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Hopping conductivity in the extended hard-core square lattice gas

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The hopping conductivity of particles on a square lattice, with infinite nearest-neighbor repulsion, is calculated by a steady-state approach. Effects of dynamic correlations are taken into account in the first nontrivial approximation. Static correlation functions needed for the conductivity formula are computed in the Bethe-Peierls approximation, for concentrations $c \leq 0.32$. The charge correlation factor is obtained, in good agreement with Monte Carlo results.

I. INTRODUCTION

Interactions between particles diffusing on a lattice have been studied through various approaches like the path-probability method,[1,2] the rate-equation formalism,[3-8] and Monte Carlo simulations (see Ref. 9 and references therein). While for some one-dimensional models the electrical conductivity was exactly calculated,[3-5] two-dimensional models are as difficult to deal with as they are interesting. References 1 and 2 are concerned with the honeycomb lattice, which is a good model for β-alumina. The square lattice gas of particles interacting with infinite nearest-neighbor (NN) repulsion, which forbids simultaneous occupation of adjoining sites, was first studied in 1981 by Murch,[10] with Monte Carlo techniques. He obtained both f_t, the tracer correlation factor, and f, the charge correlation factor, in order to determine the Haven ratio $H = f_t / f$. A second paper on the subject was published in 1986 by Chaturvedi,[11] who calculated the tracer correlation factor using the Zwanzig-Mori formalism.

In this paper we consider the same extended hard-core interaction model on the square lattice. Our purpose is the analytic calculation of the dc conductivity of the hopping particles, taking into account dynamic correlations. Thus, we attempt the theoretical determination of the charge correlation factor defined as the ratio of the conductivities at zero and infinite frequency:[3,6]

$$f = \sigma / \sigma(\infty) . \tag{1.1}$$

In Sec. II the static correlation functions needed later on are calculated by the Bethe-Peierls approximation, and compared with the results of Froböse and Jäckle.[12] In Sec. III a stationary flow formalism due to Richards[13] is used to calculate the conductivity, taking into account dynamic correlations. A summary and discussions are presented in Sec. IV.

II. STATIC CORRELATIONS

We consider a system of N_p identical particles on a square lattice with N equivalent sites. The only interaction present is an extended hard-core one, which forbids simultaneous occupation of nearest-neighbor sites. The system is described by the set of occupation numbers $\{n_i\}$, n_i taking the values 0 or 1 if the site i is empty or occupied, respectively. At equilibrium, in the absence of an external field, translational invariance holds and the mean value of n_i is equal to the concentration of particles:

$$\langle n_i \rangle_0 = c = \frac{N_p}{N} . \tag{2.1}$$

The subscript 0 indicates an equilibrium average.

The hard-core interaction limits the values of c to the interval $0 \leq c \leq 0.5$. Actually, Eq. (2.1) holds only for concentrations smaller than a critical value $c_{\mathrm{cr}} \simeq 0.37$.[14,15] For $c > c_{\mathrm{cr}}$ translational invariance is broken, so that if the lattice is divided into two disjoint parts in a checkerboard manner, one of the sublattices is preferentially occupied.

The static correlation functions of this particular model were discussed in detail by Froböse and Jäckle.[12] They have found that the analytical method which is closest to Monte Carlo results is the Bethe-Peierls approximation (BPA), which will be used here, in a modified version: while Froböse and Jäckle considered a square cluster with three sites per side, we shall take a 4×4 square. This is necessary because the formula for the conductivity obtained in Sec. III involves correlation functions for sites situated farther apart than in the 3×3 square.

We consider the cluster in Fig. 1. The notations for static correlation functions will have two indices, the first representing the number of sites involved and the second used for numbering. In order to evaluate correlators such as

$$g_{20} = \langle n_7 n_{10} \rangle_0 / c^2 , \tag{2.2a}$$

$$g_{21} = \langle n_5 n_7 \rangle_0 / c^2 , \tag{2.2b}$$

$$g_{30} = \langle n_5 n_7 n_{10} \rangle_0 / c^3 , \tag{2.2c}$$

$$g_{40} = \langle n_2 n_5 n_7 n_{10} \rangle_0 / c^4 , \tag{2.2d}$$

the chemical potential and the sum over states are needed as functions of concentration. In the BPA the chemical potential on the boundary of the cluster is different from that of the bulk, in order to take into account the effect of those lattice sites not included in the cluster. Symmetry leads to only three different values of the chemical potential: μ for the bulk, μ_1 for the sides, and μ_2 for the corners, and the respective fugacities

$$z=e^{\beta\mu},\quad z_1=e^{\beta\mu_1},\quad z_2=e^{\beta\mu_2}\,. \tag{2.3}$$

Considering c as a parameter, the z's are determined by Eq. (2.1):

$$\langle n_6\rangle_0=\langle n_2\rangle_0=\langle n_1\rangle_0=c\,. \tag{2.4}$$

The grand canonical partition function for the reduced lattice reads

$$\begin{aligned}Z={}&(z_2+1)^2(z^2+4z+1)+8z_1(z_2+1)^3(z^2+3z+1)+4z_1^2[(z_2+1)^3(z^2+3z+1)+(z_2+1)^2(2z^2+10z+5)]\\&+8z_1^3[(z_2+1)^2(z^2+4z+2)+2(z_2+1)(z+1)]+2z_1^4[(z_2+1)^2(z+1)^2+6(z_2+1)(z+1)+1]\,.\end{aligned} \tag{2.5}$$

Equations (2.4) become

$$\begin{aligned}c&=\frac{z}{4}\frac{\partial \ln Z}{\partial z}\,,\\ c&=\frac{z_1}{8}\frac{\partial \ln Z}{\partial z_1}\,,\\ c&=\frac{z_2}{4}\frac{\partial \ln Z}{\partial z_2}\,.\end{aligned} \tag{2.6}$$

Equations (2.6) are solved numerically giving results very close to those obtained by the 3×3 BPA, which also involves three fugacities. We present, for illustration, the c dependence of the bulk fugacity, obtained by both methods, in Table I.

The present calculations concern only the disordered state below $c_{\rm cr}$. If the ordered state was to be taken into account, one would have to use an adequate BPA, with broken translational symmetry, which gives $c_{\rm cr}\simeq0.32$,[12] lower than the Monte Carlo $c_{\rm cr}\simeq0.37$. For this reason we limit all calculations of the paper to the interval $0\le c\le0.32$, the results above $c=0.32$ being meaningless in the frame of BPA.

The 4×4 BPA leads to values of the correlation functions which differ from the 3×3 BPA results by only a slight displacement towards the Monte Carlo ones (one is naturally inclined to believe that by increasing the size of the cluster all approximate thermodynamic functions will monotonously approach the exact ones). The advantage, however, is not the insignificant gain of precision, but the possibility of calculating higher-order correlators, such as, say,

$$g_{41}=\langle n_3n_6n_9n_{14}\rangle/c^4\,. \tag{2.7}$$

Equation (2.7) must be evaluated by factorization in the 3×3 BPA, an additional approximation which is avoided in the 4×4 BPA. For illustration we give the best factorization estimate of g_{41}:

$$g_{41}^{\rm fa}=g_{20}g_{30}\,. \tag{2.8}$$

In Table II numerical values of Eqs. (2.7) and (2.8) are presented for comparison. Factorization turns out to be good at small concentration, while at high concentration significant discrepancies appear.

A drawback of the BPA is that it gives correlation functions which are not translationally invariant. For instance g_{41} is topologically equivalent to

$$g_{42}=\langle n_3n_6n_{11}n_{16}\rangle_0/c^4 \tag{2.9}$$

but their values are different, especially at high c. At $c=0.3$, g_{42} takes the value 5.902 which is closer to the factorization approximation. Though there are no Monte Carlo data to support the choice between g_{41} and g_{42} as a better estimate of the correlator with the given topology of sites, we believe that g_{41} is the best candidate. We base the assumption on the fact that g_{41} is more "centered" in the 4×4 cluster, as it contains no corner site. An analogous situation is that of the two-particle correlators $\langle n_2n_5\rangle_0/c^2$ and $\langle n_1n_6\rangle_0/c^2$ out of which the first is closer to the Monte Carlo one, in the 3×3 BPA.[12]

As we have said, the results for lower-order correlation functions are essentially those of Ref. 12 and will not be discussed further.

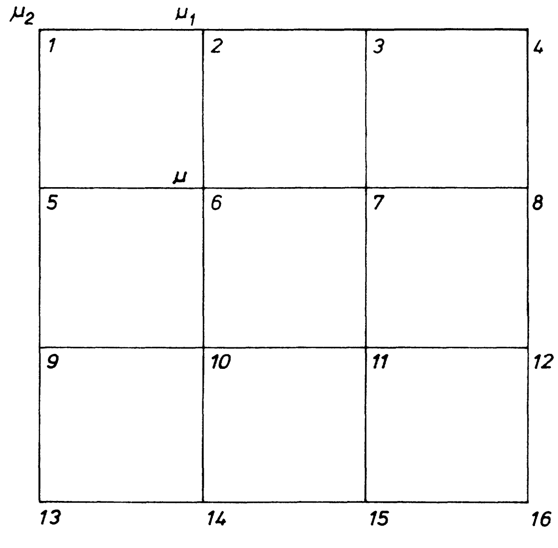

FIG. 1. The cluster replacing the lattice in the 4×4 BPA. The three different chemical potentials are indicated.

III. STATIONARY FLOW

We turn now to the dynamics of the model following a steady-state approach developed by Richards for a one-dimensional problem.[13] The lattice will be considered an $l\times m=N$ rectangle periodically repeated to cover the whole plane, in order to assure the possibility of stationary flow without sources of particles. The sites are labeled (ij) with $i=1,l$ and $j=1,m$ corresponding to the x and y axis, respectively. The particles, having positive electrical charge e, can jump to a NN vacant site with jump rate W only if all the other three NN's of the vacan-

TABLE I. The bulk fugacity obtained by two versions of the BPA: a 3×3 and a 4×4 cluster. A very slight difference is seen only at high concentration.

	c	0.1	0.15	0.2	0.25	0.3
z	3×3 BPA	0.176 78	0.372 38	0.720 13	1.3334	2.3844
	4×4 BPA	0.176 78	0.372 37	0.719 87	1.3303	2.3625

cy are empty (Fig. 2).

The average jump rate (for any time-dependent or stationary probability distribution) thus depends on the mean occupation of the four sites involved in this process. For the jump indicated in Fig. 2, it will be

$$\langle W_{ij,i+1j}\rangle = W\langle n_{ij}(1-n_{i+2j})(1-n_{i+1j+1})(1-n_{i+1j-1})\rangle \; . \tag{3.1}$$

A factor $1-n_{i+1j}$ is not needed because $n_{i+1j}=0$ in all configurations with $n_{ij}=1$.

In the presence of an electric field $\mathbf{E}=E\mathbf{e}_x$, the difference in energy between two NN sites in the x direction is eEa, a being the NN distance. In order to satisfy detailed balance, the rates for jumps parallel to the field are modified symmetrically as follows:

$$W^{E}_{ij,i\pm 1j}=W_{ij,i\pm 1j}(1\pm\Delta) \tag{3.2}$$

with

$$\Delta=\frac{eEa}{2kT} \; . \tag{3.3}$$

Equation (3.2) is written only to first order in E which is sufficient for the calculation of conductivity. In the following we will systematically discard higher-order terms.

If $\Omega=a^2$ denotes the area per site, then the charge current density is given by

$$j=\frac{N_p}{N\Omega}ev=\frac{cev}{\Omega} \; , \tag{3.4}$$

where v is the stationary flow mean velocity of the particles in the x direction. The velocity is calculated as the difference between the number of jumps to the right and of those to the left in unit time:

$$v=a\sum_{ij}(\langle W^{E}_{ij,i+1j}\rangle-\langle W^{E}_{ij,i-1j}\rangle) \; . \tag{3.5}$$

The correlation functions involved in Eq. (3.5) are steady-state averages, which are different from equilibrium ones. They retain, however, two useful symmetry properties: they are invariant under translation and under reflection with respect to the x axis. Using this and the labeling in Fig. 1 Eq. (3.5) becomes

$$\begin{aligned} v&=aN(\langle W^{E}_{6,7}\rangle-\langle W^{E}_{7,6}\rangle)\\ &=aNW[(1+\Delta)\langle n_6(1-n_3)(1-n_8)(1-n_{11})\rangle-(1-\Delta)\langle n_7(1-n_2)(1-n_5)(1-n_{10})\rangle]\\ &=aNW(\langle n_3n_6n_{11}\rangle-\langle n_2n_7n_{10}\rangle)+2aNW\Delta\langle n_6(1-n_3)(1-n_8)(1-n_{11})\rangle_0\\ &\equiv v_{\mathrm{corr}}+v_{\infty} \; . \end{aligned} \tag{3.6}$$

In the first term, v_{corr}, of the right-hand side of Eq. (3.6) all dynamic correlation functions but two cancel through symmetry. In the second term, v_{∞}, the average is taken in the equilibrium ensemble, that is zero order in E, due to the Δ prefactor, higher precision being superfluous, as previously remarked.

The vacancy availability factor is defined as[12]

$$V=\langle n_6(1-n_3)(1-n_8)(1-n_{11})\rangle_0/c \; . \tag{3.7}$$

We see that v_{∞} is proportional to V. It leads to the infinite-frequency conductivity

$$\sigma_{\infty}=\frac{j_{\infty}}{E}=\frac{cev_{\infty}}{\Omega}\frac{ea}{2kT\Delta}=\frac{N_pe^2a^2}{\Omega kT}WV \; . \tag{3.8}$$

The first term of Eq. (3.6), v_{corr}, contains all dynamic effects which make the dc conductivity σ lower than σ_{∞}. From the obvious relation

$$\langle n_3n_6n_{11}\rangle=\langle n_3n_6n_{11}\rangle_0+O(\Delta) \tag{3.9}$$

it follows that

$$\zeta\equiv\langle\langle n_3n_6n_{11}\rangle\rangle\equiv\langle n_3n_6n_{11}\rangle-\langle n_2n_7n_{10}\rangle=O(\Delta) \; . \tag{3.10}$$

TABLE I. The bulk fugacity obtained by two versions of the BPA: a 3×3 and a 4×4 cluster. A very slight difference is seen only at high concentration.

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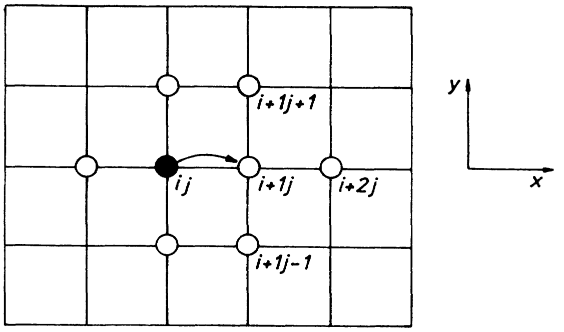

FIG. 2. A particle hops only if the vacancy has no occupied NN.

By $\langle\langle f(n)\rangle\rangle$ we denote the difference between $\langle f(n)\rangle$ and its symmetric with respect to the y axis.

While no exact calculation for ζ is possible, a good estimation can be derived from the rate equation for time-dependent ζ_t. In order to write it down, consider $\langle n_3 n_6 n_{11}\rangle_t$ in a time-dependent ensemble. It can be decreased by particles hopping from sites 3, 6, and 11 to sites 4, 18, 5, 12, and 15 (Fig. 3) and increased by the reverse jumps. This is described by the following rate equation:

$$\frac{d}{dt}\langle n_3 n_6 n_{11}\rangle_t = -(\langle W_{3,4} n_6 n_{11}\rangle_t + \langle W_{3,18} n_6 n_{11}\rangle_t + \langle W_{6,5} n_3 n_{11}\rangle_t + \langle W_{11,12} n_3 n_6\rangle_t + \langle W_{11,15} n_3 n_6\rangle_t) + \cdots , \tag{3.11}$$

where the ellipsis represents terms for reverse jumps. In the steady-state, the left-hand side of Eq. (3.11) is zero and symmetry can be used to get

$$0 = \langle (W^E_{5,6} - W^E_{6,5}) n_3 n_{11}\rangle + 2\langle (W^E_{4,3} - W^E_{3,4}) n_6 n_{11}\rangle + 2\langle (W_{18,3} - W_{3,18}) n_6 n_{11}\rangle . \tag{3.12}$$

An analog equation for $\langle n_2 n_7 n_{10}\rangle_t$ is needed to calculate ζ. Taking the difference between Eq. (3.12) and its symmetric with respect to the y axis we obtain

$$\begin{aligned} 5\zeta = {} & 2\langle\langle n_6 n_{11} n_{18}\rangle\rangle + 2\langle\langle n_4 n_6 n_{11}\rangle\rangle + \langle\langle n_3 n_5 n_{11}\rangle\rangle + 2\langle\langle n_6 n_9 n_{14} n_{17}\rangle\rangle + 4\langle\langle n_3 n_6 n_9 n_{14}\rangle\rangle \\ & + 2\langle\langle n_2 n_4 n_5 n_{10}\rangle\rangle + 2\langle\langle n_3 n_6 n_9 n_{11}\rangle\rangle + \langle\langle n_3 n_6 n_{11} n_{19}\rangle\rangle + 2\langle\langle n_3 n_6 n_{11} n_{14} n_{17}\rangle\rangle \\ & + 2\langle\langle n_1 n_3 n_6 n_{11} n_{14}\rangle\rangle + 2\langle\langle n_3 n_6 n_8 n_{11} n_{17}\rangle\rangle + 2\langle\langle n_1 n_3 n_6 n_8 n_{11}\rangle\rangle + 2\langle\langle n_1 n_3 n_8 n_{11} n_{17}\rangle\rangle \\ & - 2\langle\langle n_1 n_3 n_6 n_{11} n_{19}\rangle\rangle + \langle\langle n_1 n_3 n_6 n_9 n_{11} n_{19}\rangle\rangle - 2\langle\langle n_1 n_3 n_6 n_{11} n_{14} n_{17}\rangle\rangle - 2\Delta\zeta_0 , \end{aligned} \tag{3.13}$$

where ζ_0 is given by

$$\begin{aligned} \zeta_0 = {} & \langle n_3 n_6 n_{11}\rangle_0 + 2\langle n_4 n_6 n_{11}\rangle_0 - \langle n_3 n_5 n_{11}\rangle_0 - 2\langle n_3 n_6 n_8 n_{11}\rangle_0 - 2\langle n_3 n_6 n_9 n_{14}\rangle_0 - 2\langle n_2 n_4 n_5 n_{10}\rangle_0 \\ & + 2\langle n_3 n_6 n_9 n_{11}\rangle_0 + \langle n_3 n_6 n_{11} n_{19}\rangle_0 - 2\langle n_4 n_6 n_{11} n_{18}\rangle_0 + 4\langle n_3 n_6 n_8 n_9 n_{11}\rangle_0 \\ & + 2\langle n_3 n_6 n_8 n_9 n_{14}\rangle_0 - \langle n_1 n_3 n_6 n_9 n_{11}\rangle_0 - 2\langle n_1 n_3 n_6 n_{11} n_{19}\rangle_0 - 2\langle n_3 n_6 n_8 n_9 n_{11} n_{14}\rangle_0 + \langle n_1 n_3 n_6 n_8 n_9 n_{11}\rangle_0 . \end{aligned} \tag{3.14}$$

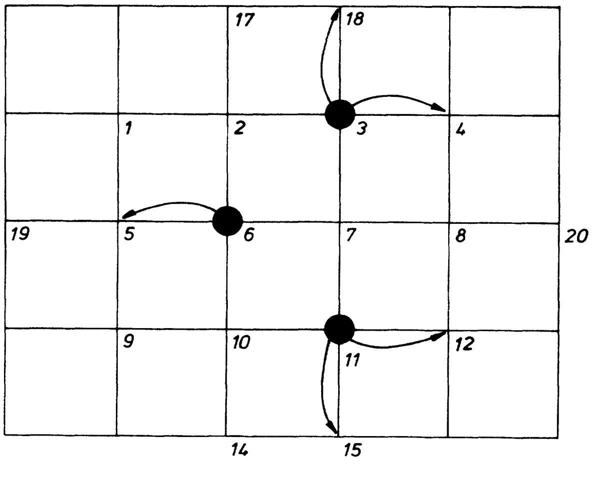

FIG. 3. Jumps contributing to the decrease of $\langle n_3 n_6 n_{11}\rangle_t$.

The dynamic correlations in Eq. (3.13) must be evaluated. For each of them an equation analog to Eq. (3.13) can be written, yielding an infinite hierarchy that must be truncated.

Let δ_i be the deviation of n_i from its equilibrium value c. Then, a correlator of, say, four particles, can be written as

$$\begin{aligned} \langle\langle n_a n_b n_c n_d\rangle\rangle &= \langle\langle (c+\delta_a)(c+\delta_b)(c+\delta_c)(c+\delta_d)\rangle\rangle \\ &= c^3 \sum_i \langle\langle \delta_i\rangle\rangle + c^2 \sum_{i,j} \langle\langle \delta_i \delta_j\rangle\rangle \\ &\quad + c \sum_{i,j,k} \langle\langle \delta_i \delta_j \delta_k\rangle\rangle + \langle\langle \delta_a \delta_b \delta_c \delta_d\rangle\rangle . \end{aligned} \tag{3.15}$$

We have, by symmetry,

$$\langle\langle \delta_i\rangle\rangle = 0, \quad \langle\langle \delta_i \delta_j\rangle\rangle = 0 \tag{3.16}$$

so we are left only with three-particle and higher correlators.

The first approximation is to retain only terms of order 3 in δ_i. Even so, we are left with the infinite number of equations relating three-particle correlation functions. We shall cut this knot by neglecting all of them, except ζ, for which we have

$$\zeta=\langle\langle \delta_3\delta_6\delta_{11}\rangle\rangle \ . \tag{3.17}$$

This is a compromise between feasibility and the quest for higher precision. The hierarchy of equations for the third-order correlators had to be truncated. We chose the simplest way of retaining only one correlation function which reduced to a minimum the amount of work. Pushing the degree of approximation further, we would have considered, beside ζ, the other three correlators of three particles involved in Eq. (3.13). Writing a similar equation for each of them, yields four coupled equations for four parameters. It turns out that by solving these equations, the value of ζ does not change significantly. We present here for illustration only a simplified version of the four equations, in which only third-order correlators were kept. The four parameters are denoted by

$$\begin{aligned}&\zeta=\langle\langle n_3n_6n_{11}\rangle\rangle, \quad \alpha=\langle\langle n_3n_5n_{11}\rangle\rangle \ ,\\ &\beta=\langle\langle n_4n_6n_{11}\rangle\rangle, \quad \gamma=\langle\langle n_6n_{11}n_{18}\rangle\rangle \ .\end{aligned} \tag{3.18}$$

The four equations, obtained in the same manner as Eq. (3.13), are

$$\begin{aligned}5\zeta-\alpha-2\beta-2\gamma&=2\Delta(\langle n_3n_5n_{11}\rangle_0-g_{30}-2\langle n_2n_5n_{11}\rangle_0) \ ,\\ -\zeta+10\alpha-2\beta&=2\Delta(2\langle n_2n_5n_{11}\rangle_0-g_{30}+\langle n_1n_7n_9\rangle_0\\ &\quad -2\langle n_2n_9n_{15}\rangle_0) \ ,\\ -\zeta-\alpha+8\beta&=2\Delta(g_{30}-\langle n_3n_5n_{11}\rangle_0+\langle n_2n_9n_{15}\rangle_0\\ &\quad -\langle n_3n_9n_{14}\rangle_0) \ ,\\ -\zeta+8\gamma&=2\Delta(\langle n_3n_6n_{14}\rangle_0+\langle n_3n_5n_{15}\rangle_0\\ &\quad -\langle n_2n_9n_{15}\rangle_0-\langle n_3n_9n_{14}\rangle_0) \ .\end{aligned} \tag{3.19}$$

Solving these, with the right-hand side (rhs) given by BPA at different concentrations shows that ζ is one order of magnitude higher than α, β, and γ. At small concentrations (for which the simplified versions of the equations are most suitable) all equilibrium correlators in the rhs are of the same approximate value (i.e., c^3) and so the rhs's of the last three equations are negligible with respect to the first rhs. It follows that $\zeta\approx 8\gamma\approx 7\beta\approx 8\alpha$ which, introduced in the first equation [which is Eq. (3.13) simplified] shows that the error made by neglecting α, β, and γ is, roughly, 10%.

In addition, let us examine the static values of the correlators in Eq. (3.13) which involve three sites. Besides g_{30} of Eq. (2.2c) we also consider the following:

$$\begin{aligned}&g_{31}=\langle n_3n_5n_{11}\rangle_0/c^3 \ ,\\ &g_{32}=\langle n_2n_5n_{11}\rangle_0/c^3 \ ,\\ &g_{33}=\langle n_2n_{11}n_{14}\rangle_0/c^3 \ .\end{aligned} \tag{3.20}$$

We have modified the site indices relative to Eq. (3.13) in order to obtain the best positioning of sites in the 4×4 cluster of Sec. I, since the BPA does not give translational invariant results.

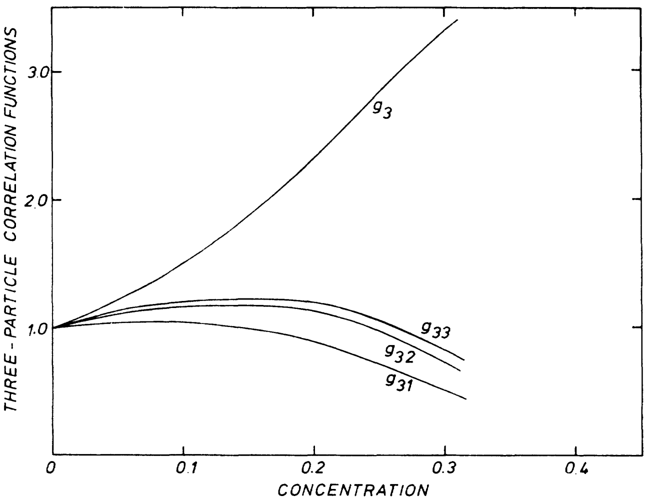

FIG. 4. The concentration dependence of four static three-particle correlators in the 4×4 BPA. g_{30} dominates at high c.

While g_{30} involves sites which in the ordered state above c_{cr} belong to the same sublattice the other three correlators contain sites of both sublattices, which leads to g_{30} being an increasing function of concentration, approaching the value 4 at $c=0.5$, while g_{31}, g_{32}, and g_{33} decrease at large c, all of them vanishing at $c=0.5$. Their dependence on c, for $c\leq 0.32$, is given in Fig. 4. While there is no apparent relation between static correlators and *differences* of dynamic ones, like $\langle\langle n_3n_6n_{11}\rangle\rangle$, it seems reasonable to consider ζ the most important of all three-particle dynamic correlators, based upon arguments like close spacing of sites and their position relative to the two sublattices.

Retaining only ζ of all three-particle correlation functions and evaluating all higher-order ones in terms of ζ

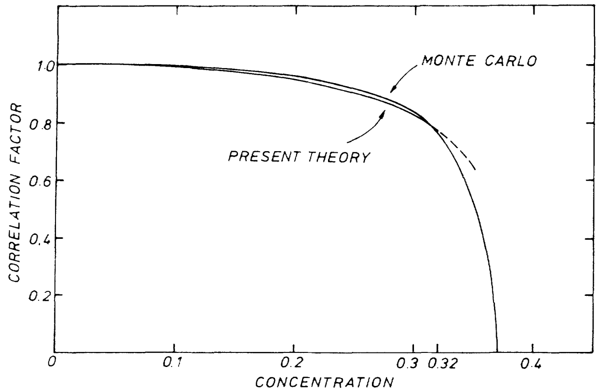

FIG. 5. The charge correlation factor of the extended hard-core model on the square lattice. The Monte Carlo results of Murch are presented along with the present theoretical evaluation.

by the δ decomposition, Eq. (3.13) becomes self-consistent. We get

$$\zeta = -\frac{2\Delta\zeta_0}{5-11c+8c^2-c^3} . \tag{3.21}$$

The charge correlation factor may now be written as

$$\begin{aligned} f &= \frac{\sigma}{\sigma_\infty} \\ &= 1 + \frac{v_{\text{corr}}}{v_\infty} = 1 + \frac{\zeta}{2\Delta c V} = 1 - \frac{\zeta_0}{cV(5-11c+8c^2-c^3)} . \end{aligned} \tag{3.22}$$

ζ_0 and V are evaluated in the 4×4 BPA and the resulting f is given in Fig. 5, together with the Monte Carlo results of Murch. We find fair agreement with the "experiment" for $c \le 0.32$, although only the first dynamic effect was taken into account.

IV. SUMMARY AND CONCLUSIONS

We have studied the square lattice gas with infinite NN repulsions with the main purpose of computing the dc hopping conductivity and its correlation factor f. This was done by a stationary flow formalism. The formula for σ involves static as well as dynamic correlation functions.

There are only two dynamic steady-state correlations in the velocity formula Eq. (3.6). A new type of correlation function was defined, so that ζ, Eq. (3.10), is the only dynamic correlator which enters the dc conductivity formula and the correlation factor Eq. (3.22). ζ was evaluated from the steady-state version of the evolution equation (3.11) by expressing all dynamic correlation functions in terms of ζ. In this approximation all other three-particle correlators vanish, while higher-order ones are dealt with as in Eq. (3.15), neglecting all products of δ-s other than ζ. The relative error was roughly estimated to be less than 0.1, but the final results for the correlation factor indicate that it is in fact of order 0.01.

For the calculation of the static correlation functions we have chosen the BPA in the version with a 4×4 cluster. This is an improvement in calculating higher-order correlators which in the 3×3 BPA would have been evaluated by factorization. Let us estimate the error induced in f by a single correlator for which 3×3 BPA is used. We consider the four-particle correlator of Eqs. (2.7) and (2.8). At $c=0.3$, g_{41}^{fa} is lower than g_{41} by 0.365. The effect on ζ_0 is lowering by $0.365c^4=0.002\,96$ which gives an f higher by 0.0265. The effect of *all* deviations of the correlators entering ζ_0, at $c=0.3$, is a value of f higher by ~4% than the Monte Carlo one, while the 4×4 BPA result is ~1% *lower*.

The final result, Fig. 5, shows good quantitative agreement with Monte Carlo experiments for $c \le 0.32$, in spite of the approximations used. Inclusion of more dynamic correlators would probably improve the calculated values of the correlation factor, but not enough to justify the amount of work required.

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