

Transport-driven reorientation in a square lattice-gas model

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We study the effect of an electric field on the rearrangement in a half-filled square lattice gas with repulsive first- and second-neighbor interactions. Below T_c the particles form parallel chains resulting in anisotropic conduction. Monte Carlo simulations show that these chains prefer the orientation parallel to the applied field. The reorientation time strongly depends on the temperature and the magnitude of electric field. A simple mean-field approximation suggests that the steady-state distribution is modified by the field component perpendicular to the chains.

In the last decades extensive progress has been made in our understanding of diffusion and conduction in concentrated lattice-gas models. Recently several lattice-gas models have been investigated which exhibit anisotropic diffusion and conduction.¹⁻³ The common feature of these models is the existence of a symmetry-breaking particle distribution pointing out a preferred direction for the particle motion. These symmetry-breaking, ordered particle distributions are degenerated, therefore, the preferred direction depends on which state is actually realized. This situation raises a question, whether an applied electric field generating a transport is able to prefer one of the degenerated states and which one.

To demonstrate the effect of the electric field on the particle redistribution a series of Monte Carlo simulations has been carried out in a two-dimensional (2D) lattice gas discussed in detail by Sadiq and Binder.¹ The choice of this model was motivated by the fact that this is the simplest model which exhibit anisotropic diffusion induced by the order. The model is described by a configuration $\{n\} = (n_1, \dots, n_N)$, with n_i either 0 or 1, defined on an $L \times L = N$ square lattice with periodic boundary conditions. Assuming repulsive interactions of equal strength between the first and second neighbors, the energy is given by the Hamiltonian

$$H = \sum_{nn, nnn} n_i n_j, \quad (1)$$

where the summation runs over the nearest-neighbor and next-nearest-neighbor pairs. Our study is restricted to a fixed number of particles corresponding to a half-filled lattice.

The system undergoes an Ising-like phase transition at a critical temperature $T_c \approx 0.55$. In the fourfold degenerated ground state the rows (or columns) are alternately occupied or empty. Figure 1 shows a typical particle distribution below T_c ($T=0.51$) where the particles form vertical chains. In this case the diffusion and conduction is higher along the chains comparing to the perpendicular direction. The long-range order may be characterized by dividing the lattice into four sublattices as indicated in Fig. 1 and introducing μ_i ($i=1,2,3,4$), the average sublattice occupations. For example, in the state represented in Fig. 1, $\mu_1 = \mu_4 \approx 0.9$ and $\mu_2 = \mu_3 \approx 0.1$.

The stochastic dynamics of the system consists of jumps

to one of the unoccupied neighboring sites. In the presence of an electric field \mathbf{e} the jump rate $\Gamma(j \rightarrow k)$ from site j to k is given as

$$\Gamma(j \rightarrow k) = \frac{1}{\tau} n_j (1 - n_k) \frac{1}{1 + \exp[\beta(E_{k,j} - \mathbf{e} \mathbf{r}_{k,j})]}, \quad (2)$$

where $E_{k,j}$ is the energy difference between the final and initial states, and $\mathbf{r}_{k,j}$ is the displacement. Here τ is an undetermined time constant which we put equal to unity when measuring times in Monte Carlo steps per particles. In the absence of electric field ($\mathbf{e}=0$) this choice of jump rate is equivalent to the Kawasaki dynamics⁴ satisfying the condition of detailed balance, i.e., the stationary state of the system is the Gibbs equilibrium state at the reservoir temperature $T=1/\beta$. In this Rapid Communication our analysis is concentrated to the effect of electric field parallel to one of the crystallographic axis.

Monte Carlo simulations have been carried out on a 60×60 square lattice with 1800 particles at different electric fields. Our analysis is restricted to a temperature region ($0.48 < T < T_c$) in which the diffusion was anisotropic and high enough to reach equilibrium within the run time. For details of Monte Carlo (MC) simulations see the review by Binder and Stauffer.⁵ Each run was

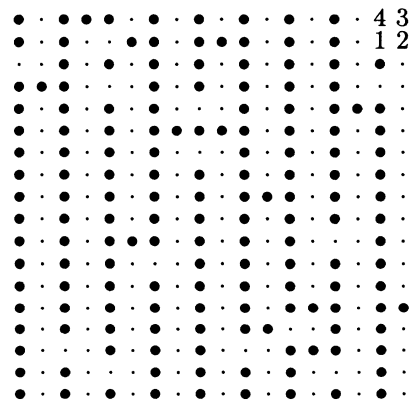


FIG. 1. A typical particle distribution on the square lattice with $\rho = \frac{1}{2}$ for $T=0.51$ and $\mathbf{e}=0$. Figures on the upper right-hand corner indicate the sublattices indices in a (2×2) cell.

started from an ordered ground state in which the particles formed vertical chains ($\mu_1 = \mu_4 = 1$ and $\mu_2 = \mu_3 = 0$). After a thermalization of a thousand of MC steps per particles at a temperature $T < T_c$, the long-range order characteristic to the initial state may be recognized as shown in Fig. 1. We have studied the time evolution of the system when switching on a horizontal [$\mathbf{e} = (e_\perp, 0)$] or vertical [$\mathbf{e} = (0, e_\parallel)$] electric field.

It is found that the electric field component perpendicular to the chains force the chains to be parallel to the field direction while the parallel field does not cause any observable change if $e_\parallel < 0.2$. This reorientation process can be monitored by introducing a direction $\phi = \arctan(b_h/b_v)$ where b_h and b_v denote the number of horizontal and vertical bonds. Figure 2 represents a typical time evolution of ϕ for $T = 0.51$ and $e_\perp = 0.15$. The real time monitoring of the particle distribution illustrates that the reorientation is analogous to the recrystallization including the nucleation and the growth of the preferred domain(s). A similar process was investigated by Binder and Müller-Krumbhaar⁶ in a spin system by a sudden reversal of the external magnetic field below T_c . In this ferromagnetic system the magnetic field drives the reversal of the magnetization via a nucleation process followed by the growth of the preferred domains. In the present lattice-gas model, however, the driving force of the reorientation is not evident because our understanding of the statistical physics in open systems is far from being satisfactory.

Introducing the concept of reorientation time t_r as indicated in Fig. 2 a series of MC simulation was carried out varying e_\perp at fixed temperature $T = 0.51$. The logarithm of the average values of t_r vs e_\perp are plotted in Fig. 3 where error bars demonstrate the standard deviation of data determined from ten runs. With increasing electric field a drastic decrease in the reorientation time and in the standard deviation has been found.

Increasing the temperature the reorientation process becomes faster at a fixed value of electric field. This effect may be explained by the higher diffusion and by the increasing role of fluctuations. Namely, the appearance of the preferred domains having sufficiently large size is more probable at higher temperatures. The separation of these two effects in the reorientation process requires a more systematic investigation.

The reorientation process shown in Fig. 2 is extremely fast in the macroscopic time scale. Because of the technical requirements of MC simulation the selected values of \mathbf{e} have to be also extremely large. The existence and the dynamics of the reorientation process may strongly modify the interpretation of the anisotropic conductivity mea-

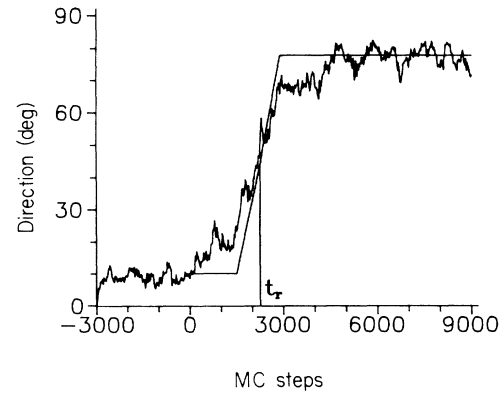


FIG. 2. Time evolution of the direction ϕ in a typical run for $T = 0.51$, $L = 60$. In the initial state the particles were arranged in vertical chains separated from each other by empty columns, the horizontal electric field ($E_\perp = 0.15$) was switched on at MCS = 0. The reorientation time t_r is given in MCS unit at the halfway between the initial and final values of ϕ .

surements because the large applied field can change the direction of chains. Unfortunately, from the present results we cannot predict the dynamical behavior in the limit $\mathbf{e} \rightarrow 0$ we are interested in from the experimental point of view.

The present results are compatible with those of Katz, Lebowitz, and Spohn.⁷ Using MC simulations of a 2D lattice gas with nearest-neighbor interactions they found an anisotropic long-range order generated by the electric field. In the low-field limit it was shown that in a lattice with repulsive interaction the phase boundary becomes parallel to the electric field. Extensive research⁷⁻¹¹ has been done on this nonequilibrium phenomenon. For example, in an electric field T_c is decreasing for repulsive interaction and increasing for attractive one. The above investigations are restricted to Ising models with nearest-neighbor interactions where the equilibrium states (or phases) are isotropic. In the present inherently anisotropic model, however, the effect of the parallel and perpendicular electric fields may well be distinguished, as demonstrated by the following simple mean-field approximation.

Using a master equation defined by the Kawasaki dynamics (2) we can determine the time derivative of the average sublattice occupations in mean-field approximation. For example, the time dependence of μ_1 is given by the equation

$$\tau \frac{d\mu_1}{dt} = -\mu_1 \{ (1 - \mu_2) [f(E_{21} - e_1) + f(E_{21} + e_1)] + (1 - \mu_4) [f(E_{41} - e_2) + f(E_{41} + e_2)] \} \\ + (1 - \mu_1) \{ \mu_2 [f(E_{12} - e_1) + f(E_{12} + e_1)] + \mu_4 [f(E_{14} - e_2) + f(E_{14} + e_2)] \}, \quad (3)$$

where $f(z) = 1/[1 + \exp(\beta z)]$ and E_{ij} is the energy difference between the sublattice i and j in mean-field approximation. In this expression we cannot use the concept of parallel and perpendicular direction, therefore, e_1 and e_2 indicate here the horizontal and vertical components of the electric field until we clarify the chain direction. Simi-

lar equations may be derived by cyclic permutation of indices for the other three sublattices. The stationary ($d/dt = 0$) solution of Eq. (3) is not unique, there exists a set of solutions corresponding to the degeneracy of ordered states. One of them describes the effect of electric field when the particles form vertical chains below T_c . In

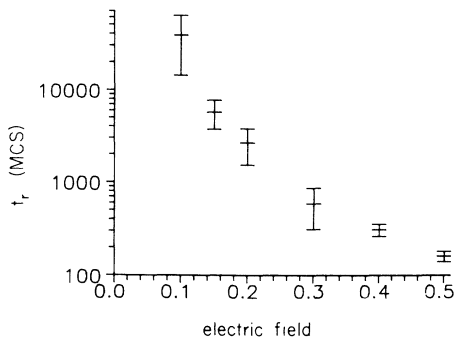


FIG. 3. Reorientation time t_r as a function of perpendicular electric field e_{\perp} for $T=0.51$ and $L=60$. Average values and standard deviations were determined from ten runs.

this case we can assume that $\mu_1=\mu_4=0.5+x$ and $\mu_2=\mu_3=0.5-x$. This assumption yields a particular solution,

$$\left(\frac{1+2x}{1-2x} \right)^2 = \frac{\exp(\beta 8x) [\cosh(\beta e_{\perp}) + \exp(\beta 8x)]}{1 + \exp(\beta 8x) \cosh(\beta e_{\perp})}, \quad (4)$$

which depends on e_{\perp} , the perpendicular component of the electric field while the effect of e_{\parallel} is eliminated. As expected, Eq. (4) reproduces the solution of the equilibrium mean-field approximation at $e_{\perp}=0$. Figure 4 shows the temperature dependence of the average sublattice occupations for $e_{\perp}=0$ and $e_{\perp}=0.5$. It is worth mentioning that in this approximation the critical temperature ($T_c=1$ for $e_{\perp}=0$) is overestimated comparing to the exact result. Figure 4 makes it clear that T_c decreases with increasing e_{\perp} and the change is proportional to e_{\perp}^2 . At fixed temperature e_{\perp} modifies the particle distribution. What is more, at sufficiently large magnitude, it can destroy the ordered state in agreement with the earlier results.^{7,9,11} A more detailed discussion of this mean-field approximation including a comparison with MC simulations will be published later.

The mean-field approximation may suggest some hints for the explanation of the reorientation process. The perpendicular component of the electric field will induce a redistribution of the particles and an increase in configurational entropy and energy. This redistribution will eliminate the degeneracy of the states and makes a distinction between the stable parallel chain domains and the nonstable or metastable perpendicular chain domains. Unfortunately, we cannot determine the difference of a “nonequilibrium free energy” between the parallel and perpendicular phases because of the lack of a consistent theory.

Finally, it is worth mentioning a possibility for the ex-

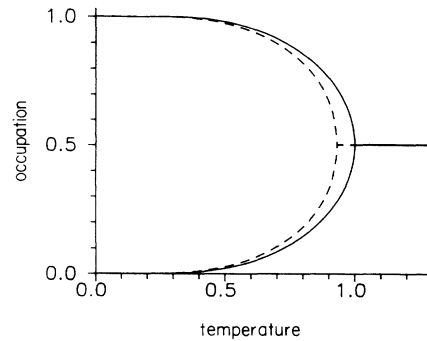


FIG. 4. Temperature dependence of sublattice occupations for $e_{\perp}=0$ (solid line) and $e_{\perp}=0.5$ (dashed line) in mean-field approximation.

perimental realization of the reorientation process found in two dimensions. Our results obtained in a square lattice may involve similar processes in three-dimensional systems because the reorientation process is closely related to the existence of anisotropic diffusion generated by the symmetry-breaking equilibrium arrangement of particles. Recently we have some indications that there exists an anisotropic superionic phase of AgI. It is to be emphasized that the properties of AgI are not understood completely, there are some contradictions between experiments, simulations, and theories. Measuring the specific heat of AgI Perrott and Fletcher¹² discovered two subsequent phase transitions contrary to earlier results.^{13,14} The existence of the second phase transition was justified by Mazzacurati *et al.*¹⁵ and by Aronsson *et al.*¹⁶ with measurement of Raman and Brillouin scattering, respectively. Within the framework of a six sublattice mean-field approximation one of us has proved the possibility of two subsequent phase transitions related to the existence of a partially ordered state¹⁷ exhibiting anisotropic diffusion.³ In molecular dynamic simulations,^{18–21} however, the second phase was not observed except by Tallon.²² The contradictions between different experiments arise from the sample preparations. At the same time the theoretical descriptions (or simulations) are very sensitive to the pair interactions taken into account. If the diffusion in the superionic phase of AgI below 700 K is really anisotropic as a consequence of the long-range order proposed by the mean-field theory then it may be considered a specimen for such experiments. Besides, this effect might be utilized to have more knowledge on the superionic phases of AgI. The present work, in fact, was inspired by this hope.

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¹A. Sadiq and K. Binder, *Surf. Sci.* **128**, 350 (1983).

²R. Kutner, K. Binder, and K. W. Kehr, *Phys. Rev. B* **28**, 1846 (1983).

³G. Szabó and J. Kertész, *J. Phys. C* **19**, L273 (1986).

⁴K. Kawasaki, *Phys. Rev.* **145**, 224 (1966).

⁵K. Binder and D. Stauffer, in *Applications of Monte Carlo*

Methods in Statistical Physics, edited by K. Binder, Topics in Current Physics Vol. 36 (Springer-Verlag, Berlin, 1983), p. 1.

⁶K. Binder and H. Müller-Krumbhaar, *Phys. Rev. B* **9**, 2328 (1974).

⁷S. Katz, J. L. Lebowitz, and H. Spohn, *Phys. Rev. B* **28**, 1655 (1983); *J. Stat. Phys.* **34**, 497 (1984).

- ⁸H. van Beijeren and L. S. Schulman, *Phys. Rev. Lett.* **53**, 806 (1984).
- ⁹J. L. Vallés and J. Marro, *J. Stat. Phys.* **43**, 441 (1986); **49**, 89 (1987); J. Marro and J. L. Vallés, *J. Stat. Phys.* **49**, 121 (1987).
- ¹⁰R. Dickman, *Phys. Rev. A* **38**, 2588 (1988).
- ¹¹K.-t. Leung, B. Schmittmann, and R. K. P. Zia, *Phys. Rev. Lett.* **62**, 1772 (1989).
- ¹²C. M. Perrott and N. H. Fletcher, *J. Chem. Phys.* **48**, 2681 (1968); **50**, 2770 (1969); **52**, 3368 (1970).
- ¹³W. Jost, H. J. Oel, and G. Schniedermann, *Z. Phys. Chem.* **17**, 175 (1958).
- ¹⁴J. Nölting and D. Rein, *Z. Phys. Chem.* **66**, 150 (1969).
- ¹⁵V. Mazzacurati, G. Ruocco, G. Signorelli, E. Cazzanelli, A. Fontana, and G. Mariotto, *Phys. Rev. B* **26**, 2216 (1982).
- ¹⁶R. Aronsson, L. Börjesson, and L. M. Torrell, *Solid State Ionics* **28-30**, 204 (1988).
- ¹⁷G. Szabó, *J. Phys. C* **19**, 3775 (1986).
- ¹⁸W. Schommers, *Phys. Rev. Lett.* **38**, 1536 (1977); *Phys. Rev. B* **17**, 2057 (1978); **21**, 847 (1980).
- ¹⁹M. Parrinello, A. Rahman, and P. Vashishta, *Phys. Rev. Lett.* **50**, 1073 (1983).
- ²⁰G. L. Chiarotti, G. Jacucci, and A. Rahman, *Phys. Rev. Lett.* **57**, 2395 (1986).
- ²¹Y. Kaneko and A. Ueda, *Phys. Rev. B* **39**, 10281 (1989).
- ²²J. L. Tallon, *Phys. Rev. B* **38**, 9069 (1988); *Solid State Ionics* **28-30**, 53 (1988).