Quantum-Mechanical, Microscopic Brownian Motion

J. McKENNA AND H. L. FRISCH

Bell Telephone Laboratories, Inc., Murray Hill, New Jersey

(Received 9 December 1965)

Recently, the Fokker-Planck (F-P) equation, satisfied by the phase-space distribution function of a Brownian (B) particle, has been derived from the Liouville equation, satisfied by the phase-space distribution function of the composite system, B particle plus fluid. We have started from a strictly quantum-mechanical formulation of the problem, using a density-matrix description of the composite system, and have shown that in an appropriate classical limit the F-P equation results. To lowest order in $\hbar$, our technique gives the same expression for the friction coefficient obtained earlier from the Liouville equation. In principle it yields corrections to the F-P equation to all orders in $\hbar$. We have shown that for a uniform fluid, the quantum corrections to the F-P equation to first order in $\hbar$ appear only as a shift in the friction coefficient. If the classical friction coefficient ζ_0 is replaced by $\zeta_0+\hbar\zeta_1$, with an appropriate ζ_1, then the F-P equation is given correctly to order $\hbar$. There is a difference in the mass dependence of ζ_0 and ζ_1 which suggests the existence of an isotope effect in the mobility of a particle moving through a quantum fluid. The technique employed in the derivation is to consider the expectation value of the density matrix of the system in a state in which the position and momentum of the particles are known with maximum accuracy (Kennard packet). This expectation value μ is an appropriate, positive, semiclassical phase-space distribution function. An appropriate equation is derived for μ, which to lowest order in $\hbar$ is the Liouville equation. The remainder of the derivation is similar to the derivation in the classical case (a suitable projection technique is used).

I. INTRODUCTION

IN the theory of Brownian motion, the Fokker-Planck equation occupies a central position.[1] Let M be the mass of a Brownian (B) particle interacting with a fluid of particles of mass m ($m\ll M$), and let $\mathbf{R}$ and $\mathbf{V}$ be the position and velocity of the B particle. Then the probability at time t that the B particle will be found in the region $d\mathbf{R}$ at $\mathbf{R}$ and that its velocity will be in the region $d\mathbf{V}$ at $\mathbf{V}$ is $f(t;\mathbf{R},\mathbf{V})d\mathbf{R}d\mathbf{V}$. The function $f(t;\mathbf{R},\mathbf{V})$ is the phase-space distribution function of the B particle and it is an appropriate solution of the Fokker-Planck equation

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

In (1.1) $\mathbf{Y}$ is the total nondissipative force acting on the B particle, ζ is the friction coefficient, and $\beta=(kT)^{-1}$, where k is Boltzmann's constant and T is the temperature.

Recently, Lebowitz and Rubin,[2] Résibois and Davis,[3] and Lebowitz and Résibois[4] have given new derivations of the Fokker-Planck equation which are independent of the usual stochastic assumptions.[1] These derivations all start with the classical Liouville equation describing the B particle interacting with the fluid particles. It is the purpose of this paper to show how the Fokker-Planck equation and its quantum-mechanical generalization can be derived starting from the quantum-mechanical density matrix describing the B particle interacting with the fluid particles.

In Ref. 4 a system is considered consisting of a B particle of mass M and charge q interacting with N fluid particles of mass m and zero charge. The whole system is enclosed in a cube of volume Ω and subject to a very small, spatially constant, but time-varying electric field $\mathbf{E}\cos(\omega t+\alpha)$. The authors start with the Liouville equation satisfied by the distribution function μ of the whole system consisting of the B particle plus the N fluid particles. By integrating out the variables of the N fluid particles, linearizing with respect to $\mathbf{E}$, and taking the limit as $N\rightarrow\infty$, $\Omega\rightarrow\infty$, $N/\Omega=\rho=$ constant, a non-Markoffian transport equation for f results. In the limit as $t\rightarrow\infty$ this becomes a Markoffian transport equation for the steady-state distribution function of the B particle, and this transport equation, when expanded in powers of the parameter $\gamma=(m/M)^{1/2}$, is to lowest order in γ just the Fokker-Planck equation. Furthermore, this derivation yields an explicit expression for the friction coefficient ζ as an autocorrelation function of the force exerted by the fluid particles on the B particle. It was also shown in Ref. 2 that higher order corrections in γ to the Fokker-Planck equation could be calculated.

In this paper[5] we also start by considering the same system of the B particle plus N fluid particles. (We consider an electric field independent of time, but our technique can easily be extended to cover fields varying as $\mathbf{E}\cos\omega t$.) However, we suppose the system to be governed by the laws of quantum mechanics, and therefore the statistical properties of the system are given by

[1] S. Chandrasekhar, Rev. Mod. Phys. **15**, 1 (1943); this paper gives a review of the theory and gives references to earlier work.

[2] J. L. Lebowitz and E. Rubin, Phys. Rev. **131**, 2381 (1963).

[3] P. Résibois and H. T. Davis, Physica **30**, 1077 (1964).

[4] J. L. Lebowitz and P. Résibois, Phys. Rev. **139**, A1101 (1965).

[5] A preliminary account of this work appeared in J. McKenna and H. L. Frisch, Phys. Letters **19**, 112 (1965). [*Note added in proof.* It has been brought to our attention that this problem has also been studied recently from the point of view of the Wigner transform by H. T. Davis, K. Hiroike, and S. A. Rice, J. Chem. Phys. **43**, 2633 (1965).]

the density matrix. Our starting point is therefore the operator equation satisfied by the density matrix. By taking the partial trace of this density matrix, we obtain a reduced density matrix giving the statistics of the B particle alone. The description of the system as well as the operator equations for the density matrix and the reduced density matrix are discussed in Sec. II.

Since the Fokker-Planck equation is an inherently classical equation, it must in some sense be the classical limit of the equation satisfied by the density matrix. The authors of this paper have shown previously, in a different context,[6] that an appropriate, semiclassical, phase-space distribution function μ can be obtained from a density matrix $\mathfrak{M}$ by considering the diagonal matrix elements of $\mathfrak{M}$ between Gaussian wave packets. The classical phase-space coordinates appear as parameters in the wave packets. This transformation was originally introduced by Husimi,[7] and we have called it the Husimi transform. It has recently been studied by several authors in different contexts.[6,8–10] We shall use the Husimi transform in this paper to get the classical limit of the equation satisfied by the density matrix. In Sec. III and Appendix I we will discuss the Husimi transform, its relation to the Wigner transform, and the modifications necessary when considering systems with periodic boundary conditions.

In Sec. IV and Appendix II we will derive the semiclassical Liouville equation satisfied by the Husimi transform of the density matrix of the system.

In Sec. V and Appendix III we will derive the non-Markoffian equation satisfied by the Husimi transform of the reduced density matrix in the limit as $N\to\infty$, $\Omega\to\infty$, $N/\Omega=\rho=$constant. Just as in Ref. 4, in the limit as $t\to\infty$, this equation becomes a Markoffian equation for the steady-state distribution function.

Finally, in Sec. VI and Appendix IV we show that to lowest order in γ and h this equation is just the Fokker-Planck equation. This derivation yields the same expression, to lowest order in γ and h, for the friction coefficient as Refs. 2–4. Furthermore, we calculate the first-order correction in h to the Fokker-Planck equation. It is shown that for spatially homogeneous systems, the form of the equation is unchanged; only a correction of order h is made to the friction coefficient. This correction involves not only the interaction between the B particle and the fluid particle, but also the interaction between the fluid particles themselves.

We feel the results of this paper are of interest for several reasons. In the first place the paper provides an interesting application of the Husimi transform (continuous representation) technique. It also provides a quantum-mechanical derivation of Brownian motion which shows how friction can arise from a quantum-mechanical system in a perfectly natural way. Finally, the fact that to first order, quantum effects manifest themselves in Brownian motion only through a shift in the friction coefficient is of considerable interest, and this may have some bearing on the study of quantum fluids.

II. FORMULATION OF THE PROBLEM

We begin by considering a heavy, spinless particle of mass M and charge $\hat{q}$ interacting with N light, spinless particles of mass m ($m\ll M$) and zero charge. (These techniques can be extended to systems of particles of half-integer spin.) Let $\mathbf{X}$ be the coordinate of the B particle, and $\mathbf{x}_j$ the coordinate of the jth fluid particle. The whole system is enclosed in a box of side $2L$ and volume $\Omega=(2L)^3$, and we assume that periodic boundary conditions are satisfied at the boundaries of Ω. In the absence of any external field, the Hamiltonian is

$$H_0=\frac{1}{2M}\mathbf{P}^2+\frac{1}{2m}\sum_{j=1}^{N}\mathbf{p}_j^2 +\sum_{j<k}\sum\hat{\varphi}(\mathbf{x}_j-\mathbf{x}_k)+\sum_{j=1}^{N}\hat{\theta}(\mathbf{x}_j-\mathbf{X}). \quad (2.1)$$

Here $\hat{\varphi}$ is the potential between two fluid particles and $\hat{\theta}$ is the potential between a fluid particle and the B particle. We assume that $\hat{\theta}$ and $\hat{\varphi}$ have a range very short in comparison with the dimension L of the box. This will allow us to throw away boundary terms when we integrate by parts in subsequent calculations. We assume that all the forces involved are central forces, so that actually $\hat{\varphi}(r)=\hat{\varphi}(\|r\|)$ and $\hat{\theta}(r)=\hat{\theta}(\|r\|)$, where $\|r\|^2=r_1^2+r_2^2+r_3^2$. We further assume that the potential functions are smooth enough so that all subsequent manipulations are valid. Otherwise, the specific nature of the interactions is arbitrary.

In the presence of an external electric field $\mathbf{E}$, the total Hamiltonian is

$$H=H_0-\hat{q}\mathbf{E}\cdot\mathbf{X}. \quad (2.2)$$

We will assume that $\mathbf{E}$ is constant, although as shown in Ref. 4, it is a straightforward matter to extend the technique to include fields of the form $\mathbf{E}\cos(\omega\hat{t}+\alpha)$.

Let $\mathfrak{M}$ be the density matrix of this system, which we assume to be normalized so that $\mathrm{tr}(\mathfrak{M})=1$. The matrix element of $\mathfrak{M}$ in the x representation is the function $\mathfrak{M}(\hat{t};\mathbf{X},\mathbf{x}_1,\cdots,\mathbf{x}_N;\mathbf{X}',\mathbf{x}_1',\cdots,\mathbf{x}_N')$, which we will generally abbreviate by $\mathfrak{M}(t;\mathbf{X},\mathbf{x};\mathbf{X}',\mathbf{x}')$. This function must be periodic with period $2L$ in each of its spatial coordinates. Now $\mathfrak{M}$ satisfies the operator equation

$$ih(\partial\mathfrak{M}/\partial\hat{t})=[H,\mathfrak{M}], \quad (2.3)$$

[6] J. McKenna and H. L. Frisch, Ann. Phys. (N.Y.) **33**, 156 (1965).
[7] K. Husimi, Proc. Phys. Math. Soc. Japan **22**, 264 (1940).
[8] J. R. Klauder, J. Math. Phys. **4**, 1058 (1963); **5**, 177 (1964).
[9] J. McKenna and J. R. Klauder, J. Math. Phys. **5**, 878 (1964).
[10] C. L. Mehta and E. C. G. Sudarshan, Phys. Rev. **138**, B274 (1965).

which in the x representation can be written

$$i\hbar\frac{\partial\mathfrak{M}(\hat{t};\mathbf{X},\mathbf{x};\mathbf{X}',\mathbf{x}')}{\partial\hat{t}}=\Bigg[-\frac{\hbar^2}{2M}\left\{\frac{\partial^2}{\partial\mathbf{X}^2}-\frac{\partial^2}{\partial\mathbf{X}'^2}\right\}-\frac{\hbar^2}{2m}\sum_{j=1}\left\{\frac{\partial^2}{\partial\mathbf{x}_j^2}-\frac{\partial^2}{\partial\mathbf{x}_j'^2}\right\}$$
$$+\sum_{j<k}\sum\{\hat{\varphi}(\mathbf{x}_j-\mathbf{x}_k)-\hat{\varphi}(\mathbf{x}_j'-\mathbf{x}_k')\}+\sum_{j=1}^{N}\{\hat{\theta}(\mathbf{x}_j-\mathbf{X})-\hat{\theta}(\mathbf{x}_j'-\mathbf{x}')\}-\hat{q}(\mathbf{X}-\mathbf{X}')\cdot\mathbf{E}\Bigg]\mathfrak{M}. \quad (2.4)$$

The reduced density matrix $\mathfrak{F}$, describing the B particle alone, has matrix elements

$$\mathfrak{F}(\hat{t};\mathbf{X};\mathbf{X}')=\int_\Omega\mathfrak{M}(\hat{t};\mathbf{X},\mathbf{x};\mathbf{X}',\mathbf{x})\prod_{j=1}^{N}d\mathbf{x}_j, \quad (2.5)$$

where the range of integration for each variable $\mathbf{x}_j$ is the volume Ω. These matrix elements satisfy the equation

$$i\hbar\frac{\partial\mathfrak{F}}{\partial\hat{t}}=\left[-\frac{\hbar^2}{2M}\left(\frac{\partial^2}{\partial\mathbf{X}^2}-\frac{\partial^2}{\partial\mathbf{X}'^2}\right)-\hat{q}(\mathbf{X}-\mathbf{X}')\cdot\mathbf{E}\right]\mathfrak{F}+\int_\Omega\sum_{j=1}^{N}\{\hat{\theta}(\mathbf{x}_j-\mathbf{X})-\hat{\theta}(\mathbf{x}_j-\mathbf{X}')\}\mathfrak{M}(\hat{t};\mathbf{X},\mathbf{x};\mathbf{X}',\mathbf{x})\prod_{j=1}^{N}d\mathbf{x}_j. \quad (2.6)$$

III. CONNECTION TO CLASSICAL PHASE SPACE

In order to relate the quantum-mechanical description of our system to a description in classical phase space, we use the Husimi transform. The essential idea of this transform, first used by Husimi,[7] is to consider the expectation value of the density matrix in a state in which the position and momentum of particles can be determined simultaneously with maximum accuracy.

In order to illustrate its properties, consider first a system having one degree of freedom, defined on the whole real line, $-\infty<x<\infty$. Let the density matrix in the x representation be $\rho(x,x')$, which is assumed to be normalized so that $\mathrm{tr}(\rho)=1$. Then, as is well known, the minimum wave packet is of the form[11]

$$\psi(x)=\left(\frac{\lambda}{\pi\hbar}\right)^{1/4}\exp\left\{-\frac{\lambda}{2\hbar}(x-r)^2+i\frac{mv}{\hbar}x\right\}. \quad (3.1)$$

The expectation values of the position and momentum operators with respect to this wave function are $\langle x\rangle=r$ and $\langle p\rangle=mv$, and $\Delta x\Delta p=\hbar/2$.

The Husimi transform of ρ is then defined to be

$$h(r,v)=\left(\frac{m}{2\pi\hbar}\right)\int_{-\infty}^{\infty}\psi^*(x)\rho(x,x')\psi(x')dxdx'. \quad (3.2)$$

(For a system of particles of spin $\frac{1}{2}$, ψ would be a vector, ρ a 2×2 matrix, and h a 2×2 matrix.) In a slightly different context, some of the physical implications of this transform have been studied recently by Klauder,[8] its mathematical properties were investigated by McKenna and Klauder,[9] its applications in the quantum theory of optical coherence have been studied by Mehta and Sudarshan,[10] and it was employed by McKenna and Frisch[6] in an investigation of high-field magnetoresistance. We list here a few of its properties which show why it is a good, semiclassical, phase-space distribution function. Some of these statements are proved in Appendix I, and proofs of the remaining statements can be found in Refs. 9 and 10.

The function $h(r,v)$ is real, non-negative, bounded, and infinitely differentiable.[9] As will be shown in the course of this paper, in the limit as $\hbar\to0$, $h(r,v)$ will tend to a solution of the classical Liouville equation. Furthermore, the following statements can be made about various moments of $h(r,v)$:

$$\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}h(r,v)drdv=\mathrm{tr}(\rho)=1, \quad (3.3)$$

$$\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}2^{-n}\left(\frac{\hbar}{\lambda}\right)^{n/2}H_n\left(r\left(\frac{\lambda}{\hbar}\right)^{1/2}\right)h(r,v)drdv=\mathrm{tr}(r^n\rho), \quad (3.4)$$

$$\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}2^{-n}(\lambda\hbar)^{n/2}H_n\left(\frac{mv}{(\lambda\hbar)^{1/2}}\right)h(r,v)drdv=\mathrm{tr}(p^n\rho), \quad (3.5)$$

[11] L. I. Schiff, *Quantum Mechanics* (McGraw-Hill Book Company, Inc., New York, 1955), p. 56.

$$\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\left\{\left(\frac{\lambda}{2\hbar}\right)^{1/2}r+i(2\lambda\hbar)^{-1/2}mv\right\}^{\alpha}\left\{\left(\frac{\lambda}{2\hbar}\right)^{1/2}r-i(2\lambda\hbar)^{-1/2}mv\right\}^{\beta}h(r,v)drdv$$
$$=\mathrm{tr}\left(\left\{\left(\frac{\lambda}{2\hbar}\right)^{1/2}r+i(2\lambda\hbar)^{-1/2}p\right\}^{\alpha}\left\{\left(\frac{\lambda}{2\hbar}\right)^{1/2}r-i(2\lambda\hbar)^{-1/2}p\right\}^{\beta}\rho\right). \quad (3.6)$$

Equation (3.3) follows almost directly from definition (3.2); in Eqs. (3.4) and (3.5), which are proved in Appendix I, $H_n(x)$ is the nth Hermite polynomial; and Eq. (3.6) is proved in Ref. 10. In these equations r and p, when appearing under the trace sign, denote the position and momentum operators, respectively. Because of the relation (3.6), Mehta and Sudarshan[10] have called the Husimi transform of the density matrix the "antinormal ordered distribution function." It should be noted in connection with Eq. (3.4), that

$$2^{-n}\left(\frac{\hbar}{\lambda}\right)^{n/2}H_n\left(r\left(\frac{\lambda}{\hbar}\right)^{1/2}\right)=\sum_{\alpha=0}^{[n/2]}\frac{n!}{\alpha!(n-2\alpha)!}\left(-\frac{\hbar}{4\lambda}\right)^{\alpha}r^{n-2\alpha}.$$

Thus if we are only interested in a low-order expansion in $\hbar$, the moment equation simplifies. A similar remark applies to (3.5).

The following important inversion formula, which is discussed in Appendix I, holds:

$$\rho(s+t,\, s-t)=e^{(\lambda/\hbar)t^2}\int_{-\infty}^{\infty}e^{(2imv/\hbar)t}\times\exp\left\{-\frac{\hbar}{4\lambda}D_s^2\right\}h(s,v)dv\,, \quad (3.7)$$

where $D_s^2=d^2/ds^2$.

Finally, we note the relationship of the Husimi transform to the Wigner transform.[12] The Wigner transform of the density matrix is defined by

$$w(r,v)=\frac{1}{\pi\hbar}\int_{-\infty}^{\infty}\rho(r+t,\, r-t)e^{-(2imv/\hbar)t}dt\,, \quad (3.8)$$

and the following two relations are easily proved:

$$h(r,v)=\frac{m^2}{\pi\hbar}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}e^{-(\lambda/\hbar)(s-r)^2-(m^2/\lambda\hbar)(t-v)^2}\times w(s,t)dsdt\,, \quad (3.9)$$

$$w(r,v)=\frac{1}{m}\exp\left\{-\frac{\hbar}{4\lambda}D_r^2-\frac{\lambda\hbar}{4m^2}D_v^2\right\}h(r,v)\,. \quad (3.10)$$

We now consider the modifications of the above ideas which are required when we consider a system defined on the interval $-L\leq x\leq L$ and for which periodic boundary conditions are prescribed. For such a system, we note that $\rho(x,x')$ is periodic in each variable, with period $2L$. The prescription for taking the Husimi transform is somewhat ambiguous in this case, for no state $\varphi(x)$ exists satisfying both $\varphi(-L)=\varphi(L)$, and $\Delta x\Delta p=\hbar/2$. We have therefore arbitrarily chosen

$$\varphi(x)=\left(\frac{\lambda}{\pi\hbar}\right)^{1/4}\sum_{n=-\infty}^{\infty}e^{-(\lambda/2\hbar)(x+2nL-r)^2+i(mv/\hbar)(x+2nL)} \quad (3.11)$$

as the state to use in defining the Husimi transform. For this state, we have

$$\langle x\rangle=r+O(e^{-\alpha L^2})\,,\quad \langle p\rangle=mv+O(e^{-\alpha L^2})\,,\quad \Delta x\Delta p=\tfrac{1}{2}\hbar+O(e^{-\alpha L^2/2})\,, \quad (3.12)$$

where $\alpha>0$. Then, we define the Husimi transform of ρ to be

$$h(r,v)=\left(\frac{m}{2\pi\hbar}\right)\int_{-L}^{L}\varphi^*(x)\rho(x,x')\varphi(x')dxdx'. \quad (3.13)$$

However, if we make use of the periodicity properties of $\rho(x,x')$, it can easily be shown that (3.13) can be written as

$$h(r,v)=\left(\frac{m}{2\pi\hbar}\right)\left(\frac{\lambda}{\pi\hbar}\right)^{1/2}\int_{-\infty}^{\infty}e^{-(\lambda/2\hbar)(x-r)^2-i(mv/\hbar)x}\times\rho(x,x')e^{-(\lambda/2\hbar)(x'-r)^2+i(mv/\hbar)x'}dxdx$$
$$=\left(\frac{m}{2\pi\hbar}\right)\int_{-\infty}^{\infty}\psi^*(x)\rho(x,x')\psi(x')dxdx'. \quad (3.14)$$

In other words, definition (3.2) holds for periodic density matrices.

The Husimi transform of a periodic density matrix is again a real, non-negative, bounded, and infinitely differentiable function. In the limit as $\hbar\to 0$, it is a solution of the classical Liouville equation. It is periodic in r with period $2L$:

$$h(r+2L,\, v)=h(r,v)\,. \quad (3.15)$$

This follows directly from (3.14) on making use of the periodicity of $\rho(x,x')$. The analogs of (3.4) and (3.6) are no longer true, but the normalization condition holds,

$$\int_{-\infty}^{\infty}dv\int_{-L}^{L}drh(r,v)=\mathrm{tr}(\rho)=1\,, \quad (3.16)$$

[12] E. P. Wigner, Phys. Rev. **40**, 749 (1932).

and

$$\int_{-\infty}^{\infty} dv \int_{-L}^{L} dr 2^{-n}(\lambda\hbar)^{n/2} H_n\left(\frac{mv}{(\lambda\hbar)^{1/2}}\right) h(r,v) = \operatorname{tr}(p^n\rho). \tag{3.17}$$

[Equations (3.16)–(3.17) are discussed further in Appendix I.] The inversion formula (3.7) is still valid. Finally, if the Wigner transform is

$$w(x,v) = \frac{1}{\pi\hbar}\int_{-L}^{L} \rho(x+t,\, x-t) e^{-(2imv/\hbar)t} dt, \tag{3.18}$$

then

$$h(x,v) = \left(\frac{m}{2L}\right)\int_{-\infty}^{\infty} \sum_{-\infty}^{\infty} e^{-(\lambda/\hbar)(s-x)^2 - (m^2/\lambda\hbar)(v-n\pi\hbar/2mL)^2} \times w\left(s, \frac{\hbar}{2m}\frac{n\pi}{L}\right) ds. \tag{3.19}$$

IV. SEMICLASSICAL LIOUVILLE EQUATION

The results of Sec. III extend directly to our system of $3N+3$ degrees of freedom. We define the two wave functions

$$\begin{aligned}\psi(\mathbf{X},\mathbf{x}) &\equiv \psi(\mathbf{X},\mathbf{x}_1,\cdots,\mathbf{x}_N)\\ &= \left(\frac{\lambda}{\pi\hbar}\right)^{(3N+3)/4} \exp\left\{-\frac{\lambda}{2\hbar}[\|\mathbf{X}-\mathbf{R}\|^2 + \sum_{j=1}^{N}\|\mathbf{x}_j-\mathbf{r}_j\|^2] + i\left[\frac{M}{\hbar}\dot{\mathbf{R}}\cdot\mathbf{X} + \frac{m}{\hbar}\sum_{j=1}^{N}\dot{\mathbf{r}}_j\cdot\mathbf{x}_j\right]\right\},\end{aligned} \tag{4.1}$$

and

$$\xi(\mathbf{X}) = \left(\frac{\lambda}{\pi\hbar}\right)^{3/4} \exp\left\{-\frac{\lambda}{2\hbar}\|\mathbf{X}-\mathbf{R}\|^2 + i\frac{M}{\hbar}\dot{\mathbf{R}}\cdot\mathbf{X}\right\}. \tag{4.2}$$

In these expressions, $\mathbf{R}$ and $\dot{\mathbf{R}}$ are the classical position and velocity of the B particle, and $\mathbf{r}_j$ and $\dot{\mathbf{r}}_j$ are the classical coordinates of the jth fluid particle. Then the Husimi transforms of $\mathfrak{M}$ and $\mathfrak{F}$ are

$$\begin{aligned}&\hat{\mu}(\hat{t};\mathbf{R},\dot{\mathbf{R}};\mathbf{r},\dot{\mathbf{r}})\\ &\equiv \hat{\mu}(\hat{t};\mathbf{R},\dot{\mathbf{R}};\mathbf{r}_1,\cdots,\mathbf{r}_N,\dot{\mathbf{r}}_1,\cdots,\dot{\mathbf{r}}_N)\\ &= \left(\frac{m}{2\pi\hbar}\right)^{3N}\left(\frac{M}{2\pi\hbar}\right)^{3}\int_{-\infty}^{\infty}\psi^*(\mathbf{X},\mathbf{x})\mathfrak{M}(\hat{t};\mathbf{X},\mathbf{x};\mathbf{X}',\mathbf{x}') \times\psi(\mathbf{X}',\mathbf{x}')d\mathbf{X}d\mathbf{X}'\prod_{j=1}^{N}d\mathbf{x}_j\,d\mathbf{x}_j'\\ &\equiv \left(\frac{m}{2\pi\hbar}\right)^{3N}\left(\frac{M}{2\pi\hbar}\right)^{3}(\psi,\mathfrak{M}\psi),\end{aligned} \tag{4.3}$$

and

$$\hat{f}(\hat{t};\mathbf{R},\dot{\mathbf{R}}) = \left(\frac{M}{2\pi\hbar}\right)^{3}\int_{-\infty}^{\infty}\xi^*(\mathbf{X})\mathfrak{F}(\hat{t};\mathbf{X};\mathbf{X}')\xi(\mathbf{X}')d\mathbf{X}\,d\mathbf{X}'. \tag{4.4}$$

In addition to the properties of $\hat{\mu}$ and $\hat{f}$ which were discussed in Sec. III, it is easy to show that

$$\int \hat{\mu}\prod_{j=1}^{N} d\mathbf{r}_j d\dot{\mathbf{r}}_j = \hat{f}. \tag{4.5}$$

In Eq. (4.5), each $\dot{\mathbf{r}}_j$ integral is over all of R_3 while each $\mathbf{r}_j$ integral is over Ω.

There are two important dimensionless parameters in the problem. The first is the square root of the mass ratio, $\gamma=(m/M)^{1/2}$. This is the expansion parameter of Lebowitz, Rubin, Résibois, and Davis,[2–4] in terms of which they introduced the scaled velocity $\hat{\mathbf{U}}$ for the B particle,

$$\dot{\mathbf{R}} = \gamma\hat{\mathbf{U}}. \tag{4.6}$$

The velocity $\hat{\mathbf{U}}$ is now of the same order of magnitude as the fluid particle velocities $\dot{\mathbf{r}}_j$.

The second important dimensionless parameter is

$$a = \lambda\hbar/2mkT. \tag{4.7}$$

This parameter is a measure of the deviation between the classical and quantum descriptions of the system. A nice physical interpretation can be obtained by noting that

$$a = \{(N+\gamma^2)(\tfrac{3}{2}kT)\}^{-1}\left\{\left(\psi,\left[\frac{\mathbf{P}^2}{2M}+\sum_{j=1}^{N}\frac{\mathbf{p}_j^2}{2m}\right]\psi\right) - [\tfrac{1}{2}M\dot{\mathbf{R}}^2+\tfrac{1}{2}m\sum_{j=1}^{N}\dot{\mathbf{r}}_j^2]\right\}. \tag{4.8}$$

In other words, a is the ratio of the difference between the expectation value of the kinetic energy and the classical kinetic energy to the mean kinetic energy when equipartition applies.

Before proceeding, we introduce a system of units in which m/kT is unity. This will make it simple to introduce dimensionless expansion parameters. Thus we define new velocities $\mathbf{V}$ and $\mathbf{v}_j$, Husimi transforms μ and f, charge q, potential functions φ and θ, and time t in terms of the corresponding quantity in the old dimensions (represented by the corresponding letter with a caret) by means of the identities:

$$\begin{aligned}&\mathbf{V}=\left(\frac{m}{kT}\right)^{1/2}\dot{\mathbf{R}},\quad \mathbf{v}_j=\left(\frac{m}{kT}\right)^{1/2}\dot{\mathbf{r}}_j,\quad \mathbf{U}=\left(\frac{m}{kT}\right)^{1/2}\hat{\mathbf{U}},\\ &\varphi=\left(\frac{m}{kT}\right)\hat{\varphi},\quad \theta=\left(\frac{m}{kT}\right)\hat{\theta},\quad \mu=\gamma^3\left(\frac{kT}{m}\right)^{(3N+3)/2}\hat{\mu},\\ &q=\left(\frac{m}{kT}\right)\hat{q},\quad f=\gamma^3\left(\frac{kT}{m}\right)^{3/2}\hat{f},\quad t=\left(\frac{kT}{m}\right)^{1/2}\hat{t}.\end{aligned} \tag{4.9}$$

(Notice that both μ and f are normalized with respect to $\mathbf{U}$ and not with respect to $\mathbf{V}$.)

In terms of the new dimensions, we rewrite (4.3) as

$$\mu(t;\mathbf{R},\mathbf{U};\mathbf{r}_j,\mathbf{v}_j)=A(\psi,\mathfrak{M}\psi), \tag{4.10}$$

where

$$A=\gamma^{-3}(m/2\pi\hbar)^{3N+3}(kT/m)^{(3N+3)/2}. \tag{4.11}$$

The inverse of this transformation can be written

$$\mathfrak{M}=\mathfrak{M}(\mu). \tag{4.12}$$

These equations are written out explicitly in Appendix II.

Now take the transform of both sides of (2.4). The resulting equation for μ can be written

$$\frac{\partial\mu}{\partial t}+\mathcal{L}\mu+\gamma\frac{q}{m}\mathbf{E}\cdot\frac{\partial\mu}{\partial\mathbf{U}}=0, \tag{4.13}$$

where

$$\mathcal{L}\mu=\left[\sum_{j=1}^{N}\mathbf{v}_j\cdot\frac{\partial}{\partial\mathbf{r}_j}+a\sum_{j=1}^{N}\frac{\partial}{\partial\mathbf{v}_j}\cdot\frac{\partial}{\partial\mathbf{r}_j}+\gamma\mathbf{U}\cdot\frac{\partial}{\partial\mathbf{R}}+a\gamma^3\frac{\partial}{\partial\mathbf{R}}\cdot\frac{\partial}{\partial\mathbf{U}}\right]\mu+\Phi(\mu)+\Theta(\mu), \tag{4.14}$$

$$\Phi(\mu)=B(\psi,[\sum_{j<k}\varphi,\mathfrak{M}(\mu)]\psi), \tag{4.15}$$

$$\Theta(\mu)=B(\psi,[\sum_{j=1}^{N}\theta,\mathfrak{M}(\mu)]\psi), \tag{4.16}$$

and $B=(i\eta/2ma)A$, $\eta=\lambda/(mkT)^{1/2}$, and A is defined in (4.11). The differential operators in $\mathcal{L}$ arise from the kinetic energy terms in the Hamiltonian and from the term $q\mathbf{E}\cdot\mathbf{X}$ by a straightforward process of integration by parts. A detailed discussion of this calculation is given in Appendix C of Ref. 6. The linear, integro-differential operators Φ and Θ, which are written out explicitly in Appendix II, are complicated in appearance but lend themselves readily to expansions in powers of a and γ.

Equation (4.13) is the analog of the classical Liouville equation, and in fact, in the limit $\hbar\to 0$, $\mathcal{L}$ becomes the classical Liouville operator.

Then, either by integrating Eq. (4.13) over the phase-space variables of the fluid particles, or by taking the transform of Eq. (2.6) with the wave function $\xi(\mathbf{x})$, we get the equation involving f:

$$\frac{\partial f}{\partial t}+\gamma\left\{\mathbf{U}\cdot\frac{\partial f}{\partial\mathbf{R}}+\frac{q}{m}\mathbf{E}\cdot\frac{\partial f}{\partial\mathbf{U}}\right\}+\gamma^3a\frac{\partial}{\partial\mathbf{R}}\cdot\frac{\partial}{\partial\mathbf{U}}f+\int\Theta(\mu)\prod_{j=1}^{N}d\mathbf{r}_jd\mathbf{v}_j=0. \tag{4.17}$$

V. TRANSPORT EQUATION FOR f

Starting from Eqs. (4.13) and (4.17), we can obtain a transport equation for f. Our methods in this section parallel the methods used in Ref. 4. The first step is to linearize Eqs. (4.13) and (4.17) with respect to $\mathbf{E}$.

Toward that end, the equilibrium phase-space distribution function is introduced:

$$\mu^0(\mathbf{R},\mathbf{U};\mathbf{r},\mathbf{v})\equiv\mu^0(\mathbf{R},\mathbf{U};\mathbf{r}_1,\cdots,\mathbf{r}_N,\mathbf{v}_1,\cdots,\mathbf{v}_N)=(A/Z)(\psi,e^{-\beta H_0}\psi), \tag{5.1}$$

where H_0 is the equilibrium Hamiltonian (2.1), $Z=\mathrm{tr}(e^{-\beta H_0})$, and A is defined in (4.11). It will be assumed that μ^0 is known; actually we will explicitly calculate the first two terms of μ^0 in an expansion in powers of a. The function μ^0 satisfies Eq. (4.13) with $\mathbf{E}=0$, and it is periodic in each spatial variable with period $2L$. In Appendix III it is shown that

$$\mu^0(\mathbf{R},\mathbf{U};\mathbf{r},\mathbf{v})=\mu^0(0,\mathbf{U};\mathbf{r}_1-\mathbf{R},\cdots,\mathbf{r}_N-\mathbf{R},\mathbf{v}_1,\cdots,\mathbf{v}_N). \tag{5.2}$$

Upon integrating μ^0 with respect to the coordinates of the fluid particles, we obtain

$$f^0(\mathbf{U})=\int\mu^0(\mathbf{R},\mathbf{U};\mathbf{r},\mathbf{v})\prod_{j=1}^{N}d\mathbf{r}_jd\mathbf{v}_j, \tag{5.3}$$

which is a solution of (4.17) with $\mathbf{E}=0$. The fact that f^0 is a function of $\mathbf{U}$ alone is a consequence of (5.2) and the periodicity of μ^0.

We now write

$$\mu=\mu^0+\mu',\quad f=f^0+f', \tag{5.4}$$

where μ' and f' are assumed to be linear in E. Then

$$\frac{\partial\mu'}{\partial t}+\mathcal{L}\mu'=-\frac{\gamma q}{m}\mathbf{E}\cdot\frac{\partial\mu^0}{\partial\mathbf{U}}, \tag{5.5}$$

and

$$\frac{\partial f'}{\partial t}+\gamma\mathbf{U}\cdot\frac{\partial f'}{\partial\mathbf{R}}+\gamma^3a\frac{\partial}{\partial\mathbf{R}}\cdot\frac{\partial}{\partial\mathbf{U}}f'+\int\Theta(\mu')\prod_{j=1}^{N}d\mathbf{r}_jd\mathbf{v}_j=-\frac{\gamma q}{m}\mathbf{E}\cdot\frac{\partial f^0}{\partial\mathbf{U}}. \tag{5.6}$$

The next step in the procedure is to formally solve (5.5) and substitute the result in (5.6). In order to do this we follow Lebowitz and Résibois[4] and employ Zwanzig's projection operator.[13] Define

$$P(\mathbf{R},\mathbf{U};\mathbf{r},\mathbf{v})=\mu^0/f^0, \tag{5.7}$$

which can be interpreted as the conditional probability of finding the fluid particles at $\mathbf{r}_j$, $\mathbf{v}_j$, given that the B particle is at $\mathbf{R}$, $\mathbf{U}$. Then if $h(\mathbf{R},\mathbf{U};\mathbf{r},\mathbf{v})$ is any phase-

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

space function, the projection operator π is defined by

$$(\pi h)(\mathbf{R},\mathbf{U};\mathbf{r},\mathbf{v}) = P(\mathbf{R},\mathbf{U};\mathbf{r},\mathbf{v})\int h(\mathbf{R},\mathbf{U};\mathbf{r},\mathbf{v})\prod_{j=1}^{N} d\mathbf{r}_j d\mathbf{v}_j. \quad (5.8)$$

If we note the fact that $\pi\mu'=Pf'$, we can write $\mu'=(1-\pi)\mu'+Pf'$, and then rewrite (5.6) as

$$\frac{\partial f'}{\partial t}+\gamma\mathbf{U}\cdot\frac{\partial f'}{\partial \mathbf{R}}+a\gamma^3\frac{\partial}{\partial \mathbf{R}}\cdot\frac{\partial}{\partial \mathbf{U}}f'+\int \Theta(Pf')\prod_{j=1}^{N} d\mathbf{r}_j d\mathbf{v}_j + \int \Theta((1-\pi)\mu')\prod_{j=1}^{N} d\mathbf{r}_j d\mathbf{v}_j = -\frac{\gamma q}{m}\mathbf{E}\cdot\frac{\partial f^0}{\partial \mathbf{U}}. \quad (5.9)$$

By operating on Eq. (5.5) with $1-\pi$, we get the equation for $(1-\pi)\mu'$,

$$\frac{\partial}{\partial t}(1-\pi)\mu'+(1-\pi)\mathcal{L}(1-\pi)\mu' = -\frac{\gamma q}{m}(1-\pi)\mathbf{E}\cdot\frac{\partial \mu^0}{\partial \mathbf{U}}-(1-\pi)\mathcal{L}(Pf'). \quad (5.10)$$

This equation can be solved formally to give

$$(1-\pi)\mu' = -\int_0^t e^{-t'(1-\pi)\mathcal{L}}(1-\pi)\mathcal{L}Pf'(t-t')dt' - \frac{\gamma q}{m}\int_0^t e^{-t'(1-\pi)\mathcal{L}}(1-\pi)\mathbf{E}\cdot\frac{\partial \mu^0}{\partial \mathbf{U}}dt', \quad (5.11)$$

where we have imposed the initial condition

$$\mu'(0;\mathbf{R},\mathbf{U};\mathbf{r}_j,\mathbf{v}_j)=P(\mathbf{R},\mathbf{U};\mathbf{r}_j,\mathbf{v}_j)f'(0;\mathbf{R},\mathbf{U}). \quad (5.12)$$

Then if (5.11) is substituted into (5.9), the following equation for f' is obtained:

$$\frac{\partial f'}{\partial t}+\gamma\mathbf{U}\frac{\partial f'}{\partial \mathbf{R}}+\gamma^3 a\frac{\partial}{\partial \mathbf{R}}\cdot\frac{\partial}{\partial \mathbf{U}}f' + \Lambda(N,\gamma)f' - \int_0^t K(t',N,\gamma)f'(t-t')dt' = -\frac{\gamma q}{m}\mathbf{E}\cdot\frac{\partial \mu^0}{\partial \mathbf{U}}+\gamma\int_0^t L(t',N,\gamma)dt'. \quad (5.13)$$

In (5.13) the operators $\Lambda(N,\gamma)$ and $K(t',N,\gamma)$,and the function $L(t',N,\gamma)$ are defined by

$$\Lambda(N,\gamma)f' = \int \Theta(Pf')\prod_{j=1}^{N} d\mathbf{r}_j d\mathbf{v}_j, \quad (5.14)$$

$$K(t',N,\gamma)f'(t-t') = \int \Theta\left(e^{-t'(1-\pi)\mathcal{L}}(1-\pi)\mathcal{L}(Pf'(t-t'))\right)\prod_{j=1}^{N} d\mathbf{r}_j d\mathbf{v}_j, \quad (5.15)$$

$$L(t',N,\gamma) = \frac{q}{m}\int \Theta\left(e^{-t'(1-\pi)\mathcal{L}}(1-\pi)\mathbf{E}\cdot\frac{\partial \mu^0}{\partial \mathbf{U}}\right)\prod_{j=1}^{N} d\mathbf{r}_j d\mathbf{v}_j. \quad (5.16)$$

The dependence of these quantities on N and γ has been emphasized in the notation, and the f' in (5.13) should be written $f'(N;t;\mathbf{R},\mathbf{U})$. These quantities, while complicated in appearance, lend themselves to expansions in powers of γ and a, as we will show in Sec. VI.

We next want to make the passage to the limit $N\to\infty$, $\Omega\to\infty$, $N/\Omega=$constant. Before doing this, the function f' must be normalized so that the density of B particles remains constant in the limit. Since we are only going to consider a uniform system, we accomplish the renormalization by replacing f by Ωf and μ^0 by $\Omega\mu^0$. This is equivalent to multiplying Eq. (5.13) by Ω. We assume that this has been done, and retain the old notation.

We now make the "physical" assumption that f tends to a limit as $N\to\infty$, $N/\Omega=$constant:

$$\lim_{N\to\infty} f'(N;t;\mathbf{R},\mathbf{U})=f'(t,\mathbf{U}). \quad (5.17)$$

Because we are dealing with a uniform system, $f'(t,\mathbf{U})$ must be independent of $\mathbf{R}$. We further assume the existence of the limits

$$\lim_{N\to\infty} \Lambda(N,\gamma)f'(N;t;\mathbf{R},\mathbf{U})=\Lambda(\gamma)f'(t,\mathbf{U}), \quad (5.18)$$

$$\lim_{N\to\infty} K(t',N,\gamma)f'(N;t-t';\mathbf{R},\mathbf{U}) = K(t',\gamma)f'(t-t',\mathbf{U}), \quad (5.19)$$

$$\lim_{N\to\infty} L(t',N,\gamma)=L(t',\gamma). \quad (5.20)$$

In (5.18)–(5.20) the limit is taken after the integration over the coordinates of the fluid particles has been performed. The resulting equation,

$$\frac{\partial f'}{\partial t}+\Lambda(\gamma)f'-\int_0^t K(t',\gamma)f'(t-t')dt' = -\frac{\gamma q}{m}\mathbf{E}\cdot\frac{\partial f^0}{\partial \mathbf{U}}+\int_0^t L(t',\gamma)dt', \quad (5.21)$$

is the non-Markoffian, irreversible equation describing the time evolution of the semiclassical distribution function of the B particle. This corresponds to Eq. (2.31) of Ref. 4.

The final step in this section is to obtain the Markoffian equation satisfied by the steady-state, semiclassical distribution function of the B particle.[14] The

[14] For an additional discussion of the relation of the Markoffian to the non-Markoffian equation, see R. Zwanzig, J. Chem. Phys. 40, 2527 (1964).

steady-state distribution function $\bar{f}'$ is defined as the "Laplace average" of f',

$$\bar{f}'(\mathbf{U})=\lim_{t_0\to\infty}\frac{1}{t_0}\int_0^\infty e^{-t/t_0}f'(t,\mathbf{U})dt, \tag{5.22}$$

which we assume to exist. If we formally take the Laplace transform of Eq. (5.21), divide by t_0, and then let $t_0\to\infty$, we get

$$\Lambda(\gamma)\bar{f}'+\frac{\gamma q}{m}\mathbf{E}\cdot\frac{\partial f^0}{\partial\mathbf{U}}=\bar{K}(\gamma)\bar{f}'+\gamma\bar{L}(\gamma), \tag{5.23}$$

where we have assumed the existence of the limits

$$\bar{K}(\gamma)\bar{f}'=\lim_{t_0\to\infty}\frac{1}{t_0}\int_0^\infty e^{-t/t_0}K(t',\gamma)f'(t-t')dt', \tag{5.24}$$

$$\bar{L}(\gamma)=\lim_{t_0\to\infty}\int_0^\infty e^{-t/t_0}L(t,\gamma)dt. \tag{5.25}$$

VI. EXPANSION IN POWERS OF γ AND a

In this final section of the paper, we discuss the expansion of Eq. (5.23) in powers of a and γ. To get this expansion, we return to (5.13), obtain the desired expansion of $\Lambda(N,\gamma)$, $K(t',N,\gamma)$, and $L(t',N,\gamma)$ and then take the appropriate limits as $N\to\infty$ and $t\to\infty$.

The first step in this direction is to obtain the first few terms in an expansion in γ of Eq. (5.13), which is correct to *all* orders in h. We write

$$\Theta=\Theta_0+\gamma\Theta_1+\gamma^2\Theta_2+\cdots. \tag{6.1}$$

In Appendix II where explicit expressions for Θ_0, Θ_1, and Θ_2 are given, it is shown that

$$\int\Theta_0(\mu)\prod_{j=1}^N d\mathbf{r}_jd\mathbf{v}_j=\int\Theta_2(\mu)\prod_{j=1}^N d\mathbf{r}_jd\mathbf{v}_j=0. \tag{6.2}$$

It is also shown there that Φ is independent of γ and

$$\int\Phi(\mu)\prod_{j=1}^N d\mathbf{r}_jd\mathbf{v}_j=0. \tag{6.3}$$

We can now write $\mathfrak{L}=\mathfrak{L}_0+\gamma\mathfrak{L}_1+\cdots$, where

$$\begin{aligned}\mathfrak{L}_0&=\sum_{j=1}^N\mathbf{v}_j\cdot\frac{\partial}{\partial\mathbf{r}_j}+a\sum_{j=1}^N\frac{\partial}{\partial\mathbf{v}_j}\cdot\frac{\partial}{\partial\mathbf{r}_j}+\Phi+\Theta_0,\\ \mathfrak{L}_1&=\mathbf{U}\cdot\frac{\partial}{\partial\mathbf{R}}+\Theta_1.\end{aligned} \tag{6.4}$$

The operator π must also be expanded in powers of γ, since in general the function $P(\mathbf{R},\mathbf{U};\mathbf{r},\mathbf{v})$ depends on γ. (We conjecture that P is actually independent of γ, but we have been unable to prove this.) If we write $P=P_0+\gamma P_1+\cdots$, then we set $\pi=\pi_0+\gamma\pi_1+\cdots$, where

$$\pi_n(h)=P_n\int h\prod_{j=1}^N d\mathbf{r}_jd\mathbf{v}_j,\quad n=1,2,\cdots. \tag{6.5}$$

There is, of course, a corresponding expansion for μ^0 and f^0,

$$\mu^0=\mu_0{}^0+\gamma\mu_1{}^0+\cdots,\quad f^0=f_0{}^0+\gamma f_1{}^0+\cdots.$$

From (6.2), (6.3), (6.4), and (6.5), we deduce that

$$\pi_j\mathfrak{L}_0=0,\quad j=0,1,\cdots. \tag{6.6}$$

The next step is to expand $e^{-t'(1-\pi)\mathfrak{L}}$ in powers of γ. First note that because of (6.6), we have

$$(1-\pi)\mathfrak{L}=\mathfrak{L}_0+\gamma(1-\pi_0)\mathfrak{L}_1+O(\gamma^2).$$

Then $e^{-t'(1-\pi)\mathfrak{L}}|_{\gamma=0}=e^{-t'\mathfrak{L}_0}$, and a relatively straightforward calculation[15] yields

$$\begin{aligned}\frac{\partial}{\partial\gamma}e^{-t'(1-\pi)\mathfrak{L}}\Big|_{\gamma=0}&=e^{-t'\mathfrak{L}_0}\alpha(-t'(1-\pi_0)\mathfrak{L}_1,-t'\mathfrak{L}_0)\\ &=\alpha(-t'(1-\pi_0)\mathfrak{L}_1,t'\mathfrak{L}_0)e^{-t'\mathfrak{L}_0},\end{aligned} \tag{6.7}$$

where

$$\begin{aligned}\alpha(u,x)&=u+\frac{1}{2!}\{u,x\}+\frac{1}{3!}\{u,x^2\}+\cdots,\\ \{u,x\}&=[u,x],\quad \{u,x^n\}=[\{u,x^{n-1}\},x].\end{aligned} \tag{6.8}$$

Therefore, we have the expansion

$$e^{-t'(1-\pi)\mathfrak{L}}=\{1+\gamma\alpha(-t'(1-\pi_0)\mathfrak{L}_1,t'\mathfrak{L}_0)\}\times e^{-t'\mathfrak{L}_0}+O(\gamma^2). \tag{6.9}$$

We can now write down the expansions of L to order γ and K and Λ to order γ^2 [notice that because of (6.2) they all start with a term of order γ]:

$$L(t',N,\gamma)=\gamma L_1(t',N)+O(\gamma^2), \tag{6.10}$$

$$L_1(t',N,\gamma)=\frac{q}{m}\int\Theta_1\left(e^{-t'\mathfrak{L}_0}(1-\pi_0)\mathbf{E}\cdot\frac{\partial\mu_0{}^0}{\partial\mathbf{U}}\right)\prod_{j=1}^N d\mathbf{r}_jd\mathbf{v}_j, \tag{6.11}$$

$$\Lambda(N,\gamma)f'=\gamma\Lambda_1(N)f'+\gamma^2\Lambda_2(N)f'+O(\gamma^3), \tag{6.12}$$

$$\Lambda_1(N)f'=\int\Theta_1(P_0f')\prod_{j=1}^N d\mathbf{r}_jd\mathbf{v}_j,\quad \Lambda_2(N)f'=\int\Theta_1(P_1f')\prod_{j=1}^N d\mathbf{r}_jd\mathbf{v}_j, \tag{6.13}$$

[15] For the details of this calculation see Sec. II in G. H. Weiss and A. A. Maradudin, J. Math. Phys. 3, 771 (1962).

$$K(t',N,\gamma)f'(t-t')=\gamma K_1(t',N)f'(t-t')+\gamma^2K_2(t',N)f'(t-t')+O(\gamma^3)\,, \tag{6.14}$$

$$K_1(t',N)f'(t-t')=\int \Theta_1(e^{-t'\mathfrak{L}_0}\mathfrak{L}_0P_0f'(t-t'))\prod_{j=1}^{N} d\mathbf{r}_jd\mathbf{v}_j\,, \tag{6.15}$$

$$\begin{aligned}K_2(t',N)f'(t-t')=&\int \Theta_1(e^{-t'\mathfrak{L}_0}(1-\pi_0)\mathfrak{L}_1P_0f'(t-t'))\prod_{j=1}^{N} d\mathbf{r}_jd\mathbf{v}_j\\ &+\int \Theta_1(\alpha[-t'(1-\pi_0)\mathfrak{L}_1,\,t'\mathfrak{L}_0]e^{-t'\mathfrak{L}_0}\mathfrak{L}_0P_0f'(t-t'))\prod_{j=1}^{N} d\mathbf{r}_jd\mathbf{v}_j+\int \Theta_1(e^{-t'\mathfrak{L}_0}\mathfrak{L}_0P_1f'(t-t'))\prod_{j=1}^{N} d\mathbf{r}_jd\mathbf{v}_j\,.\end{aligned} \tag{6.16}$$

If now in (5.13) we replace Λ, K, and L by their expansions correct to order γ^2, and then repeat the limiting procedure outlined in the remainder of Sec. V, we get the equation

$$(\gamma\Lambda_1+\gamma^2\Lambda_2)\bar{f}'+\frac{\gamma q}{m}\mathbf{E}\cdot\left(\frac{\partial f_0{}^0}{\partial\mathbf{U}}+\gamma\frac{\partial f_1{}^0}{\partial\mathbf{U}}\right)=(\gamma\bar{K}_1+\gamma^2\bar{K}_2)\bar{f}'+\gamma^2\bar{L}_1\,. \tag{6.17}$$

The operators $\bar{K}_1$ and $\bar{K}_2$ are defined from $K_1(t',N)$ and $K_2(t',N)$ via Eqs. (5.19) and (5.24), $\bar{L}_1$ arises from $L_1(t',N)$ via Eqs. (5.20) and (5.25), and Λ_j is defined from $\Lambda_j(N)$ by (5.18). The inhomogeneous term γ^2L_1 arises from quantum fluctuations which tend to drive the system away from equilibrium. As we will soon see, the expression $(\gamma\bar{K}_1+\gamma^2\bar{K}_2)\bar{f}'$ is the source of the dissipation, and $(\gamma\Lambda_1+\gamma^2\Lambda_2)\bar{f}'$ can be thought of as arising from kinematics. In terms of the Husimi transform, Eq. (6.17) is the quantum-mechanical form of the Fokker-Planck equation, correct to all orders in $\hbar$. This will be seen clearly when we expand (6.17) in powers of a.

Following our prescription of Sec. III, (3.8)–(3.10), one can obtain a transport equation for the Wigner distribution function. One should note, however, that the $\hbar$ dependence of the Husimi transform and the Wigner transform may be quite different.

We turn now to the final step in this paper, the expansion of Eq. (6.17) in powers of a, correct to first order in a. The procedure is to expand $L_1(t',N)$, $\Lambda_1(N)$, $\Lambda_2(N)$, $K_1(t',N)$, and $K_2(t',N)$ in powers of a, and then apply the limiting procedures outlined in the latter half of Sec. V. These calculations, while relatively straightforward, are lengthy and tedious, and we will only discuss the results here. An outline of some of these calculations is given in Appendix IV.

There are several important features of the calculation. In the first step, the expansion of Θ_0, Θ_1, and Φ in powers of a, each term in the expansions is a differential operator. We write $\Theta_0=\Theta_{00}+a\Theta_1+\cdots$, $\Theta_1=\Theta_{10}+a\Theta_{11}+\cdots$, $\Phi=\Phi_0+a\Phi_1+\cdots$. These operators are relatively simple. We write out those of zeroth order in a, while those of first order in a are given in Appendix II.

$$\begin{aligned}\Theta_{00}&=-\frac{1}{m}\sum_{j=1}^{N}\frac{\partial\theta(\mathbf{r}_j-\mathbf{R})}{\partial\mathbf{r}_j}\cdot\frac{\partial}{\partial\mathbf{v}_j}\,,\\ \Theta_{10}&=\frac{1}{m}\sum_{j=1}^{N}\frac{\partial\theta(\mathbf{r}_j-\mathbf{R})}{\partial\mathbf{r}_j}\cdot\frac{\partial}{\partial\mathbf{U}}\,,\\ \Phi_0&=-\frac{1}{m}\sum_{j\neq k}\sum\frac{\partial\varphi(\mathbf{r}_j-\mathbf{r}_k)}{\partial\mathbf{r}_j}\cdot\frac{\partial}{\partial\mathbf{v}_j}\,.\end{aligned} \tag{6.18}$$

We remark at this point that it can be shown from the techniques used in Appendix II, that if Θ is expanded in powers of a alone, then $\Theta|_{a=0}=\Theta_{00}+\gamma\Theta_{10}$. From this result, and Eq. (4.14) defining $\mathfrak{L}$, we see that at $a=0$, $\mathfrak{L}$ is indeed the classical Liouville operator.

The next step involves an expansion of $\mu_0{}^0$ and $\mu_1{}^0$ in powers of a. We write $\mu_0{}^0=\mu_{00}{}^0+a\mu_{01}{}^0+\cdots$, $\mu_1{}^0=\mu_{10}{}^0+a\mu_{11}{}^0+\cdots$. In Appendix III, we show that

$$\mu_{00}{}^0(\mathbf{R},\mathbf{U};\mathbf{r},\mathbf{v})=\Omega\exp\{-h_0\}\bigg/\int\exp\{-h_0\}d\mathbf{R}d\mathbf{U}\prod_{j=1}^{N}d\mathbf{r}_jd\mathbf{v}_j\,, \tag{6.19}$$

where

$$h_0\equiv h_0(\mathbf{R},\mathbf{U};\mathbf{r},\mathbf{v})=\tfrac{1}{2}\mathbf{U}^2+\tfrac{1}{2}\sum_{j=1}^{N}\mathbf{v}_j{}^2+\frac{1}{m}\sum_{j<k}\sum\varphi(\mathbf{r}_j-\mathbf{r}_k)+\frac{1}{m}\sum_{j=1}^{N}\theta(\mathbf{r}_j-\mathbf{R})\,, \tag{6.20}$$

and

$$\mu_{01}{}^0=\frac{1}{2}\left[\sum_{j=1}^{N}\frac{\partial}{\partial\mathbf{v}_j}\cdot\frac{\partial}{\partial\mathbf{v}_j}+\frac{1}{\eta^2}\left(\frac{\partial}{\partial\mathbf{R}}\cdot\frac{\partial}{\partial\mathbf{R}}+\sum_{j=1}^{N}\frac{\partial}{\partial\mathbf{r}_j}\cdot\frac{\partial}{\partial\mathbf{r}_j}\right)\right]\mu_{00}{}^0\,. \tag{6.21}$$

Furthermore, $\mu_{10}{}^0=\mu_{11}{}^0\equiv 0$. From these results one readily obtains

$$f_{00}{}^0(U)=(2\pi)^{-3/2}e^{-\frac{1}{2}U^2},\quad f_{01}{}^0(U)=f_{10}{}^0(U)=f_{11}{}^0(U)\equiv 0; \tag{6.22}$$

$$P_{00}(\mathbf{R},\mathbf{r},\mathbf{v})=\mu_{00}{}^0/f_{00}{}^0,\quad P_{10}=P_{11}\equiv 0,\quad P_{01}(\mathbf{R},\mathbf{r},\mathbf{v})=\frac{1}{2}\left[\sum_{j=1}^{N}\frac{\partial}{\partial\mathbf{v}_j}\cdot\frac{\partial}{\partial\mathbf{v}_j}+\frac{1}{\eta^2}\left(\frac{\partial}{\partial\mathbf{R}}\cdot\frac{\partial}{\partial\mathbf{R}}+\sum_{j=1}^{N}\frac{\partial}{\partial\mathbf{r}_j}\cdot\frac{\partial}{\partial\mathbf{r}_j}\right)\right]P_{00}. \tag{6.23}$$

It should be noted that P_{00} and P_{01} are independent of $\mathbf{U}$.

With the aid of these results, $L_1(t',N)$, $\Lambda_j(N)f'$, and $K_j(t',N)f'(t-t')$, $j=1$, 2, can be expanded to first order in a, and the results are as follows: Six of the terms vanish:

$$\begin{gathered}L_{10}(t',N)=L_{11}(t',N)=0,\\ \Lambda_{10}(N)=\Lambda_{00}(N)=\Lambda_{21}(N)=0,\quad K_{10}(t',N)=0.\end{gathered} \tag{6.24}$$

Two terms have the following form:

$$\begin{aligned}\Lambda_{11}(N)f'&=\Lambda(N)_{\mu\nu}\frac{\partial^2 f'}{\partial R_\mu\partial U_\nu},\\ K_{11}(t',N)f'(t-t')&=K(t',N)_{\mu\nu}\frac{\partial^2 f'(t-t')}{\partial R_\mu\partial U_\nu}.\end{aligned} \tag{6.25}$$

In (6.25), R_μ and U_μ, $\mu=1$, 2, 3, are the components of $\mathbf{R}$ and $\mathbf{U}$, and the summation convention for repeated indices is used. The coefficients $\Lambda(N)_{\mu\nu}$ and $K(t',N)_{\mu\nu}$ can be calculated explicitly, but in the limit as $N\rightarrow\infty$, $f'(t)$ will be independent of $\mathbf{R}$, and so these terms will not appear in the final formulation.[16] The remaining two terms have the form

$$K_{20}(t',N)f'(t-t')=K_0(t',N)_{\mu\nu}\frac{\partial}{\partial U_\mu}\left(U_\nu+\frac{\partial}{\partial U_\nu}\right)f'(t-t'), \tag{6.26}$$

$$K_{21}(t',N)f'(t-t')=K_1(t',N)_{\mu\nu}\frac{\partial}{\partial U_\mu}\left(U_\nu+\frac{\partial}{\partial U_\nu}\right)f'(t-t')+K_2(t',N)_\mu\frac{\partial f'(t-t')}{\partial R_\mu}. \tag{6.27}$$

Again the term involving $K_2(t',N)_\mu$, which is actually an operator, will disappear from the final formulation,[16] and we will confine our attention to $K_0(t',N)_{\mu\nu}$ and $K_1(t',N)_{\mu\nu}$.

The first of these two terms is

$$K_0(t',N)_{\mu\nu}=\beta^2\int P_{00}F_\mu e^{-t'\mathfrak{L}_{00}}F_\nu\prod_{j=1}^{N}d\mathbf{r}_jd\mathbf{v}_j. \tag{6.28}$$

In (6.28)

$$F_\mu=-\frac{1}{m\beta}\frac{\partial}{\partial R_\mu}\sum_{j=1}^{N}\theta(\mathbf{r}_j-\mathbf{R}) \tag{6.29}$$

is the force exerted on the B particle by the N fluid particles. The operator $\mathfrak{L}_{00}$ is

$$\mathfrak{L}_{00}=\sum_{j=1}^{N}\mathbf{v}_j\cdot\frac{\partial}{\partial\mathbf{r}_j}-\frac{1}{m}\sum_{j=1}^{N}\left(\frac{\partial\theta(\mathbf{r}_j-\mathbf{R})}{\partial\mathbf{r}_j}+\sum_{k\neq j}\frac{\partial\varphi(\mathbf{r}_j-\mathbf{r}_k)}{\partial\mathbf{r}_j}\right)\cdot\frac{\partial}{\partial\mathbf{v}_j}, \tag{6.30}$$

which is just the Liouville operator for a system of N fluid particles interacting with a B particle held fixed at $\mathbf{R}$. The operator $e^{-t'\mathfrak{L}_{00}}$ is the time translation operator for such a system. As Lebowitz and Rubin[2] have pointed out, $e^{-t'\mathfrak{L}_{00}}F_\nu=F_\nu(-t')$, and the integral in (6.28) is just the correlation function, $\langle F_\mu(0)F_\nu(-t')\rangle$, of the force exerted on an infinitely heavy B particle by the fluid particles.

The second of these two terms, which is an even function of t', is

$$\begin{aligned}K_1(t',N)_{\mu\nu}&=\beta\int F_\mu e^{-t'\mathfrak{L}_{00}}\frac{\partial P_{01}}{\partial R_\nu}\prod_{j=1}^{N}d\mathbf{r}_jd\mathbf{v}_j\\ &+\frac{\beta^2}{\eta^2}\int\frac{\partial}{\partial\mathbf{R}}(F_\mu)\cdot\frac{\partial}{\partial\mathbf{R}}(e^{-t'\mathfrak{L}_{00}}P_{00}F_\nu)\prod_{j=1}^{N}d\mathbf{r}_jd\mathbf{v}_j\\ &+\beta^2\int F_\mu e^{-t'\mathfrak{L}_{00}}\{\alpha(-t'\mathfrak{L}_{01},-t'\mathfrak{L}_{00})P_{00}F_\nu\}\prod_{j=1}^{N}d\mathbf{r}_jd\mathbf{v}_j.\end{aligned} \tag{6.31}$$

Upon examination we found that unless an accidental cancellation occurs, this term does not vanish. These integrals can again be written as correlation functions of forces, internal energies, and stresses. This can be shown by expanding $\partial P_{01}/\partial R_\nu$, and when this is done, it will be seen that the first integral in (6.31) also involves the interaction between the fluid particles themselves.

If, finally, in (5.13) we replace Λ, K, and L by their expansions correct to order a and γ^2, and then repeat the limiting procedure outlined in the latter half of Sec. V, we get the equation

$$-\frac{\gamma q}{m}\mathbf{E}\cdot\mathbf{U}f_{00}{}^0=(\zeta_{\mu\nu}{}^{(0)}+a\zeta_{\mu\nu}{}^{(1)})\frac{\partial}{\partial U_\mu}\left(U_\nu+\frac{\partial}{\partial U_\nu}\right)f'. \tag{6.32}$$

[16] While the analytic expressions for these terms are omitted from the paper, they are available from the authors on request.

In (6.32)

$$\zeta_{\mu\nu}^{(0)}=\lim_{t_0\to\infty}\lim_{N\to\infty}\int_0^\infty dt\, e^{-t/t_0}K_0(t,N)_{\mu\nu}, \quad (6.33)$$

$$\zeta_{\mu\nu}^{(1)}=\lim_{t_0\to\infty}\lim_{N\to\infty}\int_0^\infty dt\, e^{-t/t_0}K_1(t,N)_{\mu\nu}. \quad (6.34)$$

In form, (6.32) is just the steady-state Fokker-Planck equation for a uniform fluid which has been linearized in **E**.

Upon substituting expression (6.28) for $K_0(t,N)_{\mu\nu}$ into (6.33), the result is just the expression obtained by Lebowitz and Résibois[2] for the friction coefficient. (For an exact comparision, one must express the result in terms of the standard physical units.) Of course, under our assumption of a uniform fluid, $\zeta_{\mu\nu}^{(0)}$ and $\zeta_{\mu\nu}^{(1)}$ must both be multiples of the identity matrix.

We can summarize these results by saying that Eq. (6.17) has as its classical limit the Fokker-Planck equation (6.32), and the limiting process yields expression (6.33) for the friction constant. Furthermore, the first-order correction in $\hbar$ to (6.17) is obtained simply by replacing $\zeta_{\mu\nu}^{(0)}$ by $\zeta_{\mu\nu}^{(0)}+a\zeta_{\mu\nu}^{(1)}$. One should note at this point that the magnitude of the parameter a determines whether the system actually behaves in a quasi-classical way. A certain degree of caution should be used in merely attempting to expand in powers of the dimensional parameter $\hbar$.

One interesting conclusion can be drawn. Since $\zeta_{\mu\nu}^{(0)}$ and $\zeta_{\mu\nu}^{(1)}$ depend on M in different fashions, the quantum effects on the friction constant might be observable experimentally. Thus one could expect a nonclassical isotope effect when one studies the mobility of a heavy molecule or ion moving through a quantum fluid at sufficiently low temperatures. Possibly the isotopes of a heavier inert gas ion (Ne, etc.) in liquid He may exhibit such a phenomenon. The nonclassical portion of the mobility reflects, as we saw, the dynamical properties of the quantum fluid and could thus be a probe, albeit as yet of questionable sensitivity, for studying quantum, and in particular superfluid, fluids.

APPENDIX I

In this Appendix we discuss several properties of the Husimi transform. First we outline the proof of Eqs. (3.4) and (3.5). If one substitutes expressions (3.1) and (3.2) into the left-hand side of (3.4), the v integration can be performed, and the result is

$$\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} 2^{-n}\left(\frac{\hbar}{\lambda}\right)^{n/2} H_n\left(r\left(\frac{\lambda}{\hbar}\right)^{1/2}\right)h(r,v)drdv$$
$$=\left(\frac{\lambda}{\pi\hbar}\right)^{1/2} 2^{-n}\left(\frac{\hbar}{\lambda}\right)^{n/2}\int_{-\infty}^{\infty}\rho(x,x)dx\int_{-\infty}^{\infty}H_n\left(r\left(\frac{\lambda}{\hbar}\right)^{1/2}\right)e^{-(\lambda/\hbar)(x-r)^2}dr. \quad \text{(A1.1)}$$

In the inner right-hand integral of (A1.1) make the change of variables $r=x+(\hbar/\lambda)^{1/2}u$. Now expand $H_n(u+(\lambda/\hbar)^{1/2}x)$ in a Taylor series,

$$H_n\left(u+\left(\frac{\lambda}{\hbar}\right)^{1/2}x\right)=\sum_{\alpha=0}^{n}\binom{n}{\alpha}\left(\frac{\lambda}{\hbar}\right)^{\alpha/2}(2x)^\alpha H_{n-\alpha}(u). \quad \text{(A1.2)}$$

In (A1.2), we have made use of the recursion formula[17] $H_n'(x)=2nH_{n-1}(x)$, and the fact that $H_n(x)$ is a polynomial of degree n. Thus the inner integral on the right of (A1.1) is

$$\int_{-\infty}^{\infty}H_n\left(r\left(\frac{\lambda}{\hbar}\right)^{1/2}\right)e^{-(\lambda/\hbar)(x-r)^2}dr=\left(\frac{\hbar}{\lambda}\right)^{1/2}\sum_{\alpha=0}^{n}\binom{n}{\alpha}\left(\frac{\lambda}{\hbar}\right)^{\alpha/2}(2x)^\alpha\int_{-\infty}^{\infty}H_{n-\alpha}(u)e^{-u^2}du=\left(\frac{\pi\hbar}{\lambda}\right)^{1/2}2^n\left(\frac{\lambda}{\hbar}\right)^{n/2}x^n, \quad \text{(A1.3)}$$

since $\int_{-\infty}^{\infty}H_{n-\alpha}(u)e^{-u^2}du=\pi^{1/2}\delta_{\alpha,n}$, $\alpha\leq n$. When (A1.3) is substituted into (A1.1), the desired result is obtained. The simplest way to prove (3.5) is to go over to the p representation and then repeat the above proof of (3.4).

We next discuss the inversion formula (3.7). To start with, we make the change of variables

$$s=\tfrac{1}{2}(x+x'),\quad t=\tfrac{1}{2}(x-x'), \quad \text{(A1.4)}$$

in the integral (3.2) defining $h(r,v)$. Then we can write

$$h(r,v)=\left(\frac{m}{\pi\hbar}\right)\left(\frac{\lambda}{\pi\hbar}\right)^{1/2}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}e^{-(\lambda/\hbar)(s-r)^2-(\lambda/\hbar)t^2-(2im/\hbar)vt}\rho(s+t,\,s-t)dsdt. \quad \text{(A1.5)}$$

[17] *Handbook of Mathematical Functions*, edited by M. Abramowitz and I. A. Stegun (U. S. Department of Commerce, National Bureau of Standards, Washington, D. C., 1964), Appl. Math. Ser. 55, p. 783.

If we take the Fourier transform of both sides of (A1.5) with respect to v, and multiply both sides of the resulting equation by $\exp\{(\lambda/\hbar)t^2\}$, there results

$$\left(\frac{\lambda}{\pi\hbar}\right)^{1/2}\int_{-\infty}^{\infty} e^{-(\lambda/\hbar)(s-r)^2}\rho(s+t,\, s-t)ds = e^{(\lambda/\hbar)t^2}\int_{-\infty}^{\infty} e^{(2im/\hbar)vt}h(r,v)dv. \tag{A1.6}$$

The left-hand side of this equation is just the Weierstrass transform of the function of s, $\rho(s+t,\, s-t)$. This transform has been extensively studied in the mathematical literature.[18] In particular, it is known that the operator $\exp[-(\hbar/4\lambda)(d^2/dr^2)]$ inverts this transformation under conditions sufficiently broad to include the case under consideration here. If this operator is applied to both sides of (A1.6), Eq. (3.7) results.

We now discuss some of the mechanics of the modifications involved when a periodic system is involved. In the first place, the function $\varphi(x)$, defined in (3.11), which is used to define the Husimi transform, is not normalized to 1 when considered as a function on $-L\leq x\leq L$. In fact, a straightforward calculation yields

$$N^2=\int_{-L}^{L}|\varphi(x)|^2dx=1+2\sum_{n=1}^{\infty} e^{-(\lambda/\hbar)L^2n^2}\cos\left(\frac{2mvL}{\hbar}n\right)=1+O(e^{-\lambda L^2/\hbar}). \tag{A1.7}$$

This shows that the normalization tends exponentially fast to 1 as $L\to\infty$.

The calculations involved in (3.12) are somewhat lengthy, and since they do not have a direct bearing on the main calculations of this paper, we omit them.

The normalization condition (3.16) is proved most easily by starting from Eq. (A1.6) with $t=0$. Integrating both sides of the resulting equation with respect to r from $-L$ to L gives

$$\int_{-L}^{L}dr\int_{-\infty}^{\infty}h(r,v)dv=\left(\frac{\lambda}{\pi\hbar}\right)^{1/2}\int_{-L}^{L}dr\int_{-\infty}^{\infty}e^{-(\lambda/\hbar)(s-r)^2}\rho(s,s)ds. \tag{A1.8}$$

The integral on the right of (A1.8) can be successively transformed as follows. First write the integral over s as

$$\int_{-\infty}^{\infty}(\cdots)ds=\sum_{n=-\infty}^{\infty}\int_{(2n-1)L}^{(2n+1)L}(\cdots)ds.$$

Then in the nth double integral, make the change of variables, $r=t+2nL$, $s=u+2nL$, and make use of the periodicity of $\rho(s,s)$. Upon summing on n, one obtains

$$\int_{-L}^{L}dr\int_{-\infty}^{\infty}h(r,v)dv=\left(\frac{\lambda}{\pi\hbar}\right)^{1/2}\int_{-L}^{L}\rho(u,u)du\int_{-\infty}^{\infty}e^{-(\lambda/\hbar)(t-u)^2}dt=\int_{-L}^{L}\rho(u,u)du. \tag{A1.9}$$

The proof of (3.17) can be outlined as follows: Since $\rho(x,x')$ is periodic in x and x' with period $2L$, it can be developed into a double Fourier series

$$\rho(x,x')=\sum_{\mu=-\infty}^{\infty}\sum_{\nu=-\infty}^{\infty}\rho_{\mu\nu}e^{(\pi i/L)(\mu x-\nu x')}. \tag{A1.10}$$

It is readily shown that

$$\mathrm{tr}(p^n\rho)=2L\sum_{\mu=-\infty}^{\infty}\left(\frac{\mu\pi\hbar}{L}\right)^n\rho_{\mu\mu}, \tag{A1.11}$$

and that

$$\int_{-L}^{L}h(r,v)dr=\left(\frac{2mL}{\pi\hbar}\right)\left(\frac{\pi\hbar}{\lambda}\right)^{1/2}\sum_{\mu=-\infty}^{\infty}\rho_{\mu\mu}e^{-(1/\lambda\hbar)(mv-\mu\pi\hbar/L)^2}. \tag{A1.12}$$

Upon multiplying both sides of (A1.12) by $2^{-n}(\lambda\hbar)^{n/2}H_n(mv/(\lambda\hbar)^{1/2})$ and integrating with respect to v, calculations similar to those leading to (A1.3) yield (A1.11) on the right-hand side, which is the desired result.

---

[18] I. I. Hirschman and D. V. Widder, *The Convolution Transform* (Princeton University Press, Princeton, New Jersey, 1955), Chap. VIII. This is a standard treatise which also contains a large bibliography.

APPENDIX II

In this Appendix, we first write out explicitly the relationship between the density matrix $\mathfrak{M}$ of the system of N fluid particles plus the B particle, and the Husimi transform of the density matrix. This is most conveniently done by introducing into the integral (4.9) the change of variables

$$\mathbf{S}=\tfrac{1}{2}(\mathbf{X}+\mathbf{X}'),\quad \mathbf{T}=\tfrac{1}{2}(\mathbf{X}-\mathbf{X}'),\quad \mathbf{s}_j=\tfrac{1}{2}(\mathbf{x}_j+\mathbf{x}_j'),\quad \mathbf{t}_j=\tfrac{1}{2}(\mathbf{x}_j-\mathbf{x}_j'). \tag{A2.1}$$

Then, Eq. (4.9) can be written out explicitly as

$$\mu(t;\mathbf{R},\mathbf{U};\mathbf{r}_j,\mathbf{v}_j)=\gamma^{-3}C\int\exp\left\{-\frac{\eta^2}{2a}[\|\mathbf{S}-\mathbf{R}\|^2+\|\mathbf{T}\|^2+\sum_{j=1}^{N}(\|\mathbf{s}_j-\mathbf{r}_j\|^2+\|\mathbf{t}_j\|^2)]-i\frac{\eta}{a}\left[\frac{1}{\gamma}\mathbf{U}\cdot\mathbf{T}+\sum_{j=1}^{N}\mathbf{v}_j\cdot\mathbf{t}_j\right]\right\}$$
$$\times\mathfrak{M}(t;\mathbf{S}+\mathbf{T},\mathbf{s}+\mathbf{t};\mathbf{S}-\mathbf{T},\mathbf{s}-\mathbf{t})d\mathbf{S}d\mathbf{T}\prod_{j=1}^{N}d\mathbf{s}_jd\mathbf{t}_j, \tag{A2.2}$$

where

$$C=\left(\frac{\eta^4}{(2\pi a)^3}\right)^{(3N+3)/2},\quad \eta=\lambda/(mkT)^{1/2}. \tag{A2.3}$$

In (A2.2) the domain of integration for each variable is all of R_3. This expression can be inverted, using the technique described in Appendix I, to give

$$\mathfrak{M}(t;\mathbf{S}+\mathbf{T},\mathbf{s}+\mathbf{t};\mathbf{S}-\mathbf{T},\mathbf{s}-\mathbf{t})=\exp\left\{\frac{\eta^2}{2a}[\|\mathbf{T}\|^2+\sum_{j=1}^{N}\|\mathbf{t}_j\|^2]\right\}\int\exp\left\{i\frac{\eta}{a}\left[\frac{1}{\gamma}\mathbf{U}\cdot\mathbf{T}+\sum_{j=1}^{N}\mathbf{v}_j\cdot\mathbf{t}_j\right]\right\}$$
$$\times\exp\left\{-\frac{a}{2\eta^2}\left(\frac{\partial}{\partial\mathbf{S}}\cdot\frac{\partial}{\partial\mathbf{S}}+\sum_{j=1}^{N}\frac{\partial}{\partial\mathbf{s}_j}\cdot\frac{\partial}{\partial\mathbf{s}_j}\right)\right\}\mu(t;\mathbf{S},\mathbf{U};\mathbf{s},\mathbf{v})d\mathbf{U}\prod_{j=1}^{N}d\mathbf{v}_j. \tag{A2.4}$$

Again the domain of integration for each variable is all of R_3.

With the aid of (A2.3) and (A2.4), expressions (4.15) and (4.16) for $\Phi(\mu)$ and $\Theta(\mu)$ can be easily written down explicitly:

$$\Theta(\mu)=\gamma^{-3}\left(\frac{i\eta}{2ma}\right)C\int\exp\left\{-\frac{\eta^2}{2a}[\|\mathbf{S}-\mathbf{R}\|^2+\sum_{j=1}^{N}\|\mathbf{s}_j-\mathbf{r}_j\|^2]-i\frac{\eta}{a}\left[\frac{1}{\gamma}\mathbf{U}\cdot\mathbf{T}+\sum_{j=1}^{N}\mathbf{v}_j\cdot\mathbf{t}_j\right]\right\}$$
$$\times\sum_{j=1}^{N}\{\theta(\mathbf{s}_j+\mathbf{t}_j-\mathbf{S}-\mathbf{T})-\theta(\mathbf{s}_j-\mathbf{t}_j-\mathbf{S}+\mathbf{T})\}d\mathbf{S}d\mathbf{T}\prod_{j=1}^{N}d\mathbf{s}_jd\mathbf{t}_j$$
$$\times\int\exp\left\{i\frac{\eta}{a}\left[\frac{1}{\gamma}\mathbf{U}'\cdot\mathbf{T}+\sum_{j=1}^{N}\mathbf{v}_j'\cdot\mathbf{t}_j\right]\right\}\exp\left\{-\frac{a}{2\eta^2}\left(\frac{\partial}{\partial\mathbf{S}}\cdot\frac{\partial}{\partial\mathbf{S}}+\sum_{j=1}^{N}\frac{\partial}{\partial\mathbf{s}_j}\cdot\frac{\partial}{\partial\mathbf{s}_j}\right)\right\}\mu(t;\mathbf{S},\mathbf{U}';\mathbf{s},\mathbf{v}')d\mathbf{U}'\prod_{j=1}^{N}d\mathbf{v}_j'. \tag{A2.5}$$

The expression for $\Phi(\mu)$ is the same as the expression for $\Theta(\mu)$ except that the factor

$$\sum_{j=1}^{N}\{\theta(\mathbf{s}_j+\mathbf{t}_j-\mathbf{S}-\mathbf{T})-\theta(\mathbf{s}_j-\mathbf{t}_j-\mathbf{S}+\mathbf{T})\}\equiv\Delta\theta(\mathbf{S},\mathbf{T},\mathbf{s},\mathbf{t}) \tag{A2.6}$$

is replaced by

$$\sum_{j<k}\sum\{\varphi(\mathbf{s}_j+\mathbf{t}_j-\mathbf{s}_k-\mathbf{t}_k)-\varphi(\mathbf{s}_j-\mathbf{t}_j-\mathbf{s}_k+\mathbf{t}_k)\}\equiv\Delta\varphi(\mathbf{s},\mathbf{t}). \tag{A2.7}$$

In order to expand $\Theta(\mu)$ in powers of γ, the first step is to make the change of variables $\mathbf{T}\rightarrow\gamma\mathbf{T}$ in the integral (A2.5). This has the effect of multiplying the whole expression by γ^3, eliminating γ from all the exponential factors, and replacing $\Delta\theta(\mathbf{S},\mathbf{T},\mathbf{s},\mathbf{t})$ by $\Delta\theta(\mathbf{S},\gamma\mathbf{T},\mathbf{s},\mathbf{t})$. From (A2.3) it follows that γ appears only in the term $\Delta\theta(\mathbf{S},\gamma\mathbf{T},\mathbf{s},\mathbf{t})$. The next step is to expand $\Delta\theta(\mathbf{S},\gamma\mathbf{T},\mathbf{s},\mathbf{t})$ in powers of γ:

$$\Delta\theta(\mathbf{S},\gamma\mathbf{T},\mathbf{s},\mathbf{t})=\Delta\theta(\mathbf{S},0,\mathbf{s},\mathbf{t})+\gamma\mathbf{T}\cdot\frac{\partial}{\partial\boldsymbol{\tau}}\Delta\theta(\mathbf{S},\boldsymbol{\tau},\mathbf{s},\mathbf{t})\bigg|_{\tau=0}+\frac{\gamma^2}{2!}\left(\mathbf{T}\cdot\frac{\partial}{\partial\boldsymbol{\tau}}\right)^2\theta(\mathbf{S},\boldsymbol{\tau},\mathbf{s},\mathbf{t})\bigg|_{\tau=0}+\cdots. \tag{A2.8}$$

When this is substituted into (A2.5), the desired expansion of the operator Θ results. The $\mathbf{T}$ integrations can be carried out explicitly, and we write out the first three terms in the expansion:

$$\Theta_0(\mu)=\left(\frac{2\pi a}{\eta}\right)^3 C\int \exp\left\{-\frac{\eta^2}{2a}[\|\mathbf{S}-\mathbf{R}\|^2+\sum_{j=1}^N\|\mathbf{s}_j-\mathbf{r}_j\|^2]-i\frac{\eta}{a}\sum_{j=1}^N \mathbf{v}_j\cdot\mathbf{t}_j\right\}\Delta\theta(\mathbf{S},0,\mathbf{s},\mathbf{t})d\mathbf{S}$$

$$\times\prod_{j=1}^N d\mathbf{s}_j d\mathbf{t}_j\int \exp\left\{i\frac{\eta}{a}\sum_{j=1}^N \mathbf{v}_j'\cdot\mathbf{t}_j\right\}\exp\left\{-\frac{a}{2\eta^2}\left(\frac{\partial}{\partial\mathbf{S}}\cdot\frac{\partial}{\partial\mathbf{S}}+\sum_{j=1}^N\frac{\partial}{\partial\mathbf{s}_j}\cdot\frac{\partial}{\partial\mathbf{s}_j}\right)\right\}\mu(t;\mathbf{S},\mathbf{U};\mathbf{s},\mathbf{v}')\prod_{j=1}^N d\mathbf{v}_j', \quad (A2.9)$$

$$\Theta_1(\mu)=\left(\frac{ia}{\eta}\right)\left(\frac{2\pi a}{\eta}\right)^3 C\int \exp\left\{-\frac{\eta^2}{2a}[\|\mathbf{S}-\mathbf{R}\|^2+\sum_{j=1}^N\|\mathbf{s}_j-\mathbf{r}_j\|^2]-i\frac{\eta}{a}\sum_{j=1}^N \mathbf{v}_j\cdot\mathbf{t}_j\right\}\frac{\partial}{\partial S_\alpha}\sum_{j=1}^N\{\theta(\mathbf{s}_j+\mathbf{t}_j-\mathbf{S})+\theta(\mathbf{s}_j-\mathbf{t}_j-\mathbf{S})\}d\mathbf{S}$$

$$\times\prod_{j=1}^N d\mathbf{s}_j d\mathbf{t}_j\int \exp\left\{i\frac{\eta}{a}\sum_{j=1}^N \mathbf{v}_j'\cdot\mathbf{t}_j\right\}\exp\left\{-\frac{a}{2\eta^2}\left(\frac{\partial}{\partial\mathbf{S}}\cdot\frac{\partial}{\partial\mathbf{S}}+\sum_{j=1}^N\frac{\partial}{\partial\mathbf{s}_j}\cdot\frac{\partial}{\partial\mathbf{s}_j}\right)\right\}\frac{\partial}{\partial U_\alpha}\mu(t;\mathbf{S},\mathbf{U};\mathbf{s},\mathbf{v}')\prod_{j=1}^N d\mathbf{v}_j', \quad (A2.10)$$

$$\Theta_2(\mu)=\left(\frac{ia}{\eta}\right)^2\left(\frac{2\pi a}{\eta}\right)^3 C\int \exp\left\{-\frac{\eta^2}{2a}[\|\mathbf{S}-\mathbf{R}\|^2+\sum_{j=1}^N\|\mathbf{s}_j-\mathbf{r}_j\|^2]-i\frac{\eta}{a}\sum_{j=1}^N \mathbf{v}_j\cdot\mathbf{t}_j\right\}\left\{\frac{\partial^2}{\partial S_\alpha\partial S_\beta}\Delta(\mathbf{S},0,\mathbf{s},\mathbf{t})\right\}d\mathbf{S}$$

$$\times\prod_{j=1}^N d\mathbf{s}_j d\mathbf{t}_j\int \exp\left\{i\frac{\eta}{a}\sum_{j=1}^N \mathbf{v}_j'\cdot\mathbf{t}_j\right\}\exp\left\{-\frac{a}{2\eta^2}\left(\frac{\partial}{\partial\mathbf{S}}\cdot\frac{\partial}{\partial\mathbf{S}}+\sum_{j=1}^N\frac{\partial}{\partial\mathbf{s}_j}\cdot\frac{\partial}{\partial\mathbf{s}_j}\right)\right\}\frac{\partial^2}{\partial U_\alpha\partial U_\beta}\mu(t;\mathbf{S},\mathbf{U};\mathbf{s},\mathbf{v}')\prod_{j=1}^N d\mathbf{v}_j'. \quad (A2.11)$$

In (A2.10) and (A2.11) the summation convention for repeated indices is employed.

If one now examines

$$\int\Theta_0(\mu)\prod_{j=1}^N d\mathbf{v}_j \quad\text{and}\quad \int\Theta_2(\mu)\prod_{j=1}^N d\mathbf{v}_j,$$

with the aid of Eqs. (A2.9) and (A2.11), it is easy to see that these two integrals must vanish.

Since the expression for $\Phi(\mu)$ differs from the expression for $\Theta(\mu)$ only in that $\Delta\theta(\mathbf{S},\mathbf{T},\mathbf{s},\mathbf{t})$ is replaced by $\Delta\varphi(\mathbf{s},\mathbf{t})$, when the change of variables $\mathbf{T}\rightarrow\gamma\mathbf{T}$ is made in the integral defining $\Phi(\mu)$, γ will disappear from the resulting expression. Therefore $\Phi(\mu)$ is independent of γ.

We finally turn to the expansion of Θ_0, Θ_1, and Φ in powers of a. Consider $\Theta_0(\mu)$ as an example. In the integral in (A2.9), make the change of variables

$$\mathbf{S}\rightarrow\mathbf{R}+(2a/\eta^2)^{1/2}\mathbf{S},\quad \mathbf{s}_j\rightarrow\mathbf{r}_j+(2a/\eta^2)^{1/2}\mathbf{s}_j,\quad \mathbf{t}_j\rightarrow(a/\eta)\mathbf{t}_j.$$

After this change of variables, we have

$$\Theta_0(\mu)=\left(\frac{i\eta}{2ma}\right)\left(\frac{1}{4\pi^3}\right)^{(3N+3)/2}\int \exp\{-\|\mathbf{S}\|^2-\sum_{j=1}^N\|\mathbf{s}_j\|^2-i\sum_{j=1}^N\mathbf{v}_j\cdot\mathbf{t}_j\}$$

$$\times\sum_{j=1}^N\left\{\theta\left(\mathbf{r}_j-\mathbf{R}+\left(\frac{2a}{\eta^2}\right)^{1/2}(\mathbf{s}_j-\mathbf{S})+\frac{a}{\eta}\mathbf{t}_j\right)-\theta\left(\mathbf{r}_j-\mathbf{R}+\left(\frac{2a}{\eta^2}\right)^{1/2}(\mathbf{s}_j-\mathbf{S})-\frac{a}{\eta}\mathbf{t}_j\right)\right\}d\mathbf{S}\prod_{j=1}^N d\mathbf{s}_j d\mathbf{t}_j\int\exp\{i\sum_{j=1}^N\mathbf{v}_j'\cdot\mathbf{t}_j\}$$

$$\times\exp\left\{-\frac{a}{2\eta^2}\left(\frac{\partial}{\partial\mathbf{R}}\cdot\frac{\partial}{\partial\mathbf{R}}+\sum_{j=1}^N\frac{\partial}{\partial\mathbf{r}_j}\cdot\frac{\partial}{\partial\mathbf{r}_j}\right)\right\}\mu\left(t;\mathbf{R}+\left(\frac{2a}{\eta^2}\right)^{1/2}\mathbf{S},\mathbf{U};\mathbf{r}_j+\left(\frac{2a}{\eta^2}\right)^{1/2}\mathbf{s}_j,\mathbf{v}_j'\right)\prod_{j=1}^N d\mathbf{v}_j'. \quad (A2.12)$$

Now, each term in the integrand can be expanded in powers of $\sqrt{a}$. When this is done, the resulting integrals can be easily evaluated, and the coefficients of half-odd integer powers of a will vanish, as they should. This same technique can be applied to Θ_1 and Φ. We have already written out Θ_{00}, Θ_{10}, and Φ_0 in Sec. VI. The remaining three terms of interest are

$$\Theta_{01}(\mu)=-\frac{1}{m\eta^2}\sum_{j=1}^N\frac{\partial}{\partial r_{j\alpha}}\left(\frac{\partial^2\theta(\mathbf{r}_j-\mathbf{R})}{\partial r_{j\alpha}\partial r_{j\beta}}\frac{\partial\mu}{\partial v_{j\beta}}\right)+\frac{1}{m\eta^2}\sum_{j=1}^N\frac{\partial^2\theta(\mathbf{r}_j-\mathbf{R})}{\partial r_{j\alpha}\partial r_{j\beta}}\frac{\partial^2\mu}{\partial R_\alpha\partial v_{j\beta}}, \quad (A2.13)$$

$$\Theta_{11}(\mu)=\frac{1}{m\eta^2}\sum_{j=1}^{N}\frac{\partial}{\partial r_{j\alpha}}\left(\frac{\partial^2\theta(\mathbf{r}_j-\mathbf{R})}{\partial r_{j\alpha}\partial r_{j\beta}}\frac{\partial\mu}{\partial U_\beta}\right)-\frac{1}{m\eta^2}\sum_{j=1}^{N}\frac{\partial^2\theta(\mathbf{r}_j-\mathbf{R})}{\partial r_{j\alpha}\partial r_{j\beta}}\frac{\partial^2\mu}{\partial R_\alpha\partial U_\beta}, \tag{A2.14}$$

$$\Phi_1(\mu)=-\frac{1}{m\eta^2}\sum_{j\neq k}\sum\frac{\partial}{\partial r_{j\alpha}}\left(\frac{\partial^2\varphi(\mathbf{r}_j-\mathbf{r}_k)}{\partial r_{j\alpha}\partial r_{j\beta}}\frac{\partial\mu}{\partial v_{j\beta}}\right)-\frac{1}{m\eta^2}\sum_{j\neq k}\sum\frac{\partial^2\varphi(\mathbf{r}_j-\mathbf{r}_k)}{\partial r_{j\alpha}\partial r_{j\beta}}\frac{\partial^2\mu}{\partial r_{k\alpha}\partial v_{j\beta}}. \tag{A2.15}$$

In Eqs. (A2.13) through (A2.15), $r_{j\alpha}$, $v_{j\alpha}$, $j=1, 2, \cdots, N$, $\alpha=1, 2, 3$, denote the αth coordinates of the vectors $\mathbf{r}_j$ and $\mathbf{v}_j$, respectively; U_α is the αth coordinate of $\mathbf{U}$; and the summation convention for repeated indices is employed.

APPENDIX III

In this Appendix, the properties of the equilibrium phase-space distribution function $\mu^0(\mathbf{R},\mathbf{U};\mathbf{r},\mathbf{v})$ are studied in more detail. We first outline the proof of Eq. (5.2). This statement rests on the fact that the free Hamiltonian H_0 defined in (2.1) is invariant under the simultaneous translation $\mathbf{X}\rightarrow\mathbf{X}+\boldsymbol{\varrho}$, $\mathbf{x}_j\rightarrow\mathbf{x}_j+\boldsymbol{\varrho}$, $j=1, 2, \cdots, N$, for any fixed vector $\boldsymbol{\varrho}$. With the aid of this fact, it is easy to prove that the matrix element of $e^{-\beta H_0}$ in the X representation must satisfy

$$\langle\mathbf{X},\mathbf{x}_1,\cdots,\mathbf{x}_N|e^{-\beta H_0}|\mathbf{X}',\mathbf{x}_1',\cdots,\mathbf{x}_N'\rangle=\langle\mathbf{X}+\boldsymbol{\varrho},\mathbf{x}_1+\boldsymbol{\varrho},\cdots,\mathbf{x}_N+\boldsymbol{\varrho}|e^{-\beta H_0}|\mathbf{X}'+\boldsymbol{\varrho},\mathbf{x}_1'+\boldsymbol{\varrho},\cdots,\mathbf{x}_N'+\boldsymbol{\varrho}\rangle. \tag{A3.1}$$

If we now define $\mu^0(\mathbf{R},\mathbf{U};\mathbf{r},\mathbf{v})$ by replacing in Eq. (A2.2) the matrix element of $\mathfrak{M}$ by the matrix element of $Z^{-1}e^{-\beta H_0}$, and make use of Eq. (A3.1), then it is simple to show that

$$\mu^0(\mathbf{R}+\boldsymbol{\varrho},\mathbf{U};\mathbf{r}_1+\boldsymbol{\varrho},\cdots,\mathbf{r}_N+\boldsymbol{\varrho},\mathbf{v}_1,\cdots,\mathbf{v}_N)=\mu^0(\mathbf{R},\mathbf{U};\mathbf{r}_1,\cdots,\mathbf{r}_N,\mathbf{v}_1,\cdots,\mathbf{v}_N). \tag{A3.2}$$

Equation (5.2) follows directly from (A3.2) by setting $\boldsymbol{\varrho}=-\mathbf{R}$.

We now indicate the steps by which one calculates the first few terms in the expansion of μ^0, in powers of a. We do this using the standard physical units, and at the end of the calculation transform to units in which $m/kT=1$. The starting point is the relation

$$(\psi,e^{-\beta H_0}\psi)=\sum_{n=0}^{\infty}\frac{(-\beta)^n}{n!}(\psi,H_0^n\psi), \tag{A3.3}$$

where H_0 is defined in (2.1) and ψ is defined in (4.1). Since ψ is an exponential function, we can write

$$H_0^n\psi(\mathbf{X},\mathbf{x})=h_n(\mathbf{X},\mathbf{x})\psi(\mathbf{X},\mathbf{x}),$$

where $h_n(\mathbf{X},\mathbf{x})$ is a function of the variables $\mathbf{X}$, $\mathbf{x}_1$, $\cdots$, $\mathbf{x}_N$. A trivial calculation yields

$$h_1(\mathbf{X},\mathbf{x})=-\frac{1}{2M}\|-\lambda(\mathbf{X}-\mathbf{R})+iM\dot{\mathbf{R}}\|^2-\frac{1}{2m}\sum_{j=1}^{N}\|-\lambda(\mathbf{x}_j-\mathbf{r}_j)+im\dot{\mathbf{r}}_j\|^2$$
$$+\frac{3\lambda h}{2M}+\frac{3N\lambda h}{2m}+\sum_{j<k}\sum\hat{\varphi}(\mathbf{x}_j-\mathbf{x}_k)+\sum_{j=1}^{N}\hat{\theta}(\mathbf{x}_j-\mathbf{X}), \tag{A3.4}$$

and by noting that $h_n\psi=H_0(h_{n-1}\psi)$, we obtain the recursion relation

$$h_{n+1}=\left[-\frac{h^2}{2M}\frac{\partial}{\partial\mathbf{X}}\cdot\frac{\partial}{\partial\mathbf{X}}-\frac{h^2}{2m}\sum_{j=1}^{N}\frac{\partial}{\partial\mathbf{x}_j}\cdot\frac{\partial}{\partial\mathbf{x}_j}-\frac{h}{M}\{-\lambda(\mathbf{X}-\mathbf{R})+iM\dot{\mathbf{R}}\}\cdot\frac{\partial}{\partial\mathbf{X}}-\frac{h}{m}\sum_{j=1}^{N}\{-\lambda(\mathbf{x}_j-\mathbf{r}_j)+im\dot{\mathbf{r}}_j\}\cdot\frac{\partial}{\partial\mathbf{x}_j}+h_1\right]h_n. \tag{A3.5}$$

If we expand h_n in powers of h,

$$h_n=h_{n,0}+hh_{n,1}+h^2h_{n,2}+\cdots,$$

substitute this expansion into (A3.5), and equate like powers of h, we get a series of recursion relations which can be solved successively. The recursion relation for $h_{n,0}$ is just $h_{n,0}=h_{1,0}h_{n-1,0}$, which has the solution

$$h_{n,0}=(h_{1,0})^n. \tag{A3.6}$$

The recursion relation for $h_{n,1}$ is

$$h_{n,1}=h_{1,0}h_{n-1,1}+\left(\frac{3\lambda}{2M}+\frac{3N\lambda}{2m}\right)\{h_{1,0}\}^{n-1}+(n-1)f\{h_{1,0}\}^{n-2}, \tag{A3.7}$$

where

$$f\equiv f(\mathbf{X},\mathbf{x}_1,\cdots,\mathbf{x}_N)=\left\{\frac{\lambda}{M}(\mathbf{X}-\mathbf{R})-i\dot{\mathbf{R}}\right\}\cdot\left\{-\frac{\lambda}{M}(\mathbf{X}-\mathbf{R})+i\lambda\dot{\mathbf{R}}+\sum_{j=1}^{N}\frac{\partial}{\partial\mathbf{X}}\theta(\mathbf{x}_j-\mathbf{X})\right\}$$
$$+\sum_{j=1}^{N}\left[\left\{\frac{\lambda}{m}(\mathbf{x}_j-\mathbf{r}_j)-i\dot{\mathbf{r}}_j\right\}\cdot\left\{-\frac{\lambda}{m}(\mathbf{x}_j-\mathbf{r}_j)+i\lambda\dot{\mathbf{r}}_j+\frac{\partial}{\partial\mathbf{x}_j}\theta(\mathbf{x}_j-\mathbf{X})+\sum_{k\neq j}\frac{\partial}{\partial\mathbf{x}_j}\varphi(\mathbf{x}_j-\mathbf{x}_k)\right\}\right].\qquad\text{(A3.8)}$$

We now define the generating function

$$F(p)=\sum_{n=1}^{\infty}\frac{p^n}{n!}h_{n,1}.\qquad\text{(A3.9)}$$

If Eq. (A3.7) is multiplied by $p^n/n!$ and summed on n, it is not difficult to show that $F(p)$ satisfies the differential equation

$$\frac{dF}{dp}-h_{1,0}F=\left\{pf+\frac{3\lambda}{2M}+\frac{3N\lambda}{2m}\right\}e^{ph_{1,0}}.\qquad\text{(A3.10)}$$

Since we know that $F(0)=0$, the solution of (A3.10) is

$$F(p)=\left[\tfrac{1}{2}p^2f+\left(\frac{3\lambda}{2M}+\frac{3N\lambda}{2m}\right)p\right]e^{ph_{1,0}}.\qquad\text{(A3.11)}$$

We now notice that we can write

$$(\psi,H_0{}^n\psi)=\left(\frac{\lambda}{\pi\hbar}\right)^{(3N+3)/2}\int\exp\left\{-\frac{\lambda}{\hbar}\left[\|\mathbf{X}-\mathbf{R}\|^2+\sum_{j=1}^{N}\|\mathbf{x}_j-\mathbf{r}_j\|^2\right]\right\}h_n(\mathbf{X},\mathbf{x})d\mathbf{X}\prod_{j=1}^{N}d\mathbf{x}_j.\qquad\text{(A3.12)}$$

If in (A3.12) we make the coordinate transformation $\mathbf{X}\rightarrow\mathbf{R}+(\hbar/\lambda)^{1/2}\mathbf{X}$, $\mathbf{x}_j\rightarrow\mathbf{r}_j+(\hbar/\lambda)^{1/2}\mathbf{x}_j$, $j=1,2,\cdots,N$, replace h_n by its expansion in powers of $\hbar$, and then expand each coefficient $h_{n,j}(\mathbf{R}+(\hbar/\lambda)^{1/2}\mathbf{X},\mathbf{r}+(\hbar/\lambda)^{1/2}\mathbf{x})$ as a Taylor series in $\sqrt{\hbar}$, the resulting integrals can all be easily evaluated. All the terms involving half-odd integral powers of $\hbar$ vanish, and we have

$$(\psi,H_0{}^n\psi)=h_{n,0}(\mathbf{R},\mathbf{r})+\hbar\left[\frac{1}{4\lambda}\left(\frac{\partial}{\partial\mathbf{X}}\cdot\frac{\partial}{\partial\mathbf{X}}+\sum_{j=1}^{N}\frac{\partial}{\partial\mathbf{x}_j}\cdot\frac{\partial}{\partial\mathbf{x}_j}\right)h_{n,0}(\mathbf{X},\mathbf{x})+h_{n,1}(\mathbf{X},\mathbf{x})\right]\Bigg|_{(\mathbf{X}=\mathbf{R},\mathbf{x}=\mathbf{r})}+O(\hbar^2).\qquad\text{(A3.13)}$$

If we multiply (A3.13) by $(-\beta)^n/n!$ and sum on n, we then get

$$(\psi,e^{-\beta H_0}\psi)\,|_{\hbar=0}=e^{-\beta h_{1,0}(\mathbf{R},\mathbf{r})},$$
$$\frac{\partial}{\partial\hbar}(\psi,e^{-\beta H_0}\psi)\bigg|_{\hbar=0}=\left[\frac{1}{4\lambda}\left(\frac{\partial}{\partial\mathbf{X}}\cdot\frac{\partial}{\partial\mathbf{X}}+\sum_{j=1}^{N}\frac{\partial}{\partial\mathbf{x}_j}\cdot\frac{\partial}{\partial\mathbf{x}_j}\right)\exp[-\beta h_{1,0}(\mathbf{X},\mathbf{x})]+F(-\beta)\right]_{(\mathbf{X}=\mathbf{R},\mathbf{x}=\mathbf{r})}.\qquad\text{(A3.14)}$$

The expression on the right-hand side of (A3.14) is easily evaluated and can be written

$$\frac{\partial}{\partial\hbar}(\psi,e^{-\beta H_0}\psi)\bigg|_{\hbar=0}=\left[\frac{\lambda}{4M^2}\frac{\partial}{\partial\dot{\mathbf{R}}}\cdot\frac{\partial}{\partial\dot{\mathbf{R}}}+\frac{\lambda}{4m^2}\sum_{j=1}^{N}\frac{\partial}{\partial\dot{\mathbf{r}}_j}\cdot\frac{\partial}{\partial\dot{\mathbf{r}}_j}+\frac{1}{4\lambda}\left(\frac{\partial}{\partial\mathbf{R}}\cdot\frac{\partial}{\partial\mathbf{R}}+\sum_{j=1}^{N}\frac{\partial}{\partial\mathbf{r}_j}\cdot\frac{\partial}{\partial\mathbf{r}_j}\right)\right]e^{-\beta h_{1,0}(\mathbf{R},\mathbf{r})}.\qquad\text{(A3.15)}$$

The final step in determining the expansion of μ^0 is to note that

$$\int(\psi,e^{-\beta H_0}\psi)d\dot{\mathbf{R}}d\mathbf{R}\prod_{j=1}^{N}d\dot{\mathbf{r}}_jd\mathbf{r}_j=\left(\frac{2\pi\hbar}{m}\right)^{3N}\left(\frac{2\pi\hbar}{M}\right)^{3}Z.\qquad\text{(A3.16)}$$

Upon transforming to units in which $m/kT=1$ [defined in (4.5)], the derivation of Eqs. (6.19)–(6.21) is direct, and we omit the details. One point is worthy of comment, however. Because of the form of (A3.15), Z/A, when expanded in powers of a, has no term of first order.[19]

[19] This is a well-known result; cf., L. D. Landau and E. M. Lifschitz, *Statistical Physics* (Addison-Wesley Publishing Company, Reading, Pennsylvania, 1958), p. 99.

APPENDIX IV

In this Appendix, we outline the calculation of L_{10}, Λ_{10}, Λ_{20}, K_{10}, and K_{20}. This will demonstrate the technique involved in these calculations. The calculations of L_{11}, Λ_{11}, Λ_{21}, K_{11}, and K_{21} are longer, but involve nothing new, and will not be given here.[16]

First

$$L_{10}(t',N)=\frac{q}{m}\int\Theta_{10}\left(e^{-t'\mathfrak{L}_{00}}(1-\pi_{00})\mathbf{E}\cdot\frac{\partial\mu_{00}{}^0}{\partial\mathbf{U}}\right)\prod_{j=1}^{N}d\mathbf{r}_jd\mathbf{v}_j. \tag{A4.1}$$

From (6.19)–(6.23), we see that

$$(1-\pi_{00})\mathbf{E}\cdot\frac{\partial\mu_{00}{}^0}{\partial\mathbf{U}}=-(1-\pi_{00})\mathbf{E}\cdot\mathbf{U}\mu_{00}{}^0=-\mathbf{E}\cdot\mathbf{U}(1-\pi_{00})\mu_{00}{}^0=0. \tag{A4.2}$$

Therefore $L_{10}(t',N)=0$.

Next

$$\Lambda_{10}(N)f'=\int\Theta_{10}(P_{00}f')\prod_{j=1}^{N}d\mathbf{r}_jd\mathbf{v}_j,\quad \Lambda_{20}(N)f'=\int\Theta_{10}(P_{10}f')\prod_{j=1}^{N}d\mathbf{r}_jd\mathbf{v}_j. \tag{A4.3}$$

Now from (6.23), we see that $P_{10}=0$, so $\Lambda_{20}(N)f'\equiv 0$. In order to evaluate $\Lambda_{10}(N)f'$, we must use the expression for Θ_{10} given in (6.18), and then we get

$$\Lambda_{10}(N)f'=\int\frac{1}{m}\sum_{j=1}^{N}\frac{\partial\theta(\mathbf{r}_j-\mathbf{R})}{\partial\mathbf{r}_j}\cdot\frac{\partial}{\partial\mathbf{U}}(P_{00}f')\prod_{j=1}^{N}d\mathbf{r}_jd\mathbf{v}_j. \tag{A4.4}$$

Since P_{00} is independent of $\mathbf{U}$, and f' is independent of $\mathbf{r}$ and $\mathbf{v}$, this result can be written as

$$\Lambda_{10}(N)f'=\frac{\partial f'}{\partial\mathbf{U}}\cdot\int\frac{1}{m}\sum_{j=1}^{N}\frac{\partial\theta(\mathbf{r}_j-\mathbf{R})}{\partial\mathbf{r}_j}P_{00}\prod_{j=1}^{N}d\mathbf{r}_jd\mathbf{v}_j. \tag{A4.5}$$

Since $\partial\theta(\mathbf{r}_j-\mathbf{R})/\partial\mathbf{r}_j=-\partial\theta(\mathbf{r}_j-\mathbf{R})/\partial\mathbf{R}$, the integrand in (A4.5) can be written as

$$P_{00}\left(-\frac{1}{m}\frac{\partial}{\partial\mathbf{R}}\sum_{j=1}^{N}\theta(\mathbf{r}_j-\mathbf{R})\right)=\frac{\partial}{\partial\mathbf{R}}P_{00}.$$

The coefficient of $\partial f'/\partial\mathbf{U}$ in (A4.5) is therefore

$$\int\frac{\partial}{\partial\mathbf{R}}P_{00}\prod_{j=1}^{N}d\mathbf{r}_jd\mathbf{v}_j=\frac{\partial}{\partial\mathbf{R}}\int P_{00}\prod_{j=1}^{N}d\mathbf{r}_jd\mathbf{v}_j=0, \tag{A4.6}$$

since $\int P_{00}\prod_{j=1}^{N}d\mathbf{r}_jd\mathbf{v}_j=1$. Therefore $\Lambda_{10}(N)f'\equiv 0$.

The term

$$K_{10}(t',N)f'(t-t')=\int\Theta_{10}(e^{-t\mathfrak{L}_{00}}\mathfrak{L}_{00}P_{00}f'(t-t'))\prod_{j=1}^{N}d\mathbf{r}_jd\mathbf{v}_j$$

is also zero, because $\mathfrak{L}_{00}P_{00}=0$, and

$$\mathfrak{L}_{00}(P_{00}f'(t-t'))=f'(t-t')\mathfrak{L}_{00}P_{00}=0.$$

The final term, and the one of chief interest, is

$$K_{20}(t',N)f(t-t')=\int\Theta_{10}(e^{-t'\mathfrak{L}_{00}}(1-\pi_{00})\mathfrak{L}_{10}P_{00}f'(t-t'))\prod_{j=1}^{N}d\mathbf{r}_jd\mathbf{v}_j$$
$$+\int\Theta_{10}[\alpha(-t'(1-\pi_{00})\mathfrak{L}_{10},t'\mathfrak{L}_{00})e^{-t'\mathfrak{L}_{00}}\mathfrak{L}_{00}P_{00}f'(t-t')]\prod_{j=1}^{N}d\mathbf{r}_jd\mathbf{v}_j+\int\Theta_{10}(e^{-t'\mathfrak{L}_{00}}\mathfrak{L}_{00}P_{10}f'(t-t'))\prod_{j=1}^{N}d\mathbf{r}_jd\mathbf{v}_j. \tag{A4.7}$$

For reasons which we have discussed ($\mathfrak{L}_{00}P_{00}=0$, $P_{10}=0$), the second and third integrals in (A4.7) vanish. This leaves the first integral to evaluate. Now from (6.4) and (6.18), we see that

$$\mathfrak{L}_{10}=\mathbf{U}\cdot\frac{\partial}{\partial\mathbf{R}}+\frac{1}{m}\sum_{j=1}^{N}\frac{\partial\theta(\mathbf{r}_j-\mathbf{R})}{\partial\mathbf{r}_j}\cdot\frac{\partial}{\partial\mathbf{U}}, \tag{A4.8}$$

and a short calculation yields

$$(1-\pi_{00})\mathfrak{L}_{10}P_{00}f'(t-t')=\beta P_{00}\mathbf{F}\cdot\left(\mathbf{U}+\frac{\partial}{\partial\mathbf{U}}\right)f'(t-t'), \tag{A4.9}$$

where we have set

$$\mathbf{F}=-\frac{1}{m\beta}\frac{\partial}{\partial\mathbf{R}}\sum_{j=1}^{N}\theta(\mathbf{r}_j-\mathbf{R}). \tag{A4.10}$$

Finally, we see from (6.18) and (A4.10) that we can write

$$\Theta_{10}=\beta\mathbf{F}\cdot\frac{\partial}{\partial\mathbf{U}}. \tag{A4.11}$$

Therefore, we get the result that

$$K_{20}(t',N)f'(t-t')=\beta^2\int P_{00}F_\mu e^{-t'\mathfrak{L}_{00}}F_\nu\prod_{j=1}^{N}d\mathbf{r}_jd\mathbf{v}_j\frac{\partial}{\partial U_\mu}\left(U_\nu+\frac{\partial}{\partial U_\nu}\right)f'(t-t'). \tag{A4.12}$$

PHYSICAL REVIEW VOLUME 145, NUMBER 1 6 MAY 1966

Quantum Noise. IV. Quantum Theory of Noise Sources

MELVIN LAX
Bell Telephone Laboratories, Murray Hill, New Jersey

(Received 16 November 1965)

For a quantum system describable in a Markoffian way, via a set of variables $\mathbf{a}=\{a_1,a_2,\cdots\}$, we show that the Langevin noise sources F_μ in the operator equations of motion $da_\mu/dt=A_\mu(\mathbf{a})+F_\mu$ possess second moments $\langle F_\mu(t)F_\nu(u)\rangle=2\langle D_{\mu\nu}(\mathbf{a},t)\rangle\delta(t-u)$. The diffusion coefficients $D_{\mu\nu}$ can be determined from a knowledge of the mean equations of motion via the (exact) time-dependent Einstein relation $2\langle D_{\mu\nu}\rangle=-\langle A_\mu a_\nu\rangle-\langle a_\mu A_\nu\rangle+d\langle a_\mu(t)a_\nu(t)\rangle/dt$, where $\langle\ \rangle$ represents a reservoir average. The sources F_μ,F_ν do not commute with one another, and as a result the commutation rules of the a_μ are shown to be preserved in time. The mean motion and diffusion coefficients are calculated for a harmonic oscillator, and for a set of atomic levels. We prove that two dynamically coupled systems (e.g., field and atoms) have uncorrelated Langevin forces if they are coupled to independent reservoirs. Radiation-field–atom coupling adds no new noise sources. We thus obtain simply the maser model including noise sources used in Quantum Noise V. *Direct* calculations of the mean motion and fluctuations in a system coupled to a reservoir yield relationships in agreement with the Einstein relation. For reservoirs violating time reversal, anomalous frequency shifts are found possible that violate the Ritz combination principle since $\Delta\omega_{12}+\Delta\omega_{23}+\Delta\omega_{31}$ need not vanish.

1. INTRODUCTION AND SUMMARY

OUR treatment of quantum noise in this paper and the preceding papers in this series[1] closely parallels a corresponding discussion of noise in classical systems.[2] The first paper (I) in our classical series provides a quasilinear approach to stationary Markoffian random processes. In the quasilinear case, it was easy to obtain the corresponding Langevin theory of noise sources. This work was generalized in (III) and (IV) to include classical nonstationary nonlinear Markoffian processes treated first from a Markoffian point of view and second from a Langevin noise-source point of view. In Paper IV, we emphasized the advantages and flexibility associated with the Langevin noise-source approach.

The present paper extends the noise-source technique to quantum systems. Quantum (and classical) systems experience dissipation and fluctuations through interaction with a reservoir. Our philosophy is that the reservoir can be *completely eliminated* provided that the frequency shifts and dissipation induced by the reser-

[1] A reference to QV is a reference to the author's fifth paper on quantum noise and relaxation. QI: Phys. Rev. **109**, 1921 (1958); QII: Phys. Rev. **129**, 2342 (1963); QIII: J. Phys. Chem. Solids **25**, 487 (1964); QIV: present paper; QV: in *Physics of Quantum Electronics*, edited by P. L. Kelley, B. Lax, and P. E. Tannenwald (McGraw-Hill Book Company, Inc., New York, 1966); QVI: "Moment Treatment of Maser Noise" (unpublished); QVII: "The Rate Equations and Amplitude Fluctuations" (unpublished).

[2] A reference to IV is a reference to the author's fourth paper on classical noise. I: Rev. Mod. Phys. **32**, 25 (1960); II: J. Phys. Chem. Solids **14**, 248 (1960); III: Rev. Mod. Phys. **38**, 359 (1966); IV: Rev. Mod. Phys. (to be published); V: Bull. Am. Phys. Soc. **11**, 111 (1966); VI: *ibid.*