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From Lagrangian to Brownian Motion

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We present a Lagrangian describing an idealized liquid interacting with a particle immersed in it. We show that the equation describing the motion of the particle as a functional of the initial conditions of the liquid incorporates noise and friction, which are attributed to specific dynamical processes. The equation is approximated to yield a Langevin equation with parameters depending on the Lagrangian and the temperature of the liquid. The origin of irreversibility and dissipation is discussed.

KEY WORDS: Brownian motion; Langevin equation; dynamical processes.

1. INTRODUCTION

Dynamical problems are becoming a growing focus of attention in condensed matter physics. A partial list of subjects includes: critical dynamics, dynamics of random systems, entanglement of polimers, and $1/f$ noise. The starting point of many of the investigations in these areas is the phenomenological Langevin equation. The problem of deriving the Langevin equation from basic principles and understanding the approximations involved is thus of great importance.

Attempts to obtain Brownian motion from a mechanical description date back to the 1940s.[(1)] Some of the methods used over the years are phenomenological or qualitative in nature,[(2)] while other are very general and use powerful mathematical tools.[(3–5)] The most important result obtained by the more sophisticated treatments is the generalized fluctuation-dissipation theorem. Two difficulties are encountered in those

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methods: it seems that they rely on the assumption of thermal equilibrium,[6,7] and the Langevin parameters are not calculated explicitly in terms of the mechanical parameters.

A method that avoids those difficulties and at the same time suggests more insight into the problem was introduced by Feynman and Vernon.[8] They couple the Brownian particle linearly to a system of harmonic oscillators, thus obtaining an exactly soluble model. This method has been widely applied to many problems involving quantum dissipation.[9–13] The single-particle behaviour is shown to be affected by effective friction and noise, which are calculated as functions of the mechanical parameters.

In spite of the success of this model, one can question whether it is realistic. What may be unrealistic about linear Lagrangians? Clearly, the medium itself is never linear, but we know cases (e.g., solids at low temperatures, quantum liquids) where a linear mechanical description is very good. The difficulty lies in the linear coupling between the Brownian particle and the degrees of freedom of the medium. A simple realistic coupling is a two-body potential between the Brownian particle and the particle of the host medium. However, if the Brownian particle is not bound to a given site of the host medium *on a microscopic scale*, there is no way to expand any realistic interaction to low orders in the coordinates of the Brownian particle. The situation was illuminated by Dekker,[13] who describes a mechanical system that has linear equations of motion. The difference between such a system and a medium–particle system is evident.

The purpose of the present paper is to present a model that lies between the two extreme approaches and is more realistic than the linear models and more tractable than the full many-body system. To this aim we describe the medium as a linear system of phonons, while the interaction between the medium and the particle is a realistic short-range interaction. The interaction is linear in the density fluctuations (which for solid and liquid media correspond to the coordinates of the longitudinal phonons), but is nonlinear in the coordinates of the particle.

We study the classical equations of motion and show that the particle is affected by damping and noise. At this stage no approximation, apart from the assumption that the system is infinite, is made. Next we approximate and average over the initial conditions of the medium to obtain the simple structure of the usual Langevin equation as well as the temperature-dependent friction coefficient and noise. We finally show that the particle thermalizes. Namely, after a long time its average energy is $\frac{3}{2}k_B T$, where T is the temperature characterizing the medium *initially*.

The paper is organized as follows. In Section 2 we introduce the Lagrangian that describes the physical system we consider. In Section 3 we calculate the corresponding Euler–Lagrange equations. The exact integrodifferential equation for the Brownian particle is studied in Section 4. In Section 5 we discuss some aspects concerning the Langevin equation and the relation between the exact equation for the particle and the Langevin equation. The friction coefficient is calculated in terms of the mechanical parameters in Section 6 and the noise is calculated in Section 7. Section 8 contains a summary of results.

2. THE LIQUID–PARTICLE LAGRANGIAN

The liquid is described by the local current density $\mathbf{J}(\mathbf{r})$ and the local number density $\rho(\mathbf{r})$. Let m be the mass of a liquid particle; then we can write a kinetic term for the liquid

$$K_{\text{liq}} = \frac{1}{2} m \int \frac{\mathbf{J}^2(\mathbf{r})}{\rho(\mathbf{r})} \, d\mathbf{r} \tag{1}$$

The potential energy term for the liquid is

$$V_{\text{liq}} = \int \rho(\mathbf{r}) \, V(\mathbf{r} - \mathbf{r}') \, \rho(\mathbf{r}') \, d\mathbf{r} \, d\mathbf{r}' \tag{2}$$

where V is the two-body (effective) potential between the liquid particles.

The typical interaction between the liquid particles is a short-range interaction. We assume that the range of this interaction is much shorter than any other length scale in the system we are considering. For simplicity, we therefore replace $V(\mathbf{r} - \mathbf{r}')$ by a δ-function interaction, so that

$$V_{\text{liq}} = \frac{1}{2} \lambda \int \rho^2(\mathbf{r}) \, d\mathbf{r} \tag{3}$$

Using any other short-range potential does not change the final results qualitatively.

In order to write a Lagrangian for the liquid, we have to keep in mind that $\mathbf{J}$ and ρ are not independent variables, but obey the equation of continuity

$$\nabla \cdot \mathbf{J} + \frac{\partial \rho}{\partial t} = 0 \tag{4}$$

In order to ensure Eq. (4), we introduce a local Lagrange multiplier $\mu(\mathbf{r})$. The Lagrangian for the liquid is therefore

$$L_{\text{liq}} = \int d\mathbf{r} \left[\frac{1}{2} m \frac{\mathbf{J}^2(\mathbf{r})}{\rho(\mathbf{r})} - \frac{1}{2} \lambda \rho^2(\mathbf{r}) - \mu(\mathbf{r}) \left(\nabla \cdot \mathbf{J} + \frac{\partial \rho}{\partial t} \right) \right] \tag{5}$$

Later we shall see that μ is the potential for the velocity field in the liquid; thus, the model we are considering for the liquid allows only irrotational flows.

The Lagrangian (5) can be simplified by assuming small fluctuations in the density, that is, assuming that $(\rho - \bar{\rho})/\bar{\rho} \ll 1$, where $\bar{\rho}$ is the average density. Expanding the kinetic energy term in the small parameter and taking the leading term, we obtain

$$L_{\text{liq}} = \int d\mathbf{r} \left[\frac{1}{2} m \frac{\mathbf{J}^2(\mathbf{r})}{\bar{\rho}} - \frac{1}{2} \lambda \rho^2(\mathbf{r}) - \mu(\mathbf{r}) \left(\nabla \cdot \mathbf{J} + \frac{\partial \rho}{\partial t} \right) \right] \tag{6}$$

Consider now an external particle of mass M immersed in the liquid. The interaction between the liquid and the particle immersed in it is

$$V_{\text{int}} = \int \rho(\mathbf{r})\, U(\mathbf{r} - \mathbf{x})\, d\mathbf{r} \tag{7}$$

where $\mathbf{x}$ is the coordinate of the particle and U is the potential between a liquid particle and the external particle. The kinetic energy term for the external particle is simply $\frac{1}{2} M \dot{\mathbf{x}}^2$. The Lagrangian describing the liquid–particle system is therefore

$$L = \int d\mathbf{r} \left[\frac{1}{2} m \frac{\mathbf{J}^2(\mathbf{r})}{\bar{\rho}} - \frac{1}{2} \lambda \rho^2(\mathbf{r}) - \mu(\mathbf{r}) \left(\nabla \cdot \mathbf{J} + \frac{\partial \rho}{\partial t} \right) - \rho(\mathbf{r})\, U(\mathbf{r} - \mathbf{x}) \right] + \frac{1}{2} M \dot{\mathbf{x}}^2 \tag{8}$$

Clearly the liquid part of the Lagrangian is simplified and lacks some of the attributes of a real liquid, such as nonlinear interactions and interactions leading to vorticity. It does retain, however, the longitudinal waves as excitations. These are the only excitations that are important at low temperatures. The terms we have neglected correspond to interactions among those excitations. For details concerning such low-temperature fluids see Landau.[14]

A similar Lagrangian is obtained in the study of a harmonic solid interacting with an external particle (again the harmonic solid is a good approximation for a real solid in the low-temperature regime). The Lagrangian in this case is

$$L_S = \int \frac{1}{2} d\mathbf{q} [m(\dot{\mathbf{u}}_{\mathbf{q}} \cdot \dot{\mathbf{u}}_{-\mathbf{q}} - \omega_q^2 \mathbf{u}_{\mathbf{q}} \cdot \mathbf{u}_{-\mathbf{q}}) - i\mathbf{q} \cdot \mathbf{u}_{-\mathbf{q}} v(q) \exp(i\mathbf{q} \cdot \mathbf{x})] + \frac{1}{2} M \dot{\mathbf{x}}^2 \tag{9}$$

where m is the mass of the atom of the solid, $\mathbf{u}_q$ is the Fourier transform of the displacement field, ω_q is the corresponding frequency, and $v(q)$ is the Fourier transform of the interaction potential between the external particle and a solid atom.

Note that the transverse degrees of freedom are not coupled to the particle. The Lagrangian describing the longitudinal waves in the solid and the external particle is the same as L_{liq}. We continue the calculation with L_{liq}. The results for the harmonic solid follow in a straightforward manner.

3. EULER–LAGRANGE EQUATIONS

Consider first the equations for the liquid in the absence of the external particle,

$$\frac{m}{\bar{\rho}}\mathbf{J}(\mathbf{r}) + \nabla\mu(\mathbf{r}) = 0 \tag{10}$$

$$-\lambda\rho(\mathbf{r}) + \frac{\partial\mu(\mathbf{r})}{\partial t} = 0 \tag{11}$$

$$\nabla\cdot\mathbf{J}(\mathbf{r}) + \frac{\partial\rho(\mathbf{r})}{\partial t} = 0 \tag{12}$$

Substituting Eqs. (10) and (11) in Eq. (12), we obtain

$$\nabla^2\mu - \frac{m}{\bar{\rho}\lambda}\frac{\partial^2\mu}{\partial t^2} = 0 \tag{13}$$

Equation (13) is a homogeneous wave equation. The velocity of sound is given by

$$c = (\lambda\bar{\rho}/m)^{1/2} \tag{14}$$

Clearly, by Eqs. (10) and (11), $\mathbf{J}$ and ρ obey the same wave equation.

Adding the external particle, we obtain the equations

$$M\ddot{\mathbf{x}} + \int \rho(\mathbf{r})\,\nabla_x U(\mathbf{r}-\mathbf{x})\,d\mathbf{r} = 0 \tag{15}$$

and

$$\nabla^2\mu - \frac{1}{c^2}\frac{\partial^2\mu}{\partial t^2} = -\frac{1}{c^2}\frac{\partial}{\partial t}U(\mathbf{r}-\mathbf{x}) \tag{16}$$

Using Eqs. (10) and (11), we obtain

$$\nabla^2\rho - \frac{1}{c^2}\frac{\partial^2\rho}{\partial t^2} = -\frac{1}{\lambda}\nabla_r^2 U(\mathbf{r}-\mathbf{x}) \tag{17}$$

$$\nabla^2\mathbf{J} - \frac{1}{c^2}\frac{\partial^2\mathbf{J}}{\partial t^2} = \frac{1}{\lambda}\frac{\partial}{\partial t}\nabla_r U(\mathbf{r}-\mathbf{x}) \tag{18}$$

We see that the external particle acts as a source to the various wave equations.

We emphasize that the equations of motion (15), (17) do not contain any dissipative mechanism put in by hand. They are the exact equations obeyed by the system described by the Lagrangian. Heat modes were not introduced in the description of the liquid because one of our aims in this paper is to obtain dissipative motion for the particle from a purely mechanical Lagrangian. Introduction of heat modes certainly gives a better description of the liquid at higher temperatures. However, it might have confused the issue by associating the resulting dissipative force acting on the particle with the dissipation in the liquid.

4. GENERIC LANGEVIN EQUATION FOR THE EXTERNAL PARTICLE

The force term in a generic Langevin equation for an external particle immersed in a liquid breaks up into two parts. One part depends on the trajectory of the particle and includes the dissipative force and the regular force [e.g., $-\gamma\dot{\mathbf{x}} + \mathbf{F}(\mathbf{x})$]. The second part is a noise term that averages to zero and does not depend on the trajectory of the particle. In this section we show that the force acting on the particle we considered in the previous section is similar to that of a generic Langevin equation.

The force acting on the particle is given by Eq. (15),

$$\mathbf{F} = \int \rho(\mathbf{r})\,\nabla_r U(\mathbf{r}-\mathbf{x}) \tag{19}$$

The force depends on the trajectory $\mathbf{x}(t)$ through Eq. (17).

Suppose that the particle is immersed at $t=0$ and suppose that at that time the liquid is totally at rest ($\rho=\bar{\rho}$, $\partial\rho/\partial t=0$). Let the solution of the equation

$$\nabla^2\rho - \frac{1}{c^2}\frac{\partial^2\rho}{\partial t^2} = -\frac{1}{\lambda}\nabla_r^2 U(\mathbf{r}-\mathbf{x})\,\Theta(t) \tag{20}$$

with calm initial conditions be $\rho_0(\mathbf{r}, t)$. The solution for general initial conditions is then

$$\rho(\mathbf{r}, t) = \rho_0(\mathbf{r}, t) + \rho_f(\mathbf{r}, t) \tag{21}$$

where $\rho_f(\mathbf{r}, t)$ is a solution of the free (homogeneous) wave equation with the same initial conditions, except that $\bar{\rho} = 0$ at $t = 0$.

Clearly, ρ_0 is a functional of $\mathbf{x}(t)$, while ρ_f knows nothing about the particle. The force acting on the particle is therefore

$$\mathbf{F} = \int \rho_0(\mathbf{r}, t)\, \nabla_r U(\mathbf{r} - \mathbf{x})\, d\mathbf{r} + \int \rho_f(\mathbf{r}, t)\, \nabla_r U(\mathbf{r} - \mathbf{x})\, d\mathbf{r} \tag{22}$$

We calculate next the explicit form of the first term in Eq. (22) as a functional of the particle trajectory $\mathbf{x}(t)$. We express Eq. (20) in the form

$$\nabla^2 \rho - \frac{1}{c^2} \frac{\partial^2 \rho}{\partial t^2} = -\frac{1}{\lambda} \int \nabla_r^2 U(\mathbf{r} - \mathbf{x}(\tau))\, \Theta(\tau) \delta(\mathbf{r} - \mathbf{r}') \delta(t - \tau)\, d\mathbf{r}'\, d\tau \tag{23}$$

A particular solution of Eq. (23) is

$$\tilde{\rho} = \frac{1}{4\pi\lambda} \int d\mathbf{r}' \frac{1}{|\mathbf{r} - \mathbf{r}'|} \nabla_r^2 U\left(\mathbf{r}' - \mathbf{x}\left(t - \frac{|\mathbf{r} - \mathbf{r}'|}{c}\right)\right) \times \Theta\left(t - \frac{|\mathbf{r} - \mathbf{r}'|}{c}\right) \tag{24}$$

Since $\tilde{\rho} = 0$ for $t < 0$, the particular solution may be identified with $\rho_0 - \bar{\rho}$ (because it solves the same equation with the same initial conditions).

The force the particle exerts on itself via the liquid is therefore

$$\begin{aligned} \mathbf{F}_p &\equiv \int \rho_0(\mathbf{r}, t)\, \nabla_r U(\mathbf{r} - \mathbf{x})\, d\mathbf{r} \\ &= \frac{1}{4\pi\lambda} \int \nabla U(\mathscr{R}) \frac{1}{R} \nabla^2 U\left(\mathbf{R} + \mathscr{R} + \mathbf{x}(t) - \mathbf{x}\left(t - \frac{R}{c}\right)\right) \\ &\quad \times \Theta\left(t - \frac{R}{c}\right) d\mathbf{R}\, d\mathscr{R} \end{aligned} \tag{25}$$

where $\mathscr{R} = \mathbf{r} - \mathbf{x}(t)$ and $\mathbf{R} = \mathbf{r} - \mathbf{r}'$. The equation of motion for the particle in the liquid is

$$M\ddot{\mathbf{x}} = \mathbf{F}_p + \int \rho_f(\mathbf{r}, t)\, \nabla_r U(\mathbf{r} - \mathbf{x})\, d\mathbf{r} \equiv \mathbf{F}_{\text{tot}} \tag{26}$$

At this point we digress to discuss irreversibility and dissipation.

Equation (26) will serve as an example for the discussion. The equation of motion for the particle, Eq. (26), is *not* reversible. Namely, if at a given time we reverse the direction of the velocity of the particle, it will not track back along its path in time. The original Euler–Lagrange equations (15) and (17) are obviously reversible. The original equations, however, are reversible under a reversal of the velocities corresponding to all the degrees of freedom. Therefore, in order to reverse the motion of the particle in Eq. (26), a proper modification of the medium has to be made in addition to reversing the particle's velocity.

The irreversibility of the motion of the particle is therefore just a consequence of considering the particle (or, in general, a subsystem) and ignoring the rest of the system. Irreversibility has nothing to do with any averaging process or with the whole system being infinite. In fact, the same would be true for two interacting particles if only the velocity of one of them is reversed.

The fact that the system is infinite is important, however, to dissipation. Note that we distinguish between irreversibility and dissipation. Obviously, every dissipative system is irreversible, but not any irreversible system is dissipative. As can be seen from the particular solution for the density, Eq. (24), the medium is affected by the particle through the creation of outgoing waves radiated by the particle. Clearly, by such a procedure the particle can only lose energy to the medium, and this is the origin of dissipation.

The fact that only outgoing waves are involved relies heavily on the system being infinite. In a finite system, the Green's function of the wave equation also must include reflections from the walls. As a result, a particle in the middle of a container of size L will experience dissipation for times $\tau < L/c$, while at larger times the particle may be accelerated by waves reflected from the walls.

The origin of dissipation is radiation of phonons (for any trajectory of the particle and initial conditions of the liquid!); $\mathbf{F}_p$ in Eq. (26) is therefore a dissipative force.

The force exerted on the particle due to ρ_f is uncorrelated with the trajectory of the particle, since ρ_f depends solely on the initial conditions of the liquid. That force, for given initial conditions of the liquid and the particle, may be viewed as a realization of a random force. Its statistical properties will be discussed in subsequent sections.

So far we have shown that the equation of motion of the particle resembles a generic Langevin equation in many respects. In the next sections we tackle the problem of transforming Eq. (26) into the familiar Langevin equation and discuss the approximations involved in that process.

5. LANGEVIN EQUATION

The equation of motion for the particle, Eq. (26), gives the force acting on the particle in terms of the initial conditions of the liquid. It can therefore yield the full trajectory of the particle as a functional of the initial conditions of the liquid and the particle. Equation (26) is an exact, non-reversible equation, describing a particle affected by a fluctuating noise and by dissipation. *No averaging process* was used in order to obtain Eq. (26).

We will show that, under certain conditions, Eq. (26) can be transformed into the phenomenological Langevin equation. This step involves an averaging over the initial conditions of the liquid. No averaging over the initial conditions of the particle is performed, nor is any assumption of equilibration made. We do assume, however, that at time $t=0$ the liquid alone is in equilibrium.[4]

In order to go over to a stochastic description of the system, we will be interested in the statistical properties of representative particle trajectories rather than in the detailed study of each single trajectory. We assume that the statistical properties of the trajectories of the particle are determined by a small number of macroscopic parameters of the liquid. These parameters describe the state of the liquid at the time $t=0$, before the immersion of the particle. The average and correlations of the force that determines the statistical properties of the particle trajectories may be thus obtained by ensemble averaging over the initial conditions of the liquid. In what follows we assume that the state of the liquid at $t=0$ is described by a Boltzmann distribution corresponding to a temperature T. The Boltzmann distribution is determined by the liquid Hamiltonian, which is a functional of the denity fluctuations and their conjugate momenta. We do not assume that equilibrium is restored after the immersion of the particle.

We would like to compare the stochastic description of our system to that given by the Langevin equation. Recall that the one-dimensional Langevin equation has the form

$$M\ddot{x} + \gamma\dot{x} + F_R(t) = 0 \tag{27}$$

where $F_R(t)$ is a random force,

$$\langle F_R(t)\rangle_R = 0, \qquad \langle F_R(t)\, F_R(t')\rangle_R = \sigma\delta(t-t') \tag{28}$$

[4] It should be realized that a linear system such as the liquid we are considering can reach equilibrium only through interaction with some external system. We assume that that interaction is turned off before the immersion of the particle.

The averages in (28) are with respect to the random force distribution. The solution of Eq. (27) is

$$\dot{x}(t) = \dot{x}(t=0)\, e^{-(\gamma/M)t} + e^{-(\gamma/M)t} \int_0^t dt'\, e^{(\gamma/M)t'} \frac{F(t')}{M} \tag{29}$$

$$\langle \dot{x}(t) \rangle = \dot{x}(t=0)\, e^{-(\gamma/M)t} \tag{30}$$

$$\langle \dot{x}^2 \rangle = \langle \dot{x} \rangle^2 + \frac{\sigma}{2\gamma M} (1 - e^{-(2\gamma/M)t}) \tag{31}$$

Define $v_{\text{th}}^2 \equiv \sigma/2\gamma M$. The long-time average of $\dot{x}^2$ is therefore v_{th}^2.

In order to compare our equation to the Langevin equation, we take the ensemble average of Eq. (26),

$$\langle M\ddot{\mathbf{x}} \rangle = \langle \mathbf{F}_{\text{tot}} \rangle \tag{32}$$

We compare Eq. (32) to the average over random force distribution of Eq. (27)

$$\langle M\ddot{\mathbf{x}} \rangle_R = -\gamma \langle \dot{\mathbf{x}} \rangle_R \tag{33}$$

Identifying the ensemble average with the average over random force distribution, we conclude that the friction coefficient γ is given by the velocity and time-independent part of the coefficient of $\langle \dot{\mathbf{x}} \rangle$ in $\langle \mathbf{F}_{\text{tot}} \rangle$.

How can we determine the exact noise term and its statistical properties? We can always write $\mathbf{F}_{\text{tot}} = \mathbf{F}_{\text{tot}} - \gamma\dot{\mathbf{x}} + \gamma\dot{\mathbf{x}}$. Equation (26) therefore can be written as $M\ddot{\mathbf{x}} = -\gamma\dot{\mathbf{x}} + (\mathbf{F}_{\text{tot}} + \gamma\dot{\mathbf{x}})$. If Eq. (26) indeed corresponds to a Langevin equation, the noise must be related to $\mathbf{F}_{\text{tot}} + \gamma\dot{\mathbf{x}}$. We have to prove that the noise force has the properties of a random force in the Langevin equation. The statistical distribution of the noise is determined by studying correlation functions of the type

$$\begin{aligned} \langle [\mathbf{F}_{\text{tot}}(\mathbf{t}) + \gamma\dot{\mathbf{x}}(\mathbf{t}) - \langle \mathbf{F}_{\text{tot}}(t) + \gamma\dot{\mathbf{x}}(t) \rangle] \\ \times [\mathbf{F}_{\text{tot}}(t') + \gamma\dot{\mathbf{x}}(t') - \langle \mathbf{F}_{\text{tot}}(t') + \gamma\dot{\mathbf{x}}(t')] \rangle \end{aligned} \tag{34}$$

6. THE FRICTION COEFFICIENT

We have identified $\mathbf{F}_p$ in the equation of motion for the particle, Eq. (26), as the dissipative force. It is worthwhile, however, to check that this identification is indeed correct. We have to verify that the noise term in Eq. (26) averages to zero, as it should.

The calculation of the average of the noise term is nontrivial, since $\dot{\mathbf{x}}(t)$ is correlated with the density ρ_f at earlier times. Using the representation of the interaction $U(\mathbf{R})$ by a Fourier integral

$$U(\mathbf{R}) = \int d\mathbf{q}\,[\exp(i\mathbf{q}\cdot\mathbf{R})]\,U(\mathbf{q}) \tag{35}$$

and the fact that

$$\mathbf{x}(t) = \mathbf{x}(0) + \int_0^t dt'\,\dot{\mathbf{x}}(t')$$

it is easy to see that the average of the noise term in Eq. (26)

$$\langle \mathbf{F}_{\text{tot}} - \mathbf{F}_p \rangle = \left\langle \int d\mathbf{r}\,\rho_f(\mathbf{r}, t)\,\nabla_r U(\mathbf{r} - \mathbf{x}(t)) \right\rangle \tag{36}$$

may be written in the form $\int d\mathbf{q}\,\mathbf{h}(\mathbf{q}) \exp[-i\mathbf{q}\cdot\mathbf{x}(0)]$, where $\mathbf{h}(\mathbf{q})$ does not depend on $\mathbf{x}(0)$.

However, the final result of the average in Eq. (36) is translation-invariant and therefore cannot depend on $\mathbf{x}(0)$. The final conclusion is therefore that $\mathbf{h}(\mathbf{q})$ must vanish. The above argument proves that the only contribution to the friction coefficient comes from $\langle \mathbf{F}_p \rangle$.

In order to calculate the friction coefficient, we consider a class of liquid–particle interactions $U(\mathbf{R})$ that are spherically symmetric and short-ranged. For simplicity we consider interactions that vanish for $R > R_0$. As explained in Appendix A, the theta function in $\mathbf{F}_p$ can be dropped for this class of interactions a long time after the immersion of the particle.

The force $\mathbf{F}_p$ is therefore given by

$$\mathbf{F}_p = \frac{1}{4\pi\lambda} \int \nabla U(\mathscr{R}) \frac{1}{R} \nabla^2 U\left[\mathbf{R} + \mathscr{R} + \mathbf{x}(t) - \mathbf{x}\left(t - \frac{R}{c}\right)\right] d\mathbf{R}\,d\mathscr{R} \tag{37}$$

By the mean value theorem

$$\mathbf{x}(t) - \mathbf{x}\left(t - \frac{R}{c}\right) = \dot{\mathbf{x}}(t') \frac{R}{c} \tag{38}$$

where $t - R/c < t' < t$.

We now separate the velocity of the particle into an average part and a fluctuating part

$$\dot{\mathbf{x}}(t) = \langle \dot{\mathbf{x}}(t) \rangle + \delta\dot{\mathbf{x}}(t) \tag{39}$$

Equation (39) defines $\delta\dot{\mathbf{x}}(t)$.

We may assume that $\langle \dot{\mathbf{x}}(t) \rangle$ is a slowly varying function of time and that it is small in magnitude. A precise quantitative meaning to this statement will be given later. Note that we assume that the average velocity is small, and not that the instantaneous velocity is small. It is clear that the instantaneous velocity might have a large magnitude, since the particle interacts continuously with the liquid particles, which may have large velocities (although the probability for that is small). In addition, it would be inconsistent with any stochastic description to assume that the instantaneous velocity of the particle is small. According to any stochastic description, there is always a certain probability, though it may be small, that the velocity of the particle is large.

Using Eqs. (38) and (39), we can express $\mathbf{F}_p$ as follows:

$$\mathbf{F}_p = \frac{1}{4\pi\lambda} \int \nabla U(\mathscr{R}) \frac{1}{R} \nabla^2 U \left[\mathbf{R} + \mathscr{R} + \delta\dot{\mathbf{x}}(t') \frac{R}{c} + \langle \dot{\mathbf{x}}(t') \rangle \frac{R}{c} \right] d\mathbf{R}\, d\mathscr{R} \tag{40}$$

Expanding $\mathbf{F}_p$ in powers of $\mathbf{\Delta} = \langle \dot{\mathbf{x}}(t') \rangle R/c$, we obtain

$$\begin{aligned}
\langle \mathbf{F}_p \rangle = {} & \frac{1}{4\pi\lambda} \int \nabla U(\mathscr{R}) \frac{1}{R} \nabla^2 \left\langle U \left[\mathbf{R} + \mathscr{R} + \delta\dot{\mathbf{x}}(t') \frac{R}{c} \right] \right\rangle d\mathbf{R}\, d\mathscr{R} \\
& + \frac{1}{4\pi\lambda} \int \nabla U(\mathscr{R}) \frac{1}{R} \nabla\nabla^2 \left\langle U \left[\mathbf{R} + \mathscr{R} + \delta\dot{\mathbf{x}}(t') \frac{R}{c} \right] \right\rangle \cdot \langle \dot{\mathbf{x}}(t') \rangle \frac{R}{c}\, d\mathbf{R}\, d\mathscr{R} \\
& + \frac{1}{4\pi\lambda} \int \nabla U(\mathscr{R}) \frac{1}{2} \nabla_i \nabla_j \nabla^2 \left\langle U \left[\mathbf{R} + \mathscr{R} + \delta\dot{\mathbf{x}}(t') \frac{R}{c} \right] \right\rangle \\
& \times \langle \dot{x}_i(t') \rangle \langle \dot{x}_j(t') \rangle \frac{R^2}{c^2}\, d\mathbf{R}\, d\mathscr{R} \\
& + \cdots
\end{aligned} \tag{41}$$

where summation over the Cartesian components i and j is understood.

The first term in Eq. (41) vanishes because $U(\mathscr{R})$ is spherically symmetric. Since $\langle \dot{\mathbf{x}}(t') \rangle$ is a slowly varying function of its argument and since the R integration is effectively limited to a range of the order of R_0 (see Appendix A), we expand it in a Taylor series,

$$\langle \dot{\mathbf{x}}(t') \rangle = \langle \dot{\mathbf{x}}(t) \rangle + \langle \ddot{\mathbf{x}}(t) \rangle (t - t') + \cdots \tag{42}$$

Using Eq. (42), we obtain

$$\begin{aligned}
\langle \mathbf{F}_p \rangle = {} & \frac{1}{4\pi\lambda c} \int \nabla U(\mathscr{R})\, \nabla\nabla^2 \left\langle U \left[\mathbf{R} + \mathscr{R} + \delta\dot{\mathbf{x}}(t') \frac{R}{c} \right] \right\rangle \cdot \langle \dot{\mathbf{x}}(t) \rangle\, d\mathbf{R}\, d\mathscr{R} \\
& + \frac{1}{4\pi\lambda c} \int \nabla U(\mathscr{R})\, \nabla\nabla^2 \left\langle U \left[\mathbf{R} + \mathscr{R} + \delta\dot{\mathbf{x}}(t') \frac{R}{c} \right] \right\rangle \cdot \\
& \times \langle \ddot{\mathbf{x}}(t) \rangle (t' - t)\, d\mathbf{R}\, d\mathscr{R} + \cdots + \frac{1}{4\pi\lambda c} \int \nabla U(\mathscr{R}) \frac{1}{2} \nabla_i \nabla_j \nabla^2 \\
& \times \left\langle U \left[\mathbf{R} + \mathscr{R} + \delta\dot{\mathbf{x}}(t') \frac{R}{c} \right] \right\rangle \langle \dot{x}_i(t) \rangle \langle \dot{x}_j(t) \rangle \frac{R}{c}\, d\mathbf{R}\, d\mathscr{R} + \cdots
\end{aligned} \tag{43}$$

keeping in mind that $t - R/c < t' < t$. The dots in Eq. (43) stand for terms containing higher derivatives of $\langle \dot{\mathbf{x}}(t) \rangle$ and higher powers of $\langle \dot{\mathbf{x}}(t) \rangle$ or both.

The first term in Eq. (43) is responsible for the friction in the corresponding Langevin equation.

The term proportional to $\langle \ddot{\mathbf{x}}(t) \rangle$ signifies that the mass appearing in the Langevin equation is not the mass M appearing in the original Lagrangian. The terms proportional to higher derivatives of the average velocity are related to the existence of a nontrivial memory kernel in the corresponding stochastic equation. That is, the stochastic equation that describes the motion of the particle is actually a generalized Langevin equation. However, as long as the average velocity is a slowly varying function of time, these terms are small compared with the term proportional to the average velocity itself.

The third term in Eq. (43) corresponds to a nonlinear modification of the Langevin equation. This term is small compared to the first term as long as $\langle \dot{\mathbf{x}}(t) \rangle / c$ is small.

We point out that if, for some reason, the higher terms in the expansion are not small compared to the first term, then the Langevin equation would not give an adequate description of the system. We discuss such a situation at the end of this section.

The first term in Eq. (43) may be written in the form

$$\langle (\mathbf{F}_p)_i \rangle = A_{ij} \langle \dot{\mathbf{x}}_j(t) \rangle \tag{44}$$

where i, j are Cartesian coordinates and $A_{ij}(t)$ is given by

$$A_{ij}(t) = \frac{1}{4\pi\lambda c} \int \nabla_i U(\mathscr{R}) \nabla_j \nabla^2 \left\langle U\left[\mathbf{R} + \mathscr{R} + \delta\dot{\mathbf{x}}(t')\frac{R}{c}\right]\right\rangle \tag{45}$$

The time dependence of A_{ij} stems from the relation between t and t'.

In order to obtain the friction coefficient, we have to consider the long-time behavior of $A_{ij}(t)$. At $t \to \infty$ the average velocity approaches zero. For such times

$$x\left(t + \frac{R}{c}\right) - x(t) = \delta\dot{\mathbf{x}}(t')\frac{R}{c} \tag{46}$$

In order to calculate $\lim_{t \to \infty} A_{ij}(t)$, we represent $U(\mathscr{R})$ by a Fourier integral as in Eq. (35). Using Eq. (46), we obtain

$$\lim_{t \to \infty} A_{ij} = \lim_{t \to \infty} -\frac{(2\pi)^3}{4\pi\lambda c} \int k_i U(-k)\, k^2 U(k)\, k_j \times \left\langle \exp\left\{ i\mathbf{k} \cdot \left[\mathbf{x}\left(t + \frac{R}{c}\right) - \mathbf{x}(t)\right]\right\}\right\rangle \exp(i\mathbf{k} \cdot \mathbf{R})\, d\mathbf{R}\, d\mathbf{k} \tag{47}$$

In order to calculate the Boltzmann average in Eq. (47), we have to know the difference $\mathbf{x}(t+R/c)-\mathbf{x}(t)$ as a functional of the initial conditions of the liquid. This would involve an explicit solution of the nonlinear integrodifferential equation (26). It might be tempting to treat the noise term as a perturbation and try to obtain such an explicit solution. However, we have to know the solution $\mathbf{x}(t)$ for $t \to \infty$, and therefore the perturbative solution cannot be expected to be very useful. On the other hand, we might expect to profit from such an expansion for the difference $\mathbf{x}(t+R/c)-\mathbf{x}(t)$ when the time $R/c \sim R_0/c$ is small. In order to proceed, we assume that the interaction between the particle and the liquid is weak enough so that only the first term in the perturbation expansion can be kept. The difference $\mathbf{x}(t+R/c)-\mathbf{x}(t)$ is, in this case, a linear functional of $\rho_f(t)$. The density $\rho_f(t)$ depends linearly on the initial conditions of the liquid, which are Gaussian integration variables. We therefore conclude that

$$\begin{aligned} f(k, R) &\equiv \lim_{t \to \infty} \left\langle \exp\left\{i\mathbf{k}\cdot\left[\mathbf{x}\left(t+\frac{R}{c}\right)-\mathbf{x}(t)\right]\right\}\right\rangle \\ &= \exp\left\{-\lim_{t\to\infty}\frac{1}{2}k^2\left\langle\left[x\left(t+\frac{R}{c}\right)-x(t)\right]^2\right\rangle\right\} \end{aligned} \tag{48}$$

where $x(t)$ corresponds to one component of $\mathbf{x}(t)$.

To obtain at this stage an explicit expression for the friction coefficient in terms of the parameters appearing in the Lagrangian, we use the following procedure. We *assume* that Eq. (26) is a Langevin equation of the form of Eq. (27) with unknown coefficients γ and σ. We use the Langevin equation to calculate the average in Eq. (48). We can then check our results and see whether the quantities that we assume are small are indeed small.

We find

$$\left\langle\left[x\left(t+\frac{R}{c}\right)-x(t)\right]^2\right\rangle = 2v_{\text{th}}^2\frac{M}{\gamma}\left[\frac{R}{c}-\frac{M}{\gamma}\left(1-e^{\gamma R/Mc}\right)\right] \tag{49}$$

The function $f(k, R)$ defined in Eq. (48) behaves differently for small and large values of R,

$$f(k, R) = \begin{cases} \exp\left(-\dfrac{1}{2}k^2\dfrac{v_{\text{th}}^2}{c^2}R^2\right), & R < \dfrac{cM}{\gamma} \\ \exp\left(-k^2 v_{\text{th}}^2\dfrac{MR}{\gamma c}\right), & R > \dfrac{cM}{\gamma} \end{cases} \tag{50}$$

Taking into account that the maximal relevant values of k are of the order of $1/R_0$ and denoting the length $v_{\text{th}}(M/\gamma)$ by a, we see that as long as $a/R_0 < 1$ we may use the large-R form of $f(k, R)$ over the whole range of integration in Eq. (47). The condition $v_{\text{th}} M/\gamma R_0 < 1$ is valid if the "size" of the particle is large.

Assuming the above condition holds, we obtain

$$\lim_{t \to \infty} A_{ij}(t) = -\delta_{ij}\gamma = -\delta_{ij} \frac{(2\pi)^3}{3\lambda c} 2 \frac{v_{\text{th}}}{c} a \int U^2(k)\, k^2\, d\mathbf{k} \tag{51}$$

where γ is the friction coefficient. Note that there are corrections to Eq. (51) of the order of $(v_{\text{th}}/c)\, a/R_0$, which are small compared with the term we keep.

In Eq. (51), γ is expressed in terms of the unknown quantity $v_{\text{th}} = \sigma/2\gamma M$. We would like, however, to express γ in terms of the initial temperature of the liquid T, which has not appeared so far in the calculation. For that purpose we need an expression for σ, which we calculate in the next section [Eq. (64)]. The strength of the force–force correlation σ depends on the temperature T through the Boltzmann distribution for the initial conditions of the liquid. Combining Eqs. (51) and (64) yields $v_{\text{th}}^2 = k_B T/2M$ [Eq. (66)]. Using this expression for v_{th}^2, we obtain for γ

$$\gamma = \left[\frac{(2\pi)^3}{\lambda c^2} k_B T \int U^2(k)\, k^2\, d\mathbf{k} \right]^{1/2} \tag{52}$$

Note that as T tends to zero, so does γ. This fact does not imply that at low temperatures the particle does not transfer energy to the liquid. What it does imply is that the nature of the energy transfer is different than what is predicted by the Langevin equation. In the low-temperature regime, the terms neglected in Eq. (43) become important and dominate the dissipation mechanism.

This result is not an artifact of the model we are considering. In fact, evaluation of the average force at $T = 0$ exerted on a particle scattered elastically by a system of hard spheres exhibits the same behavior. The force does not contain a term linear in the velocity of the particle. As we show in Appendix B, the leading term is proportional to the square of the average velocity of the particle.

We are now in a position to evaluate the magnitude of the various terms neglected along the way. We can also clarify the physical conditions under which the approximations that were done can be considered good approximations.

First let us determine the magnitude of the terms containing higher derivatives of $\langle \dot{\mathbf{x}}(t) \rangle$ in Eq. (43). By our assumption,

$$\langle \dot{\mathbf{x}} \rangle = -\frac{M}{\gamma} \langle \ddot{\mathbf{x}} \rangle = \left(\frac{M}{\gamma}\right)^2 \langle \dddot{\mathbf{x}} \rangle = \cdots \tag{53}$$

Each time derivative in the expansion therefore carries with it a factor of the order of $\gamma R_0/Mc$. We conclude that as long as

$$\gamma R_0/Mc < 1 \tag{54}$$

the terms containing higher derivatives of the average velocity of the particle are small compared with the term proportional to the average velocity itself.

The terms containing higher powers of the average velocity are small if

$$\langle \dot{\mathbf{x}} \rangle / c < 1 \tag{55}$$

Recall that we have also encountered the condition

$$\gamma R_0/Mv_{\text{th}} > 1 \tag{56}$$

The conditions on the various velocities in our system are therefore

$$\langle \dot{\mathbf{x}} \rangle < v_{\text{th}} < c \tag{57}$$

Equations (54) and (56) tell us that the diffusive motion of the particle has to dominate the inertial motion. Otherwise, memory effects become important and the assumption that the particle obeys a Langevin equation is no longer valid.

7. THE NOISE

The force–force correlation strength σ in the Langevin equation is related to the velocity- and time-independent part of the correlation in (34), that is,

$$\lim_{\tau \to \infty} \langle \mathbf{N}(\tau) \cdot \mathbf{N}(\tau + t) \rangle$$

where

$$\mathbf{N}(t) = \mathbf{F}_{\text{tot}}(t) + \gamma \dot{\mathbf{x}}(t) - \langle \mathbf{F}_{\text{tot}}(t) + \gamma \dot{\mathbf{x}}(t) \rangle$$

Since we already know from the results of the previous section that $\langle \mathbf{F}_{\text{tot}} - \mathbf{F}_p \rangle = 0$ and that $\langle \mathbf{F}_p \rangle = -\gamma \langle \dot{\mathbf{x}} \rangle$, we have to calculate

$$\lim_{\tau \to \infty} \langle [\mathbf{F}_{\text{tot}}(\tau) + \gamma \dot{\mathbf{x}}(\tau)] \cdot [\mathbf{F}_{\text{tot}}(\tau + t) + \gamma \dot{\mathbf{x}}(\tau + t)] \rangle$$

In Appendix C we show that the full expression for the noise can be replaced by

$$\lim_{\tau \to \infty} \langle [\mathbf{F}_{\text{tot}}(\tau) - \mathbf{F}_p(\tau)] \cdot [\mathbf{F}_{\text{tot}}(\tau + t) - \mathbf{F}_p(\tau + t)] \rangle$$

As was anticipated, $\mathbf{F}_{\text{tot}} - \mathbf{F}_p$ is therefore the noise in the Langevin equation.

Using Fourier integral representations for the density ρ_f and the interaction U, we obtain

$$\begin{aligned} &\lim_{\tau \to \infty} \langle [\mathbf{F}_{\text{tot}}(\tau) - \mathbf{F}_p(\tau)] \cdot [\mathbf{F}_{\text{tot}}(\tau + t) - \mathbf{F}_p(\tau + t)] \rangle \\ &\quad = -(2\pi)^6 \int d\mathbf{q}\, d\mathbf{p}\, \mathbf{q} \cdot \mathbf{p} U(p) U(q) \\ &\quad\quad \times \lim_{\tau \to \infty} \langle \rho_f(\mathbf{p}, \tau)\, \rho_f(\mathbf{q}, \tau + t) \exp[i\mathbf{p} \cdot x(\tau) + i\mathbf{q} \cdot \mathbf{x}(\tau + t)] \rangle \end{aligned} \tag{58}$$

As was explained in the previous section, the average in Eq. (58) cannot depend on $\mathbf{x}(t=0)$. This implies that the rhs of Eq. (58) contains a factor of $\delta(\mathbf{p} + \mathbf{q})$. The exponential in Eq. (58) may be therefore written inside the integral as $\exp\{i\mathbf{q} \cdot [\mathbf{x}(\tau + t) - \mathbf{x}(\tau)]\}$.

Since the interaction U is short-ranged, the force–force correlation has a finite width in time. Keeping that in mind, we assume, as in the last section, that during the time interval in which the correlation is appreciable the difference $\mathbf{x}(\tau + t) - \mathbf{x}(\tau)$ depends linearly on ρ_f. The difference is therefore also linear in the initial conditions of the liquid. We conclude that, to leading order in the particle-liquid interaction,[5]

$$\begin{aligned} &\lim_{\tau \to \infty} \langle \rho_f(\mathbf{p}, \tau)\, \rho_f(\mathbf{q}, \tau + t) \exp\{i\mathbf{q} \cdot [\mathbf{x}((\tau + t) - \mathbf{x}(\tau)]\} \rangle \\ &\quad = \lim_{\tau \to \infty} \langle \rho_f(\mathbf{p}, \tau)\, \rho_f(\mathbf{q}, \tau + t) \rangle \langle \exp\{i\mathbf{q} \cdot [\mathbf{x}(\tau + t) - \mathbf{x}(\tau)]\} \rangle \\ &\quad = \frac{k_B T}{(2\pi)^3 \lambda} \cos(qct)\, \delta(\mathbf{p} + \mathbf{q})\, f(q, ct) \end{aligned} \tag{59}$$

where $f(q, ct)$ is defined in Eq. (48).

[5] The first equality in Eq. (59) is based on the multidimensional analog of the following calculation:

$$\int d\rho\, \rho^2 e^{-\rho^2/2} e^{-i\alpha\rho} \Big/ \int d\rho\, e^{-\rho^2/2} = (1 - \alpha^2)\, e^{-\alpha^2/2} = \langle \rho^2 \rangle \langle e^{i\alpha\rho} \rangle (1 - \alpha^2)$$

The force–force correlation is therefore given by

$$\lim_{\tau \to \infty} \langle [\mathbf{F}_{\text{tot}}(\tau) - \mathbf{F}_p(\tau)] \cdot [\mathbf{F}_{\text{tot}}(\tau + t) - \mathbf{F}_p(\tau + t)] \rangle$$

$$= \frac{k_B T(2\pi)^3}{\lambda} \int U^2(q)\, q^2 \cos(qct)\, f(q, ct)\, d\mathbf{q} \equiv \Phi(t) \tag{60}$$

Note that the temperature T appears in Eq. (59) and (60) as a result of averaging over the initial conditions of the liquid.

In order to understand the qualitative behavior of Φ, we employ a specific form of the potential. This enables us to make an explicit calculation of Φ. We take $U(q) = U(0) \exp(-\frac{1}{2} R_0^2 q^2)$. As in the last section, we assume that Eq. (56) is obeyed and therefore we can use the long-time, diffusive form of $f(q, ct)$, $f(q, ct) = \exp(-q^2 v_{\text{th}}^2 (M/\gamma)\, |t|)$, over the whole range of integration.

We obtain

$$\Phi(t) = \text{Const} \times \frac{(2\pi)^3}{\lambda} k_B T U^2(0)$$

$$\times \left[1 - \frac{c^2 t^2}{R_0^2 + v_{\text{th}}^2 (M/\gamma)\, |t|} + \frac{1}{12} \frac{c^4 t^4}{[R_0^2 + v_{\text{th}}^2 (M/\gamma)\, |t|]^2} \right]$$

$$\times \frac{1}{[R_0^2 + v_{\text{th}}^2 (M/\gamma)\, |t|]^{5/2}} \exp\left[-\frac{1}{4} \frac{c^2 t^2}{R_0^2 + v_{\text{th}}^2 (M/\gamma)\, |t|} \right] \tag{61}$$

The width of the correlation $\Phi(t)$ is therefore $(v_{\text{th}}^2/c^2)\, M/\gamma$.

We now wish to determine how close $\Phi(t)$ is to a δ function in time. For that purpose we have to compare the width of Φ to the characteristic time scale over which the velocity changes appreciably, which is M/γ. We conclude that if Eq. (57) holds, that is, $v_{\text{th}}/c < 1$, we can approximate $\Phi(t)$ by

$$\Phi(t) \sim 3\sigma\delta(t) \tag{62}$$

where

$$3\sigma = \int_{-\infty}^{\infty} dt\, \Phi(t) \tag{63}$$

The factor 3 in Eqs. (62) and (63) is there because σ refers to the correlation strength of only one component of the force.

Using Eq. (63), we obtain

$$\sigma = \frac{(2\pi)^3}{3\lambda} k_B T \int U^2(q)\, q^2\, d\mathbf{q} \int_{-\infty}^{\infty} f(q, ct) \cos(qct)\, dt$$
$$= \frac{(2\pi)^3}{3\lambda} k_B T 2 \frac{v_{th}}{c} a \int U^2(q)\, q^2\, dq \tag{64}$$

where $a = v_{th}(M/\gamma)$.

In Eq. (64) we neglected terms of the order of $[(v_{th}/c)(a/R_0)]^2$ compared to unity.

By comparing Eq. (64) to Eq. (51), we see that the following relation between γ and σ holds:

$$k_B T\gamma = \sigma \tag{65}$$

Equation (65) is the fluctuation-dissipation theorem.

The thermal velocity v_{th} is given by [using Eqs. (51) and (64)]

$$v_{th}^2 = \frac{\sigma}{2\gamma M} = \frac{k_B T}{2M} \tag{66}$$

Equation (66) implies that the energy of the immersed particle thermalizes to the correct value.

Using Eq. (66), we obtain for σ

$$\sigma = \left[\frac{(2\pi)^3}{\lambda c^2} (k_B T)^3 \int U^2(q)\, q^2\, d\mathbf{q}\right]^{1/2} \tag{67}$$

In Eq. (67), σ is expressed in terms of the parameters of the original mechanical Lagrangian and the temperature T.

7. SUMMARY AND RESULTS

We have introduced a model Lagrangian describing a particle interacting with an idealized medium (solid or liquid) and studied the resulting Euler–Lagrange equations.

By integrating out the degrees of freedom of the medium, we obtained an exact, nonreversible, integrodifferential equation for the particle. We showed, without resorting to any approximation, that the force acting on the particle can be broken into two parts. The first part is due to the force the particle exerts on itself via the medium. This force is a damping force. The second part is the force that the medium would have exerted on a test particle (A particle not affecting the medium). This force has the properties of a random force.

We approximated our exact equation and obtained, under certain physical conditions, the usual Langevin equation. The Langevin parameters were calculated in terms of the parameters of the Lagrangian and the temperature of the medium. We assumed that the medium is *initially* in equilibrium and is described by a Boltzmann distribution at the temperature T. After the immersion of the particle into the medium, we did not assume that equilibrium is restored. We found that the long-time average of the kinetic energy of the particle is $\frac{3}{2}k_B T$, as expected. We also considered possible situations when the Langevin equation is not expected to give an adequate description of the system.

APPENDIX A. EFFECTIVE RANGE OF INTEGRATION

The force acting on the particle is expressed in terms of an integral over $\mathbf{R}$ and $\mathscr{R}$. The purpose of this Appendix is to evaluate the effective range of integration.

Assuming that $U(R)$ vanishes for $R > R_0$, it is obvious that the integration in Eq. (25) is limited to the range

$$|\mathscr{R}| < R_0 \tag{A1}$$

and is also bounded by

$$|\mathbf{R} + \mathscr{R} + \mathbf{x}(t) - \mathbf{x}(t - R/c)| < R_0 \tag{A2}$$

The $\mathbf{x}$ increment may be expanded, using the mean value theorem, in terms of the time difference R/c and an intermediate velocity

$$\mathbf{x}(t) - \mathbf{x}\left(t - \frac{R}{c}\right) = \dot{\mathbf{x}}(t')\,\frac{R}{c} \tag{A3}$$

where $t - R/c < t' < t$. Now,

$$\left|\mathbf{R} + \frac{\dot{\mathbf{x}}(t')}{c}\,R\right| < 2R_0 \tag{A4}$$

for otherwise

$$\left|\mathbf{R} + \mathscr{R} + \frac{\dot{\mathbf{x}}(t')}{c}\,R\right| \geqslant \left|\mathbf{R} + \frac{\dot{\mathbf{x}}(t')}{c}\,R\right| - |\mathscr{R}| \geqslant R_0 \tag{A5}$$

in contradiction with (A2).

The relevant velocities for our purpose are of the order of the thermal velocity, which is assumed to be small compared to the sound velocity. The effective range of integration is therefore bounded by

$$|\mathbf{R}| \leqslant 2R_0\left(1+\frac{|\dot{\mathbf{x}}(t')|}{c}\right) \sim 2R_0 \tag{A6}$$

APPENDIX B. DISSIPATION IN A SYSTEM OF HARD SPHERES AT $T=0$

In this Appendix we consider the average force on a particle interacting with a system of hard spheres. The hard spheres are initially at rest. The temperature of the medium in this case is therefore $T=0$.

We start with a one-dimensional example. Let the mass of the external particle be M, the mass of the host particles be m, and $M>m$. The external particle is immersed into the host medium with an initial velocity $+V_0$. The coordinate of the host particle to its right is x_R, and the velocity of that host particle is V_R ($V_R=0$ at $t=0$). The velocities of the particles after they collide V_0' and V_R' are

$$V_0'=\frac{M-m}{M+m}V_0 \tag{B1}$$

$$V_R'=\frac{2M}{M+m}V_0 \tag{B2}$$

After the collision both particles continue to move to the right. Since $V_R'>V_0'$, the host particle hits the next host particle first. After this collision its velocity becomes zero, and the second host particle continues to move with a velocity V_R'. A disturbance moving with a velocity V_R' is created. This disturbance no longer affects the external particle. The external particle moves after the first collision with a velocity V_0' and collides again with the same host particle, which is again at rest, only this time at a different position. The external particle therefore loses a fraction of its momentum at each collision. The momentum loss in a single collision is given by

$$\Delta P=\frac{2Mm}{M+m}V \tag{B3}$$

where V is its velocity prior to the collision. The average number of collisions per unit time is V/l, where l is the average distance between the host particles. The average momentum loss per unit time is therefore given by $\Delta P(V/l)$. The average force acting on the particle is therefore given by

$$F = \frac{2Mm}{M+m}\frac{1}{l}V^2 \tag{B4}$$

The average force acting on the particle is therefore proportional to the square of the velocity, and not to the velocity itself.

The above result is modified for finite temperatures in the following way. Consider a system of hard spheres at a temperature T, such that the thermal velocity of the host particles $V_{\mathrm{th}}^{\mathrm{h}}$ is larger than V. In this case

$$\Delta P \sim \frac{2Mm}{M+m} V_{\mathrm{th}}^{\mathrm{h}} \tag{B5}$$

so that the average force acting on the particle becomes

$$F = \frac{2Mm}{M+m}\frac{1}{l} V_{\mathrm{th}} V \equiv \gamma V \tag{B6}$$

Note that the temperature dependence of γ here is exactly the temperature dependence of γ given by Eq. (51).

The basic ingredient that forces γ to vanish at $T=0$ is the fact that the momentum loss of the external particle when colliding with a particle at rest is proportional to its own velocity.

Consider now the situation in more than one dimension. Most of the host particles with which the external particle collides are at rest. When the external particle collides with a host particle at rest the results of the one-dimensional example are valid. Namely, the average momentum loss is proportional to the velocity of the external particle, and therefore the average force acting on the external particle due to collisions with host particles at rest is proportional to the square of its velocity.

Some of the host particles are not at rest, but are moving because they collided in the past either with the external particle or with other host particles. One may think that collisions with moving host particles would change the results of the one-dimensional example. Note that since all the host particles were initially at rest, all the velocities of the particles are proportional to $\mathbf{V}_0$, the initial velocity of the external particle. At the nth collision of the external particle, it therefore hits a host particle with a velocity given by

$$\mathbf{V}_p^{(n)} = V_0 \mathbf{k}_p^{(n)} \tag{B7}$$

where $\mathbf{k}_p^{(n)}$ is a vector which depends on the mass ratio, the initial configuration, and the direction of the initial velocity, but not on the magnitude of the initial velocity. Similarly, the velocity of the external particle at the nth collision is given by

$$\mathbf{V}_{\text{ext}}^{(n)} = V_0 \mathbf{k}_{\text{ext}}^{(n)} \tag{B8}$$

Let the direction of the initial velocity be denoted by the unit vector $\hat{e}_i$. The momentum loss in this direction due to the nth collision is

$$\Delta P = M V_0 \mathbf{k}^{(n)} \cdot \hat{e}_i \tag{B9}$$

where $\mathbf{k}^{(n)}$ has the same properties as $\mathbf{k}_p^{(n)}$. The velocity of the external particle in the direction $\hat{e}_i$ is given by

$$V_{\text{ext}} = V_0 \mathbf{k}_{\text{ext}}^{(n)} \cdot \hat{e}_i \tag{B10}$$

and therefore

$$\Delta P = M \frac{\mathbf{k}^{(n)} \cdot \hat{e}_i}{\mathbf{k}_{\text{ext}}^{(n)} \cdot \hat{e}_i} V_{\text{ext}} \tag{B11}$$

The important point here is that the coefficient of V_{ext} does not depend on V_{ext}. The momentum loss of the external particle at each collision with a moving host particle is therefore proportional to the velocity of the external particle. The average force due to such collisions is therefore also proportional to the square of the velocity of the external particle.

The above procedure may seem rather artificial, since, by the same token, ΔP is proportional to V_0 and not to V_{ext}. As a result, it could be argued that the average force on the external particle is proportional to its velocity with a friction coefficient proportional to its initial velocity. However, the coefficient of V_0 in the average momentum loss at the nth collision must be a decreasing function of n, which vanishes for large values of n.

The physical reason for that is that the moving host particles are losing kinetic energy to the other host particles. Therefore the probability that at the nth collision the external particle will hit a host particle with a given finite fraction of the initial velocity V_0 vanishes with n.

At finite temperatures, the situation in more than one dimension is qualitatively the same as in the one-dimensional example discussed in the beginning of this Appendix. Once V_{ext} becomes small compared with V_{th}^{h}, we have again only one velocity scale in the system, namely V_{th}^{h}. The average momentum loss in this case is proportional to V_{th}^{h}, resulting in $\gamma \propto T^{1/2}$.

APPENDIX C. CORRECTIONS TO THE NOISE

The full noise term in the Langevin equation is $\mathbf{F}_{\text{tot}} + \gamma\dot{\mathbf{x}}$. In Section 7 we computed the force–force correlation strength σ using $\mathbf{F}_{\text{tot}} - \mathbf{F}_p$ instead. In this Appendix we evaluate the corrections $\delta\sigma$.

The corrections $\delta\sigma$ can be expressed as $\delta\sigma = \int_{-\infty}^{\infty} \delta\Phi(t)\, dt$, where

$$\begin{aligned}\delta\Phi(t) &= \delta_1\Phi(t) + \delta_2\Phi(t)\\ &= \lim_{\tau\to\infty} \langle \boldsymbol{\Delta}_f(\tau)\cdot\boldsymbol{\Delta}_f(\tau+t)\rangle\\ &\quad + \lim_{\tau\to\infty} \{\langle \boldsymbol{\Delta}_f(\tau)\cdot[\mathbf{F}_{\text{tot}}(\tau+t) - \mathbf{F}_p(\tau+t)]\rangle\\ &\quad + \langle \boldsymbol{\Delta}_f(\tau+t)\cdot[\mathbf{F}_{\text{tot}}(\tau) - \mathbf{F}_p(\tau)]\rangle\}\end{aligned} \tag{C1}$$

The additional noise force is given by

$$\boldsymbol{\Delta}_f = (\mathbf{F}_{\text{tot}} + \gamma\dot{\mathbf{x}}) - (\mathbf{F}_{\text{tot}} - \mathbf{F}_p) = \mathbf{F}_p + \gamma\dot{\mathbf{x}} \tag{C2}$$

Consider first $\delta_1\Phi(t)$,

$$\begin{aligned}\delta_1\Phi(t) &= \lim_{\tau\to\infty} \langle \boldsymbol{\Delta}_f(\tau)\cdot\boldsymbol{\Delta}_f(\tau+t)\rangle\\ &= \lim_{\tau\to\infty} \langle [\mathbf{F}_p(\tau) + \gamma\dot{\mathbf{x}}(\tau)]\cdot[\mathbf{F}_p(\tau+t) + \gamma\dot{\mathbf{x}}(\tau+t)]\rangle\end{aligned} \tag{C3}$$

Equation (C3) consists of four terms. Consider the first term:

$$\begin{aligned}&\lim_{\tau\to\infty} \langle \mathbf{F}_p(\tau)\cdot\mathbf{F}_p(\tau+t)\rangle\\ &= \lim_{\tau\to\infty} \frac{1}{(4\pi\lambda)^2} \int\int d\mathbf{R}\, d\mathcal{R}\, d\mathbf{R}'\, d\mathcal{R}'\, \nabla U(\mathcal{R})\cdot\nabla U(\mathcal{R}') \frac{1}{RR'}\\ &\quad\times\left\langle \nabla^2 U\left[\mathbf{R} + \mathcal{R} + \mathbf{x}(\tau) - \mathbf{x}\left(\tau - \frac{R}{c}\right)\right]\right.\\ &\quad\left.\times\nabla^2 U\left[\mathbf{R}' + \mathcal{R}' + \mathbf{x}(\tau+t) - \mathbf{x}\left(\tau + t - \frac{R'}{c}\right)\right]\right\rangle\\ &= -\frac{(2\pi)^6}{(4\pi\lambda)^2} \iint d\mathbf{R}\, d\mathbf{R}'\, d\mathbf{p}\, d\mathbf{q}(\mathbf{q}\cdot\mathbf{p})\, q^2 p^2 U^2(p)\, U^2(q) \frac{1}{RR'}\\ &\quad\times\exp(i\mathbf{q}\cdot\mathbf{R})\exp(i\mathbf{p}\cdot\mathbf{R}')\\ &\quad\times\lim_{\tau\to\infty}\left\langle \exp\left\{i\mathbf{q}\cdot\left[\mathbf{x}(\tau) - \mathbf{x}\left(\tau - \frac{R}{c}\right)\right]\right\}\right.\\ &\quad\left.\times\exp\left\{i\mathbf{p}\cdot\left[\mathbf{x}(\tau+t) - \mathbf{x}\left(\tau + t - \frac{R'}{c}\right)\right]\right\}\right\rangle\end{aligned} \tag{C4}$$

Using the same techniques as in Sections 6 and 7, we calculate the average in Eq. (C4):

$$\lim_{\tau\to\infty}\left\langle \exp\left\{i\mathbf{q}\cdot\left[\mathbf{x}(\tau)-\mathbf{x}\left(\tau-\frac{R}{c}\right)\right]\right\}\right.$$

$$\left.\times\exp\left\{i\mathbf{p}\cdot\left[\mathbf{x}(\tau+t)-\mathbf{x}\left(\tau+t-\frac{R'}{c}\right)\right]\right\}\right\rangle$$

$$=f(q,R)\,f(p,R')\exp\left\{-\mathbf{p}\cdot\mathbf{q}\lim_{\tau\to\infty}\left\langle\left[\mathbf{x}(\tau)-\mathbf{x}\left(\tau-\frac{R}{c}\right)\right]\cdot\right.\right.$$

$$\left.\left.\times\left[\mathbf{x}(\tau+t)-\mathbf{x}\left(\tau+t-\frac{R'}{c}\right)\right]\right\rangle\right\}\Bigg) \tag{C5}$$

where $f(k, R)$ is defined in Eq. (48).

Using the mean value theorem, we express the difference $\mathbf{x}(\tau)-\mathbf{x}(\tau-R/c)$ as follows:

$$\mathbf{x}(\tau)-\mathbf{x}\left(\tau-\frac{R}{c}\right)=\dot{\mathbf{x}}(t')\frac{R}{c} \tag{C6}$$

where $\tau-R/c<t'<\tau$. Similarly,

$$\mathbf{x}(\tau+t)-\mathbf{x}\left(\tau+t-\frac{R'}{c}\right)=\dot{\mathbf{x}}(t'')\frac{R'}{c} \tag{C7}$$

where $\tau+t-R'/c<t''<\tau+t$.

Using Eqs. (C5)–(C7), we obtain

$$\lim_{\tau\to\infty}\langle\mathbf{F}_p(\tau)\cdot\mathbf{F}_p(\tau+t)\rangle$$

$$=-\frac{(2\pi)^6}{(4\pi\lambda)^2}\iint d\mathbf{R}\,d\mathbf{R}'\,d\mathbf{p}\,d\mathbf{q}\,d\mathbf{p}(\mathbf{q}\cdot\mathbf{p})\,q^2p^2U^2(p)\,U^2(q)\frac{1}{RR'}$$

$$\times\exp(i\mathbf{q}\cdot\mathbf{R})\exp(i\mathbf{p}\cdot\mathbf{R}')\,f(q,R)\,f(p,R')$$

$$\times\exp\left[-\mathbf{p}\cdot\mathbf{q}\frac{RR'}{c^2}\lim_{\tau\to\infty}\langle\dot{\mathbf{x}}(t')\cdot\dot{\mathbf{x}}(t'')\rangle\right] \tag{C8}$$

We now expand

$$\exp\left[-\mathbf{p}\cdot\mathbf{q}\frac{RR'}{c^2}\lim_{\tau\to\infty}\langle\dot{\mathbf{x}}(t')\cdot\dot{\mathbf{x}}(t'')\rangle\right]$$

inside the integral. All even terms in the argument of the exponential vanish because U is spherically symmetric. Inside the average in Eq. (C8) we may also expand $\dot{\mathbf{x}}(t')$ in a Taylor series as follows:

$$\dot{\mathbf{x}}(t') = \dot{\mathbf{x}}(\tau) + \ddot{\mathbf{x}}(\tau)(t' - \tau) + \cdots \tag{C9}$$

and similarly

$$\dot{\mathbf{x}}(t'') = \dot{\mathbf{x}}(\tau + t) + \ddot{\mathbf{x}}(\tau + t)(t'' - \tau - t) + \cdots \tag{C10}$$

The required correlation may therefore be expressed as

$$\begin{aligned}
&\lim_{\tau \to \infty} \langle \mathbf{F}_p(\tau) \cdot \mathbf{F}_p(\tau + t) \rangle \\
&= \frac{(2\pi)^6}{(4\pi\lambda c)^2} \iint d\mathbf{R}\, d\mathbf{R}'\, d\mathbf{p}\, d\mathbf{q} (\mathbf{q} \cdot \mathbf{p})^2\, q^2 p^2 U^2(p)\, U^2(q) \\
&\quad \times \exp(i\mathbf{q} \cdot \mathbf{R}) \exp(i\mathbf{p} \cdot \mathbf{R}') f(q, R) f(p, R') \\
&\quad \times \Big\{ \langle \dot{\mathbf{x}}(\tau) \cdot \dot{\mathbf{x}}(\tau + t) \rangle + \langle \ddot{\mathbf{x}}(\tau) \cdot \dot{\mathbf{x}}(\tau + t) \rangle (t' - \tau) \\
&\quad + \langle \dot{\mathbf{x}}(\tau) \cdot \ddot{\mathbf{x}}(\tau + t) \rangle (t'' - \tau - t) + \cdots \\
&\quad + \frac{1}{6} \frac{(RR')^2}{c^4} (\mathbf{p} \cdot \mathbf{q})^2 (\langle \dot{\mathbf{x}}(\tau) \cdot \dot{\mathbf{x}}(\tau + t) \rangle)^3 + \cdots \Big\}
\end{aligned} \tag{C11}$$

The dots in Eq. (C11) stand for terms containing correlations of higher derivatives of $\dot{\mathbf{x}}$ or higher powers of correlations of $\dot{\mathbf{x}}$ or both.

The first term in Eq. (C11) is given by

$$\begin{aligned}
&\frac{(2\pi)^6}{(4\pi\lambda c)^2} \iint d\mathbf{R}\, d\mathbf{R}'\, d\mathbf{p}\, d\mathbf{q} (\mathbf{q} \cdot \mathbf{p})^2\, q^2 p^2 U^2(p)\, U^2(q) \\
&\quad \times \exp(i\mathbf{q} \cdot \mathbf{R}) \exp(i\mathbf{p} \cdot \mathbf{R}') \\
&\quad \times f(q, R) f(p, R') \langle \dot{\mathbf{x}}(\tau) \cdot \dot{\mathbf{x}}(\tau + t) \rangle = \gamma^2 \langle \dot{\mathbf{x}}(\tau) \cdot \dot{\mathbf{x}}(\tau + t) \rangle
\end{aligned} \tag{C12}$$

The higher terms in the expansion are discussed later in this section. These terms contribute to $\delta\sigma$.

Consider now the second and third terms in Eq. (C3). The third term, for example, is

$$\begin{aligned}
&\lim_{\tau \to \infty} \langle \mathbf{F}_p(\tau) \cdot \gamma \dot{\mathbf{x}}(\tau + t) \rangle \\
&= -\frac{\gamma (2\pi)^3}{4\pi\lambda} \int i\mathbf{q} q^2 U^2(q) \frac{1}{R} [\exp(i\mathbf{q} \cdot \mathbf{R})] \cdot \\
&\quad \times \lim_{\tau \to \infty} \left\langle \left(\exp \left\{ i\mathbf{q} \cdot \left[\mathbf{x}(\tau) - \mathbf{x}\left(\tau - \frac{R}{c} \right) \right] \right\} \right) \dot{\mathbf{x}}(\tau + t) \right\rangle d\mathbf{q}\, d\mathbf{R}
\end{aligned} \tag{C13}$$

Using the equality

$$\dot{\mathbf{x}}(\tau+t) = -i\frac{\delta}{\delta\mathbf{p}}\exp[i\mathbf{p}\cdot\dot{\mathbf{x}}(\tau+t)]_{|\mathbf{p}=0}$$

we may write Eq. (C13) in the following form:

$$\begin{aligned}&\lim_{\tau\to\infty}\langle\mathbf{F}_p(\tau)\cdot\gamma\dot{\mathbf{x}}(\tau+t)\rangle\\ &\quad= -\frac{\gamma(2\pi)^3}{4\pi\lambda}\frac{\delta}{\delta\mathbf{p}_{|\mathbf{p}=0}}\cdot\int \mathbf{q}q^2U^2(q)\frac{1}{R}[\exp(i\mathbf{q}\cdot\mathbf{R})]\\ &\quad\times\lim_{\tau\to\infty}\left\langle\exp\left\{i\mathbf{q}\cdot\left[\mathbf{x}(\tau)-\mathbf{x}\left(\tau-\frac{R}{c}\right)\right]\right\}\exp[i\mathbf{p}\cdot\dot{\mathbf{x}}(\tau+t)]\right\rangle d\mathbf{q}\,d\mathbf{R}\end{aligned} \tag{C14}$$

The average in Eq. (C14) can be evaluated using the same methods as in Eq. (C4). We obtain

$$\begin{aligned}&\lim_{\tau\to\infty}\langle\mathbf{F}_p(\tau)\cdot\gamma\dot{\mathbf{x}}(\tau+t)\rangle\\ &\quad= -\gamma^2\langle\dot{\mathbf{x}}(\tau)\cdot\dot{\mathbf{x}}(\tau+t)\rangle-\gamma^2\langle\ddot{\mathbf{x}}(\tau)\cdot\dot{\mathbf{x}}(\tau+t)\rangle(t'-\tau)+\cdots\end{aligned} \tag{C15}$$

where $\tau - R/c < t' < \tau$.

Using Eqs. (C12) and (C15), we see that all the leading terms in the required correlation function sum up to zero. Consider now the corrections $\delta_1\sigma$ due to terms that contain higher derivatives of $\dot{\mathbf{x}}$. The average $\langle\mathbf{x}^{(n)}(\tau)\cdot\mathbf{x}^{(m)}(\tau+t)\rangle$ can be written as $(d/dt)\langle\mathbf{x}^{(n)}(\tau)\cdot\mathbf{x}^{(m-1)}(\tau+t)\rangle$. The corresponding $\delta_1\sigma$ therefore obeys the relation

$$\delta_1\sigma \propto \int_{-\infty}^{\infty} dt\,\frac{d}{dt}\langle\mathbf{x}^{(n)}(\tau)\cdot\mathbf{x}^{(m-1)}(\tau+t)\rangle = \langle\mathbf{x}^{(n)}(\tau)\cdot\mathbf{x}^{(m-1)}(\tau+t)\rangle|_{-\infty}^{\infty} \tag{C16}$$

The correlations $\langle\mathbf{x}^{(n)}(\tau)\cdot\mathbf{x}^{(m-1)}(\tau+t)\rangle$ can be computed using the Langevin equation. These correlations vanish for large $|t|$, for $n, m-1 \geqslant 1$. The contribution to $\delta_1\sigma$ due to those terms therefore vanishes.

Consider now the corrections $\delta\sigma$ that come from the term containing higher powers of the average $\langle\dot{\mathbf{x}}(\tau)\,\dot{\mathbf{x}}(\tau+t)\rangle$. The leading contribution comes from

$$\begin{aligned}\delta_1\Phi(t) &\sim \frac{(2\pi)^6}{(4\pi\lambda c)^2}\iint d\mathbf{R}\,d\mathbf{R}'\,d\mathbf{p}\,d\mathbf{q}(\mathbf{q}\cdot\mathbf{p})^2 q^2p^2U^2(p)\,U^2(q)\\ &\quad\times\exp(i\mathbf{q}\cdot\mathbf{R})\exp(i\mathbf{p}\cdot\mathbf{R}')\,f(q,R)\,f(p,R')\\ &\quad\times\frac{1}{6}\frac{(RR')^2}{c^4}(\mathbf{p}\cdot\mathbf{q})^2[\langle\dot{\mathbf{x}}(\tau)\cdot\dot{\mathbf{x}}(\tau+t)\rangle]^3\\ &\sim\frac{\gamma^2}{c^4}[\langle\dot{\mathbf{x}}(\tau)\cdot\dot{\mathbf{x}}(\tau+t)\rangle]^3\end{aligned} \tag{C17}$$

The average in Eq. (C17) can be computed using the Langevin equation. Since

$$\langle \dot{\mathbf{x}}(\tau) \cdot \dot{\mathbf{x}}(\tau + t) \rangle = v_{\rm th}^2 e^{-(\gamma/M)|t|} \tag{C18}$$

we conclude that

$$\delta_1 \Phi \sim \frac{\gamma^2}{c^4} v_{\rm th}^6 e^{-(3\gamma/M)|t|} \tag{C19}$$

The corresponding correction to σ is

$$\delta_1 \sigma \sim \frac{M\gamma}{c^4} v_{\rm th}^6 \tag{C20}$$

and using the definition of $v_{\rm th}$, $v_{\rm th} = \sigma/2\gamma M$, we see that

$$\delta_1 \sigma \sim \sigma(v_{\rm th}^4/c^4) \tag{C21}$$

There are also contributions to $\delta_1 \sigma$ from terms containing higher powers and higher derivatives of $\dot{\mathbf{x}}$. The leading contribution comes from terms of the form

$$\begin{aligned}\delta_1 \Phi(t) \sim & \frac{(2\pi)^6}{(4\pi\lambda c)^2} \iint d\mathbf{R}\, d\mathbf{R}'\, d\mathbf{p}\, d\mathbf{q} (\mathbf{q}\cdot\mathbf{p})^2\, q^2 p^2 U^2(p)\, U^2(q) \\ & \times \exp(i\mathbf{q}\cdot\mathbf{R}) \exp(i\mathbf{p}\cdot\mathbf{R}')\, f(q, R)\, f(p, R') \\ & \times \frac{1}{6} \frac{(RR')^2}{c^4} (\mathbf{p}\cdot\mathbf{q})^2 \, [\langle \dot{\mathbf{x}}(\tau)\cdot\dot{\mathbf{x}}(\tau+t)\rangle]^2 \langle \dot{\mathbf{x}}(\tau)\cdot\ddot{\mathbf{x}}(\tau+t)\rangle (t'' - \tau - t) \\ \sim & \frac{\gamma^2}{c^4} \frac{\gamma R_0}{Mc} [\langle \dot{\mathbf{x}}(\tau)\cdot\dot{\mathbf{x}}(\tau+t)\rangle]^3 \end{aligned} \tag{C22}$$

In Eq. (C22) we used the fact that $\tau + t - R'/c < t'' < \tau + t$ and that the effective range of the R' integration is of the order of R_0.

The corresponding $\delta_1 \sigma$ is

$$\delta_1 \sigma \sim \sigma \frac{v_{\rm th}^4}{c^4} \frac{\gamma R_0}{Mc} \tag{C23}$$

Using Eq. (54), we see that the leading contribution to $\delta_1 \sigma$ is indeed given by Eq. (C21). Recall that in Eq. (57) we already assumed that $v_{\rm th}/c < 1$. We conclude that $\delta_1 \sigma$ is small compared to σ itself.

We now discuss the other corrections to σ, $\delta_2\sigma$, defined in Eq. (C3). We have

$$\begin{aligned}\delta_2\sigma &= \int_{-\infty}^{\infty} \delta_2\Phi(t)\,dt \\ &= \langle \mathbf{\Delta}_f(\tau)\cdot[\mathbf{F}_{\text{tot}}(\tau+t)-\mathbf{F}_p(\tau+t)]\rangle + \langle \mathbf{\Delta}_f(\tau+t)\cdot[\mathbf{F}_{\text{tot}}(\tau)-\mathbf{F}_p(\tau)]\rangle\end{aligned} \tag{C24}$$

We notice that the quantity

$$\sigma_{12} = \int_{-\infty}^{\infty} dt \lim_{\tau\to\infty} \langle \mathbf{F}_1(\tau)\cdot\mathbf{F}_2(\tau+t)\rangle \tag{C25}$$

has the properties of a scalar product. It therefore obeys the Schwartz inequality. Using this fact and Eqs. (64) and (C21), we obtain

$$|\delta_2\sigma| \leqslant \sigma^{1/2}(\delta_1\sigma)^{1/2} \sim \sigma(v_{\text{th}}^2/c^2) \tag{C26}$$

and since $v_{\text{th}}/c < 1$ [Eq. (57)], we conclude that $\delta_2\sigma/\sigma < 1$.

The final conclusion of this Appendix is therefore that the corrections to the force–force correlation strength due to the additional noise force are negligable compared to σ.

REFERENCES

1. J. G. Kirkwood, *J. Chem. Phys.* **14**:180 (1946).
2. F. Reif, *Statistical Physics*, (McGraw-Hill, 1967).
3. R. Zwanzig, in *Lectures in Theoretical Physics*, Vol. 3 (Interscience, 1961).
4. H. Mori, *Prog. Theor. Phys. (Kyoto)* **34**:423 (1965).
5. B. J. Berne and G. D. Harp, *Ad. Chem. Phys.* **XVII**, 63 (1970).
6. J. L. Leibowitz and P. Resibois, *Phys. Rev. A* **139**:1101 (1965).
7. D. Forster, *Hydrodynamic Fluctuations, Broken Symmetry and Correlation Functions* (Benjamin, 1975).
8. R. P. Feynman and F. L. Vernon, *Ann. Phys (N.Y.)* **24**:118 (1963).
9. A. O. Caldera and A. J. Legget, *Ann. Phys. (N.Y.)* **149**:374 (1984).
10. G. W. Ford, M. Kac, and P. Mazur, *J. Math. Phys.* **6**:504 (1965).
11. S. Marianer and J. M. Deutch, *Phys. Rev. B* **31**:7478 (1985).
12. A. Schmid, *J. Low Temp. Phys.* **49**:609 (1982).
13. H. Dekker, *Phys. Rev. A* **31**:1067 (1985).
14. L. D. Landau, *Zh. Eksp. Teor. Fiz.* **11**:592 (1941).