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Fluctuating Hydrodynamics and Brownian Motion. II. Note on the Slip Boundary Condition

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Previous work by Hauge and Martin-Löf discussing the generalized Langevin equation for Brownian motion as a contraction from the more fundamental but still phenomenological description of a particle immersed in an incompressible fluid governed by fluctuating hydrodynamics with stick boundary condition is extended to the case of a Brownian particle with arbitrary shape and with *slip* boundary condition. The motivation for this extension is the fact that the latter condition naturally arises in a treatment of the problem from first principles.

KEY WORDS: Fluctuating hydrodynamics; boundary conditions; Brownian motion; generalized Langevin equation; fluctuation-dissipation theorem.

In a recent paper[1] (referred to as I below) Hauge and Martin-Löf discussed the contraction from the Markovian description of a Brownian particle (B) immersed in an incompressible fluid governed by fluctuating hydrodynamics, to the (in general) non-Markovian description in terms of a generalized Langevin equation [see I, Eq. (59)] for the dynamic variables of B alone. They

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proved: (i) The (time-dependent) friction tensor is symmetric for arbitrary shape of B. (ii) The (nonwhite) spectrum of the noise and the friction tensor are related by the appropriate fluctuation-dissipation theorem. These statements were proved strictly within the chosen phenomenological framework, and mechanical time reversibility was not invoked.[3]

On the other hand, the very restriction to macroscopic reasoning emphasizes the need for a derivation from first principles. The importance of a microscopic justification is, in particular, due to the fact that the phenomenological theory shows the ratio ρ/ρ_B (mass density of the fluid/mass per unit volume of B) to be crucial in determining whether the Langevin equation can be considered as Markovian to a sufficient approximation. This basic parameter has not emerged as such from previous work on the microscopic derivation of the Langevin equation.

Before one embarks on such a program, however, one minor point must be settled. In the proofs of I the stick boundary condition was used, i.e., both normal and tangential components of the fluid velocity field were assumed to coincide with the velocity of B at the surface. In a microscopic theory, however, one would like to consider the simplest case and describe the interaction between B and a fluid particle by a potential energy function $\Phi(\mathbf{r} - \mathbf{R})$ corresponding to a smooth Brownian particle of reasonable shape. But in any collision governed by Φ no tangential forces will appear. The macroscopic manifestation of this simple fact is the so-called slip condition: Tangential stresses vanish at the surface of B. It is the purpose of the present note to point out that, with slight modifications, the arguments in I carry over to the case of slip boundary condition.

The boundary condition is used explicitly in I for a single purpose only [see I, Eqs. (60)–(62) and (B.2)], namely the proof of the relation

$$-\int dS\, \mathbf{u}(t)\cdot\boldsymbol{\sigma}(t')\cdot\mathbf{n} = \mathbf{U}(t)\cdot\mathbf{F}(t') + \boldsymbol{\Omega}(t)\cdot\mathbf{M}(t') \tag{1}$$

Here $\mathbf{u}(\mathbf{x}, t)$ and $\boldsymbol{\sigma}(\mathbf{x}, t)$ are the velocity and stress tensor fields of the fluid, $\mathbf{n}(\mathbf{x})$ is a unit vector normal to, and pointing into, the surface of B at $\mathbf{x}$. The integration extends over the surface of B. The dynamic variables of B are the translational and angular velocities $\mathbf{U}$ and $\boldsymbol{\Omega}$, and the force $\mathbf{F}$ and the torque $\mathbf{M}$ exerted on B by the fluid are given as

$$\mathbf{F} = -\int dS\, \boldsymbol{\sigma}\cdot\mathbf{n} \tag{2}$$

$$\mathbf{M} = -\int dS\, \mathbf{x} \times \boldsymbol{\sigma}\cdot\mathbf{n} \tag{3}$$

[3] The two levels of reasoning are often mixed; see, e.g., Ref. 2.

The stick boundary condition reads

$$\mathbf{u}(\mathbf{x}, t) = \mathbf{U}(t) + \mathbf{\Omega}(t) \times \mathbf{x} \qquad \text{for} \quad \mathbf{x} \in S \tag{4}$$

and the proof of (1) follows immediately by insertion of (4) into the left-hand side and appeal to (2) and (3).

The slip condition, on the other hand, reads

$$\left.\begin{aligned} \mathbf{u}(\mathbf{x}, t)\cdot\mathbf{n}(\mathbf{x}) &= [\mathbf{U}(t) + \mathbf{\Omega}(t) \times \mathbf{x}]\cdot\mathbf{n}(\mathbf{x}) \quad (5)\\ \sigma_{n1} = \sigma_{n2} &= 0 \quad (6) \end{aligned}\right\} \text{for} \quad \mathbf{x} \in S$$

The normal component of (4) is a boundary condition needed already for nonviscous fluids and is of course also valid with slip. The indices in σ_{n1} and σ_{n2} refer to a local Cartesian system where n denotes the direction parallel to $\mathbf{n}$ and 1 and 2 stand for orthogonal tangential directions.

We note that with slip $\boldsymbol{\sigma}\cdot\mathbf{n} = \sigma_{nn}\mathbf{n}$ ($\boldsymbol{\sigma}$ is symmetric), and thus only the normal component $\mathbf{u}\cdot\mathbf{n}$ is needed on the left of (1). Insertion of (5) then yields $\mathbf{U}$ and $\mathbf{\Omega}$ dotted into integrals that are nothing else than the definitions (2) and (3) with $\boldsymbol{\sigma}\cdot\mathbf{n} = \sigma_{nn}\mathbf{n}$. Thus Eq. (1) is proved to be valid for the case of slip as well as stick.

The remaining arguments of I can be taken over verbatim and one has thus proved, on the basis of fluctuating hydrodynamics, the symmetry of the friction tensor appearing in the generalized Langevin equation [I, Eq. (59)] for $(\mathbf{U}, \mathbf{\Omega})$, and the fluctuation-dissipation theorem relating the autocorrelation of the random forces to this tensor.

It is curious to note that for the usual "linear combination" of stick and slip

$$u_i - [\mathbf{U} + \mathbf{\Omega} \times \mathbf{x}]_i = \beta\, \sigma_{ni}$$

where i denotes a tangential direction and β is a constant, a proof along the lines presented here does not seem to work. The fact that it *does* work for stick ($\beta \to 0$) and slip ($\beta \to \infty$) appears to be a result of accidental features of these limiting cases.

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