MEMORY EFFECT ON FORCE MEASUREMENTS AT NANOSCALES

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1. Introduction

At present, new methods of measurements of ultra-small forces down to femtonewtons are developed [1]. Due to the central role that the conception of force plays in physics and with the increased interest to the investigations of systems at small space and time scales, these methods attract great attention. The principal problem in such measurements on micro- and nano-objects is connected with the influence of ever-present thermal noise. As distinct from macroscopic systems, the noise can essentially affect the motion of small objects and the forces originating from the noise can even exceed the conservative forces in the system. This can lead to a completely incorrect interpretation of the experiments. This question is not solved today and there is an interesting discussion about it in the literature [1-4]. In [1,4], two methods of force measurements have been used to determine the influence of noise on a colloidal particle near a wall in the presence of a gravitational field. One of the methods consisted in measuring the particle drift velocity and the subsequent use of overdamped Langevin equation to determine the full force acting on the particle. In the second method the force was determined from the known particle-wall interaction potential using the equilibrium Boltzmann distribution. The results of these methods were strongly contrasting: the obtained forces deviated both in their magnitudes and even in their sign. As a resolution of these discrepancies, criticized however in [2], the choice of the anti-Itô conception of stochastic calculus has been suggested for the case of spatially inhomogeneous diffusion. In the present contribution we address one more problem of the measurements of forces acting on microscopic and nanoscopic objects that exists even in the case of homogeneous diffusion (for unbounded particles and constant forces, or the ones linearly depending on the particle position). This problem arises with lowering the time of measurement when the particles undergo unsteady motion for which the standard Langevin description becomes inappropriate. We show how due to the hydrodynamic aftereffect the experimentally determined force, even if it is known to be constant, appears to depend on the relation between the time of measurement and a characteristic time of the loss of memory in the system.

2. Drift method of force measurements at short time scales

It follows from the overdamped Langevin equation that when a microscopic body is suspended in a liquid, a force F applied to a body results in a drift velocity $\upsilon = F/\gamma$, where γ is the object´s friction coefficient. This is true for large constant forces, if the inertial effects are neglected. However, when the drift force amplitude is comparable to the effect of thermal noise and the force depends on the particle position, the equation for F must be corrected [1] since the measured velocities are statistically distributed. Moreover, an additional term -$\alpha\gamma(x)\mathrm{d}D(x)/\mathrm{d}x$, referred to as *spurious force*, should be added to the force $F(x) = \gamma(x)\langle \upsilon(x)\rangle$ (for the force changing in the direction x). Here, $D(x)$ is the position-dependent diffusion coefficient and α is a constant from the interval [0, 1]. For Brownian particles the preferred value is $\alpha = 1$ [5] but there is no common agreement as to this choice [2], which significantly affects the stochastic calculus. One more problem, to our knowledge not considered so far in the interpretations of force measurements, arises in situations when inertial effects (and, consequently, the memory in the particle dynamics) can play a role [6]. Even if the applied force is position independent, but the observation times become comparable to the vorticity time $\tau_R = R^2\rho/\eta$, where R is the particle radius and ρ and η are the density and viscosity of the solvent, the discussed method is not applicable. We will show it coming from the generalized Langevin equation [7],

$$M\dot{\upsilon}_t\left(t\right)+\gamma\upsilon_t\left(t\right)+\int_0^t\Gamma\left(t-t'\right)\dot{\upsilon}_t\left(t'\right)\mathrm{d}t'=F+\eta\left(t\right). \tag{1}$$

Here, $\eta(t)$ is a random noise force driving the particles of mass M, and F is a regular force to be determined. The force η is, due to the fluctuation dissipation theorem, connected to the dissipative properties of the system. We will consider the very realistic case when the memory in the system is of the hydrodynamic kind, i.e., the resistance force against the particle motion follows from the non-stationary Navier-Stokes equations of motion for incompressible fluids [8]. Then the kernel Γ is $\Gamma(t)=\gamma(\tau_R/\pi t)^{1/2}$. The usual Brownian relaxation time is connected to the Stokes friction coefficient as $\tau = M/\gamma$. Equation (1) for F = 0 describes the zero-mean fluctuations $\upsilon(t)$. Here we are interested in the question how the thermal noise influences the determination of the force F. Let F be constant, as it is for a freely falling particle in a fluid. We express the velocity as $\upsilon_t = \upsilon + \upsilon^*$. The deterministic part υ^* (the drift velocity) obeys the averaged Eq.(1), i.e. the equation without the random force

$$\dot{\upsilon}^* + \frac{1}{\tau}\upsilon^* + \frac{1}{\tau}\sqrt{\frac{\tau_R}{\pi}}\int_0^t \frac{\dot{\upsilon}^*(t')}{\sqrt{t-t'}}\mathrm{d}t' = \frac{F}{M}. \tag{2}$$

We can choose the initial condition as $\upsilon^*(0) = 0$. Then the Laplace-transformed Eq.(2) has the following solution for $\tilde{\upsilon}^*(s) = \Lambda\{\upsilon^*(t)\}$:

$$\tilde{\upsilon}^*(s) = \frac{F}{Ms}\frac{1}{\lambda_1 - \lambda_2}\left(\frac{1}{\sqrt{s}-\lambda_1} - \frac{1}{\sqrt{s}-\lambda_2}\right), \tag{3}$$

where $\lambda_{1,2} = -\left(\tau_R^{1/2}/2\tau\right)\left(1 \mp \sqrt{1-4\tau/\tau_R}\right)$ are the roots of equation $s + \left(\tau_R s\right)^{1/2}\tau^{-1} + \tau^{-1} = 0$. The inverse transform has the form

$$\upsilon^*(t) = \frac{F}{M}\left\{\tau + \frac{1}{\lambda_2 - \lambda_1}\sum_{i=1}^{2}(-1)^i \frac{1}{\lambda_i}\exp\left(\lambda_i^2 t\right)\mathrm{erfc}\left(-\lambda_i\sqrt{t}\right)\right\}. \tag{4}$$

The asymptotic behaviour of this solution at $t \to 0$ is $\upsilon^*(t) \approx Ft/M$. At long times we have $\upsilon^*(t) \approx (F\tau/M)\left[1 - (\tau_R/\pi t)^{1/2} + \ldots\right]$. Using these formulas, the force F can be determined through the measured mean velocity $\upsilon^*(t)$ at any time t. At long times, due to the hydrodynamic aftereffect, this velocity depends on the relation τ_R/t and very slowly approaches the limiting value F/γ. The determined force is

$$F \approx \gamma\upsilon^*\left(1 + \sqrt{\tau_R/\pi t}\right), \qquad \tau_R/t << 1. \tag{5}$$

The stochastic motion of the particle, for constant γ, does not influence the determination of the force, since its contribution to the drift velocity is zero. It is easy to see that our correction of the standard result for the force, $F = \gamma\upsilon^*$, can be significant. To demonstrate it, let us turn to the recent work [6]. The smallest observation times in this experiment were ~ 10^{-8} s. At such times the force should be proportional to υ^*/t. At longer times, when the ratio τ_R/t is small, Eq. (5) applies. For spherical particles 1μm in radius, which are suspended in water at room temperatures, we have $\tau_R \sim 10^{-6}$ s. Thus, at the times ~ 10^{-5} s the correction represents almost 20% and slowly drops with the increase of time, approaching 1% of $\gamma\upsilon^*$ at $t \cong$ 3ms.

3. Conclusion

In conclusion, we have obtained an exact solution for the drift velocity of a Brownian particle in an incompressible fluid under the action of a constant force, taking into account the hydrodynamic memory in the particle motion. This velocity is proportional to the applied force but depends in a complicated manner on the time of observation *t*. At short times it is proportional to *t* and at long times it contains algebraic tails, the longest-lived of which being $\sim t^{-1/2}$. Due to this the velocity very slowly approaches the limiting value F/γ. As a consequence, the force *F* can significantly differ from the value that would be extracted from the drift measurements neglecting the inertial effects, which is a standard assumption in the interpretation of such experiments [1-4]. The presented method can be equally applicable in the case of force linearly depending on the particle position. For nonlinear forces, first the open question about the choice of convention to be used in stochastic calculus should be resolved.

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