Simplified hydrodynamic theory of nonlocal stationary state fluctuations

David Ronis *
Department of Chemistry, University of California, Berkeley, California 97420

Seth Putterman
Physics Department, University of California, Los Angeles, California 90024
(Received 21 January 1980)

In the realsitic approximation that the expansion coefficient of a fluid vanishes, the hydrodynamic fluctuations around a steady state characterized by a small temperature gradient are determined entirely by the variations of the strength of the random forces from point to point. The assumption of local hydrodynamic equilibrium for the random forces leads to a long-range static density momentum correlation, as well as to a significant odd-in-frequency correction to the Brillouin light scattering, whose integrated intensity agrees with other work of Ronis *et al*. and Kirkpatrik *et al*. Consequences of the long-range correlations for $1/f$ noise ^{4}He and local equilibrium postulates are discussed.

The theory of low-frequency fluctuations in a state of global equilibrium as described by a linearized fluctuating hydrodynamics is well established.[1] A problem of slightly greater generality would involve a description of fluctuations about a near-equilibrium steady state such as is realized in a system with a small constant temperature gradient, no convection, and constant pressure (or, respectively, $\vec{\nabla} T_0 = \text{constant}$, $\vec{V} = 0$, $p = \text{constant}$). However, linearizing about such a state to first power in ∇T_0 already involves considerable complications.[2,3] In this paper we present a discussion of such fluctuations in terms of a physically realistic yet mathematically simplified choice of the equations of state.

In the approximation where all mode coupling and therefore all terms quadratic in the fluctuations are neglected, the mass density ρ and momentum density conservation laws are easily combined to yield

$$\frac{\partial^2 \rho}{\partial t^2} - \nabla^2 p + \nabla^2 (\rho \alpha \vec{\nabla} \cdot \vec{V}) = \nabla^2 \tilde{\tau} , \qquad (1)$$

where we assume longitudinal motion and $\rho\alpha$ is the appropriate sum of shear and bulk viscosities and $\tilde{\tau}$ is the diagonal component of the fluctuating stress tensor whose equilibrium correlation is

$$\langle \tilde{\tau}(r,t) \tilde{\tau}(r',t') \rangle = 2 k_B T_{\text{eq}} \rho \alpha \delta(r - r') \delta(t - t') , \qquad (2)$$

where the subscript eq denotes the constant equilibrium value. In general, $p = p(\rho, T)$ which has the effect of coupling in the thermal diffusion and convection, complicating the calculation, especially off equilibrium. However, if one imposes the realistic approximation that the expansion coefficient vanishes, then $p = p(\rho)$, the speed of sound is constant, and

$$\vec{\nabla} \cdot \vec{V} = -\frac{1}{\rho_{\text{eq}}} \frac{\partial \rho}{\partial t} \qquad (3)$$

so that (1) becomes

$$\frac{\partial^2 \rho}{\partial t^2} - c^2 \nabla^2 \rho - \nabla^2 \left(\alpha \frac{\partial \rho}{\partial t} \right) = \nabla^2 \tilde{\tau} \qquad (4)$$

which is one equation for the variable ρ valid even in the presence of an imposed steady-state temperature gradient.

If one further assumes that the kinematic sound-attenuation coefficient α is independent of temperature, we find that the dynamic response function of the system with $\vec{\nabla} T_0 \neq 0$ is identical with that in global equilibrium and the only manner in which the imposed temperature gradient can enter the description is through the variation of the random force from point to point. It is natural and consistent with the idea of hydrodynamic local equilibrium to assume that in the presence of a small $\vec{\nabla} T_0$ the random forces maintain the form (2) with T_{eq} replaced by

$$T_0(\vec{r}) = T_{\text{eq}} + \vec{r} \cdot \vec{\nabla} T_0 . \qquad (5)$$

Introducing the Fourier transform

$$\tilde{\tau}(\vec{k}, \omega) = \frac{1}{(2\pi)^2} \int e^{-i\vec{k}\cdot\vec{r} + i\omega t} \tilde{\tau}(\vec{r}, t) d\vec{r}\, dt \qquad (6)$$

yields from (5)

$$\langle \tilde{\tau}(\vec{k}, \omega) \tilde{\tau}^*(\vec{k}', \omega') \rangle = 2 k_B \rho \alpha \delta(\omega - \omega') T_0(\vec{k} - \vec{k}') , \qquad (7)$$

$$T_0(\vec{k} - \vec{k}') = T_{\text{eq}} \delta(\vec{k} - \vec{k}') - \vec{\nabla} T_0 \cdot \frac{\partial \delta(\vec{k} - \vec{k}')}{\partial i(\vec{k} - \vec{k}')} . \qquad (8)$$

The light scattering is immediately determined from (4) and (7) as

$$\langle\rho(\vec{k},\omega)\rho^*(\vec{k}',\omega')\rangle=\frac{2k_B\rho\alpha T_0(\vec{k}-\vec{k}')\delta(\omega-\omega')k^2k'^2}{(\omega^2-c^2k^2+i\alpha\omega k^2)(\omega^2-c^2k'^2-i\alpha\omega k'^2)}, \tag{9}$$

$$\langle\rho(\vec{r},\omega)\rho(\vec{r}',\omega')\rangle=\frac{T_0(\frac{1}{2}(\vec{r}+\vec{r}'))}{T_{eq}}S_{eq}(\vec{r}-\vec{r}',\omega)\delta(\omega-\omega')+\int S_{nl}(\vec{k},\omega)e^{i\vec{k}\cdot(\vec{r}-\vec{r}')}d^3k\,\delta(\omega-\omega'), \tag{10}$$

where the nonlocal correction to equilibrium light scattering is given by[4,5]

$$S_{nl}(\vec{k},\omega)=-S_{eq}(\vec{k},\omega)\frac{2\alpha\omega^3\vec{k}\cdot\vec{\nabla}\ln T_0}{(\omega^2-c^2k^2)^2+\alpha^2\omega^2k^4}. \tag{11}$$

In order to obtain (10) one must integrate the δ-function derivative by parts, holding $\vec{k}+\vec{k}'$ constant. The light scattering has two physically distinct contributions. The term even in the frequency shift ω is the equilibrium effect adjusted to the local value of the temperature. The term odd in frequency is due to the nonlocal correlations which result from the imposed temperature gradient. It leads to a percentage change in intensity of each Brillouin peak given by

$$-c^2\vec{k}\cdot\vec{\nabla}\ln T_0/\omega\alpha k^2, \tag{12}$$

which agrees with Ref. 2.

When the temperature dependence of α and c are included one finds no change in the nonlocal correction (11), whereas the contribution to light scattering even in ω becomes[2]

$$\left(1+\tfrac{1}{2}(\vec{r}+\vec{r}')\cdot\vec{\nabla}T_0\frac{\partial}{\partial T}\right)S_{eq}(\vec{r}-\vec{r}',\omega)\delta(\omega-\omega'). \tag{13}$$

For the remainder of the paper we ignore the temperature dependence of c and α. The static or equal-time averages are obtained by integrating over frequency so that

$$\langle\vec{J}(\vec{k},0)\rho^*(\vec{k}',0)\rangle=\int\frac{\omega\vec{k}}{k^2}\langle\rho(\vec{k},\omega)\rho^*(\vec{k}',\omega')\rangle d\omega\,d\omega'$$

$$=\frac{-k_B\rho}{2\pi\alpha k^4}(\vec{k}\cdot\vec{\nabla}T_0)\vec{k}\,\delta(\vec{k}-\vec{k}'), \tag{14}$$

$$\langle\rho(\vec{k},0)\rho^*(\vec{k}',0)\rangle=\int\langle\rho(\vec{k},\omega)\rho^*(\vec{k}',\omega')\rangle d\omega\,d\omega'$$

$$=(k_B\rho/2\pi c^2)T_0(\vec{k}-\vec{k}'). \tag{15}$$

In coordinate space the density autocorrelation function is exactly what one would expect from local equilibrium:

$$\langle\rho(\vec{r},0)\rho(\vec{r}',0)\rangle=[k_B\rho T_0(\vec{r})/c^2]\delta(\vec{r}-\vec{r}'),$$

whereas the density momentum correlation shows a nonlocal long-ranged decay going as $|\vec{r}-\vec{r}'|^{-1}$.[2,6,7] Generating the dynamics from the statics through application of the equilibrium propagators to (14) and (15) yields (9).[8]

Although the temperature gradient is essential for the long-range static $\vec{J},\rho$ correlation, the thermal conductivity K does not appear in the result. Thus in the limit where K goes to zero so that the entropy production vanishes, one is led to ask whether it should be possible to understand (14) as an equilibrium property due to the appearance of long-ranged effects in the free energy. Also the static long-range correlations bring into question the basic assumptions of *local* equilibrium and *local* momentum conservation from which these results were derived. The possible breaking of these symmetries may be accompanied by new collective modes.[2]

The long-range correlations are accompanied by a corresponding two-dimensional $1/f$ noise as can be seen from the integral

$$-\vec{\nabla}T_0\cdot\int\langle\vec{J}(\vec{k},\omega)\rho^*(\vec{k}',\omega')\rangle d^2k\,d^2k'\,d\omega'=\frac{k_B}{\alpha}\frac{(\vec{\nabla}T_0)^2}{|\omega|}.$$

A connection between experiment and this mathematical $1/f$ noise is at present unclear.

A system in which the thermal conductivity is very small is realized for superfluid ^{4}He contained in the pores formed by very fine powder, a so-called superleak. For such a system the chemical potential μ and not the pressure is constant in the steady state, so that the quantity which we approximate as zero in analogy with the expansion coefficient is

$$\frac{1}{\rho}\frac{\partial\rho_s}{\partial T}+s\frac{\partial\rho_s}{\partial p},$$

where s and ρ_s are the entropy per gram and superfluid density, respectively. In this case the dynamic equation corresponding to (4) Ref. 9 and the random force and momentum-density correlations are

$$\frac{\partial^2\rho}{\partial t^2}-c_4^2\nabla^2\rho-\rho_s\zeta_3\nabla^2\frac{\partial\rho}{\partial t}=\rho_s\nabla^2\tilde{H},$$

$$\langle\tilde{H}(\vec{r},t)\tilde{H}(\vec{r}',t')\rangle=2k_BT\zeta_3(\vec{r}-\vec{r}')\delta(t-t'),$$

$$\langle\rho_s\vec{V}_s(\vec{k},0)\rho^*(\vec{k}',0)\rangle=\frac{k_B(\vec{k}\cdot\vec{\nabla}T_0)\vec{k}\delta(\vec{k}-\vec{k}')}{2\pi\zeta_3k^4},$$

where ζ_3 is the third bulk viscosity. The percentage change in intensity of the Brillouin peaks is then given by

$$-c_4^2\vec{k}\cdot\vec{\nabla}\ln T_0/\omega\rho_s\zeta_3k^2,$$

where $c_4^2\equiv\rho_s(\partial\mu/\partial\rho)_{\rho s}$. In the superfluid state the phase of the macroscopic wave function, or velocity potential, becomes observable and its correlation with the density depends logarithmically upon the spatial separation in the presence of a temperature gradient.

Although the scattered intensity (12) agrees with Ref. 2, a comparison with the results of Ref. 3 is difficult to make. However, in a recent paper Kirkpatrick *et al.* also obtain an intensity in agreement with Eq. (12) by use of an approach which is partly hydrodynamical and partly mode coupling.[10] As is the case here, they obtain a detailed peak structure which is a square of Lorentzian, whereas Ref. 2 derived a Lorentzian peak. Thus a question arises as to whether or not the line shape is measurable. We feel that the answer is no on two grounds. First, for practical reasons, one needs to determine the frequency as well as the scattering vector to better than 1 part in 10^5 in order to resolve the line shape. Note that in the example given in Ref. 2 the peak shift is 58.4×10^6 sec^{-1} and the width is 1100 sec^{-1}. This all must be sorted out at a scattering angle of 0.4°, which is rather difficult. Secondly, the requirement that the nonlocal light-scattering effects be observable necessitates using a beam of sufficient collimation so that the variations in ordinary light scattering due to scattering from points of different temperature do not wash out a given peak. This collimation will, in general, lead to an uncertainty in the momentum transfer that averages out the peak structure. Taking a beam diameter of about 0.1 cm so as to be small compared with the length characterizing the temperature gradient yields for the spread in incident wave vector

$$\Delta k > 5 \text{ cm}^{-1}.$$

For conditions of interest the momentum transfer is 10^3 cm^{-1} and thus the uncertainty in momentum transfer is 5 parts in 10^3, more than enough to average out the line shape.

Considerable interest has been generated about this difference in peak structure due to the fact that it cannot obviously be accounted for by the Navier-Stokes vs Burnett approximations. In the remainder of this paper we present an alternate analysis of the statistical-mechanical expression given in Ref. 2 which yields line shapes consistent with those obtained here. In Ref. 2 [(cf. Eq. (3.1)] the main quantity of interest was

$$\underline{\Lambda}(\vec{k},\sigma)=\int_{-\infty}^{\infty} d\tau\langle \underline{A}_{\vec{k}}(\sigma)\underline{A}_{-\vec{k}}I_{\delta,T}(-\tau)\rangle, \tag{16}$$

where the notation is the same as in Ref. 2. Specifically, $\underline{A}_{\vec{k}}(\sigma)$ represents the set of conserved variables and $I_{\delta,T}(-\tau)$ the random flux (δ is the energy or momentum). Note the identity

$$e^{iLt}=e^{i(1-P)Lt}(1-P)+Pe^{iLt}+\int_0^t dt_1 e^{i(1-P)L(t-t_1)}(1-P)iLPe^{iLt_1}, \tag{17}$$

where L is the Liouville operator and where P is defined, for any $B_{\vec{k}}$, as

$$PB_{\vec{k}}\equiv\langle B_{\vec{k}}\underline{A}_{-\vec{k}}\rangle\langle \underline{A}_{\vec{k}}\underline{A}_{-\vec{k}}\rangle^{-1}\underline{A}_{\vec{k}}. \tag{18}$$

Thus P is just Mori's projection operator.[11] Using Eq. (17) in (16) yields

$$\underline{\underline{\Lambda}}(\vec{k},\sigma)=\underline{\underline{G}}_{\vec{k}}(\sigma)\cdot\int_{-\infty}^{\infty} d\tau\langle \underline{A}_{\vec{k}}\underline{A}_{-\vec{k}}I_{\delta,T}(-\tau)\rangle+\int_0^{\sigma} dt_1\underline{\underline{G}}_{\vec{k}}(\sigma-t_1)\cdot\int_{-\infty}^{\infty} d\tau\langle \dot{\underline{A}}^{\dagger}_{\vec{k}}(t_1)\underline{A}_{-\vec{k}}I_{\delta,T}(-\tau)\rangle, \tag{19}$$

where

$$\underline{\underline{G}}_{k}(\sigma)\equiv\langle \underline{A}_{\vec{k}}(\sigma)\underline{A}_{-\vec{k}}\rangle\langle \underline{A}_{\vec{k}}\underline{A}_{-\vec{k}}\rangle^{-1} \tag{20}$$

and

$$\dot{\underline{A}}_{\vec{k}}(t)\equiv e^{i(1-P)Lt}(1-P)iL\underline{A}_{\vec{k}} \tag{21}$$

$$\equiv i\vec{k}\cdot\underline{\vec{J}}^{\dagger}_{\vec{k}}(t). \tag{21'}$$

Equations (21) define Mori's random flux.[11] Note that

$$I_{\delta,T}(-\tau)=\lim_{\vec{k}\to 0}\vec{J}^{\dagger}_{\delta,\vec{k}}(-\tau).$$

The first term on the right-hand side (RHS) of Eq. (19) has been analyzed in Ref. 2, yielding a Lorentzian line shape. However, more care must be used in treating the second term. Denoting the second term on the RHS of Eq. (19) as $\underline{\underline{\Psi}}(\vec{k},\sigma)$, we find

$$\underline{\underline{\Psi}}(\vec{k},\sigma)=\int_0^{\sigma} dt_1\underline{\underline{G}}_{\vec{k}}(\sigma-t_1)\cdot\int_{-\infty}^{\infty} d\tau\langle[e^{iL\tau}\dot{\underline{A}}^{\dagger}_{\vec{k}}(t_1)]\underline{A}_{-\vec{k}}(\tau)I_{\delta,T}\rangle. \tag{22}$$

Using the identity

$$e^{iL\tau}e^{i(1-P)iLt_1}(1-P)iL=e^{i(1-P)L(t_1+\tau)}(1-P)iL+\int_0^{\tau} dt_2 e^{iL(\tau-t_2)}PiLe^{i(1-P)L(t_2+t_1)}(1-P)iL, \tag{23}$$

Eq. (22) gives

$$\underline{\underline{\Psi}}(\vec{k},\sigma) = \int_0^\sigma dt_1 \underline{\underline{G}}_{\vec k}(\sigma - t_1)\cdot\int_{-\infty}^{\infty} d\tau\left(\langle \dot{\underline{A}}^\dagger_{\vec k}(t_1)\underline{A}_{-\vec k}(\tau)I_{6,T}\rangle - \int_0^\tau dt_2 \langle \dot{\underline{A}}^\dagger_{\vec k}(t_2+t_1)\dot{\underline{A}}^\dagger_{-k}\rangle\cdot\langle \underline{A}_{\vec k}\underline{A}_{-\vec k}\rangle^{-1}\cdot\langle \underline{A}_{\vec k}(\tau - t_2)\underline{A}_{-\vec k}(\tau)I_{6,T}\rangle\right). \tag{24}$$

The assumption that random flux correlations decay quickly allows us to neglect the first term in Eq. (24). The remaining term may be approximated by

$$\underline{\underline{\Psi}}(\vec{k},\sigma) = -\int_0^\sigma dt_1 \underline{\underline{G}}_{\vec k}(\sigma - t_1)\cdot\int_{-\infty}^{\infty} d\tau\int_0^\tau dt_2\langle \dot{\underline{A}}^\dagger_{\vec k}(t_2+t_1)\dot{\underline{A}}^\dagger_{\vec k}\rangle\cdot\langle \underline{A}_k\underline{A}_{-k}\rangle^{-1}\cdot\langle \underline{A}_{\vec k}(\tau+t_1)\underline{A}_{-\vec k}(\tau)I_{6,T}\rangle. \tag{25}$$

For convenience, take $\sigma > 0$ (the $\sigma < 0$ behavior follows using the time-reversal properties of $\underline{\underline{\Lambda}}$). Using the assumed rapid decay properties of $\dot{\underline{A}}^\dagger_{\vec k}(t)$ allows us to rewrite Eq. (25) as

$$\Psi(\vec{k},\sigma) = -\int_0^\sigma dt_1 \underline{\underline{G}}_{\vec k}(\sigma - t_1)\cdot\int_{-\infty}^{-t_1} d\tau\int_{-\infty}^{\infty} dt_2\langle \dot{A}^\dagger_{\vec k}(t_2)\dot{\underline{A}}^\dagger_{\vec k}\rangle\langle \underline{A}_{\vec k}\underline{A}_{-\vec k}\rangle^{-1}\langle \underline{A}_{\vec k}(\tau+t_1)\underline{A}_{-\vec k}(\tau)I_{6,T}\rangle. \tag{26}$$

Recognizing

$$-\int_{-\infty}^{\infty} dt_2\langle \dot{\underline{A}}^\dagger_{\vec k}(t_2)\dot{\underline{A}}^\dagger_{-\vec k}\rangle\langle \underline{A}_{\vec k}\underline{A}_{-\vec k}\rangle^{-1} = 2\underline{\underline{M}}_{k,D}, \tag{27}$$

where $M_{k,D}$ is the Green-Kubo form[11] for the dissipative part of the hydrodynamic equations [i.e., $\dot{\underline{A}}_{\vec k}(t) =$ reversible terms $+\underline{\underline{M}}_{\vec k,D}\underline{A}_k(t)$] and using Eq. (17) shows that

$$\underline{\underline{\Psi}}(\vec{k},\sigma) = 2\int_0^\sigma dt_1\int_{t_1}^{\infty} d\tau\, \underline{\underline{G}}_{\vec k}(\sigma - t_1)\underline{\underline{M}}_{\vec k,D}\underline{\underline{G}}_{\vec k}(t_1-\tau) \times\langle \underline{A}_{\vec k}\underline{A}_{-\vec k}I_{6,T}\rangle\underline{\underline{G}}^\dagger_{\vec k}(-\tau) + \cdots, \tag{28}$$

where the ellipses represents less important terms. The superscript "†" denotes a Hermitian conjugate.

For hydrodynamics $\underline{\underline{M}}_{\vec k,D}$ is $O(k^2)$. Nonetheless, $\Psi(\vec{k},\sigma)$ is not negligible since all integrands decay on $O(k^2)$ time scales. In fact, making the hydrodynamic approximation for $\underline{\underline{G}}_{\vec k}(t)$ and $\underline{\underline{M}}_{\vec k,D}$, and using the small-$k$ form for $\langle \underline{A}_{\vec k}\underline{A}_{-\vec k}I_{6,T}\rangle$ (cf. Sec. IV of Ref. 2) yields an expression which, when Fourier transformed, is a square of Lorentzian. Combining this with the Lorentzian term in $\underline{\underline{\Lambda}}$ gives a correction to the light scattering equivalent to that found here. Physically, the source of the new term lies in the fact that the integrand in the second term on the RHS of Eq. (19) has a small part $O(k^2)$ which is long lived in time. In Ref. 2 a different definition of the random force is used and a Lorentzian result is obtained.[12] It can be shown that the difference in forces is $O(k^2)$ and long lived in time. Depending upon which definition of the random force rapidly decays, a time correlation which is either Lorentzian or Lorentzian squared can be obtained. Within the context of the assumptions of the various theories one cannot decide which peak structure is correct. Also experiments on light scattering will not be useful in this regard. Perhaps a density expansion[10] or an investigation of mode coupling[13] will yield additional insight.

One of us (D.R.) would like to thank the Department of Chemistry at U.C.L.A. for their hospitality while this work was carried out, and acknowledge a grant from the Miller Foundation. We acknowledge discussions with I. Procaccia and T. Kirkpatrick regarding the relationship of $1/f$ noise to the long-range correlations. We thank E. G. D. Cohen for critical comments.

*Permanent address: Department of Chemistry, Harvard University, Cambridge, Ma. 02138.

[1]R. Fox and G. E. Uhlenbeck, Phys. Fluids 13, 1893 (1970); 13, 2881 (1970), and references therein.

[2]D. Ronis, I. Procaccia, and I. Oppenheim, Phys. Rev. A 19, 1324 (1979).

[3]T. Kirkpatrick, E. G. D. Cohen, and J. R. Dorfman, Phys. Rev. Lett. 42, 862 (1979).

[4]It has come to our attention that A. Tremblay, E. Siggia, and M. Arai (unpublished), have obtained equivalent results with a similar calculation.

[5]S_{nl} should not be confused with the so-called nonlocality corrections of Ref. 2.

[6]Similar anomalies for other correlations in the case of steady flow are discussed by J. Machta, I. Oppenheim, and I. Procaccia, Phys. Rev. Lett. 42, 1368 (1979), and by Ref. 4.

[7]That the hydrodynamic method yields statics in agreement with those derived in Ref. 2 has been shown for fairly general macroscopic systems. [D. Ronis, I. Procaccia, and J. Machta, Phys. Rev. A (in press)]. The method used in Ref. 2 has been verified at low density by a microscopic approach [D. Ronis, I. Procaccia, and I. Oppenheim, Phys. Rev. A 19, 1307

(1979)].

[8]A general discussion of the calculation of the dynamic spectrum from the regression of a small fluctuation in a near-equilibrium stationary state is given by I. Procaccia, D. Ronis and I. Oppenheim, Phys. Rev. A 20, 2533 (1979).

[9]S. Putterman, *Superfluid Hydrodynamics* (North-Holland, Amsterdam, 1974), Chap. IV.

[10]T. Kirkpatrick, E. G. D. Cohen, and J. R. Dorfman, Phys. Rev. Lett. (in press) and private communications. The mode-coupling aspect of their calculations will be given in a future paper.

[11]See, e.g., D. Forster, *Hydrodynamic Fluctuations Broken Symmetry and Correlation Functions* (Benjamin, Reading, Mass., 1975).

[12]The form of the random force used in Ref. 2 has also been questioned by T. Kirkpatrick, J. Dorfman, and E. G. D. Cohen (private communication).

[13]I. Oppenheim (unpublished).