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Generalized Langevin equation with hydrodynamic backflow: Equilibrium properties

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ABSTRACT

We review equilibrium properties for the dynamics of a single particle evolving in a visco-elastic medium under the effect of hydrodynamic backflow which includes added mass and Basset force. Arbitrary equilibrium forces acting upon the particle are also included. We discuss the derivation of the explicit expression for the thermal noise correlation function that is consistent with the fluctuation–dissipation theorem. We rely on general time-reversal arguments that apply irrespective of the external potential acting on the particle, but also allow one to retrieve existing results derived for free particles and particles in a harmonic trap. Some consequences for the analysis and interpretation of single-particle tracking experiments are briefly discussed.

1. Introduction

Single-particle tracking experiments can access dynamical, structural and microrheological properties of complex visco-elastic media such as polymer gels or living cells [1,2]. Random displacements of a tracer are often analyzed with the help of a generalized Langevin equation which incorporates all relevant interactions of the tracer, e.g., viscous or visco-elastic Stokes force, inertial and hydrodynamic effects, active pulling by motor proteins, and eventual optical trapping [3–10]. Since several different mechanisms interplay in a complex medium, the correct formulation of the underlying phenomenological model can be sophisticated. For instance, the correlation function of the thermal noise has to be related, at equilibrium, to the memory kernels of the generalized Stokes and Basset forces according to the fluctuation–dissipation theorem. A recent experiment by Kheifets et al. [11] tracking micrometer-sized glass beads in water or acetone reveals that equipartition is broken in equilibrium by a contribution involving the mass of the displaced fluid. This raises the question of which ingredients relating to the surrounding fluid will appear in other manifestations of equilibrium, such as the fluctuation–dissipation theorem.

In this paper, we investigate the equilibrium properties of a generalized Langevin equation with hydrodynamic interactions and we provide the correct noise correlation function, consistent with the fluctuation–dissipation theorem. The role of the acceleration of the displaced fluid is sorted out, thus justifying the assumption made in Ref. [12] and amending that of Refs. [9,10]. Our analysis goes along the lines of that of Baiesi et al. [13]. Some consequences for the analysis and interpretation of single-particle tracking experiments are briefly discussed.

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2. Model

We are interested in the short time-scale motion of a tracer with mass m the displacement of which takes place in a complex visco-elastic medium, such as a gel. For simplicity, we restrict here to the one dimensional case, although generalization to two and three dimensions is straightforward. We denote by $x(t)$ the tracer's position, and we assume the tracer is subjected to an external force $F_{\rm ext}$ and we further allow ourselves the possibility to apply a small perturbation force $f_{\rm P}$. Newton's equation for the tracer reads

$$m\ddot{x} = F_{\rm S} + F_{\rm B} + F_{\rm ext} + f_{\rm P} + \xi, \tag{1}$$

where $\ddot{x}$ is the tracer's acceleration. In Eq. (1), in addition to the deterministic forces $F_{\rm ext}$ and $f_{\rm P}$, we have included a Gaussian colored noise $\xi(t)$ accounting for the interaction of the tracer with the heat bath. We have also included a generalized Stokes force $F_{\rm S}$, which expresses the viscous friction exerted by the fluid on the tracer. The latter force, when coarse-graining out the degrees of freedom of the surrounding medium, can be cast in the form [14,15]

$$F_{\rm S}(t) = -\int_{t_0}^{\infty} {\rm d}t' \gamma(t-t')\dot{x}(t'), \tag{2}$$

where the memory kernel $\gamma(\tau)$ is causal (i.e., $\gamma(\tau) = 0$ for $\tau < 0$), and the starting time t_0 is typically set either to $-\infty$ or to 0. A number of experiments [16–19] in living cells or in synthetic polymer solutions point to γ being accurately described by a power law [4,6], thereby expressing that a hierarchy of time-scales is involved in viscous friction for these complex media. Much less studied in a visco-elastic medium is the Basset force $F_{\rm B}$ which we have also included in Eq. (1) following Refs. [9,10]. As much as the usual inertia contribution $m\ddot{x}$, the Basset force is usually negligible at the macroscopic observation time scales considered in standard tracking experiments, but its effects have been shown to be prominent at short time-scales in Refs. [20,21,7–10]. This force is related to the inertia of the boundary layer surrounding the tracer. While the initial derivation for the expression of the Basset force in terms of the tracer's position dates back to Boussinesq for Newtonian fluids, Zwanzig and Bixon [22,23] provided a derivation of that force for a visco-elastic fluid characterized by a memory kernel γ as in Eq. (2). The generalized Basset force then reads

$$F_{\rm B}(t) = -\frac{m_{\rm f}}{2}\ddot{x}(t) - \int_{t_0}^{\infty} {\rm d}t' \zeta_{\rm B}(t-t')\ddot{x}(t'), \tag{3}$$

where $m_{\rm f}$ is the mass of the fluid displaced by the tracer. The memory kernel $\zeta_{\rm B}$ is causal as well, and can be argued to be related to γ in the following fashion:

$$\hat{\zeta}_{\rm B}(\omega) = 3\sqrt{\frac{m_{\rm f}\hat{\gamma}(\omega)}{2i\omega}}, \qquad \tilde{\zeta}_{\rm B}(s) = 3\sqrt{\frac{m_{\rm f}\tilde{\gamma}(s)}{2s}} \tag{4}$$

where the hat and the tilde stand for the Fourier and the Laplace transforms, respectively. In order to arrive at Eq. (4), the argument put forward in Ref. [22] goes as follows: for a Newtonian fluid, one has $\hat{\zeta}_{\rm B}(\omega) = 6\pi a^2\sqrt{\frac{\rho_{\rm f}\eta}{i\omega}}$, where a is the tracer's radius. For a visco-elastic medium, the viscosity is to be replaced with its frequency-dependent expression $\hat{\eta}(\omega)$, thus leading to $\hat{\zeta}_{\rm B}(\omega) = 6\pi a^2\sqrt{\frac{\rho_{\rm f}\hat{\eta}}{i\omega}}$. Finally, with the generalized Stokes law $\hat{\gamma} = 6\pi\hat{\eta}a$ for spherical tracers, we obtain Eq. (4). Note that the following derivation does not rely on relation (4) between memory kernels $\gamma(t)$ and $\zeta_{\rm B}(t)$, and it is thus valid in a more general situation.

The question we now ask regards to thermal noise correlations $\sigma(t-t') = \langle\xi(t)\xi(t')\rangle$ that we must impose to ensure that in the absence of a perturbing force ($f_{\rm P} = 0$) and for a conservative external force $F_{\rm ext}$ that derives from a potential, the tracer undergoes equilibrium and reversible dynamics, in agreement with, *e.g.*, the fluctuation–dissipation theorem. In the absence of the Basset force, this issue has been settled in the seminal paper by Kubo [24] and further discussed in the nice reviews by Mainardi et al. [25] or by Hänggi [26]. We begin by recalling the expression of the fluctuation–dissipation theorem.

3. Stating the fluctuation–dissipation theorem

The response of a position-dependent observable A to an infinitesimal external perturbation $f_{\rm P}(t')$ is denoted by χ and it is defined by

$$\chi(t,t') = \left.\frac{\delta\langle A(t)\rangle}{\delta f_{\rm P}(t')}\right|_{f_{\rm P}=0}. \tag{5}$$

Equilibrium first requires stationarity, namely time-translation invariance, so that $\chi(t,t') = \chi(t-t')$ in the regime of interest. Causality ensures the response function vanishes if the measurement is performed before the perturbation, when $t \le t'$. The fluctuation–dissipation theorem (FDT) states that in equilibrium the response is related to the correlation between the observable and the perturbation as [27]:

$$\chi(t-t') = \beta\frac{\partial\langle A(t)x(t')\rangle}{\partial t'}\Theta(t-t'), \tag{6}$$