Tracer correlation factor in a square-lattice gas with extended hard-core interactions

D. K. Chaturvedi

Department of Physics, Kurukshetra University, Kurukshetra (Haryana), India

(Received 19 May 1986)

A lattice-gas model with extended hard-core interactions is used to study tracer diffusion in a square planar lattice. For the vacancy availability factor a simple approximation, which decouples higher order correlations in terms of the pair-correlation functions, is suggested. The results for the third- and fourth-order correlation functions and the vacancy availability factor are compared with the recent Monte Carlo data of Fröböse and Jäckle. The tracer correlation factor is in qualitative agreement with that of Murch.

I. INTRODUCTION

Lattice-gas models have been extensively used to study diffusion in solids through the vacancy mechanism in which an atom or ion move through successive jumps from a lattice site to a vacant site. It was long recognized by Bardeen and Herring[1] and LeClare[2] that the successive jump directions are not random but are correlated with one another. The reason is that when an atom has just made a jump there is a vacancy left behind. Therefore the probability for a forward jump is always smaller than the probability for a backward jump. These correlation effects are described by a correlation factor, known as the tracer correlation factor. Bardeen and Herring have given accurate results for the three cubic Bravais lattices that contain a single vacancy. With the discovery of many nonstoichiometric compounds there is now a need to know the tracer correlation factor in the whole concentration range, that is from a single vacancy to an almost empty lattice.

In the past, several theoretical methods[3-8] have been used to study tracer diffusion in a lattice gas that excludes multiple occupation of a site. The results for the tracer correlation factor agree within one percent of the Monte Carlo[9-11] data. Lattice gas models with nearest-neighbor[12-14] and the long-range Coulomb[15-18] interactions have also been studied. Such models are relevant in the study of fast ionic diffusion in some superionic conductors. A few years ago, Murch[19] obtained Monte Carlo results for the Haven ratio $H_R(=f_t/f_c$, where f_t is the tracer correlation factor and f_c is the charge correlation factor[20]) in a square lattice gas with extended hard-core interactions, which excludes simultaneous occupation of nearest-neighbor sites. The results are very interesting, especially for the concentration $c > 0.3$ where f_c decreases rapidly to a value close to zero around $c = 0.375$. At this concentration a second order disorder/order phase transition is also observed. f_t, however, show no such catastrophic behavior at the transition.

So far much attention is devoted to the thermodynamic[21,22] properties and the phase transitions[23,24] in a lattice gas with finite and infinite nearest-neighbor repulsions. To the best of our knowledge no theoretical attempt has

been made to study correlation factors in a square-lattice gas with extended hard-core interactions. In this paper, we are mainly concerned with a tracer atom diffusing against a dynamic background of particles with hard-core interactions that excludes both the multiple occupation of the site and the simultaneous occupation of its nearest-neighbor sites. In Sec. II, we describe the lattice-gas model and the master rate equation which governs the dynamics. For the transition rate we have proposed a form which is compatible with the detailed balance condition and is nonzero only when the nearest neighbors of the diffusing particle and the vacancy to which jump is taken are simultaneously vacant. The result for the average transition rate is compared with the very recent Monte Carlo data of Fröböse and Jäckle.[25] In Sec. III, the tracer correlation factor is obtained from the incoherent structure factor by using the projection operator technique of Mori.[26] Numerical results and their discussion is finally given in Sec. IV.

II. LATTICE-GAS MODEL

We consider a regular square lattice of equivalent site l, which is partly occupied by N identical particles. Let p_l and n_l denote the stochastic occupancy variables for a marked ("tracer") atom and for any atom of the system, respectively. Then, p_l equals unity if, at time t, site l is occupied by the tracer and is zero otherwise. Similarly, n_l equals unity when the site l is occupied and is zero when it is vacant. p_l and n_l satisfy the properties

$$n_l^2 = n_l, \quad n_l p_l = p_l ,$$
$$p_l p_{l'} = p_l \delta_{l,l'}, \quad \sum_l p_l = 1 . \tag{2.1}$$

The last relation holds because we consider only a single tracer atom. The mean occupation $\langle n_l \rangle$ gives the average concentration $c = 1 - v$ of diffusing atoms (v is the vacancy concentration, with $0 < v < 1$).

Now we allow the system to evolve in time through a many-particle jump diffusion process, where jumps are assumed between nearest-neighbor sites only. The change in the probability $P(n,t)$ to find the configuration $n = \{n_l\}$ at time t is governed by the rate equation

$$\frac{dP(n,t)}{dt} = \frac{1}{2} \sum_{l,\delta} [W_{l+\delta,l}(\tilde{n})P(\tilde{n},t) - W_{l,l+\delta}(n)P(n,t)] .$$

$$(2.2)$$

Here $\tilde{n}$ denotes the configuration which results from n by interchanging the occupation at l and its nearest neighbor $l+\delta$, i.e., $\tilde{n}_{l+\delta}=n_l$ and $\tilde{n}_l=n_{l+\delta}$. $W_{l,l+\delta}$ denotes the corresponding transition rate.

Let $f(n,t)$ denote the average of some function $f(n)$ at time t for a given initial configuration n at time $t=0$. Starting from the master rate equation the time evolution of $f(n)$ is formally[15,18] given by the operator L, defined by

$$Lf(n) = \frac{1}{2} \sum_{l,\delta} W_{l,l+\delta}(n)[f(\tilde{n})-f(n)] .$$

$$(2.3)$$

The problem now is to find the transition rates. They embody basic statistics of the problem and in equilibrium satisfy the condition of detailed balance. For a lattice gas with hard-core interaction, detailed balance requires that

$$W_{l,l+\delta}(n) = W_{l,l+\delta}(\tilde{n}) ,$$

$$(2.4)$$

or, $W_{l,l+\delta}(n) = W_{l+\delta,l}(n)$. In the simplest case, where double occupancy of a site is excluded, one assumes the form

$$W_{l,l+\delta}^0(n) = \alpha[n_l(1-n_{l+\delta})+n_{l+\delta}(1-n_l)] ,$$

$$(2.5)$$

which allows a jump only if $n_l \neq n_{l+\delta}$. α is the hopping frequency. With extended hard-core interactions, we take the form

$$W_{l,l+\delta}(n) = W_{l,l+\delta}^0(n)$$
$$\times \prod_{\delta_1(\neq \delta)} (1-n_{l+\delta_1}) \prod_{\delta_2(\neq -\delta)} (1-n_{l+\delta+\delta_2}) .$$

$$(2.6)$$

Here δ_2 is the nearest-neighbor vector of the site $l+\delta$. The product terms in (2.6) ensure that the transition rate is zero when any of the nearest-neighbor site of l or $l+\delta$ is occupied. The form of (2.6) also satisfies the detailed balance condition (2.4).

To proceed further we note that the extended hard-core interactions of the particle at l guarantees that the pair correlation function $g_2(\delta) = \langle n_l n_{l+\delta} \rangle/c^2$ is zero. Therefore, while taking the averages we can drop the terms that contain occupancies of nearest-neighbor sites. The average transition rate, $\langle W \rangle = \langle W_{l,l+\delta} \rangle$, can thus be written from (2.6) and (2.5) as

$$\langle W \rangle = \alpha[\langle n_1 \prod_{\delta_2(\neq -\delta)} (1-n_{l+\delta+\delta_2}) \rangle$$

$$+ \langle n_{l+\delta} \prod_{\delta_1(\neq \delta)} (1-n_{l+\delta_1}) \rangle]$$

$$= 2\alpha c V ,$$

$$(2.7)$$

where

$$V = \langle n_l \sum_{\delta_2(\neq -\delta)} (1-n_{l+\delta+\delta_2}) \rangle/c$$

$$(2.8)$$

is called the vacancy availability factor. For a square lattice, (2.8) can be expressed as (using the notation of Fig. 1)

$$V = 1 - c[2g_2(1,2)+g_2(1,3)]$$
$$+ 3c^2 g_3(1,2,3) - c^3 g_4(1,2,3,2') .$$

$$(2.9)$$

g_3 and g_4 are, respectively, the third- and fourth-order correlation functions, which are defined for general n by

$$g_n(l_1,l_2,\longrightarrow l_n) = \langle n_{l_1} n_{l_2} \ldots n_{l_n} \rangle/c^2 .$$

$$(2.10)$$

Equation (2.9) is the same as used by Fröböse and Jäckle[25] in their Monte Carlo calculations. These authors have shown that the Kirkwood superposition approximation[27] is very inaccurate for the correlation functions of third and fourth orders, but gives better results for V. The decoupling approximation, is, however, needed as we shall come across several higher-order correlations, which are not known a priori. In this paper, we use a simple approximation and write (2.8) as

$$V = \prod_{\delta_2(\neq -\delta)} [1-cg_2(\delta+\delta_2)] .$$

$$(2.11)$$

We may remark here that (2.11) is in agreement with O'Keefe's cluster approximation result[22,25] on the square lattice. Now comparing (2.11) with (2.9) we find that the results are exact up to the pair correlations. The higher-order correlations correspond to a superposition approximation (SA):

$$g_n(l_1,l_2,\ldots,l_n) = g_2(l_1,l_2)g_2(l_1,l_3)\ldots g_2(l_1,l_n) ,$$

$$(2.12)$$

which is different from Kirkwood in the sense that here pair correlations are taken with respect to the site l_1 only.

III. TRACER CORRELATION FACTOR

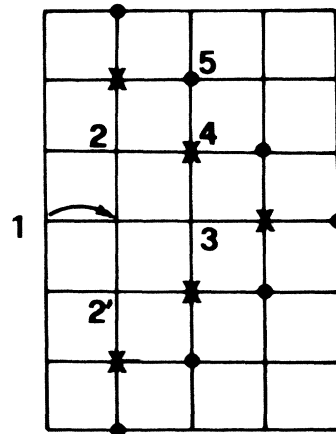The motion of the tracer atom is described by the incoherent structure factor

FIG. 1. Indicated notation is used throughout the paper. The sites marked with $\times$ and $\bullet$ are assumed to be equivalent to sites 4 and 5, respectively.

$$S_{\text{inc}}(q,t) = \langle p(q,t) \mid p(q) \rangle \; , \tag{3.1}$$

defined in terms of the tracer density

$$p(q) = \sum_l p_l \exp(-iql) \; . \tag{3.2}$$

The Laplace transform $S_{\text{inc}}(q,z)$ of (3.1) can be written in the standard form[26]

$$S_{\text{inc}}(q,z) = [z + \Omega(q) + M(q,z)]^{-1} \; , \tag{3.3}$$

where

$$\Omega(q) = -\langle p(q) \mid Lp(q) \rangle \; , \tag{3.4}$$

and $M(q,z)$ is the corresponding memory function.

To calculate $Lp(q)$ we substitute $f = p_l$ in (2.3), which yields

$$Lp(q) = \sum_{l,\delta} (e^{-iq\delta} - 1) p_l W_{l,l+\delta} e^{-iql} \; . \tag{3.5}$$

Now substituting (3.5) in (3.4) and observing the conditions (2.1), we get

$$\Omega(q) = \frac{\langle W \rangle}{2c} \sum_\delta (1 - \cos q\delta) = V\omega(q) \; , \tag{3.6}$$

where

$$\omega(q) = \alpha \sum_\delta (1 - \cos\delta) \tag{3.7}$$

is the unblocked tracer diffusion frequency and V gives the blocking factor. In the absence of correlations the value of V is equal to $(1-c)^3$. In a mean field approximation, where the memory function is neglected, a particle makes independent hops with an average jump rate $\langle W \rangle/2c$. The tracer atom, however, performs a correlated random walk because of the presence of other atoms. This fact is accounted for by a wave-vector- and frequency-dependent correlation factor:

$$f(q,z) = 1 + M(q,z)/\Omega(q) \; . \tag{3.8}$$

Its long-wavelength and zero-frequency limit represents the conventional tracer correlation factor f_t, in terms of

which tracer diffusion constant D_t can be obtained from (3.3) and (3.8) as

$$D_t = \lim_{\omega \to 0} \lim_{q \to 0} \Omega(q) f(q, -i\omega)/q^2 = D_0 V f_t \; . \tag{3.9}$$

Here, $D_0 = \alpha a^2$ is the diffusion constant for infinite dilution and a is the lattice constant.

In order to calculate the tracer correlation factor we need to evaluate the memory function $M(q,z)$. One way is to use a simple factorization approximation.[15,16] Since the extended-hard-core interaction model involves multiple correlations such a procedure will not be convenient. The other method is to use a continued fraction for $M(q,z)$. In the simple-hard-core model (site blocking only) it was observed[7,8] that the first stage of the continued fraction (obtained by neglecting the second-order memory function) accounts for about 80% of the correlation effects. Therefore, we use this procedure and write the first-stage continued fraction as

$$M(q,z) = \frac{\Delta_1^2(q)}{z + \Omega_1(q)} \; , \tag{3.10}$$

where

$$\Delta_1^2(q) = -\langle A_1(q) \mid A_1(q) \rangle \tag{3.11}$$

and

$$\Omega_1(q) = -\langle A_1(q) \mid LA_1(q) \rangle \langle A_1(q) \mid A_1(q) \rangle^{-1} \; . \tag{3.12}$$

$A_1(q)$ is the $t = 0$ value of the random force, defined as

$$A_1(q) = QLp(q) \tag{3.13}$$

with

$$Q = 1 - \mid p(q) \rangle \langle p(q) \mid \; . \tag{3.14}$$

Substituting (3.13) in (3.11) and making use of (3.4), we write

$$\Delta_1^2(q) = -\langle Lp(q) \mid Lp(q) \rangle + \Omega^2(q) \; . \tag{3.15}$$

Now using (3.5) the first term on the right-hand side can be written as

$$\langle Lp(q) \mid Lp(q) \rangle = \sum_{\delta,l} (1 - e^{iq\delta}) \left[(1 - e^{-iq\delta}) \langle p_l W_{l,l+\delta}^2 \rangle + \sum_{\delta_1(\neq\delta)} (1 - e^{-iq\delta_1}) \langle p_l W_{l,l+\delta} W_{l,l+\delta_1} \rangle \right] \; . \tag{3.16}$$

Equation (3.16) can be solved easily by substituting the transition rates from (2.6) and making use of the properties given in (2.1). Care, however, must be taken in counting the distinct terms. Finally, dropping the terms with nearest-neighbor occupancies and making the approximation discussed in (2.11), we get

$$\langle Lp(q) \mid Lp(q) \rangle = 2\alpha(1 - V)\Omega(q) + \Omega^2(q) \; . \tag{3.17}$$

Thus, from (3.17) and (3.15), we have

$$\Delta_1^2(q) = -2\alpha(1 - V)\Omega(q) \; , \tag{3.18}$$

which is the same as obtained for site blocking with

$V = 1 - c$. We may mention here that in deriving (3.17) we have used

$$\sum_l \langle p_l W_{l,l+\delta} W_{l,l+\delta_1} \rangle$$

$$= \frac{\alpha^2}{c} \left\langle n_l \prod_{\delta_2(\neq -\delta)} (1 - n_{l+\delta+\delta_2}) \prod_{\delta_2'(\neq -\delta_1)} (1 - n_{l+\delta_1+\delta_2'}) \right\rangle \tag{3.19a}$$

$$= \alpha^2 V^2 \; . \tag{3.19b}$$

Obviously, (3.19a) involve correlations upto the seventh

order which are expressed in a simple form, (3.19b), through the approximation of (2.11).

We now need to evaluate $\Omega_1(q)$, which can be rewritten as

$$\Omega_1(q) = \frac{1}{\Delta_1^2(q)} [\langle Lp(q) | L^2 p(q) \rangle$$

$$+ \Omega(q)\langle p(q) | L^2 p(q) \rangle] - \Omega(q) . \quad (3.20)$$

In order to obtain the equation of motion for $Lp(q)$, i.e., $L^2 p(q)$, we substitute $f(n) = p_l W_{l,l+\delta}$ in (2.3). With the transition rate (2.6), we see that $f(\tilde{n})$ differs from $f(n)$ only when the transition occurs on the bonds connecting the site l, $l+\delta_1$, or $l+\delta+\delta_2$. The second case, however, does not contribute because the transition from $l+\delta_1$ requires the site l to be empty, which is already occupied by the tracer atom. Thus, we write

$$L(p_l W_{l,l+\delta}) = \alpha \left[\sum_{\delta_1'} W_{l,l+\delta_1'}[p_{l+\delta_1'}(1-n_l) - p_l(1-n_{l+\delta_1'})] \prod_{\delta_1(\neq\delta_1')} (1-n_{l+\delta_1}) \prod_{\delta_2(\neq-\delta)} (1-n_{l+\delta+\delta_2}) \right.$$

$$\left. + \sum_{\delta_2(\neq-\delta)} \sum_{\delta_3(\neq-\delta_2)} W_{l+\delta+\delta_2,l+\delta+\delta_2+\delta_3}(n_{l+\delta+\delta_2} - n_{l+\delta+\delta_2+\delta_3})p_l \prod_{\delta_1} (1-n_{l+\delta_1}) \prod_{\delta_2'(\neq-\delta,\delta_2)} (1-n_{l+\delta+\delta_2'}) \right] . \quad (3.21)$$

Substituting the values of the transition rates in (3.21) and using (3.5) we get, after some simplifications,

$$L^2 p(q) = \alpha^2 \sum_{l,\delta} (e^{-iq\delta} - 1)[A + B]e^{-iql} , \quad (3.22)$$

where

$$A = p_l \left[\left[-2 + e^{-iq\delta} \prod_{\delta_1'(\neq-\delta)} (1-n_{l+2\delta+\delta_1'}) - \prod_{\delta_1'(\neq\delta)} (1-n_{l-\delta+\delta_1'}) \right] \prod_{\delta_1(\neq-\delta)} (1-n_{l+\delta+\delta_1}) \right.$$

$$\left. + \sum_{\delta_1(\neq\pm\delta)} \left[e^{-iq\delta_1} \prod_{\delta_2(\neq-\delta,-\delta_1)} (1-n_{l+\delta+\delta_1+\delta_2}) - \prod_{\delta_2(\neq-\delta,\delta_1)} (1-n_{l+\delta+\delta_2}) \right] \prod_{\delta_1'(\neq-\delta_1)} (1-n_{l+\delta_1+\delta_1'}) \right] \quad (3.23)$$

and

$$B = \sum_{\delta_1(\neq-\delta)} \sum_{\delta_2(\neq-\delta,-\delta_1)} p_l \left[n_{l+\delta+\delta_1} \prod_{\delta_3(\neq-\delta_2)} (1-n_{l+\delta+\delta_1+\delta_2+\delta_3}) \right.$$

$$\left. - n_{l+\delta+\delta_1+\delta_2} \prod_{\delta_2'(\neq-\delta,-\delta_1,\delta_2)} (1-n_{l+\delta+\delta_1+\delta_2'}) \right] \prod_{\delta_1'(\neq-\delta,\delta_1)} (1-n_{l+\delta+\delta_1'}) . \quad (3.24)$$

With $Lp(q)$ from (3.5) and $L^2 p(q)$ from (3.22) one can get $\Omega_1(q)$ from (3.20). It can be seen again that the calculations involve several site correlations, which according to approximation (2.11) can be expressed in the simpler form. In the long-wavelength limit (in which we are interested) the contributions from the second and third term of (3.20) vanishes. The first term then yields

$$\lim_{q\to 0} \Omega_1(q) = \alpha \left[2 - F^3(4) + \frac{2F(2)F(3)}{(1-V)}[1-F(2)F(3)] \right.$$

$$+ \frac{c}{1-V} \{ 3g_2(1,3)F^2(2)F^3(5) + 4g_2(1,2)F(2)F(3)F^3(5) $$

$$\left. + 3g_2(1,4)F^2(4)[1-F^2(2)] + 4g_2(1,4)F(4)[1-F(2)F(3)] \} \right] . \quad (3.25)$$

where we have used the notation (see Fig. 1)

$$F(n) = 1 - cg_2(1,n), \quad n = 2,3,\ldots,5 . \quad (3.26)$$

Finally, substituting (3.25) and (3.18) in (3.10) and making use of (3.8) and (3.9), we get the tracer correlation factor

$$f_t(c) = 1 - \frac{2\alpha(1-V)}{\Omega_1(q=0)} . \quad (3.27)$$

The numerical results for the dependence of tracer correlation factor on concentration will be discussed in the next section.

IV. SUMMARY AND RESULTS

We have studied self-motion in a square lattice gas with extended-hard-core interactions which prevents the simultaneous occupation of the nearest-neighbor sites. The basic statistics of the problem is incorporated through the transition rate which describes the probability of a jump between the nearest-neighbor sites. Our expression for the average jump rate is in agreement with that of Fröböse and Jäckle,[25] who have also obtained Monte Carlo results for the pair, triplet, and quadruplate correlation functions in a cluster around the test particle.

For the vacancy availability factor we have used a simple superposition approximation which decouples the higher-order correlation functions in terms of the pair correlation function g_2. We have tested the accuracy of the approximation by comparing our results for the correlation functions $g_3(1,2,3)$ and $g_4(1,2,3,2')$ of third and fourth order and for the vacancy availability factor V, with the results of the Monte Carlo calculation. In Figs. 2 and 3 we have plotted, respectively, the values of g_3 and g_4 obtained from (2.12) with the Monte Carlo data for g_2, and the corresponding Monte Carlo results. To make some comparison we have also shown the values obtained in Kirkwood superposition approximation (KSA), which can be expressed as

$$g_3^{\text{KSA}}(1,2,3)=[g_2(1,2)]^2 g_2(1,3) , \qquad (4.1a)$$

$$=g_2(1,2)g_3^{\text{our}}(1,2,3) , \qquad (4.1b)$$

and

$$g_4^{\text{KSA}}(1,2,3,2')=[g_2(1,2)]^4[g_2(1,3)]^2 , \qquad (4.2a)$$

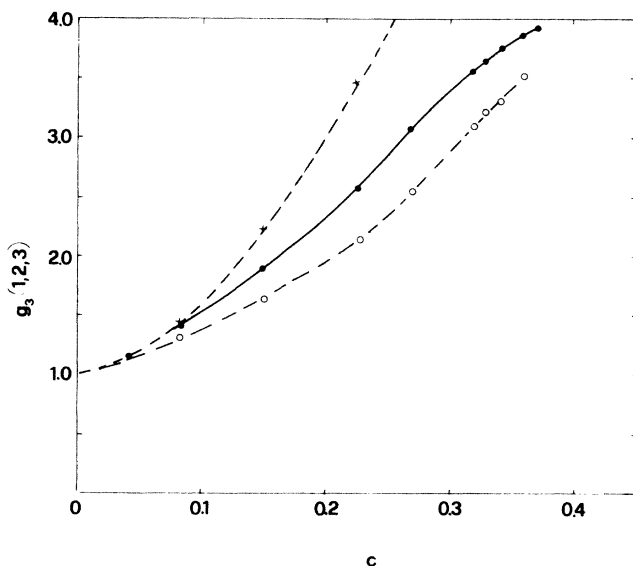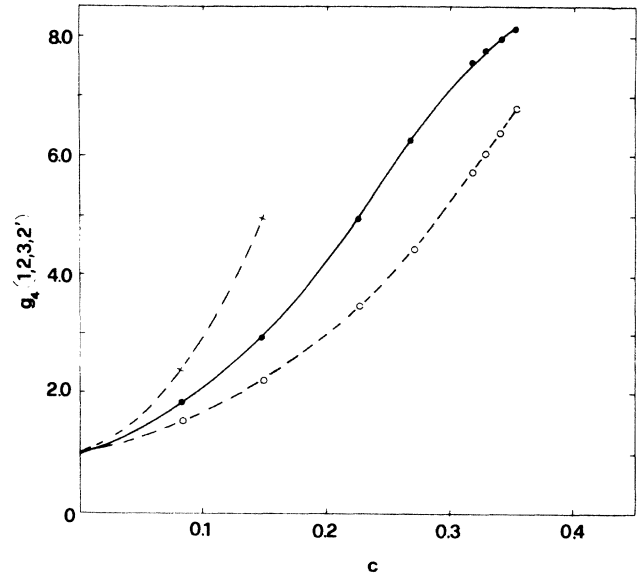$$=[g_4^{\text{our}}(1,2,3,2')]^2 , \qquad (4.2b)$$

FIG. 3. Fourth-order correlation function $g_4(1,2,3,2')$ in simple SA (- -O- -) and in KSA (- -×- -) compared with the result of Monte Carlo calculation (—●—). The lines are guides to the eye.

where g_3^{our} and g_4^{our} are obtained from (2.12). As already pointed out by Fröböse and Jäckle, KSA gives very inaccurate results, specially for g_4 where it increases very fast at concentrations $c > 0.12$. On the other hand, our values are slightly lower but are in much better agreement with the Monte Carlo data in the entire concentration range ($c \leq 0.37$). This fact is further reflected in the vacancy availability factor V, which is shown in Fig. 4. Since for V the errors in the approximation for g_3 and g_4 get somewhat compensated, the results in KSA are reasonable for concentration up to $c = 0.3$, but then deteriorates rapidly, leading to very small and finally negative values of V. Our values of V remain positive in the entire concentration range (up to $c = 0.37$) and are in good agreement with the Monte Carlo data; they are lower by about 30% for $c \geq 0.3$. We may also remark here that the Gaussian
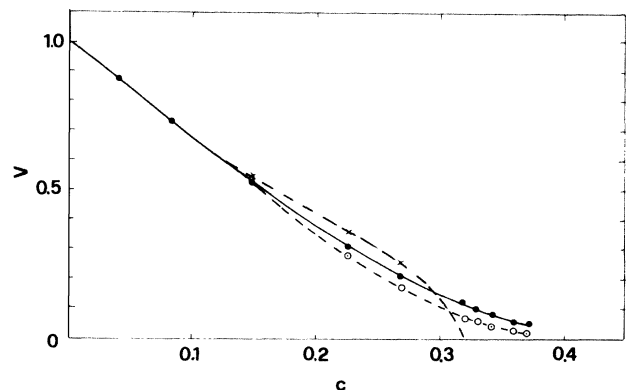

FIG. 2. Triplet correlation function $g_3(1,2,3)$ in simple SA (- -O- -) and in KSA (- -×- -) compared with the result of Monte Carlo calculation (—●—). The lines are guides to the eye.

FIG. 4. Vacancy availability factor in simple SA (- -⊙- -) and in KSA (- -×- -) compared with the Monte Carlo calculation (—●—). The lines are guides to the eye.
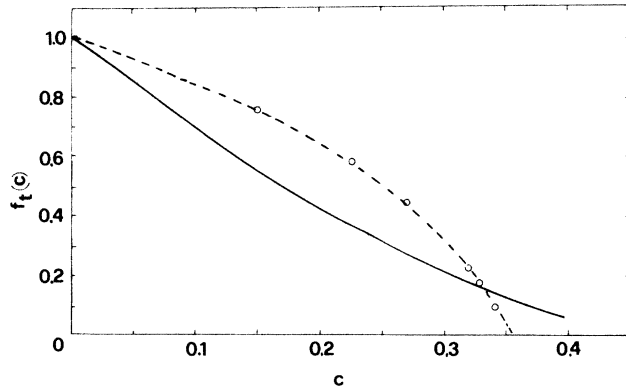
FIG. 5. Tracer correlation factor as a function of concentration. Monte Carlo calculation (——) compared with present calculation (- -O- -). The lines are guides to the eye.

factorization and Bethe-Peirels cluster approximation used by Fröböse and Jäckle give good results for g_3, g_4, and for V. The present approximation, however, has the advantage that it simplifies the calculations considerably, as seen in the previous sections.

An important question arises why KSA, which describes short-range correlations so well in the liquids, fails in the present case. Our analysis indicates that if instead of (2.12) one uses the approximation

$$g_3(1,2,3) = [g_2(1,2)]^2 , \qquad (4.3)$$

the results are in complete agreement (up to the first place in decimal figure) with the MC data. A plausible reason may be that $g_3(1,2,3)$ gives the probability to find particles at sites 2 and 3 when a particle is situated at site 1. Thus, the vacancy between sites 1 and 3 makes the occu-

pancies of these sites uncorrelated, which is equivalent to replacing $g_2(1,3)$ in (4.1a) by unity. With a similar reasoning, one can write

$$g_4(1,2,3,2') = \frac{[g_3(1,2,3)]^2}{g_2(1,3)} \qquad (4.4a)$$

$$= \frac{[g_2(1,2)]^4}{g_2(1,3)} . \qquad (4.4b)$$

Again with this approximation results are in very good agreement with the MC data. A similar analysis, however, may be difficult when one is dealing with several site correlations in a bigger cluster. Therefore, in this paper we have continued with the approximation of (2.12).

Finally, in Fig. 5 we have shown the dependence of the tracer correlation factor, $f_t(c)$, on concentration c. The results of (3.27) when compared with the Monte Carlo data of Murch[19] show higher values up to $c = 0.3$ and then decreases to values close to zero around $c = 0.35$. The quantitative disagreement is not surprising because we have neglected the second-order memory function and have used a superposition approximation for higher-order correlation functions, which introduces inaccuracies especially at higher concentrations. The inclusion of a higher-order memory function will reduce our results in line with the MC data. However, since the calculations becomes tedious we do not attempt it here.

ACKNOWLEDGMENTS

[1]J. Bardeen and C. Herring, *Imperfections in Nearly Perfect Crystals* (Wiley, New York, 1952), p. 261.

[2]A. D. LeClare, in *Physical Chemistry—An Advanced Treatise*, edited by H. Eyring, D. Henderson, and W. Jost (Academic, New York, 1970), Vol. 10.

[3]O. F. Sankey and P. A. Fedders, Phys. Rev. B 15, 3586 (1977).

[4]P. A. Fedders and O. F. Sankey, Phys. Rev. B 18, 5938 (1978).

[5]N. Nakazato and K. Kitahara, Prog. Theor. Phys. 64, 2261 (1980).

[6]R. A. Tahir-Kheli and R. J. Elliott, Phys. Rev. B 27, 844 (1983).

[7]D. K. Chaturvedi, Phys. Rev. B 28, 6868 (1983).

[8]D. K. Chaturvedi, J. Phys. C 17, L449 (1984).

[9]H. H. deBruin and G. E. Murch, Philos. Mag. 27, 1475 (1973).

[10]K. W. Kehr, R. Kutner, and K. Binder, Phys. Rev. B 23, 4931 (1981).

[11]G. E. Murch, Philos. Mag. A 49, 21 (1984).

[12]H. Sato and R. Kikuchi, Phys. Rev. B 28, 648 (1983).

[13]G. E. Murch and R. J. Thorn, Philos. Mag. 35, 493 (1977).

[14]H. Singer and I. Peschel, Z. Phys. B 39, 333 (1980).

[15]A. Bunde, D. K. Chaturvedi, and W. Dieterich, Z. Phys. B 47, 209 (1982).

[16]D. K. Chaturvedi and W. Dieterich, Z. Phys. B 49, 17 (1982).

[17]A. Bunde, D. K. Chaturvedi, and W. Dieterich, Solid State Ionics, 9&10, 1415 (1983).

[18]A. Bunde and W. Dieterich, Phys. Rev. B 31, 6012 (1985).

[19]G. E. Murch, Philos. Mag. A 44, 699 (1981).

[20]G. E. Murch and R. J. Thorn, Philos. Mag. 36, 529 (1977).

[21]G. Boureau, J. Phys. Chem. Solids 42, 743 (1983).

[22]M. O'Keeffe, in *Chemistry of Extended Defects in Non-Metallic Solids*, edited by L. Eyring and M. O'Keeffe (North-Holland, Amsterdam, 1971), p. 609.

[23]J. Orban and D. Van Belle, J. Phys. A 15, L501 (1982).

[24]D. A. Huckaby, J. Stat. Phys. 30, 23 (1983).

[25]K. Fröböse and J. Jäckle, J. Stat. Phys. 42, 551 (1986).

[26]H. Mori, Progr. Theor. Phys. 34, 423 (1969).

[27]J. G. Kirkwood, J. Chem. Phys. 18, 380 (1950).