Tracer diffusion on two coupled lines: The long-time tail of the velocity autocorrelation function compared to the mode-coupling prediction

H. van Beijeren
Institut für Theoretische Physik A, Rheinisch-Westfälische Technische Hochschule Aachen, D-5100 Aachen, West Germany

R. Kutner
*Institut für Festkörperforschung der Kernforschungsanlage Jülich, D-5170 Jülich, West Germany and Institut of Experimental Physics, University of Warsaw, Hoża 69, PL-00-681 Warszawa, Poland**

K. W. Kehr
Institut für Festkörperforschung der Kernforschungsanlage Jülich, D-5170 Jülich, West Germany
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Mode-coupling predictions for the long-time behavior of the mean-square displacement of tracer particle diffusion in a quasi-one-dimensional lattice-gas model are compared with the results of Monte Carlo simulations. Furthermore, the internal consistency of an approximate theory is investigated under the assumption that mode-coupling theory is valid.

In a recent publication,[1] to be referred to as I, we investigated tracer diffusion in the following model: The sites of an $N\times 2$ rectangular lattice with periodic boundary conditions are randomly occupied by particles with an average concentration (occupation number per site) c. Double occupancy is forbidden and the particles may jump to unoccupied neighboring sites with jump rates Γ_x and Γ_y for jumps in the $+$ or $-x$ direction and the $+$ or $-y$ direction, respectively. We obtained an approximate expression for the velocity autocorrelation function or, equivalently, the mean-square displacement of a tagged particle and found this expression to agree quite well with the results of Monte Carlo simulations. We also found that for large jump-rate ratios the coefficient of the $t^{1/2}$ contribution to the mean-square displacement shows a strong variation as a function of c. Therefore, we feel that the present model provides a rather sensitive test of the validity of the mode-coupling predictions for the long-time behavior of correlation functions in purely diffusive systems.

For long times the mean-square displacement was found to behave as

$$\langle [x(t)-x(0)]^2\rangle = 2D_{\rm tr}t + c_1 t^{1/2} + \cdots \ . \tag{1}$$

In this equation the brackets denote an average both over the equilibrium distribution of particles and over the stochastic hopping process. The tracer-diffusion coefficient depends on the jump rates and the concentration through the following set of relations:

$$D_{\rm tr} = b\Gamma_x \ , \tag{2}$$

$$2cb(1+b)+[b^2+3cb-(1-c)](\gamma+q)=0, \tag{3}$$

$$q=\sqrt{\gamma(1+b+\gamma)} \ , \tag{4}$$

where we set the lattice spacing equal to unity and introduced the quantity $\gamma=(2-c)\Gamma_y/\Gamma_x$. The quantity b can be solved for numerically from these equations. The coefficient c_1 is given as

$$c_1 = 2c(1-c)\sqrt{(1+b)\Gamma_x} \times\left(\frac{2c(1+b)-(1+b-2c)(\gamma+q)}{4c^2(1+b)-(1+b-2c)(1+b+2c)\gamma}\right)^2 . \tag{5}$$

The asymptotic form (1) of the mean-square displacement is also predicted by phenomenological mode-coupling theory[2] with the coefficient c_1 given as

$$c_1^{\rm MC} = (D+D_{\rm tr})^{-3/2}\langle \hat{c}\hat{c}\rangle\left(\frac{D-D_{\rm tr}}{2c}-\frac{1}{2}\frac{\partial D_{\rm tr}}{\partial c}\right)^2 . \tag{6}$$

In our case the bulk-diffusion coefficient D equals Γ_x. The factors 2 in the denominators inside the parenthesis occur because the average density per lattice unit in the x direction is $2c$. The compressibility $\langle \hat{c}\hat{c}\rangle$ is given as

$$\langle \hat{c}\hat{c}\rangle = \sum_{n_1=1}^{2}\sum_{m_2=-\infty}^{\infty}\sum_{n_2=1}^{2}\left(\langle \eta_{0n_1}\eta_{m_2n_2}\rangle - \langle \eta_{0n_1}\rangle\langle \eta_{m_2n_2}\rangle\right) = 2c(1-c) \ . \tag{7}$$

Here the η_{mn} are stochastic variables equaling unity if the site (m,n) is occupied and zero if it is empty. In the equilibrium state these variables are entirely uncorrelated to each other; hence, the only nonvanishing contributions result from $(m_2,n_2)=(0,n_1)$.

Our Monte Carlo simulations do not cover a fine enough grid of concentration values to allow for a reliable numerical determination of the derivative $\partial D_{\rm tr}/\partial c$. However, we found such good agreement between the simulation values of $D_{\rm tr}$ and the values obtained from Eqs. (2)–(4) that we believe the value of $\partial D_{\rm tr}/\partial c$ deduced from these equations cannot be in error by more than a few percent. In addition mode-coupling theory provides a consistency check on our theory: assuming mode-coupling theory to be correct we

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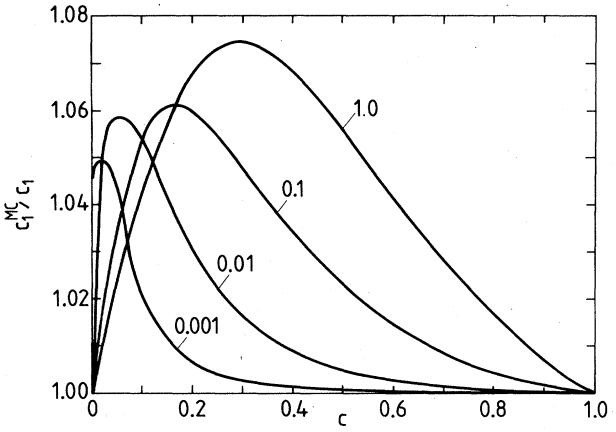

FIG. 1. The ratio c_1^{MC}/c_1 plotted as a function of concentration for four values of Γ_y/Γ_x.

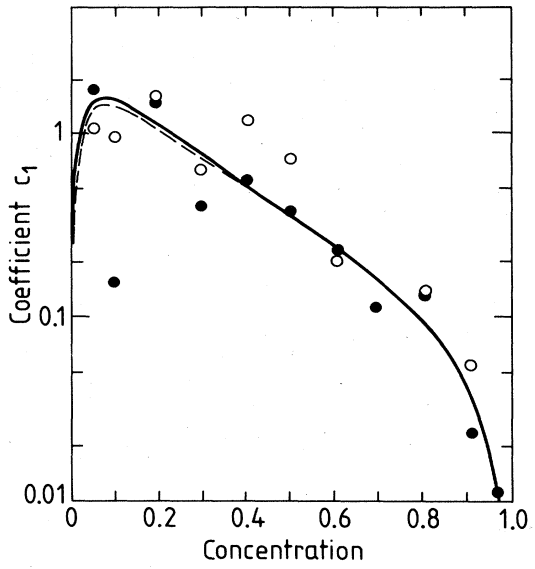

FIG. 2. The coefficients c_1^{MC} (solid line) and c_1 (dashed line) compared with the Monte Carlo predictions (open and full points represent two- and three-parameter fit, respectively), over the full range of concentrations.

ought to obtain identical results from (5) and (6), using (2) and (3) to determine D_{tr} and $\partial D_{tr}/\partial c$.

In Fig. 1 we show the results of this consistency check. The ratio of c_1^{MC}, obtained by calculating (6) using our theoretical expressions (2)–(4) for D_{tr}, and c_1, as given by (5), is plotted as a function of c for four different values of Γ_y/Γ_x. The deviations from unity are nowhere larger than might be expected on the basis of the few percent of difference we typically found to exist between theory and simulation results.[1]

In Fig. 2 we compare c_1^{MC} obtained by using Eqs. (2)–(4) for calculating D_{tr} and its derivative with respect to c, with Monte Carlo values for c_1, which were obtained for several values of c at the constant jump-rate ratio $\Gamma_y/\Gamma_x=0.01$. The Monte Carlo values of c_1 were determined both by a two-parameter fit of the Monte Carlo mean-square displacement for large time to a function of the form (1) and by a three-parameter fit to a function of the form

$$\langle [x(t)-x(0)]r^2\rangle = 2D_{tr}t + c_1 t^{1/2} + c_2 \ . \tag{8}$$

For reference we also plotted c_1 as given by (5) and (6) in this figure.

One sees that the Monte Carlo values for c_1 show large scatter around the predicted mode-coupling values as well as around the theoretical prediction (5). In addition its value in several cases depends rather sensitively on the choice of the fitting procedure. Yet one may conclude that the Monte Carlo results within the present precision are consistent with the mode-coupling predictions. Clearly, longer Monte Carlo runs would be desirable; however, the large extra cost in computer time has prevented us from performing these.

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*Permanent address.

[1]R. Kutner, H. van Beijeren, and K. W. Kehr, Phys. Rev. B **30**, 4382 (1984).

[2]H. van Beijeren, J. Stat. Phys. **35**, 399 (1984); **39**, 449(E) (1985).