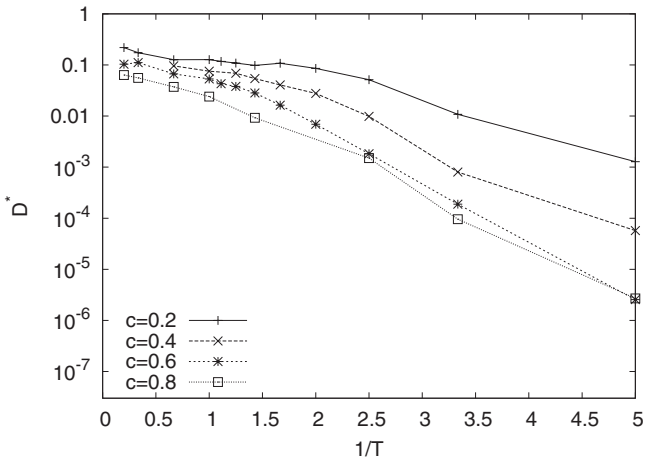

FIG. 10. Tracer diffusion coefficient D^* versus inverse temperature for interactions to NN and NNN.

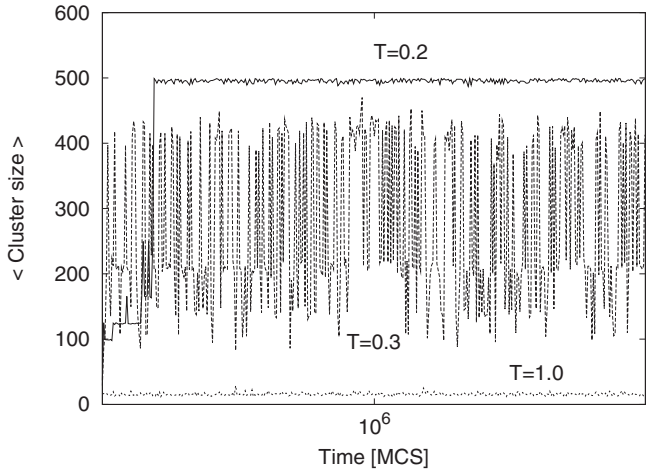

FIG. 11. Average cluster size versus time for $c=0.2$ for interactions to NN and NNN.

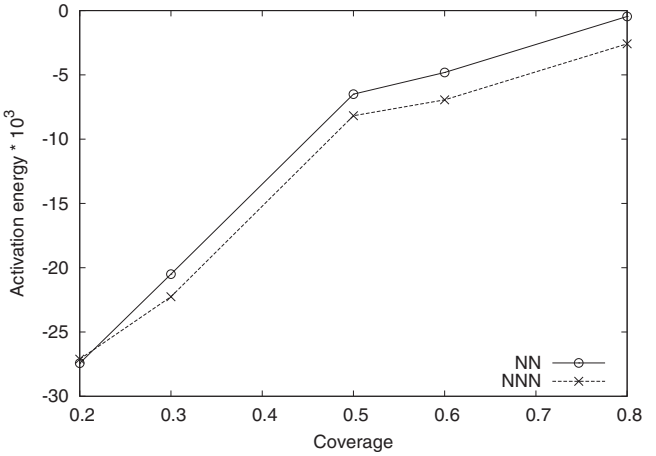

FIG. 12. Activation energy E_a versus coverage for interactions to NN and NNN.

negative, indicating that the repulsion interactions are dominating.[15] One could also notice that the shape of the curves does not depend on the range of interactions. The character of the activation energy dependence on coverage is quite different from the one reported by UG.[15] This is not surprising since, as could be seen from comparison of the snapshots in Refs. 15 and 14, spatial configurations are quite different due to different character of the acting forces.

We turn now to investigation of differences between lattices with odd and even number of sites. All previously reported results were obtained for lattices with even number of nodes in columns and rows. Will the patterns remain the same if we increase the size of the lattice from, say, 50 to 51? For interactions to NN and at low coverages, below $c=0.5$, we do not observe any changes. At higher densities differences do occur. For interactions restricted to NN we observe an "antiferromagnetic" structure (see Fig. 4). If there are no vacancies for $L=51$ then the system (with periodic boundary conditions) would be frustrated. Since there are vacancies, empty space appears, making a kind of an irregular cross (periodic boundary conditions are in both X and Y directions). This enables the system to avoid frustration (see Fig. 13). Generally the cross arms (empty spaces) have odd number of sites and the ions facing each other across the empty space have opposite charges. Sometimes the arms have even number of sites and then the facing atoms have the same charge. Such long-range organization, despite forces acting only to NN or NN and NNN, is possible because, as we have shown earlier,[14] correlations act on much longer distances. This organization of ions across the empty space prevents gluing of a diffusing ion and filling the gap. Sometimes instead of a cross a diagonal (never horizontal or vertical) stripe is formed. Change in the cluster shape is realized when an empty space appears inside the cluster. A part of it could break away, could be reorganized in the process of diffusion of individual particles, and then the small cluster could be glued again, probably at a different place, into a main one. On lattices with odd number of nodes there are no blocked directions, therefore diffusion has normal character (see Fig. 14). For interactions to NN and NNN the picture is roughly

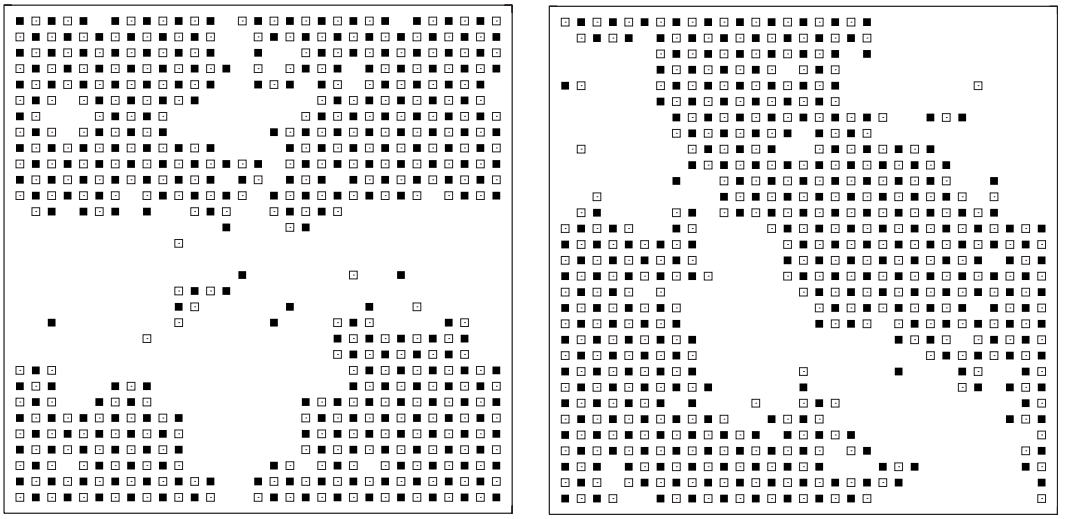

FIG. 13. Typical patterns on a lattice with odd number of nodes. $T=0.4$, $c=0.6$.

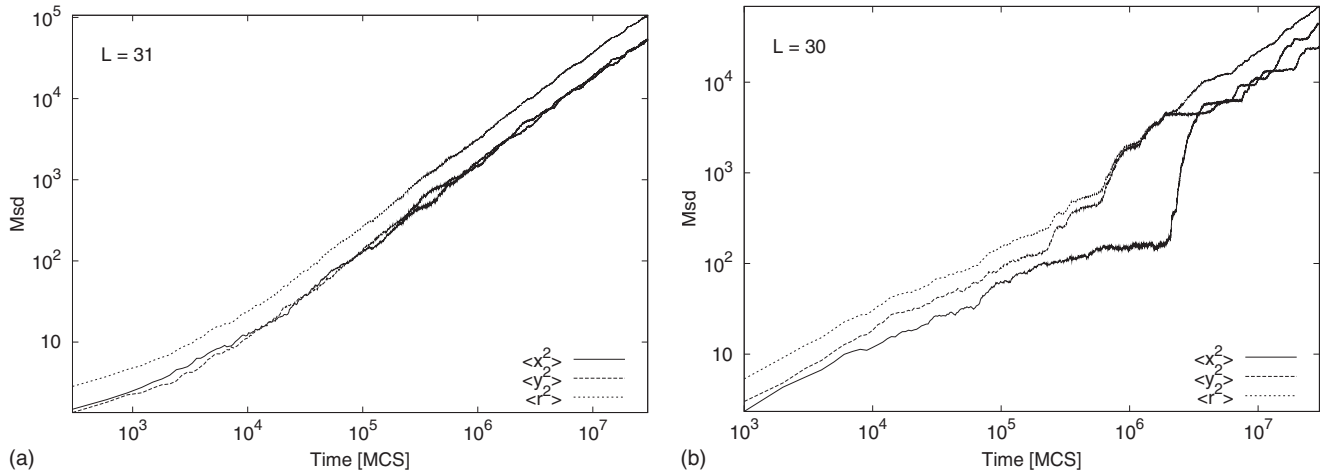

FIG. 14. Mean-square displacement along the X and Y axes for a lattice with odd (left panel) and even (right panel) number of nodes for the same parameters—interactions to NN, $T=0.4$, and $c=0.6$.

the same—for $L=51$ anisotropic diffusion disappears and is replaced by a normal one.

IV. CONCLUSIONS

In this paper we have investigated and discussed dynamic characteristics of a system of two types of oppositely charged particles and vacancies. The same type of particles repel each other while particles of different type attract themselves. Interactions are screened; hence the resulting potential between two particles is exponentially falling off with the distance.[22] Particles could move to an empty NN site following the standard Metropolis algorithm. At low temperatures we have observed either subdiffusion or the mean-square displacement could not be presented as a power-type dependence. At high temperatures diffusion had normal character, regardless of the coverage or interaction range. The most interesting, as is often the case, was the region of intermediate temperatures. For NN interactions, even at rather low ($c=0.1$) coverage, a single cluster is formed, spanning the whole lattice, either in the X or Y direction. It blocks diffusion in the direction perpendicular to it but allows for costless gliding along the border of the cluster. This kind of behavior is visible as a strong anisotropy of $\langle x^2(t)\rangle$ and $\langle y^2(t)\rangle$. The situation is, however, not stable and after some time the big cluster may break down into several smaller ones. They, in turn, could coalesce again into one cluster but this time it could be oriented along another axis. The effect is better visible in small ($L=20-40$) systems. For larger ones it appears at later times and also the chances that a flip in the cluster orientation will occur go down with the size of the lattice. After sufficiently long time the diffusion returns to normal. Blocking of the diffusion in one direction is therefore a temporary effect, which, however, for large systems could last very long. This type of asymmetric diffusion caused by formation of a single domain spanning the lattice in one direction could also happen, and at lower coverages, if the interactions are extended to second neighborhoods. At higher coverages and sufficiently low temperatures, a perfect pattern appears and the particles remain in their positions. As a result we observe a large slowing down of diffusion. Patterns and diffusion characteristics depend to a large extent on whether the model system size has even or odd number of nodes. For lattices with odd number of sites, only normal diffusion is observed since avoiding frustration prevents formation of a single cluster reducing diffusion. The Arrhenius plots for NN and NNN interactions have similar character, and agree with the results reported by UG,[15] which means that such plots are not sensitive to, even large, modifications of the models. Average cluster size does not depend much, as could be expected, on the range of forces in the region of high temperatures. At low temperatures the process of cluster formations is faster for NNN interactions and the average value stabilizes sooner. For NN interactions it takes much longer time to reach this state. At intermediate temperatures we observe, for both types of interactions, very fast processes of formation and splitting of clusters. Finally, the activation energy as a function of coverage does not depend in any essential way on the range of acting forces.

The model presented here is a simple one and extends, probably quite interestingly, like longer-ranged interactions; different types of A and B particles, asymmetric interactions, etc. are of course possible. This would however greatly complicate the model and we believe that, before embarking on such research, good understanding of the basic mechanisms is needed. Such an insight is better deduced from simple models.

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