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Self-consistent kinetic theory for the Lorentz gas

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A new kinetic theory for a particle moving in a random array of static spherical scatterers is derived. By decoupling a higher-order dynamical correlation function, a kinetic equation is obtained comprising known approximations like ring, repeated-ring theory, and their self-consistent versions and, in particular, the equivalence of a recent mode-coupling theory and self-consistent ring theory is established. Vertex corrections, as represented by a new class of collision sequences, are shown to be essential for the Lorentz gas. New results for the Burnett coefficient are also presented.

I. INTRODUCTION

The Lorentz gas, a gas of mutually noninteracting particles, or equivalently one single particle, moving in a random array of fixed scatterers, is one of the simplest models in statistical mechanics.[1,2] The particular version that will be discussed mainly in this work deals with a hard-core interaction between the point particle and scatterers, but no interaction is assumed among the scatterers, so that they are allowed to overlap.

At low density of scatterers, cluster expansions and resummations of collision sequences were applied, and a nonanalytic density dependence of the diffusion constant was found.[3] Also, by means of a ring kinetic theory, a power-law long-time decay of the velocity correlation function (VCF) similar to the one in fluids, albeit with a different exponent, was derived.[4]

Both results were confirmed by molecular-dynamic experiments in two dimensions.[5-7] Moreover, these experiments in two, as well as in three, dimensions[5] show that with increasing density of scatterers, particle diffusion becomes more and more difficult, and a critical density n_c was found where diffusion stops.[6] Above n_c, the particle is localized.

Following the kinetic-theory approach, various approximations, especially the ring and repeated-ring theory, were examined.[8-10] These theories, while being quite accurate at low density, were found to fail to yield a critical density n_c and therefore a satisfactory description of the system at moderate and high densities of scatterers in two and three dimensions.

On the other hand, in a different line of approach, a mode-coupling theory, developed originally to investigate the dynamical conductivity of a quantum particle moving in a random potential,[11] was considerably improved and applied to the hard-core Lorentz gas.[12] It is based on the idea that the particle's current relaxation depends on its way of propagation and therefore should be evaluated self-consistently. The theory was shown to yield a diffusion-localization transition, and a critical density was predicted where the diffusion constant vanishes. Furthermore, the particle's dynamics as reflected in the diffusion constant and the velocity correlation function was studied in detail in the diffusion regime below n_c as well as in the localization regime above n_c and compared with experimental results. Almost quantitative agreement was found in two and three dimensions, an exception being the value of the critical density in two dimensions. Also, new predictions for the localization length and the dynamical structure factor were made. For a recent survey of this self-consistent current relaxation theory approach to describe the motion of a classical or quantum particle in a random potential, especially in two dimensions, we refer the interested reader to Ref. 13.

The purpose of the present work is twofold. Firstly, the connection between the mode-coupling theory[12] for the Lorentz gas and kinetic theory shall be examined. It is shown that the mode-coupling theory is equivalent to a self-consistent kinetic ring theory. Also, a new kinetic equation is suggested that allows us to discuss various other approximations like ring and repeated-ring theory and their self-consistent versions as special cases. It furthermore includes a new class of collision sequences which are shown to be equally important as the ring collisions. Secondly, a divergency problem of the collision operator for small frequency originating from its wave-number dependence shall be discussed. In the solution of the self-consistent ring and repeated-ring theories presented so far, the additional approximation of a wave-number independent collision operator was made.[12,10] It is shown that by abandoning this approximation, one encounters serious difficulties in the form of stronger infrared singularities peculiar to the Lorentz gas, which may be overcome only by including vertex corrections. The new collision sequences serve precisely this purpose and appear quite naturally in our theory.

The paper is organized as follows. In Sec. II, the formally exact generalized Boltzmann equation is derived. An approximate solution is presented which includes the exact expression for the Burnett correlation function in terms of collision operator matrix elements and results for Boltzmann's theory are presented. In Sec. III, various approximations to the collision operator are discussed. New results for the Burnett coefficient in a ring approximation are reported. Then the equivalence of the mode-coupling theory[12] and self-consistent ring theory is discussed. A new self-consistent kinetic equation is derived in Sec. IV, and the importance of vertex corrections is demonstrated. The work closes in Sec. V with a summary and conclusions.

II. GENERALIZED BOLTZMANN EQUATION

A. Derivation of the kinetic equation

In a particular version of the Lorentz gas we want to discuss in this paper, a point particle whose position and velocity are denoted by $\vec{r}_0$ and $\vec{v}_0$ is moving in a static array of randomly distributed scatterers of hard disks or spheres which are allowed to overlap. The positions of the scatterers in a certain configuration are denoted by $\vec{r}_1, \ldots, \vec{r}_N$ and their average density in volume V is $n=N/V$. The particle moves according to Newton's equation of motion generated by the "pseudo"-Liouville operator[14] $L_\pm = L_0 + L'_\pm$ and $A(t)=\exp(iL_\pm t)A(0)$, $t \gtrless 0$, for a particle's variable A. Here, $L_0=-i\vec{v}_0\vec{\nabla}_0$ and $L_\pm=\sum_{k\neq 0} T_\pm(0k)$ describe free streaming and collisions where

$$T_\pm(0k)=iv_0\sigma^{d-1}\int d\hat{r}\,\Theta(\mp\vec{v}_0\cdot\hat{r})\delta(\hat{r}\sigma-\vec{r}_{0k})\times(b_0-1)\ . \quad (1)$$

Here and in the following, $\hat{r}=\vec{r}/|\vec{r}|$ denotes a unit vector, σ is the scatterer's radius set equal to unity, d is the dimension of space, and the operator b_0 changes the particle's velocity $\vec{v}_0$ to $\vec{v}'_0 = b_0\vec{v}_0=\vec{v}_0-2(\vec{v}_0\hat{r})\hat{r}$.

The particle's phase-space distribution is shortly abbreviated as

$$f(1,t)=\delta(1-0)=\delta(\vec{r}_1-\vec{r}_0(t))\delta(\vec{v}_1-\vec{v}_0(t))\ ,$$

where $1\equiv(\vec{r}_1,\vec{v}_1)$ is an external field point. In the following, we are interested in the temporal and spatial evolution of the particle's phase-space correlation function $\phi(12,t)=(f(1,t)\,|\,f(2))$, where the parentheses denote a microcanonical average $(A\,|\,B)=\langle A^*B\rangle$ over scatterer configurations and particle variables with fixed magnitude v_0 of velocity. It is convenient to introduce the Laplace transform of $\phi(12,t)$ by

$$\phi(12,z)=i\int_0^\infty dt\,e^{izt}\phi(12,t)=(f(1)\,|\,(L_- - z)^{-1}\,|\,f(2))$$

for $\mathrm{Im}z>0$ and its spatial Fourier transform by

$$\phi(\vec{q},z)=\int d\vec{r}_{12}\exp(-i\vec{q}\cdot\vec{r}_{12})\phi(12,z)\ .$$

The kinetic equation for $\phi(12,z)$ is easily derived by applying the Zwanzig-Mori projection technique[15] with $P=|f(\bar{1}))(f(\bar{1})|=1-Q$ projecting onto the particle phase-space density. Here and in the following, a bar over a variable means integration over this variable. It may be written

$$[\phi^{B-1}(1\bar{2},z)-m(1\bar{2},z)]\phi(\bar{2}3,z)=\delta(13)\ , \quad (2)$$

where $\phi^B(12,z)$ is the Boltzmann propagator. It satisfies Boltzmann's kinetic equation

$$[\phi^{(0)-1}(1\bar{2},z)-t(1\bar{2})]\phi^B(\bar{2}3,z)=\delta(13) \quad (3)$$

with $\phi^{(0)}_{\vec{v}\vec{v}'}(\vec{q},z)=\delta_{\vec{v},\vec{v}'}/(\vec{q}\cdot\vec{v}-z)$ describing the free motion of the particle and $t(12) = -nT_-(1\bar{4})\delta(12)$ the instantaneous and local collision with a scatterer. The kernel $m(12,z)$ is the nontrivial part of the general collision operator

$$\mathscr{C}(12,z)=t(12)+m(12,z)\ . \quad (4)$$

Its formal expression as provided by the projection technique is

$$m(12,z)=-nT_+(1\bar{1})G_2(1\bar{1},2\bar{2};z)T_-(2\bar{2})\ , \quad (5a)$$

where the operators T_- and T_+ are operating on functions of the velocity on the right- and left-hand side, respectively. Introducing a phase-space particle-scatterer distribution

$$f(12)=\delta(1-0)\sum_{k\neq 0}\delta(2-k)/\sqrt{N}\ ,$$

the particle-scatterer correlation function G_2 may be written as

$$G_2(12,34;z)=(Qf(12)\,|\,(QL_-Q-z)^{-1}\,|\,Qf(34))\ . \quad (5b)$$

The main part of the present work will be concerned with the derivation and discussion of various approximations to G_2 describing the particle's propagation from phase point 3 to point 1 in the presence of scatterers at positions $\vec{r}_4$ and $\vec{r}_2$.

B. Solution of the kinetic equation

For a given approximation to $G_2(12,34;z)$ and $m(12,z)$ one then faces the problem of solving the kinetic equation, Eq. (2), for $\phi(12,z)$, which can be accomplished exactly only in exceptional cases. Therefore one has to introduce approximations in solving the kinetic equation, too, which, however, are controllable and may be improved systematically. The simplest approximation to the kinetic equation,

$$[\phi^{(0)-1}(1\bar{2},z)-\mathscr{C}(1\bar{2},z)]\phi(\bar{2}3,z)=\delta(13)\ , \tag{6}$$

which preserves particle-number conservation and allows for an analytic solution, is

$$\mathscr{C}_{\vec{v}\,\vec{v}'}(\vec{q},z)=\mathscr{C}_{11}(q,z)(\delta_{\vec{v},\,\vec{v}'}-|0\rangle\langle 0|)\ . \tag{7a}$$

Here, $|0\rangle\langle 0|$ is the projector onto the density state $|0\rangle=1$ in velocity space, and $\mathscr{C}_{11}(q,z)=\langle 1|\mathscr{C}(\vec{q},z)|1\rangle$ is the matrix element of the collision operator taken with the longitudinal current state $|1\rangle=\hat{v}_z\sqrt{d}$ (the wave vector $\vec{q}$ defines the z direction). Notice that particle-number conservation implies that $\mathscr{C}_{\vec{v}\,\vec{v}'}(\vec{q},z)$ integrated over velocities $\hat{v}$ or $\hat{v}'$ yields zero, which is easily verified using Eqs. (5a) and (1).

The approximation equation (7a), used in previous work on the electron localization problem[11] and the Lorentz gas,[12,9,10] is designed so as to yield the exact representation of the VCF in terms of its velocity relaxation kernel. In order to discuss the Burnett transport coefficient we have to improve upon this approximation. Quite generally in the kinetic model representation the collision operator $\mathscr{C}_{\vec{v}\,\vec{v}'}(\vec{q},z)$ is approximated by a $N+1$, $N+1$ matrix and a diagonal part in such a way that the chosen $N+1$ velocity states are treated exactly. Thus one writes

$$\mathscr{C}_{\vec{v}\,\vec{v}'}(\vec{q},z)=\sum_{\mu,\mu'=0}^{N}|\mu\rangle\gamma_{\mu\mu'}(q,z)\langle\mu'|+\alpha(q,z)\delta_{\mu\mu'} \tag{7b}$$

with $\alpha(q,z)=\langle N+1|\mathscr{C}(\vec{q},z)|N+1\rangle$ and $\gamma_{\mu\mu'}=\mathscr{C}_{\mu\mu'}-\alpha\delta_{\mu\mu'}$. Note that Eq. (7a) corresponds to Eq. (7b) with the simplest choice $N=0$. We employ in the following that $N=1$ approximation by including in addition to the density state $|0\rangle$ and the velocity state $|1\rangle$ the further state

$$|2\rangle=(\hat{v}_z^2-1/d)d\left[\frac{(d+2)}{2(d-1)}\right]^{1/2}.$$

With the use of Eq. (7b) the kinetic equation, Eq. (6), may be solved analytically.

Of greatest interest is the density correlation function $\phi(q,z)=\phi_{00}(q,z)$ of the particle. In general, it may be written in the following way which stresses the small frequency and wave-number behavior due to particle-number conservation:

$$\phi(q,z)=-\frac{1}{z+q^2D(q,z)} \tag{8a}$$

defining the generalized diffusion constant $D(q,z)$. Two other important correlation functions are intimately related to the density correlations by the continuity equation, namely, the cross correlation $\phi_{10}(q,z)=\phi_{01}(q,z)$ between density and longitudinal current and the longitudinal current correlation function $\phi_{11}(q,z)$. They may be expressed by $D(q,z)$ in the following way:

$$\phi_{10}(q,z)=\frac{\sqrt{d}}{v_0}\,\frac{qD(q,z)}{z+q^2D(q,z)}\ , \tag{8b}$$

$$\phi_{11}(q,z)=\frac{d}{v_0^2}\,\frac{zD(q,z)}{z+q^2D(q,z)}\ . \tag{8c}$$

These equations hold irrespective of any approximation to the general collision operator as long as it satisfies particle-number conservation.

The small wave-number limit of $D(q,z)$ is the velocity correlation function

$$D(z)=\lim_{q\to 0}D(q,z)=\left(\vec{v}_z\left|\frac{1}{L_- - z}\right|\vec{v}_z\right)\ . \tag{9}$$

Furthermore, the diffusion coefficient D is the zero frequency limit

$$D=-i\lim_{z\to 0}D(z)\ , \tag{10}$$

if that limit exists.

Assuming that a small wave-number expansion of $D(q,z)$ is possible in the form

$$D(q,z)=\sum_{n=0}^{\infty}(-q^2)^nD_{2n+2}(z)\ , \tag{11}$$

one may define the Burnett correlation function $D_4(z)$ and higher-order correlation functions.[16,17] The Burnett coefficient

$$D_4=-i\lim_{z\to 0}D_4(z) \tag{12}$$

contains new information on the spatial extension of the transport process, and some results were obtained in molecular-dynamic experiments[16,6] for the two-dimensional system.

The solution of the kinetic equation (6) with Eq. (7b) for $N=1$ yields for the generalized diffusion coefficient,

$$D(q,z) = -\frac{v_0^2/d}{z+\mathscr{C}_{11}(q,z)+M_{11}^{(0)}(q,z+\mathscr{C}_{22}(q,z))}\ , \tag{13}$$

where $M_{11}^{(0)}(q,z')$ is the free-particle current relaxation kernel. It may be obtained from the known free-particle density correlation function

$$\phi_{00}^{(0)}(q,z) = \int \frac{d\hat{v}}{\Omega_d}\frac{1}{\vec{q}\,\vec{v}_0 - z} = -\frac{1}{z}\,{}_2F_1\left[\tfrac{1}{2},1;d/2;\left[\frac{qv_0}{z}\right]^2\right] \tag{14}$$

by using Eqs. (8a) and (13) for zero density of scatterers. In Eq. (14), Ω_d is the surface of the d-dimensional unit sphere and ${}_2F_1$ denotes the hypergeometric function.[18] From Eq. (13) one easily obtains the velocity and Burnett correlation function. Assuming that $\mathscr{C}(q,z)$ allows for a small wave-number expansion in the form

$$\mathscr{C}_{11}(q,z) \simeq \mathscr{C}_{11}(z) + (qv_0)^2\mathscr{C}_{11}^{(2)}(z) + \cdots\ ,$$

$$\mathscr{C}_{22}(q,z) \simeq \mathscr{C}_{22}(z) + \cdots\ ,$$

and using

$$M_{11}^{(0)}(q,z') = -\frac{2(d-1)}{d(d+2)}\frac{(qv_0)^2}{z'}\left[1+O\left[\frac{qv_0}{z'}\right]^2\right]$$

for small qv_0/z', one finds

$$D(z) = -\frac{v_0^2/d}{z+\mathscr{C}_{11}(z)}\ , \tag{15a}$$

$$D_4(z) = D^2(z)\left[\frac{2(d-1)}{(d+2)}\frac{1}{z+\mathscr{C}_{22}(z)} - d\,\mathscr{C}_{11}^{(2)}(z)\right]. \tag{15b}$$

Let us stress that Eqs. (15) are exact formulas. They express the velocity and Burnett correlation functions in terms of matrix elements of the general collision operator $\mathscr{C}(\vec{q},z)$. The element $\mathscr{C}_{11}(z)$, called velocity relaxation kernel or dynamical friction coefficient, was shown to be the crucial quantity in the dynamical theory of the diffusion versus localization transition in the Lorentz gas,[12] diverging for small frequency at the critical density, while $\mathscr{C}_{22}(z)$ stays finite.[19] Note that the wave-number dependence of the collision operator $\mathscr{C}(\vec{q},z)$ is important for the Burnett as well as higher-order correlation functions. Exact expressions for these in analogy to Eq. (15b) may be obtained by increasing the order N of approximation in Eq. (7b) in a straightforward way.

C. Boltzmann theory results

Here, we want to report the results of Boltzmann's theory, Eq. (3), which only considers uncorrelated binary collisions and is expected to be a good approximation at low density of scatterers. It therefore often serves as a reference in comparison to other theories in order to study the effects of dynamic correlations. The eigenfunctions and eigenvalues of the Lorentz-Boltzmann operator can be evaluated in all dimensions and so any correlation function may be calculated to any desired degree of accuracy using, for instance, Eq. (7b) or the continued fraction method.[19] In one and three dimensions even at exact solution is possible.[3,8]

Since the Lorentz-Boltzmann operator is rotational invariant, its eigenfunctions are the Gegenbauer polynomicals[18] $C_k^{(\alpha)}(\hat{q}\,\hat{v})$ ($\alpha=(d-2)/2$, $k=0,1,2,\ldots$), the generalizations of the Legendre polynomials in $d=3$ to d dimensions. For the eigenvalues we find

$$\nu_k = \gamma\frac{4k(k+d-2)}{(2k+d-2)^2-1}\ , \tag{16}$$

where $\gamma = n\sigma^{d-1}v_0\pi^{(d-1)/2}/\Gamma((d+1)/2)$ is the collision frequency. Of course, $\nu_0=0$ due to particle-number conservation. In $d=3$, $\nu_k=\gamma$ for all $k\neq 0$; for $d\neq 3$, the eigenvalues are rapidly and monotonically converging to $\nu_\infty=\gamma$ for increasing k. Thus in the Boltzmann approximation we find $\mathscr{C}_{kn}(q,z)=i\nu_k\delta_{kn}$. This independence of wave number and frequency reflects the fact that the hard-core collisions are local and instantaneous. Inserting into Eqs. (15), the Boltzmann results for the velocity and Burnett correlation functions are

$$D^{(0)}(z) = -\frac{v_0^2/d}{z+i\nu_1}\ , \tag{17a}$$

$$D_4^{(0)}(z) = \frac{2(d-1)}{(d+2)}[D^{(0)}(z)]^2\frac{1}{z+i\nu_2}\ . \tag{17b}$$

While the VCF decays purely exponentially in time,

$$\phi_{vv}^{(0)}(t) = \frac{v_0^2}{d}\exp(-\nu_1 t)\ , \tag{18a}$$

the Burnett correlation function $\phi_4^{(0)}(t)$ starts from zero at time zero, is increasing proportional t^2, and decays exponentially at long times

$$\phi_4^{(0)}(t)=\begin{cases}\frac{2}{5}\left[\frac{v_0^2}{3}\right]^2 t^2 e^{-\nu_1 t} & \text{for } d=3\\ \frac{2(d-1)}{(d+2)}\left[\frac{v_0^2}{d}\right]^2\frac{1}{(\nu_2-\nu_1)}\left[te^{-\nu_1 t}-\frac{1}{(\nu_2-\nu_1)}(e^{-\nu_1 t}-e^{-\nu_2 t})\right] & \text{for } d\neq 3.\end{cases} \tag{18b}$$

The diffusion and Burnett coefficients in the Boltzmann approximation are, using Eq. (17),

$$D^{(0)}=v_0^2/d\nu_1\ , \tag{19a}$$

$$D_4^{(0)}=\frac{2(d-1)}{(d+2)}(D^{(0)})^2/\nu_2\ . \tag{19b}$$

Higher-order Burnett correlation functions of Boltzmann's theory may be obtained by making a small wave-number expansion of the exact solution of Boltzmann's equation in the form of a continued fraction,

$$D^{(0)}(q,z)=-\frac{v_0^2/d}{z+i\nu_1+}\,\frac{a_1^2}{z+i\nu_2+}\,\frac{a_2^2}{z+i\nu_3+}\cdots \tag{20}$$

with

$$a_n^2=-(qv_0)^2\frac{(n+1)(n+d-2)}{(2n+d-2)(2n+d)}$$

and comparing coefficients of powers of q^2. This continued fraction is especially convenient for numerical evaluation.

III. APPROXIMATIONS OF THE COLLISION OPERATOR

A. Second-order kinetic equation

In order to find approximations to the general collision kernel $m(12,z)$, we will in this section derive a second kinetic equation for the particle-scatterer correlation function G_2, Eq. (5b), and examine some approximations to its memory kernel. Again applying the projection technique, this time to G_2 with the projector

$$P_2=|F(\bar{1}\bar{2}))\frac{1}{g(\bar{1}\bar{2})}(F(\bar{1}\bar{2})|=1-Q_2\ ,$$

where $F(12)=Qf(12)$, we find the second-order kinetic equation

$$[\phi^{B-1}(1\bar{3})\delta(2\bar{4})+T_-(12)\delta(1\bar{3})\delta(2\bar{4})-m_2(12,\bar{3}\bar{4};z)]G_2(\bar{3}\bar{4},56;z)=\delta(15)\delta(26)g(12)\ . \tag{21}$$

For the overlapping Lorentz gas the static pair correlation $g(12)$ between the particle and a scatterer is $g(12)=\theta(r_{12}-\sigma)$. The memory kernel m_2 and G_2 may be expressed by a particle-two-scatterer correlation function G_3,

$$m_2(12,34;z)=-nT_+(1\bar{1})G_3(1\bar{1}2,3\bar{3}4;z)T_-(3\bar{3}) \tag{22a}$$

with

$$G_3(123,456;z)=(F_3(123)|(Q_2QL_-QQ_2-z)^{-1}|F_3(456))\ , \tag{22b}$$

where $F_3(123)=Q_2f(123)$ is the particle-two-scatterer phase-space density

$$f(123)=\delta(1-0)\sum_{k\neq 0,i\neq 0}\delta(2-k)\delta(3-i)/N\ ,$$

properly orthogonalized.

B. Ring and repeated-ring theory

Let us first discuss the simplest approximation for G_2. Neglecting all but the first term in the bracket on the left-hand side (lhs) of Eq. (21), one finds $G_2(12,34;z)=\phi^B(13,z)\delta(24)g(12)$, which, inserted in Eq. (5a), yields the so-called ring-collision operator[20,3]

$$m_R(12,z) = -nT_+(1\bar{3})\phi^B(12,z)T_-(2\bar{3}) \ . \tag{23}$$

It describes a dynamically correlated process where the particle collides with a certain scatterer, then propagates in the system according to Boltzmann's equation, which means suffering only uncorrelated binary collisions, and finally collides with the initial scatterer again. This may be represented graphically as in Fig. 1(a).

A well-known consequence of the ring collisions is the nonanalytic small frequency expansion[4] of the VCF

$$D(z)/iD^{(0)} = 1 + \frac{\Omega_d}{nd(2\pi)^d}\begin{cases}\frac{1}{2}\pi(-1)^{(d-1)/2}\zeta^d, & d=1,3,\dots\\ (-1)^{d/2}\zeta^d \ln\zeta, & d=2,4,\dots\end{cases} \tag{24a}$$

where $\zeta = (z/iD^{(0)})^{1/2}$, which implies for long times a power-law decay[4]

$$\phi_{vv}(t) \sim -\frac{(D^{(0)})^2 2\pi}{n(4\pi D^{(0)}t)^{d/2+1}} \ . \tag{24b}$$

Evaluating analogously the velocity relaxation kernal at finite wave number in lowest order in density we find that the leading nonanalytic term in frequency may be simply expressed by its zero wave-number limit

$$\mathscr{C}_{11}(q,z) \simeq f_l^2(q)\mathscr{C}_{11}(z) \ , \tag{25}$$

Here, $f_l(q) = df_1'(q)$ with $f_1(x) = \Gamma(d/2) \times (2/x)^{d/2-1}J_{d/2}(x)$ may be replaced by its small wave-number limit $f_l(q=0)=1$, since at small density the mean free path is large compared to the scatterer radius $\sigma = 1$. It is not difficult to realize that the other matrix elements of the ring-collision operator show no stronger singularity than $\mathscr{C}_{11}(q=0,z)$. Together with Eq. (25) this then implies that the higher-order Burnett correlation functions decay at large times with the same exponent as the VCF. This was conjectured by Alley and Alder[16] on the basis of results from molecular-dynamic experiments in two dimensions. For the Burnett coefficient this conjecture was confirmed recently on the basis of ring theory also.[21] Using Eqs. (25) and (15b) and noticing that $\mathscr{C}_{22}(z)$ is less singular than $\mathscr{C}_{11}(z)$ we find in particular for the leading nonanalytic part for small frequency

$$D_4(z) \simeq a\frac{D_4}{D}D(z) \ , \tag{26}$$

where in the low-density limit $a = 2$. Alley and Alder[16] tested Eq. (26) in $d = 2$ at a medium density $n\sigma^2 = 0.2$ with $a = 1$ and found good agreement with experimental results, which suggests that the ring theory is no longer accurate at this density.

The ring-collision events were also shown to lead to logarithmic terms in the low-density expansion of the diffusion constant, for instance, in two dimensions[3]

$$D/D^{(0)} = 1 + \tfrac{4}{3}n\ln n + \cdots \ . \tag{27a}$$

One may also evaluate the contribution of the ring collisions to the Burnett coefficient, and we find in $d = 2$ a similar expansion as for the diffusion constant

$$D_4/D_4^{(0)} = 1 + \tfrac{8}{5}n\ln n + \cdots \ , \tag{27b}$$

and in $d = 3$ the nonanalytic term is proportional to $n^2\ln n$, also similar to the diffusion constant.[22]

Returning to the discussion of the second-order kinetic equation, we note that the second term on the lhs of Eq. (21) gives rise to the so-called repeated-ring collisions. The repeated-ring kinetic equation, obtained by keeping the first two terms on the lhs of Eq. (21), solving for G_2, and inserting into Eq. (5a), allows the particle to collide with the same scatterer more than once while its propagation between these scattering events occurs according to Boltzmann's equation. A simple example is shown in Fig. 1(b). In one dimension, the exact solution of the repeated-ring equation yields the correct result of a zero diffusion coefficient.[8] But besides this, the VCF of the repeated-ring equation shows a long-time power law decay proportional to $t^{-3/2}$, in contrast to the exact solution for the one-dimensional Lorentz gas which shows exponential decay.[8] In two and three dimensions, the repeated-ring equation was demonstrated[8] to exhibit no critical density n_c, where the diffusion constant vanishes and the

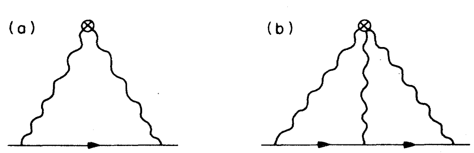

FIG. 1. Dynamically correlated collision events contribution to $m(12,z)$. ——▶——, Boltzmann phase-space propagator ϕ^B; ∿∿∿, binary-collision operator T; and ⊗, scatterer. (a) Ring collision. (b) Simple repeated-ring collision.

theory fails[10] above roughly one-fourth the experimental n_c.

C. Self-consistent ring theory

Notice that the ring-collision operator could be obtained more easily directly from Eq. (5a) by factorizing the dynamical particle-scatterer correlation

$$G_2(12,34;z) \simeq \phi(13,z)\delta(24) \tag{28}$$

and replacing the phase-space propagator by its Boltzmann's approximation ϕ^B. Inserting G_2 into Eq. (5a), the resulting memory kernel

$$m_2(12,z) = -nT_+(1\bar{3})\phi(12,z)T_-(2\bar{3}) \tag{29}$$

is a very natural extension of the ring memory kernel. It may be called a self-consistent ring approximation (SRA) since the memory kernel $m(12,z)$ has to be determined by solving Eq. (29) and the kinetic equation (2) simultaneously. The self-consistency requirement, as discussed previously in work on the localization problem of a quantum particle in a random potential,[11] expresses the intuitive physical picture that the friction the particle experiences depends on the way the particle is propagating. The importance of this fact becomes most obvious at high density of scatterers when the particle is localized.

With the additional approximation of ignoring the wave-number dependence of $m(q,z)$, that is, replacing $m(q,z)$ by $m(q=0,z)$, Eq. (29) represents the mode-coupling theory discussed extensively,[12] and we will therefore only outline briefly its main results.

At low density it reproduces the results of the ring theory as is to be expected. But at higher densities the feedback mechanism introduced by the self-consistency leads to important new qualitative effects. The diffusion constant is reduced relative to its Boltzmann value, and at a certain critical density n_c it drops to zero linearly with density. Above n_c the particle is trapped in a finite region, the extension of which defines a localization length. Approaching the critical density from above, the localization length increases with an inverse square root. The critical density is found to be $n_c = d/V_d$, where V_d is the volume of the d-dimensional unit sphere.[23] While this value is in surprisingly good agreement with molecular-dynamics[5] and Monte Carlo[24] results in $d=3$, its values turns out to be too large by a factor of 1.7 in two dimensions, compared to molecular-dymamics[6] and percolation theory[25] results (see Table I).

The VCF exhibits a $|\omega|^{d/2}$ small-frequency singularity below n_c signifying a $t^{-(d+2)/2}$ long-time tail, while at the critical density the singularity is $|\omega|^{1/2}$ with logarithmic corrections in $d=2$, and above n_c the long-time decay is exponential. This was used successfully to explain[12] the experimentally observed[6] apparent density variation of the long-time exponent in two dimensions.

The repeated-ring approximation may also be extended to its self-consistent version[8] (SRRA) and for $d=1$ a zero diffusion coefficient and an exponential decay of the VCF at long times in agreement with the exact solution were found.[8] Recently, some results for the SRRA using the single relaxation kernel approximation, Eq. (7a), ignoring the wave-number dependence of the collision operator, were obtained in one, two, and three dimensions with a variational method.[26] Extending their method to d dimensions we find that the diffusion constant goes to zero at a critical density $n_c = (d/V_d)(1-1/d)$, which is compared with the result $n_c = d/V_d$ of the self-consistent ring theory and experimental results in Table I.

IV. A NEW SELF-CONSISTENT KINETIC EQUATION

There are mainly two ways to improve the theories presented so far. The first one is to abandon the simplifying assumption of ignoring the

TABLE I. Critical density n_c for the Lorentz gas of d-dimensional spherial overlapping scatterers. MD is the molecular-dynamic experiments. MC is the Monte Carlo experiments. SRA, Refs. 12 and 23. SRRA, Ref. 26.

Dimension	1	2	3	d
MD		≈ 0.37[a]	≈ 0.72[b]	
MC		0.359 ± 0.002[c]	0.81 ± 0.05[d]	
SRA	$\frac{1}{2}$	0.64	0.72	d/V_d
SRRA	0	0.32	0.48	$(d/V_d)(1-1/d)$

[a]Reference 6.
[b]Reference 5 (extrapolation).
[c]Reference 25.
[d]Reference 24.

wave-number dependence of the general collision operator $\mathscr{C}(\vec{q},z)$. The second one is to include new classes of collision sequences, in particular, those yielding infinite contributions to the velocity relaxation kernel at the critical density. It turns out that these two points are closely related. It is easy to verify that the inclusion of a wave-number dependence in the collision operator of the self-consistent ring or repeated-ring theory would change the results drastically, e.g., one would find zero diffusion in $d=2$ at all densities and a long-time power-law exponent $d/2$ instead of $d/2+1$ for $d>2$.

The reason for this, which will be elaborated on later, is that in these self-consistent theories certain collision sequences with strong infrared singularities are summed; but other collision sequences with the same strong infrared singularities, up to a sign by reasons of symmetry, are completely ignored. Hence these new collision sequences have to be identified and included to allow for a cancellation of these singularities. A hint for the nature of these new collisional events may be obtained from the density expansion of the diffusion constant. It was found[3] that there are collisional events contributing to the leading density correction of D which are not included in the ring or repeated-ring theory, or their self-consistent versions.

A. Factorization approximation, vertex function, and kinetic equation

Continuing with the discussion of Eq. (21) we suggest a simple factorization approximation for the particle-two-scatterer correlation function G_3,

$$G_3(123,456;z) \simeq \phi(14,z)[\delta(25)\delta(36)+\delta(26)\delta(35)] \; . \tag{30a}$$

Notice that G_3 by definition has to be symmetric under the interchange of scatterers at positions $\vec{r}_2$ with $\vec{r}_3$ or at $\vec{r}_5$ with $\vec{r}_6$. We want to point out here that the theories discussed so far violated this important symmetry by keeping only the first term on the right-hand side (rhs) of Eq. (30a) which leads to the difficulties discussed above.

Inserting Eq. (30a) into Eq. (22a), one readily obtains

$$m_2(12,34;z) = -nT_+(1\bar{1})\phi(13,z)T_-(3\bar{1})\delta(24) - nT_+(14)\phi(13,z)T_-(32) \; . \tag{30b}$$

This is the new approximation to be discussed further after insertion into Eq. (21). Let us introduce Fourier transforms by

$$G_2(\vec{k},\vec{q}-\vec{k};\vec{\kappa},\vec{q}-\vec{\kappa};z) = \int d1234\, e^{-i\vec{k}\cdot\vec{r}_1 - i(\vec{q}-\vec{k})\cdot\vec{r}_2} G_2(12,34;z) \times e^{i\vec{\kappa}\cdot\vec{r}_3 + i(\vec{q}-\vec{\kappa})\cdot\vec{r}_4}$$

and analogously for the other correlation functions. Defining a vertex function $\Gamma(\vec{k},\vec{q}-\vec{k};z)$ by

$$\phi^B(\vec{k},z)\Gamma(\vec{k},\vec{q}-\vec{k};z) = \sum_{\vec{\kappa}} G_2(\vec{k},\vec{q}-\vec{k};\vec{\kappa},\vec{q}-\vec{\kappa};z)T_-(\vec{\kappa}-\vec{q}) \; , \tag{31}$$

the integral equation (21) may be rewritten in the following way:

$$\Gamma(\vec{k},\vec{q}-\vec{k};z) = T_-(\vec{k}-\vec{q}) - \sum_{\vec{\kappa}} [T_-(\vec{k}-\vec{\kappa}) - m_2(\vec{k},\vec{q}-\vec{k};\vec{\kappa},\vec{q}-\vec{\kappa};z)]\phi^B(\vec{\kappa},z)\Gamma(\vec{\kappa},\vec{q}-\vec{\kappa};z) \; , \tag{32a}$$

where transforming Eq. (30b),

$$m_2(\vec{k},\vec{q}-\vec{k};\vec{\kappa},\vec{q}-\vec{\kappa};z) = -n\sum_{l} T_+(\vec{k}-\vec{l})\phi(\vec{l},z)T_-(\vec{l}-\vec{k})\delta_{\vec{k},\vec{\kappa}} - nT_+(\vec{q}-\vec{\kappa})\phi(\vec{k}+\vec{\kappa}-\vec{q},z)T_-(\vec{k}-\vec{q}) \; , \tag{32b}$$

and $\sum_{\vec{\kappa}}$ denotes the wave-number integral $\int d^d\kappa/(2\pi)^d$. The generalized Boltzmann kinetic equation (2) for the phase-space propagator $\phi(\vec{q},z)$ may be rewritten as

$$[\phi^{B\,-1}(\vec{q},z) - m(\vec{q},z)]\phi(\vec{q},z) = 1 \; , \tag{32c}$$

where the memory kernel $m(\vec{q},z)$ may be expressed by the vertex function by

$$m(\vec{q},z) = -n\sum_{\vec{k}} T_+(\vec{q}-\vec{k})\phi^B(\vec{k},z)\Gamma(\vec{k},\vec{q}-\vec{k};z) \; . \tag{32d}$$

Notice that in these equations the velocity indices are omitted to simplify the notation. Equations (32a)–(32d) have to be solved self-consistently.

B. Relation to self-consistent ring theory

It is quite instructive to reconsider the previously discussed theories as various special cases of Eq. (32) by dropping certain collision sequences. The ring theory is equivalent to truncating Eq. (32a) for the vertex function in lowest order yielding $\Gamma_R(\vec{k},\vec{q}-\vec{k};z)\simeq T_-(\vec{k}-\vec{q})$, which leads to the following approximation for $m(\vec{q},z)$:

$$m_R(\vec{q},z)=-n\sum_{\vec{k}} T_+(\vec{q}-\vec{k})\phi^B(\vec{k},z)T_-(\vec{k}-\vec{q}) , \tag{33}$$

where the subscript R denotes ring theory. On the other hand, in the self-consistent ring theory, one also keeps the first term on the rhs of Eq. (32b). The vertex function is thus approximated by

$$\Gamma_{SR}(\vec{k},\vec{q}-\vec{k};z)= T_-(\vec{k}-\vec{q}) + m_{SR}(\vec{k},z)\phi^B(\vec{k},z) \times\Gamma_{SR}(\vec{k},\vec{q}-\vec{k};z) , \tag{34a}$$

where

$$m_{SR}(\vec{q},z)=-n\sum_{\vec{k}} T_-(\vec{q}-\vec{k})\phi(\vec{k},z) \times T_-(\vec{k}-\vec{q}) , \tag{34b}$$

and the subscript SR denotes self-consistent ring theory. It is easy to verify by solving Eq. (34a) for Γ_{SR} and inserting in Eq. (32c) that m_{SR} as defined in Eq. (34b) is indeed the memory kernel of the phase-space propagator of the self-consistent ring theory. In Eqs. (34a) and (34b), a very special class of an infinite number of collision sequences is summed that has the effect of dressing the Boltzmann propagator and replacing it by the self-consistent one. This is illustrated graphically in Fig. 2. The first graph on the rhs in Fig. 2 represents the ring collisions, the next graph a ring within a ring, and so on.

As will be shown in Sec. IV C, the ring diagram $m_R(q,z)$ has at finite wave number q a stronger small-frequency singularity than at zero wave number, e.g., in $d=2$ it is diverging as $\ln z$ at $q\neq 0$, while at $q=0$ it behaves as $z\ln z$. Hence if $m_R(q,z)$ is iterated in the self-consistent theory and inserted, for example, in the second diagram on the rhs in Fig. 2, this stronger singularity stemming from nonzero wave numbers will dominate the singularity of the self-consistent ring diagram even at zero wave number. This has various consequences which, however, are not supported by experiments, e.g., a long-time power-law exponent $d/2$ instead of $d/2+1$ for the VCF in $d>2$, or even zero diffusion at all densities in $d=2$.

This stronger singularity of $m_R(q,z)$ for $q\neq 0$ can be suppressed artificially within self-consistent ring theory by neglecting the wave-number dependence of $m_R(q,z)$ altogether, thereby cutting off this unwanted feedback mechanism at finite wave numbers.[12] The same arguments apply for the self-consistent repeated-ring theory.[26]

The derivation of the self-consistent ring theory presented here indicates a solution to this problem. As explained before, in the SRA only the first part on the rhs of Eq. (32b), or equivalently in Eq. (30a), is taken into account, which leads to the dressing of the propagator ϕ^B in the memory kernel $m_R(q,z)$. But in that way, one violates the symmetry of interchangeability of scatterers in G_3 [see Eq. (30a)]. The expansion of the rhs of Eq. (32a) in powers of the T operator makes it very clear that to every diagram included in the SRA corresponds a diagram which is not included and may be obtained from the former by interchanging scatterer labels. These new diagrams are generated by the second term on the rhs of Eq. (30a) or (32b) and they have the form of vertex corrections. The simplest pair of graphs con-

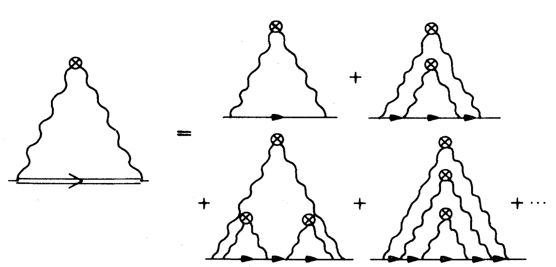

FIG. 2. Memory kernel $m(12,z)$ in SRA. $\Rightarrow$, self-consistent phase-space propagator ϕ (see also Fig. 1).

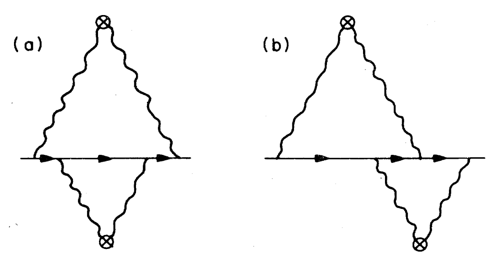

FIG. 3. Contributions to memory kernel $m(12,z)$. (a) Insertion of a ring diagram in a ring [Fig. 1(a)], generated by self-consistent ring theory. (b) Vertex correction, not contained in self-consistent ring theory.

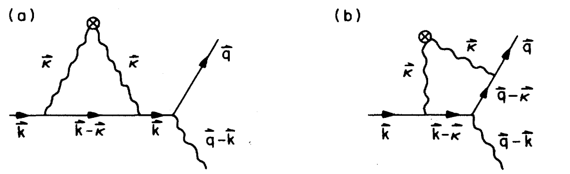

FIG. 4. Vertex functions $\Gamma(\vec{k},\vec{q}-\vec{k};z)$ corresponding to Fig. 3.

tributing to $m(q,z)$ is depicted in Fig. 3. Let us mention in passing that the collision sequence described by Fig. (3b) was shown[3] to contribute to the leading density correction of the diffusion constant in $d=3$. In Sec. IV C, it will be shown that these new diagrams also exhibit strong small-frequency singularities, and moreover, exactly cancel the leading singularities of the ring diagrams.

C. Cancellation of leading singularities

In the following, we will demonstrate the cancellation of the leading singularities in the lowest-order diagrams shown in Fig. 3. It is convenient to introduce a complete set of states in velocity space, the simplest of which are the density state $|0\rangle$ and the current states $|k\rangle$ $(k=1,\ldots,d)$. To discuss the small-frequency and wave-number properties of the present theory, it is sufficient to keep only these states since possible singularities are caused by the presence of a diffusive particle mode in the system.

First, we consider the ring-theory graph, Fig. 3(a), or rather the corresponding expression for the vertex function Γ. An examination of the diagram with the relevant velocity states attached to it leads us to the following special diagram, depicted in Fig. 4(a), as a candidate for a singularity. Its mathematical expression is

$$\Gamma^{(a)}(\vec{k},\vec{q}-\vec{k};z)=nV_d^3(v_0^2/d)f(\vec{k}-\vec{q})\sum_{\vec{\kappa}}f^2(\vec{\kappa})(\vec{k}\cdot\hat{\kappa})\phi_{00}^B(\vec{k}-\vec{\kappa},z)\frac{(\vec{k}-\vec{q})\hat{q}}{k^2+z/D(k,z)}\ , \tag{35}$$

where

$$\Gamma^{(a)}(\vec{k},\vec{q}-\vec{k};z)=\sum_{n,m}\hat{k}_n\langle n\,|\,\Gamma^{(a)}(\vec{k},\vec{q}-\vec{k};z)\,|\,m\rangle\hat{q}_m\ .$$

Here $\phi_{00}^B(k,z)$ denotes the density correlation function, and the cross-correlation function between density and current as given in Eq. (8b) was inserted together with the relevant matrix element of the T operator $\langle 0\,|\,T_-(\vec{k})\,|\,i\rangle=-(v_0/\sqrt{d}\,)V_d\vec{k}_i f(k)$ with $f(x)=(d/x)f_1(x)$ and $f_1(x)=\Gamma(d/2)(2/x)^{(d-2)/2}J_{d/2}(x)$. Equation (35) may be rewritten as

$$\Gamma^{(a)}(\vec{k},\vec{q}-\vec{k};z)=V_d f(\vec{q}-\vec{k})\frac{k(\vec{k}-\vec{q})\hat{q}}{k^2+z/D(k,z)}R(k,z)\ , \tag{36a}$$

where $R(k,z)$ is the lowest-order ring diagram

$$R(k,z)=nV_d^2(v_0^2/d)\sum_{\vec{\kappa}}f^2(\kappa)(\vec{\kappa}\cdot\hat{k})^2\phi_{00}^B(\vec{k}-\vec{\kappa},z)\ . \tag{36b}$$

In two dimensions $R(k,z)$ exhibits an infrared divergency for $k\neq 0$ due to the diffusion mode. The singular part $R_s(k,z)$ of R is

$$R_s(k,z)=nV_d^2(v_0^2/d)k^2f^2(k)\sum_{\vec{\kappa}}\phi_{00}^B(\kappa,z) \tag{36c}$$

diverging proportional to ln$z$ for small frequency at $k\neq 0$. Inserting this into Eq. (32d) this would entail a divergency of $m(q,z)$ at zero wave number.

We now turn to the vertex correction, depicted in Fig. 4(b), and show that it cancels the divergency of the ring diagram in Eq. (36b). Inserting special velocity states the corresponding expression is

$$\Gamma^{(b)}(\vec{k},\vec{q}-\vec{k};z)=nV_d^3(v_0^2/d)f(\vec{k}-\vec{q})\sum_{\vec{\kappa}}f^2(\kappa)\phi_{00}^B(\vec{k}-\vec{\kappa},z)\frac{(\vec{\kappa}\cdot\hat{k})(\vec{q}-\vec{\kappa})\cdot(\vec{k}-\vec{q})(\vec{\kappa}\cdot\hat{q})}{(\vec{\kappa}-\vec{q})^2+z/D(\vec{\kappa}-\vec{q},z)}\ . \tag{37}$$

This vertex correction also exhibits an infrared divergency in $d=2$ for $k\neq 0$, and this divergent part may be written

$$\Gamma_s^{(b)}(\vec{k},\vec{q}-\vec{k};z)=-nV_d^3(v_0^2/d)f(\vec{k}-\vec{q})f^2(k)\frac{k(\vec{q}-\vec{k})^2(\vec{k}\cdot\hat{q})}{(\vec{k}-\vec{q})^2+z/D(\vec{k}-\vec{q},z)}\sum_{\vec{\kappa}}\phi_{00}^B(\kappa,z)\ . \tag{38}$$

This singular contribution thus cancels the singular counterpart of $\Gamma^{(a)}(\vec{k},\vec{q}-\vec{k};z)$, Eq. (36a), at $q=0$, as can easily be verified. Hence both diagrams together lead to a memory kernel $m(q,z)$ which is not divergent at $q=0$. These arguments may be repeated for higher-order diagrams.

It is interesting to note that a cancellation similar to the one of the leading singularities of the two diagrams in Fig. 3 for zero wave number described above occurs also in the related quantum-mechanical problem of the Anderson localization, and it was shown[27,28] that the classical diffusion pole does not yield divergent contributions to the current relaxation kernel at zero wave number in a diagrammatic weak-coupling expansion.

V. SUMMARY AND CONCLUSIONS

The difficulties discussed in Sec. IV C are connected with the fact that the vertex describing the coupling of the current mode to the product of the particle and scatterer density vanishes at zero wave number. On the other hand, in the case of a fluid,[29] there is in addition a coupling to the transverse current of the fluid. The corresponding vertex does not vanish at zero wave number. This first of all implies that the small-frequency singularity of the VCF is stronger in the fluid than in the Lorentz gas and, secondly, that the singularity of $m_R(q,z)$ at nonzero wave number is not stronger than at zero wave number. In this sense, the Lorentz gas is very peculiar and different from the fluid. In particular, the mode-coupling theory applied successfully in liquids has to be modified by vertex corrections. The present work is a first step in this direction in the framework of kinetic theory. In this way, it is possible to clarify and elucidate the relation between the mode-coupling theory presented recently for the Lorentz gas[12] and different versions of kinetic theories. It is shown that the mode-coupling theory is a self-consistent ring theory. Moreover, it is shown that the existing self-consistent ring and repeated-ring theories for the Lorentz gas are incomplete because of missing collision sequences in the form of vertex corrections. A kinetic equation is presented in Eq. (32) which includes an important new class of collisional events, and these are shown to cancel the spurious small-frequency singularities of the self-consistent ring theory. The simplest of these collision sequences was found also to contribute to the leading density correction to Boltzmann's theory in a low-density expansion of the diffusion constant.[3]

In view of the cancellation of the spurious infrared singularities of the straightforward extension of the mode-coupling theory to finite wave numbers the present kinetic theory appears to be promising. But its detailed predictions, especially for long times or near the critical density, which are entailed by the wave-number dependence of the current relaxation kernel, have still to be worked out. Owing to this wave-number dependence which leads to integral equations instead of transcendental equations[12] and due to the necessary inclusion of quite intricate collision sequences, the solution of the self-consistency equations derived here is more difficult than before and is deferred to a separate investigation.

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