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.

Keywords: Generalized Langevin equation, Fluctuation-Dissipation Theorem, Basset Force, Hydrodynamics, Subdiffusion, Optical Tweezers

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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 \cite{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 \cite{3, 4, 5, 6, 7, 8, 9, 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 \textit{et al.} \cite{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 [12] and amending that of [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.

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 $F_{\text{ext}}$ and we further allow ourselves the possibility to apply a small perturbation force $f_P$. Newton’s equation for the tracer reads

$$m \dddot{x} = F_S + F_B + F_{\text{ext}} + f_P + \xi ,$$  

(1)

where $\dddot{x}$ is the tracer’s acceleration. In Eq. (1), in addition to the deterministic forces $F_{\text{ext}}$ and $f_P$, 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 $F_S$, 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]

$$F_S(t) = -\int_{t_0}^{\infty} dt' \gamma(t - t') \dddot{x}(t'),$$  

(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, 17, 18, 19] in living cells or in synthetic polymer solutions point to $\gamma$ 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 $F_B$ which we have also included in Eq. (1) following [9, 10]. As much as the usual inertia contribution $m \dddot{x}$, the Basset force in usually negligible at the macroscopic observation time scales considered in standard tracking experiments, but its effects have been shown to be prominent at short timescales in [20, 21, 7, 8, 9, 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 $\gamma$ as in Eq. (2). The generalized Basset force then reads

$$F_B(t) = -\frac{m}{2} \dddot{x}(t) - \int_{t_0}^{\infty} dt' \zeta_B(t - t') \dddot{x}(t'),$$  

(3)
where $m_f$ is the mass of the fluid displaced by the tracer. The memory kernel $\zeta_s$ is causal as well, and can be argued to be related to $\gamma$ in the following fashion

$$\hat{\zeta}_s(\omega) = 3\frac{m_f\gamma(\omega)}{2\omega}, \quad \tilde{\zeta}_s(s) = 3\frac{m_f\gamma(s)}{2s}. \tag{4}$$

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 [22] goes as follows: for a Newtonian fluid, one has $\hat{\zeta}_s(\omega) = 6\pi a^2 \sqrt{\frac{m_f}{\eta}},$ 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 $\hat{\zeta}_s(\omega) = 6\pi a^2 \sqrt{\frac{m_f}{\eta}}/\omega.$ Finally, with the generalized Stokes law $\hat{\jmath} = 6\pi \eta a$ for spherical tracers, we obtain Eq. (4). Note that the following derivation does not rely on relation (4) between memory kernels $\gamma(t)$ and $\zeta_s(t),$ and 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 ($f_t = 0$) and for a conservative external force $F_{ext}$ 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 Bas-set 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 $f_t(t')$ is denoted by $\chi$ and is defined by

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

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 $t \leq t'.$ The fluctuation–dissipation theorem (FDT) states that in equilibrium the response is related to the correlation between the observable and the perturbation as [27]:

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

where $\beta = 1/(k_BT), T$ is the bath temperature, and $\Theta$ denotes the Heaviside function. Stationarity also leads to $\langle A(t)x(t') \rangle = \langle A(t-t')x(0) \rangle.$ The FDT can be written without enforcing explicit causality as

$$\chi(\tau) - \chi(-\tau) = -\beta \frac{d\langle x(\tau)A(0) \rangle}{d\tau}. \tag{7}$$

In single-particle tracking experiments the observable $A$ is the tracer’s position $x(t)$ and $\langle A(t)x(t') \rangle = \langle x(t)x(t') \rangle = C_x(t,t')$ is the position auto-correlation function, which, in equilibrium, is a function of $t-t'$ only, $C_x(t,t') = C_x(t-t').$ The FDT in Eq. (7) has the equivalent Fourier formulation

$$k_BT = \frac{-\omega\hat{C}_x(\omega)}{2\hat{\chi}(\omega)} \tag{8}$$
where $\tilde{\chi}''$ denotes the imaginary part of the response Fourier transform (and where our convention for the Fourier transform is $\hat{f}(\omega) = \int_{-\infty}^{\infty} dt \ e^{-i\omega t} f(t)$). Alternatively, the FDT can be stated in the Laplace domain in terms of the mean square displacement (MSD) $\langle \Delta x^2(t) \rangle = 2(C_x(0) - C_x(t))$ as:

$$k_B T = \frac{s \langle \Delta \tilde{x}^2 \rangle(s)}{2 \tilde{\chi}(s)}.$$  (9)

where the Laplace transform is defined by $\tilde{f}(s) = \int_0^{\infty} dt \ e^{-st} f(t)$.

In systems with a very small Reynolds number such as living cells, that is when inertial effects are negligible—which includes the Basset force—the response function is simply related to the Stokes memory kernel in the Laplace domain by $\tilde{\chi}(s) = 1/(s \tilde{\gamma}(s))$. The FDT is then usually stated in terms of the complex modulus $G^*(s) = s \tilde{\eta}(s)$ as [28, 29, 30]:

$$\langle \Delta \tilde{x}^2 \rangle(s) = \frac{k_B T}{3 \pi a s G^*(s)}.$$  (10)

4. Noise correlations in equilibrium

Our goal is now to explicitly derive the expression of the thermal noise correlations $\langle \xi(t)\xi(t') \rangle = \sigma(t-t')$, as imposed by the FDT in the presence of inertial effects. By definition, the function $\sigma$ is even, $\sigma(t) = \sigma(-t)$. Here we follow the approach presented in [31, 32]. Since the thermal noise has Gaussian statistics, the probability weight $P$ associated with a given realization of the thermal noise is $P[\xi] \propto e^{-S[\xi]}$, where $S[\xi] = \frac{1}{2} \int_{t_0}^{\infty} dt_1 dt_2 \xi(t_1) \Gamma(t_1-t_2) \xi(t_2)$. The expression of $\xi$ in this formula is determined by the tracer’s dynamics in Eq. (1), and the symmetric function $\Gamma$ is related to the thermal noise correlations by $\int_{t_0}^{\infty} dt_1 \sigma(t-t_1) \Gamma(t_1-t') = \delta(t-t')$. The application of the external perturbation $f_s$ results in a variation $\delta S$ of $S$, so that the response function is expressed as:

$$\chi(t, t') = - \left. \left( A(t) \frac{\delta S}{\delta f_s(t')} \right)_{f_s=0} \right|_{f_s=0}.$$  (11)
Substituting $\xi$ from Eq. (1) into $\mathcal{S}[\xi]$ and calculating the functional derivative in Eq. (11) yields the expression of the response function $\chi = \chi_{\text{in}} + \chi_{\text{ext}} + \chi_{s} + \chi_{n}$, with four contributions:

\begin{align}
\chi_{\text{in}}(t, t') &= m^* \int_{t_0}^{\infty} \int_{t_0}^{\infty} dt_1 \Gamma(t_1 - t') \langle \ddot{x}(t_1) A(t) \rangle , \\
\chi_{\text{ext}}(t, t') &= - \int_{t_0}^{\infty} \int_{t_0}^{\infty} dt_1 \Gamma(t_1 - t') \langle F_{\text{ext}}(t_1) A(t) \rangle , \\
\chi_{s}(t, t') &= \int_{t_0}^{\infty} \int_{t_0}^{\infty} dt_1 dt_2 \Gamma(t_1 - t') \gamma(t_1 - t_2) \langle \ddot{x}(t_2) A(t) \rangle , \\
\chi_{n}(t, t') &= \int_{t_0}^{\infty} \int_{t_0}^{\infty} dt_1 dt_2 \Gamma(t_1 - t') \zeta_0(t_1 - t_2) \langle \ddot{x}(t_2) A(t) \rangle ,
\end{align}

where $m^* = m + m_{\text{f}}/2$. In order to compare this prediction with the FDT, we focus on the regime where the system reaches an equilibrium state, namely when the dynamics does no longer depend on initial conditions, by setting $t_0 \to -\infty$. In this regime, the correlation functions are time–translation invariant, and the response functions depend only on the time lag $\tau = t - t'$. We split the difference $\chi(\tau) - \chi(-\tau)$ into four contributions corresponding to the functions defined in Eq. (12). The first contribution is expressed as:

$$
\chi_{\text{in}}(\tau) - \chi_{\text{in}}(-\tau) = m^* \int_{-\infty}^{\infty} dt_1 \left[ \Gamma(t_1 - \tau) \langle \ddot{x}(t_1) A(0) \rangle - \Gamma(t_1 + \tau) \langle \ddot{x}(t_1) A(0) \rangle \right].
$$

(13)

We perform the change of variable $t_1 \to -t_1$ in the second integral. The key trademark of equilibrium that we now make use of is time reversibility, which implies, $\langle \ddot{x}(t_1) A(0) \rangle = \langle \ddot{x}(-t_1) A(t) \rangle$. Given that $\Gamma$ is even, it follows $\chi_{\text{in}}(\tau) = \chi_{\text{in}}(-\tau)$, and we show similarly that $\chi_{\text{ext}}(\tau) = \chi_{\text{ext}}(-\tau)$. We perform the changes of variable $t_1 \to -t_1$ and $t_2 \to -t_2$ in the expression of $\chi_{s}$ and $\chi_{n}$, and given the parity of the observables in the correlation functions of Eqs. (12c) and (12d), we deduce:

\begin{align}
\chi_{s}(\tau) - \chi_{s}(-\tau) &= \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} dt_1 dt_2 \Gamma(t_1 - \tau) \langle \ddot{x}(t_2) A(0) \rangle \\
&\times \left( \gamma(t_1 - t_2) + \gamma(t_2 - t_1) \right), \\
\chi_{n}(\tau) - \chi_{n}(-\tau) &= \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} dt_1 dt_2 \Gamma(t_1 - \tau) \langle \ddot{x}(t_2) A(0) \rangle \\
&\times \left( \zeta_0(t_1 - t_2) - \zeta_0(t_2 - t_1) \right).
\end{align}

(14a, 14b)
As a result, we finally obtain the expression of the difference $\chi(\tau) - \chi(-\tau)$ in terms of the kernels appearing in the generalized Stokes force and the Basset force. For an equilibrium process, this expression should be identical to the prediction of the FDT in Eq. (7), which enforces that

$$\beta \left\langle \dot{x}(\tau) A(0) \right\rangle = \int_{-\infty}^{\infty} dt_1 dt_2 \Gamma(t_1 - \tau) \times \left[ \left( \gamma(t_1 - t_2) + \gamma(t_2 - t_1) \right) \left\langle \dot{x}(t_2) A(0) \right\rangle \right. \left. + \left( \zeta_\alpha(t_1 - t_2) - \zeta_\alpha(t_2 - t_1) \right) \left\langle \ddot{x}(t_2) A(0) \right\rangle \right].$$

(15)

This relation is independent of the parity of the observable we consider for the response function. In the case where a more general perturbation force $f_P(x) = -a_P(t) U'(x(t))$ is applied to the tracer, it is also possible to define the response function with respect to the parameter $a_P$:

$$\chi_U(t, t') = \delta \left\langle A(t) \right\rangle \bigg|_{a_P=0} \delta a_P(t') \bigg|_{a_P=0}.$$  

(16)

We then recover the standard FDT (analogous to Eq. (6)):

$$\chi_U(t - t') = \beta \frac{\partial \left\langle A(t) U(x(t')) \right\rangle}{\partial t'} \Theta(t - t').$$

(17)

In the Fourier domain, and since the Fourier transform of thermal correlations is related to $\hat{\Gamma}$ as: $\hat{\sigma}(\omega) = 1/\hat{\Gamma}(\omega)$, we obtain from Eq. (15)

$$\hat{\sigma}(\omega) = 2k_B T \left( \hat{\gamma}'(\omega) - \omega \hat{\zeta}''(\omega) \right),$$

(18)

where $\hat{\gamma}'$ and $\hat{\zeta}''$ denote the real part of the $\gamma$ Fourier transform and the imaginary part of the $\zeta$ Fourier transform, respectively. Hence, we deduce the thermal noise correlations read:

$$\left\langle \xi(t) \xi(t') \right\rangle = k_B T \left[ \gamma \left( |t - t'| \right) + \frac{d\zeta}{dt} \left( |t - t'| \right) \right].$$

(19)

This result can be decoded as an effective visco–elastic memory kernel $\gamma^* = \gamma + \frac{d\zeta}{dt}$, which could have been guessed by integrating by parts the Basset memory term. In that case however integration by parts involves a $\zeta_\alpha(0)$ term which at best is not well defined, while our derivation encompasses this problem by using an anti–symmetric function $\zeta_\alpha(t) - \zeta_\alpha(-t)$. Note that $m_t$ does not appear in this expression, so that only the terms with memory kernels in the Basset force and the generalized Stokes force contribute to the dissipation of the tracer with the heat bath as expressed by the FDT. This is in fully consistent with the free-particle situation considered by Felderhof [33] (his Eq. (2.10)) or by Indei et al. [8] (their Eqs. (64) and (65)). In the Laplace domain, the thermal correlation function is expressed as:

$$\left\langle \hat{\xi}(s) \hat{\xi}(s') \right\rangle = k_B T \left[ \frac{\hat{\gamma}(s) + \hat{\gamma}(s')}{{s} + s'} + \frac{s \hat{\zeta_\alpha}(s) + s' \hat{\zeta_\alpha}(s')}{{s} + s'} \right].$$

(20)
The equipartition theorem represents an alternative method to characterize equilibrium properties. It relates the initial value of the velocity autocorrelation function $C_v(t-t') = \langle \dot{x}(t) \dot{x}(t') \rangle$ to the bath temperature as: $C_v(0) = k_B T/m$. By using the FDT prediction in Eq. (9), and given the velocity autocorrelation function is simply related to the MSD in the Laplace domain as: $\hat{C}_v(s) = \frac{1}{2} s^2 \langle \Delta x^2 \rangle \langle s \rangle$, we deduce: $\hat{C}_v(s) = k_B T s \hat{G}(s)$, where $G$ denotes the “usual” response function [4, 6]. Considering the dynamics described by Eq. (1) with an external force $F_{ext} = -k x$, we compute the response function in the Laplace domain, and we use Eq. (4) to obtain:

$$\hat{G}(s) = \frac{1}{s^2 m^* + 3 s^{3/2} \sqrt{m \bar{\gamma}(s)/2} + s \bar{\gamma}(s) + k}.$$  \hspace{1cm} (21)

From the initial value theorem, we finally deduce:

$$\frac{C_v(0)}{k_B T} = \lim_{s \to \infty} \frac{1}{m^* + 3 \sqrt{m \bar{\gamma}(s)/(2s)} + \bar{\gamma}(s)/s + k/s^2}.$$  \hspace{1cm} (22)

As discussed in Sec. 2, the Laplace transform of the Stokes memory kernel in the high frequency regime behaves like $s^{\alpha-1}$, where $\alpha < 2$, so that: $\bar{\gamma}(s)/s \to 0$. It follows that the initial value of the velocity autocorrelation function $\langle \dot{x}^2 \rangle = k_B T/m^*$ is different from the “usual” equipartition theorem prediction, as already noticed in [12]. Earlier works on this subject, like those of Widom [34] or Case [35] used to determine correlation functions by assuming the “usual” equipartition, leading to slightly wrong results. Here we show that using the FDT as starting point avoids such issues. We also note that this result remains the same if we consider a constant external force $F_{ext}$ is applied to the tracer, the initial value of the velocity autocorrelation function $\gamma(t)$ is positive, $\gamma(t)$ can be replaced by $-\langle \dot{x} F_{ext} \rangle_{eq}/\langle \dot{x}^2 \rangle_{eq} = k_B T/\langle \dot{x}^2 \rangle_{eq}$. Given the process defined in Eq. (1) has a Gaussian statistics, an experimental method to verify the validity of this result lies in measuring the stationary distribution of the tracer’s velocity [11], for which the variance should equal the initial value of the velocity autocorrelation function. For an overdamped system in the absence of external force, the condition $\langle \Delta x^2 \rangle(0) = 0$ associated with the FDT prediction in Eq. (9) imposes $\alpha$ is positive, meaning the Stokes kernel necessarily diverges in the short time limit for such a system.

In summary, we have revised some equilibrium properties of generalized Langevin equation with hydrodynamic interactions. Under the fluctuation-dissipation theorem, the memory kernels $\gamma(t)$ and $\zeta_\alpha(t)$ of generalized Stokes and Basset forces have been related to the noise correlation function $\langle \xi(t) \xi(t') \rangle$ according to Eq. (19). The derivation is valid in both Fourier and Laplace domains. This relation allows one to refine phenomenological models that are used for the analysis and interpretation of single-particle tracking experiments in complex visco-elastic media, notably in living cells. In particular, we showed that the noise correlation function in [9, 10] should not contain the term $m_t s/2$ which came from a naive extension of the fluctuation-dissipation theorem to the Basset force (since this term could alter tracer’s dynamics only at very short time scales, its presence does not affect the results reported in [9, 10]). Note also that relation (4) between the memory kernels of the generalized Stokes and Basset forces allows one to reduce the number of model parameters in [9, 10] yielding potentially more robust fits. Future optical tweezers single-particle tracking experiments at short time scales can further clarify hydrody-
namic interactions between the tracer, the solvent, and semi-flexible polymers such as, e.g., actin filaments.

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