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# Viscous–dissipative correlations in fluctuating hydrodynamics

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**Abstract.** The time and spatial correlations of the equilibrium fluctuations in isotropic incompressible fluids are studied. The reasoning is based on the example of Brownian motion using the memory function formalism and the hydrodynamic expression of the Langevin force as an integral of the stress tensor fluctuations over the solid particle surface. Relations thus obtained allow the determination of some correlation characteristics of the stress tensor fluctuations and the Langevin force, which depend on both dissipative and thermodynamic parameters. These results are in a good agreement with the known experimental data from ellipsometric studies of spatial correlations of thermal fluctuation capillary waves on gas–simple liquid interfaces.

Although the collective fluctuation modes in fluids are well known [1–4], there is no complete description of their time and spatial distributions. The present paper does not treat this problem directly but, on the assumption of such distributions, their effects on a freely moving macroparticle are studied and some of their mean characteristics are obtained. The concept of this analysis is that the Brownian particle does not disturb the medium fluctuations but just captures their characteristics.

The main goal of this paper is to describe in more detail the hydrodynamic fluctuations compared with the classical theory, based on the linearized stochastic Landau–Lifshitz equation governing the motion of an incompressible fluctuating fluid [5–7]:

$$\rho(\partial/\partial t)V(r,t) = -\nabla \cdot \mathbf{P}(r,t)$$

$$\nabla \cdot V(r,t) = 0$$

$$\mathbf{P}(r,t) = p(r,t)\mathbf{I} - \rho\nu[(\nabla V) + (\nabla V)^{\dagger}] + \mathbf{S}(r,t).$$

Here $V$ is the fluctuating hydrodynamic velocity, $\mathbf{P}$ is the stress tensor, $\rho$ and $\nu$ are the mass density and the kinematic viscosity, respectively, of the fluid, $p$ is the fluctuating pressure, $\mathbf{I}$ is the unit tensor and $\mathbf{S}$ is the random stress tensor, with the following stochastic properties:

$$\langle \mathbf{S}(r,t) \rangle_{\mathrm{G}} = 0$$

and

$$\langle \mathbf{S}(r,t)\mathbf{S}(r+R,t+\tau) \rangle_{\mathrm{G}} = 2k_{\mathrm{B}}T\rho\nu\mathbf{L}\delta(\tau)\delta(R)$$

with

$$L_{ij}^{kl} \equiv \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} - \tfrac{2}{3}\delta_{ij}\delta_{kl}.$$

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In these equations the angular brackets $\langle\cdot\rangle_G$ denote averages over an equilibrium canonical Gibbs ensemble with temperature $T$, and $k_B$ is the Boltzmann constant.

A known shortcoming of the classical model is that infinite local dispersions of the fluctuating hydrodynamic quantities such as $\langle VV\rangle_G$, $\langle \mathbf{PP}\rangle_G$ and $\langle \mathbf{SS}\rangle_G$ are obtained which is valid for both compressible and incompressible fluids [1, 3]. This occurs for two reasons. The first is the simplified modelling of the dissipation in the classical hydrodynamic model, which affects the time dependence of the hydrodynamic fluctuation correlations [1, 2]. The second reason is the use of the classical fluctuation–dissipation theorem [2, 3, 8] which is strongly related to the equilibrium canonical Gibbs distribution, to obtain the statistical properties of the random stress tensor. The canonical distribution is not applicable to fluctuations in small systems since the terms accounting for the system–thermostat interactions are neglected [3, 4]. For the correct description of such fluctuations (particularly the local hydrodynamic fluctuations) it is necessary to extend the classical fluctuation theory [1–4] by taking into account the significant influence of the dissipative interactions on these objects [9].

A convenient way to study the hydrodynamic fluctuations is their application to Brownian motion. It has been theoretically shown by Langevin [10] that the interaction of the medium with a macroparticle can be represented by two forces: a frictional force, linked with the concept of the fluid hydrodynamic viscous resistance [6], and a driving force, most commonly called a Langevin force which reflects the stochastic nature of the collisions. The momentum balance of the motion according to this description has the form of the generalized Langevin equation [1, 8, 11]

$$\begin{aligned} &M(\mathrm{d}/\mathrm{d}t)\boldsymbol{U}(t) + \hat{\mathbf{G}}(t)\boldsymbol{U} = \boldsymbol{F}(t) \\ &\langle \boldsymbol{F}(t)\rangle = \mathbf{0} \end{aligned} \tag{1}$$

where $M$ and $\boldsymbol{U}$ are the mass and velocity, respectively, of the Brownian particle, $\hat{\mathbf{G}}\boldsymbol{U}$ is the frictional force and $\boldsymbol{F}$ is the Langevin force. In the present paper the angular brackets $\langle\cdot\rangle$ denote averages over the real equilibrium distribution, which could be different from the Gibbs canonical distribution for the reasons mentioned above.

Because of the common molecular–kinetic origin of the two *ad hoc* introduced forces $\hat{\mathbf{G}}\boldsymbol{U}$ and $\boldsymbol{F}$, they are not independent. Their link, called by Kubo [8] the second fluctuation–dissipation theorem, was formulated most generally by Mori [11] and Zwanzig [12]:

$$\hat{\mathbf{G}}(t)\boldsymbol{U} = (k_B T)^{-1}\int_0^t \mathbf{C}_{\boldsymbol{FF}}(t-t_1)\cdot\boldsymbol{U}(t_1)\,\mathrm{d}t_1 \tag{2}$$

where

$$\mathbf{C}_{\boldsymbol{FF}}(\tau) = \langle \boldsymbol{F}(t)\boldsymbol{F}(t+\tau)\rangle$$

is the Langevin force autocorrelation function (LFACF) and the temperature is introduced via the relation

$$M\langle \boldsymbol{UU}\rangle = k_B T\mathbf{I}.$$

The operator $\hat{\mathbf{G}}$ is proportional to the friction coefficient $\beta$. The relationship between $\hat{\mathbf{G}}$ and $\beta$ follows from the long-term limit of (2) and is given by the well known relation [1, 2, 8]

$$(k_B T)^{-1}\int_0^\infty \mathbf{C}_{\boldsymbol{FF}}(\tau)\,\mathrm{d}\tau = \beta\mathbf{I}. \tag{3}$$

When the frictional force $\hat{\mathbf{G}}U$ is approximated by $\beta U$, the LFACF tends to $2k_B T\beta\mathbf{I}\delta(\tau)$ (see (2)).

In the traditional statistical mechanical treatments the $\hat{\mathbf{G}}U \to \beta U$ transition is analysed on the microscopic level as a function of the ratio of the mass $M$ of a Brownian particle to the mass $m$ of the particles in the environment. It is shown in the mass point approximation [1, 2] that $\hat{\mathbf{G}}(m/M \to 0)U \to \beta U$. For objects of finite, sufficiently large size, with a macroscopically averaged interaction with the surrounding medium, the order of magnitude of the mass $m$ cannot be that of the mass of the fluid microparticles [13]. The magnitude of $m$ should be determined by the order of the mass of the fluid displaced by the particle, $m \sim \rho\pi R_p^3$ with $R_p$ being the radius of the Brownian particle. This can be demonstrated on the basis of the classical result for the Laplace transform (hereafter denoted by a tilde (˜) of the LFACF [13–16]:

$$\tilde{\mathbf{C}}_{FF}(s) = k_B T 6\pi\rho\nu R_p \mathbf{I}\left[1 + \sqrt{sR_p^2/\nu} + \tfrac{1}{9}sR_p^2/\nu\right] \tag{4}$$

which is obtained by means of the Landau–Lifshitz model [14–16]. Here the friction coefficient $\beta$ is equal to $6\pi\rho\nu R_p$. Equation (4) shows again the inconsistency of the model discussed above, resulting in the divergence of the dispersion of the random force and a wrong result for the dispersion of the Brownian particle velocity:

$$\langle U \cdot U\rangle = 3k_B T/(M + \tfrac{2}{3}\rho\pi R_p^3).$$

This is easy to explain since these results present an incorrect extension of a formula valid for small $s$ to derive quantities related mainly to the highest $s$-value.

From the hydrodynamic point of view [15, 16], the force $F$ in (1) is the sum of the normal projections of the fluctuating part of the stress tensor $\Delta\mathbf{P} = \mathbf{P} - \langle\mathbf{P}\rangle_U$ upon the Brownian particle:

$$\begin{aligned} F(t) &= \int_{S_p} \Delta\mathbf{P}(R(t) + R_p n, t)\cdot n\,\mathrm{d}S \\ R(t) &= \int U(t)\,\mathrm{d}t \end{aligned} \tag{5}$$

where the integration is carried out along the particle surface $S_p$. The stress tensor $\langle\mathbf{P}\rangle_U$ comprises the equilibrium stress tensor of the fluid $\langle\mathbf{P}\rangle$ and the perturbation caused by the Brownian particle and determines the drag force $\hat{\mathbf{G}}U$. It represents the mean value of the stress tensor $\mathbf{P}$ averaged along the coordinates and momenta of the fluid particles without averaging along the coordinates and the momentum of the Brownian particle and, owing to the random nature of the Brownian motion, it is also a random quantity with a mean value $\langle\mathbf{P}\rangle$.

According to (5), the mean value of the Langevin force $\langle F\rangle$ is equal to zero. Its autocorrelation function $\mathbf{C}_{FF}$ is the first characteristic which is sensitive to the scale of stress tensor fluctuations $\Delta\mathbf{P}$ and, using (5), it takes the form

$$\mathbf{C}_{FF}(\tau) = \int_{S_p}\int_{S_p} n\cdot\langle\Delta\mathbf{P}(R_p n, t)\Delta\mathbf{P}(R_p n_1, t+\tau)\rangle\cdot n_1\,\mathrm{d}S\,\mathrm{d}S_1. \tag{6}$$

To arrive at (6), one uses the fact that in the fully linearized scheme the time dependence of $R(t)$ is negligible and that the origin of the coordinate may be assumed to be in the centre of the Brownian particle [16].

In equilibrium homogeneous systems the fluctuations are described as stationary and uniform processes [1–4], which justifies expressing the stress tensor autocorrelation as

$$\mathbf{C_{PP}}(\boldsymbol{R},\tau)=\langle\Delta\mathbf{P}(\boldsymbol{r},t)\Delta\mathbf{P}(\boldsymbol{r}+\boldsymbol{R},t+\tau)\rangle=\langle\Delta\mathbf{P}\Delta\mathbf{P}\rangle C(R/l_c,\tau/\tau_c) \tag{7}$$

where $\tau_c$ is the correlation time and $l_c$ is the correlation length. The direct substitution of $\mathbf{C_{PP}}$ from (7) into (6) elucidates the link between the LFACF and the stress tensor fluctuations $\Delta\mathbf{P}$:

$$\mathbf{C}_{FF}(\tau)=\pi l_c^2 S_p\langle\Delta\mathbf{P}\cdot\Delta\mathbf{P}\rangle f(\tau/\tau_c;R_p/l_c). \tag{8}$$

In order to determine the dispersion $\langle\Delta\mathbf{P}\cdot\Delta\mathbf{P}\rangle$, reflecting the order of the magnitude of the LFACF, the equilibrium fluctuation amplitudes of the stress tensor fluctuations $(\Delta\mathbf{P})_V$ in a macroscopic volume $V\gg\pi l_c^3$ could be used. At constant $\rho$ and $T$ the only source of the fluid fluctuations is the non-uniform distribution of the fluid particle velocities and for this reason the variance $\langle(\Delta\mathbf{P})_V\cdot(\Delta\mathbf{P})_V\rangle$ of equilibrium stress tensor fluctuations can be expressed as follows [3, 17]:

$$\langle(\Delta\mathbf{P})_V\cdot(\Delta\mathbf{P})_V\rangle=\frac{1}{V^2}\int_V\int_V\langle\Delta\mathbf{P}(\boldsymbol{r},t)\cdot\Delta\mathbf{P}(\boldsymbol{r}_1,t)\rangle\,\mathrm{d}\boldsymbol{r}\,\mathrm{d}\boldsymbol{r}_1=\frac{k_BT\rho c^2}{V}\mathbf{I}.$$

The equilibrium canonical Gibbs distribution is used to obtain this last equation since $V\gg\pi l_c^3$, and $c$ is the Maxwell velocity of the fluid particles. When the autocorrelation function $\mathbf{C_{PP}}$ of the stress tensor fluctuations given by (7) is replaced in this result, the following expression for the variance of the stress tensor fluctuations is obtained: $\langle\Delta\mathbf{P}\cdot\Delta\mathbf{P}\rangle\sim k_BT\rho c^2/(\pi l_c^3)\mathbf{I}$. By substituting it in (8) the LFACF takes the form

$$\tilde{\mathbf{C}}_{FF}(s)\sim k_BT\rho c^2 S_p\tau_c/l_c\mathbf{I}\tilde{f}(s\tau_c;R_p/l_c). \tag{9}$$

The dependence of $\tau_c$ and $l_c$ on the parameters of the medium can be obtained from $\sqrt{s\tau_c}$ by a power expansion of (9):

$$\tilde{\mathbf{C}}_{FF}(s)\sim k_BT\rho c^2 S_p\tau_c/l_c\mathbf{I}[\tilde{f}_0+\tilde{f}_1\sqrt{s\tau_c}+\tilde{f}_2 s\tau_c+\ldots]$$

where it is supposed that

$$\tilde{f}_n(R_p/l_c)\equiv(1/n!)[\partial^n\tilde{f}(u^2;R_p/l_c)/\partial u^n]_{u=0}$$

are finite. The LFACF should contain as a limit the classical result (4) at small $s\tau_c$, when classical hydrodynamics are valid. Comparison of these equations leads to the following expressions for the correlation time $\tau_c$ and correlation length $l_c$:

$$\tau_c\sim(\nu/c^2)[(l_c/R_p)/\tilde{f}_0]\sim\nu/c^2 \tag{10a}$$

$$l_c\sim\sqrt{c^4\tau_c^3/\nu}[\tilde{f}_1]\sim(c\tau_c)^2\left[\sqrt{(l_c/R_p)\tilde{f}_2}\right]\sim\nu/c. \tag{10b}$$

The expressions in square brackets in (10) must be constants because $\tau_c$ and $l_c$ are characteristics of the medium and should not depend on $R_p$. Note that $f(\tau/\tau_c;R_p/l_c)$ depends on $R_p$ since it represents an integral of the stress tensor autocorrelation over the

Brownian particle surface. According to (10), $\tau_c$ and $l_c$ are functions of the dissipative and non-dissipative parameters and are proportional to the mean free time and path [9].

One could obtain the same results [9] if $l_c$ is considered as the distance at which the perturbation $\Delta\mathbf{P}$ would spread for the time $\tau_c$ ($l_c \sim c\tau_c$) and if the time of persistence of the perturbation $\tau_c$ is interpreted as a characteristic which reflects the loss of correlation as a result of viscous friction ($l_c^2 \sim \nu\tau_c$). It is notable that these results are in good agreement with the experimental data from the known ellipsometric investigations of spatial correlations of fluctuation capillary waves on simple gas–liquid interfaces [9].

New results also derived are the finite values of the variance of the stress tensor fluctuations $\langle \Delta\mathbf{P} \cdot \Delta\mathbf{P} \rangle \sim k_B T \rho c^5/(\pi\nu^3)\mathbf{I}$ and of the Langevin force $\langle \boldsymbol{F}\boldsymbol{F} \rangle \sim k_B T \rho S_p (c^3/\nu) f(0; R_p c/\nu)\mathbf{I}$ which depend on the kinematic viscosity of the fluid. In the case when $R_p \gg l_c$ it follows from (8) that $f(0; R_p c/\nu) \sim 1$.

According to the present work the dynamics of the stress tensor fluctuations in the bulk at constant $\rho$ and $T$ are analogous to a pure deformation field with a correlation length $l_c \sim \nu/c$ and a correlation time $\tau_c \sim \nu/c^2$. The classical description of the hydrodynamic fluctuations follows from the present theory in the limit of infinite thermal velocity of the fluid particles $c$ at finite kinematic viscosity $\nu$. As noted in the introduction, the Landau–Lifshitz model is directly related to the canonical Gibbs distribution. The limit $c \to \infty$ at finite $\nu$ is equivalent to neglecting the dissipative interactions between the system and the thermostat with respect to the system's kinetic energy, and the equilibrium distribution of the system can be approximated by a canonical distribution function [3, 4].

The physical nature of the viscous–dissipative correlation as a self-organisation structure could be elucidated by means of the laws of the local turbulence [3, 6]. In 1941, Oboukhov and Kolmogorov established a characteristic length $l$ of the small-scale turbulence, below which the motion is laminar. The size of these turbulent subdomains obeys the relation $l^4 \sim \nu^3/w$, where $w$ is the local power (per unit mass) of the dissipation of energy as viscous friction. Because of stationarity, the power $w$ has to be equal to that of the external sources. In the description of fluctuations the thermal molecular motion plays the role of 'external' source and $w$ could be estimated as $w \sim \epsilon/\tau$, $\epsilon$ and $\tau$ being the specific energy of the thermal motion and the excitation time of the fluctuations, respectively. Because of stationarity, the characteristic time $\tau$ of the source has to be equal to the mean relaxation time $l^2/\nu$ of the energy dissipation. From this point of view the Oboukhov–Kolmogorov law has the alternative expression $\epsilon \sim (\nu/l)^2$. Since the specific energy of the thermal motion of molecules is proportional to their mean square velocity, i.e. $\epsilon \sim c^2$, the size and lifetime of the fluctuations are estimated from this law as $l \sim \nu/c$ and $\tau \sim \nu/c^2$, i.e. they are identical with the correlation length $l_c$ and time $\tau_c$ obtained above in (10).

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