Thermal fluctuations in the presence of two dissipative steady-state currents

Claude Tremblay and A.-M.S. Tremblay
Département de Physique, Université de Sherbrooke, Sherbrooke, Québec J1K 2R1, Canada
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Nonlinear corrections to the density-density correlation function are calculated for a nonturbulent fluid subjected to both a temperature gradient and a shear flow. Linear corrections vanish in the chosen geometry.

I. INTRODUCTION

Considerable interest in fluctuations about hydrodynamic dissipative steady states has been generated by the recent prediction[1] and observation[2] of long-range correlations in a noncritical fluid carrying a constant heat current. These long-range correlations are experimentally observable as an asymmetry in the small-angle Brillouin light scattering spectrum. Numerous methods of calculations have led to the same theoretical results.[3-5] One of these methods is a simple extension of the well-known Langevin formalism for fluctuations about equilibrium.[4] The problems studied up to now have concentrated on corrections to the equilibrium fluctuations which are proportional to the first power of the externally imposed temperature gradient or shear velocity. In that regime, a straightforward generalization of the Langevin equilibrium formalism is physically justified.[4] To nonlinear order, however, there exists at least one calculation[6] which shows that the Langevin formalism gives results that differ by a few percent from microscopic calculations. The reason for the discrepancy is related to the fact that, to nonlinear order, the direction-independent part of the microscopic velocity distribution function does not have a local-equilibrium form. Unfortunately the above-mentioned calculation applies to an electron gas scattering off impurities and is very hard to check experimentally.

In this paper, we use the Langevin formalism to compute nonlinear corrections to the density fluctuations which can, in principle, be observed by light scattering. Some authors[7] have calculated nonlinear corrections of second and higher order in an applied temperature gradient[8] but have neglected many parasitic effects, such as the influence of boundaries and the temperature dependence of the dielectric constant, which to that order influence the observations.[4] We propose instead to study a fluid in the presence of both a shear and a temperature gradient. If $\vec{\nabla}T$ is the temperature gradient, $\vec{\nabla}\vec{v}_0$ the shear tensor, and $\hat{k}$ a unit vector in the direction of the scattering wave vector, then $\vec{\nabla}T\cdot\vec{\nabla}\vec{v}_0\cdot\hat{k}$ is one of the scalars to which the corrections to the light scattering spectrum may be proportional. Contributions of that type to the light scattering spectrum are extremely interesting because none of the extraneous effects mentioned above contribute to that order. Furthermore, it is possible to choose a geometry for which the linear corrections, proportional to $\hat{k}\cdot\vec{\nabla}T$ and $\hat{k}\cdot\vec{\nabla}\vec{v}_0\cdot\hat{k}$, both vanish while the nonlinear corrections are finite. The physical interpretation of the results and discussion of possible experiments are presented at the end of this paper. The integrated intensity for the light scattering spectrum is most easily calculated from the Einstein relation. This is done in the Appendix. The steady-state problem is solved in Sec. II and the density fluctuations are calculated in Sec. III. The reader is referred to Ref. 4 for a very detailed discussion of the methods we use.

II. STEADY STATE

We start from the full nonlinear hydrodynamic equations[9]

$$\frac{\partial\rho}{\partial t}+\vec{\nabla}\cdot(\rho\vec{v})=0\,, \tag{1}$$

$$\frac{\partial v_i}{\partial t}+v_j\nabla_j v_i=-\frac{1}{\rho}\nabla_i p+\frac{1}{\rho}\nabla_j[\eta(\nabla_i v_j+\nabla_j v_i-\tfrac{2}{3}\vec{\nabla}\cdot\vec{v}\delta_{ij})+\xi\vec{\nabla}\cdot\vec{v}\delta_{ij}]\,, \tag{2}$$

$$\rho T\left(\frac{\partial s}{\partial t}+(\vec{v}\cdot\vec{\nabla})s\right)=\vec{\nabla}\cdot(\kappa\vec{\nabla}T)+\frac{\eta}{2}(\nabla_i v_j+\nabla_j v_i-\tfrac{2}{3}\vec{\nabla}\cdot\vec{v}\delta_{ij})^2+\xi(\vec{\nabla}\cdot\vec{v})^2\,, \tag{3}$$

where ρ is the mass density, $\vec{v}$ the velocity, s the entropy per unit mass, p the pressure, T the temperature, κ the thermal conductivity, and η and ξ, respectively, the shear and bulk dynamic viscosities. The stationary state may be found by setting the time derivatives equal to zero and applying the boundary conditions. We take a system which is bounded in the y direction and may be considered infinite in the other two directions. The fluid may flow in the x direction. We choose the following boundary conditions:

$$T(x, y=\pm L/2, z)=T_0 \pm \Delta T/2\,, \tag{4}$$

$$v_x(x, y=\pm L/2, z)=v_0 \pm \Delta v_0/2\,, \tag{5}$$

$$v_y = v_z = 0\,. \tag{6}$$

Neglecting the viscous heating terms, assuming that $(\partial \ln\kappa/\partial \ln T)(\Delta T/T) \ll 1$ and using the fact that the velocity field is perpendicular to $\vec{\nabla} s$ we find from Eqs. (3) and (4) a linear temperature profile:

$$T(\vec{r}) = T_0 + \frac{y}{L}\Delta T \approx T_0 + \delta\tilde{T} \sin\vec{q}_2 \cdot \vec{r}\,. \tag{7}$$

The latter equality will be useful to calculate correlation functions in Fourier space. We will always work in the limit where $\vec{q}_2 \cdot \vec{r} \ll 1$. Hence, $\vec{q}_2 \delta\tilde{T} = \vec{\nabla} T = \hat{e}_y(\Delta T/L)$. Similarly, from Eqs. (1) and (2) and the boundary conditions (5) and (6) we find that the pressure is a constant and that

$$\vec{v}_0(\vec{r}) = [v_0 + \Delta v_0(y/L)]\hat{e}_x = \vec{V}_0 + \tilde{\vec{v}}_0 \sin\vec{q}_1 \cdot \vec{r}\,. \tag{8}$$

Here also we will only use the limit $\vec{q}_1 \cdot \vec{r} \ll 1$ and make the identification $\vec{q}_1 \tilde{\vec{v}}_0 = \vec{\nabla}\vec{v}_0 = \hat{e}_y \hat{e}_x (\Delta v_0/L)$. Note that because $\vec{\nabla} T \cdot \vec{\nabla}\vec{v}_0 \neq 0$ in Eq. (2), the linear velocity profile (8) is a good approximation only if $(\partial \ln\eta/\partial \ln T)(\Delta T/T) \ll 1$. Note that $\Delta T/T \ll 1$ implies that $(\nabla \ln T)^{-1} \gg L$, where L is the system size. This was one of the restrictions also imposed in Ref. 4.

III. FLUCTUATIONS

To find the fluctuations, we linearize the hydrodynamic equations around the steady state and include Langevin-force terms. We find

$$\frac{\partial \delta\rho}{\partial t} + \vec{v}_0 \cdot \vec{\nabla}\delta\rho + \rho \vec{\nabla}\cdot \delta\vec{v} = 0\,, \tag{9}$$

$$\frac{\partial \delta\vec{v}}{\partial t} + \vec{v}_0 \cdot \vec{\nabla}\delta\vec{v} + \delta\vec{v}\cdot \vec{\nabla}\vec{v}_0 = -\frac{1}{\rho}\vec{\nabla}\delta p + \frac{1}{\rho}\nabla_j[\eta(\nabla_j\,\delta\vec{v} + \vec{\nabla}\delta v_j)] + \frac{1}{\rho}\vec{\nabla}[(\xi - \tfrac{2}{3}\eta)\vec{\nabla}\cdot\delta\vec{v}] + \vec{\nabla}\cdot\tilde{\mathrm{S}}\,, \tag{10}$$

$$\frac{\partial \delta s}{\partial t} + \vec{v}_0 \cdot \vec{\nabla}\delta s + \delta\vec{v}\cdot\vec{\nabla} s = \frac{1}{\rho T}\vec{\nabla}\cdot(\kappa \vec{\nabla}\delta T) + \frac{1}{\rho T}\vec{\nabla}\cdot\vec{g}\,, \tag{11}$$

where $\tilde{\mathrm{S}}$ and $\vec{g}$ are, respectively, the fluctuating stress tensor and heat flux.[9] In Eqs. (9) to (11) we must use the local steady-state value of all transport coefficients and nonfluctuating thermodynamic quantities. To close the set of equations (9) to (11) we need the thermodynamic relations

$$\delta p = \left(\frac{\partial p}{\partial \rho}\right)_s \delta\rho + \left(\frac{\partial p}{\partial s}\right)_\rho \delta s\,, \tag{12}$$

$$\delta T = \left(\frac{\partial T}{\partial \rho}\right)_s \delta\rho + \left(\frac{\partial T}{\partial s}\right)_\rho \delta s\,. \tag{13}$$

If $(\partial V/\partial T)_p = 0$, then $(\partial p/\partial s)_\rho = 0$ and $(\partial T/\partial \rho)_s = 0$. Thus, to zeroth order in $\delta\tilde{T}$ the entropy equation (11) decouples from the other fluctuating variables. As demonstrated in Ref. 4, this remains true to first order in $\delta\tilde{T}$ and $\tilde{v}_0$. This is also true to order $\tilde{v}_0\delta\tilde{T}$. The proof will be given later. Henceforth we neglect the effect of entropy fluctuations on the density fluctuations. In the case where the mean-free path of the excitations is much less than the system size, we may Fourier-transform Eqs. (9) and (10) and Eqs. (7) and (8) and neglect the boundary conditions to compute the fluctuations. We find

$$\begin{aligned}
\begin{pmatrix} \delta\rho_{k,\omega} \\ \vec{k}\cdot\delta\vec{v}_{\vec{k},\omega} \end{pmatrix} &= \frac{1}{-\tilde{\omega}^2 - i\tilde{\omega}(\vec{k}\cdot\vec{\nabla}\vec{v}_0\cdot\vec{k} + D_l k^2) + c^2k^2} \begin{pmatrix} -i\tilde{\omega} + \vec{k}\cdot\vec{\nabla}\vec{v}_0\cdot\vec{k} + D_l k^2 & -i\rho_0 \\ -ic^2k^2/\rho_0 & -i\tilde{\omega} \end{pmatrix} \\
&\times \Bigg[\begin{pmatrix} 0 \\ \frac{i}{\rho_0}\vec{k}\cdot\tilde{\mathrm{S}}_{\vec{k},\omega}\cdot\vec{k} \end{pmatrix} \\
&\quad - \frac{\tilde{\vec{v}}_0}{2i}\cdot \begin{pmatrix} i(\vec{k}-\vec{q}_1)\delta\rho_{\vec{k}-\vec{q}_1,\omega} - i(\vec{k}+\vec{q}_1)\delta\rho_{\vec{k}+\vec{q}_1,\omega} \\ i(\vec{k}-\vec{q}_1)\left(1+\frac{\vec{k}\cdot\vec{q}_1}{k^2}\right)(\vec{k}-\vec{q}_1)\cdot\delta\vec{v}_{\vec{k}-\vec{q}_1,\omega} - i(\vec{k}+\vec{q}_1)\left(1-\frac{\vec{k}\cdot\vec{q}_1}{k^2}\right)(\vec{k}+\vec{q}_1)\cdot\delta\vec{v}_{\vec{k}+\vec{q}_1,\omega} \end{pmatrix} \\
&\quad - \frac{\partial D_l}{\partial T}k^2\frac{\delta\tilde{T}}{2i} \begin{pmatrix} 0 \\ (\vec{k}-\vec{q}_2)\cdot\delta\vec{v}_{\vec{k}-\vec{q}_2,\omega} - (\vec{k}+\vec{q}_2)\cdot\delta\vec{v}_{\vec{k}+\vec{q}_2,\omega} \end{pmatrix} - \frac{ik^2}{\rho_0}\frac{\partial c^2}{\partial T}\frac{\delta\tilde{T}}{2i} \begin{pmatrix} 0 \\ \delta\rho_{\vec{k}-\vec{q}_2,\omega} - \delta\rho_{\vec{k}+\vec{q}_2,\omega} \end{pmatrix} \Bigg]\,,
\end{aligned} \tag{14}$$

where $c^2=(\partial p/\partial\rho)_s$, $D_l=(\xi+\frac{4}{3}\eta)\rho_0^{-1}$, and $\tilde{\omega}=\omega-\vec{k}\cdot\vec{v}_0$. Note that we have completely neglected the transverse part of the velocity components $\delta\vec{v}_{\vec{k}\pm\vec{q}_1,\omega}$ and $\delta\vec{v}_{\vec{k}\pm\vec{q}_2,\omega}$ because these fluctuations are centered at zero frequency instead of the sound frequency like the longitudinal fluctuations. Thus, around the Brillouin-peak frequency, these transverse contributions are a factor D_lk^2/ck smaller than the corresponding longitudinal contributions and they can be neglected. For the Langevin force, we assume again that since they represent microscopic (fast) degrees of freedom, they are delta correlated and hence the following local equilibrium form is valid:

$$\langle\vec{k}\cdot\vec{\vec{S}}_{\vec{k},\omega}\cdot\vec{k}\vec{k}\cdot\vec{\vec{S}}^*_{\vec{k},\omega}\cdot\vec{k}\rangle=2T_0\rho_0D_lk^4 2\pi\delta(\omega-\omega)(2\pi)^3\delta^3(0)\,, \tag{15a}$$

$$\langle(\vec{k}\pm\tfrac{1}{2}\vec{q}_2)\cdot\vec{\vec{S}}_{\vec{k}\pm\vec{q}_2/2,\omega}\cdot(\vec{k}\pm\tfrac{1}{2}\vec{q}_2)(\vec{k}\mp\tfrac{1}{2}\vec{q}_2)\cdot\vec{\vec{S}}^*_{\vec{k}\mp\vec{q}_2/2,\omega}\cdot(\vec{k}\mp\tfrac{1}{2}\vec{q}_2)\rangle=\pm 2\frac{\delta\tilde{T}}{2i}\left(1+\frac{\partial\ln D_l}{\partial\ln T}\right)\rho_0D_lk^4 2\pi\delta(\omega-\omega)(2\pi)^3\delta^3(0)\,. \tag{15b}$$

We have neglected terms of order q_2^2.

A light scattering experiment measures a quantity proportional to

$$I=\int\frac{d^3p}{(2\pi)^3}\int\frac{d^3p'}{(2\pi)^3}\Delta(\vec{p}-\vec{k})\Delta(\vec{p}\,'-\vec{k})\langle\delta\rho_{\vec{p},\omega}\delta\rho^*_{\vec{p}\,',\omega}\rangle\,, \tag{16}$$

where $\Delta(\vec{l})$ is a function which is peaked around $\vec{l}=0$ with a width δk inversely proportional to the size of the scattering volume and which reduces to a Dirac delta function in the infinite-volume limit. Using the delta functions contained in the Langevin-force correlations Eqs. (15a) and (15b), it is possible to evaluate the integral over $\vec{p}\,'$ trivially. Since $\delta k>q_1,q_2$, it is possible to choose the remaining dummy integration variable such that (16) looks like an integral over $\vec{p}$ with a weight function peaked at $\vec{p}\sim\vec{k}$ multiplying[10]

$$\{[\langle\delta\rho_{\vec{p}+\vec{q}_2/2,\omega}\delta\rho^*_{\vec{p}-\vec{q}_2/2,\omega}\rangle+(\vec{q}_2\leftrightarrow-\vec{q}_2)] +[\langle\delta\rho_{\vec{p}+\vec{q}_2/2+\vec{q}_1/2,\omega}\delta\rho^*_{\vec{p}-\vec{q}_2/2-\vec{q}_1/2,\omega}\rangle+(\vec{q}_1\leftrightarrow-\vec{q}_1) +(\vec{q}_2\leftrightarrow-\vec{q}_2)+(\vec{q}_1\leftrightarrow-\vec{q}_1)(\vec{q}_2\leftrightarrow-\vec{q}_2)]\}[(2\pi)^3\delta^3(0)]^{-1}\,. \tag{17}$$

If the width of the weight function is small enough, then (17) evaluated at $\vec{p}=\vec{k}$ becomes a good approximation to the integral. In practical experimental situations, however,[2] the width of the weight function is much larger than the intrinsic Brillouin linewidth hence only the overall integrated intensity of the line may be observed. We have calculated detailed line shapes (they are not Lorentzian) but the results are rather lengthy and unilluminating. Hence, the final result we will quote shortly is only for the integrated intensities since it is the most relevant quantity in the rather extreme experimental conditions needed to observe nonlinear corrections to the spectrum.

To compute the corrections to the correlation function to order $\tilde{v}_0\delta\tilde{T}$, we need $\delta\rho$ to the same order. That may be obtained by iterating Eq. (14).[11] We may substitute for the $\delta\rho$ and δv terms multiplying $\tilde{v}_0$ on the right-hand side of Eq. (14) the solution up to first order in $\delta\tilde{T}$ and for the terms multiplying $\delta\tilde{T}$ the solution up to first order in $\tilde{v}_0$. In the correlation functions, terms of order $\tilde{v}_0\delta\tilde{T}$ come in either because one of the $\delta\rho$ is of order $\tilde{v}_0$ and the other one of order $\delta\tilde{T}$, or because one of the $\delta\rho$ is of order $\tilde{v}_0\delta\tilde{T}$ and the other of zeroth order, or, finally, because one of the $\delta\rho$ is of order $\tilde{v}_0$ and Eq. (15b) instead of (15a) is used to compute the Langevin-force correlation. In other words, terms of order $\delta\tilde{T}$ may come either from the hydrodynamic matrix or from the Langevin-force correlation while terms of order $\tilde{v}_0$ come only from the hydrodynamic matrix. Note also that in $\langle\delta\rho_{\vec{k}+\vec{q}_2/2,\omega}\delta\rho^*_{\vec{k}-\vec{q}_2/2,\omega}\rangle+(\vec{q}_2\leftrightarrow-\vec{q}_2)$, the terms of order $\tilde{v}_0$ come from couplings to $\tilde{v}_0$ which are diagonal in momentum space, i.e., are obtained by expanding the denominators on the right-hand side of Eq. (14) to first order in $\tilde{v}_0$. (There are no contributions from the upper left-hand corner element of the matrix multiplying the square bracket.) The procedure is straightforward but very tedious.

To order $\tilde{v}_0\delta\tilde{T}$, we find terms proportional to $(c\hat{k}\cdot\vec{\nabla}\ln T)(\hat{k}\cdot\vec{\nabla}\vec{v}_0\cdot\hat{k})/(D_lk^2)^2$, which is just the product of the expansion parameters for linear corrections, terms of order $(c\hat{k}\cdot\vec{\nabla}\vec{v}_0\cdot\vec{\nabla}\ln T)/(D_lk^2)^2$ which vanish in the steady state given in Eqs. (7) and (8), and finally, terms of order $(c\vec{\nabla}\ln T\cdot\vec{\nabla}\vec{v}_0\cdot\hat{k})/(D_lk^2)^2$ which we are interested in. These last terms are the only ones which contribute if we choose $\hat{k}$ perpendicular to both $\vec{\nabla}T$ and $\vec{\nabla}|\vec{v}_0|$ because then the linear terms and the terms proportional to their product, $(c\hat{k}\cdot\vec{\nabla}\ln T)\times(\hat{k}\cdot\vec{\nabla}\vec{v}_0\cdot\hat{k})/(D_lk^2)^2$, vanish. Note that the coefficients of the latter terms are of the same order as those of the terms we are interested in, hence $(c\hat{k}\cdot\vec{\nabla}\ln T)(\hat{k}\cdot\vec{\nabla}\vec{v}_0\cdot\hat{k})/(D_lk^2)^2$ terms do not complicate the interpretation of the spectrum even if $\hat{k}$ is not strictly perpendicular to $\vec{\nabla}T$ and $\vec{\nabla}|\vec{v}_0|$.

For the steady state described in the preceding section and for $\vec{k}$ perpendicular to $\vec{\nabla}T$ and $\vec{\nabla}|\vec{v}_0|$, we predict an asymmetry in the Doppler-shifted

Brillouin peaks which may be characterized as follows. ΔN, the difference between the integrated intensity of the $\tilde{\omega}=ck$ Brillouin peak and that of the $\tilde{\omega}=-ck$ peak, divided by N, the sum of the integrated intensities of both peaks at equilibrium, is given by

$$\frac{\Delta N}{N}=-\frac{c\vec{\nabla}\ln T\cdot\vec{\nabla}\vec{\mathrm{v}}_0\cdot\hat{k}}{(D_l k^2)^2}\,. \qquad (18)$$

Note that the terms proportional to $(\partial \ln c^2/\partial \ln T)$ and $(\partial \ln D_l/\partial \ln T)$ drop out from (18). The integrals over frequency which must be done to obtain (18) are most easily performed if $\tilde{\omega}^2$ is used as an integration variable and if the usual method is used to differentiate under the integral sign with respect to the roots of $[(\tilde{\omega}^2-c^2k^2)^2+(\tilde{\omega}D_l k^2)^2]=0$ or with respect to $(D_l k^2)^2$ or $(ck)^2$. One must be particularly careful to keep all terms of the same order in $D_l k^2/(ck)$.

We may now discuss parenthetically the reasons for neglecting the contributions from the entropy fluctuations Eq. (11) in the computation of the density fluctuations. The important point is that when $(\partial V/\partial T)_p=0$ to zeroth order, then the coupling between the entropy fluctuations and the other fluctuations are only proportional to $\delta\tilde{T}$, not to both $\bar{v}_0$ and $\delta\tilde{T}$. They come from $\vec{\nabla}[(\partial p/\partial s)_p\,\delta s]$ in the velocity equation (10) and from $-\delta\vec{\mathrm{v}}\cdot\vec{\nabla}s+(pT)^{-1}\vec{\nabla}\cdot\{\kappa\vec{\nabla}[(\partial T/\partial\rho)_s\delta\rho]\}$ in the entropy equation (11). [To first order in $\delta\tilde{T}$, $(\partial T/\partial\rho)_s\neq 0$ and $(\partial p/\partial s)_\rho\neq 0$.] Evaluating $\delta\rho$ and δs in these coupling terms up to first order in $\bar{v}_0$ will only introduce, in the equation for the density, terms proportional to $\delta\tilde{T}\vec{\mathrm{g}}$ or to $\bar{v}_0\delta\tilde{T}\vec{\mathrm{g}}$, where $\vec{\mathrm{g}}$ is the fluctuating Langevin heat flux. But the fact that the Langevin heat flux is uncorrelated with the fluctuating Langevin stress tensor leads to a vanishing contribution to the density-density correlation function to order $\bar{v}_0\delta\tilde{T}$. That should be contrasted with what would have happened if there also had been a term of order $\bar{v}_0$ coupling the entropy and density fluctuations: Then, for example, the terms proportional to $\delta\tilde{T}\vec{\mathrm{g}}$ in one of the $\delta\rho$ would correlate with the terms of order $\bar{v}_0\vec{\mathrm{g}}$ in the other $\delta\rho$. If $(\partial V/\partial T)_p\neq 0$ those arguments do not apply and the entropy fluctuations are coupled to the density fluctuations. However, we expect that since entropy fluctuations are at low frequency, their only effect on the Brillouin lines is to change D_l into Γ_s, the usual sound damping constant which also contains the thermal conductivity, not only the viscosities.

IV. CONCLUSION

The key to the physical interpretation of result (18) is that excitations do not propagate along straight lines when a shear flow is present. One can use Eqs. (9) and (10) to obtain a wave equation from which the sound-wave trajectory may be calculated in the eikonal approximation. The result is illustrated in Fig. 1. Sound waves with wave vectors $\vec{\mathrm{k}}$ parallel to $\hat{x}$ at point 0 arrive from the left along the trajectory $y=-\frac{1}{2}(|\vec{\nabla}\vec{\mathrm{v}}_0|/c)x^2$ or from the right along $y=\frac{1}{2}(|\vec{\nabla}\vec{\mathrm{v}}_0|/c)x^2$; hence, the latter, emited by a hotter part of the fluid, are generated at higher rate, leading to an asymmetry in the Brillouin peaks. By this argument, the result (18) may be reproduced. Note that the rate at which sound waves are generated contains a factor $\partial\ln D_l/\partial\ln T$ [see Eq. 15(b)] which does not appear in Eq. (18) because it is compensated by terms representing the change in mean-free path proportional to that same factor.

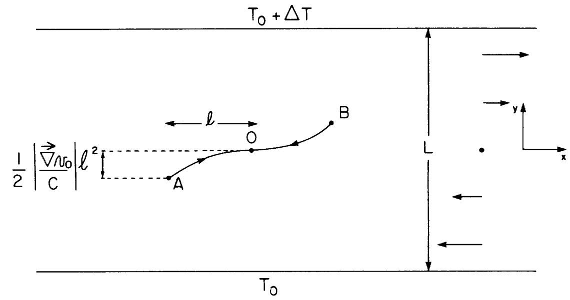

FIG. 1. Sound-wave trajectories in a shear flow: the velocity profile is represented by horizontal arrows. When a temperature gradient is present, more sound waves are generated at point B than at point A, which are roughly one mean-free path l away from the origin.

Let us discuss the observability of the effect. The experiment could be performed with two coaxial cylinders with the outer (instead of the inner) cylinder rotating to avoid instability. If, further, the inner cylinder is heated and the outer one cooled, then we have achieved a stable flow in which both the temperature gradient and the gradient in the nonzero component of the velocity are parallel. A scattered wave vector perpendicular to both gradients may be achieved by sending in the laser beam from the cylinder's top or bottom and collecting the scattered light at the other end. By measuring the dependence of the Brillouin-peak asymmetry on the magnitude of the two gradients and on the direction of $\vec{\mathrm{k}}$ in the plane perpendicular to the gradients, one can single out the term we are interested in.

The inequalities which define the domain of applicability of the theory are basically the same as those discussed for the linear regime. An experiment in the presence of a temperature gradient has already been performed[2] (somewhat outside the range of validity of the theory) with acceptable results but it appears that shear flow experiments would be much more difficult.[12] The following set of inequalities summarizes the range of applicability of the theory[13]:

$$\left(\frac{c}{D_l L}\right)^{1/2} < k < \frac{c}{D_l}\,, \tag{19}$$

$$L < |(\vec{\nabla} \ln T)|^{-1}\,, \tag{20}$$

$$\frac{|\vec{\nabla}\vec{v}_0|}{D_l k^2} < 1\,, \tag{21}$$

$$\frac{\Delta v_0 L}{D_l} < R_c\,. \tag{22}$$

The first inequality in (19) guarantees that the sound-wave mean-free path is less than the system size. That inequality combined with (20), which comes from $\Delta T/T \ll 1$, guarantees that the expansion parameter $|c\vec{\nabla}\ln T|/(D_l k^2)$ is smaller than unity. Note that to make that expansion parameter as large as possible, it is preferable to do the experiment in a low-temperature fluid where it is in principle possible to achieve $|\vec{\nabla}\ln T|^{-1} \sim L$ while staying in the liquid phase. The second inequality in (19) ensures that the sound waves are well-defined excitations ($ck > D_l k^2$) while (21) must be satisfied since $|\vec{\nabla}\vec{v}_0|/(D_l k^2)$ is an expansion parameter. Finally, (22) restricts the Reynolds number to be smaller than the critical Reynolds number R_c where the laminar flow becomes unstable.

To achieve $|\vec{\nabla}\vec{v}_0|/(D_l k^2) \equiv \epsilon$ less than, but close to, unity we must take the smallest possible value of k consistent with (19). Taking $k = a[c/(D_l L)]^{1/2}$ where a is larger than unity, we obtain $\epsilon = \Delta v_0/(a^2 c)$. Hence we need velocities close to the speed of sound to achieve sizable values of ϵ. This unfortunately implies that we must also use very small size systems to stay in the laminar region. Indeed, (22) and (19) imply that

$$L < \frac{R_c}{\epsilon a^2}\frac{D_l}{c} < \frac{R_c}{\epsilon a^2} k^{-1}\,. \tag{23}$$

For example, with water, $D_l = 2\times 10^{-2}$ cm^2/s and $c = 1.42\times 10^5$ cm/s hence with $\epsilon = 0.3$ and $a^2 = 3$ the velocity must be close to the sound velocity and if $R_c = 10^3$ then $L \lesssim 10^{-4}$ cm and $k \sim 10^6$ cm^{-1}. A fluid with the largest possible viscosity is preferable since it allows a maximization of the system size. The heat conductivity must simultaneously be large enough to be able to neglect viscous heating.

In summary, we have shown that the theory developed in Ref. 4 may be used to compute nonlinear corrections to the fluctuations about dissipative steady states in rather extreme conditions at the limit of stability of laminar flow. A check of the theory would require very delicate experimentation or computer simulations. Details of this calculation will appear elsewhere.[14]

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APPENDIX

In this Appendix, we demonstrate how the equal-time correlation functions can be obtained from an Einstein relation. Equations (9) and (10) are first written in matrix form

$$\frac{\partial}{\partial T} A_\alpha(\vec{k},t) + \int \frac{d^3k'}{(2\pi)^3} M_{\alpha\beta}(\vec{k},\vec{k}')A_\beta(\vec{k}',t) = f_\alpha(\vec{k},t)\,, \tag{A1}$$

where $A_1 = \delta\rho(\vec{k},t)$, $A_2 = \vec{k}\cdot\delta\vec{v}(\vec{k},t)$; $M_{\alpha\beta}(\vec{k},\vec{k}')$ is the hydrodynamic matrix, and $f_\alpha(\vec{k},t)$ is the Langevin force term with

$$f_1 = 0,\quad f_2 = i\vec{k}\cdot\vec{S}(\vec{k},t)\cdot\vec{k}/\rho_0\,.$$

A generalized Einstein relation[4] may be obtained from (A1):

$$\underline{D}(\vec{k},\vec{k}') = \underline{M}(\vec{k},\vec{p})\underline{\chi}(\vec{p},\vec{k}') + \underline{\chi}(\vec{k},\vec{p})[\underline{M}(\vec{p},\vec{k}')]^\dagger\,, \tag{A2}$$

where

$$\langle f_\alpha(\vec{k},t) f_\beta(\vec{k}',t')\rangle = D_{\alpha\beta}(\vec{k},\vec{k}')\delta(t-t')$$

and

$$\chi_{\alpha\beta}(\vec{k},\vec{k}') = \langle A_\alpha(\vec{k},0)A_\beta(\vec{k}',0)\rangle$$

is the equal-time correlation function. Integration over repeated momentum indices is implied. Equation (A2) must be understood as a matrix equation in $\vec{k}$ space as well as in the $\delta\rho$ and δv space. In particular, note that

$$[M_{\alpha\beta}(\vec{p},\vec{k}')]^\dagger = M_{\beta\alpha}(\vec{k}',\vec{p})^* \,.$$

$\underline{\chi}$ is the unknown matrix we wish to compute from Eq. (A2). We write the hydrodynamic matrix as

$$\underline{M}(\vec{k},\vec{k}') = \underline{M}_0(\vec{k})\delta^3(\vec{k}-\vec{k}') + \underline{M}_1(\vec{k})[\delta^3(\vec{k}-\vec{k}'-\vec{q}_1) - \delta^3(\vec{k}-\vec{k}'+\vec{q}_1)] + \underline{M}_2(\vec{k})[\delta^3(\vec{k}-\vec{k}'-\vec{q}_2) - \delta^3(\vec{k}-\vec{k}'+\vec{q}_2)] \,, \tag{A3}$$

where $\underline{M}_0$ is the equilibrium matrix while $\underline{M}_1$ and $\underline{M}_2$ are purely off-diagonal terms proportional, respectively, to the shear flow and the temperature gradient. Note that there is no term proportional to both $\vec{\nabla} T$ and $\vec{\nabla}\vec{v}_0$ in the hydrodynamic matrix, and that for the sake of simplicity, we have momentarily neglected the diagonal term in $\vec{\nabla}\vec{v}_0$. Similarly,

$$\underline{D}(\vec{k},\vec{k}') = \underline{D}_0(\vec{k})\delta^3(\vec{k}-\vec{k}') + \underline{D}_2(\vec{k})[\delta^3(\vec{k}-\vec{k}'-\vec{q}_2) - \delta^3(\vec{k}-\vec{k}'+\vec{q}_2)] \,.$$

There is no term proportional to $\vec{\nabla}\vec{v}_0$ in that diffusion constant. To solve Eq. (A2), we write

$$\begin{aligned}\underline{\chi}(\vec{k},\vec{k}') = {} & \underline{\chi}_0(\vec{k})\delta^3(\vec{k}-\vec{k}') + \underline{\chi}_1(\vec{k})[\delta^3(\vec{k}-\vec{k}'-\vec{q}_1) + \delta^3(\vec{k}-\vec{k}'+\vec{q}_1)] \\ & + \underline{\chi}_2(\vec{k})[\delta^3(\vec{k}-\vec{k}'-\vec{q}_2) + \delta^3(\vec{k}-\vec{k}'+\vec{q}_2)] \\ & + \underline{\chi}_{1,2}(\vec{k})[\delta^3(\vec{k}-\vec{k}'-\vec{q}_1-\vec{q}_2) + (\vec{q}_1 \leftrightarrow -\vec{q}_1) + (\vec{q}_2 \leftrightarrow -\vec{q}_2) + (\vec{q}_1 \leftrightarrow -\vec{q}_1, \vec{q}_2 \leftrightarrow -\vec{q}_2)]\end{aligned} \tag{A4}$$

and equate powers of $\vec{\nabla} T$ and $\vec{\nabla}\vec{v}_0$ on both sides of Eq. (A2). To zeroth order, we recover for $\underline{\chi}_0$ the equilibrium result. The first-order equation in $\vec{\nabla} T$ involves $\underline{\chi}_0$. The result is identical to that found in Ref. 4. An analogous calculation reveals that $\underline{\chi}_1 = 0$. Finally, the equation containing $\underline{\chi}_{1,2}$ involves $\underline{\chi}_1$ and $\underline{\chi}_2$ which have been computed in the previous steps. To that order, Eq. (A2) reads

$$\underline{M}_0(\vec{k},\vec{p})\underline{\chi}_{1,2}(\vec{p},\vec{k}') + \underline{\chi}_{1,2}(\vec{k},\vec{p})[\underline{M}_0(\vec{p},\vec{k}')]^\dagger = -\underline{M}_1(\vec{k},\vec{p})\underline{\chi}_2(\vec{p},\vec{k}') - \underline{\chi}_2(\vec{k},\vec{p})[\underline{M}_1(\vec{p},\vec{k})]^\dagger \,. \tag{A5}$$

Using the δ functions to perform the $\vec{p}$ integrals we obtain

$$\begin{aligned}& \underline{M}_0(++,++)\underline{\chi}_{1,2}(++,--) + \underline{\chi}_{1,2}(++,--)[\underline{M}_0(--,--)]^\dagger \\ & \quad = -\underline{M}_1(++,-+)\underline{\chi}_2(-+,--) - \underline{\chi}_2(++,+-)[\underline{M}_1(+-,--)]^\dagger \,,\end{aligned} \tag{A6}$$

where $(\pm\pm,\pm\pm)$ stands for $(\vec{k}\pm\vec{q}_1/2,\pm\vec{q}_2/2, \vec{k}\pm\vec{q}_1/2\pm\vec{q}_2/2)$. There are equations similar to (A6) for $(\vec{q}_1 \leftrightarrow -\vec{q}_1)$, $(\vec{q}_2 \leftrightarrow -\vec{q}_2)$, $(\vec{q}_1 \leftrightarrow -\vec{q}_1, \vec{q}_2 \leftrightarrow -\vec{q}_2)$. $\underline{\chi}_{1,2}$ is practically our final result. The diagonal term proportional to $\vec{\nabla}\vec{v}_0$ in the hydrodynamic matrix may be taken into account without further difficulties.

Finally, one can relate $\underline{\chi}_{1,2}$ to the integrated intensities. Using the equation of continuity (9), one can write (omitting the frequency indices)

$$\begin{aligned}& \frac{\tilde{\omega}}{ck}\{\langle\delta\rho_{\vec{k}}\delta\rho^*_{\vec{k}}\rangle + [\langle\delta\rho_{\vec{k}+\vec{q}_2/2}\delta\rho^*_{\vec{k}-\vec{q}_2/2}\rangle + (\vec{q}_2 \leftrightarrow -\vec{q}_2)] \\ & \quad + [\langle\delta\rho_{\vec{k}+\vec{q}_1/2+\vec{q}_2/2}\delta\rho^*_{\vec{k}-\vec{q}_1/2-\vec{q}_2/2}\rangle + (\vec{q}_1 \leftrightarrow -\vec{q}_1) + (\vec{q}_2 \leftrightarrow -\vec{q}_2) + (\vec{q}_1 \leftrightarrow -\vec{q}_1, \vec{q}_2 \leftrightarrow -\vec{q}_2)]\} \\ & \quad = \frac{\rho_0}{ck}\{\langle\vec{k}\cdot\delta\vec{v}_{\vec{k}}\rho^*_{\vec{k}}\rangle + [\langle(\vec{k}+\vec{q}_2/2)\cdot\delta\vec{v}_{\vec{k}+\vec{q}_2/2}\delta\rho^*_{\vec{k}-\vec{q}_2/2}\rangle + (\vec{q}_2 \leftrightarrow -\vec{q}_2)] \\ & \qquad + [\langle(\vec{k}+\vec{q}_1/2+\vec{q}_2/2)\cdot\delta\vec{v}_{\vec{k}+\vec{q}_1/2+\vec{q}_2/2}\delta\rho^*_{\vec{k}-\vec{q}_1/2-\vec{q}_2/2}\rangle + (\vec{q}_1 \leftrightarrow -\vec{q}_1) + (\vec{q}_2 \leftrightarrow -\vec{q}_2) + (\vec{q}_1 \leftrightarrow -\vec{q}_1, \vec{q}_2 \leftrightarrow -\vec{q}_2)]\} \\ & \qquad + i\,\frac{\vec{\nabla}\vec{v}_0\cdot\vec{k}}{2ck}\{[\langle\delta\rho_{\vec{k}-\vec{q}_1/2+\vec{q}_2/2}\delta\rho^*_{\vec{k}-\vec{q}_1/2-\vec{q}_2/2}\rangle + (\vec{q}_2 \leftrightarrow -\vec{q}_2)] - (\vec{q}_1 \leftrightarrow -\vec{q}_1)\} \,.\end{aligned} \tag{A7}$$

The last line involves only the correction for a temperature gradient alone, evaluated at $\vec{k}\pm\vec{q}_1/2$, which is odd in $\tilde{\omega}$; hence, integrating both sides of Eq. (A7) over $\tilde{\omega}$, we are left with a relation between ΔN, defined before Eq. (18), and the element 1, 2 of matrix $\underline{\chi}_{1,2}$. Using the result for that matrix then leads to Eq. (18).

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[3]A.-M. S. Tremblay, Eric D. Siggia, and M. R. Arai, Phys. Lett. 76A, 57 (1980); T. Kirkpatrick, E. G. D. Cohen, and J. R. Dorfman, Phys. Rev. Lett. 44, 472 (1980).

[4]A.-M. S. Tremblay, M. Arai, and E. D. Siggia, Phys. Rev. A 23, 1451 (1981).

[5]Reference 1 contained conflicting predictions. Reference 3 arrived simultaneously at the now generally accepted result. See conclusion of Ref. 4 for disscussion and more complete references.

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[7]T. R. Kirkpatrick and E. G. D. Cohen, Phys. Lett. 78A, 350 (1980).

[8]The expansion parameter is the sound-wave mean-free path divided by $|\nabla \ln T|^{-1}$, the length scale defined by the temperature gradient, or equivalently, it is the sound-wave decay time divided by the time taken by the sound wave to cross a distance $|\vec{\nabla} \ln T|^{-1}$.

[9]L. D. Landau and E. M. Lifshitz, *Fluid Mechanics* (Pergamon, Oxford, 1959).

[10]The calculations of Appendix A of Ref. 4 can easily be extended to the case of interest here.

[11]The linearized hydrodynamic equations do not contain terms of that order. There is no inconsistency since the expansion parameters $[c|\vec{\nabla} \ln T|/(D_l k^2)]$ and $[|\vec{\nabla}\vec{v}_0|/(D_l k^2)]$ involved in iterating the linearized equations to find the fluctuations are related to the correlation times of the fluctuations and are not the same as those involved in deriving the hydrodynamic equations. A similar agrument allows us to neglect terms like $\delta\rho\,\delta v$ in Eqs. (9) to (11).

[12]J. Machta, I. Oppenheim, and I. Procaccia, Phys. Rev. A 22, 2809 (1980).

[13]The inequalities $(\partial \ln \kappa/\partial \ln T)\Delta T/T \ll 1$ and $(\partial \ln \eta/\partial \ln T)\Delta T/T \ll 1$ of Sec. II would add a factor $(\partial \ln \kappa/\partial \ln T)$ or $(\partial \ln \eta/\partial \ln T)$ on the left-hand side of Eq. (20). These extra factors may reduce the range of validity of the theory and should be accounted for in each practical case.

[14]Claude Tremblay, Master's thesis, Université de Sherbrooke (unpublished).