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Dynamical theory of diffusion and localization in a random, static field

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An approximation scheme is derived to calculate the velocity-autocorrelation function for the Lorentz model of overlapping hard discs and hard spheres. The theory describes a feedback between the particle-density correlations and the current relaxation rate, and is shown to give a percolation edge, a transition from the normal diffusion phase to nondiffusion phase characterized by a finite localization length l_0. Near the edge the low-frequency velocity spectrum for either phase is evaluated, thereby finding diffusivity to approach zero linearly with the separation parameter $\epsilon = (n - n_c)/n_c$, where n_c is the critical density, while l_0 diverges like $1/\sqrt{\bar{\epsilon}}$. A power-law long–time decay of the velocity-autocorrelation function is found for the diffusion phase. Upon approaching n_c the hydrodynamic regime shrinks to zero, and a transition in the power-law exponent from its low-density value which is dependent on dimension to a value of 3/2 for both dimensions is predicted.

I. INTRODUCTION

Among the various models in statistical mechanics used to study diffusion is a class called the Lorentz models in which classical point particles without mutual interaction move in a medium of randomly distributed stationary scatterers.[1] Different Lorentz models are distinguished by the shape of the scatterers and the scattering interaction. In addition one can specify whether the scatterers are allowed to overlap, and consider different space dimension. Even though dynamics is absent in the scattering medium, such models are of fundamental interest because they exhibit nontrivial correlation effects which are also characteristic of diffusion in normal fluids. In a density expansion of the diffusion coefficient D one encounters divergences of the same kind and resummation leads to nonanalytic terms.[2] It is known from kinetic theory analysis of ring collisions that the velocity-autocorrelation function $K(t)$ also decays nonexponentially at long times, albeit the power law differs from the corresponding fluid.[3] More recently, the velocity-autocorrelation function at moderately low density, which shows the same qualitative deviations from exponential decay at intermediate times as the fluid, has been analyzed by a calculation that includes the effects of repeated ring collisions.[4]

Theoretical results on the hard-sphere Lorentz model can be checked by computer molecular dynamics simulation. Such studies have been carried out to observe the nonanalytic behavior[5] in the density variation of D and the power-law decay of $K(t)$ in two dimensions.[5-7] Moreover, computer results on overlapping hard discs have established the critical density when D vanishes and the detailed behavior of $K(t)$ below and above the critical region.[6] These results serve to emphasize the importance of understanding the model behavior over the entire density range.

At present no theory exists for the spherical Lorentz model that is valid outside the low-density range, although abnormal diffusion or localization has been studied in a special case, the Ehrenfest windtree model.[1,8] The purpose of the present paper is to analyze both phases of the hard-sphere Lorentz model with overlap, including the region of phase transition. Our approach is based on a recently developed theory to study the electrical conductivity of quantum particles moving in random potentials.[9] We will derive for two and three dimensions self-consistent equations for the density and velocity-autocorrelation functions and examine the general features of the theory. It will be shown that the critical density n_c emerges quite naturally from the analysis of either phase. Moreover, one finds a density variation of the long-time decay of $K(t)$ in the same way as observed by computer simulation.

The paper is organized as follows. In Sec. II the correlation functions for the density and current fluctuations of the moving particle are introduced and related to the velocity-autocorrelation function $K(t)$. The diffusivity D and the localization length l_0 are used as hydrodynamic parameters to characterize the diffusion and localization phases respectively. The current relaxation kernel $M(z)$, which is the memory function for $K(t)$, is the central quantity in the present formulation. In Sec. III all phase space correlation functions for the moving particle, in particular the density fluctuation function $\phi(q,z)$, are expressed approximately in terms of $M(z)$. A mode-coupling approximation is used to express $M(z)$ in terms of the various correlation functions thereby establishing a closed nonlinear equation for $M(z)$. In Sec. IV various consequences of the theory are discussed by finding asymptotic solutions of the self-consistency equations. The results are summarized in Sec. V.

II. THE CORRELATION FUNCTIONS

Let us consider a point particle of unit mass whose position and momentum will be denoted by $\vec{r}_0(t)$ and $\vec{v}_0(t)$. This particle is assumed to move through an ensemble of N identical stationary scatterers distributed over the d-dimensional ($d=2,3$) unit volume $V=1$ at random positions $\vec{r}_1, \vec{r}_2, \ldots, \vec{r}_N$. Each scatterer represents a hard core potential of radius σ, so that the total potential felt by the particle reads $V=\sum_{i=1}^N v(|\vec{r}_0 - \vec{r}_i|)$, $v(r)=\infty$ for $r<\sigma$, and $v(r)=0$ for $r>\sigma$. For such a system the kinetic energy is a conserved quantity and we take the equilibrium momentum distribution to be $\phi(\vec{k})=a_d(v_0)\delta(k-v_0)$. Here and in the following, $|\vec{p}|=p$, $|\vec{k}|=k$, etc. The normalization factor $a_d(v_0)$ is determined from $\sum_{\vec{k}}\phi(\vec{k})=1$, therefore $a_2=2\pi/v_0$ and $a_3=2\pi^2/v_0^2$.

The correlation function of variable A and B is defined as $\phi(t)=\langle\delta A^*(t)\delta B\rangle$, where $\delta X=X-\langle X\rangle$ and the average refers to the various distributions of scatterer positions. The correlation function for complex frequency z is given as the Laplace transform

$$\phi(z)=\pm i\int dt\,\theta(\pm t)e^{izt}\phi(t),\quad \mathrm{Im}z\gtrless 0\,, \tag{1}$$

with the usual spectral representation. The spectral function is the discontinuity across the real axis, $\phi(\omega\pm i0)=\phi'(\omega)\pm i\phi''(\omega)$, and can be obtained as the Fourier transform of $\frac{1}{2}\phi(t)$. By defining a scalar product in the linear space of dynamical variables by $(A|B)=\langle\delta A^*\delta B\rangle$, the correlation function $\phi(z)$ can be expressed as the resolvent matrix element of the Liouville operator L, $\phi(z)=(A|(L-z)^{-1}|B)$. Operator L generates the time evolution, $L|A)=-i|\dot{A})$. For autocorrelation functions, $A=B$, the spectral function is real and non-negative. In this case the Zwanzig-Mori projection operator formalism[10] can be applied to rewrite $\phi(z)$ in terms of the normalization $g=(A|A)$, the restoring force $\Omega=\omega/g$, with $\omega=(A|LA)$, and a relaxation kernel $\Gamma(z)=\gamma(z)/g$ formed with the fluctuating forces $F_A=QLA$,

$$\gamma(z)=(F_A|(QLQ-z)^{-1}|F_A)\,. \tag{2}$$

One obtains

$$\phi(z)=-[z-\Omega+\Gamma(z)]^{-1}g\,. \tag{3}$$

Here $P=|A)g^{-1}(A|$ is the projector onto $|A)$, and $Q=1-P$ projects on the complement. The preceding equations hold also for a set of k linear independent variables $A_1, A_2, \ldots, A_k$, provided all quantities are interpreted as k by k matrices.[11]

To describe the motion of the particle through the system, the autocorrelation function of the density fluctuations $\rho(\vec{q})=\exp(i\vec{q}\cdot\vec{r}_0)$ is of primary interest,

$$\phi(q,t)=(\rho(\vec{q},t)|\rho(\vec{q}))\,. \tag{4}$$

We will also need the correlation function of current and density

$$\phi_{\mathrm{CD}}(q,t)\hat{q}_i=(j_i(\vec{q},t)|\rho(\vec{q}))\,, \tag{5}$$

and the current correlation

$$\phi_{ik}(\vec{q},t)=(j_i(\vec{q},t)|j_k(\vec{q})) =\hat{q}_i\hat{q}_k\phi_l(q,t)+(\delta_{ik}-\hat{q}_i\hat{q}_k)\phi_t(q,t)\,, \tag{6}$$

where $\hat{\vec{q}}$ denotes the unit vector $\vec{q}/q$. The current density is given by $\vec{j}(\vec{q})=\vec{v}_0\rho(\vec{q})$, and the continuity equation, $L\rho(\vec{q})=\vec{q}\,\vec{j}(\vec{q})$, allows one to express the mixed correlation function and the longitudinal current correlation function in terms of $\phi(q,z)$,

$$\phi_{\mathrm{CD}}(q,z)=[1+z\phi(q,z)]/q\,, \tag{7a}$$

$$\phi_l(q,z)=[1+z\phi(q,z)]z/q^2\,. \tag{7b}$$

While in principle ϕ_{CD} and ϕ_l do not exhibit any new physics beyond that expressed by $\phi(q,z)$, they will enter the following theory in quite a different context.

The density correlation function describes how the probability distribution $\langle\delta(\vec{r}-\vec{r}_0(t))\rangle$ spreads out in space within the time interval t, if such a distribution was created at time zero.[12,13] In particular one finds for the root mean square displacement $C_2(t)=\langle[\Delta x(t)]^2\rangle/2$ with $\Delta x(t)=r_0^x(t)-r_0^x(0)$,

$$C_2(t)=-\frac{1}{2}\frac{d^2\phi(q,t)}{dq_x^2}\,,\quad q=0\,. \tag{8}$$

In the case of self-diffusion the generalized Langevin equation, Eq. (3), is particularly simple. With $g=1$ and the restoring force $\Omega=0$, one finds

$$\phi(q,z)=\frac{1}{z+q^2K(q,z)}\,, \tag{9a}$$

where

$$K(q,z)=(j(\vec{q})|(QLQ-z)^{-1}|j(\vec{q}))\,, \tag{9b}$$

and $QL\rho(\vec{q})/q = j(\vec{q})$ is the longitudinal current. When $q \to 0$, $Q \to 1$, the homogeneous kernel $K(q=0,z) = K(z)$ is again a correlation function, the Laplace transform of the velocity autocorrelation of the moving particle, $K(t) = \langle v_0^x(t) v_0^x \rangle$. Applying the Zwanzig-Mori formalism to $K(z)$ one gets

$$K(z) = -\frac{1}{z + M(z)}(v_0^2/d), \tag{10}$$

with a memory function $M(z)$ determined, according to Eq. (2), by the fluctuating forces the particle experiences due to scatterers.[14] We will refer to $M(z)$ as the current relaxation kernel.

The expressions for time correlation functions $\phi(q,z)$ and $K(z)$ in terms of their respective memory functions $K(q,z)$ and $M(z)$ are completely general and equally applicable to the diffusion and localization phases of the system. In the diffusion phase the long-wavelength and low-frequency behavior of $K(q,z)$ may be expressed as

$$K(q, \omega \pm i0) = \pm iD, \quad q < q_h, \text{ and } \omega < \omega_h, \tag{11a}$$

where D is the self-diffusion coefficient and is given by

$$D = v_0^2 / dM''(\omega), \quad \omega = 0, \tag{11b}$$

with $M''(\omega)$ being the imaginary part of $M(z=\omega + i0)$. In Eq. (11a), ω_h and q_h define a hydrodynamic regime of frequencies and wavelengths. Equation (11a) leads to the result that on a time scale larger than $t_h = 1/\omega_h$ and a length scale exceeding $l_h = 1/q_h$ the density fluctuation spectrum is given by

$$\phi''(q,\omega) = \frac{Dq^2}{\omega^2 + (Dq^2)^2}. \tag{11c}$$

By analogy with the hydrodynamics of a normal fluid one would expect ω_h and l_h to be determined by the collision frequency and mean-free path. One has therefore the classical behavior of the particle spreading out over any finite volume within a finite time interval. Correspondingly, density fluctuations propagate according to a simple diffusion equation, and for large times $C_2(t) \propto Dt$. From Eqs. (8), (11b), and (11c) one has the classical relations between the mean square displacement, the velocity autocorrelation and diffusivity.[13]

In the localization phase one can continue to describe the system behavior in terms of $K(q,z)$ and $M(z)$. Since D is now zero, it is useful to introduce the concept of a stiffness of the medium by considering the displacement response to an external homogeneous time dependent force due to a potential $r_0^x \cdot E(t)$. This response is expressed by the dynamical polarizability $\chi(z)$ associated with the displacement correlation function, or

$$\chi''(\omega)/\omega = \tfrac{1}{2}\int dt\, e^{i\omega t}\langle r_0^x(t) r_0^x \rangle. \tag{12a}$$

Since $\dot{r}_0^x = v_0^x$, $\chi(z)$ is related to the velocity-autocorrelation function[11,15]

$$\chi(z) = \frac{1}{z}K(z). \tag{12b}$$

The static polarizability $\chi^0 = \chi(z=0)$ is the displacement induced by a static unit field. One can associate with it a length l_0 such that $l_0^2 = \chi^0 = 1/s$, where s is the stiffness.

The long-wavelength, low-frequency behavior of $K(q,z)$ when the particle is localized may be expressed as

$$K(q, \omega \pm i0) = \omega/s, \quad q < q_h, \text{ and } \omega < \omega_h, \tag{13a}$$

with

$$s = -(d/v_0^2)\omega M'(\omega), \quad \omega = 0. \tag{13b}$$

One derives from Eq. (9a) in leading order of the small parameters q and z

$$\phi(q,z) = -\frac{1}{z}f(q), \tag{13c}$$

with $f(q) = [1 + (ql_0)^2]^{-1}$. The density correlation function exhibits a zero frequency pole, therefore density fluctuations are nonergodic variables.[15] One can define an isolated compressibility $\kappa(q) = 1 - f(q)$,

$$\kappa(q) = (ql_0)^2/[1 + (ql_0)^2], \tag{13d}$$

which is different from the thermodynamic compressibility. Consequently, a density fluctuation does not disappear. Rather, it gets redistributed in a region of characteristic extension l_0. In particular, Eq. (8) yields

$$C_2(t) = l_0^2, \quad t > t_h. \tag{13e}$$

Hence, kernel $M(z)$ is a convenient tool to characterize the diffusion as well as the localization phase in physical terms. In one phase the low-frequency part of M is dissipative and determines the transport coefficient D, in the other phase it is reactive and fixes the thermodynamical quantity s.

III. SELF-CONSISTENT CURRENT RELAXATION APPROXIMATION

A. A single kernel approximation

To ensure the correct free particle asymptote $\Phi^{(0)}(q,z)$ for the density correlation function in the limit of small interaction or large $\vec{q}$ it is a convenient detour to consider the correlation function for the phase space density $f_{\vec{k}}(\vec{q}) = \delta[\vec{k} - \vec{v}_0(t)] \exp i\vec{q}\cdot\vec{r}_0(t)$. The generalized Langevin equation of Zwanzig-Mori then reads

$$(z - \vec{k}\cdot\vec{q}/m)\Phi_{\vec{k}\vec{p}}(\vec{q}, z) = -\delta_{\vec{k}\vec{p}}\Phi(\vec{k}) - \sum_{\vec{l}} C_{\vec{k}\vec{l}}(\vec{q}, z)\Phi_{\vec{l}\vec{p}}(\vec{q}, z), \quad (14)$$

where we denote by C the sum of the kernel Γ in Eq. (3) and the interaction part of Ω. In a single relaxation-time approximation one would write[16]

$$\sum_l C_{kl}(\vec{q}, z)\Phi_{\vec{l}\vec{p}}(\vec{q}, z) \cong \frac{i}{\tau}\left(\Phi_{\vec{k}\vec{p}}(\vec{q}z) - \Phi(\vec{k})\sum_{\vec{k}}\Phi_{\vec{k}\vec{p}}(\vec{q}, z)\right), \quad (15)$$

where the relaxation time τ would be determined in terms of the self-diffusion coefficient. We suggest that as an improvement of this approximation i/τ should be replaced by a frequency-dependent kernel $\tilde{M}(z)$. Equation (14) then can be solved easily

$$\Phi_{\vec{k}\vec{p}}(\vec{q}, z) = \Phi^{(0)}_{\vec{k}\vec{p}}(\vec{q}, z) - \left(\sum_{\vec{l}}\Phi^{(0)}_{\vec{k}\vec{l}}(q, \tilde{z})\right)\frac{\tilde{M}(z)}{1 + \tilde{M}(z)\Phi^{(0)}(q, \tilde{z})} \times\left(\sum_{\vec{l}}\Phi^{(0)}_{\vec{l}\vec{p}}(q, \tilde{z})\right). \quad (16)$$

Here $\tilde{z} = z + \tilde{M}(z)$ and the superscript (0) denotes the free system functions. One gets in particular

$$\Phi^{(0)}(q, z) = \frac{1}{qv_0}\phi_0(\tilde{z}/qv_0), \quad (17a)$$

$$\phi_0(x) = \begin{cases} -1/(x^2-1)^{1/2}, & d=2 \\ \frac{1}{2}\log\frac{x-1}{x+1}, & d=3 \end{cases} \quad (17b)$$

and the corresponding function for the interacting system reads in our approximation

$$\Phi(q, z) = \frac{\Phi^{(0)}(q, z + \tilde{M}(z))}{1 + \tilde{M}(z)\Phi^{(0)}(q, z + \tilde{M}(z))}. \quad (17c)$$

Exploiting the small qv_0 expansion of $\Phi^{(0)}(q, z)$

$$1/\Phi^{(0)}(q, \tilde{z}) + M(z) = -z + \frac{1}{d}\frac{(qv_0)^2}{z}[1 + O((qv_0/\tilde{z})^2)], \quad (18)$$

one verifies that approximation (17c) implies Eq. (9a) with $K(z, q=0)$ given by Eq. (10) where $M(z) = \tilde{M}(z)$. Hence, $\tilde{M}(z)$ has been identified as the current relaxation kernel. One checks easily that the solution (16) also implies Eqs. (7). Approximation Eq. (17) therefore expresses $\Phi(q, z)$ in terms of the current relaxation kernel $M(z)$ such that $\Phi(q, z)$ has the correct analytical properties, obeys the leading sum rules, preserves hydrodynamical behavior with particle conservation, and leads to the exact expression for $K(z)$. It generalizes the single relaxation-time approximation such that the constant i/τ is replaced by a kernel $M'(\omega) + iM''(\omega)$. The absorptive part $M''(\omega)$ generalizes $1/\tau$ to a frequency-dependent function, while reactive processes now can be described by the real part $M'(\omega)$. It is the $M'(\omega)$ term, which plays the essential role in the present theory.

B. A mode-coupling approximation

In this subsection the essential approximation of the present theory will be formulated. The current relaxation kernel $M(z)$ will be approximately expressed in terms of integrals over the correlation functions. To treat the singular hard core dynamics the pseudo Liouville operator introduced by Ernst *et al.*[17] will be employed. One introduces non-Hermitian operators $L_\pm$ in place of L in all the resolvent matrix elements for $\mathrm{Im}z \gtrless 0$ such that the discontinuous velocity change during a collision process is reproduced correctly. The use of the pseudo Liouville operator in the Zwanzig-Mori formalism has been studied before for hard-sphere gases to extend the Boltzmann equation to higher densities[18] and to develope a mode-coupling theory for hard-sphere liquids.[19]

For the present problem one writes $L_\pm = L_0 + L'_\pm$ where L_0 is the free-particle part and $L'_\pm$ is the interaction part. One finds

$$L'_\pm\vec{v}_0 = -2i\int d\vec{r}\,\delta(r-\sigma)(\vec{v}_0\cdot\hat{\vec{r}})^2\hat{\vec{r}}\,\theta(\mp\vec{v}_0\cdot\hat{\vec{r}}) \times\sum_{n=1}^{N}\delta(\vec{r} - \vec{r}_0 + \vec{r}_n). \quad (19)$$

To evaluate matrix elements with $L'_\pm$ one has to make use of the model property that the scatterer positions $\vec{r}_n$ are completely uncorrelated. Hence, $\langle\sum_{n=1}^{N}\delta(\vec{r} - \vec{r}_0 + \vec{r}_n)\rangle = n$ for $|\vec{r}_0 - \vec{r}_n| > \sigma$, with n denoting the scatterer density. For the structure factor of the scatterers formed with the scatterer density fluctuation $P(\vec{\kappa}) = \sum_n \exp(i\vec{\kappa}\cdot\vec{r}_n)$, one gets $\langle P^*(\vec{\kappa})P(\vec{\kappa})\rangle = n$. Because hard core collisions are instantaneous $M(z)$ will approach a nonzero constant for large frequencies. It is advantageous to separate out this constant by writing

$$M(z) = \pm i\nu + m(z), \quad \mathrm{Im}z \gtrless 0. \quad (20)$$

In the Zwanzig-Mori formalism the constant ν appears naturally as the restoring force term; from Eq. (3) $-i\nu=(v_0^* L'_- v_0^*)d/v_0^2$, so ν is proportional to the binary collision rate. From this expression one finds $\nu=8\sigma n v_0/3$ for $d=2$ and $\nu=\pi n\sigma^2 v_0$ for $d=3$. The kernel $m(z)$ is defined by Eq. (2),

$$m(z)=\sum_{ab}(F_\pm|F_a)\left(F_a\left|\frac{1}{QLQ-z}\right|F_b\right)(F_b|F_\pm)\,. \tag{21}$$

Here $F_\pm$ are the fluctuating forces for the longitudinal current $F_\pm=QL'_\pm j(\vec{q})\sqrt{d}/v_0$, $q=0$, and F_a represents a complete orthonormalized set of modes with momentum $q=0$.

The simplest modes are the particle excitations f_α, such as the density and current fluctuations. They read, e.g.,

$$\begin{aligned} f_0(\vec{q})&=\sum_{\vec{k}} f_{\vec{k}}(\vec{q})\,,\\ f_1(\vec{q})&=\sum_{k}(\vec{k}\cdot\vec{q}/qmv_0)f_{\vec{k}}(q)\sqrt{d}\,,\\ f_2(\vec{q})&=\sum_{\vec{k}}[d(\vec{k}\cdot\vec{q}/qmv_0)^2-1][(d+2)/2(d-1)]^{1/2}\,. \end{aligned} \tag{22}$$

These modes do not contribute to Eq. (21) since they are projected out by Q. The next simplest modes are pair excitations with momentum $-\vec{k}$ and particle excitations with momentum $\vec{k}$,

$$F^{(2)}_{\vec{k}\alpha}=P(-\vec{k})f_\alpha(\vec{k})\sqrt{N}\,. \tag{23a}$$

If the particle motion were unaffected by the scatterers, the propagator in Eq. (21) would become

$$\left(F^{(2)}_{\vec{k}\alpha}\left|\frac{1}{QLQ-z}\right|F^{(2)}_{\vec{p}\beta}\right)=\delta_{\vec{k}\vec{p}}\,\Phi_{\alpha\beta}(\vec{k},z)\,. \tag{23b}$$

As approximation for $m(z)$ we will (i) take into account only the pair modes given by Eq. (23a) and (ii) factorize the correlations of particle and potential fluctuations according to Eq. (23b):

$$m(z)=\frac{1}{N}\sum_{\vec{k}}\sum_{\alpha\beta}V_\alpha(\vec{k})\Phi_{\alpha\beta}(\vec{k}z)V_\beta(\vec{k})\,, \tag{24a}$$

$$V_\alpha(k)=(F_\pm|P(-\vec{k})f_\alpha(\vec{k}))\,. \tag{24b}$$

If the propagator on the right-hand side of Eq. (24a) is replaced by $\Phi^{(0)}_{\alpha\beta}$, one gets zero.[2] This reflects the fact that all binary collision contributions are taken care of already by ν in Eq. (20). To improve convergence of the α-β sum in Eq. (24a) let us replace $\Phi_{\alpha\beta}(\vec{q},z)$ by $\Phi_{\alpha\beta}(\vec{q},z)-\Phi^{(0)}_{\alpha\beta}(\vec{q},z+M(z))$. Thereby one guarantees that every term in Eq. (24a) vanishes for small $M(z)$ or large q. With Eq. (16) one gets

$$m(z)=-\frac{1}{N}\sum_{\vec{k}}\sum_{\alpha\beta}M(z)V_\alpha(\vec{k})\Phi^{(0)}_{\alpha 0}(k\tilde{z})V_\beta(\vec{k})\Phi^{(0)}_{0\beta}(k\tilde{z})/[1+M(z)\Phi^{(0)}(q,\tilde{z})]\,. \tag{24c}$$

Not all single particle modes contribute to the preceding sum. Classifying the modes $f_\alpha(\vec{k})$ by their magnetic quantum number with respect to the axis q, rotational invariance implies $\Phi^{(0)}_{\alpha 0}(k,z)=0$ unless α belongs to the quantum number zero.

The first three of these modes are given in Eq. (22). These modes can all be generated by applying L_0, L_0^2, etc. on f_0 and hence, the corresponding correlation functions $\Phi_{\alpha 0}$ can all be expressed in terms of Φ_0, Eqs. (17a) and (17b). Φ_{10} has been given in Eq. (7a) already in terms of

$$\phi_1(x)=1+x\phi_0(x)\,, \tag{25a}$$

and similarly one gets ϕ_{20} in terms of

$$\phi_2(x)=[dx\,\phi_1(x)-\phi_0(x)]/(d-1)\,. \tag{25b}$$

Summarizing the preceding formulas one arrives at

$$M(z)=i\nu-\frac{\lambda}{d}M(z)\int_0^\infty d\kappa\,\kappa^{d-3}\frac{F\left(k,\dfrac{z+M(z)}{\kappa\sigma v_0}\right)^2}{1+M(z)\Phi^{(0)}(\kappa\sigma,z+M(z))}\,, \tag{26a}$$

where for $d=2$ and 3, respectively,

$$F(\kappa,x)=\begin{cases} J_0'(\kappa)\,\phi_0(x)-i\,\dfrac{16}{3\pi}J_1'(\kappa)\,\phi_1(x)-J_2'(\kappa)\,\phi_2(x)\cdots\\[2ex] j_0'(\kappa)\,\phi_0(x)-i\,\frac{9}{4}\,j_1'(\kappa)\,\phi_1(x)-2j_2'(\kappa)\,\phi_2(x)\cdots\,. \end{cases} \tag{26b}$$

Here $J_n(\kappa)$ are Bessel functions of index n and $j_n(\kappa)$ are spherical Bessel functions[20]; $\lambda=\pi n\sigma^2$ and $8n\sigma^3/3$ for $d=2$ and $d=3$.

Equation (26a) is the general result of the present paper. It is a transcendental equation to determine the current relaxation kernel $M(z)$. The solution then yields all phase space correlation functions, Eq. (16), in particular the density correlations, Eq. (17c). $M(z)$ determines the velocity correlation function, Eq. (10), in particular the diffusivity and the polarizability. Equation (26a) is a mode-coupling approximation expressing the kernel $M(z)$ for single mode propagation in terms of the correlations for two mode propagation. Mode-coupling approximations have been introduced originally to calculate transport coefficients near critical points[21] or long-time tails of correlation functions.[22] In the present work the mode-coupling approximation is used to evaluate the current relaxation kernel $M(z)$ in the entire frequency regime. In the analysis of liquid dynamics such an approximation has been demonstrated to be quite useful.[23] In the present context the self-consistent formulation of the relaxation kernel $M(z)$ in terms of the correlation function $\Phi(q,z)$ has been introduced first to study the electrical conductivity in strongly disordered systems.[9] In the low-density limit, where the feedback is removed by writing $M(z)=i\nu$ our Eq. (26a) for three dimensions reduces to a result derived by Weyland.[24]

Equation (26a) is too complicated to allow for an analytical solution but it can be solved numerically by a straightforward iteration procedure. The interesting features of the present theory, however, can be understood by examining various asymptotic expansions of Eqs. (26) as we shall demostrate now.

IV. DISCUSSION

A. Self-consistency and feedback

The crucial element in the present theory is the feedback mechanism introduced by the mode-coupling contribution to the current relaxation kernel. Equation (26a) formalizes the idea that the current relaxation rate $M''(\omega)$ has to be calculated self-consistently with the density fluctuation spectrum $\Phi''(q,\omega)$ which in turn is ruled by $M(z)$. The feedback between the spectra of the motion to the rate of relaxation is described by the nonlinear integral contribution in Eq. (26a). In the low-density limit feedback is unimportant $M^{(0)}(z)=i\nu$, i.e.,

$$K^{(0)}(z)=\frac{-1}{z+i\nu}\,(v_0^2/d)\,. \tag{27}$$

This result implies an exponentially decaying velocity-autocorrelation function. In this case $M''(\omega)$, which may be interpreted as the rate current relaxation due to the transfer of recoil momentum for the random scatterers to the particle, is simply given by the binary collision rate ν. The low-density limit is seen to be the analogue of the Boltzmann-Enskog theory with the diffusion coefficient determined in the first Sonine approximation.[16] Here the scatterer density n merely sets the time and length scales for the diffusion process, and one is justified in treating the collisions as uncorrelated events. As the density increases, the probability that the particle will be reflected such that it can collide again with some of the previous scatterers increases. This effect is expressed by $m(z)$ in Eq. (20). A change in the relaxation rate $m''(\omega)$ affects $K''(\omega)$ and therefore the propagator $\Phi''(q,\omega)$ which in turn will affect $m''(\omega)$. In the following, the various consequences of this feedback process will be investigated.

B. Suppression of diffusivity

In lowest order the diffusivity $D^{(0)}=v_0^2/(d\nu)$ decreases proportional to $1/n$:

$$D^{(0)}=(v_0\sigma)\frac{1}{n^*}\begin{cases}3/16, & \text{for } d=2\\ 1/3\pi, & \text{for } d=3\end{cases} \tag{28}$$

with $n^*=n\sigma^d$ denoting the dimensionless density. The most important effect of the feedback is a suppression of D below $D^{(0)}$ causing a density variation of $D/D^{(0)}$. After a binary collision event there is a certain probability for the particle to return to the original scattering center. Hence, more recoil can be transferred leading to an enhancement of $M''(\omega)$ and thus to a decrease of D. For $z=i0$, Eq. (26a) yields with Eq. (11b):

$$D/D^{(0)}=1-(\lambda/d)\int_0^\infty d\kappa\,\frac{\kappa^{d-3}}{1+iy\,\phi_0(iy)}\,F(\kappa,iy)^2\,, \tag{29a}$$

$$y=\frac{D^{(0)}}{D}\left(\frac{\sigma}{l\kappa}\right), \tag{29b}$$

where $l=v_0/\nu$ is the particles mean-free path.

For $n^*\to 0$ one gets $l\propto 1/n^*\to\infty$; hence, the low-density expansion of $D/D^{(0)}$ is obtained by expanding the right-hand side of Eq. (29a) in powers of y. For $d=2$, Eq. (26b) yields $F(\kappa,i0)=-i8/3\pi+O(\kappa)$ and therefore putting $y=0$ in the denominator of Eq. (29a) leads to a logarithmically divergent integral. The small κ divergency, however, is cut off for $\kappa\sim 1/l$, since $\kappa\to 0$, $y\to\infty$. One gets in leading order

$$D/D^{(0)}=1+\tfrac{4}{3}\,(8/3\pi)\,n^*\log n^*+O(n^*),\quad d=2\,. \tag{30}$$

So the present theory reproduces, up to the factor $8/3\pi$, the known[2] nonanalytic density expansion of D in leading order. For $d=3$ all modes contribute to the leading density correction for $D/D^{(0)}$. The first three terms noted in Eq. (26b) yield the result within about 1% error,

$$D/D^{(0)}=1-1.48\,n^*+O(n^{*2}\log n^*),\quad d=3\,. \tag{31}$$

The prefactor of the n^* term differs by about 25% from the results arrived at previously.[2]

C. Diffusion blocking

To find out whether or not there exists a small D solution of our self-consistency equations one has to perform the large y expansion in Eq. (29a). Since $\phi_n(x) = O(1/x^{n+1})$ one can restrict function F in Eq. (26b) to the first term in order to arrive at

$$D/D^0 = 1 - n^*/n_c^*, \quad \text{if } n^* \to n_c^{*-}, \tag{32a}$$

$$n_c^* = \begin{cases} 2/\pi, & \text{for } d = 2 \\ 9/4\pi, & \text{for } d = 3\,. \end{cases} \tag{32b}$$

Hence, there appears a critical density n_c where the diffusion constant vanishes. For n approaching n_c the diffusivity vanishes linearly with the separation parameter $\epsilon = (n - n_c)/n_c$. For n exceeding n_c there is absence of diffusion. The linear variation of D with density has been observed in computer simulations in both two and three dimensions.[5,6] The theoretical value of $n_c^* = 9/4\pi$ for $d = 3$ agrees well with extrapolation of the computer data of Bruin,[5] whereas for $d = 2$ the value of $n_c^* = 2/\pi$ is about twice that found in the simulation results of Alder and Alley.[6]

Our results show clearly how the feedback mechanism leads to a blocking of diffusion. Renormalization effects are expressed through the requirement of self-consistency. One finds an exhanced current relaxation rate $M''(\omega \to 0)$ because the recoil spectrum $\Phi''(q \to 0, \omega = 0)$ determining $m''(\omega \to 0)$ via Eq. (26) is enhanced for $q \to 0$ (Ref. 25). The enhancement of $M''(\omega = 0)$ implies a suppression of D, Eq. (11b), and this in turn enhances $\Phi''(q \to 0, \omega = 0)$. Consequently there is a tendency for an instability which leads to $D \to 0$ for $n \to n_c$. Since the particle diffuses, it returns again and again to a given scattering center, and so the same center can transfer recoil momentum more effectively than that described in a kinetic equation which considers only uncorrelated collisions. The repeated scattering can be so effective that the probability of escape vanishes—diffusion becomes blocked.

D. The localization phase and its instability

To examine the possibility of a localization phase, one can carry out the $z \to 0$ limit of Eqs. (26) observing $zM(z) \to -sv_0^2/d$. Again one finds only the first term in Eq. (26b) to contribute and $\phi_0(x)$ can be expanded in leading order. The resulting integral in Eq. (26a) can be expressed in terms of Bessel functions for imaginary arguments I, K (Ref. 20) to yield the transcendental equation for the stiffness s:

$$1 = \frac{n^*}{n_c^*} F_0(\sigma\sqrt{s}), \tag{33a}$$

$$F_0(\zeta) = d I_{d/2}(\zeta) K_{d/2}(\zeta)\,. \tag{33b}$$

For large densities this implies a stiffness increasing proportional to n^{*2}

$$s\sigma^2 = n^{*2} \begin{cases} \pi^2/4, & d = 2 \\ 8\pi^2/27, & d = 3\,. \end{cases} \tag{34a}$$

To find out the small s solution one can expand the right-hand side of Eq. (32) to obtain for $n^* \to n_c^{*+}$

$$s\sigma^2 = (n^*/n_c^* - 1) \begin{cases} (-4)\log(n^*/n_c^* - 1)^{-1}, & d = 2 \\ 5/2, & d = 3\,. \end{cases} \tag{34b}$$

Thus for high density the self-consistency mechanism yields a localization phase. The localization length l_0, Eq. (13a), decreases inversely proportional to the density n. With decreasing n the stiffness becomes smaller, and approaching n_c^*, the polarizability $\chi_0 \propto 1/s$ diverges according to a Curie-Weiss law, Eq. (34). The instability point for the insulator n_c is the same as the blocking point of the conductor.

E. The low-frequency velocity spectrum

While the zero-frequency limit of $K(z)$ gives the self-diffusion coefficient, the low-frequency behavior of $K(z)$ reflects the way in which the velocity-auto correlation function $K(t)$ decays at long times. We will find that the diffusion and localization phases are characterized by different asymptotic decay behavior and that the dependence on dimension is also different between the two phases. Moreover, in the vicinity of the critical density still another type of behavior appears. For the localized phase the small-frequency expansion of the kernel reads

$$M(\omega + i0) = \frac{-s}{\omega}\frac{v_0^2}{d} + i\tilde{\nu}\,. \tag{35a}$$

Substitution of this result into Eq. (10) yields the regular frequency dependence for the velocity spectrum

$$K''(\omega) = \frac{d}{v_0^2 s^2}\,\tilde{\nu}\,\omega^2 + O(\omega^4), \quad n > n_c\,. \tag{35b}$$

Hence, the velocity-autocorrelation function relaxes to zero exponentially in time; it behaves like the correlation function of an oscillator with frequency $\omega_0^2 \propto s$ and friction constant $\tilde{\nu}$. s has been discussed above and $\tilde{\nu}$ is obtained from Eq.

(26a) by working out the expansion term of the integral proportional to the small parameter z, yielding

$$\tilde{\nu} = 2\frac{\nu}{s\sigma^2 n^*/n_c^*}\left(1 - \frac{2}{d}F_1(\zeta)\right)\Big/\left(-\frac{1}{\zeta}\frac{\partial F_0(\zeta)}{\partial\zeta}\right), \zeta = \sigma\sqrt{s} \qquad (35c)$$

$$F_1(\zeta) = \frac{d-2}{2}F_0(\zeta) - \frac{1}{2}\zeta\frac{\partial}{\partial\zeta}F_0(\zeta).$$

For the diffusion phase the denominator in the mode-coupling integral, Eq. (26a), develops the structure $z + i\kappa^2 D$, provided frequencies and wave numbers are small. The singular behavior, reflecting the very slow motion of density fluctuations in the hydrodynamic limit, produces a $z = 0$ branch point for $M(z)$. For small frequencies one gets

$$M''(\omega) - v_0^2/(dD) \propto |\omega|^{d/2}. \qquad (36a)$$

With Eq. (10) one finds the irregular velocity spectrum

$$K''(\omega)/D^{(0)} - D/D^{(0)} = \gamma(|\omega|D^{(0)}/\nu D)^{d/2} \times[1 + O(\omega)], \quad n < n_c. \qquad (36b)$$

To evaluate γ one merely has to pick up the leading singularity of the mode-coupling integrand for $\kappa \to 0$. The first two contributions in Eq. (26b) vanish proportional to κ^2; the others yield higher powers in κ for the long-wavelength limit and therefore they do not contribute to γ. Working out the integrals one finds

$$\gamma = \begin{cases} \dfrac{8n^*}{9}\left(\dfrac{D}{D^{(0)}} + \pi n^*\right)^2, & d = 2 \\[2ex] \dfrac{3\pi n^*}{4}\left(\dfrac{\pi n^*}{\sqrt{6}}\right)\left(\dfrac{D}{D}(0) + \dfrac{4\pi}{3}n^*\right)^2, & d = 3. \end{cases} \qquad (36c)$$

The nonanalytic ω dependence implies a long-time power-law decay proportional to $t^{-5/2}$ and t^{-2} for $d = 3$ and 2. These power laws and their prefactors agree with the kinetic theory calculations of Ernst and Weyland[2] in the case of low density. Compared to the earlier results, Eqs. (36) are noteworthy in two respects. First, besides mode coupling to the longitudinal current which gives the leading density contribution in Eq. (36c), the decay of the current relaxation kernel into density modes is included. The latter leads to contributions higher order in density as shown by the second term in the bracket in Eq. (36c). This effect also has been discussed by Keyes and Mercer.[4,26] Second, the full diffusion constant D, instead of the low-density approximation $D^{(0)}$, appears on the right-hand side of Eqs. (36). The significance of this point lies in the density variation of frequency ω_h which delineates the hydrodynamic regime. In the case of $d = 3$, the hydrodynamic regime is given by that frequency where the ω-dependent term in Eq. (36b) becomes of order $D/D^{(0)}$. Hence, $\omega_h \sim \nu n^{-4/3}(1 - n/n_c)^{5/3}$. At low densities the amplitude of the long-time tail increases with density and the frequency regime where the long-time singularity can be observed expands. However, ω_h decreases with approaching n_c and therefore the time interval $t_h \sim 1/\omega_h$ for diffusive motion and the time for the onset of power-law decay will increase. The hydrodynamic regime will continue to shrink with increasing density and at n_c it disappears completely. One can associate with ω_h a wave vector $q_h = (\omega_h/D)^{1/2}(\nu/v_0)n^{-2/3}(1 - n/n_c)^{1/3}$ so that the shrinking of the hydrodynamic regime is observed also in wave vector space. These results refer to $d = 3$. A similar discussion can be given for $d = 2$. One should expect that these estimates for ω_h and q_h are not correct quantitatively as n approaches n_c, since near the edge Eq. (26a) cannot be solved by the expansion leading to Eqs. (36).

At the critical density $M(z)$ will diverge for $z \to 0$ since $D = 0$ but $zM(z)$ will approach zero for $z \to 0$ since $s = 0$. Using these properties one can make use of the simplifications of Eqs. (26) discussed in connection with Eqs. (32) and (34). One finds for small ω in leading order,

$$K''(\omega)/D^{(0)} = \sqrt{\omega}/\nu \begin{cases} \dfrac{4\sqrt{2}}{3\pi}\log\left(\dfrac{\nu}{\omega}\right), & d = 2 \\[2ex] \dfrac{9}{4}\left(\dfrac{3}{5}\right)^{1/2}, & d = 3, \quad n = n_c. \end{cases} \qquad (37)$$

According to Eq. (37), when $n \to n_c$, a frequency window opens $\omega_h \ll \omega < \nu$ where the frequency spectrum of the velocity-autocorrelation function exhibits a nonanalytic $\omega^{1/2}$ variation. Hence, there will be a preasymptotic long-time tail, $K(t) \propto t^{-3/2}\log 1/t$ and $t^{-3/2}$ for $d = 2$ and 3. This tail appears in the density region $3n_c/2 > n > 2n_c/3$, where $\omega_h \ll \nu$, and it extends from ν^{-1} to t_h. At the critical point, characterized by $t_h = \infty$, the $t^{-3/2}$ tail extends up to infinity.

It follows from the foregoing discussion that a transition phenomenon exists in the long-time decay, $t^{-\beta}$, of the velocity-autocorrelation function. In the time interval $t > \nu^{-1}$ $K(t)$ should show a power-law decay with exponent $\beta = 2$ or 5/2, for $d = 2$ or 3, when $n \ll n_c$. If the density exceeds about $2n_c/3$, one has to go out to longer and longer times in order to observe this exponent. At a critical density it is possible to observe only a power-law decay with exponent 3/2. For $d = 2$ there should also appear at $n = n_c$ a logarithmic deviation from a power law, but as usual this will be difficult to detect. When n is increased above n_c, the long-time decay of $K(t)$ may be fitted to a power law with an exponent greater than 3/2 since in the hydrodynamic limit of the localization phase the exponent is effectively infinite. The value of this apparent exponent should increase with increasing density. The behavior of the long-time decay of the velocity autocorrelation as described here has indeed been observed in computer simulation results of Alder and Alley[6] for $d = 2$.

F. The transition regime

The preceding asymptotic results, Eqs. (32), (34b), (35), and (37) can be summarized if one defines implicitly transition regime in the two-dimensional space of variables (ω, n) by[9]

$$|z/\nu| \ll 1\,,$$

$$|\nu/M(z)| \ll 1\,,$$

$$|zM(z)/\nu^2| \ll 1\,. \tag{38a}$$

For $n = n_c$ the three inequalities are valid if $z \to 0$. Continuity of the integral (26a) implies that there will be a certain density interval around n_c such that for low frequencies all equations (38a) are obeyed. Performing the expansion of the right-hand side of Eq. (26a) in terms of the mentioned small parameters, as discussed before, and using $K(z) \simeq -(v_0^2/d)1/M(z)$, one arrives at

$$\hat{K}^2 + i\epsilon\hat{K} - ia\hat{z} = 0\,. \tag{38b}$$

Here $\hat{K} = K/D^{(0)}$, $\hat{z} = z/\nu$, $\epsilon = (n - n_c)/n_c$, and $a = 2(8/3\pi)^2 \log(\hat{K}/\hat{z})$ for $d = 3$, $a = (6/5)(9/4)^2$ for $d = 3$. So for $d = 3$,

$$K(z) = D^{(0)}\epsilon f(z/\nu\epsilon^2)\,, \tag{38c}$$

and a similar formula exists for $d = 2$ with f containing a logarithmic correction term. The function f is found by solving the quadratic equation (38b) and choosing the branch that is compatible with the Kramers-Kronig relation for $K(z)$. Equation (38c) formulates a scaling law for the velocity-autocorrelation function. The scaling frequency $\omega_h = \nu\epsilon^2$ has been identified previously. It separates the low-frequency hydrodynamic regime from the critical regime. At the transition point there is a critical slowing down of the modes and a corresponding critical wavelength expansion, $q_h^2 = \omega_h/D$ (or $q_h^2 = s$). If one approaches the critical point, one has to wait longer and longer and also to average over larger and larger spatial regions in order to detect the hydrodynamic motion. If $q > q_h$ or $\omega > \omega_h$ one cannot decide whether the system is in the diffusion phase or the localization phase.

V. CONCLUSIONS

The novel idea of the present paper is the suggestion to evaluate the current relaxation kernel $M(z)$ for a classical particle moving in a certain static field self-consistently with the correlation functions for the particle density, momentum, etc. The current relaxation spectrum $M''(\omega)$, describing the rate for momentum transfer from the particle to the field, depends on the particles mode of propagation. A particle flying around freely will relax its momentum differently than one diffusing slowly or oscillating in a trap. The spectra for the particle motion in turn are ruled by $M(z)$ and so a strong feedback mechanism is established whenever $M(z)$ is large compared to its binary collision value ν. The mode-coupling-approximation technique was used to formalize the self-consistent current relaxation concept leading to a closed but highly nonlinear equation for $M(z)$, Eq. (26).

The self-consistency equations have been shown to reproduce in leading order the known long-time anomalies[3] as well as the known density expansion for the diffusivity.[2] Thus, the present theory presents a unified extension of the classical kinetic equation results in frequency and coupling-constant space. Those results, Eqs. (30), (31), and (36), can be considered as a successful check of the present theory against those results obtained earlier[2,3] with quite different mathematical reasoning.

The equations derived are the first proposal to obtain correlation functions for a classical dynamical system valid for all densities. The new outcome of the theory is a first principle description of a transition from a diffusion to a localization phase if the density crosses a critical value n_c. Obviously, the phase transition found represents a classical percolation point. However, so far classical theories[27] have not been applied to calculate physical quantities like the diffusion constant D or the localization length l_0 for the model studied in this paper. Our treatment of the percolation problem in the Lorentz model is quite different from the known approaches towards phase transitions. Usually one concentrates on anomalies for thermodynamic variables and then one analyzes their influence on the fluctuation spectra. In the present theory a frequency-dependent correlation function, a transport coefficient, or a spectral density is the critical variable. The mentioned phase transition has been observed in computer experiment[5,6] and it is a reassuring yet somewhat surprising outcome of our theory that the value for n_c, Eq. (32b), agrees with the experimental one for $d = 3$ and differs from the experimental number for $d = 2$ only by a factor of 2.

In the transition region there is a small parameter for our problem, viz. the separation $\epsilon = (n - n_c)/n_c$ from the percolation edge allowing for a closed solution of the self-consistency equations for small frequencies, Eq. (38). Thereby, besides the Curie-Weiss divergency of the polarizability of the localized phase, Eq. (34b), and the linear decrease of the diffusivity near the n_c, Eq. (32a), a critical slowing down of all dynamical processes has been found. The characteristic frequency ω_h and wave number q_h, defining the regime of hydrodynamic motion, shrink proportional to ϵ^2 and $\sqrt{\epsilon}$, respectively, as the critical point is approached. Consequently, the previously unexplained characteristic variation of the effective long-time-tail exponent for the velocity correlations in $d = 2$[6] now can be described qualitatively, Sec. IV E.

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[26]We believe there is an error in Eq. (30c) of Ref. 4, namely, the factor of $\sqrt{\pi}$ should be deleted. This means that in Eq. (40) of Ref. 4 the factor of $4\sqrt{\pi}$ should be 4π. With this change our results are in agreement with Ref. 4.
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