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A note on the fluctuation–dissipation relation for the generalized Langevin equation with hydrodynamic backflow

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ABSTRACT

This paper is devoted to finding the fluctuation–dissipation relation (FDR) for the generalized Langevin equation (GLE) with the Boussinesq–Basset (BB) force in which the Stokes friction is generalized to a convolution of a memory kernel with the velocity of a Brownian particle. First, the solution of such GLE with hydrodynamic backflow is obtained. Using this solution, we find in a simple and easily controllable way the time correlation function of the thermal force driving the particles. If the GLE is used with the original BB force for pure liquids, the FDR known from the literature is corrected. It is shown that in this case the FDR contains, in addition to the known term $\sim t^{-3/2}$, a more slowly decaying contribution $\sim t^{-1/2}$.

1. Introduction

In the original Langevin equation (LE) [1], designed to describe the Brownian motion (BM) of particles due to collisions with surrounding molecules within a thermal bath, the force acting on the particle splits into the macroscopic Stokes friction force and a microscopic random force. The latter force is delta-correlated in time and thus called white noise. The assumption of instantaneous interaction with the environment is an idealization that significantly restricts the applicability of the LE on real physical systems [2]. Whereas the LE very well describes the experiments with Brownian particles in gases [3], it fails to describe the chaotic motion of freely buoyant particles in fluids, except long times when the LE possesses the same results as the classical Einstein theory [4]. The limitations of the LE have been established long ago in the work [5], where a hydrodynamic theory of the BM has been created (for a review of this and related works on the translational and rotational BM see [6]). In the hydrodynamic theory, the Stokes friction is replaced by the Boussinesq (or Boussinesq–Basset) force [7,8] that appears naturally as a solution to the linearized Navier–Stokes equations for incompressible fluids [9]. The resulting integro-differential equation contains a convolution of the particle acceleration with the hydrodynamic memory kernel decaying in time as $\sim t^{-1/2}$ and probably represents the first generalization of the LE taking into account the histories in the stochastic dynamics. Nowadays, the generalized LE (GLE) is widely used in different areas of science from physics, chemistry and biology to financial markets (for a number of examples see, e.g., the recent papers [10–14]) to implement memory effects in the behavior of stochastic systems. Owing to the wide use of the GLE one could take for granted that this equation is fully investigated and formal problems concerning its properties and solution have been already solved. Although basically this thought is correct, there still remains at least one problem that is incorrectly treated in the literature. One of the goals of the present note is to show that the so-called second fluctuation–dissipation theorem (FDT) (below called the fluctuation–dissipation relation, FDR) for the GLE describing the hydrodynamic BM with the Boussinesq force, as it was reported, e.g., in [15–18], and as it appears also in the recent papers [3,19], should be corrected.

The concepts of the GLE and the corresponding to it FDR were well established in the middle of the 20th century [20,21]. The GLE (the equation of motion for Brownian particles) considered in these works contains as a generalization of the Stokes friction force the convolution

$$F_S(t) = -\int_0^t dt' \gamma(t-t') \upsilon(t') \tag{1}$$

of a memory kernel $\gamma(t)$ with the velocity of the particle, $\upsilon(t) = \dot{x}(t)$. Then the FDR connects the correlation function of the ran-

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dom force $\xi(t)$ driving the Brownian particle with $\gamma(t)$ as follows: $\sigma(t) = \langle\xi(0)\xi(t)\rangle = k_B T\gamma(t)$, $t > 0$. As distinct from this relation, the equation for motion with the Boussinesq force is more difficult to analyze. If the memory kernel in this equation is $\zeta_B(t)$, the corresponding FDR in the time domain at $t > 0$ has been obtained as $\sigma(t) = k_B T\dot{\zeta}_B(t)$ [15–18], which, since $\zeta_B \sim t^{-1/2}$, behaves as $t^{-3/2}$. In [22–24] it has been shown by different methods that this result misses an important contribution $\sim t^{-1/2}$. In this paper a more general problem is being considered when the equation of motion for the Brownian particle contains a generalized Boussinesq force, in which the original Stokes friction force is written in a more general form of the force $F_S(t)$. Such a model was studied in detail in the works [25,26] with $\zeta_B(t)$ as it appears in the Boussinesq force. The corresponding FDR has been found in [19] on the basis of the FDT for a more general case, when the kernel $\zeta_B(t)$ may not be specified. Below this result is obtained in a much more simple and controllable way. Moreover, for the special case of the hydrodynamic Brownian motion in pure liquids we correct the FDR derived in [25,26,19] and a number of other papers.

2. Generalized Langevin equation with hydrodynamic interactions

The linear model [25,26] under consideration is aimed to describe the motion of a tracer with mass m in a complex viscoelastic medium. With no loss of generality, it restricts to the one-dimensional case. The equation of motion for the tracer reads

$$m\dot{\upsilon} = F_S + F_B + F_{ext} + \xi, \tag{2}$$

where $F_S(t)$ is given by Eq. (1), $F_{ext}(t)$ is an external force, e.g., the elastic one, $\xi(t)$ is a colored noise accounting for the interaction of the tracer with the heath bath, and the force

$$F_B(t) = -\frac{m_f}{2}\dot{\upsilon}(t) - \int_0^t dt'\zeta_B(t-t')\dot{\upsilon}(t'), \tag{3}$$

comes from the Boussinesq force, the full expression for which is $-\gamma\upsilon + F_B(t)$. Here, m_f is the mass of the fluid displaced by the tracer and γ is the Stokes friction coefficient ($\gamma = 6\pi\eta a$ for a particle of radius a in a fluid of viscosity η). Note that although most often the initial moment for the forces $F_S(t)$ and $F_B(t)$ is set to zero, to reflect the fact that the forces are determined by the particle velocity and acceleration in all the moments of time preceding t, we will use the integrals from t_0 to t, with t_0 infinitely remote from t. This choice, however, does not affect the results presented below. It will be shown that obtaining the correct solution of Eq. (2) requires that the correlation function $Z(t) = \langle\upsilon(0)\xi(t)\rangle$ is nonzero at $t > 0$. In the case of the hydrodynamic BM the correct velocity autocorrelation function (VAF) for the Brownian particle is obtained only if $\tau\langle\upsilon(0)\xi(t)\rangle = -k_B T(\tau_f/\pi t)^{1/2}$, where $\tau = m^*/\gamma$, $\tau_f = a^2\rho_f/\eta$, ρ_f is the fluid density and $m^* = m + m_f/2$ [22–24]. A similar expression can be obtained in the considered more general model by Grebenkov and co-workers [25,26,19].

It should be noted that the model proposed in [25] has so far no microscopic substantiation. In spite of this, it seems to be a promising step to incorporate a description of viscoelastic properties of a medium (through the generalized Stokes force $F_S(t)$) in the successful description of hydrodynamic interactions of a spherical tracer with the surrounding fluid (through the Boussinesq force), the latter being confirmed in a number of experiments. Generalizing the arguments put forward in [27] for Newtonian fluids, it has been proposed in [19] that the kernels $\gamma(\tau)$ and $\zeta_B(\tau)$ are in visco-elastic media functionally related to one another: e.g., their Laplace transforms are connected as follows:

$$\tilde{\gamma}(s) = \frac{2s}{9m_f}\tilde{\zeta}_B^2(s). \tag{4}$$

3. Solution of the model

The basic equation of the model (2) that includes also the external force $F_{ext} = -kx$,

$$m^*\dot{\upsilon}(t) + \int_{t_0}^t dt'\gamma(t-t')\upsilon(t') + \int_{t_0}^t dt'\zeta_B(t-t')\dot{\upsilon}(t') + kx(t) = \xi(t), \tag{5}$$

is most easily solved by transforming it to the equation [28,29]

$$m^*\dot{V}(t) + \int_0^t dt'\gamma(t-t')V(t') + \int_0^t dt'\zeta_B(t-t')\dot{V}(t') + k\int_0^t dt'V(t') = 2k_B T, \tag{6}$$

with the initial condition $V(0) = 0$. Here, $V(t) = \dot{X}(t) = 2D(t)$, where $X(t)$ is the mean square displacement (MSD) of the tracer and $D(t)$ is its time-dependent diffusion coefficient. The transformation from (5) to (6), equivalent to the FDT [30], was used for the first time in building the hydrodynamic theory of the BM in incompressible fluids [5] (for some historical remarks and a review on this pioneering work and related papers see [6]; a generalization to compressible fluids is given in another remarkable work [30], also exploring the FDT). The velocity autocorrelation function (VAF) is related to $V(t)$ by the formula $C_\upsilon(t) = \langle\upsilon(t)\upsilon(0)\rangle = \dot{V}(t)/2$. It is seen from (6) that $\dot{V}(0) = 2k_B T/m^*$, so that $C_\upsilon(0) = k_B T/m^*$. This result from the hydrodynamic BM [5], confirmed experimentally [31], does not mean that the fundamental energy equipartition theorem $m\langle\upsilon^2\rangle/2 = k_B T/2$ is broken in equilibrium fluids [19] or that the above expression for $C_\upsilon(0)$ is incorrect. This misunderstanding has arisen in some early studies of the VAF for one-dimensional BM in a viscous incompressible fluid [27,32–35]. The modification of the relation for $C_\upsilon(0)$ is merely due to the limitation on the applicability of the Boussinesq force to incompressible fluids, i.e., to times $t >> a/c$, where c is the velocity of sound. At shorter times the compressibility of the fluid must be taken into account. As it was explained already in the work [5] and later proven in [30,36,37], for compressible fluids the equipartition holds in its commonly known form.

The solution to Eq. (6) can be immediately obtained by Laplace transform $\tilde{V}(s) = \{V(t)\} = \int_0^\infty dt V(t)\exp(-st)$:

$$\tilde{V}(s) = \frac{2k_B T}{s}\left[m^*s + \tilde{\gamma}(s) + s\tilde{\zeta}_B(s) + \frac{k}{s}\right]^{-1}. \tag{7}$$

From here, the Laplace transform of the VAF is $\tilde{C}_\upsilon(s) = s\tilde{V}(s)/2$. The obtaining of these functions in the time domain is now just a technical problem. If the kernel $\gamma(t)$ is replaced by $\gamma(t) = 2\gamma\delta(t)$, $F_S(t)$ becomes the Stokes force $-\gamma\upsilon(t)$ present in the original form of the Boussinesq force when $\zeta_B = \gamma(\tau_f/\pi t)^{1/2}$ and $\tilde{\zeta}_B(s) = \gamma(\tau_f/s)^{1/2}$. Having $\tilde{\gamma}(s) = \gamma$, in accordance with Eq. (4) one obtains a less general form of the solution (7),

$$\tilde{C}_\upsilon(s) = \frac{k_B T}{m^*}\left[s + \frac{(\tau_f s)^{1/2}}{\tau} + \frac{1}{\tau} + \frac{k}{m^* s}\right]^{-1}. \tag{8}$$

Below we skip the term $\sim k$ in Eq. (7). Since we want to establish the FDR, this term is insignificant in the linear theory where the correlation properties of the thermal force do not depend on the external forces applied to the system [21].