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Hydrodynamic Memory in the Motion of Charged Brownian Particles across the Magnetic Field

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An exact solution of the Langevin equation is given for a charged Brownian particle driven in an incompressible fluid by the magnetic field, taking into account the hydrodynamic aftereffect. The stochastic integro-differential Langevin equation is converted to a deterministic equation for the particle mean square displacement. We have found the mean square displacement and other time correlation functions describing the particle motion. For the motion along the field the known results from the theory of the hydrodynamic motion of a free Brownian particle are recovered. The correlation functions across the field contain at long times the familiar Einstein terms and additional algebraic tails. The longest-lived tail in the mean square displacement is proportional to $t^{1/2}$. At short times the motion is ballistic and independent of the magnetic field.

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1. Introduction

Beginning from the works by Taylor [1] and Kurţunoďlu [2], the Brownian motion (BM) of charged particles in magnetic fields has been studied in a number of papers (for a review see [3–5]). The dynamics of the Brownian particle (BP) was modeled by a Langevin equation (LE) [6] with the Stokes friction and the white-noise stochastic force. However, such a description of the BM in fluids is appropriate only for steady motion, i.e., at long times. Alternatively, it is valid for short times but for particles with the density much larger than that of the surrounding fluid [7]. In a more general description the friction force should reflect the memory in the BP motion. Accordingly, the fluctuation-dissipation theorem dictates that the stochastic force in the LE must be changed to a colored-noise force [6]. This results in a generalized LE, which is a Volterra-type integro-differential stochastic equation. For a charged BP in an external magnetic field such equation has been solved in [4] in the case when the memory kernel in the LE exponentially decreases in time. In the present contribution a different kind of memory is considered, the so-called hydrodynamic memory or viscous aftereffect (see [8] and references therein). It naturally arises within the nonstationary Navier–Stokes hydrodynamics and for particles moving in a liquid it reveals in the appearance of a resistance force (the Boussinesq–Basset force) that in the time t depends on the state of the particle motion in all the preceding moments of time. We give an exact solution to the corresponding LE using a method that is in linear consideration applicable for systems with any other kind of memory. The mean square displacement (MSD) for the BP along and across the magnetic field has been found together with other relevant time correlation functions (CFs), such as the time-dependent diffusion coefficient or the velocity autocorrelation function (VAF). The found CFs are characterized by interesting peculiarities, the algebraic long-time tails.

2. Generalized Langevin equation and its solution

The Boussinesq–Basset resistance force on a spherical particle in an incompressible liquid is [8]:

$$\boldsymbol{F}(t) = -\gamma\left[\boldsymbol{v}(t) + \frac{\rho R^2}{9\eta}\frac{\mathrm{d}\boldsymbol{v}}{\mathrm{d}t} + \sqrt{\frac{\rho R^2}{\pi\eta}}\int_{-\infty}^{t}\frac{\mathrm{d}\boldsymbol{v}}{\mathrm{d}t'}\frac{\mathrm{d}t'}{\sqrt{t-t'}}\right], \quad (1)$$

where $\boldsymbol{v}$ is the particle velocity, R — its radius, $\gamma = 6\pi\eta R$ is the friction factor, and ρ and η are the density and viscosity of the solvent. This equation is valid for all times $t \gg R/c$ (c is the sound velocity), i.e. very short times when the compressibility effects play a role are excluded from the consideration. If Q is the charge of the particle of mass m and density ρ', $\boldsymbol{B}$ is a constant induction of magnetic field along the axis z, then the LE of motion for the BP has the form

$$m\dot{\boldsymbol{v}}(t) + \boldsymbol{F}(t) = Q\boldsymbol{v}\times\boldsymbol{B} + \boldsymbol{f}(t)\,, \quad (2)$$

with $\boldsymbol{f}$ being the stochastic force. The projection of this equation onto the axis z does not contain the magnetic

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force so that along the field we deal with the motion of a free BP. If we are interested in the MSD of the BP, the following efficient method for solving Eq. (2) can be applied [7]. It consists in a simple replacement of the projections v_i, $i = x, y, z$, with the functions $V_i(t) = \mathrm{d}\xi_i/\mathrm{d}t$, where $\xi_i(t)$ is the MSD in the i direction (i.e. $\xi_x(t) = \langle \Delta x^2(t)\rangle = \langle [x(t)-x(0)]^2\rangle$, etc.). At the same time, the stochastic force must be replaced with $2k_\mathrm{B}T$, where k_B is the Boltzmann constant and T — the temperature. The resulting deterministic equation has to be solved with the initial conditions $\xi_i(0) = V_i(0) = 0$. So, the x projection of Eq. (2) converts to

$$\dot{V}_x(t) + \frac{1}{\tau}\sqrt{\frac{\tau_\mathrm{R}}{\pi}}\int_0^t \frac{\dot{V}_x(t')}{\sqrt{t-t'}}\mathrm{d}t' + \frac{1}{\tau}V_x(t) + \frac{1}{\tau_\mathrm{c}}V_y = \frac{2k_\mathrm{B}T}{M}. \tag{3}$$

Here $M = m + m_\mathrm{s}/2$ and m_s is the mass of the solvent displaced by the particle. The characteristic times in this equation are $\tau = M/\gamma$ (the usual Brownian relaxation time), $\tau_\mathrm{R} = R^2\rho/\eta$ (the vorticity time), and τ_c, which is connected to the cyclotron frequency $\omega_\mathrm{c} = QB/M = 1/\tau_\mathrm{c}$. Analogously, $\xi_y(t)$ is determined by the equation for $V_y(t)$ that differs from Eq. (2) only by the changes V_x to V_y and V_y/τ_c to $-V_x/\tau_\mathrm{c}$. Finally, the equation for $V_z(t)$ does not contain the magnetic field at all. The MSD across the field is determined by the sum $\xi_{xy} = \xi_x + \xi_y$. This quantity can be found using the Laplace transformation Λ. For $\tilde{V}_{xy}(s) = \Lambda[V_x(t)+V_y(t)]$ one obtains from Eq. (3)

$$\tilde{V}_{xy}(s) = \frac{4k_\mathrm{B}T}{Ms}\frac{\psi(s)}{\psi(s)^2 + \tau_\mathrm{c}^{-2}}, \tag{4}$$

where $\psi(s) = s + \tau^{-1}\sqrt{\tau_\mathrm{R}s} + \tau^{-1}$. For what follows we need the roots κ_j, $j = 1, \ldots 4$ of the equation $\psi(s)^2 + \tau_\mathrm{c}^{-2} = 0$ and the roots $\lambda_{1,2}$ of the quadratic equation $\psi(s) = 0$. The solutions are $2\kappa_{1,2} = \lambda_1 + \lambda_2 \pm \sqrt{(\lambda_1-\lambda_2)^2 - 4\mathrm{i}\omega_\mathrm{c}}$, $2\kappa_{3,4} = \lambda_1 + \lambda_2 \pm \sqrt{(\lambda_1-\lambda_2)^2 + 4\mathrm{i}\omega_\mathrm{c}}$, and $\lambda_{1,2} = -(\sqrt{\tau_\mathrm{R}}/2\tau)[1 \mp \sqrt{1-4\tau/\tau_\mathrm{R}}]$. Using these roots, the s-dependent fraction in Eq. (4) can be decomposed into simple fractions,

$$\frac{2\psi(s)}{\psi(s)^2+\tau_\mathrm{c}^{-2}} = \frac{1}{\kappa_2-\kappa_1}\left(\frac{1}{\sqrt{s}-\kappa_2} - \frac{1}{\sqrt{s}-\kappa_1}\right) + \frac{1}{\kappa_4-\kappa_3}\left(\frac{1}{\sqrt{s}-\kappa_4} - \frac{1}{\sqrt{s}-\kappa_3}\right),$$

after which the inverse Laplace transform of Eq. (4) yields

$$V_{xy}(t) = \frac{2k_\mathrm{B}T}{M}\left\{\left(\frac{1}{\kappa_1\kappa_2} + \frac{1}{\kappa_2-\kappa_1}\left[f(\kappa_2,t) - f(\kappa_1,t)\right]\right) + (\kappa_1\to\kappa_3, \kappa_2\to\kappa_4)\right\}, \tag{5}$$

where $f(\kappa,t) = \kappa^{-1}\exp(\kappa^2 t)\mathrm{erfc}(-\kappa\sqrt{t})$. From here, the MSD is readily found by the integration of $V_{xy}(t)$ from 0 to t,

$$\xi_{xy}(t) = \frac{2k_\mathrm{B}T}{M}\left\{\left(\frac{t}{\kappa_1\kappa_2} + 2\sqrt{\frac{t}{\pi}}\frac{\kappa_1+\kappa_2}{(\kappa_1\kappa_2)^2} + \frac{\kappa_1^2+\kappa_1\kappa_2+\kappa_2^2}{(\kappa_1\kappa_2)^3} + \frac{1}{\kappa_2-\kappa_1} \times\left[\frac{1}{\kappa_2^3}\exp\left(\kappa_2^2 t\right)\mathrm{erfc}\left(-\kappa_2\sqrt{t}\right) - \frac{1}{\kappa_1^3}\exp\left(\kappa_1^2 t\right)\mathrm{erfc}\left(-\kappa_1\sqrt{t}\right)\right]\right) + (\kappa_1\to\kappa_3, \kappa_2\to\kappa_4)\right\}. \tag{6}$$

In the absence of the field $\omega_\mathrm{c} = 0$, $\kappa_1 = \kappa_3 = \lambda_1$, and $\kappa_2 = \kappa_4 = \lambda_2$, so that we obtain $\xi_{xy} = \xi_x + \xi_y$, where $\xi_x = \xi_y = \xi_z$ exactly correspond to the known result [9] found by a much more complicated method, and to the earlier little known solution [7]

$$V_z(t) = \frac{2k_\mathrm{B}T}{M}\frac{1}{\lambda_2-\lambda_1}\left[\frac{1}{\lambda_2}\exp\left(\lambda_2^2 t\right)\mathrm{erfc}\left(-\lambda_2\sqrt{t}\right) - \frac{1}{\lambda_1}\exp\left(\lambda_1^2 t\right)\mathrm{erfc}\left(-\lambda_1\sqrt{t}\right)\right]. \tag{7}$$

The results from [10] are different due to the difference in the roots $\lambda_{1,2}$. The time-dependent diffusion coefficient is $D(t) = V(t)/2$ and the VAF $\Phi(t) = \dot{D}(t)$. The latter function is expressed by Eq. (7), if $V_z(t)$ is divided by 2 and $1/\lambda_{1,2}$ in $[\ldots]$ replaced by $\lambda_{1,2}$. The VAF decay is not exponential as in the usual BM: at $t\to\infty$ the VAF contains a tail $\sim t^{-3/2}$. Algebraic tails are present also in the solution (6). Up to the longest-lived tail Eq. (6) gives

$$\xi_{xy}(t) \approx \frac{4k_\mathrm{B}T}{M}\frac{\tau t}{1+(\tau/\tau_\mathrm{c})^2} \times\left(1 - 2\sqrt{\frac{\tau_\mathrm{R}}{\pi t}}\frac{1-(\tau/\tau_\mathrm{c})^2}{1+(\tau/\tau_\mathrm{c})^2} + \ldots\right), \quad t\to\infty. \tag{8}$$

Even the Einstein limit $\sim t$ differs from the previous result [10] (the agreement is only when $M = m$ (or at $\omega_\mathrm{c} = 0$)). At short times the MSD is independent of B, $\xi_x(t) = \xi_y(t) \approx (k_\mathrm{B}T/M)t^2$ as $t\to 0$. An apparent contradiction with the equipartition theorem (one would expect m in the denominator instead of $M = m + m_\mathrm{s}/2$) is just a consequence of the assumed incompressibility of the solvent, due to which the limit $t\to 0$ cannot be really accomplished. The correct result is achieved when the compressibility of the solvent is taken into account. Also this task can be straightforwardly solved by the presented method.

3. Conclusions

In conclusion, we have solved the problem of the hydrodynamic BM of a charged particle in a fluid under influence of a constant magnetic field. The obtained solution is exact for incompressible fluids described by the nonstationary Navier–Stokes equations. The found MSD along the field corresponds to the known result in the absence of the external field. Across the field the time CFs of the

BP are affected by the field and significantly differ from the results on the basis of the traditional Langevin equation of motion. In particular, the memory in the fluid and particle motion manifests itself in algebraic decay of the CFs that characterizes the transition from ballistic to the Einstein diffusion. Although the detection of this long-time tail effect requires high spatial and temporal resolution, the current experimental possibilities make it possible. The presented theory could be verified in similar experiments as in [11], where the hydrodynamic theory of the BM of a free particle has been definitely confirmed.

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