

## One-dimensional harmonic liquid: A Fokker-Planck description of fluctuations from the nonequilibrium steady state

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(Received 10 February 1982)

The one-dimensional harmonic liquid, subject to a temperature gradient, is studied using the Fokker-Planck (FP) equation. A formalism is set up for solution of the FP equation and calculation of (1) the phase-space distribution function for the nonequilibrium steady state (NESS), (2) the conditional probability for evolution through phase space in the NESS, (3) phase-function averages in the NESS, (4) the correlation of phase functions in the NESS, etc. For the harmonic liquid this formalism can be implemented without approximation. Some properties of the harmonic liquid in equilibrium are examined to illustrate use of the formalism; so are some phase-function averages in the NESS. The displacement-displacement correlation function  $D(k, \omega)$  in the NESS is calculated and found to have the well-established, interesting amplitude and frequency dependence. The completeness of the Fokker-Planck description makes it possible to identify the physical processes responsible for the behavior of  $D(k, \omega)$  and the dynamic structure factor  $S(k, \omega)$ . The interesting features of light scattering from a liquid subject to a temperature gradient, seen in  $S(k, \omega)$  or  $D(k, \omega)$ , are due to light scattering from width fluctuations induced by the gradient; the interesting features in light scattering from a liquid-supporting shear are due to an attenuation mechanism that arises because of the velocity field induced by the shear. The results in this paper constitute a partial demonstration of the usefulness of the method employed in handling the Fokker-Planck equation.

### I. INTRODUCTION

The problem of characterization of a nonequilibrium steady state (NESS) and of a proper description of fluctuations about such a steady state is known to be far more difficult than the equivalent equilibrium problem. Renewed interest in the behavior of fluctuations about the NESS has arisen in response to the description of a fluid supporting flows given by Procaccia, Ronis, and Oppenheim<sup>1,2</sup> and by Kirkpatrick, Cohen, and Dorfman.<sup>3</sup> These authors and others have found a wave vector  $k$  and frequency  $\omega$  dependent contribution to  $S(k, \omega)$ , in the presence of flow, that involves "long-range correlations", the breaking of time-reversal symmetry, etc. Attempts to understand these results have led to a number of kinds of contributions; for example, those that confirm the results finding them in a different limit or in a different kind of calculation of  $S(k, \omega)$ , those that attempt to construct a suitable formal apparatus in which proper questions can, in principle, be asked exactly, answered exactly, etc.<sup>4-8</sup> This state of affairs is described nicely in the introduction and conclusion of the paper by

Tremblay, Arai, and Siggia (TAS).<sup>4</sup>

In this paper we describe the harmonic liquid<sup>9</sup> (the one-dimensional harmonic chain) subject to a temperature gradient through the introduction of suitable stochastic forces. This liquid is described by a Fokker-Planck equation that is able to be solved exactly. Thus, within this model, we are able to give an exact description of the NESS, and of fluctuations about the NESS. We are able to examine the way in which the temperature gradient makes itself known in physical phenomena. To be specific as to what is meant by this last remark let us briefly consider a light-scattering experiment. Light in entering a fluid couples to the density fluctuations present in the fluid. The frequency structure of the scattered light arises from the time evolution of these density fluctuations. A nonequilibrium state in the fluid could make itself known in two ways: (1) it could induce an additional fluctuation structure in the fluid from which the light will scatter and/or (2) it could modify the time evolution of the fluctuations to which the light couples. We look at  $S(k, \omega)$  to learn the relative importance of (1) and (2). We examine fluctuations about the

NESS and learn that width fluctuations play a major role in a fluid supporting a temperature gradient.

In Sec. II we introduce the harmonic liquid, the Fokker-Planck equation with which we describe this liquid in a temperature gradient, and the formal apparatus for solution of the FP equation that we employ. In Sec. III we review the equilibrium properties of the harmonic liquid in preparation for Sec. IV in which we calculate various single-particle averages in the NESS, various correlation functions in the NESS, etc. Particular attention is paid to the displacement-displacement correlation function that is related to  $S(k, \omega)$ . We summarize our results in Sec. V. There some time is spent in discussing a view of the physics that is involved in the scattering of light from a fluid supporting a temperature gradient and from a fluid in shear. Certain cumbersome details are found in the Appendices.

## II. HARMONIC LIQUID FOKKER-PLANCK EQUATION, ETC.

### A. Harmonic liquid and the Fokker-Planck equation

Consider a one-dimensional harmonic chain with Hamiltonian

$$\mathcal{H} = \sum_{i=1}^N \left[ \frac{1}{2} m v_i^2 + \frac{1}{2} \Gamma (x_{i+1} - x_i - a)^2 \right], \quad (1)$$

where  $x_i$  and  $m v_i$  are the position and momentum of the  $i$ th particle along the chain (see Fig. 1). The chain is attached to a system of temperature reservoirs that bring each particle (if uncoupled from its neighbors) to temperature  $T_i = k_B / \beta_i$ . Then, the FP equation for the chain is

$$\frac{\partial F}{\partial t} + L F = \frac{\gamma}{m} \sum_i \frac{\partial}{\partial v_i} \left[ v_i + \frac{1}{m \beta_i} \frac{\partial}{\partial v_i} \right] F, \quad (2)$$

$$L = \sum_i \left[ v_i \frac{\partial}{\partial x_i} + K_i \frac{\partial}{\partial v_i} \right]; \quad (3)$$

( $m K_i = -\partial \mathcal{H} / \partial x_i$ ) is the Liouville operator that gives the classical dynamics of the particles and  $\gamma$  is the damping constant that occurs in the associated Langevin equation

$$m \dot{v}_i + \gamma v_i = -m K_i + f_i(t). \quad (4)$$

The stochastic forces in Eq. (4), due to the reservoirs, obey

$$\langle f_i(t) f_j(t') \rangle = 2 \gamma k_B T_i \delta(t - t') \delta_{ij}. \quad (5)$$

The function  $F$  in Eq. (2) is the phase-space distribution function. The inhomogeneous ( $i$ -dependent) velocity terms on the right-hand side of Eq. (2) describe the diffusion of a particle in momentum space toward an equilibrium value of  $\langle v_i^2 \rangle = 2 k_B T_i / m$  that is enforced by the stochastic forces (the reservoirs). The physical picture of a fluid carrying a heat current, to which the model implied by Eq. (1) corresponds, is described further in Sec. V. Here it suffices to say that the heat current is carried by the fast modes that are the source of the stochastic forces but not by the slow modes that are driven by those forces.

It is useful in developing a solution to Eq. (2) to go to a set of dimensionless variables:

$$m v_0^2 = k_B T_0,$$

$$t = v_0 t / a,$$

$$y_i = v_i / v_0,$$

$$u_n = (x_n - n a) / a,$$

$$\eta = \gamma a / m v_0;$$

$$\begin{aligned} \frac{\partial F}{\partial t} + \sum_i \left[ y_i \frac{\partial}{\partial u_i} - \frac{\partial \beta \mathcal{H}}{\partial u_i} \frac{\partial}{\partial y_i} \right] F \\ = \eta \sum_i \frac{\partial}{\partial y_i} \left[ y_i + \frac{1}{\epsilon_i} \frac{\partial}{\partial y_i} \right] F, \end{aligned} \quad (6)$$

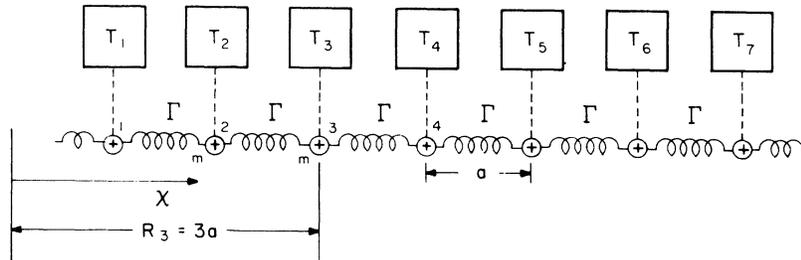


FIG. 1. Linear chain with temperature gradient. Each particle on the chain, of mass  $m$ , is connected to its neighbors by spring  $\Gamma$  and to its own thermal reservoir that drives it with stochastic force  $f_i(t)$ .

where  $\epsilon_i = \beta_i / \beta_0$  and  $\beta_0$  is a convenient reference temperature (in thermal equilibrium  $\epsilon_i = 1 \forall i$ ). [A remark about time reversal is in order. The momentum damping term in Eq. (6) describes forward evolution in time. Backward evolution in time is described by an equation like Eq. (6) except with a minus sign in front of  $\eta$  on the right-hand side  $\eta \rightarrow -\eta$ . See below, Eqs. (38) and (39).]

Equation (6) can be put in a particularly useful form upon writing it as an equation for  $\hat{F}$  defined by

$$F = \Phi_0 \hat{F}, \quad (7)$$

where  $\Phi_0 = \Psi_0 = \exp(-\beta\mathcal{H}/2)$ . We have

$$\frac{\partial \hat{F}}{\partial t} + L\hat{F} = \sum_i (-\eta a_i^\dagger a_i + p_i a_i^\dagger a_i) \hat{F},$$

with

$$L = \sum_i (a_i^\dagger B_i - a_i B_i^\dagger), \quad (8)$$

where

$$a_i^\dagger = -\frac{\partial}{\partial y_i} + \frac{1}{2} y_i,$$

$$a_i = \frac{\partial}{\partial y_i} + \frac{1}{2} y_i,$$

$$B_i^\dagger = -\frac{\partial}{\partial u_i} + \frac{1}{2} \frac{\partial \beta \mathcal{H}}{\partial u_i} = \sqrt{\lambda_2} \left[ -\frac{\partial}{\partial z_i} + \frac{1}{2} \frac{\partial \beta \mathcal{H}}{\partial z_i} \right],$$

$$B_i = \frac{\partial}{\partial u_i} + \frac{1}{2} \frac{\partial \beta \mathcal{H}}{\partial u_i} = \sqrt{\lambda_2} \left[ \frac{\partial}{\partial z_i} + \frac{1}{2} \frac{\partial \beta \mathcal{H}}{\partial z_i} \right], \quad (9)$$

and  $p_i = \eta(\epsilon_i^{-1} - 1)$ . The operators  $a_i^\dagger$  and  $a_i$  are harmonic oscillator creation and annihilation operators that create excitations in momentum space  $\{a_i^\dagger a_i$  has eigenvalues  $n = 0, 1, 2, 3, \dots$ ,  $[a_i, a_j^\dagger] = \delta_{i,j}$ , etc; the ground-state wave function  $\phi_0(y_i) = \exp(-y_i^2/4)$  is the equilibrium momentum-space distribution function  $\phi_0^2 = \exp(-\frac{1}{2}\beta m v_i^2)$ , etc.}. The operators  $B_i^\dagger$  and  $B_i$  are many-particle operators in configuration space. Unlike the momentum-space operators, in general, these operators have no simple algebra. However, for the harmonic liquid these operators take on a simple form when Eq. (8) is Fourier analyzed. Upon writing

$$a_i^\dagger = \sum_q \frac{1}{\sqrt{N}} a_q^\dagger e^{-iqR_i},$$

$$a_i = \sum_q \frac{1}{\sqrt{N}} a_q e^{iqR_i},$$

$$B_i^\dagger = \sum_q \sqrt{[(\omega_q \lambda_2)/N]} b_q^\dagger e^{-iqR_i},$$

$$B_i = \sum_q \sqrt{[(\omega_q \lambda_2)/N]} b_q e^{iqR_i},$$

$[\omega_q^2 = 4 \sin^2(qa/2); \lambda_2 = \beta \Gamma a^2, R_n = na]$ , we find

$$\frac{\partial \hat{F}}{\partial t} + L\hat{F} = \sum_q \left[ -\eta a_q^\dagger a_q + \sum_{q'} p(q+q') a_q^\dagger a_{q'} \right] \hat{F}, \quad (10)$$

with

$$L = \sum_q \sqrt{\lambda_2 \omega_q} (a_q^\dagger b_q - a_q b_q^\dagger) \quad (11)$$

and

$$p(q) = \frac{1}{N} \sum_i p_i e^{iqR_i}. \quad (12)$$

The operators  $b_q^\dagger, b_q$  obey the algebra  $[b_q, b_{q'}^\dagger] = \delta_{q,q'}$ ,  $[a_q, b_{q'}^\dagger] = 0 \forall q, q'$ , etc. It is the FP equation in the form given by Eq. (10) that we will use in this paper. The form of the Fokker-Planck equation in Eq. (8) is a particularly useful place from which to initiate the use of a variety of many body techniques for its solution.<sup>10</sup>

## B. Solution of the Fokker-Planck equation

To proceed to a formal (but useful) solution to Eq. (10) we view it as involving an unperturbed operator  $\mathcal{F}_0$  and a perturbation due to the temperature gradient  $\mathcal{F}'$ ;

$$\frac{\partial F}{\partial t} = -\mathcal{F}_0 \hat{F} + \mathcal{F}' \hat{F}, \quad (13)$$

where

$$\begin{aligned} \mathcal{F}_0 &= \sum_q \mathcal{F}_0(q) \\ &= \sum_q [ +\eta a_q^\dagger a_q + \sqrt{\lambda_2 \omega_q} (a_q^\dagger b_q - a_q b_q^\dagger) ] \end{aligned} \quad (14)$$

and

$$\mathcal{F}' = \sum_{qq'} p(q+q') a_q^\dagger a_{q'}. \quad (15)$$

We solve Eq. (13) by using perturbation theory on  $\mathcal{F}'$  employing the states and the algebra generated by  $\mathcal{F}_0$ . In preparation for this we note some of the properties of  $\mathcal{F}_0$  and  $\mathcal{F}'$ . For  $p_i = 0 \forall i$  the chain is at uniform temperature  $T_0$  and described by

$$\frac{\partial \hat{F}}{\partial t} = -\mathcal{F}_0 \hat{F}. \quad (16)$$

The eigenvalue problem associated by  $\mathcal{F}_0$ , a non-Hermitian operator,<sup>11</sup> is

$$\mathcal{F}_0 \Psi_\nu = \Lambda_\nu \psi_\nu, \quad (17)$$

with  $\Lambda_0=0$ ,  $\Psi_0=\Phi_0=\exp(-1/2\beta\mathcal{H})$ . Here we use  $\Phi_\nu$  to denote the left-hand eigenfunction associated with  $\Lambda_\nu$ . From Eq. (7)  $F=\Phi_0\hat{F}$ . The harmonic oscillator states generated from  $\{a_q^\dagger\}$  are complete for the purposes of describing motion in momentum space; the harmonic oscillator states generated from  $\{b_q^\dagger\}$  are complete for the purposes of describing motion in configuration space. Thus we assume the set of states  $\{\Psi_\nu\}$  is complete for the purposes of describing motion in phase space. Note that  $\mathcal{F}'$  creates excitations only; there are no matrix elements  $\langle 0 | \mathcal{F}' | \nu \rangle$ , etc., where we use the notation

$$\langle \nu | A | \mu \rangle = \int d\Gamma \Phi_\nu A \Psi_\mu; \quad (18)$$

$$d\Gamma = dy_1 \cdots dy_{N+1} du_1 \cdots du_{N+1},$$

$$\Phi_\nu = \Phi_\nu(y_1 \cdots y_{N+1}, u_1 \cdots u_{N+1}),$$

$$A = A(y_1 \cdots y_{N+1}, u_1 \cdots u_{N+1}),$$

etc.

$$T(tt')_{nm} = e^{-\Lambda_n t} T \left[ \delta_{n,m} + \int_{t'}^t dt'' \mathcal{F}'(t'')_{nm} + \sum_l \int_{t'}^t dt'' \int_{t'}^t dt''' \mathcal{F}'(t'')_{nl} \mathcal{F}'(t''')_{lm} + \cdots \right], \quad (21)$$

where  $T$  is the time-ordering operator and

$$\mathcal{F}'(tt')_{nm} = e^{(\Lambda_n - \Lambda_m)(t-t')} \mathcal{F}'_{nm}. \quad (22)$$

It is useful to view  $T(tt')$  as a power series in the addition of excitations, i.e.,  $T = T^{(0)} + T^{(2)} + T^{(4)} + \cdots$ ;  $T^{(0)}$  leaves the excitation level unchanged,  $T^{(2n)}$  increases the excitation level by  $2n$ . If at  $t \rightarrow -\infty$  the system is in thermal equilibrium with  $\beta_i = \beta_0 \forall i$ , then at time  $t$  the phase-space distribution function (the distribution function for the nonequilibrium steady state) NESS, is

$$F_s(lt | -\infty) = \sum_n \Phi_0(l) \Psi_n(l) T(t, -\infty)_{n0}, \quad (23)$$

and the average of  $A(l)$  in the NESS is ( $d\Gamma = dl$ )

$$\begin{aligned} \langle 0 | A(t) \rangle &\equiv \int dl A(l) F_s(lt | -\infty) \\ &= \sum_n \langle 0 | A | n \rangle T(t, -\infty)_{n0}. \end{aligned} \quad (24)$$

Similarly, for a correlation function defined by

$$\begin{aligned} \langle 0 | A(t) B(t') \rangle &\equiv \int dl \int dl' A(l) B(l') \\ &\quad \times F(lt | l't') F_s(l't' | -\infty), \end{aligned}$$

Let us construct a formal solution by perturbation theory to Eq. (13) for  $\hat{F}$ , and develop expressions for single-particle averages, two-particle averages, etc., that will be called for below. We write

$$\hat{F} = \sum_\nu c_\nu(t) e^{-\Lambda_\nu t} \Psi_\nu. \quad (19)$$

Then, for the conditional probability, the probability that the system is at  $l$  at time  $t$ , given that it is at  $l'$  at time  $t'$ , we have

$$F(lt | l't') = \Phi_0(l) \sum_{nm} \Psi_m(l) T(tt')_{mn} \Phi_n(l') / \Phi_0(l'), \quad (20)$$

where we have required  $F(lt | l't') = \delta(l-l')$ . Here we use the notation

$$l = (y_1 \cdots y_{N+1}, u_1 \cdots u_{N+1}),$$

$$l' = (y'_1 \cdots y'_{N+1}, u'_1 \cdots u'_{N+1}).$$

The time evolution operator in Eq. (20),  $T(tt')$ , follows from applying perturbation theory to  $\mathcal{F}'$ , in Eq. (13) and is given by

we have

$$\begin{aligned} \langle 0 | A(t) B(t') \rangle &= \sum_{lmn} \langle 0 | A | n \rangle T(tt')_{nm} \\ &\quad \times \langle m | B | l \rangle T(t', -\infty)_{l0}. \end{aligned} \quad (25)$$

The structure of this equation is clear. At  $t = -\infty$  the system is in equilibrium ( $\beta_i = \beta_0 \forall i$ ). Evolution from the equilibrium state at  $-\infty$  to the NESS at time  $t'$  is brought about by  $T(t', -\infty)$ . [It is during this evolution that the temperature gradient drives the system into the additional fluctuation structure described under (1) in the Introduction.] At time  $t'$  the NESS is probed by  $B(l')$ . The disturbance produced at  $t'$  by  $B$  propagates from  $t'$  to  $t$  according to  $T(tt')_{nm}$ . [The terms  $T^{(2)}(tt')$ ,  $T^{(4)}(tt')$ , . . . , produce the modification in the time evolution of fluctuations described under (2) in the Introduction.] At time  $t$  the system is probed by  $A$  and returned to the ground state, the equilibrium state. In principle,  $\langle 0 | A(t) B(t') \rangle$  is a function of  $t, t'$  and the initiation time (here  $-\infty$ ) at which the perturbation is turned on.

### C. Form of $\mathcal{F}'$ , diagonalization of $\mathcal{F}_0$ , etc.

Implementation of the formalism outlined above for calculation of  $\langle 0|A(t)\rangle$ ,  $\langle 0|A(t)B(t')\rangle$ , ..., requires use of Eqs. (25), (24), (22), and (21) for a particular choice of  $\mathcal{F}_0$  and  $\mathcal{F}'$ . For the harmonic liquid  $\mathcal{F}_0$  is given by Eq. (14) and  $\mathcal{F}'$  is given by Eq. (15).

To specify  $\mathcal{F}'$  we need the temperature gradient, we use

$$k_B T_i = \beta_0^{-1} (1 + \delta\epsilon \sin QR_i) \quad (26)$$

and consider (later) the  $Q \rightarrow 0$  limit. Thus,

$$p_i = \eta(\epsilon_i^{-1} - 1) = \eta \delta\epsilon \sin QR_i$$

and

$$p(q+q') = \frac{\eta \delta\epsilon}{2i} (\delta_{q', q-q} - \delta_{q', -q-q}) \quad (27)$$

It is convenient in working with  $\mathcal{F}_0$  to diagonalize the quadratic form in Eq. (14) for each  $q$ . To this end we employ the transformation, described in detail in Appendix A, which leads to

$$\mathcal{F}_0 = \sum_q \epsilon_\alpha(q) \alpha_q^\dagger \alpha_q + \epsilon_\beta(q) \beta_q^\dagger \beta_q,$$

where  $\epsilon_\alpha$ ,  $\epsilon_\beta$ ,  $a_q^\dagger$  in terms of  $\alpha_q^\dagger$  and  $\beta_q^\dagger$ , etc., are found in Eqs. (A10), (A11), etc. As discussed elsewhere,<sup>10</sup> the modes created by  $\alpha_q^\dagger$  and  $\beta_q^\dagger$  are a set of modes with which the phase-space distribution function can be described. These modes are constructed from the normal modes for motion in momentum space (created by  $a_q^\dagger$ ) and the normal modes for motion in configuration space (created by  $b_q^\dagger$ ). As  $\eta \rightarrow +\infty$ , the heavy damping limit, the  $\alpha$  modes are momentumlike and the  $\beta$  modes are configurationlike (i.e.,  $\alpha^\dagger \simeq a^\dagger$  and  $\beta^\dagger \simeq b^\dagger$ ). As  $\eta \rightarrow 0$ , no damping, the  $\alpha$  and  $\beta$  modes go over to modes that permit the distribution function to describe the motions given by the Liouville operator, i.e., the motions that solve the  $F = ma$  problem, the normal modes of a harmonic chain. This can be seen by

calculating  $\langle 0|y\rangle$ ,  $\langle 0|z\rangle$ , for  $F = \alpha_q^\dagger |0\rangle$ , etc. For  $q > 0$  the  $\beta$  modes are right-hand-going waves and the  $\alpha$  modes are left-hand-going waves. We call the modes created by  $\alpha^\dagger$  and  $\beta^\dagger$  the displacement modes.

The transformation that diagonalizes  $\mathcal{F}_0(q)$  has two forms according to whether the mode  $q$  is overdamped or underdamped, see Appendix A. As we proceed we write *all* equations in the form appropriate to the underdamped case [case 2(b) in Appendix A]. Thus for  $q \rightarrow 0$  some equations will have to be modified. To be more precise, in the  $\eta \rightarrow 0$  limit the eigenvalues  $\epsilon_\alpha(q)$ ,  $\epsilon_\beta(q)$  go over to

$$\epsilon_{\beta,\alpha}(q) = \pm i\omega(q),$$

where  $\omega(q) = 2\sqrt{\lambda_2} \sin(qa/2)$ . The parameter that determines the transition from underdamped to overdamped is  $P(q) = 2\omega(q)/\eta$ . For  $P(q) < 1$ , large  $\eta$ , a mode is overdamped, both  $\epsilon_\alpha(q)$  and  $\epsilon_\beta(q)$  are real; for  $P(q) > 1$ , small  $\eta$ , a mode is underdamped, both  $\epsilon_\alpha(q)$  and  $\epsilon_\beta(q)$  are complex [ $\epsilon_\alpha(q) = \epsilon_\beta^*(q)$ ]. Thus for fixed  $\eta$  the  $q \rightarrow 0$  modes are always overdamped. We ought, in principle, to separate  $\mathcal{F}_0(q)$  into two parts at  $\bar{q}$  given by  $P(\bar{q}) = 1$  and to write out all equations with due note taken of the change at  $\bar{q}$ . We do not do this.

Although we make no explicit use of it in this paper, it is useful to observe that the Smolouchoski equation (valid in the heavy damping limit,  $\eta \gg 1$ ) is derived from Eq. (8) by applying perturbation theory to  $\eta^{-1}$ . The result is

$$\frac{\partial \hat{F}}{\partial t} = -\mathcal{S} \hat{F},$$

where

$$\mathcal{S} = \frac{1}{\eta} \sum_i B_i^\dagger B_i = \frac{\lambda_2}{\eta} \sum_q \omega_q b_q^\dagger b_q. \quad (28)$$

See, for example, Ref. 12. The first equality here is general; the second equality is special to the harmonic liquid.

### III. EQUILIBRIUM PROPERTIES OF THE HARMONIC LIQUID

In this section we calculate the equilibrium properties of the harmonic liquid: the displacement-displacement correlation function, the static structure factor, the diffusion constant, etc. We do this in part to illustrate use of the FP formalism outlined above, to display some useful results, and to make comparisons with known results.

(1) *The displacement-displacement correlation function.* To calculate  $D(ij; tt')$ , defined by

$$D(ij; tt') = \langle 0|z_i(t)z_j(t')\rangle \quad (29)$$

in thermal equilibrium, we use Eq. (25) with  $T(t', -\infty) = T(t', -\infty)_{00} = 1$  and

$T(t, t')_{nm} = T^{(0)}(t, t')_{nm} = \delta_{n,m} \exp -\Lambda_n t$ . We have  $\langle 0 | z_i(t) z_j(0) \rangle = \langle 0 | z_i T^{(0)}(t) z_j | 0 \rangle$ . For  $z_i$ , we have

$$z_i = \sum_q \frac{1}{\sqrt{N\omega_q}} \{ [M(q)\beta_q^\dagger - iM^*(q)\alpha_q^\dagger] e^{-iqR_i} + [M(q)\beta_q + iM^*(q)\alpha_q] e^{iqR_i} \}, \quad (30)$$

from  $z_i \rightarrow b_q \rightarrow \cdots \beta_q + \cdots \alpha_q + \cdots$ , as shown in Eqs. (9) and (A11). Substituting Eqs. (30) into Eq. (29) we find for  $D^{(0)}$ , the equilibrium value of  $D$ ,

$$D^{(0)}(ij; t; 0) = \sum_q \frac{1}{N\omega_q} [M(q)^2 e^{-\epsilon_\beta(q)t} + M^*(q)^2 e^{-\epsilon_\alpha(q)t}] e^{iq(R_i - R_j)}, \quad t > 0. \quad (31)$$

To make comparison with the results of YMS we let  $\eta \rightarrow 0$  so that all modes  $q$  are underdamped. Then  $\epsilon_\alpha = -i\sqrt{\lambda_2\omega_q} = \epsilon_\beta^*$ ,  $M(q)^2 = M^*(q)^2 = \frac{1}{2}$ , and

$$D^{(0)}(ij; t; 0) = \sum_q \frac{1}{N\omega_q} \cos\sqrt{\lambda_2\omega_q} t \cos q(R_i - R_j). \quad (32)$$

Similarly, as  $\eta \rightarrow 0$  we have

$$\begin{aligned} \langle 0 | [z_i(t) - z_j(0)]^2 \rangle &= 2[D^{(0)}(ii; 00) - D^{(0)}(ij; t; 0)] \\ &= \sum_q \frac{2}{N\omega_q} [1 - \cos\sqrt{\lambda_2\omega_q} t \cos q(R_i - R_j)] \end{aligned} \quad (33)$$

$$= \frac{|R_i - R_j|}{a} + \Delta_{|R_i - R_j|}(S), \quad (34)$$

where  $S \propto t$ , and  $\Delta_{|R|}$  is defined by Yashida, Shobu, and Mori (YSM),<sup>9</sup>  $\Delta(0) = 0$ .

(2) *The static structure factor.* The static structure factor is defined by

$$S(k) = \frac{1}{N} \sum_{ij} e^{ik(R_i - R_j)} \langle 0 | e^{ikz_i} e^{-ikz_j} \rangle. \quad (35)$$

Upon using the well-known identities that exponentiate the average

$$\langle 0 | \exp(ikz_i) \exp(-ikz_j) \rangle = \exp[-k^2 D^{(0)}(ij; 00)/2],$$

and inserting Eq. (34) at  $t = 0$  ( $S = 0$ ) to find ( $K = ka$ ):

$$S(k) = \frac{\sinh(\frac{1}{2}K^2/\lambda_2)}{\cosh(\frac{1}{2}K^2/\lambda_2) - \cos K}. \quad (36)$$

The static structure factor has pseudo-Bragg peaks with width determined by the structural fluctuations, i.e.,  $K^2/\lambda_2$  (see Fig. 2).

(3) *Phonons.* The phonon modes supported by the harmonic liquid in equilibrium are found from examination of the  $k - \omega$  Fourier components of the displacement-displacement correlation function. Here we employ a slight generalization of this function that will be useful later:

$$D(kk'; \omega\omega') = \frac{1}{N} \sum_i \sum_j e^{ikR_i} e^{ik'R_j} \int dt \int dt' e^{-i\omega t} e^{-i\omega' t'} D(ij; tt'), \quad (37)$$

with  $D(ij; tt')$  given by Eq. (29) and in this case, thermal equilibrium, Eq. (31) for  $t \rightarrow 0$ . The time evolution of  $z(t)$  in Eq. (31) is forward time evolution. This is implied by our convention that unless otherwise noted time evolution is forward,  $t > 0$ . To denote the time evolution in Eq. (31) explicitly we write

$$D^{(0)}(ij; t > 0, 0) = \sum_q \frac{1}{N\omega_q} [M(q)^2 e^{-\epsilon_\beta^>(q)t} + M^*(q)^2 e^{-\epsilon_\alpha^>(q)t}] e^{iq(R_i - R_j)}, \quad (38)$$

where  $\epsilon_\alpha^>(q) = (\eta/2) - i\Omega_q$ ,  $\epsilon_\beta^>(q) = [\epsilon_\alpha^>(q)]^*$ . For backward time evolution we have

$$D^{(0)}(ij; t < 0, 0) = \sum_q \frac{1}{N\omega_q} [M(q)^2 e^{-\epsilon_\beta^<(q)t} + M^*(q)^2 e^{-\epsilon_\alpha^<(q)t}] e^{iq(R_i - R_j)},$$

where  $\epsilon_\alpha^<(q) = -\epsilon_\beta^>(q)$ ,  $\epsilon_\beta^<(q) = -\epsilon_\alpha^>(q)$ . We need  $D^{(0)}(;t>0)$  and  $D^{(0)}(;t<0)$  to learn about the frequency structure of the phonons because of the time integration. Making use of Eqs. (38) and (39) we find

$$D^{(0)}(kk';\omega\omega') = 2\pi\delta(\omega+\omega')\delta_{k',-k} \frac{\eta}{\omega_k} \left[ M(k)^2 \frac{1}{(\omega-\Omega_k)^2 + \frac{\eta^2}{4}} + M^*(k)^2 \frac{1}{(\omega+\Omega_k)^2 + \frac{\eta^2}{4}} \right], \tag{40}$$

where  $\Omega_k = \eta P(k)\Lambda(k)/2$ ,  $\Lambda(k)^2 = 1 - P(k)^{-2}$ , and  $P(k) = 2\sqrt{\lambda_2\omega_k}/\eta$ . Equation (40) describes two displaced Lorentzian lines at  $\pm\Omega_k$  with width determined by  $\eta$ . Had we been dealing with modes  $k$  that are overdamped,  $P(k) < 1$ , we would have found two Lorentzian lines centered at  $\omega=0$ ; a line that narrows as  $\eta \rightarrow +\infty$  and results from diffusion in configuration space and a line that broadens as  $\eta \rightarrow +\infty$  and results from diffusion in momentum space. The momentum-space Lorentzian has an amplitude that goes to zero as  $\eta \rightarrow +\infty$ . At fixed  $\eta$  as  $k$  evolves the displacement response function evolves as shown qualitatively in Fig. 3.

(4) *Velocity correlations and diffusion.* The velocity-velocity correlation function is defined by

$$\begin{aligned} \langle 0 | v_i(t)v_j(t') \rangle &= \frac{1}{m\beta_0} \langle 0 | y_i(t)y_j(t') \rangle \\ &= \frac{1}{m\beta_0} V(ij;tt'). \end{aligned} \tag{41}$$

To calculate  $V(ij;tt')$  we use

$$\begin{aligned} y_i &= \sum_q \frac{1}{\sqrt{N}} \{ [M(q)\alpha_q^\dagger + iM^*(q)\beta_q^\dagger] e^{-iqR_i} \\ &\quad + [M(q)\alpha_q - iM^*(q)\beta_q] e^{iqR_i} \} \end{aligned} \tag{42}$$

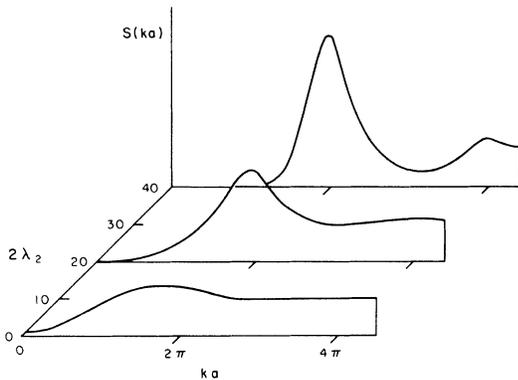


FIG. 2.  $S(k)$ : Static structure factor  $S(k)$ , Eq. (36), as a function of  $ka$  for several values of  $\lambda_2$  ( $\lambda_2$  measures the width of simple particle displacements).

and in thermal equilibrium we find

$$\begin{aligned} V^{(0)}(ij;t0) &= \sum_q \frac{1}{N} [M(q)^2 e^{-\epsilon_\alpha(q)t} + M^*(q)^2 e^{-\epsilon_\beta(q)t}] \\ &\quad \times e^{iq(R_i - R_j)}. \end{aligned} \tag{43}$$

The diffusion constant is related to  $V^{(0)}$  by

$$D = \int_0^\infty dt \frac{1}{m\beta_0} V(ii;t0). \tag{44}$$

To calculate  $D$  from Eq. (44) using Eq. (43) we take  $\eta \rightarrow 0$  [ $P(q) \rightarrow +\infty$  for all  $q$ ],  $M^2 = (M^*)^2 = \frac{1}{2}$ ,  $\epsilon_\beta = i\Omega$ ,  $\epsilon_\alpha = -i\Omega$ ,

$$\begin{aligned} V^{(0)}(ii;t0) &= \sum_q \frac{1}{N} \cos(\sqrt{\lambda_2\omega_q}t) \\ &= J_0(2\sqrt{\lambda_2}t). \end{aligned} \tag{45}$$

Thus  $D = 1/(2m\beta_0\sqrt{\lambda_2})$  in agreement with Eq. (3.8) of YSM. This result for  $D$  may also be found from Eq. (32) by using  $\langle 0 | [z_i(t) - z_i(0)]^2 \rangle \propto Dt$ .

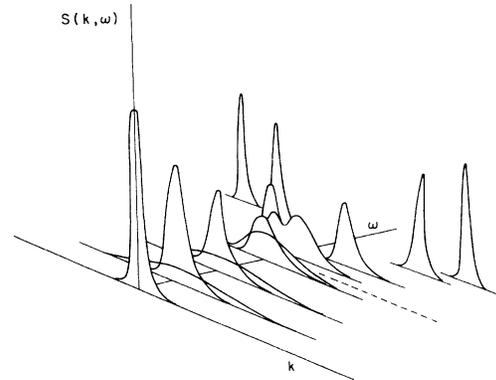


FIG. 3.  $S(k,\omega)$  at fixed  $\eta$ . Qualitative behavior of  $S(k,\omega)$  is shown as a function of  $k,\omega$ .  $S(k,\omega)$  has two Lorentzian peaks at  $\omega=0$  for  $P(k) \leq 1$ ; for  $P(k) > 1$  a pair of Brillouin lines appear.  $P(k)=1$  is shown by a dashed line in the  $k-\omega$  plane.

#### IV. FLUCTUATIONS FROM THE NONEQUILIBRIUM STEADY STATE

In this section we calculate a number of properties of the harmonic liquid that are sensitive to the presence of the temperature gradient; e.g., the average kinetic energy of a particle, the kinetic energy current, the displacement-displacement correlation function, etc. The formulation in Sec. II of calculations with  $\mathcal{F}_0, \mathcal{F}'$  suggests the use of diagrammatic perturbation theory. While such a perturbation theory can be developed, the use we would make of it is so modest as to preclude undertaking such a development. However, as we proceed we will use the language and physical ideas that such a development provides. In carrying through calculations that involve Eqs. (24) and (25), in which the non-trivial part of the time-evolution operator is involved, an important point to note is that  $T(tt')$  works in only one direction, it works only to increase the excitation level: Since matrix elements in  $\langle 0|A\rangle$  and  $\langle 0|AB\rangle$  are always from ground state to ground state it is up to  $A$  or  $AB$  to undo excitation produced by  $T(tt')$ . Thus for simple  $A$ 's and  $B$ 's the possibilities for the influence of  $T(tt')$  are extremely limited. This fact will be continually useful as will the fact that the series in excitation level  $T = T^{(0)} + T^{(2)} + T^{(4)} + \dots$  is also a series in  $\delta\epsilon$ ,  $T = 1 + \delta\epsilon + \delta\epsilon^2 + \dots$ .

(1) *The kinetic energy in NESS.* The average kinetic energy in the motion of the atom at the  $i$ th site is given by Eq. (24),

$$\langle 0|y_i^2\rangle = \langle 0|y_i^2|0\rangle + \sum_{n \neq 0} \langle 0|y_i^2|n\rangle T(t, -\infty)_{n0}. \quad (46)$$

To calculate  $\langle 0|y_i^2\rangle$  we use Eq. (15) and equations

$$\begin{aligned} \langle 0|y_i y_j\rangle - \delta_{i,j} \langle 0|y_i^2|0\rangle &= \frac{2\eta\delta\epsilon}{2i} \sum_{qq'} \frac{1}{N} e^{iqR_i} e^{iq'R_j} \\ &\times \left[ \delta_{q', Q-Q} \left( \frac{M(q)^2 M(Q-Q)^2}{\epsilon_\alpha(q) + \epsilon_\alpha(Q-Q)} + \frac{M(q)^2 M^*(Q-Q)^2}{\epsilon_\alpha(q) + \epsilon_\beta(Q-Q)} + \frac{M^*(q)^2 M(Q-Q)^2}{\epsilon_\beta(q) + \epsilon_\alpha(Q-Q)} \right. \right. \\ &\left. \left. + \frac{M^*(q)^2 M^*(Q-Q)^2}{\epsilon_\beta(q) + \epsilon_\beta(Q-Q)} \right) - \delta_{q', -Q-Q} (\dots) \right], \quad (48) \end{aligned}$$

where the ellipsis represents replacement of  $-Q$  by  $+Q$ . Each of the diagrams in Fig. 4 corresponds to the creation, by the temperature gradient, of a fluctuation structure in the NESS. This fluctuation

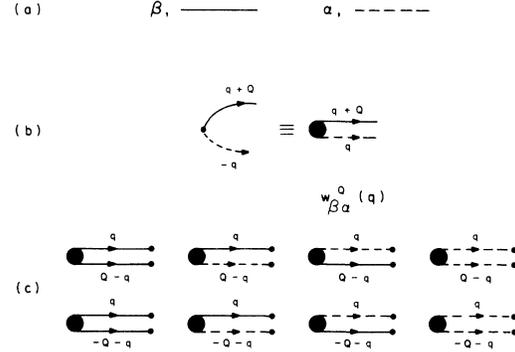


FIG. 4. Diagrams for  $\langle 0|y_i^2\rangle$ . Calculation of  $\langle 0|y_i^2\rangle$  involves use of Eq. (42) for  $y_i$  and Eq. (47) for  $\mathcal{F}'$ . Contributions to calculation of  $\langle 0|y_i^2\rangle$  can be found from eight diagrams that can be evaluated using rules like; —, represents a  $\beta$  displacement mode; ---, represents an  $\alpha$  displacement mode; each closed  $\beta$  line  $\rightarrow$  has a factor  $M(q)^2$ ; each closed  $\alpha$  line  $\leftarrow$  has a factor  $M^*(q)^2$ , etc. Excitation created by the temperature gradient, a pair of displacement modes, is termed a width fluctuation and denoted as shown in (b).

from Appendix A to find

$$\begin{aligned} \mathcal{F}' &= \sum_{qq'} p(q+q') [M(q)\alpha_q^\dagger + iM^*(q)\beta_q^\dagger] \\ &\times [M(q')\alpha_{q'}^\dagger + iM^*(q')\beta_{q'}^\dagger] \quad (47) \end{aligned}$$

and Eq. (42) for  $y_i$ . Then, there are eight contributions, shown in Fig. 4, that lead to the departure of  $\langle 0|y_i^2\rangle$  from  $\langle 0|y_i^2|0\rangle$ . These contributions are due to  $T^{(2)}$ , they are proportional to  $\delta\epsilon$ , and are given by

structure has no dynamics; it is established by the temperature gradient  $T(t, -\infty)$  and is time independent; its presence is probed by  $\langle 0|y_i^2\rangle$ . If we take the limit  $Q \rightarrow 0$  we find (following considerable

algebra)

$$\langle 0 | y_i y_j \rangle - \delta_{i,j} \langle 0 | y_i^2 | 0 \rangle = \delta_{i,j} \delta \epsilon \sin QR_i \quad (49)$$

as we would expect; cf. Eq. (26).

(2) *The kinetic energy current in NESS.* The kinetic energy current, a component of the heat current, is zero in the NESS. This result follows from the construction of the model system we are describing, see Secs. II and IV below. It also follows from direct calculation since the kinetic energy current, proportional to  $(y_i^2)y_i$ , has no nonzero matrix elements of the form  $\langle 0 | y_i^3 | n \rangle T(t, -\infty)_{n0}$  since  $T(t, -\infty)$  creates even numbers of excitations.

(3) *Phonons in the NESS.* We calculate the  $kk' - \omega\omega'$  Fourier component of the displacement-displacement correlation function defined in Eq. (37) with

$$D(ij, tt') = \sum_{lmn} \langle 0 | z_i | n \rangle T(t, t')_{nm} \times \langle m | z_j | l \rangle T(t', -\infty)_{l0}. \quad (50)$$

We note that  $z_i z_j$  can at most destroy two excitations; the nontrivial part of  $D$  must involve  $T(tt')_{nm} = T^{(0)}(t, t') \propto \delta_{n,m}$  and  $T(t', -\infty)_{l0} = T_{l0}^{(2)}$ . Thus we see, even before the details are displayed, that  $D(kk'; \omega\omega')$  probes the same fluctuation structures present in the NESS that are probed by  $\langle 0 | y_i^2 \rangle$ .  $D(kk'; \omega\omega')$  does not probe modification in the time evolution brought about by the temperature gra-

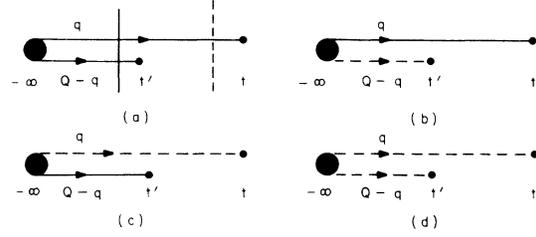


FIG. 5. Diagrams for  $\delta D$ . Calculation of  $\delta D$  requires evaluation of terms that correspond to the two-step destruction of the various width fluctuations. Temperature gradient begins at  $-\infty$  to establish a width fluctuation in the system that is probed by  $z_j$  at  $t'$  and decays into a displacement mode that propagates to  $t$  where it is destroyed by  $z_i$ . Diagrams correspond to contributions that are like those described in Fig. 4; there is a denominator with the “energy” of the modes present at the location of the vertical solid line. For each contribution with  $+Q$  there is also a term with  $-Q$ .

dient. In carrying through the calculation called for by Eq. (50) we note that  $t' > -\infty$  for all  $t'$  so that  $T(t', -\infty)$  involves forward time evolution only. On the other hand, we have  $t > t'$  as well as  $t < t'$  in  $T(tt')$  so that we must be careful with the time integration. In Fig. 5 we show four contributions to  $\delta D(ij; tt') = D - D^{(0)}$ . For one of them we have, Fig. 5(a), e.g.,

$$\delta D(ij; t > t') = + \sum_{qq'} \frac{2}{N \sqrt{\omega_q \omega_{q'}}} |M(q)|^2 |M(q')|^2 p(q+q') e^{iqR_i} e^{iq'R_j} \frac{1}{\epsilon_\beta(q) + \epsilon_\beta(q')} e^{-\epsilon_\beta^>(q)(t-t')}. \quad (51)$$

In this formula the denominator, involving  $\epsilon_\beta(q)$  and  $\epsilon_\beta(q')$ , comes from the time evolution from  $-\infty$  to  $t'$ ,  $T(t', -\infty)$ , and  $\exp[-\epsilon_\beta^>(t-t')]$  comes from the time evolution from  $t'$  to  $t$ . For  $t < t'$  we have the same result with  $\exp[-\epsilon_\beta^>(t-t')]$  replaced by  $\exp[-\epsilon_\beta^<(t-t')]$ . Assembling the contributions corresponding to the diagrams in Fig. 5 leads to ( $t = t - t' > 0$ )

$$\delta D(ij; t > 0) = \sum_{qq'} e^{iqR_i} e^{iq'R_j} \frac{p(q+q')}{i\Omega_q} \left[ \frac{1}{\epsilon_\beta(q) + \epsilon_\beta(q')} \frac{1}{\epsilon_\beta(q) + \epsilon_\alpha(q')} e^{-\epsilon_\beta^>(q)t} + \frac{1}{\epsilon_\alpha(q) + \epsilon_\alpha(q')} \frac{1}{\epsilon_\alpha(q) + \epsilon_\beta(q')} e^{-\epsilon_\beta^<(q)t} \right]. \quad (52)$$

Thus for  $\delta D(kk'; \omega\omega')$  we have (after some considerable algebra)

$$\delta D(kk'; \omega\omega') = 4\pi\delta(\omega + \omega') p(k+k') P(k, \omega) P(k', \omega'), \quad (53)$$

where

$$P(k, \omega) = \frac{1}{[i\omega - \epsilon_\beta(k)][i\omega - \epsilon_\alpha(k)]}. \quad (54)$$

Further algebraic manipulations are made easier upon using  $\bar{K} = (k+k')/2$ ,  $\kappa = k-k'$ . An example of this algebra is given in Appendix B. We find as  $Q \rightarrow 0$  [cf. Eq. (5.11) of TAS],

$$\delta D(kk';\omega\omega')=2\pi\delta(\omega+\omega')\delta_{k',-k}4\eta^2\delta\epsilon Qa\omega\Omega_k\frac{d\Omega_k}{d(ka)}(|P_\alpha|^2|P_\beta|^2)^2, \quad (55)$$

where  $P_\nu^{-1}=i\omega-\epsilon_\nu(k)$ . This extra contribution to  $D$  is to be compared to

$$D^{(0)}(kk';\omega\omega')=2\pi\delta(\omega+\omega')\delta_{k',-k}\left[\frac{M(k)^2}{\omega_k}|P_\beta|^2+\frac{M^*(k)^2}{\omega_k}|P_\alpha|^2\right]. \quad (56)$$

The following remarks are in order: (1)  $\delta D$  is proportional to the temperature gradient through the factor  $\delta\epsilon Qa$ ; (2)  $\delta D$  is antisymmetric in  $\omega$  so that upon doing more algebra (try Appendix B again) we have

$$D(kk';\omega\omega')=2\pi\delta(\omega+\omega')\delta_{k',-k}\frac{\eta}{2\omega k}\{|P_\alpha|^2[1-\epsilon(k,\omega)]+|P_\beta|^2[1+\epsilon(k,\omega)]\}, \quad (57)$$

where

$$\epsilon(k,\omega)=8\delta\epsilon Qa\frac{\sqrt{\lambda_2}}{\eta}\times\left[\frac{\eta^2\omega(q)^2}{[\omega^2-\omega(q)^2]^2+\eta^2\omega(q)^2}\right] \quad (58)$$

and  $\omega(q)^2=\lambda_2\omega_q$ ; (3) the amplitude asymmetry in  $D$  goes as  $\eta^{-1}$  [in TAS this factor is replaced by  $(Dq^2)^{-1}$ ].

An extensive discussion is given by TAS about results like Eq. (57) in terms of the physics that leads to their essential features. We understand results like that in Eq. (57) somewhat differently from TAS and we describe that understanding below. Before we go on to this let us remark further. Employing the procedure sketched above for obtaining  $\delta D$  we are able to calculate the velocity-velocity correlation function (it is much like the displacement-displacement correlation function) and the velocity-displacement correlation function. These correlation functions probe the fluctuation structure in the NESS; like  $\delta D$  they do not probe modification of the time evolution brought about by the temperature gradient. On the other hand, the energy-energy correlation function has contributions that are sensitive to modification of the time evolution of fluctuations. Note that the formalism we have employed provides a prescription for calculating the phase-space distribution function appropriate to the NESS and a prescription for determining the conditional probability for motion through phase space.

## V. DISCUSSION AND CONCLUSION

We begin here by describing the physical picture that develops from the results above, of light scattering from a liquid supporting a temperature

gradient. We also discuss the physical picture of light scattering from a fluid in shear. Some general and speculative concluding remarks follow this discussion.

*Temperature Gradient.* To review; a description of light scattering from a solid supporting a temperature gradient has been developed by Griffin.<sup>13</sup> The basic microscopic process considered by Griffin involves a photon  $(k,\omega)$ -phonon  $(q,\Omega)$  interaction of the type shown in Fig. 6. The Stokes and anti-Stokes components, at frequencies  $\omega+\Omega$  and  $\omega-\Omega$ , come from phonons that are moving toward or away from the direction of momentum transfer  $\vec{K}=\vec{k}-\vec{k}'$ . The amplitude of the two components is proportional to the number of phonons in the system at wave vector  $q$  in thermal equilibrium,  $N_0(q)$ . When a temperature gradient is present this picture is modified. The temperature gradient induces a heat current; it accomplishes this by driving the phonon distribution function away from  $N_0(q)$ . From simple modeling the phonon distribution function is given by

$$N(q)=N_0(q)+\tau_q\vec{C}\cdot\vec{\nabla}T\frac{\partial N_0}{\partial T}.$$

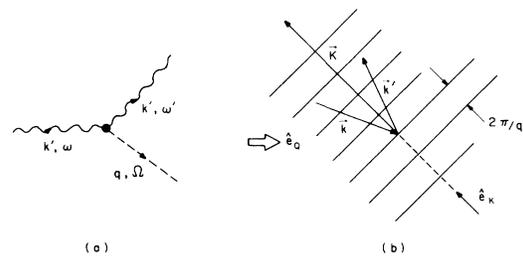


FIG. 6. Light scattering and the heat current. Basic microscopic process in Griffin's description of light scattering from a solid is shown in (a), a photon transfers energy and momentum to a phonon. This process involves momentum transfer  $\vec{K}$  to the photon. This momentum transfer has an additional amplitude when a heat current is present due to the component of phonon flux along the direction  $\hat{e}_x$ .

This nonequilibrium phonon distribution function has a shift at wave vector  $q$  proportional to  $\tau_q |\vec{C}|$ , where  $|\vec{C}|$  is the phonon velocity and  $\tau_q$  is a measure of the time required for the phonon distribution function at wave vector  $q$  to relax to local equilibrium (e.g., a phonon collision time). A heat current  $\bar{Q}$  results from the shift in the phonon distribution function. There are more phonons going in direction  $\hat{e}_Q(\hat{e}_K)$  than in direction  $-\hat{e}_Q(-\hat{e}_K)$ , Fig. 6. The difference in phonon populations, proportional to  $N(q) - N_0(q)$ , gives rise to the modification of the amplitude of the Stokes and anti-Stokes lines. The picture of the amplitude asymmetry developed by Griffin stresses the involvement of the phonon flux or heat current, induced by the temperature gradient, in the process.

In the picture of light scattering we have developed here a very different process is involved. The modes of the system we have examined are the long-wavelength modes, the hydrodynamic modes. By construction these modes carry no heat current. The heat current is carried by the high-frequency modes. It is participation in the heat current process that informs the high-frequency modes of the local temperature. The local temperature characterizes the reservoirs that drive the long-wavelength modes. That is, each particle on the chain is driven by a local reservoir; a manifestation of the aggregate of local high-frequency heat current carrying modes. Thus, the long-wavelength modes are in local thermal equilibrium. The light scattering is from modes in local thermal equilibrium that carry no heat current and no momentum flux. An argument like Griffin's in explanation of what is going on would be incorrect.

Before going further we emphasize once again that the modes with which we build up the phase-space distribution function, the phase-space modes, describe the damped, classical motions of the particles. In the state  $\alpha_q^\dagger |0\rangle$  a particle on the chain has a nonzero average value for its displacement, velocity, etc.,  $\langle 0 | y_i \alpha_q^\dagger | 0 \rangle \neq 0$ . This is to be contrasted with the phonon description of the chain. In the phonon state  $a_q^\dagger |0\rangle$  the expectation value of  $y_i$  is zero;  $\langle 0 | a_q y_i a_q^\dagger | 0 \rangle = 0$ . The phase-space modes are like to the coherent states that can be built up from the phonon modes.

To identify the source of the asymmetry of the amplitude in the light scattering from the hydrodynamic modes we begin by looking at the equilibrium result  $D^{(0)}(kk';\omega\omega')$ . The diagram, Fig. 7(a), shows that  $z_j$  probes the system at  $t'$ , excites it out of the equilibrium state, and that the frequency

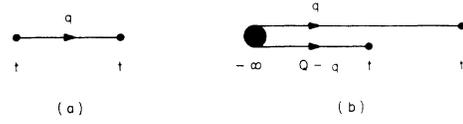


FIG. 7. Contributions to  $D^{(0)}$  and  $\delta D$ . Time evolution in  $D^{(0)}$  is the same as that in  $\delta D$ , compare (a) and (b) even though the source of the displacement mode that propagates in the two cases is different. In  $D^{(0)}$  the mode is created at  $t'$  by  $z_j$ ; in  $\delta D$  the mode is left over after part of a width fluctuation is destroyed at  $t'$  by  $z_j$ .

structure in  $D^{(0)}$  is a consequence of the time evolution of excitation from  $t'$  to  $t$ ; (at time  $t$  the excitation is destroyed by the probe  $z_i$ ). The amplitude of this process depends on the amplitude of various excitations in the probes,  $z_i, z_j$ ; recall Eq. (30). The diagram, Fig. 7(b), for  $D(kk';\omega\omega')$  shows that the source of the additional scattering is in the static, fluctuation structure induced by the temperature gradient. The probe  $z_j$  at  $t'$  couples to this fluctuation structure and creates an excitation of the same kind as that created from the equilibrium state by the action of  $z_j$  in  $D^{(0)}$ . The time evolution of this excitation is the same as that in  $D^{(0)}$  so that the frequency structure is essentially the same in  $\delta D$  as in  $D^{(0)}$ . Thus we must understand the source and characteristics of the static fluctuation structure.

The static fluctuation structure arises from local heating of the chain. This local heating is produced by pairs of displacement modes  $a_{q+Q}^\dagger a_{-q}^\dagger$  that carry no heat current, lead to no average velocity or average displacement, etc. We call these pairs of displacement modes the width fluctuations; they have  $\langle 0 | y_i^2 \rangle \neq 0$ ,  $\langle 0 | z_i^2 \rangle \neq 0$ . Light incident on the chain finds the width fluctuation modes to be present and is coupled to them. The size of the asymmetry in the light scattering depends upon the response of the system to the temperature gradient. That is, the width fluctuation modes are driven by the temperature gradient and acquire an amplitude that measures the systems response to that gradient. The temperature gradient drives four pairs of modes,  $\beta^\dagger \beta^\dagger$ ,  $\beta^\dagger \alpha^\dagger$ ,  $\alpha^\dagger \beta^\dagger$ , and  $\alpha^\dagger \alpha^\dagger$ . It is the mixed pairs  $\alpha^\dagger \beta^\dagger$  and  $\beta^\dagger \alpha^\dagger$  that are responsible for the essential features of the light scattering. (In Appendix C we repeat the phonon calculation from Sec. IV, considering these modes only, to demonstrate this point.) Perhaps the most important of these features is the dependence of the amplitude of the width fluctuation on  $\eta^{-1}$ , Eq. (58). This dependence comes about because the two displacement modes involved, e.g., propagate in the same direction (their group velocity is to the right) and have

time dependence  $\exp -[\epsilon_\beta(q+Q) + \epsilon_\alpha(-q)]t$ . As  $Q \rightarrow 0$ ,  $\exp -[\epsilon_\beta(q) + \epsilon_\alpha(q)]t \rightarrow \exp -\eta t$ ; the two displacement modes are “in phase” and the width fluctuation is driven coherently by the perturbation for times of order  $\eta^{-1}$ . The width fluctuation acquires an amplitude proportional to  $\eta^{-1}$  and becomes important as  $Q \rightarrow 0$ . By way of contrast, the pair of displacement modes  $\beta_{q+Q}^\dagger \beta_{-q}^\dagger$  evolve in time as  $\exp -[\epsilon_\beta(q+Q) + \epsilon_\beta(-q)]t$  which goes as  $[\exp(-\eta t)][\exp(-2i\Omega_q t)]$  as  $Q \rightarrow 0$ . Thus the width fluctuation mode  $\beta_{q+Q}^\dagger \beta_{-q}^\dagger$  is incoherent in its response to the temperature gradient because of the factor  $\exp(-2i\Omega_q t)$ ; it acquires amplitude proportional to  $(\eta + 2i\Omega_q)^{-1}$  and as  $\eta \rightarrow 0$  it is much less important. The time response we are describing here is not in the evolution initiated by the probe  $z_j(t')$ ; it is the time evolution in  $T(t', -\infty)$  that gives rise to the NESS.

Finally, we note that the width fluctuation mode  $\beta_{q+Q}^\dagger \beta_{-q}^\dagger$ , while involving right-hand going and left-hand going displacement modes, has group velocity to the right (the width fluctuation mode  $\alpha_{q+Q}^\dagger \beta_{-q}^\dagger$  has group velocity to the left). It is this group velocity that retains the sense of the temperature gradient and gives direction to the asymmetry in the light scattering. Several observations are in order. They are as follows.

(1) There is discussion of the result of analysis of calculation of light scattering from a fluid subject to a heat current that pays attention to the long-range correlations that are present, for example, in the amplitude of the asymmetry. These long-range correlations have their source in the response of various modes of a fluid system to the temperature gradient; i.e., in the structure of the NESS. The  $k \rightarrow 0$  modes of a fluid recover very slowly from a perturbation [from the Navier-Stokes equation  $\tau_k \simeq (Dk^2)^{-1}$ ] and the temperature gradient is able to drive them to particularly large amplitudes. Thus the long-range correlations are brought about by a mechanism that has none of the qualities of the long-range correlation in a system near its critical point. On the other hand, a system near its critical point can have modes with especially slow recovery from a perturbation so that a large amplitude NESS can be created in such systems.

(2) In the simplest kind of modeling of light scattering from the width fluctuations, the  $\beta^\dagger \alpha^\dagger$  and  $\alpha^\dagger \beta^\dagger$  modes only are retained, all of the important features are found and the line shape is Lorentzian; see Appendix C. Thus departures of the line shape from the Lorentzian have their source in the detailed features that are brought about by coupling of

the light to the  $\beta^\dagger \beta^\dagger$  and  $\alpha^\dagger \alpha^\dagger$  width fluctuation modes.

(3) The physics we have described is not at all that of Griffin. The mechanism that he gives attention to does not operate in the hydrodynamic regime. Thus in contrast to the remarks of TAS we find Griffin’s assessment of what is not going on to be correct.

*Shear.* Here we briefly discuss the physics operating in light scattering from a fluid in shear. Calculations which deal with this situation are described in detail in TAS and elsewhere. The principle observation we want to make is the following. Light scattering from a fluid arises from coupling to the density fluctuations that propagate in the fluid. The light senses the velocity of the fluctuations as well as their lifetime (i.e., the attenuation mechanisms). When a density fluctuation propagates in a fluid supporting a shear it is propagating in a fluid on which a velocity field  $\vec{v}_0$  has been impressed. Because of the nonlinear flow term in the Navier-Stokes equation  $(\vec{v} \cdot \vec{\nabla})\vec{v}$  the velocity field  $\vec{v}_0$  drives the density fluctuation *out of phase* and gives rise to an additional attenuation mechanism.<sup>14</sup> Thus the velocity field obeys [TAS Eq. (4.7)]

$$\left[ \frac{\partial}{\partial t} + \vec{v}_0 \cdot \vec{\nabla} \right] \delta \vec{v} = -\frac{c^2}{\rho_0} \vec{\nabla} \delta \rho + \eta \vec{\nabla}^2 \delta \vec{v} - (\delta \vec{v} \cdot \vec{\nabla}) \vec{v}_0$$

instead of the same equation with  $\vec{v} = 0$  (we have left out the stochastic force and the more complex form of the viscous damping). The terms  $\eta \vec{\nabla}^2 \delta \vec{v}$  and  $(\delta \vec{v} \cdot \vec{\nabla}) \vec{v}_0$  cause attenuation. The first by causing the velocity field to diffuse and the second by driving fluid in the opposite direction from that which will restore  $\delta \vec{v}$  to zero. The first of these attenuation mechanisms has an amplitude that is related to the strength of the stochastic forces; the second has an amplitude that is unrelated to the stochastic forces. Thus the characteristics of these two attenuation mechanism makes themselves known in  $S(k, \omega)$  in slightly different ways [see, for example, the treatment of  $S(k, \omega)$  by TAS], and give rise to the features noted.<sup>1-4, 15</sup>

In this paper we have discussed fluctuations from a nonequilibrium steady state for a particular model system, the harmonic liquid, using the Fokker-Planck equation. We exhibited a formalism for employing the Fokker-Planck description of the liquid that permits calculation of the NESS distribution function, the conditional probability for evolution through phase space in the NESS, single-particle

averages, two-particle correlation functions, etc. Application of this formalism to the equilibrium problem yields known results. Application of this formalism to calculation of correlation functions in the NESS yields results in essential agreement with those found earlier. By virtue of the completeness of the Fokker-Planck description it is possible to ask and answer specific questions about the physics that underlies the results. We find that the "excess" light scattering from a fluid supporting a temperature gradient is due to width fluctuation modes established in the fluid by the temperature gradient; that the excess light scattering from a fluid supporting shear is due to the attenuation mechanism brought into play by the velocity field established in the fluid by the shear. To the degree that one is interested in phenomena as well as their mathematical description an understanding of the physics of the phenomena is useful. Several variations on the standard experiments that probe the NESS are suggested; e.g., light scattering from liquid  $^4\text{He}$ , solid  $^4\text{He}$ , NaI, etc., supporting a standing second sound wave; light scattering from fluid in which a velocity field has been established by other than a shear flow, e.g., a shallow water wave; the propagation of an energy probe, second sound, in a fluid (solid) supporting a temperature gradient, etc.

#### ACKNOWLEDGMENTS

The author is pleased to acknowledge helpful conversations with M. C. Marchetti. This work was supported in part by the National Science Foundation.

#### APPENDIX A: DIAGONALIZATION OF THE QUADRATIC HAMILTONIAN

We want to diagonalize

$$\mathcal{H} \equiv Aa^\dagger a + Ba^\dagger b + Cab^\dagger + Db^\dagger b, \quad (\text{A1})$$

where  $[a, a^\dagger] = [b, b^\dagger] = 1$ ,  $[a, b^\dagger] = 0$ . For the variety of choices for  $A$ ,  $B$ ,  $C$ , and  $D$  this diagonalization, although always tedious, is easiest to carry through using the transformation

$$\hat{\mathcal{H}} = e^S \mathcal{H} e^{-S}, \quad (\text{A2})$$

$S = Ua^\dagger b + Vab^\dagger$  with  $U$  and  $V$  chosen to remove the cross terms,  $a^\dagger b$ ,  $ab^\dagger$ . Because the commutator of  $S$  with  $\mathcal{H}$  repeats itself,  $[S, S, S, \mathcal{H}] \propto [S, \mathcal{H}]$ , the transformation can be written out as a power series

in  $S$  and summed. We find,  $x = \sqrt{4UV}$ ,

$$\hat{\mathcal{H}} = \mathcal{H} + (b^\dagger b - a^\dagger a) \left[ (VB - UC) \frac{\sinh x}{x} - \frac{(A - D)}{2} (\cosh x - 1) \right] \quad (\text{A3})$$

with the conditions

$$B + U \left[ (D - A) \frac{\sinh x}{x} + 2(VB - UC) \frac{(\cosh x - 1)}{x^2} \right] = 0$$

and

$$C - V \left[ (D - A) \frac{\sinh x}{x} + 2(VB - UC) \frac{(\cosh x - 1)}{x^2} \right] = 0. \quad (\text{A4})$$

The operators  $a$ ,  $a^\dagger$ ,  $b$ , and  $b^\dagger$  transform thus:

$$\begin{aligned} \hat{a}^\dagger &= \cosh \frac{x}{2} \alpha^\dagger + \frac{2V}{x} \sinh \frac{x}{2} \beta^\dagger, \\ \hat{a} &= \cosh \frac{x}{2} \alpha - \frac{2U}{x} \sinh \frac{x}{2} \beta, \\ \hat{b}^\dagger &= \cosh \frac{x}{2} \beta^\dagger + \frac{2U}{x} \sinh \frac{x}{2} \alpha^\dagger, \\ \hat{b} &= \cosh \frac{x}{2} \beta - \frac{2V}{x} \sinh \frac{x}{2} \alpha, \end{aligned} \quad (\text{A5})$$

where  $\alpha$ ,  $\alpha^\dagger$ ,  $\beta$ , and  $\beta^\dagger$  denote the operators that work on the transformed states. Here we display the results of the transformation for several special cases.

*Case 1;*  $A > 0$ ,  $B > 0$ ,  $C = B$ ,  $D = 0$ . Equation (A4) is solved by  $V = -U$ ,  $\tan 2U = 2B/A \equiv Q$ . Then,

$$\hat{\mathcal{H}} = \frac{A}{2} (1 + R) \alpha^\dagger \alpha + \frac{A}{2} (1 - R) \beta^\dagger \beta, \quad (\text{A6})$$

$R = (1 + Q^2)^{1/2}$ , and

$$\begin{aligned} \hat{a}^\dagger &= \sqrt{[(R + 1)/2R]} \alpha^\dagger - \sqrt{[(R - 1)/2R]} \beta^\dagger, \\ \hat{a} &= \sqrt{[(R + 1)/2R]} \alpha - \sqrt{[(R - 1)/2R]} \beta, \\ \hat{b}^\dagger &= \sqrt{[(R + 1)/2R]} \beta^\dagger + \sqrt{[(R - 1)/2R]} \alpha^\dagger, \\ \hat{b} &= \sqrt{[(R + 1)/2R]} \beta + \sqrt{[(R - 1)/2R]} \alpha. \end{aligned} \quad (\text{A7})$$

Let us examine several limits. As  $A \rightarrow +\infty$ , the "damping" becomes large,  $Q \rightarrow 0$ ,  $R \rightarrow 1$  and

$\alpha^\dagger \rightarrow a^\dagger, \beta^\dagger \rightarrow b^\dagger$ . Thus in this limit the  $\alpha$  modes are momentumlike, the  $\beta$  modes are configurationlike and the two sets of modes involve only weak momentum-configuration coupling. This limit is the Smolouchowski equation limit in which momentum-space evolution and configuration-space evolution proceed independently. As  $A \rightarrow 0$ , there is little damping,  $Q \rightarrow +\infty, R \rightarrow +\infty$ , and  $\alpha^\dagger \rightarrow a^\dagger + b^\dagger, \beta^\dagger \rightarrow a^\dagger - b^\dagger$ ; both the  $\alpha$  and  $\beta$  modes involve combined momentum-space and configuration-space motion. In this limit the motion proceeds with little damping; the motion is that given by the Liouville operator.

Case 2;  $A > 0, B > 0, C = -B, D = 0$ . Equation (A4) is solved by  $V = U, \tanh 2U = 2B/A = P$ . There are two cases to be considered: case 2(a),  $P^2 \leq 1, \tanh x = P$  and case 2(b),  $P^2 \geq 1, x = X + iY, \tanh X = P^{-1}, Y = \pi/2$ .

Case 2(a). Here,

$$\hat{\mathcal{H}} = \frac{A}{2}(1+r)\alpha^\dagger\alpha + \frac{A}{2}(1-r)\beta^\dagger\beta, \quad (\text{A8})$$

$$r = (1 - P^2)^{1/2},$$

$$\hat{a}^\dagger = E\alpha^\dagger + F\beta^\dagger,$$

$$\hat{a} = E\alpha - F\beta,$$

$$\hat{b}^\dagger = E\beta^\dagger + F\alpha^\dagger,$$

$$\hat{b} = E\beta - F\alpha,$$

where  $E = \sqrt{(1+r)/2r}$  and  $F = \sqrt{(1-r)/2r}$ .

Case 2(b). Now,

$$\hat{\mathcal{H}} = \frac{A}{2}(1 - iP\Lambda)\alpha^\dagger\alpha + \frac{A}{2}(1 + iP\Lambda)\beta^\dagger\beta, \quad (\text{A10})$$

$$\Lambda = (1 - P^{-2})^{1/2},$$

$$\hat{a}^\dagger = M\alpha^\dagger + iM^*\beta^\dagger,$$

$$\hat{a} = M\alpha - iM^*\beta,$$

$$\hat{b}^\dagger = M\beta^\dagger + iM^*\alpha^\dagger,$$

$$\hat{b} = M\beta - iM^*\alpha,$$

where

$$M = \frac{1}{2}[\sqrt{(1+\Lambda)/\Lambda} + i\sqrt{(1-\Lambda)/\Lambda}].$$

Note that the eigenvalues  $\epsilon_\alpha$  and  $\epsilon_\beta$  are complex.

Case 3;  $A > 0, B > 0, C = B, 0 \leq D \leq A$ . Here,

$$\hat{\mathcal{H}} = \epsilon_\alpha\alpha^\dagger\alpha + \epsilon_\beta\beta^\dagger\beta, \quad (\text{A12})$$

$$\epsilon_{\alpha,\beta} = \frac{A+D}{2} \pm \frac{(A-D)}{2}R,$$

$$\hat{a}^\dagger = P\alpha^\dagger - N\beta^\dagger,$$

$$\hat{a} = P\alpha - N\beta,$$

$$\hat{b}^\dagger = P\beta^\dagger + N\alpha^\dagger,$$

$$\hat{b} = P\beta + N\alpha,$$

where  $P^2 = (R+1)/2R, N^2 = (R-1)/2R, R = (1+P^2)^{1/2}, P^2 = 4B^2/(A-D)^2$ .

## APPENDIX B: SIMPLIFICATIONS

We exhibit the algebraic manipulations that simplify equations like Eq. (55). Equation (55) is of the form

$$\delta D = 2\pi A \delta(\omega + \omega') p(k+k') P(k, \omega) P(k', \omega'),$$

where  $P(k, \omega) = P_\beta(k, \omega) P_\alpha(k, \omega); P_\nu(k, \omega)^{-1} = i\omega - \epsilon_\nu(k)$ . Use  $\bar{K} = (k+k')/2, \kappa = k-k'$  to write

$$\delta D = 2\pi A \delta(\omega + \omega') p(2\bar{K}) P\left[\bar{K} + \frac{\kappa}{2}, \omega\right] P\left[\bar{K} - \frac{\kappa}{2}, -\omega\right].$$

Take  $p(2\bar{K})$  to be given by Eq. (27); then as  $Q \rightarrow 0$  find

$$\delta D = \lim_{Q \rightarrow 0} 2\pi A \delta_{k', -k} \delta(\omega + \omega') \frac{\epsilon}{2i} \left[ P\left[\frac{\kappa}{2} + 2Q, \omega\right] P\left[\frac{\kappa}{2} - 2Q, -\omega\right] - P\left[\frac{\kappa}{2} - 2Q, \omega\right] P\left[\frac{\kappa}{2} + 2Q, -\omega\right] \right],$$

where we use the fact that  $P$  is an even function of  $k$ . Taylor-series expansion of  $P$  to first order in  $Q$  leads to

$$D = 16\pi A \delta_{k', -k} \delta(\omega + \omega') \frac{\delta\epsilon}{2i} Q \left[ P\left[\frac{\kappa}{2}, -\omega\right] P'\left[\frac{\kappa}{2}, \omega\right] - P\left[\frac{\kappa}{2}, \omega\right] P'\left[\frac{\kappa}{2}, -\omega\right] \right],$$

where the prime denotes differentiation with respect to  $\kappa$ . Using the product form of  $P$  ( $P = P_\alpha P_\beta$ ) and  $(d/d\kappa)P_\nu(\kappa/2, \omega) = +P_\nu(\kappa/2, \omega)[d\epsilon_\nu(\kappa/2)/d\kappa], P_\beta(k, \omega) = P_\alpha^*(k, -\omega), P_\alpha(k, \omega) = P_\beta^*(k, -\omega)$ , leads to

$$\begin{aligned}
& P(k, -\omega)P'(k, +\omega) - P(k, \omega)P'(k, -\omega) \\
&= |P_\alpha(k, \omega)|^2 |P_\beta(k, \omega)|^2 \\
&\quad \times \left[ [P_\beta(k, \omega) - P_\beta(k, -\omega)] \frac{d\epsilon_\beta(\kappa/2)}{d\kappa} + [P_\alpha(k, \omega) - P_\alpha(k, -\omega)] \frac{d\epsilon_\alpha(\kappa/2)}{d\kappa} \right].
\end{aligned}$$

Recall  $\epsilon_{\beta, \alpha} = \eta/2 \pm i\Omega$ ,  $d\epsilon_\alpha/d\kappa = -d\epsilon_\beta/d\kappa$ , and use  $P_\beta(k, \omega) - P_\beta(k, -\omega) = -2i\omega P_\beta(k, \omega)P_\beta(k, -\omega)$ ,  $P_\alpha(k, \omega) - P_\alpha(k, -\omega) = -2i\omega P_\alpha(k, \omega)P_\alpha(k, -\omega) = -2i\omega P_\beta^*(k, \omega)P_\beta^*(k, -\omega)$  to find

$$\delta D = 16A\delta_{k', -k}\delta(\omega + \omega')\delta\epsilon Q\omega |P_\alpha(k, \omega)|^2 |P_\beta(k, \omega)|^2 \frac{d\Omega(\kappa/2)}{d\kappa} \text{Im}P_\beta(k, \omega)P_\beta(k, -\omega).$$

Finally, use

$$\text{Im}P_\beta(k, \omega)P_\beta(k, -\omega) = -\eta\Omega |P_\beta(k, \omega)|^2 |P_\alpha(k, \omega)|^2$$

to achieve

$$\delta D = -16\pi\eta^2\delta_{k', -k}\delta(\omega + \omega')Q\delta\epsilon\omega\Omega(\kappa/2)\frac{d\Omega(\kappa/2)}{d\kappa} (|P_\alpha(k, \omega)|^2)^2 (|P_\beta(k, \omega)|^2)^2.$$

Manipulation to achieve the form in Eq. (57) proceeds as follows. The real part of  $D$  is [aside from the factor  $2\pi\delta_{k', -k}\delta(\omega + \omega')$ ]

$$D \propto \frac{\eta}{2\omega_k} (|P_\alpha|^2 + |P_\beta|^2) + A\omega (|P_\alpha|^2)^2 (|P_\beta|^2)^2.$$

We want to write this in the form

$$D \propto \frac{\eta}{2\omega_k} [ |P_\alpha|^2(1 - \epsilon) + |P_\beta|^2(1 + \epsilon) ].$$

Comparison of these two equations and suitable algebra leads to  $[\omega(q)^2 = \lambda_2\omega_q]$

$$\epsilon = 8\delta\epsilon Qa \left[ \frac{\lambda_2}{\eta} \right]^{1/2} \frac{\eta^2\omega(q)^2}{[\omega^2 - \omega(q)^2]^2 + \eta^2\omega(q)^2}.$$

See also Appendix C.

### APPENDIX C: EVALUATION OF $\delta D(kk'; \omega\omega')$

In this appendix we evaluate  $\delta D(kk'; \omega\omega')$  in the approximation in which the  $\beta^\dagger\alpha^\dagger$  and  $\alpha^\dagger\beta^\dagger$  terms alone are used. From Eq. (52) we have

$$\begin{aligned}
\delta D(kk'; \tau = t - t') &= \frac{-2}{\sqrt{\omega_k\omega_{k'}}} p(k+k') |M(k)|^2 |M(k')|^2 \\
&\quad \times \left[ \frac{1}{\epsilon_\beta(k) + \epsilon_\alpha(k')} e^{-\epsilon_\beta(k)\tau} + \frac{1}{\epsilon_\alpha(k) + \epsilon_\beta(k')} e^{-\epsilon_\alpha(k)\tau} \right], \tag{C1}
\end{aligned}$$

where  $p(k+k') = \eta\delta\epsilon(\delta_{k', -k+Q} - \delta_{k', -k-Q})/2i$ . The first term in the large parentheses comes from  $\beta^\dagger\alpha^\dagger$  and the second term comes from  $\beta^\dagger\alpha^\dagger$ . Use  $|M|^2 = 1/2\Lambda$  and  $\sqrt{\Lambda_2\omega_q}\Lambda = \Omega$  to write

$$\delta D(kk'; t) = -\frac{\lambda_2}{2} p(k+k') \frac{1}{\Omega_k\Omega_{k'}} \left[ \frac{1}{\epsilon_\beta(k) + \epsilon_\alpha(k')} e^{-\epsilon_\beta(k)\tau} + \frac{1}{\epsilon_\alpha(k) + \epsilon_\beta(k')} e^{-\epsilon_\alpha(k)\tau} \right]. \tag{C2}$$

Upon using  $p(k+k')$  and considering the limit  $Q \rightarrow 0$  we find

$$\delta D(kk';t) = -\frac{\lambda_2 \delta \epsilon Q \Omega'_k}{2i \Omega_k^2} \left[ \left( \frac{1}{\Omega_k} - \frac{i}{\eta} \right) e^{-\epsilon_{\beta}(k)\tau} + \left( \frac{1}{\Omega_k} + \frac{i}{\eta} \right) e^{-\epsilon_{\alpha}(k)\tau} \right], \quad (C3)$$

where  $\Omega'_k = d\Omega_k/dk$ . For the real part of  $\delta D$  we have

$$\text{Re} \delta D = \frac{\lambda_2 \delta \epsilon Q \Omega'_k}{2\Omega_k^2 \eta} (e^{-\epsilon_{\beta}(k)\tau} - e^{-\epsilon_{\alpha}(k)\tau}). \quad (C4)$$

This is to be compared to the real part of  $D^{(0)}(kk';\tau)$ ;

$$\text{Re} D^{(0)}(kk';\tau) = \frac{1}{2\omega_k} (e^{-\epsilon_{\beta}(k)\tau} + e^{-\epsilon_{\alpha}(k)\tau}). \quad (C5)$$

In proceeding from Eq. (C3) to Eq. (C4) and in writing Eq. (C5) we have ignored the complex structure in the exponential time dependence. The justification for this is that when using  $D^{(0)}(\tau)$  or  $\delta D(\tau)$  to calculate  $D^{(0)}(\omega)$  or  $\delta D(\omega)$  we use forward and backward time evolution so that the exponential time dependence like  $e^{-\epsilon_{\beta}(k)\tau}$  leads to a real result. Combining Eqs. (C4) and (C5) leads to

$$\text{Re} D(kk';\tau) = \frac{1}{2\omega_k} [(1 + \epsilon_k) e^{-\epsilon_{\beta}(k)\tau} + 1(1 - \epsilon_k) e^{-\epsilon_{\alpha}(k)\tau}], \quad (C6)$$

where

$$\epsilon_k = \frac{\delta \epsilon Q \Omega'_k}{2\eta} \frac{1}{\Lambda_k}, \quad (C7)$$

which should be compared to Eq. (57).

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<sup>8</sup>M. C. Marchetti (unpublished).

<sup>9</sup>T. Yoshida, K. Shobu, and H. Mori, Prog. Theor. Phys. **66**, 759 (1981), referred to as YSM.

<sup>10</sup>The methods employed here have been successfully

used on a variety of problems; e.g., the dynamics of the Toda chain [R. A. Guyer (unpublished)], the motion of a "wedgie" (a model for a flexible macromolecule) [R. A. Guyer and D. Prato (unpublished)].

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<sup>12</sup>R. A. Guyer, Phys. Rev. A **21**, 4484 (1980).

<sup>13</sup>P. A. Griffin, Can. J. Phys. **46**, 2843 (1968).

<sup>14</sup>J. Machta, I. Oppenheim, and I. Procaccia, Phys. Rev. Lett. **42**, 1368 (1979); this picture could be developed along the lines of the discussion of  $(\vec{v} \cdot \vec{\nabla}) \vec{v}$  given by Lovesey in *Condensed Matter Physics* (Benjamin, Reading, Mass., 1980).

<sup>15</sup>A.-M. S. Tremblay, Eric D. Siggia, and M. R. Arai, Phys. Lett. **76A**, 57 (1980).