equal to $N^2/16$ for zero energy,[10] the radiation rate

$$\langle R_+R_-\rangle = \tfrac{1}{2}N(\tfrac{1}{2}N+1) - \sigma + W - W^2 \approx \tfrac{3}{16}N^2 \qquad (21)$$

is proportional to N^2. So, at time τ_0, the state of our system obeys the superradiant conditions $(W\approx 0, \langle R_+R_-\rangle \approx N^2)$. But these conditions do not seem to ensure a coherent radiation field.

We have also performed calculations with systems prepared by an intense laser $\frac{1}{2}\pi$ pulse, i.e.,

$$\rho_{m,m}(0) = \frac{N!}{(\frac{1}{2}N+m)!\,(\frac{1}{2}N-m)!}\,(2)^{-N}\,. \qquad (22)$$

The variation of $g^{(2)}(\tau, 0)$ against time $N\tau$ is plotted in Fig. 5 for different N. For sufficiently large N, $g^{(2)}(\tau, 0)$ decays from roughly unity to a nonzero value. A straightforward calculation shows that with the initial conditions given by Eq. (22), the relation (13), which expresses the correlations as a function of the radiation rates, is valid for large N[i.e., the quantity $\langle R_+(t_2)R_-(t_2)R_+(t_1)R_-(t_1)\rangle$ factorizes]. Then, by introducing the classical radiation rates[9] [Eq. (13)] the asymptotic value of $g^{(2)}(\tau, 0)$ is found to be $1-4/(N+1)$. Clearly, for a large system, $g^{(2)}(\tau, 0)$ is constant and equal to 1. The radiation field emitted by a system prepared as Eq. (22) is coherent (at least) to the second-order.

As the initial values of the energy and the radiation rate are approximately the same in the two cases illustrated by Figs. 4 and 5, the different behaviors of the intensity-fluctuation function are due to the $\rho_{m,m}(\tau_0)$ distribution.[2] If one hypothesizes that a property of a superradiant state is coherent emission. This implies not only conditions for the energy and radiation rate of the system but also for its preparation.

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Absence of Nonlinear Response in the Quasistochastic Model*

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The full nonlinear governing equations for the quasistochastic model, which describes the motion of a Brownian charge carrier in an external electric field, are solved for the stationary current response. We find that the nonlinear response is identical to the linear response obtained earlier by Lebowitz and Rubin.

I. INTRODUCTION

The classical theory of Brownian motion, which is based on an *ab initio* stochastic description, can be formulated in terms of the Fokker-Planck equation[1]

$$\frac{\partial f}{\partial t} + \vec{V}\cdot\frac{\partial f}{\partial \vec{R}} + \frac{e}{M}\,\vec{E}\cdot\frac{\partial f}{\partial \vec{V}} = D\frac{\partial}{\partial \vec{V}}\cdot\left(f\,\frac{\partial}{\partial \vec{V}}\ln f/f_0\right). \qquad (1)$$

In order to describe the higher-order response it is necessary to consider a more detailed description of Brownian motion than that given by (1). This is of interest since at present only formal results[2] for nonlinear response coefficients have been obtained in the literature. The case of conduction by Brownian charge carriers offers a physically relevant problem, e.g., for the area of electrolyte theory,[3] which should be easier to treat than the general problem of conduction by arbitrary carriers.

A generalization of (1) which eliminates the stochastic content in the description and proceeds instead directly from the Liouville equation has been obtained by several authors[4] (the *first* of these references will be referred to as LR hereafter). They find a kinetic equation for the Brownian particle in which the collision term [corresponding to the right-hand side of (1)] is expanded in the square root of the ratio $\gamma^2 = m/M$, m the solvent particle mass. The leading term in this expansion is of the Fokker-Planck form, so that higher-order terms (in γ) would have to be considered to obtain the corrections to the Ohmic current. This was not done by the above-mentioned authors, who only consider the linear (in E) kinetic equation. Another possible means for obtaining the higher-order response is to consider the quasistochastic model introduced by LR. This is a formal model with both stochastic and dynamical features, and may be considered as being midway between the pure dynamical and pure stochastic descriptions. The advantage shown for this model by LR was that it was amenable to explicit calculations to all orders of γ in the linearized (in E) response regime, the results obtained being formally equivalent to those found in the dynamical description. Since models which are both instructive and exactly solvable are not very common in nonequilibrium statistical mechanics it seems worthwhile to consider nonlinear response theory in the context of the quasistochastic model. The main result of this paper will be to show that the full nonlinear response can be computed for this model, and that, in fact, there is no additional contribution beyond the Ohmic current. Thus, in order to describe the higher-order response either a generalization of the stochastic theory[5] or, preferably, the higher-order dynamical description must be used. This latter approach will be discussed in connection with the related problem of Hall conduction in a separate report.

II. QUASISTOCHASTIC MODEL

The fully dynamical theory of Brownian motion proceeds from the Liouville equation, the various particle pair interactions that occur being described by specific potential functions. In the quasistochastic model the description is simplified by assumming that the Brownian particle interacts with only one solvent particle through a potential function, U, and the effect of the rest of the solvent is described stochastically. Instead of Liouville's equation, the basic governing equation is the generalized two-particle Liouville equation

$$\frac{\partial\mu}{\partial t} + H\mu + \frac{\vec{E}}{M}\cdot\frac{\partial\mu}{\partial\vec{V}} = \int d\vec{r}\,'d\vec{v}\,'\,[K(\vec{r},\vec{v},\vec{r}\,',\vec{v}\,',\vec{R},\vec{V})\,\mu(\vec{R},\vec{V},\vec{r}\,',\vec{v}\,',t) - K(\vec{r}\,',\vec{v}\,',\vec{r},\vec{v},\vec{R},\vec{V})\,\mu(\vec{R},\vec{V},\vec{r},\vec{v},t)]. \quad (2)$$

Here $\mu = \mu(\vec{R},\vec{V},\vec{r},\vec{v},t)$ is the joint distribution function for the Brownian particle and interacting solvent particle, $\vec{R},\vec{V}$ and $\vec{r},\vec{v}$ the position and velocity of these particles,

$$H = \vec{V}\cdot\frac{\partial}{\partial\vec{R}} + \vec{v}\cdot\frac{\partial}{\partial\vec{r}} - \frac{1}{M}\frac{\partial U}{\partial\vec{R}}\cdot\frac{\partial}{\partial\vec{V}} - \frac{\partial U}{\partial\vec{r}}\cdot\frac{\partial}{\partial\vec{v}}$$

is the Hamilton operator for the motion of the two-particle system in the absence of $\vec{E}$, and K is a stochastic kernel describing the effect of the remaining solvent, which acts as a resevoir and is assumed to only indirectly interact with the Brownian particle. For convenience we have set the charge on the Brownian particle and the solvent particle mass both equal to one. As in LR we now take

$$K(\vec{r},\vec{v},\vec{r}\,',\vec{v}\,',\vec{R},\vec{V}) = \mu_0/\tau f_0 = P_0/\tau$$

and

$$U = \tfrac{1}{2}\omega_0^2(\vec{R}-\vec{r})^2 = \tfrac{1}{2}\omega_0^2 R_0^2 .$$

The first of these assumptions is discussed by LR; the second appears reasonable in the context of the model and allows us to carry out exact calculations. Although we know of no other work based on this specific model, various of its distinguishing features have appeared in several independent descriptions of the liquid state.[6]

With the above form for K the model Liouville equation becomes

$$\frac{\partial\mu}{\partial t} + H\mu + \frac{\vec{E}}{M}\cdot\frac{\partial\mu}{\partial\vec{V}} = \frac{fP_0 - \mu}{\tau}. \quad (3)$$

LR only treat the linearized (in E) version of this equation, but we shall be interested in the full equation. However, we are only interested in the steady-state current, so we will only need to determine the moment

$$\vec{j} = \lim_{t\to\infty}\int d\vec{R}\,d\vec{V}\,d\vec{r}\,d\vec{v}\,\vec{V}\,\mu(t)$$

of the solution which we can do without explicitly finding μ.

III. STEADY-STATE CURRENT

We proceed by formally solving (3). First, we write $\mu = \mu_0 + \mu'$, where μ_0 is the equilibrium value of μ, so that μ' satisfies the following equation:

$$\frac{\partial\mu'}{\partial t} + H\mu' + \frac{\vec{E}}{M}\cdot\frac{\partial\mu'}{\partial\vec{V}} - \beta\vec{V}\cdot\vec{E}\,\mu_0 = \frac{(f'P_0 - \mu')}{\tau}, \quad (4)$$

with $f' = \int d\vec{r}d\vec{v}\,\mu'$. A formal stationary solution to (4) is

$$\mu'(\infty) = \frac{1}{\tau}\int_0^\infty dt'\exp\left[-t'\left(H + \frac{\vec{E}}{M}\cdot\frac{\partial}{\partial\vec{V}} + \frac{1}{\tau}\right)\right] \times \mu^0\left(\frac{f'(\infty)}{f_0} + \tau\beta\vec{V}\cdot\vec{E}\right), \quad (5)$$

which differs from the linear solution found by LR [their Eq. (C19)] in that the exponential streaming operator contains the full, field-dependent Hamilton operator $H+(\vec{E}/M)\cdot\partial/\partial\vec{V}$ instead of the field-free operator. Note also that we have not used the scaled velocity variable $\vec{V}\gamma^{-1}$ here as we are not interested in a γ expansion, but rather an exact result for the model. For the full streaming operator we find that

$$\exp\left[s\left(H+\frac{\vec{E}}{M}\cdot\frac{\partial}{\partial\vec{V}}\right)\right]\vec{V}$$
$$=\vec{V}+\gamma^2 m^*\left[(\vec{v}-\vec{V})(1-\cos\omega S)+\omega\vec{R}_0\sin\omega s\right]$$
$$+\gamma^2 m^*\vec{E}\left(S=\frac{\gamma^2}{\omega}\sin\omega s\right), \quad (6)$$

where the reduced mass m^*, and ω are identical to the quantities defined in LR [note (C22) has a misprint],

$$m^*=(1+\gamma^2)^{-1}, \quad \omega=\omega_0(1+\gamma^2)^{1/2}.$$

Since the full streaming operator which appears in (5) does not commute with μ_0 the integral equation for f' becomes considerably less tractable than the corresponding linear equation, and it is no longer practical to attempt to solve directly for this quantity. However, the steady-state current can be calculated fairly directly

$$\vec{j}=\int d\vec{V}d\vec{R}\,d\vec{v}\,d\vec{r}\,\vec{V}\mu(\infty)$$
$$=\int d\vec{V}d\vec{R}d\vec{v}d\vec{r}$$
$$\times\int_0^\infty dt'\left\{\exp\left[t'\left(H+\frac{\vec{E}}{M}\cdot\frac{\partial}{\partial\vec{V}}-\frac{1}{\tau}\right)\right]\vec{V}\right\}$$
$$\times\frac{\mu_0}{\tau}\left(\frac{f'(\infty)}{f_0}+\tau\beta\vec{V}\cdot\vec{E}\right)$$
$$=\vec{j}\left(1-\frac{\gamma^2 m^*(\omega\tau)^2}{1+(\omega\tau)^2}\right)$$
$$+\tau\beta\left(\frac{1+(\omega\tau)^2-\gamma^2 m^*(\omega\tau)^2}{1+(\omega\tau)^2}\right)I\vec{E}, \quad (7)$$

where

$$I=\tfrac{1}{3}\int d\vec{V}\,\vec{V}^2 f_0.$$

The second equation above follows from the first and (5) after an integration by parts (or change of variable). Rearranging terms in (7) we then find that

$$\vec{j}=(\beta/\gamma^2\omega_0^2\tau)[1+(\omega_0\tau)^2]I\vec{E} \quad (8)$$

which is identical to the linear result which one obtains from the results of LR when these are expressed in terms of the physical velocity $\vec{V}$ [cf. (C24)].

The result that there is no nonlinear response in the quasistochastic model is instructive in that it indicates that the transport coefficients associated with this current cannot be expressed solely in terms of functions of the linear-transport coefficient, i.e., the friction coefficient, but depend on higher-order correlation functions, which vanish for this model. This conclusion could not have been made on the basis of the linear results found previously from the dynamical theory.[4] Finally, it is interesting to note that the same "solution technique" used in this paper can be used to obtain exact solutions for some other nonequilibrium problems, e.g., the nonlinear heat flow in a linear harmonic chain. We are currently carrying out the computations for this problem.

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