Fluctuations in finite systems: Fluctuations in fluids in thermocapillary motion

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We compute nonequilibrium correlation functions about the stationary state in which the fluid moves as a consequence of tangential stresses on the liquid surface, related to a varying surface tension (thermocapillary motion). The nature of the stationary state makes it necessary to take into account that the system is finite. We then extend a previous analysis on fluctuations about simple stationary states to include some effects related to the finite size of the sample.

I. INTRODUCTION

Recently, a considerable amount of work has shown that fluctuating hydrodynamics provides a consistent scheme to analyze fluctuations in systems away from equilibrium (see, for example, Refs. 1 and 2 and references quoted therein). The procedure to arrive at the correlation functions from that theory has become standard. One formulates the stochastic differential equations for the fluctuating fields as Langevin-like equations, where the random sources are related to the random parts of the dissipative currents, accounting for fast variables. Then one formally solves such equations and computes the correlations in real or Fourier representations.

Although systems in stationary states are essentially of finite size, because of the energy or mass transfer through their boundaries, it turns out that the penetration lengths for fluctuations or the mean-free path of collective modes are sometimes much smaller than a characteristic length of the system. Thus for practical purposes one assumes that the system is infinite. This is what is normally assumed when dealing with density fluctuations in a fluid under a temperature gradient. In that case the preceding condition is $c/D_1k^2 \ll L$, where c is the sound velocity in the medium, L the length between plates, and $D_1=(4/3\eta+\xi)/\rho$, with ρ the mass density and η and ξ the shear and bulk viscosities, respectively. Such a condition imposes the cutoff wave number $k_L=1/L$.[7]

There are a number of interesting situations in which the preceding assumption cannot be made. This is what happens, for example, when the boundaries themselves act not only by providing simple boundary conditions but by modifying the nature of the stationary states or adding sochastic sources of noise.[3–5] In Refs. 3–6 fluctuating hydrodynamics was applied to get correlation functions in finite systems.

The purpose of this paper is precisely to provide an example of those cases in which the stationary state is a consequence of the existence of a temperature-dependent surface tension. Our work is distributed as follows. In Sec. II we comment on the nature of the stationary state. We also establish the stochastic differential equations for the evolution of the fluctuations. In Sec. III we give our expressions of the velocity and temperature correlation functions, which follow from an extension of the method used in.[7,8] We also evaluate the nonequilibrium corrections. In Sec. IV we will be concerned with some discussion remarks. In particular, we will stress some analogies and differences between the correlation functions in our system and in fluids under external gradients, such as the ones studied in Refs. 7 and 8.

II. FLUCTUATIONS ABOUT THE STATIONARY STATE

To study fluctuations we must, first of all, specify the nature of the stationary state. Let us consider a thin layer of an incompressible fluid of infinite extent in the x and z directions. In a realistic case the surface tension α of the liquid will depend on temperature. Then the action of a constant temperature gradient parallel to the surface will give rise to tangential stresses and, as a consequence, a motion of the fluid layer (referred to as thermocapillary motion[9]) will be produced. The temperature and velocity profiles then follow from the differential equations of nonequilibrium thermodynamics.[10] If the imposed temperature gradient ∇T is sufficiently small the velocity of the liquid will be also small. Therefore the convective and viscous heating terms of the internal energy balance equation can be neglected and the stationary temperature T_s reads

$$T_s(x)=T_0+x\,|\nabla T|\ , \tag{1}$$

where T_0 is a reference temperature. The stationary velocity follows from the Navier-Stokes equation when imposing the conditions

$$v_y \ll v_x,\quad v_x\frac{\partial v_x}{\partial x} \ll \nu\frac{\partial^2 v_x}{\partial y^2}\ , \tag{2}$$

where ν is the kinematic viscosity. The first condition is a consequence of the fact that the thickness of the fluid layer h is small compared with a characteristic length L along the x direction, in which thermocapillary motion takes place. The second condition is accomplished when $\mathcal{R}(h/L)\ll 1$, $\mathcal{R}$ being the Reynolds number equal to uh/ν, with $u=(h/4\eta)\partial\alpha/\partial x$. In view of the preceding hypothesis about the aspect ratio, Reynolds numbers of the order of 1 or smaller will satisfy that inequality.

When considering the boundary conditions

$$v_x = 0 \quad (y=0), \quad \eta \frac{\partial v_x}{\partial y} = \frac{\partial \alpha}{\partial x} \quad (y=h) , \tag{3}$$

together with the incompressibility condition written in the form

$$\int_0^h v_x dy = 0 , \tag{4}$$

where we have assumed that the total flow is zero, one arrives at[9]

$$\mathbf{v}_s = \frac{\nabla \alpha}{4\eta h}(3y^2 - 2hy) . \tag{5}$$

This stationary solution clearly indicates that the motion is due to the existence of a nonconstant surface tension. Moreover, that equation reveals the appearance of the characteristic frequency $\omega = |\nabla \alpha| / \eta$, which will be also present in our analysis of the correlation functions.

The velocity and temperature fluctuations evolve according to the stochastic differential equations dictated by fluctuating hydrodynamics[11]

$$\nabla \cdot \delta \mathbf{v} = 0 , \tag{6}$$

$$\frac{\partial \delta \mathbf{v}}{\partial t} + \mathbf{v}_s \cdot \nabla \delta \mathbf{v} + \delta \mathbf{v} \cdot \nabla \mathbf{v}_s = -\frac{1}{\rho} \nabla \delta p + \nu \nabla^2 \delta \mathbf{v} - \frac{1}{\rho} \nabla \cdot \underline{\mathbf{\Pi}}^R , \tag{7}$$

$$\frac{\partial \delta T}{\partial t} + \mathbf{v}_s \cdot \nabla \delta T + \delta \mathbf{v} \cdot \nabla T_s = \chi \nabla^2 \delta T + \frac{4\eta}{\rho c_v} [\nabla \mathbf{v}_s]:[\nabla \delta \mathbf{v}] - \frac{1}{\rho c_v} \nabla \cdot \mathbf{J}_q^R , \tag{8}$$

where we have taken into account that the fluid is incompressible[12] and linearized around the stationary hydrodynamic fields. Here $\delta \mathbf{v}$, δp, and δT are the velocity, pressure, and temperature fluctuations, χ the thermal diffusivity, and c_v the specific heat at constant volume. The square brackets denote the symmetric and traceless part of a tensor. The stochastic sources $\mathbf{\Pi}^R$ and $\mathbf{J}_q^R$ satisfy the fluctuation-dissipation theorems

$$\langle \Pi_{ij}^R(\mathbf{r},t) \Pi_{kl}^R(\mathbf{r}',t') \rangle = 2k_B T_s(\mathbf{r}) \eta_{ijkl} \delta(\mathbf{r}-\mathbf{r}') \delta(t-t') , \tag{9}$$

$$\langle J_{q,i}^R(\mathbf{r},t) J_{q,j}^R(\mathbf{r}',t') \rangle = 2k_B T_s^2(\mathbf{r}) \lambda \delta_{ij} \delta(\mathbf{r}-\mathbf{r}') \delta(t-t') , \tag{10}$$

where k_B is the Boltzmann constant, λ the thermal conductivity, and the fourth-rank tensor η_{ijkl} is equal to $\eta(\delta_{ij}\delta_{kl} + \delta_{il}\delta_{jk})$. The stochastic differential equations and the fluctuation-dissipation theorems introduced above where also used in Ref. 8 to compute correlation functions in the presence of external gradients.

III. CORRELATION FUNCTIONS

Our aim in this section is to compute the velocity and temperature correlation functions. To arrive at the velocity correlation function we will apply the operator $\nabla \times \nabla \times$ to Eq. (7) and Fourier transform that equation in the vector parallel to the surface $\mathbf{r}_\parallel$. We then obtain

$$\left[k_z^2 - \frac{\partial^2}{\partial y^2} \right] \left\{ \left[-i\omega + \nu \left[k_z^2 - \frac{\partial^2}{\partial y^2} \right] \right] \mathbf{1} + \frac{\partial v_s}{\partial y} \hat{\mathbf{e}}_x \hat{\mathbf{e}}_y \right\} \cdot \delta \mathbf{v} = -\frac{1}{\rho} \left[\left[ik_z \hat{\mathbf{e}}_z + \frac{\partial}{\partial y} \hat{\mathbf{e}}_y \right] \left[ik_z \hat{\mathbf{e}}_z + \frac{\partial}{\partial y} \hat{\mathbf{e}}_y \right] + \left[k_z^2 - \frac{\partial^2}{\partial y^2} \right] \mathbf{1} \right] \cdot \nabla \cdot \mathbf{\Pi}^R , \tag{11}$$

where $\mathbf{e}_i$, with $i = x,y,z$, are the unit vectors and $\mathbf{1}$ the unit matrix. For simplicity's sake we have considered wave vectors such that $k_x = 0$. Because our system is finite in the y direction we need to use Fourier series for the fluctuating fields. We will follow the steps outlined in Ref. 13. Thus we consider a mirror image of our system between $-h$ and 0 and split the velocity fluctuation and its corresponding random source as

$$\delta \dot{v}(k_z,y,\omega) = \frac{1}{2h} \sum_{n=-\infty}^{\infty} \delta \mathbf{v}_n(k_z,\omega) e^{in\pi y/h} , \tag{12}$$

$$\nabla \cdot \mathbf{\Pi}^R(k_z,y,\omega) = \frac{1}{2h} \sum_{n=-\infty}^{\infty} \mathbf{F}_n^R(k_z,\omega) e^{in\pi y/h} , \tag{13}$$

where $\delta \mathbf{v}_n$ and $\mathbf{F}_n^R$ are the Fourier components of the fluctuating fields corresponding to the nth mode.

If (12) and (13) are used in (11) one arrives at

$$\tilde{k}_n^2 \left[[G_n^v(\tilde{k}_n,\omega)]^{-1} \delta \mathbf{v}_n + \frac{i}{2\eta\pi} \frac{\partial \alpha}{\partial x} \sum_{\substack{k=-\infty \\ k \neq n}}^{\infty} \frac{1-2(-1)^{k-n}}{k-n} \delta \mathbf{v}_k \cdot \hat{\mathbf{e}}_y \hat{\mathbf{e}}_x \right] = -\frac{1}{\rho} \left[\left[ik_z \hat{\mathbf{e}}_z + \frac{in\pi}{h} \hat{\mathbf{e}}_y \right] \left[ik_z \hat{\mathbf{e}}_z + \frac{in\pi}{h} \hat{\mathbf{e}}_y \right] + \tilde{k}_n^2 \mathbf{1} \right] \cdot \mathbf{F}_n^R , \tag{14}$$

where we have defined the discrete propagator

$$G_n^v(\tilde{k}_n,\omega)\equiv\frac{1}{-i\omega+\nu\tilde{k}_n^2}\ , \tag{15}$$

with $\tilde{k}_n^2=k_\parallel^2+(n\pi/h)^2$. In the discretized version of the fluctuation-dissipation theorem (9) we must replace the wave vector along the y direction by $n\pi/h$ and use the Kronecker δ. We then obtain

$$\langle F_{nk}^R(k_z,\omega)F_{ml}^R(k_z',\omega')\rangle=2hD\eta\delta_{n,-m}\left[\tilde{k}_n^2\delta_{k,n}+\left[k_z\delta_{k,z}+\frac{n\pi}{h}\delta_{k,y}\right]\left[k_z\delta_{l,z}+\frac{n\pi}{h}\delta_{l,y}\right]\right]\ , \tag{16}$$

where use has been made of the definition

$$D\equiv 2k_BT_0(2\pi)^3\delta(\omega+\omega')\delta(k_z+k_z')\ .$$

Notice that the effects of the temperature gradient disappear in (16) as a consequence of the fact of considering wave vectors such that $k_x=0$. On the other hand the geometry of the problem, or in other words the absence of the convolution term in (7), enables one to compute the different components of the velocity correlation function matrix without any approximation. For the same reason only some components of that matrix will contain nonequilibrium corrections which are proportional to the surface tension gradient. Then Eqs. (14)–(16) lead to the velocity correlation function matrix

$$\begin{bmatrix}\langle\delta v_x\delta v_x\rangle^{(0)}+\langle\delta v_x\delta v_x\rangle^{(2)} & \langle\delta v_x\delta v_y\rangle^{(1)} & \langle\delta v_x\delta v_z\rangle^{(1)}\\ \langle\delta v_y\delta v_x\rangle^{(1)} & \langle\delta v_y\delta v_y\rangle^{(0)} & \langle\delta v_y\delta v_z\rangle^{(0)}\\ \langle\delta v_z\delta v_x\rangle^{(1)} & \langle\delta v_z\delta v_y\rangle^{(0)} & \langle\delta v_z\delta v_z\rangle^{(0)}\end{bmatrix}\ , \tag{17}$$

where a superindex specifies the order in $\nabla\alpha$. Then only the x-i or i-x components, with $i=x,y,z$, exhibit nonequilibrium corrections. Furthermore, the correction to the x-x component is quadratic, and therefore can be neglected in our approximation, whereas the remaining components vanish at equilibrium. To illustrate their behavior let us compute, as an example, the static correlation function $\langle\delta v_x(k_z,t)\delta v_y(k_z',t)\rangle$. The analysis of its discrete modes yields

$$\langle\delta v_{nx}(k_z,t)\delta v_{my}(k_z',t)\rangle=\begin{cases}0, & n=-m\\ -4\dfrac{h^2i}{\eta^2}2\pi k_BT_0\,|\,\nabla\alpha\,|\,\dfrac{2(-1)^{n+m}-1}{n+m}\dfrac{k_z^2}{\tilde{k}_m^2(\tilde{k}_n^2+\tilde{k}_m^2)}\delta(k_z+k_z'), & n\neq -m\ .\end{cases} \tag{18}$$

To study the nonequilibrium corrections originated from the surface tension gradient we will compare it with the equilibrium correlation function

$$\langle\delta v_{ny}(k_z,t)\delta v_{my}(k_z',t')\rangle\ .$$

Then we compute the quantities $B_{n,m}$ defined as

$$B_{n,m}\equiv\left|\frac{\langle\delta v_{nx}\delta v_{my}\rangle}{\langle\delta v_{ny}\delta v_{my}\rangle}\right|=\frac{\rho\nabla\alpha}{\pi\eta^2}\left|\frac{2(-1)^{n+m}-1}{n+m}\right|\frac{1}{\tilde{k}_n^2+\tilde{k}_m^2}\ , \tag{19}$$

which are essentially proportional to the ratio between the characteristic frequency ω_c, defined above, and the frequencies related to the viscous modes $\nu\tilde{k}_n^2$. One arrives at

$$B_{n,m}=\frac{4}{\pi}\mathcal{R}\left|\frac{2(-1)^{n+m}-1}{n+m}\right|\frac{1}{h^2(\tilde{k}_n^2+\tilde{k}_m^2)}\le\frac{12}{\pi}\mathcal{R}[h^2(\tilde{k}_1^2+\tilde{k}_0^2)]^{-1} \tag{20}$$

from which we conclude that the most important corrections will occur for the lowest modes and at small viscosities, provided that the condition (2b) is fulfilled. To evaluate the corrections let us assume a fluid layer under a temperature gradient of 0.1 °C/cm. If the fluid is water at 20 °C and $h=0.3$ cm, $\omega_c=1.5\ \mathrm{s}^{-1}$ and $\mathcal{R}=3.4$, then if $k_z=100\ \mathrm{cm}^{-1}$ the correction is about 0.72%. That correction increases for mercury at 50 °C (6.2%) and is very important when the viscosity decreases dramatically as occurs for liquid CO_2 at 20 °C (80%).

The temperature correlation function can be also computed by means of the procedure outlined above. In this case we need to use the Fourier series

$$\delta T(\mathbf{k}_\parallel,y,\omega)=\frac{1}{2h}\sum_{n=-\infty}^{\infty}\delta T_n(\mathbf{k}_\parallel,\omega)e^{in\pi y/h}\ , \tag{21}$$

$$\nabla\cdot\mathbf{J}_q^R(\mathbf{k}_\parallel,y,\omega)=\frac{1}{2h}\sum_{n=-\infty}^{\infty}Q_n^R(\mathbf{k}_\parallel,\omega)e^{in\pi y/h}\ . \tag{22}$$

From inspection of Eq. (8) we conclude that, due to the fact that the stochastic sources are not correlated, the terms containing the stationary velocity introduce corrections of first order in $\nabla\alpha$, whereas those proportional to the velocity fluctuations give rise to corrections of second order in the expansion parameter that will be neglected. As a consequence, in order to obtain corrections to the temperature correlation function we must consider in this case wave vectors for which $k_x\neq 0$. Our final expression for the temperature fluctuation corresponding to the nth mode is

$$\delta T_n(k_z,\omega) = -\frac{1}{\rho c_v} G_n^T(\tilde{k}_n,\omega) Q_n^R + \frac{ik_x h \mid \nabla\alpha \mid}{2\pi^2 \eta \rho c_v} G_n^T(\tilde{k}_n,\omega) \times \sum_{\substack{k=-\infty \\ k\neq n}}^{\infty} \frac{2(-1)^{k+n}+1}{(k-n)^2} G_k^T(\tilde{k}_k,\omega) \times Q_k^R(\tilde{k}_k,\omega) + \cdots , \tag{23}$$

where now the propagator is defined as $G_n^T(k_n,\omega) = -i\omega + \kappa \tilde{k}_n^2$, this last expression we have indicated by elipses those terms that give corrections to the equilibrium temperature correlation function of second order or higher in $\nabla\alpha$ [or ∇T due to the relation $\nabla\alpha = (\partial\alpha/\partial T)\nabla T$].

As before, the fluctuation-dissipation theorem for the discrete modes reads

$$\langle Q_n^R(\mathbf{k}_\parallel,\omega) Q_m^R(\mathbf{k}'_\parallel,\omega') \rangle = 2h\lambda E(\mathbf{q}) \delta_{n,-m} \left[\left[\frac{n\pi}{h} \right]^2 - \mathbf{k}_\parallel \cdot \mathbf{k}'_\parallel \right] \delta(\omega+\omega') , \tag{24}$$

with the definition

$$E(\mathbf{q}) = 2k_B (2\pi)^3 \{ T_0^2 \delta(\mathbf{k}_\parallel + \mathbf{k}'_\parallel) - iT_0 \delta\tilde{T} [\delta(\mathbf{k}_\parallel + \mathbf{k}'_\parallel + \mathbf{q}) - \delta(\mathbf{k}_\parallel + \mathbf{k}'_\parallel - \mathbf{q})] \} , \tag{25}$$

in which we have used the form of the temperature gradient $\nabla T = i\delta T \mathbf{q}$, $\mathbf{q}$ being a vector in the direction of the gradient.[7]

The temperature correlation function in the $\mathbf{k}$-t representation then follows from Eqs. (23)–(25). The equilibrium contribution and the first correction in the surface tension gradient are given, respectively, by

$$\langle \delta T_n(\mathbf{k}_\parallel,t) \delta T_m(\mathbf{k}'_\parallel,t') \rangle^{(0)} = \frac{(2\pi)^2 2hk_B T_0^2}{\rho c_v} e^{-\lambda(t-t')\tilde{k}_n^2} \delta(\mathbf{k}_\parallel + \mathbf{k}'_\parallel) \delta_{n-m} , \tag{26}$$

$$\langle \delta T_n(\mathbf{k}_\parallel,t) \delta T_m(\mathbf{k}'_\parallel,t') \rangle^{(1)} = \begin{cases} \dfrac{4h^2 k_B T_0^2}{\eta\lambda} \mid \nabla\alpha \mid \dfrac{ik_x}{\tilde{k}_n^2 - \tilde{k}_m^2} \dfrac{2(-1)^{n+m}+1}{(n+m)^2} (e^{-\chi(t-t')\tilde{k}_n^2} - e^{-\chi(t-t')\tilde{k}_m^2}) \\ \quad \times \delta(\mathbf{k}_\parallel + \mathbf{k}'_\parallel) \quad (n \neq -m) \\ 0 \quad (n = -m) . \end{cases} \tag{27}$$

Notice that in this case the nonequilibrium correction vanishes for $t = t'$. The fact that the temperature correlation function is now a development in powers of $\mid \nabla\alpha \mid$ introduces a restriction in our theory which follows from the calculation of the quantities

$$C_{n,m} \equiv \left| \frac{\langle \delta T_n(\mathbf{k}_\parallel,t) \delta T_m(\mathbf{k}'_\parallel,t') \rangle^{(1)}}{\langle \delta T_n(\mathbf{k}_\parallel,t) \delta T_m(\mathbf{k}'_\parallel,t') \rangle^{(0)}} \right| . \tag{28}$$

Using (26) and (27) one arrives at

$$C_{n,m} = \frac{hk_x \rho c_v}{2\pi^2\lambda} \frac{\mid \nabla\alpha \mid}{\eta} \frac{\mid 2(-1)^{n+m}+1 \mid}{(n+m)^2} \times \left| \frac{1 - e^{\chi(t-t')(\tilde{k}_n^2 - \tilde{k}_m^2)}}{\tilde{k}_n^2 - \tilde{k}_m^2} \right| \ll 1 , \tag{29}$$

which shows that the quantities $C_{n,m}$ are proportional not only to the ratio between ω_c and the frequencies corresponding to the viscous modes but to the quantity hk_x. As before, the corrections increase for the lowest modes. Then to satisfy the inequality in (29) it is sufficient to impose

$$C_{1,0} \simeq \frac{hk_x}{2\pi^2} \omega_c (t-t') = \tau^{-1}(t-t') \ll 1 , \tag{30}$$

from which we conclude that the relaxation time τ depends on the wave vector and constitutes an upper bound for times in our theory. This quantity can be evaluated for some liquids. For a layer of water of $h = 0.1$ cm at 20 °C, under a temperature gradient of 0.1 °C/cm, the characteristic frequency is $\omega_c = 1.5\ \mathrm{s}^{-1}$, then for wave vectors of the order 1000 cm^{-1} we have $\tau = 0.07$ s. Under the same conditions and for glycerin the value of τ is about 140 s.

IV. DISCUSSION

To clarify and elaborate on some of the points raised in this paper the following comments may be useful. We have shown how to extend the formalism of fluctuating hydrodynamics outlined in Refs. 7 and 8 to compute correlation functions for finite systems. The stationary state is a consequence of the presence of a temperature gradient parallel to the surface of the fluid. For real cases the surface tension depends on temperature, therefore the existence of tangential stresses will induce motion of the fluid layer. Our case provides an example of a system away from equilibrium in which the physics depends crucially on the existence of a finite dimension in the problem. The domain of applicability of the present theory, $\mathcal{R}(h/L) \ll 1$, illustrates our contention. In fact, the larger the aspect ratio, the smaller the Reynolds number, therefore the thermocapillary motion fades away when increasing h/L.

It is interesting to realize that the nonconstant surface tension introduces a natural frequency $\omega_c = \mid \nabla\alpha \mid / \eta$ in our problem. In fact the capillary and viscous effects can be contrasted through the relation

$$|\mathbf{F}_\alpha|/|\mathbf{F}_\eta| = \left|\frac{\nabla\alpha/\eta}{\nabla\mathbf{v}}\right| , \tag{31}$$

where $\mathbf{F}_\alpha$ and $\mathbf{F}_\eta$ are the forces related to surface tension and viscous effects. Such a frequency constitutes the expansion parameter in which the correlation functions should be developed and plays a role similar to that of the velocity gradient in fluids under external gradients.[8]

As a consequence of the chosen wave vectors our expression of the velocity correlation function is exact as follows from the fact that the component k_x enters the convolution term in Eq. (7). Our result (17) illustrates the contention that some components of the correlation function matrix are different from zero due to the surface tension gradients. The nonequilibrium corrections given through the quantities $B_{n,m}$ are essentially proportional to $\mathcal{R}(hk_\parallel)^{-2}$.

To study the temperature correlation function we need to keep the terms proportional to k_x since they provide the nonequilibrium corrections up to first order in $\nabla\alpha$. A convolution term is then present and therefore an expansion in $\nabla\alpha$ is involved. As a consequence, we must introduce a cutoff frequency τ^{-1} and a cutoff wave vector $(\hbar\omega_c t)^{-1}$. Our theory is then valid for $\omega \gg \tau^{-1}$ or $k_x \ll (h\omega_c t)^{-1}$. Notice that in this case we have found an upper cutoff wave vector instead of the ones usually encountered when dealing with fluids under external gradients[7,8].

It is also interesting to realize from (27) that the nonequilibrium part corresponding to the static temperature correlation function vanishes and therefore no long-range correlations are present at equal time. This result should be compared with that for a fluid under a temperature gradient for which we have correlations of the form $1/|\mathbf{r}-\mathbf{r}'|^8$. On the other hand, the leading terms of some components of the velocity correlation function matrix given in (17) are nonequilibrium contributions proportional to $\nabla\alpha$. This result is a consequence of our stationary temperature and velocity profiles given through (1) and (5).

Finally, we will point out that the nonequilibrium corrections to the correlation functions we compute are the result of a nonconstant surface tension. Our situation is then different to what happens when considering light scattering from a liquid surface subject to a temperature gradient, in which surface tension is assumed to be constant.[14]

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