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Statistical mechanics of stationary states. V. Fluctuations in systems with shear flow

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Long-wavelength thermal fluctuations in a fluid with a linear shear are investigated. Certain equal-time correlations are found to have a long-range part. The dynamic structure factor is modified in such a way that the Landau-Plazcek ratio no longer holds. A light-scattering experiment is proposed to test some of these results.

I. INTRODUCTION

In the previous papers in this series we developed a statistical mechanical theory of nonequilibrium stationary states (NESS). In particular, we examined the fluctuations that occur in NESS of simple fluids and discovered some interesting modifications of the equilibrium fluctuations. The formal properties of NESS correlation functions were discussed in I.[1] In III[2] we examined the dynamic structure factor (density-density time correlation function) for a fluid subject to a temperature gradient but without a velocity gradient. We found an asymmetry in the Brillouin components of the light-scattering spectrum, which was linked to the breaking of time reversal symmetry.[2,3] This asymmetry is pronounced for small scattering angles (small-k vectors), and is negligible for large angles. It has been experimentally observed by Beysens, Garrobos, and Zalcyer.[4]

In this paper, we examine the question of fluctuations in a system with a steady, linear, shear flow. We shall show that the couplings to the dissipative momentum flux induce fluctuations that, for small k, differ significantly from the fluctuations predicted on the basis of a local equilibrium assumption. In particular, we evaluate below the dynamic structure factor and the momentum autocorrelation function. One interesting result is that, although the symmetry of the two Brillouin components of the spectrum is not affected now, the Landau-Plazcek ratio between the Brillouin and the Rayleigh components no longer holds.[5-7] We link this finding to the appearance of long-range correlations in the pair distribution function. Another interesting result is that the nonequilibrium contributions to the correlation functions arise solely from couplings to the shear part of the velocity field and there is no contribution due to the rotational part.

As in the previous papers, we consider time-dependent correlation functions in nonequilibrium steady states, $\langle \underline{A}_k(t)\underline{A}_{-k}\rangle_{NE}$. We restrict our considerations to k vectors that satisfy

$$\left(\frac{c_0}{L\Gamma_s}\right)^{1/2} < k \ll \frac{c_0}{\Gamma_s}\,, \tag{1.1}$$

where L is a linear dimension of the system in which the steady state exists, c_0 is the adiabatic sound speed, and Γ_s is the sound attenuation coefficient. These restrictions imply that sound modes in the fluid with wave vector k decay over a distance smaller than the size of the system but are still only lightly damped. In this regime we may neglect the boundaries of the system. It turns out that the upper bound on k will also allow us to neglect terms of order $1/k$ relative to terms of order $1/k^2$. In Sec. III we will discuss the restrictions on k in more detail in the context of a proposal for a light-scattering experiment to test our results.

A short report of the results concerning light scattering was given in Ref. 8. Here, we expand the treatment and consider also the interesting changes in the momentum autocorrelation functions.

In Sec. II we review the ideas involved in our method. In Sec. III we compute the dynamic structure factor and discuss the implications of the result for the spectrum of light scattered from a linear shear flow. Section IV deals with the momentum autocorrelation functions. We summarize and discuss the results in Sec. V.

II. SUMMARY OF PREVIOUS RESULTS

We consider a classical simple fluid whose macroscopic evolution is described by the equations of fluid mechanics. We start with the classical phase functions for the densities of the conserved variables; number, energy, and momentum: $N(\vec{r},t)$, $E(\vec{r},t)$, $\vec{P}(\vec{r},t)$. Denoting this set by the symbol $\underline{A}(\vec{r},t)$, we write the equations of continuity:

$$\dot{\underline{A}}(\vec{r},t) = -\nabla\cdot\underline{J}(\vec{r},t)\,, \tag{2.1}$$

where $\underline{J}(\vec{r},t)$ is the set of microscopic fluxes. The set $\underline{A}$ is assumed to span the space of slow variables of the system. Associated with this set are the conjugate thermodynamic variables denoted by $\underline{\Phi}(\vec{r},t)$, which are related to the local temperature, chemical potential and velocity field. The set $\underline{\Phi}$ appears in the definition of the local equilibrium distribution function:

$$f_L(X)=\frac{\exp[\int\underline{\Phi}(\vec{r})\cdot\underline{A}(\vec{r};X)d\vec{r}]}{\sum_N\int dX\exp[\int\underline{\Phi}(\vec{r})\cdot\underline{A}(\vec{r};X)d\vec{r}]}\,,\qquad(2.2)$$

where X is the phase point.

In I[1] and IV[9] we derived the expression for the NESS average of an arbitrary dynamical variable, and, in particular, for correlation functions. To linear order in the gradients of the conjugate thermodynamic variables the equal-time correlation of any of the conserved variables is

$$\langle\hat{\underline{A}}(\vec{r})\hat{\underline{A}}(\vec{r}_1)\rangle_{NE}=\langle\hat{\underline{A}}(\vec{r})\hat{\underline{A}}(\vec{r}_1)\rangle+\int\langle\hat{\underline{A}}(\vec{r})\hat{\underline{A}}(\vec{r}_1)\hat{\underline{A}}(\vec{r}_2)\rangle\cdot[\underline{\Phi}(\vec{r}_2)-\underline{\Phi}(\vec{r})]\,d\vec{r}_2-\int_0^\infty d\tau\langle\hat{\underline{A}}(\vec{r},\tau)\hat{\underline{A}}(\vec{r}_1,\tau)I_T\rangle\cdot\nabla\underline{\Phi}(\vec{r})\,.\qquad(2.3)$$

The unsubscripted angular brackets refer to an equilibrium average where the uniform values of the conjugate thermodynamic variables are given by the values of $\underline{\Phi}$ at the point $\vec{r}$ in the fluid and $\hat{\underline{A}}(\vec{r}_1,t)\equiv\underline{A}(\vec{r}_1,t)-\langle\underline{A}(\vec{r}_1)\rangle$. The second term on the right-hand side (RHS) of (2.3) comes from an expansion of local equilibrium around the total equilibrium defined by $\underline{\Phi}(\vec{r})$. This nonlocality correction can be shown to be very small[2] and will be ignored henceforth. The third term is a nonequilibrium effect arising from couplings to the total dissipative current $\underline{I}_T$ defined by

$$\underline{I}_T\equiv\underline{J}_T-\langle\underline{J}_T\hat{\underline{A}}_T\rangle\cdot\langle\hat{\underline{A}}_T\hat{\underline{A}}_T\rangle^{-1}\cdot\hat{\underline{A}}_T-\langle\underline{J}_T\rangle\,,\qquad(2.4)$$

where, for example,

$$\underline{A}_T\equiv\int d\vec{r}_1\underline{A}(\vec{r}_1,\tau)\,.$$

It is easy to verify that in simple fluids there are only two dissipative currents—one associated with the energy flux and the other with the momentum flux. The number flux is purely convective and has no irreversible part. Thus the third term on the rhs of Eq. (2.3) has two contributions, one from $\nabla\underline{\Phi}(\vec{r})=-\nabla\beta(\vec{r})$ and one from $\nabla\underline{\Phi}(\vec{r})=\nabla\beta(\vec{r})\vec{v}(\vec{r})$. We restrict our attention to systems which are linearly displaced from equilibrium, in which case we can consider systems with temperature gradients and no flow, and systems with no temperature gradients with linear shears. The former case was analyzed in III.[2] The latter case is analyzed in this paper. The steady-state conditions for the systems considered here are

$$\nabla p_h=\nabla\beta=0,\quad\nabla\cdot\vec{v}=0,\quad\nabla^2\vec{v}=0\,.\qquad(2.5)$$

To simplify our analysis we choose our frame of reference so that the velocity vanishes at $\vec{r}$. It is also convenient to work in Fourier space; then Eq. (2.3) becomes[1]

$$\langle\underline{A}_k\underline{A}_{-k}\rangle_{NE}=\langle\underline{A}_k\underline{A}_{-k}\rangle-\int_0^\infty d\tau\,\Gamma(\vec{k},\tau)\cdot\nabla\beta\vec{v},\qquad(2.6)$$

where

$$\frac{1}{V}\langle\underline{A}_k\underline{A}_{-k}\rangle_{NE}\equiv\int d\vec{r}_1e^{i\vec{k}\cdot(\vec{r}-\vec{r}_1)}\langle\hat{\underline{A}}(\mathrm{r})\hat{\underline{A}}(\vec{r}_1)\rangle_{NE}$$

and

$$\frac{1}{V}\,\tilde{\underline{\Gamma}}(\vec{k},\tau)\equiv\int d\vec{r}_1e^{i\vec{k}\cdot(\vec{r}-\vec{r}_1)}\langle\hat{\underline{A}}(\vec{r},\tau)\hat{\underline{A}}(\vec{r}_1,\tau)\tilde{I}_{P,T}\rangle\,.$$

The total dissipative momentum current is given by

$$\tilde{I}_{P,T}=\tilde{\tau}-\left[\left(\frac{\partial P_h}{\partial e}\right)_n\hat{E}_T+\left(\frac{\partial P_h}{\partial n}\right)\hat{N}_T+P_h\right]\tilde{1}\,.\qquad(2.7)$$

Here, $\tilde{\tau}$ is the total stress tensor, P_h is the hydrostatic pressure, e is the average energy density, and n is the average number density.

We have now reduced the problem of computing nonequilibrium correlation functions to one of computing equilibrium correlation functions. The first term on the rhs of Eq. (2.6) is easily evaluated using thermodynamics. The nonequilibrium correction is more difficult since it involves a time correlation function. We showed in Sec. III of paper III[2] of this series that the leading contribution to $\Gamma(k,t)$ can be evaluated using linearized hydrodynamics with the result that

$$\tilde{\underline{\Gamma}}(\vec{k},t)=\exp(\tilde{\underline{M}}_kt)\,\tilde{\underline{\Gamma}}(k)\exp(\tilde{\underline{M}}_k^\dagger t)\,,\qquad(2.8)$$

where $\tilde{\underline{\Gamma}}(\vec{k})\equiv\tilde{\underline{\Gamma}}(\vec{k},0)$; $\tilde{\underline{M}}_k$ is the k Fourier component of the hydrodynamic matrix linearized around equilibrium:

$$\tilde{\underline{M}}_k=\begin{pmatrix}0&0&ik/m&0&0\\-k^2\kappa_n&-k^2\kappa_e&ikh/mn&0&0\\ik\chi_n&ik\chi_e&-k^2\nu_l&0&0\\0&0&0&-k^2\nu&0\\0&0&0&0&-k^2\nu\end{pmatrix},\qquad(2.9)$$

where m is the particle mass, h the enthalpy density, n the number density,

$$\chi_n\equiv\left(\frac{\partial P_h}{\partial n}\right)_e,\quad\chi_e\equiv\left(\frac{\partial P_h}{\partial e}\right)_n,$$

$$\kappa_n \equiv \lambda\left(\frac{\partial T}{\partial n}\right)_e, \quad \kappa_e \equiv \lambda\left(\frac{\partial T}{\partial e}\right)_n,$$

$$\nu_l \equiv (\zeta + \tfrac{4}{3}\eta)/mn, \quad \nu \equiv \eta/mn,$$

λ is the thermal conductivity, ζ the bulk viscosity, η the shear viscosity, and P_h the hydrostatic pressure. $\underline{\tilde{M}}_k^\dagger$ is the Hermitian conjugate of $\underline{\tilde{M}}_k$.

In order to compute the dynamic structure factor $\langle N_k(t)N_{-k}\rangle_{NE}$, and hence the light-scattering intensity, we must generalize Eq. (2.3). In this paper, we shall be interested in the total intensities in each of the Lorentzians in the spectrum and thus may use the regression hypothesis

$$\langle \underline{A}_k(t)\underline{A}_{-k}\rangle_{NE} = e^{\underline{\tilde{M}}_k t}\langle \underline{A}_k \underline{A}_{-k}\rangle_{NE}. \tag{2.10}$$

Other authors[10] have pointed out that this regression equation is not correct and that there should be additional terms on the rhs of (2.10) if we wish to describe the detailed shape of the spectrum. In the case of the dynamic structure factor, these additional terms contribute only to a small renormalization of the sound attenuation coefficient in $e^{\underline{\tilde{M}}_k t}$. This renormalization induces small changes in the shape of the Brillouin peaks of the light-scattering spectrum. Since, in the regime of interest, line shapes are too narrow to observe experimentally, we will not bother to compute the modifications of them here.

III. THE DYNAMIC STRUCTURE FACTOR

In order to evaluate the time-dependent density autocorrelation function, the regression hypothesis, Eq. (2.10), demands that we compute $\langle N_k N_{-k}\rangle_{NE}$, $\langle E_k N_{-k}\rangle_{NE}$, and $\langle \vec{P}_k N_{-k}\rangle_{NE}$. However, as we will discuss below, the contribution from $\langle \vec{P}_k N_{-k}\rangle_{NE}$ is much smaller than the contributions from $\langle N_k N_{-k}\rangle_{NE}$, and $\langle E_k N_{-k}\rangle_{NE}$ and can be ignored for k's in the regime defined by Eq. (1.1).

We now focus our attention on the calculation of $\langle N_k N_{-k}\rangle_{NE}$, $\langle E_k N_{-k}\rangle_{NE}$, and $\langle E_k E_{-k}\rangle_{NE}$, starting from Eqs. (2.5) and (2.8). Since these correlation functions are scalars under rotation, we know that the nonequilibrium correction term in Eq. (2.5) must be the contraction of a second-rank tensor with $\nabla\vec{v}$. The only two second-rank tensors available to us are $\hat{k}\hat{k}$ and the unit tensor $\overline{1}$. Thus, considering only the nonequilibrium term we may write

$$\beta\int_0^\infty dt \exp(\underline{\tilde{M}}_k t)\,\underline{\bar{\bar{\Gamma}}}(k)\exp(\underline{\tilde{M}}_k t):\ \nabla\vec{v}$$
$$= \beta[\underline{\tilde{\alpha}}_1(k)\hat{k}\hat{k} + \underline{\tilde{\alpha}}_2(k)\overline{1}]:\ \nabla\vec{v}. \tag{3.1}$$

The term containing $\underline{\tilde{\alpha}}_2(k)$ couples only to $\nabla\cdot\vec{v}$. In steady states that are not far from equilibrium $\nabla\cdot\vec{v} = 0$ and therefore $\underline{\tilde{\alpha}}_2(k)$ need not be calculated. The left-hand side (lhs) of Eq. (3.1) seems rather complicated. It is a product of three 5×5 matrices and is a second-rank tensor. Its calculation can be greatly simplified if we invoke the relevant symmetries and the orders of magnitude. First, we consider the structure of $\underline{\bar{\Gamma}}(\vec{k})$.

A. The structure of $\Gamma(k)$

We will order the five conserved variables according to $\underline{A}_k = [N_k, E_k, P_k^x, P_k^y, P_k^z]$, and choose the x direction to coincide with that of $\vec{k}$. As N_k, E_k, and I_P are even subject to time reversal, whereas the components of P_k are odd, $\underline{\bar{\Gamma}}(\vec{k})$ is block diagonal:

$$\underline{\bar{\bar{\Gamma}}}(\vec{k}) \equiv \langle \underline{A}_k \underline{A}_{-k}\overline{I}_{P,T}\rangle \sim \begin{bmatrix} X & X & 0 & 0 & 0\\ X & X & 0 & 0 & 0\\ 0 & 0 & X & X & X\\ 0 & 0 & X & X & X\\ 0 & 0 & X & X & X\end{bmatrix}. \tag{3.2}$$

Since $\underline{\bar{\bar{\Gamma}}}$ is an equal-time equilibrium correlation function, it can be evaluated in the $k\to 0$ limit. Since the equilibrium state is rotationally invariant, the upper left block of $\underline{\bar{\bar{\Gamma}}}(k\to 0)$ is a unit tensor in the indices that couple to $\nabla\vec{v}$, so this block contributes only to $\underline{\tilde{\alpha}}_2(k)$. The lower right block of $\underline{\bar{\bar{\Gamma}}}(\vec{k})$ is of the form $\langle\vec{P}\vec{P}\overline{I}_{P,T}\rangle$. Using Eq. (2.7) we may write this explicitly as

$$\langle\vec{P}\vec{P}\overline{I}_{P,T}\rangle = \langle\vec{P}\vec{P}\overline{\tau}\rangle - \langle\vec{P}\vec{P}\hat{E}\rangle\left(\frac{\partial P_h}{\partial e}\right)_n\overline{1}$$
$$-\langle\vec{P}\vec{P}\hat{N}\rangle\left(\frac{\partial P_h}{\partial n}\right)_e\overline{1} - \langle\vec{P}\vec{P}\rangle P_h\overline{1}. \tag{3.3}$$

The subtracted terms couple to $\nabla\vec{v}$ as unit tensors and thus contribute only to $\underline{\alpha}_2$. Thus, only $\langle\vec{P}\vec{P}\overline{\tau}\rangle$ contributes to $\underline{\alpha}_1(k)$ and we evaluate this using thermodynamic derivatives in an ensemble with a mean velocity v,

$$\Gamma_{rs}^{ij} \equiv \langle P^i P^j \hat{\tau}^{rs}\rangle = \frac{\partial}{\beta\partial v_i}\langle P^j\hat{\tau}^{rs}\rangle_v\Big|_{v=0}$$
$$= \frac{1}{\beta^2}\,\frac{\partial^2\langle\hat{\tau}^{rs}\rangle_v}{\partial v_i\partial v_j}\Big|_{v=0}, \tag{3.4}$$

Since

$$\langle\hat{\tau}^{rs}\rangle_v = mnv_r v_s, \tag{3.5}$$

$$\Gamma_{rs}^{ij} = (k_B T)^2 mn(\delta_{ir}\delta_{js} + \delta_{is}\delta_{jr}). \tag{3.6}$$

Finally, notice that $\underline{\tilde{M}}_k$ does not couple N_k (or E_k) to the transverse components of momentum, P_k^y or P_k^z, so that the computation of $\langle N_k N_{-k}\rangle_{NE}$, $\langle E_k N_{-k}\rangle_{NE}$, and $\langle E_k E_{-k}\rangle_{NE}$ involves only Γ_{xx}^{xx}. The corresponding values of $\alpha_1(k)$ are

$$\alpha_1^{NN}(k)=2(k_BT)^2mn\int_0^\infty dt(e^{M_kt})_{NP^x}(e^{M_k^\dagger t})_{P^xN}\,, \tag{3.7a}$$

$$\alpha_1^{EN}(k)=2(k_BT)^2mn\int_0^\infty dt(e^{M_kt})_{EP^x}(e^{M_k^\dagger t})_{P^xN}\,, \tag{3.7b}$$

and

$$\alpha_1^{EE}(k)=2(k_BT)^2mn\int_0^\infty dt(e^{M_kt})_{EP^x}(e^{M_k^\dagger t})_{P^xE}\,.$$

The matrix $e^{\tilde{M}_kt}$ was computed in III[2] and is given in the Appendix of this paper. The integrals in Eqs. (3.7) are straightforward and lead to the following results:

$$\frac{\langle N_kN_{-k}\rangle_{NE}}{V}=\frac{\langle N_kN_{-k}\rangle}{V}-\frac{1}{2}\frac{n}{k^2\Gamma_s}\frac{k_BT}{mc_0^2}\hat{k}\hat{k}:\nabla\vec{v}\,, \tag{3.8a}$$

$$\frac{\langle E_kN_{-k}\rangle_{NE}}{V}=\frac{\langle E_kN_{-k}\rangle}{V}-\frac{1}{2}\frac{h}{k^2\Gamma_s}\frac{k_BT}{mc_0^2}\hat{k}\hat{k}:\nabla\vec{v}\,, \tag{3.8b}$$

and

$$\frac{\langle E_kE_{-k}\rangle_{NE}}{V}=\frac{\langle E_kE_{-k}\rangle}{V}-\frac{1}{2}\frac{h^2}{nk^2\Gamma_s}\frac{k_BT}{mc_0^2}\hat{k}\hat{k}:\nabla\vec{v}\,, \tag{3.8c}$$

where c_0 is the adiabatic sound speed, Γ_s is the sound attenuation coefficient, h is the enthalpy density, n is the number density, and m is the mass of a particle.

The dissipative contribution to $\langle P_k^xN_{-k}\rangle_{NE}$ can be computed in the same way as we have computed $\langle N_kN_{-k}\rangle_{NE}$ and $\langle E_kN_{-k}\rangle_{NE}$. In place of the integrals of Eq. (3.7) we would then have $\int_0^\infty dt(e^{M_kt})_{P^xP^x}\times(e^{M_k^\dagger t})_{P^xN}$. Evaluating this integral we find that the largest terms are proportional to $1/kc_0$. Thus $\langle P_k^xN_{-k}\rangle_{NE}$ is smaller by a factor of $c_0/k\Gamma_s$ than the NN, EE, and EN nonequilibrium corrections and may be neglected for k's given by Eq. (1.1).

B. The dynamic structure factor

The calculation of the time-dependent density autocorrelation function is accomplished according to Eq. (2.10):

$$\langle N_k(t)N_{-k}\rangle_{NE}=(e^{M_kt})_{NN}\langle N_kN_{-k}\rangle_{NE}+(e^{M_kt})_{NE}\langle E_kN_{-k}\rangle_{NE}\,. \tag{3.9}$$

This equation holds only for $t>0$. The extension to negative times is done on the basis of the stationarity of the time correlation functions in the NESS that was proved in paper I,[1] Sec. IV, which implies that

$$\langle \underline{A}_k(-t)\underline{A}_{-k}\rangle_{NE}=\langle \underline{A}_k(t)\underline{A}_{-k}\rangle_{NE}^\dagger\,. \tag{3.10}$$

Using Eqs. (3.8), (3.9), and (3.10) we can evaluate $\langle N_k(t)N_{-k}\rangle_{NE}$ easily. Performing the Fourier transform according to the definition

$$S_{k\omega}\equiv\int_{-\infty}^\infty dt\,e^{-i\omega t}\,\frac{\langle N_k(t)N_{-k}\rangle_{NE}}{V}\,,$$

we obtain the final result

$$S_{k\omega}=\left[\frac{2k_BTn}{mc_0^2}\left(\frac{c_p}{c_v}-1\right)\frac{\Gamma_Tk^2}{\omega^2+(\Gamma_Tk^2)^2}\right]+\frac{2k_BTn\Gamma_sk^2}{mc_0^2}\times\left(\frac{1-\epsilon}{(\omega-kc_0)^2+(\Gamma_sk^2)^2}+\frac{1-\epsilon}{(\omega+kc_0)^2+(\Gamma_sk^2)^2}\right), \tag{3.11}$$

where

$$\epsilon=\frac{c_0}{2k^2\Gamma_s}\hat{k}\hat{k}:\frac{\nabla\vec{v}}{c_0} \tag{3.12}$$

and Γ_T is the thermal diffusion constant. The quantity ϵ contains the nonequilibrium contribution to the structure factor.

We recall that Eq. (3.11) is correct for the total intensities of the Brillouin peaks but does not contain the changes in line shapes due to a renormalization of Γ_s. We emphasize that these latter effects are not experimentally accessible.

Because of our choice of $\vec{v}(\vec{r})$ in writing Eq. (2.6), our result for $S_{k\omega}$ is valid for a point in the fluid that is at rest. $S_{k\omega}$ is transformed to a moving point in the fluid by replacing ω by $\omega+\vec{k}\cdot\vec{v}$, where $\vec{v}$ is the velocity of the fluid at the point where $S_{k\omega}$ is required.

To demonstrate the effects of a velocity gradient on the spectrum of scattered light, we envision scattering a laser beam from a dilute gas with a steady, approximately linear, shear. Such a shear can be achieved by rotating the outer of two concentric cylinders with the fluid contained between them. The light-scattering geometry is depicted in Fig. 1. The velocity $\vec{v}$ is parallel to the x axis and the velocity gradient ∇v is in the y direction. The incident beam is in the positive z direction. We choose two scattering angles that share a small polar angle but differ in their azimuthal angles (in the $\vec{v}$-∇v plane) by 90°. The $\vec{k}$ vectors $\vec{k}^{(1)}$ and $\vec{k}^{(2)}$ lie in the $\vec{v}$-∇v plane and have $k_x^{(1)}=-k_x^{(2)}$, $k_y^{(1)}=k_y^{(2)}$, $|k_x^{(1,2)}|=|k_y^{(1,2)}|$. With this geometry, the only contribution to ϵ arises from

$$\epsilon=\pm\frac{c_0}{4k^2\Gamma_s}\frac{\partial}{\partial y}\left(\frac{v_x}{c_0}\right), \tag{3.13}$$

with the plus sign for $\vec{k}^{(1)}$ and the minus sign for $\vec{k}^{(2)}$. The resulting spectra are shown in Fig. 2. The Brillouin components associated with $\vec{k}^{(1)}$ are diminished compared to the equilibrium spectrum,

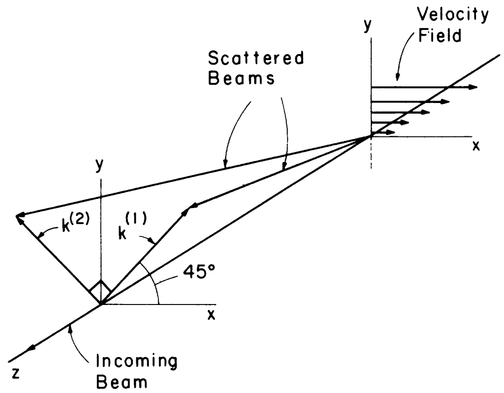

FIG. 1. Geometry for scattering light from a linear shear. The velocity $\vec{v}$ is parallel to the x axis and the velocity gradient ∇v is in the y direction. The incident beam is in the positive z direction. We choose two scattering angles that share a small polar angle but differ in their azimuthal angles (in the $\vec{v}$-$\nabla\vec{v}$ plane) by 90°. The $\vec{k}$ vectors $\vec{k}^{(1)}$ and $\vec{k}^{(2)}$ lie in the $\vec{v}$-$\nabla\vec{v}$ plane and have $k_x^{(1)}=-k_x^{(2)}$, $k_y^{(1)}=k_y^{(2)}$, $|k_x^{(1,2)}|=|k_y^{(1,2)}|$.

whereas those associated with $\vec{k}^{(2)}$ are enhanced.

Notice that the Rayleigh peak is not affected. As a result, the Landau-Placzek ratio between the intensities of the Rayleigh and Brillouin components no longer holds. This ratio becomes k dependent and will change according to the scattering geometry. This finding indicates that the pair distribution function has a long-range part, as is discussed in Sec. V.

Under what conditions can we expect a shear to produce observable changes in the light-scattering spectrum? We will examine this question for the case of a dilute gas. It will be clear from the discussion that the effect will be less pronounced for dense fluids than for dilute gases. For a dilute atomic gas $\gamma\equiv c_p/c_v=\frac{5}{3}$ and the Prandtl number $P=\nu/\Gamma_T=\frac{2}{3}$; thus

$$\Gamma_s=\tfrac{1}{2}[(\gamma-1)/P+\tfrac{4}{3}]\nu=\tfrac{7}{6}\nu, \tag{3.14}$$

where ν is the kinematic viscosity. Letting v_0 be

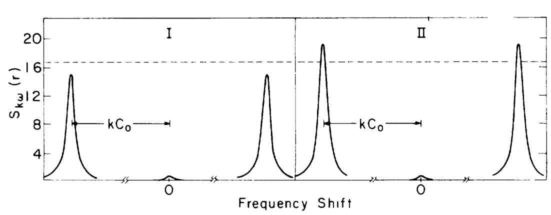

FIG. 2. A schematic spectrum of light scattered from a fluid with a linear shear. In panel I $\hat{k}\hat{k}$: $(\nabla\vec{v}/c_0)$ is positive whereas in panel II it is negative. The dashed line marks the height of the Brillouin components in the same system at equilibrium. Notice that the Landau-Placzek ratio no longer holds.

the velocity difference between the two cylinders, we can express ϵ in terms of the Reynold's number

$$R\equiv\frac{v_0L}{\nu}=\frac{7}{6}\frac{v_0L}{\Gamma_s}, \tag{3.15}$$

where L is the difference between the radii of the two cylinders. Writing Eq. (3.13) in terms of R gives

$$|\epsilon|=\frac{3}{14}\frac{R}{k^2L^2}. \tag{3.16}$$

Since laminar flow cannot be maintained in this geometry[11] much above $R=10^3$ and since L must be a macroscopic length, we may have difficulty choosing a k that satisfies Eq. (1.1) and makes $|\epsilon|$, say, 10%.

The domain of k vectors for which the theory is valid is bounded from below by considerations of the dimensions of the system. It is bounded from above by the constraint that the modes we are observing be weakly damped. Since here k refers to the wave vector of sound modes whose decay length is $c_0/2k^2\Gamma_s$ we must have

$$A(c_0/2k^2\Gamma_s)\leq L/2, \tag{3.17}$$

with A at least 3 in order to justify ignoring boundary effects. In a dilute monoatomic gas

$$\Gamma_s=a(k_BT/m)^{1/2}l, \tag{3.18}$$

where l is the mean-free path and $a\approx 1$. The adiabatic sound speed is given by

$$c_0=(\gamma k_BT/m)^{1/2}. \tag{3.19}$$

Thus the lower bound on k is

$$k\geq\left[\frac{A}{lL}\left(\frac{\gamma^{1/2}}{a}\right)\right]^{1/2}. \tag{3.20}$$

The upper bound on k is given by

$$c_0k\geq k^2\Gamma_sB, \tag{3.21}$$

where B should be at least 10 in order to ensure the validity of ignoring $1/k$ terms relative to $1/k^2$ terms as we have done. This gives

$$k\leq\frac{1}{lB}\left(\frac{\gamma^{1/2}}{a}\right). \tag{3.22}$$

Notice that this bound also ensures the validity of a hydrodynamic treatment. Combining Eqs. (3.16), (3.20), and (3.22) we find an upper bound for ϵ:

$$|\epsilon|\leq\frac{R}{4A^2B^2}. \tag{3.23}$$

By letting all the inequalities become equalities we can obtain $|\epsilon|\sim 0.1$ by choosing

$$L=0.06\text{ cm},\quad R=10^3,\qquad k=800\text{ cm}^{-1},\quad l=1\times10^{-4}\text{ cm}. \tag{3.24}$$

Unfortunately these parameters correspond to a gas pressure of roughly $\frac{1}{10}$ atmospheric pressure, at scattering angles less than 1°, and tangential velocities of the cylinder walls on the same order as thermal velocities. Thus, the experiment may be better suited to computer simulation than physical realization.

IV. EQUAL-TIME MOMENTUM AUTOCORRELATION FUNCTION

In this section we compute $\langle \vec{P}_k \vec{P}_{-k}\rangle_{NE}$, the equal-time momentum autocorrelation function, in the rest frame of the fluid. In the case of the corrections to the NN and NE static correlation function, we found contributions due only to sound modes in the fluid. This is evidenced by the factor $1/k^2\Gamma_s$ in the dissipative terms. In the case of $\langle \vec{P}\vec{P}\rangle_{NE}$ the correction term will be due to both sound modes and shear modes. Since $\langle \vec{P}\vec{P}\rangle_{NE}$ is a second-rank tensor, we will encounter greater difficulties in determining the tensorial form of the correction terms. Apart from this, however, the computation is analogous to the one presented in Sec. III for the static NN and EN correlation functions.

We showed in Sec. III that the upper left 2×2 block of Γ can be ignored since it produces terms proportional to $\vec{\nabla}\cdot\vec{v}$ which vanish in the steady state. Thus, when we apply Eqs. (2.6) and (2.8) to the computation of $\langle \vec{P}\vec{P}\rangle_{NESS}$ only the momentum-momentum entries of Γ and $(e^{M_k t})$ will enter the calculation. This allows us to simplify the notation by using lower case letters to indicate momentum components:

$$\frac{\langle P^i P^j\rangle_{NE}}{V}=\frac{\langle P^i P^j\rangle}{V}-\beta\int_0^\infty (e^{Mt})_{ir}\Gamma^{rs}_{lp}(e^{M^+t})_{sj}\frac{\partial v_l}{\partial r_p}\,. \tag{4.1}$$

All k dependences have been suppressed and summation over repeated indices is implied. The symmetry of Γ^{rs}_{lp} [cf. Eq. (3.6)] under interchange of l and p implies that only the symmetric part of ∇v survives contraction with Γ. Defining the shear tensor

$$S_{ij}\equiv\frac{1}{2}\left(\frac{\partial v_i}{\partial r_j}+\frac{\partial v_j}{\partial r_i}\right), \tag{4.2}$$

we may effect the contraction of Γ and ∇v to obtain

$$\frac{\langle P^i P^j\rangle_{NE}}{V}=\frac{\langle P^i P^j\rangle}{V}-2(k_BT)mn\int_0^\infty dt(e^{Mt})_{il}(e^{M^+t})_{pj}S_{lp}\,. \tag{4.3}$$

Since $(e^{Mt})_{il}$ couples two vectors, it must be a second-rank tensor. Since the only two second-rank tensors available to us are $\vec{k}\vec{k}$ and $\vec{1}$, we can write $(e^{Mt})_{il}$ in the form

$$(e^{M_k t})_{il}=A(t)\frac{k_ik_l}{k^2}+B(t)\delta_{il}\,. \tag{4.4}$$

In the Appendix we show that

$$A(t)=\tfrac{1}{2}(e^{\xi_+t}+e^{\xi_-t})-e^{-k^2\nu t},$$
$$B(t)=e^{-k^2\nu t}, \tag{4.5}$$

where $\xi_\pm=\pm ikc_0-k^2\Gamma_s$, and ν is the kinematic viscosity. Combining Eqs. (4.3)–(4.5), performing the integrations, and keeping only terms of order $1/k^2$, we obtain the following result for the momentum autocorrelation function:

$$\frac{\langle\vec{P}\vec{P}\rangle_{NE}}{V}=\frac{\langle\vec{P}\vec{P}\rangle}{V}-k_BTmn\left(\frac{1}{2k^2\Gamma_s}\hat{k}\hat{k}\hat{k}\hat{k}:\vec{S}-\frac{1}{k^2\nu}(\hat{k}\hat{k}\cdot\vec{S}+\vec{S}\cdot\hat{k}\hat{k}-\vec{S}-\hat{k}\hat{k}\hat{k}\hat{k}:S)\right). \tag{4.6}$$

Using the identity $\vec{A}\times(\vec{B}\times\vec{C})=\vec{A}\cdot\vec{C}\vec{B}-\vec{A}\cdot\vec{B}\vec{C}$, we may rewrite Eq. (4.6) in the form

$$\langle\vec{P}\vec{P}\rangle=k_BTmn\left(\vec{1}-\frac{1}{2k^2\Gamma_s}\hat{k}\hat{k}\cdot\vec{S}\cdot\hat{k}\hat{k}-\frac{1}{k^2\nu}\hat{k}\times(\hat{k}\times\vec{S}\times\hat{k})\times\hat{k}\right). \tag{4.7}$$

V. DISCUSSION

We have shown in this paper that laminar shear flow in a fluid induces interesting changes in the fluctuations of the hydrodynamic variables. The static correlation functions that are strongly affected are the scalars $\langle N_k N_{-k}\rangle_{NE}$, $\langle N_k E_{-k}\rangle_{NE}$, and $\langle E_k E_{-k}\rangle_{NE}$ and the second-rank tensor $\langle \vec{P}_k\vec{P}_{-k}\rangle_{NE}$. This is in contrast with our observation that in a fluid with a heat flux it is the vector static correlation functions such as $\langle \vec{P}_k N_{-k}\rangle_{NE}$ that are strongly affected. The conclusion is that, when the product of the time reversal signatures of two variables is the same as the signature of the dissipative flux, then the correlation function of those variables is significantly changed.

The nonequilibrium contributions to the static correlation functions have a $1/k^2$ dependence. This is, of course, equivalent to a $1/r$ dependence in physical space, which is an indication of a long-range order. We remind the reader, however, that our theory is limited at the present time by Eq. (1.1). Thus the $1/k^2$ dependence cannot be taken as a divergence for $k\to 0$ and the $1/r$ dependence is valid for intermediate distances. This growth of the correlation length has particularly interesting implications for the properties of the light-scattering spectrum. As was found above, the total scattering intensity becomes k dependent and the Landau-Placzek ratio does not hold. To see the relation of this to the correlation length, we recall that the total scattering intensity is proportional to $\langle N_k N_{-k}\rangle_{NE}$. In equilibrium this quantity is given by

$$\langle N_k N_{-k}\rangle = \rho + \rho^2 \int [g^{(2)}(\vec{r}-\vec{r}\,') - 1] \times e^{i\vec{k}\cdot(\vec{r}-\vec{r}\,')} d(\vec{r}-\vec{r}\,'),$$

where $g^{(2)}(r - r')$ is the pair correlation function. For a small k we can expand the exponent in a Taylor series to obtain

$$\langle N_k N_{-k}\rangle = \rho + \rho^2 \int [g^{(2)}(\vec{r}-\vec{r}\,') - 1] \times [1 - \tfrac{1}{2}k^2(\vec{r}-\vec{r}\,')^2] d(\vec{r}-\vec{r}\,'),$$

where the linear term in the expansion is dropped by symmetry. If the correlation length is small, the k-dependent term contributes a negligible correction and the static correlation function is k independent (i.e., purely thermodynamic).

This example can be generalized; as long as the correlation length is small, static correlations may be computed in the $k=0$ limit, which yields a purely thermodynamic quantity. This conclusion pertains in particular to the static correlations of the heat and sound modes, respectively, which determine the ratio of intensities of the Brillouin and Rayleigh peaks in an equilibrium system. This is why in an equilibrium system the Landau-Placzek ratio $(c_p/c_v - 1)$ is purely thermodynamic and has no k dependence.

Once the correlation length grows, however, the above argument breaks down, the k dependence of the static correlation functions becomes important and the Landau-Placzek ratio does not hold.

What are the physical processes underlying the long-range behavior of these correlation functions? To understand these processes it is easiest to think in terms of the eigenmodes[6] of the hydrodynamic equations linearized around equilibrium. For each wave vector there are five modes: two longitudinal-momentum (sound) modes, two transverse-momentum (shear) modes, and one heat mode. We will see that the presence of a velocity gradient modifies the intensities of the fluctuating sound modes and shear modes.

First, consider thermally generated sound. Figure 3 shows a small box in the fluid fixed with respect to the laboratory frame in which we will estimate the intensity of sound propagating through various faces of the box. Let us first assume that

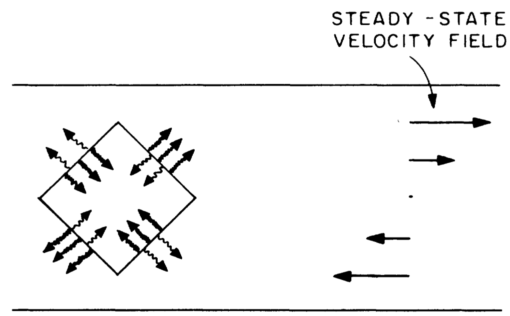

FIG. 3. Propagation of sound waves (wavy lines) across the faces of a small box in a linear shear.

the fluid is in local equilibrium and then see how the velocity gradient perturbs this. In local equilibrium, the intensity of sound entering each face of the box is given by the sound velocity at that face times the local sound intensity. Since the fluid has a homogeneous temperature (neglecting a small quadratic temperature profile) the local sound intensity is uniform. However, the local sound velocity is a sum of the equilibrium velocity c_0 and the local velocity $\vec{v}(\vec{r})$.

Sound waves entering the lower-right and upper-left faces of the box have a higher speed than c_0, whereas those leaving the same faces of the box have a lower speed than c_0. The effect of this is that the sound intensity in the box is constantly increasing over its equilibrium value. The balance between this increase which is proportional to $\hat{k}\hat{k}:\nabla\vec{v}$ and the return to equilibrium at the rate $2k^2\Gamma_s$ determine the nonequilibrium intensity of sound in the box with wave vector $\hat{k}$. If we considered sound going between the other two faces of the box, the opposite effect would have resulted and the sound intensity would be less than in equilibrium. It is the $(1/k^2\Gamma_s)\hat{k}\hat{k}:\nabla\vec{v}$ behavior in the random sound intensity which induces similar behavior in the correlation functions $\langle NN\rangle_{NE}$, $\langle NE\rangle_{NE}$, $\langle EE\rangle_{NE}$, and $\langle P^x P^x\rangle_{NE}$.

Notice that it is the sound modes which carry momentum in the same direction as the dissipative momentum flux (proportional to ν) which are enhanced. Conversely, sound modes which carry momentum in the opposite direction are depleted. Thus, we find that the long-wavelength thermally generated sound positively renormalizes the viscosity of the system. This is a general property of all of the physical processes underlying the $1/k^2$ behavior of static correlations in nonequilibrium systems. In the momentum conducting steady state these processes enhance the viscosity of the system and in the heat conducting steady state they enhance the thermal conductivity of the system. In systems of dimensionality greater than two this renormalization is very small.

To understand the $1/k^2$ behavior of the fluctuating transverse momentum, consider Fig. 4. The dots represent lines of vorticity directed out of the page, whereas the crosses represent lines of vorticity going into the page. The whole array represents a thermally excited shear mode whose wave vector is 45° to both the velocity and the velocity gradient. Now, imagine how this pattern is affected by the velocity field. Since the vorticity is perpendicular to the velocity, it is merely convected along and so after a time Δt the array has been changed from that of Fig. 4(a) to that of Fig. 4(b). This causes a change in the wave vector $\hat{k}$ but, most importantly, it causes an increase in the amplitude of the shear mode because the dots and crosses on their respective lines have been moved closer together. The Kelvin circulation theorem tells us that the line integral of the velocity around each dot and cross is constant except for the reduction due to viscosity. Thus, as the dots and crosses move together, the velocity perturbation perpendicular to $\hat{k}$ is amplified. The balance between this amplification and the decay of transverse momentum back to equilibrium produces the $1/k^2\nu\, \hat{k}\cdot(\hat{k}\cdot \overleftrightarrow{S}\cdot\hat{k})\cdot\hat{k}$ contribution in $\langle PP\rangle$. Again, this process positively renormalizes the shear viscosity of the system. This amplification of shear modes was first discussed by Orr in a 1907 paper.[12] These same processes near walls cause the slipping length in fluids to have a weak divergence.[13]

We have suggested an experiment to test our predictions. The idea is to scatter light from a steady state system with a linear shear and observe the spectrum associated with the wave vectors that have different orientations with respect to the velocity field and its gradient. The prediction is that the ratio of the intensities of the Rayleigh and Brillouin components will be different for the two wave vectors. We find that the onset of turbulence constrains the experiment to small shears and thus a significant effect can be seen only in a dilute gas.

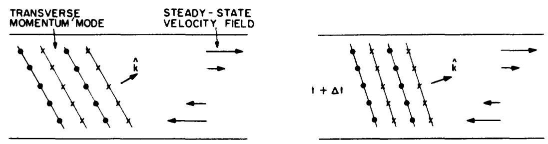

FIG. 4. Evolution of a transverse momentum mode in a linear shear. Dots represent lines of vorticity directed out of the plane, while crosses represent vorticity lines into the plane.

The findings with respect to the light-scattering spectrum are related to our investigation of the fluctuation-dissipation theorem in NESS. In IV[9] we argued that the total dissipation associated with a small perturbation of a NESS relates to the local equilibrium part of the appropriate correlation function, and not to the true NESS correlation function. In the context of light scattering, this statement means that the total scattering amplitude should be independent of the new, dissipative terms found above for the dynamic structure factor. Examining Eqs. (3.11) and (3.12), we see that this is indeed the case. The total scattering is proportional to the integral of $S_{k\omega}$ over all $\vec{k}$ and ω. The contributions to ϵ have a quadrupole form in $\vec{k}$ space and thus their integral vanishes as is predicted by the modified fluctuation-dissipation relation.

Note added in proof. Some of the results presented here have appeared in a paper by Onuki.[14]

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APPENDIX

In Appendix A of III we computed $e^{\tilde{\underline{M}}_k t}$. Here we rewrite that result for the couplings between N_k, E_k, and P_k^x:

$$\exp(\tilde{\underline{M}}_k^t\sigma)=\frac{1}{2c_0^2}\begin{bmatrix}(e^{\xi_+\sigma}+e^{\xi_-\sigma})\chi_n/m+2e^{\xi_T\sigma}\chi_e h/mn, & \chi_e(e^{\xi_+\sigma}+e^{\xi_-\sigma}-2e^{\xi_T\sigma})/m, & c_0(e^{\xi_+\sigma}-e^{\xi_-\sigma})/m\\ \chi_n h(e^{\xi_+\sigma}+e^{\xi_-\sigma}-2e^{\xi_T\sigma})/mn, & \chi_e h(e^{\xi_+\sigma}+e^{\xi_-\sigma})/mn+2\chi_n e^{\xi_T\sigma}/m, & c_0 h(e^{\xi_+\sigma}-e^{\xi_-\sigma})/mn\\ c_0\chi_n(e^{\xi_+\sigma}-e^{\xi_-\sigma}), & c_0\chi_e(e^{\xi_+\sigma}-e^{\xi_-\sigma}), & c_0^2(e^{\xi_+\sigma}+e^{\xi_-\sigma})\end{bmatrix}, \qquad \text{(A1)}$$

where

$$\xi_\pm(k)\simeq \pm ikc_0-k^2\Gamma_s$$

and (A2)

$$\xi_T(k)=-k^2\Gamma_T\,,$$

with

$$\Gamma_T\equiv\lambda/nc_p$$

and (A3)

$$\Gamma_s\equiv\tfrac{1}{2}[(c_p/c_v-1)\Gamma_T+\nu_l]\,.$$

We also need the tensor form of the momentum-momentum couplings in $e^{\tilde{\underline{M}}_k t}$. In terms of P_k^x, P_k^y, and P_k^x with $\hat{x}=\hat{k}$, we find

$$e^{M_k t}=\begin{bmatrix} \frac{1}{2}(e^{\xi_+ t}+e^{\xi_- t}) & 0 & 0 \\ 0 & e^{-k^2\nu t} & 0 \\ 0 & 0 & e^{-k^2\nu t}\end{bmatrix}, \qquad (A4)$$

with ν the kinematic shear viscosity. Since the only two second-rank tensors available are $k_i k_j$ and δ_{ij}, we must be able to write this in the form

$$e^{M_k t}_{il}=A(t)k_i k_l/k^2+B(t)\delta_{il}\,, \qquad (A5)$$

where

$$A(t)=\tfrac{1}{2}(e^{\xi_+ t}+e^{\xi_- t})-e^{-k^2\nu t}$$

and (A6)

$$B(t)=e^{-k^2\nu t}\,.$$

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