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Microscopic Theory of Brownian Motion in an Oscillating Field; Connection with Macroscopic Theory*

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Recently, Lebowitz and Rubin, and Résibois and Davis, showed that the Fokker-Planck equation for the distribution function of a Brownian particle (B particle) of mass M, in a fluid of particles of mass m, may be derived directly from the Liouville equation for the joint distribution of fluid and B particle. It is the lowest order term in a $(m/M)^{1/2}$ expansion of the effect of the fluid on the distribution of the B particle. These authors studied in particular the steady-state distribution function of B particles acted on by a small constant external field $\mathbf{E}$, which results from a balance between the effects of the driving force and those of the fluid. In this paper we extend these studies to the case where the B particle is acted on by a time-dependent field $\mathbf{E}\,e^{i\omega t}$. We find that the effect of the fluid on the distribution function of the B particle is again given, to lowest order in $(m/M)^{1/2}$, by a Fokker-Planck term, albeit one with a frequency-dependent friction constant, $M\zeta(\omega)\sim\int_0^\infty\langle\mathfrak{F}(0)\cdot\mathfrak{F}(t)\rangle\,e^{i\omega t}dt$. Here $\mathfrak{F}$ is the microscopic, N-body force acting on a *stationary* B particle and the average is over the equilibrium distribution function of the fluid in the presence of this *fixed* B particle. We further show that $-M\zeta(\omega)\mathbf{V}_0\,e^{i\omega t}$ is equal to the force acting on a B particle moving through the fluid with a *prescribed* small velocity $\mathbf{V}_0\,e^{i\omega t}$. Under appropriate circumstances this latter force may be computed from kinetic theory or from hydrodynamics. We thus have complete agreement between our microscopic theory and that obtained from stochastic considerations. We also clarify the relation between the different formalisms used by Lebowitz and Rubin and by Résibois and Davis.

I. INTRODUCTION

THE theory of Brownian motion was developed initially by Einstein and Smoluchowsky and later elaborated by Langevin and others.[1] Describing the effect of the fluid particles of mass m on a Brownian particle (B particle) of mass M, $M\gg m$, in a schematic stochastic fashion, their results are summarized in the Fokker-Planck equation for the distribution function of the B particle in its position and velocity space $f(\mathbf{R},\mathbf{V},t)$,

$$\partial f(\mathbf{R},\mathbf{V},t)/\partial t+\mathbf{V}\cdot\partial f/\partial\mathbf{R}+M^{-1}\mathbf{Y}\cdot\partial f/\partial\mathbf{V}=\zeta\partial/\partial\mathbf{V}\cdot[\mathbf{V}+(\beta M)^{-1}\partial/\partial\mathbf{V}]f. \quad (1.1)$$

Here $\mathbf{Y}$ is the total external nondissipative force acting on the B particle, ζ is the friction constant of the B particles in the fluid, and $\beta=(kT)^{-1}$ is the reciprocal temperature of the fluid. The right side of (1.1) represents the effect of the fluid on the B particles, and will be called the Fokker-Planck term.

Recently Lebowitz and Rubin,[2] and Résibois and Davis[3] have developed a "microscopic" theory of Brownian motion.[4] They start with the Liouville equation for the distribution function μ of the whole system consisting of the host fluid and B particle. A transport equation for f then results after integration over the variables of the fluid particles, in certain limits involving the size of the fluid and the time scale. The equation they arrive at is of the same form as (1.1), *to the lowest order in the mass ratio of fluid and B particle*, with an explicit, if unevaluated, molecular expression for ζ,

$$M\zeta=\tfrac{1}{3}\beta\lim_{t_0\to\infty}\int_0^\infty e^{-t/t_0}\langle\mathfrak{F}(0)\cdot\mathfrak{F}(t)\rangle dt. \quad (1.2)$$

Here $\mathfrak{F}(t)$ is the total force exerted on the B particle at time t by the molecules in the surrounding fluid. Its time dependence is determined by the solution of the molecular equations of motion subject to the condition that the B particle is *held fixed* in position. The average is over an equilibrium ensemble at temperature T.

Lebowitz and Rubin and Résibois and Davis considered in particular the case where there is a small constant external force, say an electric field $\mathbf{E}$, acting on the B particle. This force corresponds to the term $\mathbf{Y}$ in (1.1) and has the effect of preventing the B-particle distribution f from reaching equilibrium. Instead f reaches, as $t\to\infty$, a stationary nonequilibrium value in which the "driving" effect of the external force on the B particle is balanced by the dissipative effect of its interaction with the fluid which is represented to lowest order in $(m/M)^{1/2}$ by a Fokker-Planck term. Higher order terms in $(m/M)^{1/2}$ were also computed by these authors.

In this note we carry further, and show the complete agreement between, the ideas developed in Refs. 2 and 3. In Sec. II we derive a "generalized transport" or master

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[1] For details and references see S. Chandrasekhar, Rev. Mod. Phys. **15**, 1 (1943); M. C. Wang and G. E. Uhlenbeck, *ibid.* **17**, 323 (1945).
[2] J. Lebowitz and E. Rubin, Phys. Rev. **131**, 2381 (1963).
[3] P. Résibois and R. Davis, Physica **30**, 1077 (1964).
[4] For a "kinetic" derivation see also M. S. Green, J. Chem. Phys. **19**, 1936 (1951); J. Lebowitz, Phys. Rev. **114**, 1192 (1959); R. M. Mazo, J. Chem. Phys. **35**, 831 (1961).

equation for the distribution f of a B particle acted on by a *small* oscillatory electric field $\mathbf{E}e^{i\omega t}$. The method used here is simpler than that used previously and we try to point out along the way the precise nature of the limits taken and assumptions made in deriving an irreversible equation for f.

In Sec. III we consider the steady-state distribution achieved by f as $t\to\infty$, which represents a balance between the external oscillatory force and the effect of the fluid. We find that the effect of the fluid may still be represented, to lowest order in $(m/M)^{1/2}$, by a Fokker-Planck term albeit the friction constant ζ is, in general, a function of ω,

$$M\zeta(\omega)=\tfrac{1}{3}\beta \lim_{t_0\to\infty}\int_0^\infty e^{-t/t_0}e^{-i\omega t}\langle\mathfrak{F}(0)\cdot\mathfrak{F}(t)\rangle dt \quad (1.3)$$

reducing to (1.2) when $\omega=0$.

In Sec. IV it is shown that the friction constant $\zeta(\omega)$ is precisely the same as one would obtain by considering the force $\mathbf{F}_V(\omega)$ acting on a B particle moving through the fluid with an *externally imposed small velocity* $\mathbf{V}_0e^{i\omega t}$; $\beta m V_0^2\ll 1$,

$$\mathbf{F}_V(\omega)=-M\zeta(\omega)\mathbf{V}_0e^{i\omega t}. \quad (1.4)$$

In this latter case the B particle does not have any degrees of freedom but acts merely as a source of external potential for the fluid. The relation (1.4) is, of course, the one generally used in the macroscopic[1] theories of Brownian motion for defining the friction constant ζ (for $\omega=0$). Our result thus shows a complete agreement between the dynamical and stochastic theories of Brownian motion. The evaluation of $\zeta(\omega)$ from the appropriate kinetic theory (e.g., the Boltzmann equation for a dilute gas) or from hydrodynamics is also discussed in this section.

In Sec. V we show the complete agreement (despite different appearances) between the transport equation for f derived here (which coincides in form with that of Ref. 2) and that obtained from the general Prigogine-Résibois[5] theory (which coincides with that of Ref. 3).

II. GENERAL FORMULATION

The Hamiltonian of our system, consisting of host fluid and B particle will have the form,[2]

$$H=[\tfrac{1}{2}MV^2+\chi(\mathbf{R})]+[\sum_{i=1}^N \tfrac{1}{2}mv_i^2+\sum_{i<j}\varphi(r_{ij})]$$
$$+[\sum_{i=1}^N u(\mathbf{r}_i-\mathbf{R})]=H_1+H_l+U, \quad (2.1)$$

where $\mathbf{r}_i$ and $\mathbf{v}_i$ are the position and velocity of the ith fluid particle. Here H_1 and H_l are, respectively, the Hamiltonians of the isolated B particle and the isolated fluid and U is the interaction between them: $\chi(\mathbf{R})$ is an external potential acting on the B particle and $\varphi(r_{ij})$ the interaction between two fluid particles. The whole system is enclosed in a periodic box of volume Ω.

Consider now a situation where at time $t=0$ we turn on an external electric field[6] equal to the real part of $\mathbf{E}e^{i\omega t}$. This field acts only on the B particle which has a unit charge. The joint distribution function of the whole system μ will obey the Liouville equation, for $t\geq 0$,

$$\partial\mu(x,y,t)/\partial t=-(\mu,H)-\gamma e^{i\omega t}\mathbf{E}\cdot\partial\mu/\partial\mathbf{v}$$
$$\equiv-\sum_{i=1}^N[\mathbf{v}_i\cdot\partial\mu/\partial\mathbf{r}_i-\sum_{j\neq i}\partial\varphi(r_{ij})/\partial\mathbf{r}_i\cdot\partial\mu/\partial\mathbf{v}_i$$
$$-\partial u(\mathbf{r}_i-\mathbf{R})/\partial\mathbf{r}_i\cdot\partial\mu/\partial\mathbf{v}_i]$$
$$-\gamma[\mathbf{v}\cdot\partial\mu/\partial\mathbf{R}-\partial\chi/\partial\mathbf{R}\cdot\partial\mu/\partial\mathbf{v}+\mathbf{F}\cdot\partial\mu/\partial\mathbf{v}]$$
$$-\gamma[e^{i\omega t}\mathbf{E}\cdot\partial\mu/\partial\mathbf{v}], \quad (2.2)$$

where (μ,H) is the Poisson bracket between μ and H and we have set $m=1$, $M=\gamma^{-2}$, $\mathbf{V}=\gamma\mathbf{v}$,

$$x=(\mathbf{R},\mathbf{v}), \quad y=(\mathbf{r}_1,\cdots\mathbf{r}_n,\mathbf{v}_1,\cdots\mathbf{v}_n). \quad (2.3)$$

Also,

$$\mathbf{F}=-\partial U/\partial\mathbf{R}=-\textstyle\sum_i\partial u(\mathbf{r}_i-\mathbf{R})/\partial\mathbf{R} \quad (2.4)$$

is the microscopic force acting on the B particle. Equation (2.2) is to be solved subject to some initial condition $\mu(x,y,0)$.

The terms in the first square bracket on the right side of (2.2) may be written in the form,

$$(\mu,H_2(y;\mathbf{R}))=iL\mu, \quad (2.5)$$

where

$$H_2(y;\mathbf{R})=H_l(y)+U(y;\mathbf{R}) \quad (2.6)$$

is the Hamiltonian of the fluid in the presence of a B particle *fixed* at $\mathbf{R}$, i.e., $\mathbf{R}$ is a parameter in H_2, not a dynamic variable and L operates only on the fluid variables y. The second and third bracket on the right side of (2.2) may be writen in the form

$$\gamma(\mu,H_1)+\gamma\mathbf{F}\cdot\partial\mu/\partial\mathbf{v}=i\gamma J\mu, \quad (2.7)$$

$$\gamma e^{i\omega t}\mathbf{E}\cdot\partial\mu/\partial\mathbf{v}=i\mathcal{E}\mu \quad (2.8)$$

with the Poisson bracket in (2.7) taken with respect to $\mathbf{R}$ and $\mathbf{v}$. Equation (2.2) may now be written symbolically,

$$i\partial\mu/\partial t=(L+\gamma J+\mathcal{E})\mu\equiv(\mathfrak{L}+\mathcal{E})\mu. \quad (2.9)$$

The distribution of the B particle as a function of $\mathbf{v}$ and $\mathbf{R}$, normalized to unity, is given by

$$f(x,t)=\int\mu(x,y,t)dy. \quad (2.10)$$

The equilibrium distributions in the absence of the

[5] I. Prigogine and P. Résibois, Physica **27**, 629 (1961).

[6] The problem of introducing a spacially uniform electric field in a system with periodic boundary conditions is discussed in Appendix A of the paper by W. Kohn and J. Luttinger, Phys. Rev. **108**, 590 (1957).

electric field are

$$\mu_0(x,y)=Z^{-1}e^{-\beta H},\quad Z=\int e^{-\beta H}dxdy,$$
$$f_0(x)=\int \mu_0(x,y)dy=e^{-\beta\mathcal{H}_1(x)}\Big/\int e^{-\beta\mathcal{H}_1(x)}dx, \tag{2.11}$$

where $\mathcal{H}_1(x)$ is the "effective" Hamiltonian of the B particle

$$\mathcal{H}_1(x)=\tfrac{1}{2}v^2+\chi(\mathbf{R})+w(\mathbf{R}), \tag{2.12}$$

w being the potential of "average force,"

$$e^{-\beta w(\mathbf{R})}=\Omega\int e^{-\beta H_2}dy\Big/\int e^{-\beta[H_2+\chi]}dyd\mathbf{R}. \tag{2.13}$$

The average force vanishes, $w=\text{constant}$, when the system described by H_l is really a fluid, but does not vanish when the B particle is embedded in a crystal (or when we deal with a composite B particle) to which our formalism also applies (cf. Appendix E, Ref. 2). We also introduce the conditional distribution function $P(x,y,t)$ which gives the probability density of finding the fluid at y given that the B particle is at x,

$$P(x,y,t)=\mu(x,y,t)/f(x,t),$$
$$P_0(x,y)=\mu_0(x,y)/f_0(x)$$
$$=\Omega\exp[-\beta(H_2-w)]\Big/\int\exp[-\beta(H_2+\chi)]dyd\mathbf{R}. \tag{2.14}$$

In order to obtain the time evolution of $f(x,t)$, the aim of this whole formalism, we utilize the technique of projection operators developed by Zwanzig.[7,8] Defining the projection operator $\mathcal{P}$,

$$\mathcal{P}(\cdots)=P_0\int(\cdots)dy, \tag{2.15}$$

we have

$$\mathcal{P}\mu(x,y,t)=P_0(x,y)f(x,t). \tag{2.16}$$

Applying the operator $\mathcal{P}$ to the Liouville equation (2.9) we obtain in a straightforward manner

$$\partial f(x,t)/\partial t+\gamma(f,\mathcal{H}_1)+\gamma e^{i\omega t}\mathbf{E}\cdot\partial f/\partial\mathbf{v}$$
$$=-\gamma\partial/\partial\mathbf{v}\cdot\int\mathfrak{F}(1-\mathcal{P})\mu(x,y,t)dy, \tag{2.17}$$

where the Poisson bracket is taken with respect to the variable $\mathbf{v}$, and

$$\mathfrak{F}=\mathbf{F}-\int\mathbf{F}P_0dy=\mathbf{F}-(-\partial w(\mathbf{R})/\partial\mathbf{R}).$$

Applying now the operator $(1-\mathcal{P})$ to (2.9) yields,

$$i(\partial/\partial t)(1-\mathcal{P})\mu=(1-\mathcal{P})(\mathcal{L}+\mathcal{E})[(1-\mathcal{P})\mu+\mathcal{P}\mu]. \tag{2.18}$$

Equation (2.18) can now be solved formally for $(1-\mathcal{P})\mu(x,y,t)$ in terms $f(x,t')$, $t'\leq t$, and $\mu(x,y,0)$ and this solution substituted in (2.17) to obtain a "transport-like" equation for f. Before carrying through this procedure, however, it is convenient at this stage to linearize (2.17) and (2.18) with respect to $\mathbf{E}$, since we are not interested in the complications arising from a strong field. We therefore write

$$\mu(x,y,t)=\mu_0(x,y)+\mu'(x,y,t)+\mathrm{O}(\mathrm{E}^2),\quad t\geq 0, \tag{2.19}$$
$$f(x,t)=f_0(x)+f'(x,t)+O(E^2),\qquad t\geq 0, \tag{2.20}$$

with μ' (and thus f') linear in $\mathbf{E}$ (including their initial values at $t=0$).

Actually the terms $O(E^2)$ cannot be expected to be uniformly small for all times since we expect the system to absorb energy from the external field at a rate proportional to E^2. Thus, for t sufficiently large, μ will be very far from μ_0, no matter how small E is.[9] We are however interested here solely in the B-particle distribution function when the size of the fluid becomes infinitely large while t is fixed, (cf. end of section). Under these conditions we expect that the $O(E^2)$ term in (2.20) can indeed be made arbitrarily small for any fixed x and all t.

The linearized equations (2.17) and (2.18) now assume the form,

$$\partial f'/\partial t+\gamma(f',\mathcal{H}_1)-\beta\gamma e^{i\omega t}\mathbf{E}\cdot\mathbf{v}f_0$$
$$=-\gamma\partial/\partial\mathbf{v}\cdot\int\mathfrak{F}(1-\mathcal{P})\mu'(x,y,t)dy, \tag{2.21}$$

$$i\partial/\partial t[(1-\mathcal{P})\mu']=(1-\mathcal{P})\mathcal{L}[(1-\mathcal{P})\mu'+P_0f'(x,t)]. \tag{2.22}$$

There is no term linear in $\mathbf{E}$ in (2.22) since

$$(1-\mathcal{P})\mathcal{E}\mu_0=(1-\mathcal{P})\mu_0\gamma\beta\mathbf{v}\cdot\mathbf{E}e^{i\omega t}=0. \tag{2.23}$$

The solution of (2.22) may now be written formally in the form

$$(1-\mathcal{P})\mu'(t)=\exp[-it(1-\mathcal{P})\mathcal{L}](1-\mathcal{P})\mu'(0)$$
$$+(1/i)\int_0^t\exp[-it'(1-\mathcal{P})\mathcal{L}]$$
$$\times(1-\mathcal{P})\mathcal{L}P_0\cdot f'(t-t')dt'. \tag{2.24}$$

We shall now assume, for convenience, that at $t=0$, the conditional fluid distribution P is equal to its equilibrium value P_0, i.e.,

$$\mu'(x,y,0)=P_0(x,y)f'(x,0). \tag{2.25}$$

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[7] R. W. Zwanzig, *Lectures in Theoretical Physics* (Interscience Publishers, Inc., New York, 1961), Vol. 3, p. 106.

[8] R. W. Zwanzig (private communication); see also, J. Chem. Phys. **40**, 2527 (1964).

[9] We also have for the total momentum of the system

$$\int[\gamma^{-1}\mathbf{v}+\Sigma\mathbf{v}_i]\mu(t)dxdy=\int_0^t e^{i\omega t'}\mathbf{E}dt'[=\mathbf{E}t,\text{ for }\omega=0].$$

Under these conditions the first term on the right side of (2.24) vanishes and upon substitution of the remainder into (2.21) we are led to the following equation for f':

$$\partial f'(x,t)/\partial t+\gamma(f',\mathcal{H}_1)-\beta\gamma e^{i\omega t}\mathbf{E}\cdot\mathbf{v}f_0$$
$$=i\gamma(\partial/\partial\mathbf{v})\cdot\int dy\mathfrak{F}\int_0^t \exp[-it'(1-\mathcal{P})\mathcal{L}]\times(1-\mathcal{P})\mathcal{L}P_0f'(t-t')dt'$$
$$=\int_0^t\left[\gamma^2\frac{\partial}{\partial\mathbf{v}}\cdot\int dy\mathfrak{F}\exp\{-it'[L+\gamma(1-\mathcal{P})J]\}P_0\mathfrak{F}\times\left(\frac{\partial}{\partial\mathbf{v}}+\beta\mathbf{v}\right)\right]f'(t-t')dt'$$
$$\equiv\int_0^t\mathcal{K}(t',N,\gamma)f'(t-t')dt', \qquad (2.26)$$

where we have used the easily verifiable relations:

$$\mathcal{P}iL(\cdots)=-P_0\int dy(H_2,(\cdots))=0$$

and

$$(1-\mathcal{P})i\mathcal{L}P_0f_0[f'(t-t')/f_0]$$
$$=(1-\mathcal{P})iP_0f_0\gamma J[f'(t-t')/f_0]$$
$$=\gamma P_0f_0\mathfrak{F}\cdot(\partial/\partial\mathbf{v})[f'(t-t')/f_0].$$

Equation (2.26) is to be solved subject to some initial condition $f'(x,0)$, and we have indicated explicitly that $\mathcal{K}$ depends on the size of the fluid, $N=n\Omega$, and on the square root of the mass ratio γ.

It should be emphasized here that the derivation of (2.26) for the B-particle distribution function from the Liouville equation (2.2) for μ, linearized with respect to $\mathbf{E}$, is exact subject to the assumed initial condition (2.25). In particular, if $\mathbf{E}$ is set equal to zero in (2.26) then (2.26) will be an *exact* equation for f subject to μ satisfying the initial condition (2.25). Thus the fact that we want to deal with a macroscopic size fluid, which might be expected to show dissipative-type behavior, or that we shall be interested in $\gamma\ll 1$ has not been introduced yet. Indeed, if $\mathbf{E}=0$ (or if $\mathbf{E}e^{i\omega t}$ is present for all t, but linearization is still meaningful), (2.26) is equally valid for $t\geq 0$ or $t\leq 0$.

Before considering the transition to the limit of an infinite size fluid, $N\to\infty$, $N/\Omega=n$ fixed we must ensure that the *density* of B particles remains finite in this limit. We shall therefore renormalize f by multiplying it through by the constant term, $\Omega'=\int e^{-\beta[\chi+w]}d\mathbf{R}$:

$$\rho(x,t)=\Omega' f(x,t)=\rho_0(x)+\psi(x,t), \qquad (2.27)$$

$$\rho_0(x)=(2\pi/\beta)^{-3/2}\exp-\beta[v^2/2+\chi(\mathbf{R})+w(\mathbf{R})]. \qquad (2.28)$$

Ω' will be proportional to Ω when the B particles are not confined to a limited region of space by their effective potential $\chi+w$, and will equal Ω in a uniform system, f_0 independent of $\mathbf{R}$, in which case $\rho(\mathbf{v},t)$ is just the velocity distribution of the B particles. ψ will satisfy the same equation as f', Eq. (2.26), with f_0 replaced by ρ_0.

We shall now assume, what appears physically as obvious, that ψ considered as a function of t and N, $\psi(t,N)$, will approach a definite limit as $N\to\infty$, $N/\Omega=n$ fixed,

$$\lim_{N\to\infty}\psi(t,N)=\psi(t), \qquad (2.29)$$

and similarly for the operator $\mathcal{K}$ defined in (2.26),

$$\lim_{N\to\infty}\mathcal{K}(t,N,\gamma)=\mathcal{K}(t,\gamma), \qquad (2.30)$$

where the limit $N\to\infty$ of $\mathcal{K}(t,N,\gamma)$ is to be taken *after* the integration over y, in the definition of $\mathcal{K}$, Eq. (2.26) has been performed.

Combining Eqs. (2.26) and (2.30) we finally obtain the following equation for ψ

$$\partial\psi(x,t)+\gamma(\psi,\mathcal{H}_1)-\gamma\beta e^{i\omega t}\mathbf{E}\cdot\mathbf{v}\rho_0(x)=\int_0^t\mathcal{K}(t',\gamma)\psi(x,t-t')dt'. \qquad (2.31)$$

Equation (2.31) is a non-Markoffian irreversible equation which describes the evolution of the B-particle distribution, linearized with respect to $\mathbf{E}$, towards its stationary value.

III. STEADY-STATE DISTRIBUTION FUNCTION

Defining now,

$$\Phi(x,t)=e^{-i\omega t}\psi(x,t), \qquad (3.1)$$

Φ will obey the equation

$$\frac{\partial\Phi(x,t)}{\partial t}+i\omega\Phi+\gamma(\Phi,\mathcal{H}_1)-\beta\gamma\mathbf{E}\cdot\mathbf{v}\rho_0=\int_0^t dt'\mathcal{K}(t',\gamma)e^{-i\omega t'}\Phi(t-t'). \qquad (3.2)$$

We shall now make the physical assumption that $\Phi(x,t)$ approaches a limiting value as $t\to\infty$. The validity of this assumption might be expected to depend on the nature of the potentials $\varphi(r_{ij})$ and $u(\mathbf{r}_i-\mathbf{R})$. This assumption can be weakened by requiring only the existence of the limit of the "Laplace average" of Φ, i.e.,

$$\lim_{t_0\to\infty}\frac{1}{t_0}\int_0^\infty e^{-t/t_0}\Phi(t)dt\equiv\lim_{t_0\to\infty}\bar{\Phi}(t_0)=\bar{\Phi}, \qquad (3.3)$$

exists. $\bar{\Phi}(t_0)$ will satisfy the equation

$$\frac{\Phi(x,0)}{t_0}+\left[\frac{1}{t_0}+i\omega\right]\bar{\Phi}(t_0)+\gamma(\bar{\Phi}(t_0),\mathcal{H}_1)-\beta\gamma\mathbf{E}\cdot\mathbf{v}\rho_0=\tilde{K}(t_0,-\omega,\gamma)\bar{\Phi}(t_0), \qquad (3.4)$$

where $\Phi(x,0)=\psi(x,0)$, and

$$\tilde{K}(t_0,-\omega,\gamma)=\int_0^\infty e^{-t/t_0}e^{-i\omega t}\mathcal{K}(t,\gamma)dt. \qquad (3.5)$$

Assuming now also the existence of

$$\lim_{t_0\to\infty}\tilde{K}(t_0,\omega,\gamma)=\bar{K}(\omega,\gamma), \qquad (3.6)$$

we obtain finally the following equation for $\bar{\Phi}$

$$i\omega\bar{\Phi}(x,\gamma)+\gamma[\mathbf{v}\cdot\partial\bar{\Phi}/\partial\mathbf{R}-(\partial(\chi+w)/\partial\mathbf{R})\cdot\partial\bar{\Phi}/\partial\mathbf{v}] -\beta\gamma\mathbf{E}\cdot\mathbf{v}\rho_0=\bar{K}(-\omega,\gamma)\Phi(x,\gamma), \qquad (3.7)$$

where we have indicated explicitly the γ dependence of Φ.

Expansion in Powers of γ

We consider now the case where $\gamma=M^{-1/2}\ll 1$, and treat the variables $\mathbf{y}_i$ and $\mathbf{v}$ as quantities of $O(1)$, i.e., we assume that the velocity of the B particle $\mathbf{V}=\gamma\mathbf{v}\sim O(\gamma)$ is much smaller (in the range of interest), than the velocities of the fluid particles. We then formally expand $\bar{K}$ in powers of γ and obtain,

$$\bar{K}(-\omega,\gamma)=\beta^{-1}\boldsymbol{\zeta}(\omega):(\partial/\partial\mathbf{v})[\beta\mathbf{v}+\partial/\partial\mathbf{v}]+O(\gamma^3), \qquad (3.8)$$

with the "friction tensor" $\boldsymbol{\zeta}(\omega)$ given by

$$\boldsymbol{\zeta}(\omega)=\beta\gamma^2\lim_{t_0\to\infty}\lim_{N\to\infty}\int_0^\infty dte^{-t/t_0}e^{-i\omega t}\int\mathfrak{F}\mathfrak{F}(-t)P_0dy. \quad (3.9)$$

Here,

$$\mathfrak{F}(-t)=e^{-itL}\mathfrak{F}(y,\mathbf{R}) \qquad (3.10)$$

is the microscopic fluctuating force, $(\mathfrak{F}=\mathbf{F}-\langle\mathbf{F}\rangle_0)$, acting on the B particle at $-t$, when the state of the fluid at $t=0$ is specified by the point y in its own phase space and the B particle is *kept fixed at its position R.* When the fluid is isotropic (our "fluid" can actually be a crystal. Cf. Appendix E, Ref. 2), $\boldsymbol{\zeta}(\omega)$ is diagonal $\boldsymbol{\zeta}(\omega)=\zeta(\omega)\mathbf{1}$ and (3.11) becomes equal to the Fokker-Planck operator on the right-hand side of (1.1) albeit with a frequency-dependent friction constant. The higher order terms in $\bar{K}(\omega,\gamma)$ can also be evaluated readily and are discussed extensively for the case of a spatially uniform system and $\omega=0$, in Ref. 2.

IV. CONNECTION WITH MACROSCOPIC THEORY

In order to prove the equivalence of our microscopic approach and the usual stochastic theory, we need a demonstration of the equivalence between the definitions (1.3) and (1.4) for the friction coefficient.

As will be shown presently, this property is easily demonstrated in a very general way, and does not require a detailed analysis of the behavior of the fluid distribution functions. However, the explicit evaluation of ζ depends on the stationary state which is attained by the one-body fluid distribution function $W_1(\mathbf{r},\mathbf{v},t)$ in the presence of a moving B particle; this problem requires the derivation of a transport equation for $W_1(\mathbf{r}_1,\mathbf{v}_1)$ and is much more complicated.[10] As such an analysis falls outside the general scope of this paper, we shall merely illustrate it here for the case of a dilute gas where W_1 is assumed to obey a Boltzmann equation.

Consider a B particle moving in a fluid with a *prescribed* velocity equal to the real part of $\mathbf{V}_0e^{i\omega t}$, for $t\geq 0$. If we call the fluid coordinates $y'=(\mathbf{r}_1',\cdots\mathbf{r}_N',\mathbf{v}_1',\cdots\mathbf{v}_N')$, the fluid will now be characterized by the time-dependent Hamiltonian $H_2(y';\mathbf{R}(t))$ given in (2.6),

$$H_2(y';\mathbf{R}(t)) =H_l(y')+\sum u[\mathbf{r}_i'-\mathbf{R}_0-\mathbf{V}_0(e^{i\omega t}-1)/i\omega], \quad (4.1)$$

where $\mathbf{R}_0$ is the position of the B particle at $t=0$ and we have set $m=1$. The N-particle distribution function of the fluid (normalized to unity) $W_N'(y',t)$ will evolve according to the Liouville equation,

$$\partial W_N'/\partial t+[W_N',H_2(y';\mathbf{R}(t))]=0. \qquad (4.2)$$

We now make the transformation of variables, $y'\to y=(\mathbf{r}_1,\cdots\mathbf{v}_N)$

$$\mathbf{r}_i=\mathbf{r}_i'-\mathbf{R}(t), \quad \mathbf{v}_i=\mathbf{v}_i'-\mathbf{V}_0e^{i\omega t}. \qquad (4.3)$$

The distribution function $W_N(y,t)$ will now obey the equation,

$$\partial W_N(y,t)/\partial t=(H_2(y),W_N) +i\omega\mathbf{V}_0e^{i\omega t}(\sum_{j=1}^N\partial/\partial\mathbf{v}_j)W_N, \quad (4.4)$$

i.e., $W_N(y,t)$ evolves under the action of the time-independent Hamiltonian $H_2(y)$ and a time-dependent "force"-$i\omega\mathbf{V}_0e^{i\omega t}$ acting on each fluid particle.

We shall now assume that the fluid was in equilibrium at $t=0$,

$$W_N'(y',0)=P_0(y';\mathbf{R}_0) =e^{-\beta H_2(y';\mathbf{R}_0)}\Big/\int e^{-\beta H_2(y';\mathbf{R}_0)}dy', \quad (4.5)$$

and that $\mathbf{V}_0$ is small compared to the molecular velocities, $\beta V_0^2\ll 1$. We may then write

$$W_N(y,t)=P_0(\mathbf{r}_1,\cdots\mathbf{r}_N,\mathbf{v}_1+\mathbf{V}_0e^{i\omega t},\cdots,\mathbf{v}_N+\mathbf{V}_0e^{i\omega t}) +P'(y,t)=P_0(y)-\beta\mathbf{V}_0e^{i\omega t}\cdot(\sum\mathbf{v}_j)P_0+P', \quad (4.6)$$

[10] The general transport equation for $W_1(\mathbf{r}_1,\mathbf{v}_1)$ can easily be obtained using the techniques of I. Progogine and co-workers [for a similar case, see R. Balescu, Physica **27**, 693 (1961); P. Resibois, J. Chem. Phys. **41**, 2979 (1964)]. In particular, the Boltzmann equation (4.18) may be rigorously proved for a dilute gas, provided that the momentum transfer due to the B force during a collision is small with respect to the exchange of momentum between the two particles [P. Resibois (unpublished)].

where P' is linear in $\mathbf{V}_0$, and satisfies the equation

$$\begin{aligned}\partial P'(y,t)/\partial t &= -iLP' + \beta \mathbf{V}_0 e^{i\omega t} iL(\sum v_j P_0) \\ &= -iLP' - \beta \mathbf{V}_0 e^{i\omega t} \mathbf{F} P_0 . \end{aligned} \tag{4.7}$$

The operator L and $\mathbf{F}$ are defined in (2.5) and (2.7), $\mathbf{F}$ being the force on the moving B particle. Equation (4.7) is to be solved with the initial condition $P'(y,0)=0$. This gives

$$\begin{aligned}P'(y,t) &= -\int_0^t e^{-i(t-t')L} \beta \mathbf{V}_0 e^{i\omega t'} \cdot \mathbf{F} P_0 dt' \\ &= -\beta \mathbf{V}_0 e^{i\omega t} \cdot \int_0^t e^{-it'L} e^{-i\omega t'} \mathbf{F} P_0 dt' \\ &= P_V'(y,\omega,t) e^{i\omega t} . \end{aligned} \tag{4.8}$$

Computing now the *macroscopic* force on the moving B particle at time t gives

$$\begin{aligned}\langle \mathbf{F} \rangle &= \int \mathbf{F} W_N(y,t) dy \\ &= -\beta \mathbf{V}_0 e^{i\omega t} \cdot \int_0^t dt' e^{-i\omega t'} \int \mathbf{F}\mathbf{F}(-t') P_0 dy \\ &= \mathbf{F}_V(\omega,t,N) . \end{aligned} \tag{4.9}$$

Upon taking the limit $N\rightarrow\infty$, and $t\rightarrow\infty$ we finally obtain

$$e^{-i\omega t}\mathbf{F}_V(\omega) = \lim_{t\to\infty} \lim_{N\to\infty} e^{-i\omega t} \mathbf{F}_V(\omega,t,N) = -M\zeta(\omega)\mathbf{V}_0 \tag{4.10}$$

in agreement with (1.4): assuming that these limits exist. If the limit $t\rightarrow\infty$ does not exist we have to introduce the convergence factor e^{-t/t_0} as in (3.9).

We have thus shown the complete equivalence between the friction constant appearing in the Fokker-Plank equation for an unconstrained heavy B particle and the "friction" constant appearing in the force acting on a B particle moving with a small prescribed velocity. In the latter case the mass of the B particle does not, of course, enter the analysis.

Evaluation of ζ: Kinetic Theory

In order to actually compute ζ we note that Eq. (4.9) may also be written in the form

$$\langle F \rangle = \int \mathbf{F}_1 W_1(\mathbf{r}_1,\mathbf{v}_1,t) d\mathbf{r}_1 d\mathbf{v}_1 , \tag{4.11}$$

where

$$\mathbf{F}_1 = \partial/\partial \mathbf{r}_1 u(\mathbf{r}_1); \quad \mathbf{F} = \sum_{i=1}^N \mathbf{F}_1(\mathbf{r}_i) , \tag{4.12}$$

and $W_1(\mathbf{r}_1\mathbf{v}_1,t)$ is the one-particle distribution function, of the fluid,

$$W_1(\mathbf{r}_1,\mathbf{v}_1,t) = N\int W_N(y,t) d\mathbf{r}_2 \cdots d\mathbf{r}_N d\mathbf{v}_2 \cdots d\mathbf{v}_N . \tag{4.13}$$

According to (4.6) and (4.8), W_1 may be written for small $\mathbf{V}_0$ in the form

$$\begin{aligned}W_1(\mathbf{r}_1,\mathbf{v}_1,t) &= W_1{}^0(\mathbf{r}_1,\mathbf{v}_1) \\ &\quad - \mathbf{V}_0 e^{i\omega t}[\beta \mathbf{v}_1 W_1{}^0(\mathbf{r}_1,\mathbf{v}_1) + \varphi_1(\mathbf{r}_1,\mathbf{v}_1,t)] , \end{aligned} \tag{4.14}$$

where $W_1{}^0$ is the equilibrium one-particle distribution of the fluid in the presence of an external one-body potential $u(\mathbf{r})$ and

$$\varphi_1 = \beta N \int d\mathbf{r}_2 \cdots d\mathbf{v}_N \int_0^t dt' \times \{\exp[-i(L+\omega)t']\} \mathbf{F} P_0(y) . \tag{4.15}$$

It is now clear that if W_1 obeys any transport equation in the presence of an external potential $u(\mathbf{r}_1)$ and a small oscillating external force, $-i\omega\mathbf{V}_0 e^{i\omega t}$ (in the proper thermodynamic limit and time scale), then $\mathbf{F}_V$ and thus ζ can be computed from the stationary solution of this transport equation subject to the boundary conditions,

$$\begin{aligned}\lim_{|\mathbf{r}_1|\to\infty} W_1(\mathbf{r}_1,\mathbf{v}_1) &= W_1{}^0(\mathbf{r}_1, \mathbf{v}_1 + \mathbf{V}_0 e^{i\omega t}) \\ &= W_1{}^0(\mathbf{r}_1,\mathbf{v}_1)[1 - \beta \mathbf{V}_0 e^{i\omega t} \mathbf{v}_1] \end{aligned} \tag{4.16}$$

linearized with respect to $\mathbf{V}_0$; $\beta \mathbf{V}_0{}^2 \ll 1$. Thus, for the case where the fluid is a dilute gas the appropriate transport equation would be the Boltzmann equation (see footnote, Ref. 10)

$$\begin{aligned}\partial W_1(\mathbf{r}_1,\mathbf{v}_1,t)/\partial t + \mathbf{v}_1 \cdot \partial W_1/\partial \mathbf{r}_1 - \mathbf{F}_1 \cdot \partial W_1/\partial \mathbf{v}_1 \\ - i\omega \mathbf{V}_0 e^{i\omega t} \cdot \partial W_1/\partial \mathbf{v}_1 = \bar{J}(W_1,W_1) , \end{aligned} \tag{4.17}$$

where $\bar{J}$ is the Boltzmann collision operator. Linearizing W_1 in the form (4.14) we obtain the following equation for φ_1,

$$\begin{aligned}i\omega\varphi_1 + \partial\varphi_1/\partial t + (\mathbf{v}_1 \cdot \partial/\partial \mathbf{r}_1 - \mathbf{F}_1 \cdot \partial/\partial \mathbf{v}_1)\varphi_1 - \beta \mathbf{F}_1 W_1{}^0 \\ = \bar{J}(W_1{}^0,\varphi_1) + \bar{J}(\varphi_1,W_1{}^0) , \end{aligned} \tag{4.18}$$

where $W_1{}^0$ is now given by

$$W_1{}^0(\mathbf{r}_1,\mathbf{v}_1) = \bar{n}(2\pi/\beta)^{-3/2} e^{-\beta[\frac{1}{2}v_1{}^2 + u(\mathbf{r}_1)]} , \tag{4.19}$$

where $\bar{n}$ is the particle density far from the source of the external potential. The stationary state achieved by φ_1 as $t\rightarrow\infty$ from which $\mathbf{F}_V$ and ζ may be computed can be found from (4.18) by setting $\partial\varphi_1/\partial t = 0$ there and solving the resulting time-independent equation with the boundary condition $\varphi_1 \rightarrow 0$ as $|\mathbf{r}_1| \rightarrow \infty$.

Evaluation of ζ: Hydrodynamics

It is seen from (4.11) that if we are only interested in computing $\langle \mathbf{F} \rangle$, it is not necessary to know the complete $W_1(\mathbf{r}_1,\mathbf{v}_1,t)$. Rather it is sufficient to know the one-particle fluid density

$$n(\mathbf{r}_1,t) = \int W_1(\mathbf{r}_1,\mathbf{v}_1,t) d\mathbf{v}_1 , \tag{4.20}$$

since

$$\langle \mathbf{F} \rangle = \int \mathbf{F}_1(\mathbf{r}_1) n(\mathbf{r}_1,t) d\mathbf{r}_1 . \tag{4.21}$$

For obtaining $\mathbf{F}_V(\omega)$ and $\zeta(\omega)$ only the steady-state value of n is necessary.

The density $n(\mathbf{r},t)$ satisfies the continuity equation[11]

$$\partial n(\mathbf{r},t)/\partial t + \partial \cdot n\mathbf{u}(\mathbf{r},t)/\partial \mathbf{r} = 0 , \tag{4.22}$$

where $\mathbf{u}(\mathbf{r},t)$ [not to be confused with the external potential $u(\mathbf{r})$] is the local velocity

$$\mathbf{u}(\mathbf{r},t) = [n(r,t)]^{-1} \int \mathbf{v}_1 W_1(\mathbf{r},\mathbf{v}_1,t) d\mathbf{v}_1 . \tag{4.23}$$

The momentum density $n\mathbf{u}$ obeys in turn the dynamical equation, appropriate to the present problem,

$$\partial (n\mathbf{u})/\partial t + \partial \cdot \mathbf{p}(\mathbf{r},t)/\partial \mathbf{r} + n\mathbf{F}_1(\mathbf{r}) + i\omega \mathbf{V}_0 e^{i\omega t} n = 0 , \tag{4.24}$$

where $\mathbf{p}$ is the stress tensor of the fluid which will be some functional of W_1 whenever W_1 obeys a transport equation. In particular, for a *dilute gas*,

$$\mathbf{p}(\mathbf{r},t) = \int \mathbf{v}_1 \mathbf{v}_1 W_1(\mathbf{r},\mathbf{v}_1,t) d\mathbf{v}_1 . \tag{4.25}$$

Equation (4.24) may also be used to compute $\langle \mathbf{F} \rangle$. The result is especially simple for $\omega = 0$, when we obtain for the stationary value of $\langle \mathbf{F} \rangle$,

$$\mathbf{F}_V = -\oint \mathbf{p} \cdot d\mathbf{S} , \tag{4.26}$$

where the integration is over any surface enclosing the origin but sufficiently far from it that $\mathbf{F}_1(\mathbf{r})$ vanishes outside this surface.

Equations (4.22) and (4.24) are not closed as they stand since they contain in addition to n and $\mathbf{u}$ also $\mathbf{p}$. It is only when $\mathbf{p}$ is expressible as a functional of n and $\mathbf{u}$ (or of n, $\mathbf{u}$, and some other variables for which new equations are supplied[11]) that there will exist a closed set of hydrodynamic equations, e.g., the Navier-Stokes equations, for the fluid. When such a set of equations exists for our system, i.e., for the fluid in the presence of an external potential $u(\mathbf{r})$ and a small oscillating external force $-i\omega \mathbf{V}_0 e^{i\omega t}$ then $\mathbf{F}_V(\omega)$ and $\zeta(\omega)$ may be computed from the stationary solution of these equations (linearized with respect to $\mathbf{V}_0$) subject to the appropriate boundary conditions at infinity.

An investigation of the conditions necessary to ensure the validity of the hydrodynamic equations for the description of the steady state is outside the range of this paper. Intuitively, the requirement would seem to be that the length and time scale associated with $u(\mathbf{r}_1)$ and ω^{-1} should be large compared to the microscopic length and time scales associated with the fluid. For a dilute gas these latter are, respectively, the mean free path and mean free time between collisions.[12]

The hydrodynamic equations and hence $\zeta(\omega)$ will depend explicitly on the parameters of the fluid, such as its viscosity η, as well as on the nature of the "external" potential $u(\mathbf{r})$. In some cases the effect of $u(\mathbf{r})$ may be represented by appropriate boundary conditions. Thus, if $u(\mathbf{r})$ represents a rigid-sphere type of interaction

$$\begin{aligned} u(\mathbf{r}) &= \infty , \quad |\mathbf{r}| < \tfrac{1}{2}a , \\ &= 0 , \quad |\mathbf{r}| > \tfrac{1}{2}a , \end{aligned} \tag{4.27}$$

then $n(\mathbf{r})$ will be zero for $|\mathbf{r}| < \frac{1}{2}a$. The hydrodynamic equations will then have to be solved[13] subject to the conditions that the normal component of $\mathbf{u}(\mathbf{r})$ as well as the tangential components of the stress $\mathbf{p}$ vanish at $|r| = \frac{1}{2}a$. A solution of the Navier-Stokes equations with these boundary conditions is given by Lamb,[14] for the case $\omega = 0$, and yields

$$\mathbf{F}_V(0) = -4\pi\eta a \mathbf{V}_0 = -M\zeta(0)\mathbf{V}_0 . \tag{4.28}$$

As mentioned before the validity of the Navier-Stokes equations for this problem requires that a be very large compared to the mean free path in the fluid. Hence the value of $\zeta(0)$ given in (4.28) does not conflict with the value of $\zeta(0)$ computed for a heavy sphere moving in an ideal gas[15] where the mean free path is infinite.

Finally we might mention that it is not clear to us whether there exists any potential $u(\mathbf{r})$ which leads to Navier-Stokes equations with a boundary condition that the fluid velocity, both normal and tangential, vanish at the "surface" of the B particle. It is this boundary condition which leads to Stokes' law $\mathbf{F}_V = -6\pi\eta a \mathbf{V}_0$, commonly used for B particles.[16] Perhaps such a boundary condition arises when one considers a "composite B particle" with internal structure. A fluid particle can then be "absorbed" by the B particle and re-emitted with zero average velocity.

V. AN ALTERNATIVE FORMULATION

Let us now return to the general time-dependent equation (2.31),

$$\partial \psi(x,t)/\partial t + \gamma(\psi, H_1) - \gamma\beta e^{i\omega t} \mathbf{E} \cdot \mathbf{v} \rho_0(x) = \int_0^t \mathcal{K}(t-t', \gamma) \psi(x,t') dt' , \tag{5.1}$$

[11] See, e.g., H. S. Green, *Molecular Theory of Fluids* (North-Holland Publishing Company, Amsterdam, 1952).

[12] H. Grad, *Encyclopedia of Physics*, edited by S. Flügge (Springer-Verlag, Berlin, 1958), Vol. XII, p. 205.

[13] G. Grad (private communication) believes that this can be shown rigorously for the case of a dilute gas satisfying the Boltzmann equation.

[14] H. Lamb, *Hydrodynamics* (Dover Publications, Inc., New York, 1945), 6th ed., Art. 337, p. 602.

[15] J. L. Lebowitz, Phys. Rev. **114**, 1192 (1959), and references cited there.

[16] A very interesting derivation of Stokes law which expresses ζ in the form (1.2) but with $\mathfrak{F}$ representing a "hydrodynamic force" has been given recently by R. Zwanzig, J. Res. Natl. Bur. Std. **68B**, 143 (1964).

where we have replaced, for a fluid $\mathcal{H}_1$ by H_1. A remarkable feature of this equation is that, except for the trivial flow terms, the complete and exact dynamics of the problem has been incorporated in the collision operator $\mathcal{K}(t,\gamma)$. This result seems at first sight in contradiction with the general theory of approach to equilibrium developed by Prigogine and one of us[5] (P.R.). Indeed it is expected in general that the transport equation for ψ should have the following form:

$$\frac{\partial\psi(x,t)}{\partial t}+\gamma(\psi,H_1)-\gamma\beta e^{i\omega t}\mathbf{E}\cdot\mathbf{v}\left[\rho_0+\int_0^t dt'\mathfrak{D}_0(t-t',\gamma)\rho_0\right]=\mathfrak{D}_0(t,\gamma)\psi(x,0)+\int_0^t G(t-t',\gamma)\psi(x,t')dt'. \quad (5.2)$$

Here $G(t,\gamma)$ is a generalized collision operator, to be defined below; the so-called "destruction term" $\mathfrak{D}_0(t,\gamma)\psi(x,0)$ represents the effect of the initial spatial correlations while the last term on the left side describes the acceleration of the particle due to the external field during a collision process; these two latter terms are not present in (5.1). A similar paradox was found in the stationary situation discussed in Ref. 3.

We shall now show that it is possible indeed to write down an alternative transport equation for $\psi(x,t)$ which has precisely the structure (5.2); this equation is the exact analog of the general kinetic equation of Prigogine-Résibois, as applied to the present problem. However, the operators $\mathfrak{D}_0$ and G are not independent of $\mathcal{K}$: they are connected in such a way that the solutions of the two equations (5.1) and (5.2) are identical for all times. In order to avoid long calculations in the main text, some proofs are left for the Appendix.

Let us start again with the Liouville equation (2.9), linearized in the external field:

$$i\partial\mu'/\partial t-\mathcal{E}P_0f_0=(L+\gamma J)\mu', \quad (5.3)$$

and let us assume again the initial condition (2.25).

We define a projection operator I by

$$I(\cdots)=\frac{\rho_0^f(\{\mathbf{v}\})}{\Omega^N}\int dy(\cdots), \quad (5.4)$$

where $\rho_0^f(\{\mathbf{v}\})$ is the fluid equilibrium *velocity* (distribution function):

$$\rho_0^f(\{\mathbf{v}\})=(2\pi/\beta)^{-3N/2}\exp[-\beta\textstyle\sum_i v_i^2/2]. \quad (5.5)$$

As will be seen later (Appendix A) the motivation for introducing the operator I is that it is closely related to the so-called "irreducibility condition" in Fourier space, which plays a very important role in the calculation of Ref. 3. We then notice the two useful identities:

$$IL=0, \quad (5.6)$$

$$f'(\mathbf{R},\mathbf{v},t)=\{\rho_0^f(\{\mathbf{v}\})/\Omega^N\}^{-1}I\mu'(x,y,t), \quad (5.7)$$

and introduce the abbreviation:

$$\Gamma(x,y,t)=(1-I)\mu'(x,y,t). \quad (5.8)$$

We now apply the operators I and $(1-I)$ to the Liouville equation (5.3); we get, respectively,

$$\frac{\partial f'(x,t)}{\partial t}+\gamma(f',H_1(x))-\gamma\beta e^{i\omega t}\mathbf{E}\cdot\mathbf{v}f_0=-\gamma\frac{\partial}{\partial v}\int dy\mathbf{F}\Gamma(x,y,t), \quad (5.9)$$

$$\frac{i\partial\Gamma(x,y,t)}{\partial t}-(1-I)P_0\mathcal{E}f_0=\{L+\gamma(1-I)J\}\frac{\rho_0^f(\{\mathbf{v}\})}{\Omega^N}f(x,t)+\{L+\gamma(1-I)J\}\Gamma(x,y,t). \quad (5.10)$$

The similarity between (5.9), (5.10), and (2.21), (2.22) is striking; the formal manipulations followed here will thus be the same as in Sec. II: one writes the formal solution of (5.10), which is then inserted into (5.9). The limit process $N\to\infty$, $(N/\Omega)=n$ finite is applied as before; assuming that this limit exists, we immediately obtain Eq. (5.2) with the following definitions:

$$\mathfrak{D}_0(t,\gamma)=-\gamma\frac{\partial}{\partial\mathbf{v}}\int dy\ \mathbf{F}(\exp\{-i[L+\gamma(1-I)J]t\})\times(1-I)P_0(x,y), \quad (5.11)$$

$$G(t,\gamma)=-i\gamma\frac{\partial}{\partial\mathbf{v}}\int dy\ \mathbf{F}(\exp\{-i[L+\gamma(1-I)J]t\})\times[L+\gamma(1-I)J]IP_0(x,y), \quad (5.12)$$

where we have used the identity:

$$IP_0(x,y)=\rho_0^f(\{v\})/\Omega^N. \quad (5.13)$$

The formal device of introducing I has thus allowed us to express $\mathfrak{D}_0$ and G in a closed form; this provides us with an explicit transport equation for $\psi(x,t)$ which has exactly the same structure as the Prigogine-Résibois transport equation. More precisely, it may be shown that Eq. (5.2) together with (5.11) and (5.12) is identical to this equation, when this latter is specialized to our particular Brownian problem. This result is discussed in Appendix A for the stationary solution of (5.2).

Let us now show that the solutions of Eqs. (5.1) and (5.2) are identical. We introduce the Laplace transform of ψ:

$$\bar{\psi}(x,z)=\int_0^\infty \exp[izt]\psi(x,t)dt, \quad (5.14)$$

$$\psi(x,t)=\frac{-1}{2\pi i}\oint dz\exp[-izt]\bar{\psi}(x,z), \quad (5.15)$$

where the contour C is parallel to the real axis above the singularities of $\bar{\psi}$. Similarly we define $\bar{K}(z,\gamma)$ [see also (3.5)], $\bar{G}(z,\gamma)$ and $\bar{D}_0(z,\gamma)$. From (5.1), we obtain easily:

$$-iz\bar{\psi}(x,z)-\psi(x,0)+\gamma(\bar{\psi},H_1)-\gamma(i/(\omega+z))\beta\mathbf{E}\cdot\mathbf{v}\rho_0(x) \\ =\bar{K}(z,\gamma)\bar{\psi}(x,z)\,, \quad (5.16)$$

while Eq. (5.2) gives

$$-iz\bar{\psi}(x,z)-\psi(x,0)+\gamma(\bar{\psi},H_1) \\ -\gamma(i/(\omega+z))[1+\bar{D}_0(z,\gamma)]\beta\mathbf{E}\cdot\mathbf{v}\rho_0(x) \\ =\bar{D}_0(z,\gamma)\psi(x,0)+\bar{G}(z,\gamma)\bar{\psi}(x,z)\,. \quad (5.17)$$

If we set,

$$i\gamma L_0\cdots=\gamma(\cdots,H_1)\,, \quad (5.18)$$

$$i\gamma\delta L^I=-\gamma\mathbf{F}\cdot(\partial/\partial\mathbf{v})\,, \quad (5.19)$$

and solve (5.16) and (5.17) formally, we get

$$\bar{\psi}(x,z)=[-iz+i\gamma L_0-\bar{K}(z,\gamma)]^{-1} \\ \times\{\psi(x,0)+(i/(\omega+z))\beta\gamma\mathbf{E}\cdot\mathbf{v}\rho_0(x)\}\,, \quad (5.16')$$

$$\bar{\psi}(x,z)=[-iz+i\gamma L_0-\bar{G}(z,\gamma)]^{-1}[1+\bar{D}_0(z,\gamma)] \\ \times\{\bar{\psi}(x,0)(i/(\omega+z))\beta\gamma\mathbf{E}\cdot\mathbf{v}\rho_0(x)\}\,, \quad (5.17')$$

where we have explicitly:

$$\bar{K}(z,\gamma)=i\int dy\gamma\delta L^I\frac{1}{(1-\mathcal{P})\mathcal{L}-z}(1-\mathcal{P})\mathcal{L}P_0(x,y)\,, \quad (5.20)$$

$$\bar{G}(z,\gamma)=i\int dy\gamma\delta L^I\frac{1}{(1-I)\mathcal{L}-z}(1-I)\mathcal{L}IP_0(x,y)\,, \quad (5.21)$$

$$\bar{D}_0(z,\gamma)=-\int dy\gamma\delta L^I\frac{1}{(1-I)\mathcal{L}-z}(1-I)P_0(x,y)\,. \quad (5.22)$$

Obviously, the solutions of the two transport equations will be the same provided the following identity holds:

$$\{-iz+i\gamma L_0-\bar{K}(z,\gamma)\}^{-1} \\ =\{-iz+i\gamma L_0-\bar{G}(z,\gamma)\}^{-1}[1+\bar{D}_0(z,\gamma)]\,, \quad (5.23)$$

which may be written as

$$\bar{G}(z,\gamma)-iz\bar{D}_0(z,\gamma)+i\gamma\bar{D}_0(z,\gamma)L_0 \\ =[1+\bar{D}_0(z,\gamma)]\bar{K}(z,\gamma)\,. \quad (5.24)$$

Using Eqs. (5.20), (5.21), and (5.22), the validity of (5.24) may be proven by direct algebraic manipulations. The explicit proof is left to Appendix A.

This establishes the link between the methods used respectively in Refs. 2 and 3. We just want to add the following remarks:

(1) The simplicity of (5.1) is related to the very particular choice of an initial condition (2.25); in a more general case, we would also find in (5.1) a "destruction term" which keeps trace of the initial correlations in the system [see Eq. (2.24) and Ref. 2, Sec. V].

(2) If one considers the limit of small γ, the two transport equations (5.1) and (5.2) lead to the same explicit form. In particular, if we study the relaxation of the B particle toward its equilibrium value in absence of external field ($E=0$):

$$\psi(x,t)=\rho(x,t)-\rho_0(x)\to 0 \quad \text{as} \quad t\to\infty\,. \quad (5.25)$$

In the limit $\gamma^2\ll 1$, one has to take the time scale $t\to\infty$ t/γ^2 finite[2,3]; in this case both Eqs. (5.1) and (5.2) reduce to the time-dependent Fokker-Planck equation (1.1) with $\zeta=\zeta(0)$ given by (3.12).

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APPENDIX A: THE EQUIVALENCE OF EQS. (5.1) AND (5.2)

Let us first prove the identity (5.24). Using the definitions (5.20), (5.21), and (5.22), together with the obvious identity valid for an arbitrary function $A(x,y)$

$$(\mathcal{P}-I)A(x,y)=\left(P_0-\frac{\rho_0{}^f(\{\mathbf{v}\})}{\Omega^N}\right)\int dy\,A(x,y)$$

$$=(1-I)P_0(x,y)\int dy\,A(x,y)\,, \quad (\text{A1})$$

one obtains by straightforward calculations:

$$-iz\bar{D}_0(z,\gamma)+\bar{G}(z,\gamma)+i\gamma\bar{D}_0(z,\gamma)L_0 \\ =i\int dy\gamma\delta L^I\frac{1}{(1-I)\mathcal{L}-z}(1-\mathcal{P})\mathcal{L}P_0\,, \quad (\text{A2})$$

while

$$[1+\bar{D}_0(z,\gamma)]\bar{K}(z,\gamma) \\ =i\int dy\gamma\delta L^I\frac{1}{(1-I)\mathcal{L}-z}(1-\mathcal{P})\mathcal{L}P_0 \\ +i\left\{\int dy\gamma\delta L^I\frac{1}{(1-I)\mathcal{L}-z}(1-I)P_0\right\} \\ \times\gamma L_0\left\{\int dy\frac{1}{(1-I)\mathcal{L}-z}(1-\mathcal{P})\mathcal{L}P_0\right\}\,. \quad (\text{A3})$$

We see thus that (5.24) will be satisfied provided the second term on the right side of (A3) vanishes. However, one has

$$\int dy\bar{\Gamma}(x,y,z)=0\,, \quad (\text{A4})$$

where

$$\bar{\Gamma}(x,y,z)=\frac{1}{(1-\mathcal{P})\mathcal{L}-z}(1-\mathcal{P})P_0\,. \quad (\text{A5})$$

Indeed, we have

$$[(1-\mathcal{P})\mathcal{L}-z]\bar{\Gamma}(x,y,z)=(1-\mathcal{P})\mathcal{L}P_0. \tag{A6}$$

Applying the operator $\mathcal{P}$ to both sides of (A6), we get

$$-z\mathcal{P}\Gamma(x,y,z)\equiv -zP_0\int dy\Gamma(x,y,z)=0, \tag{A7}$$

which is precisely (A4).

This completes the proof of the equivalence between the two methods used in the text.

In order to establish the contact with our previous work, we still have to prove that in the case where $\mathbf{E}$ is independent of time ($\omega=0$) and in the absence of external force ($\chi(\mathbf{R})=0$), Eq. (5.2) which becomes now in the limit $t\rightarrow\infty$:

$$-\gamma\beta\mathbf{E}\cdot\mathbf{v}\rho_0(\mathbf{v})-\gamma\beta\bar{D}_0(0,\gamma)\mathbf{E}\cdot\mathbf{v}\rho_0(v) =\bar{G}(0,\gamma)\psi(x,t\rightarrow\infty) \tag{A8}$$

is identical to Eq. (4.15) of Ref. 3.

Let us consider the collision operator:

$$\bar{G}(0,\gamma)=i\int dy\gamma\delta L^I\frac{1}{L+\gamma(1-I)J-i0} \times[L+\gamma(1-I)J]IP_0(x,y). \tag{A9}$$

Here, some care has to be taken in going to the limit $z\rightarrow i0$; indeed, at first sight it seems that

$$\lim_{z\rightarrow i0}\bar{G}(z,\gamma)=i\int dy\gamma\delta L^IIP_0(x,y)=0,$$

which is obviously wrong. However, for finite z, Eq. (A9) may be rewritten as

$$\bar{G}(z,\gamma)=i\int dy\gamma\delta L^I\frac{1}{L+\gamma(1-I)J-z} \times(1-I)\gamma J\left(\frac{-z}{L-z}IP_0\right), \tag{A10}$$

and we have

$$\lim_{z\rightarrow i0}\frac{-z}{L-z}IP_0(x,y)=P_0(x,y). \tag{A11}$$

This result was proved in Ref. 3 using a perturbation expansion for L [see Ref. 3, Eq. (4.12)].

Using (A10) and (A11), we get immediately the expansion:

$$\bar{G}(i0,\gamma)=i\int dy(-\gamma\delta L^I) \times\sum_{n=1}^{\infty}\left[\left(\frac{-1}{L-i0}\right)\gamma(1-I)J\right]^nP_0. \tag{A12}$$

We now write explicitly

$$\int dy\equiv\int d\{\mathbf{r}\}d\{\mathbf{v}\}$$

and take the Fourier transform of the operators involved in (A10) with respect to $\{\mathbf{r}\}$; we use for this the completeness relation

$$\sum_{\{\mathbf{k}\}}|\{\mathbf{k}\}\rangle\langle\{\mathbf{k}\}|=\delta(\mathbf{r}-\mathbf{r}'), \tag{A13}$$

where $|\{\mathbf{k}\}\rangle$ is the eigenvector in Hilbert space which has the spatial representation

$$\langle\{\mathbf{r}\}|\{\mathbf{k}\}\rangle=\Omega^{-N/2}\exp[i\textstyle\sum_j\mathbf{k}_j\cdot\mathbf{r}_j]. \tag{A14}$$

We obtain then from (A12)

$$\begin{aligned}\bar{G}(i0,\gamma)=i\int d\{\mathbf{v}\}\sum_{\{\mathbf{k}'\}\{\mathbf{k}''\}\cdots}\Bigg[\langle 0|\gamma\delta L^I\frac{1}{L-i0}|\{\mathbf{k}'\}\rangle \\ \times\Big\{\langle\{\mathbf{k}'\}|(1-I)\gamma JP_0|0\rangle \\ +\langle\{\mathbf{k}'\}|(1-I)\gamma J\frac{(-1)}{L-i0}|\{\mathbf{k}''\}\rangle \\ \times\langle\{\mathbf{k}''\}|(1-I)\gamma JP_0|0\rangle+\cdots \\ +\langle\{\mathbf{k}'\}|(1-I)\gamma J\frac{(-1)}{L-i0}|\{\mathbf{k}''\}\rangle\cdots \\ \times\langle\{\mathbf{k}^{(n)}\}|(1-I)\gamma JP_0|0\rangle\Big\}\Bigg].\end{aligned} \tag{A15}$$

But we have, for two arbitrary functions or operators F_1 and F_2 depending only on a finite number of fluid particles

$$\begin{aligned}&\int d\{\mathbf{v}\}F_1(\{\mathbf{v}\})|\{\mathbf{k}\}\rangle\langle\{\mathbf{k}\}|(1-I)F_2(\{\mathbf{v}\})P_0 \\ &\equiv\int d\{\mathbf{v}\}F_1(\{\mathbf{v}\})|\{\mathbf{k}\}\rangle \\ &\quad\times\left[\langle\{\mathbf{k}\}|F_2P_0-\rho_0{}^f\int d\{\mathbf{v}\}\langle 0|F_2P_0\delta_{\{k\},0}{}^{Kr}\right] \qquad (A16)\\ &=\int d\{\mathbf{v}\}F_1(\{\mathbf{v}\})|\{\mathbf{k}\}\rangle\langle\{\mathbf{k}\}|F_2P_0,\quad \{\mathbf{k}\}\neq 0 \\ &=0 \qquad\qquad \{\mathbf{k}\}\equiv 0. \qquad (A17)\end{aligned}$$

This latter formula holds in the thermodynamic limit, because the contributions involving the same particle in both F_1 and F_2 are of negligible weight as compared to the others (see Ref. 3, Appendix 1 for more details).

We get then

$$\bar{G}(i0,\gamma)=i\int d\{\mathbf{v}\}\langle 0|-\gamma\delta L^I \sum_{n=1}^{\infty}\left(\frac{-1}{L-i0}\gamma J\right)^n P_0|0\rangle\,, \tag{A18}$$

where the prime means that all intermediate states should correspond to nonvanishing wave numbers $\{\mathbf{k}\}\neq 0$: Equation (A18) is precisely the complete expansion of the right side of Eq. (4.15), Ref. 3. The same calculation may be developed for the left side of (A8).

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Energy Spectrum of a Simple Bose-Einstein Model*

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A soluble model of a Bose-Einstein system is examined, in which the interaction energy between particles is attractive as the momentum approaches zero. The model is based on one proposed by Bassichis and Foldy.

I. INTRODUCTION

THE second-quantized form of the Hamiltonian for a system of Bose-Einstein particles interacting via a two-body potential is given by

$$H=\sum_k \omega_k a_\omega{}^* a_k+(1/2\Omega)\sum_{k_1,k_2,k_3,k_4} v(|\mathbf{k}_1-\mathbf{k}_3|) \times a_{k_4}{}^* a_{k_3}{}^* a_{k_2} a_{k_1}\delta(\mathbf{k}_4+\mathbf{k}_3-\mathbf{k}_2-\mathbf{k}_1)\,, \tag{1}$$

where $a_{\mathbf{k}}$ and $a_{\mathbf{k}}{}^*$ are the annihilation and creation operators for particles in the state of momentum $\mathbf{k}$, Ω is the volume occupied by the system, and $v(|\mathbf{k}|)$ is the Fourier transform of the two-body potential. The Bogoliubov[1] approximation consists of assuming that most of the particles are in the state $k=0$ and that it is thus permissible to replace $a_0{}^*$ and a_0 by c numbers equal to $\sqrt{N}$, where N is the number of particles in the system. One then retains those portions of the interaction which are of order N. This procedure yields an energy spectrum

$$E(m_k)=\sum_{k\neq 0} m_k(\omega_k{}^2+2\omega_k v(k)N/\Omega)^{1/2}+E_g\,, \tag{2}$$

where m_k is the number of elementary excitations of momentum $\mathbf{k}$, and E_g is the energy of the ground state. This result may be improved by using either a pseudopotential[2,3] or the single-particle Green's function.[4] In addition, the number of particles in the state $k=0$ is taken to be N_0 rather than N, where N_0 is the average value of $a_0{}^* a_0$. The energy of an elementary excitation of momentum $\mathbf{k}$ is then modified to

$$\epsilon_{\mathbf{k}}=(\omega_k{}^2+8\pi\omega_k N_0 a/\Omega m)^{1/2} \tag{3}$$

where a is the S-wave scattering length. ($\hbar=1$). For sufficiently small momenta this expression is real only if a is positive. In this case we obtain an acoustic dispersion, and hence a zero energy gap between the ground state and the lowest excited states. However, if a is positive these low-momenta excitations are unstable against decay into two or more excitations, which is not observed in liquid helium II. Unfortunately, a negative scattering length means a Bose-Einstein system cannot be dilute, which is an important assumption for the derivation of the above excitation spectrum. It is thus of interest to examine a soluble model in which the interaction energy between particles is attractive in the limit that the momentum approaches zero. The model we propose examining is one that was studied by Bassichis and Foldy[5] earlier.

II. THE BASSICHIS AND FOLDY MODEL

This model consists of extracting from H of Eq. (1) only those terms involving three single-particle levels of momenta k, $-k$, and zero. Thus, we consider the Hamiltonian[6]

$$\begin{aligned} h=&\,\omega_k(n_k+n_{-k}) \\ &+\frac{v(k)}{\Omega}[n_0(n_k+n_{-k})+a_0{}^{*2}a_k a_{-k}+a_0{}^2 a_k{}^* a_{-k}{}^*] \\ &+\frac{v(0)}{2\Omega}[2n_0(n_k+n_{-k})+2n_k n_{-k}+n_k{}^2-n_k \\ &\qquad +n_{-k}{}^2-n_{-k}+n_0{}^2-n_0]+\frac{v(2k)}{\Omega}n_k n_{-k}\,, \end{aligned} \tag{4}$$

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