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On the Physical Origin of Long-Ranged Fluctuations in Fluids in Thermal Nonequilibrium States

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Thermodynamic fluctuations in systems that are in nonequilibrium steady states are always spatially long ranged, in contrast to fluctuations in thermodynamic equilibrium. In the present paper we consider a fluid subjected to a stationary temperature gradient. Two different physical mechanisms have been identified by which the temperature gradient causes long-ranged fluctuations. One cause is the presence of couplings between fluctuating fields. Secondly, spatial variation of the strength of random forces, resulting from the local version of the fluctuation-dissipation theorem, has also been shown to generate long-ranged fluctuations. We evaluate the contributions to the long-ranged temperature fluctuations due to both mechanisms. While the inhomogeneously correlated Langevin noise does lead to long-ranged fluctuations, in practice, they turn out to be negligible as compared to nonequilibrium temperature fluctuations resulting from the coupling between temperature and velocity fluctuations.

KEY WORDS: Fluctuating hydrodynamics; long-ranged correlations; nonequilibrium fluctuations; nonequilibrium steady states; temperature fluctuations.

1. INTRODUCTION

Equal-time (or static) correlation functions of fluctuating dynamical variables in fluids in thermodynamic equilibrium states are generically spatially short ranged, except in the vicinity of a critical point. On the other hand, in nonequilibrium steady states, correlations of thermodynamic fluctuations become spatially long ranged,(1–3) typically encompassing the spatial size of the system.(2, 4)

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A theoretical treatment of fluctuations in nonequilibrium states is commonly based on fluctuating hydrodynamics. Fluctuating hydrodynamics was originally formulated for dealing with fluctuations of hydrodynamic variables in fluids in thermodynamic equilibrium states. The ordinary balance equations of hydrodynamics are transformed into a set of stochastic partial differential equations by assuming that the dissipative fluxes contain an average part (related to the corresponding forces by the usual linear laws), plus an stochastic part (which averages to zero). Since fluctuations in equilibrium are small, the resulting equations for the fluctuating fields are linearized, yielding a set of coupled Langevin equations where stochastic dissipative fluxes play the role of random-noise terms.[5, 6] The autocorrelation functions of the different components of the random dissipative fluxes are then related to the relevant thermodynamic and transport properties by application of the fluctuation-dissipation theorem[7] (FDT).

In nonequilibrium steady states one is interested in the fluctuations of the dynamical variables around their local equilibrium values. For this application fluctuating hydrodynamics is extended by applying a local version of the FDT.[8] A consequence of this extension is that the noise correlation functions in inhomogeneous steady states now, in principle, may become dependent on the position through the spatial dependence of the corresponding physical quantities. Thus, in nonequilibrium states, the correlations of the noise appearing in the fluctuating hydrodynamic equations are in many cases inhomogeneous.

A review of the literature indicates that nonequilibrium constraints may cause long-ranged nonequilibrium fluctuations by two different mechanisms:

1. A first mechanism results from the spatial dependence of the noise autocorrelation functions,[9–15] specified in accordance with a local FDT, as explained above.

2. A second mechanism is the generic existence of nonequilibrium hydrodynamic couplings between fluctuating fields.[3, 16–19] This generates long-ranged nonequilibrium fluctuations even in the absence of any spatial dependence of the noise correlation, as in the case of an isothermal fluid subjected to shear,[20–23] or isothermal reaction-diffusion problems.[24, 25]

The nonequilibrium hydrodynamic couplings (second mechanism) usually arise from the advection terms in the hydrodynamic equations. Since these same terms cause the equilibrium long-time tails in the correlation functions,[19] it is not a surprise that the importance of such mode-coupling effects for generating long-ranged fluctuations was initially demonstrated within the framework of kinetic theory by researchers familiar with long-time tails.[26–28] Generally the two mechanisms mentioned above have been investigated independently: some authors have focussed on the spatial dependence of the autocorrelation function of the Langevin forces and others have focussed on the effect of mode couplings. As a consequence, the relative importance of these two physical mechanisms for originating long-ranged fluctuations in actual systems has remained unclear.

To elucidate the relative importance of the two mechanisms we consider here an incompressible fluid with a nonzero thermal expansion coefficient ($\alpha \neq 0$) subjected to a stationary temperature gradient. This situation can be realized in practice by confining the fluid between two horizontal plates that are maintained at two different temperatures. We shall refer to this case as the Rayleigh–Bénard problem[29] with the understanding that we only consider stationary nonequilibrium states below the onset of convection. This condition is satisfied for states with negative Rayleigh numbers (the density gradient induced by the temperature gradient coinciding with the direction of gravity) and for states with positive Rayleigh numbers (the density gradient induced by the temperature gradient being opposite to the direction of gravity) below the critical Rayleigh number R_c. The Rayleigh–Bénard problem, thus defined, is of special interest, since for this problem we are able to evaluate the autocorrelation function of the nonequilibrium temperature fluctuations taking into account both mechanisms so as to obtain an assessment of their mutual importance. Moreover, for this case the theoretical predictions can be readily tested by light-scattering experiments[30, 31] or shadowgraph experiments.[32, 33]

Our calculation of the contributions to the long-ranged nonequilibrium temperature fluctuations from the two mechanisms will proceed in two steps. First, in Section 2, we neglect the hydrodynamic coupling between temperature and velocity fluctuations, so that the Rayleigh–Bénard problem reduces to a simple heat-conduction problem. Specifically, we show how the spatial dependence of the autocorrelation function of the random heat flux leads to the presence of long-ranged temperature fluctuations. Next, in Section 3, we consider the same Rayleigh–Bénard problem but no longer neglect the hydrodynamic coupling between temperature and velocity fluctuations. Specifically, we show how the existence of a coupling between temperature and velocity fluctuations further contributes to the generic long-ranged nature of the equal-time autocorrelation function of the temperature fluctuations. For mathematical simplicity we do not include any effects from gravity, which for negative Rayleigh numbers turns out to be a good approximation.[34] Finally, in Section 4, we analyze the relative importance of the nonequilibrium fluctuations arising from both mechanisms. We conclude that the effects due to the spatial inhomogeneity of the noise correlations in the Rayleigh–Bénard problem are completely negligible as compared to the long-ranged nonequilibrium fluctuations induced by the coupling between temperature and transverse-velocity fluctuations. We also discuss in Section 4 how this conclusion may be understood in a more general context.

2. NONEQUILIBRIUM FLUCTUATIONS DUE TO INHOMOGENEOUSLY CORRELATED RANDOM HEAT FLUX

We consider a system bounded by two horizontal plates located at $z=0$ and $z=L$ and maintained at different temperatures. The local temperature in the system between the plates can be decomposed as:

$$T(\mathbf{r}, t) = T_0(\mathbf{r}) + \delta T(\mathbf{r}, t), \tag{1}$$

where $T_0(\mathbf{r})$ is the average temperature at position $\mathbf{r}$. If the temperatures of the two plates are kept constant, the system reaches a steady state and $T_0(\mathbf{r})$ does not actually depend on the time t. If we assume the thermal conductivity λ of the sample to be constant in the relevant range of temperatures, the average temperature T_0 will vary linearly with the vertical coordinate z:

$$T_0(z) = \overline{T_0}\left[1+\beta\left(\frac{z}{L}-\frac{1}{2}\right)\right], \tag{2}$$

where $\overline{T_0} = T_0(L/2)$ is the average temperature in the layer and $\beta = L(\nabla T_0)/\overline{T_0}$, with ∇T_0 being the magnitude of the uniform vertical temperature gradient.

In this section we consider only temperature fluctuations $\delta T(\mathbf{r}, t)$ and disregard any possible fluctuations of the fluid velocity. As mentioned in Section 1, we do so as to concentrate here only on nonequilibrium effects arising from inhomogeneously correlated noise, but the analysis can also be considered as applying to heat conduction in a solid. If we neglect the coupling between velocity and temperature fluctuations, the spatiotemporal evolution of the temperature fluctuations $\delta T(\mathbf{r}, t)$ will be governed by the heat-conduction equation:

$$\frac{\partial}{\partial t}\,\delta T(\mathbf{r}, t) = D_T \nabla^2\, \delta T(\mathbf{r}, t) - \frac{1}{\rho c_P}\,\nabla\cdot\delta\mathbf{Q}(\mathbf{r}, t), \tag{3}$$

where D_T is the local thermal diffusivity, ρ the local density, and c_P the local isobaric specific heat capacity. In Eq. (3), $\delta\mathbf{Q}(\mathbf{r}, t)$ denotes a random heat flux in accordance with the principle of fluctuating hydrodynamics.[5, 6]

The average value of $\langle \delta\mathbf{Q}(\mathbf{r}, t)\rangle = 0$, and the correlations among their components are given by:[5, 17]

$$\begin{aligned}\langle \delta Q_i^*(\mathbf{r}, t)\cdot\delta Q_j(\mathbf{r}', t')\rangle &= 2k_{\mathrm{B}}\lambda T_0^2(\mathbf{r})\,\delta(\mathbf{r}-\mathbf{r}')\,\delta(t-t')\,\delta_{ij}\\ &= 2k_{\mathrm{B}}\lambda\overline{T_0}^{\,2}\left[1+\beta\left(\frac{z}{L}-\frac{1}{2}\right)\right]^2\delta(\mathbf{r}-\mathbf{r}')\,\delta(t-t')\,\delta_{ij},\end{aligned}\tag{4}$$

where k_{B} is Boltzmann's constant. Equation (4) is the fluctuation-dissipation theorem (FDT) for fluctuations around equilibrium, but the equilibrium temperature has been replaced with its local value $T_0(\mathbf{r})$ as given by Eq. (2) In applying a local equilibrium version of the FDT we are making an assumption which, ultimately, requires justification from microscopic nonequilibrium statistical mechanics.[8, 35] We interpret Eq. (3) as a linear Langevin equation where the term $\nabla\cdot\delta\mathbf{Q}(\mathbf{r}, t)$ plays the role of an inhomogeneously correlated random force. This random force is expressed as the divergence of a vector to assure that fluctuations preserve local energy balance.

To solve Eq. (3), we perform Fourier transforms in time and in the horizontal XY-plane. However, to incorporate boundary conditions for the fluctuations $\delta T(\mathbf{r}, t) = 0$ at $z = 0$ and $z = L$, we do not Fourier transform in the z-coordinate, but instead expand the solution in a Fourier sine series:

$$\delta T(\omega, \mathbf{q}_{\parallel}, z) = \sum_{N=1}^{\infty} T_N(\omega, \mathbf{q}_{\parallel})\sin\left(\frac{N\pi}{L}z\right),\tag{5}$$

where ω is the frequency of the fluctuations and $\mathbf{q}_{\parallel} = \{q_x, q_y\}$ is the wave vector of the fluctuations in the horizontal XY-plane. Substituting Eq. (5) into Eq. (3), assuming that all the thermophysical properties are constants, we readily solve for the coefficients $T_N(\omega, \mathbf{q}_{\parallel})$ and obtain:

$$T_N(\omega, \mathbf{q}_{\parallel}) = \frac{-F_N(\omega, \mathbf{q}_{\parallel})}{\rho c_P[\mathrm{i}\omega + D_T(\frac{N^2\pi^2}{L^2}+q_{\parallel}^2)]},\tag{6}$$

where $q_{\parallel}^2 = q_x^2 + q_y^2$, and where $F_N(\omega, \mathbf{q}_{\parallel})$ are the Fourier sine series coefficients of the random force $\nabla\cdot\delta\mathbf{Q}(\mathbf{r}, t)$:

$$F_N(\omega, \mathbf{q}_{\parallel}) = \frac{2}{L}\int_0^L dz\,\sin\left(\frac{N\pi}{L}z\right)[\nabla\cdot\delta\mathbf{Q}](\omega, \mathbf{q}_{\parallel}, z).\tag{7}$$

In Eq. (7),

$$[\nabla\cdot\delta\mathbf{Q}](\omega,\mathbf{q}_{\parallel},z)=\mathrm{i}q_x\,\delta Q_x(\omega,\mathbf{q}_{\parallel},z)+\mathrm{i}q_y\,\delta Q_y(\omega,\mathbf{q}_{\parallel},z)+\partial_z\,\delta Q_z(\omega,\mathbf{q}_{\parallel},z)$$

is the Fourier transform of $\nabla\cdot\delta\mathbf{Q}(\mathbf{r},t)$ in time and in the horizontal x and y coordinates. To calculate the correlation function $\langle\delta T^*(\omega,\mathbf{q}_{\parallel},z)\cdot\delta T(\omega',\mathbf{q}'_{\parallel},z')\rangle$ of the temperature fluctuations we need the correlation functions $\langle F_N^*(\omega,\mathbf{q}_{\parallel})\cdot F_M(\omega',\mathbf{q}'_{\parallel})\rangle$ of the Fourier components of the random force $[\nabla\cdot\delta\mathbf{Q}](\omega,\mathbf{q}_{\parallel},z)$. These correlation functions can be computed from the definition (7) of $F_N(\omega,\mathbf{q}_{\parallel})$ by applying a double Fourier transform in t and t' and in the horizontal variables x and y, to the right-hand side of Eq. (4). We then find that these correlation functions can be expressed as:

$$\langle F_N^*(\omega,\mathbf{q}_{\parallel})\cdot F_M(\omega',\mathbf{q}'_{\parallel})\rangle=\frac{8k_{\mathrm{B}}\lambda\overline{T_0}^{\,2}}{L^3}\,(2\pi)^3\,\delta(\omega-\omega')\,\delta(\mathbf{q}_{\parallel}-\mathbf{q}'_{\parallel})\,\tilde{A}_{NM}(q_{\parallel}),\tag{8}$$

where the dimensionless functions $\tilde{A}_{NM}(q_{\parallel})$ are given by:

$$\begin{aligned}\tilde{A}_{NM}(q_{\parallel})=L\int_0^L\int_0^L dz\,dz'\,\sin\left(\frac{N\pi}{L}z\right)\sin\left(\frac{M\pi}{L}z'\right)\\ \times(q_{\parallel}^2+\partial_z\partial_{z'})\cdot\left\{\left[1+\beta\left(\frac{z}{L}-\frac{1}{2}\right)\right]^2\delta(z-z')\right\}.\end{aligned}\tag{9}$$

To evaluate the double integral of the derivatives of the delta function contained in Eq. (9), we perform a couple of integrations by parts to move the differential operators from the delta function to the preceding sine functions. The resulting integrals can be readily evaluated and the final expression for $\tilde{A}_{NM}(q_{\parallel})$ is presented in the Appendix (see Eq. (28)).

We now have all the information needed for the calculation of the autocorrelation function $\langle\delta T^*(\omega,\mathbf{q}_{\parallel},z)\cdot\delta T(\omega',\mathbf{q}'_{\parallel},z')\rangle$ of the temperature fluctuations. In view of Eq. (8) it is evident that it will be expressed as:

$$\langle\delta T^*(\omega,\mathbf{q}_{\parallel},z)\cdot\delta T(\omega',\mathbf{q}'_{\parallel},z')\rangle=F(\omega,q_{\parallel},z,z')(2\pi)^3\,\delta(\omega-\omega')\,\delta(\mathbf{q}_{\parallel}-\mathbf{q}'_{\parallel}),\tag{10}$$

which is expected, since translational symmetries in time and in the horizontal plane make the correlation function $\langle\delta T^*(\mathbf{r}_{\parallel},z,t)\cdot\delta T(\mathbf{r}'_{\parallel},z',t')\rangle$ to depend only on the differences $t-t'$ and $\mathbf{r}_{\parallel}-\mathbf{r}'_{\parallel}$. In Eq. (10) the function

$F(\omega, q_{\parallel}, z, z')$ is represented by a double Fourier series in the variables z and z':

$$F(\omega, q_{\parallel}, z, z') = \frac{8k_B \lambda \overline{T_0}^2}{L^3 \rho^2 c_P^2} \sum_{N=1}^{\infty} \sum_{M=1}^{\infty} \sin\left(\frac{N\pi}{L} z\right) \sin\left(\frac{M\pi}{L} z'\right) \times \frac{\tilde{A}_{NM}(q_{\parallel})}{[-\mathrm{i}\omega + D_T(\frac{N^2\pi^2}{L^2} + q_{\parallel}^2)][\mathrm{i}\omega + D_T(\frac{M^2\pi^2}{L^2} + q_{\parallel}^2)]}. \tag{11}$$

Our goal here is to calculate the *equal-time* (or static) autocorrelation function, $\langle \delta T^*(\mathbf{q}_{\parallel}, z, t) \cdot \delta T(\mathbf{q}'_{\parallel}, z', t) \rangle$. Applying a double inverse Fourier transform in ω and ω' to Eq. (10), we obtain:

$$\langle \delta T^*(\mathbf{q}_{\parallel}, z, t) \cdot \delta T(\mathbf{q}'_{\parallel}, z', t) \rangle = F(q_{\parallel}, z, z')(2\pi)^2\, \delta(\mathbf{q}_{\parallel} - \mathbf{q}'_{\parallel}), \tag{12}$$

where $F(q_{\parallel}, z, z') = (2\pi)^{-1} \int d\omega\, F(\omega, q_{\parallel}, z, z')$. The ω integration of Eq. (11) for $F(\omega, q_{\parallel}, z, z')$ yields:

$$F(q_{\parallel}, z, z') = \frac{8k_B \overline{T_0}^2}{\rho c_P L} \sum_{N=1}^{\infty} \sum_{M=1}^{\infty} \frac{\tilde{A}_{NM}(\tilde{q}_{\parallel})}{(N^2\pi^2 + M^2\pi^2 + 2\tilde{q}_{\parallel}^2)} \times \sin\left(\frac{N\pi}{L} z\right) \sin\left(\frac{M\pi}{L} z'\right), \tag{13}$$

where $\tilde{q}_{\parallel} = q_{\parallel} L$ is a dimensionless wave number, and where we have made use of the definition of the thermal diffusivity $D_T = \lambda / \rho c_P$. With the help of Eqs. (30a) and (30b) presented in the Appendix, the sum of the trigonometric series above over the index M can be performed exactly, and we finally obtain:

$$F(q_{\parallel}, z, z') = \frac{k_B}{\rho c_P} T_0^2(z)\, \delta(z - z') + F_{\mathrm{NE}}(q_{\parallel}, z, z'), \tag{14}$$

where we have collected the nonequilibrium contribution into the function:

$$F_{\mathrm{NE}}(q_{\parallel}, z, z') = \frac{2k_B \overline{T_0}^2}{\rho c_P L} \sum_{N=1}^{\infty} \frac{\beta^2}{N^2\pi^2 + \tilde{q}_{\parallel}^2} \sin\left(\frac{N\pi}{L} z\right) \sin\left(\frac{N\pi}{L} z'\right). \tag{15}$$

We observe in Eq. (14) that the equal-time autocorrelation function for the temperature fluctuations is expressed as a sum of a short-ranged local equilibrium contribution and a long-ranged nonequilibrium contribution, the latter given by Eq. (15). The local-equilibrium contribution is the same as the one obtained from the theory of equilibrium fluctuations,[36, 37] but with the uniform equilibrium temperature replaced with the local nonequilibrium value, $T_0(\mathbf{r})$, which in our case is given by Eq. (2).

It is evident that the nonequilibrium contribution (15) to the temperature fluctuations arises from the fact that we have used a local version of the FDT, so that the right-hand side of Eq. (4) has become dependent on the coordinate z.

Since we have neglected here the presence of velocity fluctuations, the problem of fluctuations in a fluid subjected to a stationary ∇T_0 reduces to that of fluctuations in the nonequilibrium heat-conduction equation, which case has been considered by a number of other authors. Actually, if we take $q_{\parallel}=0$ in Eq. (15), we reproduce previous results of Garcia *et al.*[11] and of Malek Mansour *et al.*,[12] for the horizontal average of the temperature fluctuations. Alternatively, taking $q_{\parallel}=0$ in Eq. (15) can be interpreted as reducing the original problem to one spatial dimension, in which case we reproduce the result of Breuer and Petruccione,[14] obtained by solving the one-dimensional Fokker–Planck equation corresponding to the Langevin equation considered here.

The sum of the series in Eq. (15) may be performed exactly, yielding:

$$F_{\mathrm{NE}}(q_{\parallel}, z, z') = \frac{k_{\mathrm{B}} \overline{T_0}^2 \beta^2}{2\rho c_P L} \left\{ \frac{\cosh[\tilde{q}_{\parallel}(1-\frac{|z-z'|}{L})]}{\tilde{q}_{\parallel}\sinh(\tilde{q}_{\parallel})} - \frac{\cosh[\tilde{q}_{\parallel}(1-\frac{z+z'}{L})]}{\tilde{q}_{\parallel}\sinh(\tilde{q}_{\parallel})} \right\}. \tag{16}$$

The nonequilibrium contribution to the equal-time correlation function in real space is obtained by applying a double inverse Fourier transform to $\langle \delta T^*(\mathbf{q}_{\parallel}, z, t) \cdot \delta T^*(\mathbf{q}'_{\parallel}, z', t) \rangle$. As is to be expected from the translational symmetry of the problem, it will depend on the difference $\mathbf{r}_{\parallel} - \mathbf{r}'_{\parallel}$. Specifically, separating $\langle \delta T^*(\mathbf{r}, t) \cdot \delta T(\mathbf{r}', t) \rangle$ into an equilibrium part and a nonequilibrium part, we obtain for the nonequilibrium contribution to the intensity of temperature fluctuations:

$$\langle \delta T^*(\mathbf{r}, t) \cdot \delta T(\mathbf{r}', t) \rangle_{\mathrm{NE}} = \frac{2k_{\mathrm{B}} \overline{T_0}^2 \beta^2}{\rho c_P L} \int_0^{\infty} dq_{\parallel} \sum_{N=1}^{\infty} \frac{2\pi q_{\parallel} J_0(q_{\parallel} r_{\parallel})}{N^2\pi^2 + \tilde{q}_{\parallel}^2} \sin\left(\frac{N\pi}{L} z\right) \sin\left(\frac{N\pi}{L} z'\right), \tag{17}$$

where $r_{\parallel}$ is the distance in the horizontal XY-plane between the two points $\mathbf{r}$ and $\mathbf{r}'$ at which the correlation function is evaluated. Equation (17) diverges when $\mathbf{r} = \mathbf{r}'$, indicating that, in three-dimensional heat conduction, some short-range nonequilibrium contribution is present in addition to the long-ranged contribution.

We emphasize that some previous results in the literature[11, 12] refer to averages over the horizontal plane, while we have obtained here the complete expression for the temperature fluctuations as a function of the wave number $q_{\parallel}$. The divergence of $\langle \delta T^{*}(\mathbf{r}, t) \cdot \delta T(\mathbf{r}', t) \rangle_{\mathrm{NE}}$ in real space when $\mathbf{r} = \mathbf{r}'$, is eliminated by the averaging procedure used elsewhere.[11, 12] Such horizontal averaging makes the three-dimensional problem equivalent to one-dimensional heat conduction in the segment $[0, L]$, studied by Breuer and Petruccione,[14] where such a divergence does not exist. Probably the presence of a short-range contribution manifested as a divergence in real space when $\mathbf{r} = \mathbf{r}'$, is the cause of the disagreement found[11] with the results of Liu and Oppenheim,[38] the latter referring to the actual correlation function, as considered here.

It is also interesting to note that, in the heat-diffusion problem considered here, the long-ranged nonequilibrium contribution to the temperature fluctuations is proportional to ∇T_0^2; in contrast to a nonequilibrium correction proportional to $\nabla \ln(T_0)$ found for the mass-diffusion problem by Tremblay *et al.*[10] The reason for that is the fact that in the FDT for the random heat flow the temperature appears squared, while in the FDT for random diffusive fluxes considered by Tremblay *et al.*,[10] the temperature itself appears as a multiplicative factor.

We conclude this section by pointing out that the validity of Eq. (17) to describe the nonequilibrium temperature fluctuations, averaged over a horizontal plane ($q_{\parallel} = 0$), has been verified by numerically solving the stochastic heat-conduction equation.[11]

3. NONEQUILIBRIUM FLUCTUATIONS DUE TO HYDRODYNAMIC COUPLING BETWEEN FLUCTUATING FIELDS

To evaluate the long-ranged nonequilibrium temperature fluctuations that originate from the coupling between hydrodynamic modes, we consider the same problem as in Section 2, but we now take into account that velocity fluctuations are present in addition to temperature fluctuations. We consider fluctuations around the steady-state conductive solution, where the temperature distribution $T_0(z)$ is again given by Eq. (2), and where the local fluid velocity averaged over fluctuations is everywhere zero. Specifically, we need to account for a coupling between temperature fluctuations and the fluctuations of the velocity component parallel to the gradient, the contribution of which was neglected in Section 2. To describe the spatiotemporal evolution of fluctuations around (2) we shall use here the Boussinesq approximation for the equations of fluctuating hydrodynamics. Use of the Boussinesq approximation implies that we neglect the sound modes responsible for the Brillouin component of the structure factor and consider only contributions from the temperature fluctuations to the structure factor.[4, 29]

To further simplify the calculations of this section, we shall neglect gravity effects, which turns out to be a good approximation in the case of negative Rayleigh numbers,[4] for which the conductive state (2) will always be stable. Then the spatiotemporal evolution of the temperature and velocity fluctuations will be given by:

$$\frac{\partial}{\partial t}\,\delta T(\mathbf{r},t)=D_T\nabla^2\,\delta T(x,t)-\nabla T_0\,\delta u_z(\mathbf{r},t)-\frac{1}{\rho c_P}\,\nabla\cdot\delta\mathbf{Q}(\mathbf{r},t), \tag{18a}$$

$$\frac{\partial}{\partial t}\,[\nabla^2\,\delta u_z(\mathbf{r},t)]=\nu\,\nabla^2[\nabla^2\,\delta u_z(\mathbf{r},t)]+\frac{1}{\rho}\,\{\nabla\times\nabla\times[\nabla\cdot\delta\Pi(\mathbf{r},t)]\}_z, \tag{18b}$$

where $\delta u_z(\mathbf{r},t)$ are the fluctuations in the Z-component of the fluid velocity, and where ν is the kinematic viscosity of the fluid. Equations (18) are the linearized Boussinesq equations supplemented with random noise terms in the absence of gravity.[4, 29] In accordance with fluctuating hydrodynamics, we now need to consider in addition to $\delta\mathbf{Q}(\mathbf{r},t)$ a second random dissipative flux, which is a random viscous stress tensor $\delta\Pi(\mathbf{r},t)$, appearing in Eq. (18b) for $\delta u_z(\mathbf{r},t)$. The correlations among the components of $\delta\Pi(\mathbf{r},t)$ are again obtained by applying the FDT locally, so that:

$$\begin{aligned}
&\langle\delta\Pi_{ij}(\mathbf{r},t)\cdot\delta\Pi_{kl}(\mathbf{r}',t')\rangle\\
&\quad=2k_{\mathrm{B}}\eta T_0(z)(\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk})\,\delta(\mathbf{r}-\mathbf{r}')\,\delta(t-t')\\
&\quad=2k_{\mathrm{B}}\eta\overline{T_0}\left[1+\beta\left(\frac{z}{L}-\frac{1}{2}\right)\right](\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk})\,\delta(\mathbf{r}-\mathbf{r}')\,\delta(t-t'),
\end{aligned} \tag{19}$$

where η is the shear viscosity ($\eta=\nu\rho$). As regards a comparison with the classical work of Landau and Lifshitz,[5, 17] we note that we are using here the FDT for an incompressible fluid, so that the contribution of the bulk viscosity ζ is neglected. Just as in Eq. (4) for the random heat flux, Eq. (19) contains the local average temperature T_0, which depends on z in accordance to Eq. (2). Regarding the boundary conditions, like in Section 2, we continue to use perfectly conducting walls for the temperature fluctuations: $\delta T(\mathbf{x}_\parallel,0,t)=\delta T(\mathbf{x}_\parallel,L,t)=0$. For the fluctuations in the vertical component of the velocity we adopt here, for the sake of mathematical simplicity, stress-free boundary conditions. These allow us to look for the solution of the Fourier-transformed Eqs. (18) in terms of a Fourier sine series:

$$\begin{aligned}
\delta T(\omega,\mathbf{q}_\parallel,z)&=\sum_{N=1}^{\infty}T_N(\omega,\mathbf{q}_\parallel)\sin\left(\frac{N\pi}{L}z\right),\\
\delta u_z(\omega,\mathbf{q}_\parallel,z)&=\sum_{N=1}^{\infty}w_N(\omega,\mathbf{q}_\parallel)\sin\left(\frac{N\pi}{L}z\right).
\end{aligned} \tag{20}$$

Fourier transforming Eqs. (18) in time t and in the horizontal coordinates x and y, representing the random-forces in a Fourier sine series, we readily deduce for the functions $T_N(\omega, \mathbf{q}_\parallel)$ and $w_N(\omega, \mathbf{q}_\parallel)$:

$$T_N(\omega, \mathbf{q}_\parallel) = \frac{F_N(\omega, \mathbf{q}_\parallel) - \nabla T_0\, w_N(\omega, \mathbf{q}_\parallel)}{\rho c_P[\mathrm{i}\omega + D_T(\frac{N^2\pi^2}{L^2} + q_\parallel^2)]}, \tag{21a}$$

$$w_N(\omega, \mathbf{q}_\parallel) = \frac{-G_N(\omega, \mathbf{q}_\parallel)}{\rho(\frac{N^2\pi^2}{L^2} + q_\parallel^2)[\mathrm{i}\omega + \nu(\frac{N^2\pi^2}{L^2} + q_\parallel^2)]}. \tag{21b}$$

In Eq. (21a) $F_N(\omega, \mathbf{q}_\parallel)$ are again the coefficients of the Fourier sine series for the Fourier-transform $[\nabla \cdot \delta\mathbf{Q}](\omega, \mathbf{q}_\parallel, z)$ of the random force $\nabla \cdot \delta\mathbf{Q}(\mathbf{r}, t)$ given by Eq. (7), while $G_N(\omega, \mathbf{q}_\parallel)$ in Eq. (21b) are the coefficients of the Fourier sine series for the corresponding Fourier transform $\{\nabla\times\nabla\times[\nabla\cdot\delta\mathit{\Pi}]\}_z\, (\omega, \mathbf{q}_\parallel, z)$ for the second random force $\{\nabla\times\nabla\times[\nabla\cdot\delta\mathit{\Pi}(\mathbf{r}, t)]\}_z$:

$$G_N(\omega, \mathbf{q}_\parallel) = \frac{2}{L}\int_0^L dz\, \sin\left(\frac{N\pi}{L} z\right) \{\nabla\times\nabla\times[\nabla\cdot\delta\mathit{\Pi}]\}_z\, (\omega, \mathbf{q}_\parallel, z). \tag{22}$$

To determine the autocorrelation function for the temperature fluctuations, we observe from Eq. (21a) that we need various correlation functions: $\langle F_N^*(\omega, q_\parallel) \cdot F_M(\omega', q_\parallel')\rangle$, $\langle F_N^*(\omega, q_\parallel) \cdot w_M(\omega', q_\parallel')\rangle$, and $\langle w_N^*(\omega, q_\parallel) \cdot w_M(\omega', q_\parallel')\rangle$. We first note that the random heat flux and the random stress tensor are uncorrelated, so that $\langle F_N^*(\omega, q_\parallel) \cdot w_M(\omega', q_\parallel')\rangle = 0$. For the same reason, the correlation function $\langle F_N^*(\omega, q_\parallel) \cdot F_M(\omega', q_\parallel')\rangle$ remains the same as the one calculated in Section 2 in the absence of velocity fluctuations (see Eqs. (8) and (9): this is the main reason why we presented the calculation in two steps). Finally, the correlation functions $\langle w_N^*(\omega, \mathbf{q}_\parallel) \cdot w_M(\omega', \mathbf{q}_\parallel')\rangle$ can be calculated from the definition (21) of $w_N(\omega, \mathbf{q}_\parallel)$, the definition (22) of $G_N(\omega, \mathbf{q}_\parallel)$, and the multiple Fourier transform of Eq. (19) in t and t' and in the horizontal variables x, x', y, and y'. After some long algebra we obtain

$$\begin{aligned}&\langle w_N^*(\omega, \mathbf{q}_\parallel) \cdot w_M(\omega', \mathbf{q}_\parallel')\rangle \\ &\quad = \frac{8k_\mathrm{B}\overline{T_0}}{\rho^2 L^5} \frac{(2\pi)^3\, \delta(\omega-\omega')\, \delta(\mathbf{q}_\parallel - \mathbf{q}_\parallel')\, q_\parallel^2\, \tilde{B}_{NM}(q_\parallel)}{(\frac{N^2\pi^2}{L^2} + q_\parallel^2)(\frac{M^2\pi^2}{L^2} + q_\parallel^2)[-\mathrm{i}\omega + \nu(\frac{N^2\pi^2}{L^2} + q_\parallel^2)][\mathrm{i}\omega + \nu(\frac{M^2\pi^2}{L^2} + q_\parallel^2)]}\end{aligned} \tag{23}$$

with

$$\tilde{B}_{NM}(q_{\parallel}) = L^3 \int_0^L dz \int_0^L dz' \sin\left(\frac{N\pi}{L} z\right) \sin\left(\frac{M\pi}{L} z'\right)$$
$$\times \left\{ \left[q_{\parallel}^4 + q_{\parallel}^2 \left(\frac{\partial^2}{\partial z^2} + \frac{\partial^2}{\partial z'^2} + 4 \frac{\partial}{\partial z} \frac{\partial}{\partial z'} \right) + \frac{\partial^2}{\partial z^2} \frac{\partial^2}{\partial z'^2} \right] \right.$$
$$\left. \times \left[1 + \beta \left(\frac{z}{L} - \frac{1}{2} \right) \right] \delta(z - z') \right\}. \tag{24}$$

To calculate the coefficients $\tilde{B}_{NM}(q_{\parallel})$, we follow the same procedure described in Section 2 for the calculation of $\tilde{A}_{NM}(q_{\parallel})$. The resulting expression is long and not very informative, so we give it in the Appendix (see Eq. (29)).

We now have all the ingredients needed to actually calculate the autocorrelation function of the temperature fluctuations. We first observe that, since both Eq. (8) and Eq. (23) contain products of delta functions, the autocorrelation $\langle \delta T^*(\omega, \mathbf{q}_{\parallel}, z) \cdot \delta T(\omega', \mathbf{q}'_{\parallel}, z') \rangle$, as expected, can be cast in the same form, Eq. (10), obtained in Section 2 in the absence of velocity fluctuations. In addition, since $\langle F_N^*(\omega, q_{\parallel}) \cdot w_M(\omega', q'_{\parallel}) \rangle = 0$, the function $F(\omega, q_{\parallel}, z, z')$ will be the same obtained in Eq. (11), plus some contribution from the coupling of the hydrodynamic modes. The expression is long, not very informative, and can be easily obtained by the reader, so we skip it here.

As in Section 2, we focus our attention on the equal-time (or static) autocorrelation function $\langle \delta T^*(q_{\parallel}, z, t) \cdot \delta T(q'_{\parallel}, z', t) \rangle$. Since the frequency-dependent correlation function is proportional to a delta function $\delta(\omega - \omega')$, the equal-time autocorrelation of the temperature fluctuations can be expressed in a form as given in Eq. (12), where the function $F(q_{\parallel}, z, z')$ is to be obtained by the integration in frequency of $F(\omega, q_{\parallel}, z, z')$. If we perform the integration, incorporating Eqs. (4) and (19) for the correlations of the random forces, and use some formulas presented in the Appendix, we arrive at

$$F(q_{\parallel}, z, z') = \frac{k_{\mathrm{B}}}{\rho c_P} T_0^2(z)\, \delta(z - z') + \frac{2 k_{\mathrm{B}} (\nabla T_0)^2 L}{\rho c_P}$$
$$\times \sum_{N=1}^{\infty} \left[\frac{1}{N^2\pi^2 + \tilde{q}_{\parallel}^2} + \frac{2 L^2 c_P \overline{T_0}}{D_T(\nu + D_T)} \frac{\tilde{q}_{\parallel}^2}{(N^2\pi^2 + \tilde{q}_{\parallel}^2)^3} \right]$$
$$\times \sin\left(\frac{N\pi}{L} z\right) \sin\left(\frac{N\pi}{L} z'\right), \tag{25}$$

where we have neglected some higher-order terms that are cubic in ∇T_0.

From Eqs. (12) and (25) we observe that there are three contributions to $\langle \delta T^*(\mathbf{q}_\parallel, z, t) \cdot \delta T(\mathbf{q}'_\parallel, z', t) \rangle$: First the local-equilibrium correlation function. The second contribution in Eq. (25) (in the form of a series expansion) is the same already found in Eq. (14) and it accounts for the inhomogeneously correlated random heat flux, in accordance with the local formulation of the FDT. The third contribution in Eq. (25) (again in the form of a series expansion) is due to the coupling between temperature and velocity fluctuations; it is the same as obtained previously,[4] where $T_0(z)$ was identified with T_0 in both Eqs. (4) and (19). The Z-dependence of the correlation function (19) of the random stress tensor only contributes terms cubic in ∇T_0, which we have neglected in Eq. (25). Our final result (25) shows how both mechanisms mentioned in the Introduction yield contributions to nonequilibrium fluctuations that are spatially long-ranged. To our knowledge, this is the first time that both effects have been evaluated for the same system.

To show the spatially long-ranged nature of the correlations more clearly, we plot in Fig. 1 the third contribution to Eq. (25), denoted $G(z, z')$, as a function of z', for two values of z. Figure 1 shows the nonequilibrium contribution in real space, so it is actually obtained by applying a double inverse Fourier transform to Eq. (25) in the variables $\mathbf{q}_\parallel$ and $\mathbf{q}'_\parallel$. The real-space correlation function depends on the horizontal distance $r_\parallel$ between the two points at which is evaluated. The data shown in Fig. 1 correspond to $r_\parallel = 0$. In addition $G(z, z')$ has been normalized, so as to make it dimensionless. A simple inspection of the figure shows that the nonequilibrium contribution to the correlation function is spatially long-ranged, does not involve any intrinsic length scale, and encompasses the entire system.

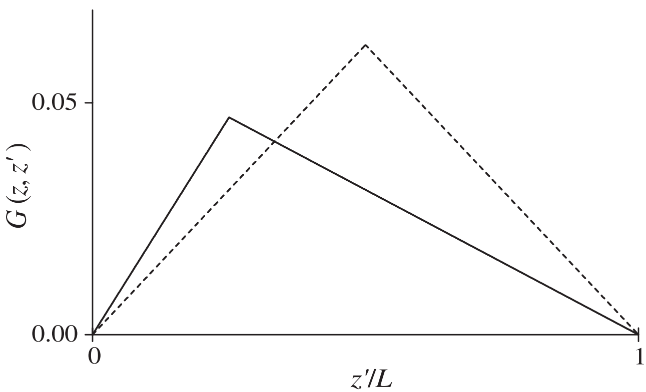

Fig. 1. Normalized nonequilibrium contribution to the structure factor arising from the hydrodynamic coupling between temperature and velocity fluctuations, $G(z, z')$, as a function of z' for $z = L/4$ (solid curve), and for $z = L/2$ (dashed curve). The spatially long-ranged nature of the nonequilibrium fluctuations is evident.

The long-ranged nature of nonequilibrium fluctuations can be probed by light-scattering experiments performed at different scattering wave vectors $\mathbf{q}$ (different scattering angles). As discussed elsewhere,[3, 4, 39] the intensity of light scattered by a fluid with scattering wave vector $\mathbf{q} = \{\mathbf{q}_\parallel, q_\perp\}$ is determined by a structure factor $S(\mathbf{q})$, defined by:

$$\alpha^2 \rho^2 S(\mathbf{q})(2\pi)^2\, \delta(\mathbf{q}_\parallel - \mathbf{q}'_\parallel) \\ = \frac{1}{L}\int_0^L dz \int_0^L dz'\, \mathrm{e}^{-\mathrm{i}\, q_\perp (z-z')}\, \langle \delta T^*(\mathbf{q}_\parallel, z, t) \cdot \delta T(\mathbf{q}'_\parallel, z', t)\rangle. \tag{26}$$

If we substitute in Eq. (26) the local-equilibrium contribution to $\langle \delta T^*(\mathbf{q}_\parallel, z, t) \cdot \delta T(\mathbf{q}'_\parallel, z', t)\rangle$, which is proportional to the first term in the right-hand side of Eq. (25), we find that in equilibrium the intensity of the scattered light does not depend on $\mathbf{q}$; hence, it is independent of the scattering angle. However, both nonequilibrium contributions in Eq. (25) depend on the wave number $q_\parallel$ and, hence, yield contributions to the scattered-light intensity that will depend on the scattering angle. In this way, the spatial long-ranged nature of the fluctuations may be probed experimentally. It should be noted that the divergence in real space of the nonequilibrium fluctuations due to the inhomogeneously correlated random heat flux, mentioned at the end of Section 2, does not cause any problem in real experiments. Integration over z and z' of Eq. (16), as required by Eq. (26), yields a convergent result for any value of $\mathbf{q}$.

In the discussion above we have assumed that the fluid has a nonzero thermal expansion coefficient α. When $\alpha = 0$, the temperature fluctuations do not contribute to the structure factor $S(q)$ (see Eq. (26)) so that the scattering function will only contain contributions from the sound modes yielding two Brillouin lines neglected here.

As noted at the end of Section 2, the contributions to the long-ranged temperature fluctuations from inhomogeneous correlated noise have to some extent been verified by computer simulations.[11] We do not know of any computer simulations of the contributions due to hydrodynamic coupling. However, the dependence of the Rayleigh component of the nonequilibrium structure factor on q^4 implied by Eq. (25) for large q has been verified experimentally.[30, 31]

4. DISCUSSION

Our initial motivation was to compare the contribution from the inhomogeneity of the thermal noise with the contribution from the non-equilibrium coupling between temperature and velocity fluctuations to the Rayleigh component of the nonequilibrium structure factor of a fluid in the presence of a temperature gradient. Previous studies[3, 39] have not considered the effect of the inhomogeneity in the thermal noise. As far as we know, we have performed for the first time a complete calculation for a case where both mechanisms, namely inhomogeneity of the noise correlation and coupling of hydrodynamic modes, are present. The resulting expression for the equal-time autocorrelation $\langle \delta T^*(\mathbf{q}_{\parallel}, z, t) \cdot \delta T(\mathbf{q}'_{\parallel}, z', t) \rangle$ of the temperature fluctuations is given by Eqs. (12) and (25). From Eq. (25) we observe that the contribution from a nonequilibrium coupling between temperature and velocity fluctuations (proportional to the second term inside the square brackets) relative to the contribution from the inhomogeneously correlated noise terms (proportional to the first term inside the square brackets) is determined by the dimensionless ratio:

$$\tilde{\mathcal{R}} = \frac{2L^2 c_P \overline{T_0}}{D_T(\nu + D_T)}. \tag{27}$$

If we consider a liquid layer with height $L = 2$ mm and if we adopt values of the physical quantities for toluene[40] at 25°C, we find $\tilde{\mathcal{R}} = 6 \times 10^{13}$. While long-ranged correlations due to inhomogeneously correlated random forces exist in principle, they are totally negligible in practice as compared to the long-ranged correlations resulting from a coupling between the temperature and velocity fluctuations. This conclusion is also supported by measurements of the nonequilibrium temperature fluctuations obtained from light-scattering experiments,[30, 31] which could be interpreted as arising exclusively from the mode-coupling effects.

While our analysis was specifically concerned with a fluid in the presence of a stationary temperature gradient, it is of interest to consider whether one can draw some more general conclusions about the role of the two mechanisms for generating long-ranged fluctuations in nonequilibrium states in fluids. First we note that there are cases where one of the two mechanisms through which nonequilibrium constraints cause long-ranged nonequilibrium fluctuations may be absent. One example is the case of an isothermal fluid under shear,[20–23] where the noise correlation is spatially uniform (uniform temperature), so that the first of the two mechanisms is absent. Nonequilibrium effects do appear in this case due exclusively to the hydrodynamic coupling between the uniform shear rate (nonequilibrium constraint) and the velocity fluctuations, through the advective term in the Navier–Stokes equation. Hence, in this case only the second of the mechanisms mentioned in Section 1 is responsible for the presence of long-ranged fluctuations.

Another example, of the opposite kind, are nonequilibrium effects in the Brillouin doublet. As mentioned earlier in Section 3, we have considered here the nonequilibrium Rayleigh component of the structure factor of a fluid in the presence of a temperature gradient. In addition, there are also long-ranged nonequilibrium fluctuations in the Brillouin components (an asymmetry in the Brillouin doublet) that, if studied in the simpler $\alpha = 0$ approximation, turn out to be exclusively due to the inhomogeneity of the thermal noise.[9, 10]

Finally, we note that the ratio $\tilde{\mathscr{R}}$, given by Eq. (27), is proportional to L^2. In molecular simulations with extremely small L the contribution from inhomogeneously correlated noise could become more important.

We conclude that a complete theory of fluctuations in nonequilibrium states requires that two different mechanisms need to be considered by which the nonequilibrium constrains induce spatially long-ranged fluctuations, as we have done for the Rayleigh–Bénard problem. However, as discussed above, in some particular cases one of the two mechanisms may be absent. The possibility of long-ranged fluctuations generated by anisotropic (but spatially uniform) correlated noise has been discussed by Maes and Reding[41, 42] and is not considered here.

Our conclusions are based on the application of fluctuating hydrodynamics to nonequilibrium states. The validity of this approach has been verified through microscopic simulations.[43] Fluctuating hydrodynamics is a mesoscopic theory which, ultimately, requires justification from microscopic models (note that the FDT is only fully rigorous for global equilibrium states). Unfortunately, a rigorous implementation of such a program is difficult, even for systems without dissipation.[35] Very recently, advances have been made by exactly solving some simple microscopic models.[44, 45] Evidently, a fully rigorous justification of nonequilibrium fluctuating hydrodynamics for real fluid systems continues to be one of the challenges of nonequilibrium statistical physics.

APPENDIX

In this appendix we first present the expressions for $\tilde{A}_{NM}(\tilde{q}_{\parallel})$ and $\tilde{B}_{NM}(\tilde{q}_{\parallel})$, obtained by evaluating the integrals contained in Eqs. (9) and (23), respectively:

$$\begin{aligned}\tilde{A}_{NM}(\tilde{q}_{\parallel}) = {} & \left\{\frac{N^2\pi^2+\tilde{q}_{\parallel}^2}{2}+\frac{\beta^2}{24}\left[N^2\pi^2+6+\frac{\tilde{q}_{\parallel}^2}{\pi^2N^2}(N^2\pi^2-6)\right]\right\}\delta_{NM} \\ & +2\beta NM\,\frac{(N^2\pi^2+M^2\pi^2+2\tilde{q}_{\parallel}^2)}{\pi^2(N^2-M^2)^2}\,[\cos(N\pi)\cos(M\pi)-1] \\ & +\beta^2 NM\,\frac{(N^2\pi^2+M^2\pi^2+2\tilde{q}_{\parallel}^2)}{\pi^2(N^2-M^2)^2}\,[\cos(N\pi)\cos(M\pi)+1](1-\delta_{NM}),\end{aligned} \tag{28}$$

$$\tilde{B}_{NM}(\tilde{q}_{\|}) = \frac{(N^2\pi^2+\tilde{q}_{\|}^2)^2}{2}\,\delta_{NM} + 2\beta NM\,\frac{(N^2\pi^2+\tilde{q}_{\|}^2)(M^2\pi^2+\tilde{q}_{\|}^2)}{\pi^2(N^2-M^2)^2}\,[\cos(N\pi)\cos(M\pi)-1]. \tag{29}$$

Secondly, to obtain Eqs. (14) and (15) for $F(q_{\|}, z, z')$ we need the sum of two trigonometric series. For this purpose use has been made of the following identities:

$$\sum_{M=1}^{\infty} \frac{M[\cos(N\pi)\cos(M\pi)-1]}{(M^2-N^2)^2}\sin(M\pi z) = \frac{\pi^2(2z-1)}{8}\sin(N\pi z), \tag{30a}$$

$$\sum_{M=1}^{\infty} \frac{M[\cos(N\pi)\cos(M\pi)+1](1-\delta_{MN})}{(M^2-N^2)^2}\sin(M\pi z) = \left[\frac{\pi^2(6z^2-6z+1)}{24N}+\frac{1}{8N^3}\right]\sin(N\pi z). \tag{30b}$$

Equations (30a) and (30b) are valid for nonzero positive integers N and for $z \in [0, 1]$. These expressions can be obtained from formulas 1.445 in Gradstein and Ryzhik,[46] after some lengthly algebra. They can be easily verified numerically.

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