Neighboring interaction effect on surface diffusion using Monte Carlo simulation

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We are interested on studying diffusion process of a half occupied clean surface. Our calculations are performed in the framework of lattice gas model using Monte Carlo simulation. We analyze the behavior of the relevant physical quantities as diffusion coefficient tracer and specific heat. We consider first JFN and second JSN neighboring repulsive interactions with the ratio R = J_{SN} / J_{FN} in the [0.1] range. We show that the second repulsive neighboring interactions induce an order/disorder transition for some critical values of R that affect the surface diffusion mechanism.

1. Introduction

Diffusion process in layered systems is still a typical question [1-7] as it is present in many different phenomena such as adsorption/desorption [8,9], crystal growth or catalytic reactions [10,11]. Effort has been devoted to the understanding of the dynamics of this process because of its technological importance. Investigations deal with experimental and theoretical study. Scanning tunneling microscopy [12,13] as well as field microscopy technics [14-16] have shown that effects of lateral interactions on diffusion process are relevant for the establishment of ordered structures.

Theoretical studies of the dynamics of diffusion process in systems undergoing phase transition suffer from the absence of a systematic method that takes into account time evolution of cooperative phenomena. Only the crude approximation named mean field approach allows analytical progress [17-19] that helps in understanding the studied phenomenon at the price of overestimating the critical regions. For the convincing works are no doubt those using numerical methods especially the stochastic Monte Carlo simulation of diffusion in two-dimensional surface. The latter was successfully used in many different situations [20-22]. However the dynamics of diffusion with the presence of ordering in the case of first and second neighboring repulsive interactions has been omitted even its potential technological applications. In fact, experimental studies of some layered crystals such as K_2CoF_4 and Rb_2CoF_4 [23-25] clearly show anisotropic antiferromagnets suggesting the presence of the mentioned interaction regime. This is the aim of the present work.

From simple physical consideration, it is intuitively expected that repulsion between adatoms accelerates surface migration. In contrast, attractive interactions should inhibit adatom diffusion. These simple rules qualitatively describe the characteristics in many adsorption systems. However, more sophisticated arguments are required for the description of surface diffusion in case of phase transition when strong lateral interactions force the system to order near a critical temperature [25,26]. In the present work, we study statics and dynamics proprieties of the adparticles on a square lattice using Monte Carlo method.

We consider a regular clean surface where adsorbate particles can perform activated jumps. Our calculations are limited to a half-filling lattice ($\theta = 0.5$). Interaction is extended to second neighboring. The ratio $R = J_{SN} / J_{FN}$ between first and second nearest neighbors is in the $[0,1]$ interval. This study is performed in the canonical ensemble and deals with effects of the critical behavior on diffusion process.

The paper is organized as follows: In the next section, we present the lattice gas model. In section 3, we describe technic and the algorithm used. In section 4, the results are presented. The conclusion is given in the last section.

2. Lattice gas model

The potential minima of the crystalline structure are nodes of a square lattice that can be occupied by adsorbate moving particles. The state of each node "i" is quantified by a Boolean variable "n_i" called occupation number for which the values $"0"$ and $"1"$ denote empty and occupied site, respectively. We consider first neighboring JFN and second neighboring JSN static interactions

between pair of particles. The following Hamiltonian gives the interaction energy:

$$H = -\frac{1}{2}J_{FN}\sum_{\langle i,j\rangle} n_i n_j - \frac{1}{2}J_{SN}\sum_{\langle\langle i,k\rangle\rangle} n_i n_k - \mu\sum_i n_i \quad (1)$$

The symbols $\langle...\rangle$ and $\langle\langle...\rangle\rangle$ represent nearest and next nearest neighboring sites respectively and μ is the chemical potential. JFN is taking to be negative to represent repulsive nature of the interaction.

For a conserved concentration of the lattice there are so many different possible configurations. Let $P(\{n\},t)$ be the probability of finding the configuration $\{n\}$ at time t. Time evolution of the lattice consists on change of configurations as a consequence of particles hope to empty sites. Focusing the desired configuration there are many jumps that contribute positively to its establishment and others that just do the opposite. This is formally expressed by the phenomenological master equation hence defined [26]:

$$\frac{\partial}{\partial t}P(\{n\},t) = \sum_{\{n'\}}\left[W(\{n'\},\{n\})P(\{n'\},t) - W(\{n\},\{n'\})P(\{n\},t)\right]$$

(2)

$W(\{n'\},\{n\})$ is the transition frequency from $\{n\}$ to $\{n'\}$ and should include the physics of the system. Hence it is a function of the Hamiltonian H. Equation (2) can be seen as a continuity equation expressing that total probability is conserved at all times. The master equation has been solved for the unique case of one particle diffusing on uniform regular lattice [27]. Transition rate was taking to be of Arrhenius-like form as jumps are promoted by thermal activation. The equation has been diagonalized in reciprocal space and led to Lorentzian behavior whose width is related to quasi-elastic incoherent neutron scattering [28]. A part from this case the master equation is completely non-linear and is suitable only for numerical solution when the studied phenomenon can be assimilated to a Markovian process i.e. there is complete loss of history and time evolution is only determined by the state at present time. Equilibrium state is reached when transition between two configurations is made without loose of energy. Resulting expression is called detailed balance condition.

$$W(\{n'\},\{n\})P_{eq}(\{n'\}) = W(\{n\},\{n'\})P_{eq}(\{n\}) \quad (3)$$

Where $P_{eq}(\{n\})$ is given by

$$P_{eq}(\{n\}) = \frac{\exp(-\beta\Delta H)}{Z} \quad (4)$$

β denotes the inverse reduced temperature and ΔH corresponds to the energy difference between the final and the initial configuration. We recall that in the metropolis algorithm [31,32] a site is allowed to change its state if the global energy of the system is reduced or a response to a local deformation of the lattice represented by the comparison of the adsorption/desorption length to a number chosen at random. Z denotes the partition function and is unknown but luckily cancelled by taking the ratio of individual probabilities relative to each configuration of the Markov chain states. As there is not unique choice of the transition probability any expression written versus energy difference is acceptable.

3. Method and algorithm

Transition rate is chosen according to Metropolis algorithm [29]:

$$W = \min(1, \exp(-\beta\Delta H)) \quad (5)$$

For this algorithm any elementary process is realized once the finale state contributes to the stability of the system. Hence, it accepts the configuration changes that lower energy of the system. Otherwise, occurrence of the process is conditioned by the comparison of the transition rate to a random number. Metropolis algorithm suffers from allowing adsorption/desorption processes for every attempt in the high temperature limit leading to an oscillation between states of the system. Ergodicity principle is then violated. But this has no effect on the temperature range of interest in this study, a periodically bounded bidirectional lattice of square geometry and lateral size L is considered. Tendency towards equilibrium is done by means of metropolis algorithm. The specific heat is defined as:

$$C_V = \beta.(\langle E^2\rangle - \langle E\rangle^2) \quad (6)$$

E denote the system energy and β represents the inverse reduced temperature. Diffusion process is investigated by computing the diffusion coefficient tracer D^*. The latter is defined as the mean square displacement of tagged particles [33]:

$$D^* = \lim_{t\to\infty}\frac{1}{2Ntd}\sum_{i=1}^{N}\left\langle\left|r_i(t) - r_i(0)\right|^2\right\rangle \quad (7)$$

where d is the system dimension $(d = 2)$, t is the elapsed time expressed in units of Monte Carlo step (LXL random interrogations) and $\left|r_i(t) - r_i(0)\right|$ is expressed in unity of the lattice constant[31].

4. Results and discussion

To establish critical values for R, we compute the specific heat. Their effects on diffusion process are deduced from calculating the tracer diffusion coefficient.
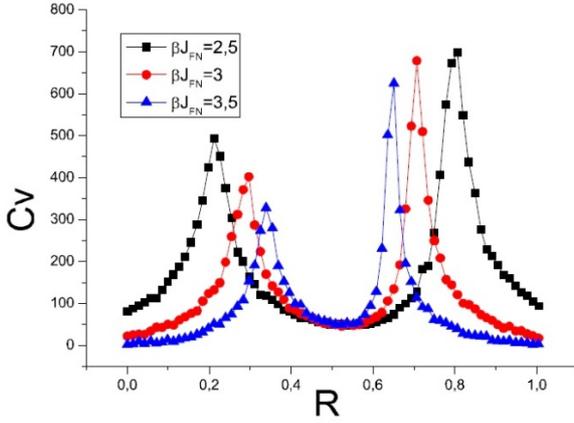
Fig. 1. Specific heat versus R at different values of invers reduced temperature and $\theta = 0.5$.

In Fig. 1 we plot the curve representing the specific heat C_V as a function of R, the ratio between second J_{SN} and first J_{FN} nearest neighbors. We note that each graph presents two peaks for different values of R. The first peak appears at a value of R lower than 0.5, which it shows that our system goes through a phase transition. This phase is characterized by an ordered structure such $\theta(2X2)$.

Second peak is reached when R is greater than 0.5. In this case, our system also passes through a phase transition, but the structure is $\theta(1X2)/\theta(2X1)$ ordered type. The peaks found, agree with the phase transition diagram of reference [34].
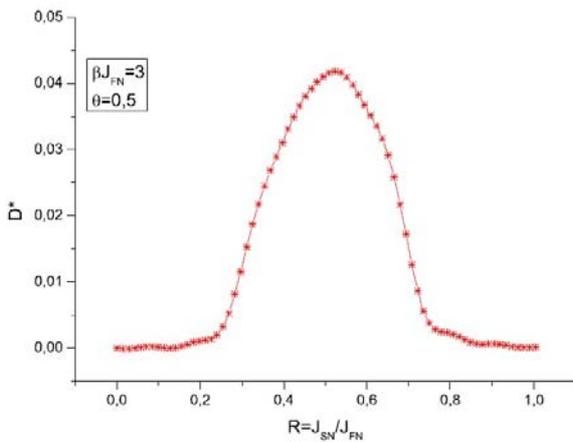
Fig. 2. Tracer diffusion coefficient versus R at $\beta J_{FN} = 3$ and $\theta = 0.5$.

Fig. 2 represents the evolution of the tracer diffusion coefficient for only one value of temperature used in Fig. 1 $(\beta J_{FN} = 3)$. We note that the graph of Fig. 2 presents two inflection points. The study of the function derivative from each curve, reveals the coordinates of each point inflexion (Fig. 2,3). This allowed us to identify critical properties of our system.
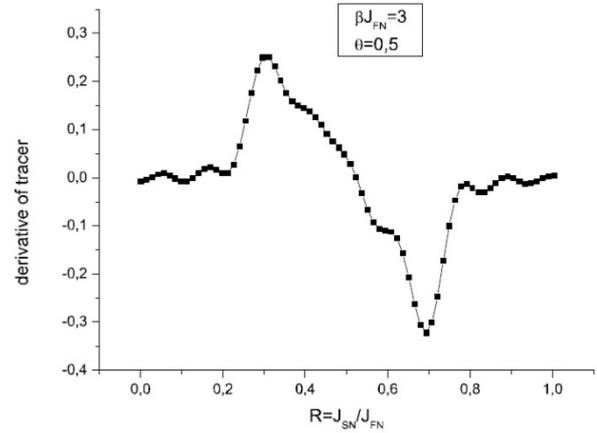
Fig. 3. Derivative function of tracer diffusion coefficient versus R at $\beta J_{FN} = 3$ and $\theta = 0.5$.

5. Conclusion

In this work, we are interested on studying diffusion process of a half occupied clean surface. We analyze the behavior of the tracer diffusion coefficient, and specific heat. We consider first J_{FN} and second J_{SN} neighboring repulsive interactions with the ratio $R = J_{SN} / J_{FN}$ in the [0.1] range, and show that the second repulsive neighboring interactions induce an order/disorder transition for some critical values of R that affect the surface diffusion mechanism. This allows to note that repulsive interactions are not always favorable to diffusion process, but can lead to ordering on the lattice.

References

[1] A. Sadiq, K. Binder, Surf. Sci. **128**, 350 (1983).
[2] M. Biehl, M. Ahr. Europhys. Lett., **53**(2), 169 (2001).
[3] A. A. Chumak, C. Uebing, Eur. Phys. J. B **17**, 713 (2000).
[4] Ya. G. Groda, P. Argyrakis, G. S. Bokun, V. S. Vikhrenko, Eur. Phys. J.B **32**, 527 (2003)
[5] A. A. Tarasenko et al. Surf. Sci. **536**, 1 (2003).
[6] M. Masin, Z. Chvoj, P. Jelinek, J. Heinonen. Surf. Sci. **516**, 169 (2002).
[7] T. Ala-Nissila, R. Ferrando, S. C. Ying, Adv. Phys. **51**(3), 949 (2002).
[8] Adsorption and Diffusion, H. G. Karge.J. Weitkamp, Springer **7**, (2008).
[9] D. A. Reed, G. Ehrlich, Surf. Sci. **105**, 603 (1981).

[10] S. C. Singhal, MRS Bull. **253**, 16 (2000).
[11] M. M. Thackeray, J. O. Thomas, M. S. Whittingham, MRS Bull. **253**, 16 (2000).
[12] P. Ebert, M. G. Lagally, K. urban, phys. Rev. Lett. **70**, 1437 (1993).
[13] J. M. Carpinelli, B. S. Swartzentruber, Phys. Rev. B **58**, 13423 (1998).
[14] R. Gomer, Field Emission and Field Ionization, Harvard University Press, Cambridge, (1961).
[15] R. Gomer, in: V.T. Binh (Ed), Surface Mobilities on Solid Materials, Plenum Press, New York p.127, (1981).
[16] R. Gomer, Surf. Sci. 299/300 (1994).
[17] R. Nassif, Y. Boughaleb, A. Hekkouri, J. F. Gouyet, M. Kolb. Eur. Phys. J. **31**, 453 (1998).
[18] H. Ennamiri, R. Nassif, Y. Boughaleb, J. F. Gouyet, J. phys: Condensed Matter, **9**, 2433 (1997).
[19] A. Memsouk, Y. Boughaleb, R. Nassif, H. Ennamiri, Eur. Phys. J. B; M. J. Condensed Matter **2**, 35 (2000).
[20] M. Tringides, Gomer, Surf. Sci. **166**, 419 (1986).
[21] C. Uebing and R. Gomer, J. Chem. Phys. **95**, 7626, 7636, 7641, 7648 (1991).
[22] C. Uebing, R. Gomer, J. Chem. Phys. **100**, 7759 (1994).
[23] L. J.de Jongh, A. R. Miedema, Experiments on Simple Magnetic Model Systems (Taylor and Francis, London, 1974).
[24] H. Ikeda, K. Hirakawa, Solid State Commun. **14**, 529 (1974).
[25] E. J. Samuelsen, J. Phys. Chem. Solids **35**, 785 (1974).
[26] K. Binder, D. W. Heermann. Monte Carlo Simulation in Statistical physics, Springer (2002).
[27] K. W. Kehr et al. Diffusion in Condensed Matter Springer (2005).
[28] T. Springer: Quasielastic neutron scarttering for the investigation of diffusive Motions in Solids Liquids Tracs in Modern physics, **64** (Springer -Verlag, Berlin Heidelberg New York 1972).
[29] N. Metropolis, A. W. Rosenbluth, M. N. Rosenbluth, A. H. Teller, E. Teller, J. Chem. Phys. **21**, 1087 (1953).
[30] D. A. Reed, G. Ehrlich, Surf. Sci. **102**, 588 (1981).
[31] R. Gomer, Rep. Prog. Phys. **53**, 917 (1990).
[32] D. P. Landau, K. Binder, Phys. Rev. B **31**, 5946 (1985).
[33] K. W. Kehr, R. Kutner, K. Binder, Phys. Rev. B **23**, 4931 (1981).
[34] Y. Boughaleb, M. Nouredine, M. Snina, R. Nassif, M. Bennai, Phys. Research Internationnal 284231 (2010).

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