HYDRODYNAMIC BROWNIAN MOTION OF PARTICLES IN A HARMONIC TRAP

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Received 10 May 2011; accepted 29 May 2011.

1. Introduction

At the late sixtieth and early seventieth of the last century the famous long-time "tails" of the molecular velocity autocorrelation function (VAF) have been discovered. First in the computer experiments, and later they have been confirmed theoretically and experimentally. This discovery doubted the commonly accepted view on the microscopic and macroscopic properties of liquids as being characterized by very different time scales, and extended the range of the applicability of classical hydrodynamics. It is less known that these tails have been correctly predicted much earlier in the work by Vladimirsky and Terletzky [1], the authors of the first hydrodynamic theory of the Brownian motion (BM). The hydrodynamic approach has essentially enriched the classical Einstein theory valid only for $t \to \infty$. It has also revealed the limits of its later generalization for arbitrary times. Such a generalization was made by Langevin who proposed the first stochastic differential equation for the description of the memoryless BM. In the hydrodynamic theory the Langevin equation (LE) is modified to take into account a possible memory in the particle motion. In the present contribution we give an exact solution to the LE with hydrodynamic memory. It is essential that we use a simple method that is in linear consideration applicable for systems with any other kind of memory. The VAF has been found together with the mean square displacement (MSD) for Brownian particles (BP) trapped in a harmonic well.

2. Langevin equation for the Brownian motion with hydrodynamic memory

The standard LE for the velocity $\upsilon(t) = \mathrm{d}x/\mathrm{d}t$ of the BP has the form

$$m\frac{\mathrm{d}\upsilon}{\mathrm{d}t} = -\gamma\upsilon + \sqrt{2D}\xi(t), \tag{1}$$

where the coefficient of friction γ for a spherical particle with the radius R and the mass m is the Stokes one, $\gamma = 6\pi R\eta$ (η is the dynamic viscosity), and the erratic motion of the particle, resulting from random, uncompensated impacts of the molecules of the surrounding fluid is described by the stochastic (white noise) force ~ $\xi(t)$ with the statistical properties $\langle\xi(t)\rangle = 0$, $\langle\xi(t)\xi(t')\rangle = \delta(t-t')$ and the intensity $D = k_B T\gamma$ (k_B is the Boltzmann constant and T is the temperature). The Stokes friction force, which is traditionally used to describe the friction that a particle feels during its motion in a liquid, is in fact valid only for the steady motion of the particle (at long times), and should be replaced by the expression [2]

$$F(t) = -\gamma\left\{\upsilon(t) + \frac{\rho R^2}{9\eta}\frac{\mathrm{d}\upsilon}{\mathrm{d}t} + \sqrt{\frac{\rho R^2}{\pi\eta}}\int_{-\infty}^{t}\frac{\mathrm{d}\upsilon}{\mathrm{d}t'}\frac{\mathrm{d}t'}{\sqrt{t-t'}}\right\}, \tag{2}$$

where ρ is the density of the solvent. Equation (2) is valid for all times $t >> R/c$ (c is the sound velocity), i.e., except the very short times when the solvent compressibility must be taken into account. This expression has been derived by Boussinesq in 1885 [3], used in the mentioned work [1], and later brought to wider attention by Hinch [4]. It is seen from Eq.(2) that for fluids with the density comparable to the density of the BP (which is the usual case of freely buoyant particles), the terms additional to the Stokes one cannot be neglected since in the equation of motion for the particle they are of the same order as the inertial term. Here we will consider a more complicated problem of the movement of the BP, when the particle is subjected to an external harmonic potential.

3. Solution of the hydrodynamic Langevin equation

Now we show that the solution can be obtained very easily as follows. Based on another little known work [5], instead of Eq.(1) with the force (2) we can solve the deterministic "equation of motion" for the quantity $V(t) = \mathrm{d}X(t)/\mathrm{d}t$ with $X(t)$ being the particle MSD,

$$\dot{V}(t) + \frac{1}{\tau}\sqrt{\frac{\tau_R}{\pi}}\int_0^t\frac{\dot{V}(t')}{\sqrt{t-t'}}\mathrm{d}t' + \frac{1}{\tau}V(t) + \omega_0^2\int_0^t V(t')\mathrm{d}t' = 2\Phi_0, \qquad \Phi_0 = \frac{k_B T}{M}, \tag{3}$$

where $M = m + m_s/2$ (m_s is the mass of the solvent displaced by the particle) and $\omega_0^2 = k/M$ (k is the force constant of an external harmonic potential). The characteristic times in this equation are $\tau = M/\gamma$ (the relaxation time of the BP) and $\tau_R = \rho R^2/\eta$ (the vorticity time). The constant "force" $2k_BT$ at the right begins to act on the particle at the time $t = 0$; up to this moment the particle is at rest together with the liquid. The problem has to be solved with the evident initial conditions $V(0) = X(0) = 0$. It is also seen from Eq.(3) that $\dot{V}(0) = 2\Phi_0$. Taking the Laplace transformation Λ of Eq.(3), we obtain for $\tilde{V}(s) = \Lambda\{V(t)\}$

$$\tilde{V}(s) = 2\Phi_0 s^{-1}\left(s + \sqrt{\tau_R}\tau^{-1}s^{1/2} + \tau^{-1} + \omega_0^2 s^{-1}\right)^{-1}. \tag{4}$$

Its inversion gives the solution

$$V(t) = 2\Phi_0 \sum_{i=1}^{4} b_i z_i \exp\left(z_i^2 t\right)\mathrm{erfc}\left(-z_i\sqrt{t}\right), \tag{5}$$

where z_i are the roots of the quartic equation $z^4 + \tau_R^{1/2}\tau^{-1}z^3 + \tau^{-1}z^2 + \omega_0^2 = 0$ and the coefficients b_i can be easily determined decomposing the right hand side of Eq.(4) in simple fractions. The VAF $\Phi(t)$ is expressed by a similar equation, if one divides $V(t)$ by 2 and replaces $b_i z_i$ with $b_i z_i^3$. For $\omega_0^2 \to 0$ this expression exactly corresponds to the solutions found in Refs. [4, 6] and contains the long-time tail discovered already in the computer experiments [7, 8]. In our more general case it follows from Eq.(5) for the VAF at $t \to \infty$ that

$$\Phi(t) = \frac{\Phi_0}{2\sqrt{\pi}}\frac{1}{t^{3/2}}\sum_{i=1}^{4} b_i \sum_{m=1}^{\infty}(-1)^{m+1}\frac{(2m-1)!!}{\left(2z_i^2 t\right)^{m-1}}, \tag{6}$$

i.e., the longest-lived tail is $\sim t^{-3/2}$. Finally, the MSD of the BP is found integrating the function $V(t)$ from 0 to t,

$$X(t) = -2\Phi_0 \sum_{i=1}^{4}\frac{b_i}{z_i}\left[1 + 2z_i\sqrt{\frac{t}{\pi}} - \exp\left(z_i^2 t\right)\mathrm{erfc}\left(-z_i\sqrt{t}\right)\right]. \tag{7}$$

In the long time limit it agrees very well with experiment [9]. The asymptotic expansion of this equation for $t \to \infty$ is

$$X(t) = -2\Phi_0 \sum_{i=1}^{4}\frac{b_i}{z_i}\left\{1 + 2z_i\sqrt{\frac{t}{\pi}} + \frac{1}{z_i\sqrt{\pi t}}\left[1 + \sum_{m=1}^{\infty}(-1)^m\frac{(2m-1)!!}{\left(2z_i^2 t\right)^m}\right]\right\}. \tag{8}$$

4. Conclusions

For the BP moving in a fluid the standard Langevin equation does not represent a good model and the more correct hydrodynamic description should be used. We have solved the problem of the hydrodynamic BM of a particle in an external harmonic potential. Our description corresponds to a number of recent experiments on particles in optical traps. The obtained solution is exact for incompressible fluids described by the nonstationary Navier-Stokes equations. The found VAF and MSD generalize the known results in the absence of the potential well. Although the detection of the predicted long-time tail effect requires high spatial and temporal resolution, our results could be verified in similar experiments as in [9], where the hydrodynamic theory of the BM of a free particle has been definitely confirmed.

Acknowledgement

This work was financially supported by the Agency for the Structural Funds of the EU within the projects NFP 26220120021 and 26220120033, and by the grant VEGA 1/0300/09.

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