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Effective temperatures of hot Brownian motion

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We derive generalized Langevin equations for the translational and rotational motion of a heated Brownian particle from the fluctuating hydrodynamics of its nonisothermal solvent. The temperature gradient around the particle couples to the hydrodynamic modes excited by the particle itself so that the resulting noise spectrum is governed by a frequency-dependent temperature. We show how the effective temperatures at which the particle coordinates and (angular) velocities appear to be thermalized emerge from this central quantity.

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I. INTRODUCTION

Hot Brownian motion [1] is the diffusive dynamics of a colloidal particle persistently maintained at higher temperature than the surrounding fluid, so that the fluid temperature field $T(\boldsymbol{r}) = T(r)$ decays radially around the particle. It is of practical relevance, e.g., for laser-heated suspended nanoparticles involved in several experimental applications ranging from particle trapping and tracking [2,3] to self-thermophoretic microswimmers [4,5]. Besides, it is also of considerable theoretical interest, since it can be thought of as an archetypical example of a system in contact with a nonisothermal bath, hence far from thermal equilibrium. Nevertheless, for important conceptual and practical purposes, the hot particle can often be treated like an equivalent Brownian particle in equilibrium, with appropriate effective transport coefficients.

In particular, it has been shown analytically [6,7] that free and confined diffusion of a hot spherical particle are, in the long-time limit, governed by effective "positional" temperatures, denoted by T^X and T^Θ for translation in the X direction and rotation along an angle Θ, respectively. The positional temperatures enter the effective Stokes-Einstein relations and Boltzmann factors for translation and rotation of the particle, respectively. Additionally, extensive numerical simulations [6,8] have shown that the Maxwellian (angular) velocity distribution and the short-time response of the hot Brownian particle are characterized by yet other, somewhat higher, effective temperatures, so-called kinetic temperatures T^V and T^Ω. None of these effective temperatures are generally equal to the solvent temperature $T_s \equiv T(r \to R)$ at the particle surface or to the ambient temperature $T_0 \equiv T(r \to \infty)$. This complex behavior has led to the conclusion that an effective Langevin description of hot Brownian motion is restricted to the Markov limit [8].

Here we show constructively how this limitation can be overcome, starting from the fluctuating hydrodynamics of a solvent maintained at local thermal equilibrium with a temperature field $T(\boldsymbol{r})$. On this basis, we derive the generalized Langevin equation (GLE) for a heated spherical particle. Conceptually, nonspherical particles can be treated along the same lines, albeit with additional complications [7]. The most conspicuous feature of the theory is a frequency-dependent noise temperature $\mathcal{T}(\omega)$ [9]. It arises from the hydrodynamic coupling between the particle and distant solvent volume elements that are locally equilibrated at different temperatures $T(\boldsymbol{r})$. From this central quantity analytical predictions for the mentioned kinetic and positional effective temperatures are derived.

The characteristic frequency scales that primarily select the dominant modes from the "temperature spectrum" $\mathcal{T}(\omega)$ are (for a translating sphere of mass m, density ϱ_p, and radius R)

$$\omega_f \equiv \frac{2\nu}{R^2} \quad \text{and} \quad \omega_p \equiv \frac{6\pi\eta R}{m} = \frac{9\varrho}{4\varrho_p}\omega_f, \tag{1}$$

namely, the inverse time scale for vorticity diffusion across the particle, and the inverse Stokes relaxation time of the particle momentum, respectively. The former characterizes how efficiently the particle momentum is spatially dispersed in a solvent of kinematic viscosity $\nu = \eta/\varrho$ and density ϱ, and the latter how, as a result, the motion of the particle adjusts to that of the fluid. The meaning of slow and fast processes, or low and high frequencies of the noise spectrum, is primarily provided by these rates. It should be clear, though, that any externally imposed additional time scale that interferes with these rates can be expected to yield additional features.

The paper is structured as follows. In the next section we introduce the theoretical model of a Brownian particle in a nonisothermal solvent. We then sketch the contraction of the coupled solvent-particle system to the GLE for the particle motion, alone. Details of the calculation are given in Appendix A. In Sec. III we examine the frequency-dependent temperatures $\mathcal{T}(\omega)$ that govern the Langevin noise for the translational and rotational degrees of freedom of a heated sphere and give a qualitative physical interpretation of their functional form, while some technicalities are deferred to Appendix B. From this central quantity, we derive the effective rotational and translational kinetic temperatures of a free particle in Sec. IV. We analyze their explicit dependence on the characteristic time scales for the velocity relaxation of the particle and the solvent by varying their density ratio. Also we regain the known positional temperatures for translation and rotation [6,7] as the low-frequency limits of $\mathcal{T}(\omega)$. In Sec. V, we consider a hot Brownian particle trapped in a harmonic

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potential. While the known effective-equilibrium description can be retrieved in the Markov limit, we point out that the kinetic and positional temperatures may differ from those in the free case for very stiff traps, due to the interference of the characteristic trap time scale with the rates defined in Eq. (1). We conclude with a summary and short outlook.

II. FLUCTUATING HYDRODYNAMICS OF A HEATED PARTICLE

We consider a spherical particle of radius R immersed in an incompressible fluid of density ϱ. The time evolution of fluid momentum is described by the linearized fluctuating hydrodynamic equations [10–13]

$$\varrho\partial_t \boldsymbol{v}(\boldsymbol{r},t) - \nabla\cdot\boldsymbol{\sigma}(\boldsymbol{r},t) = \nabla\cdot\boldsymbol{\tau}(\boldsymbol{r},t), \tag{2a}$$

$$\nabla\cdot\boldsymbol{v}(\boldsymbol{r},t) = 0, \tag{2b}$$

$$\boldsymbol{v}(\boldsymbol{r},t) = \boldsymbol{V}(t) + \boldsymbol{\Omega}(t)\times\boldsymbol{r} \quad \text{on } \mathcal{S}, \tag{2c}$$

where the velocity field $\boldsymbol{v}$ of the fluid is defined in the volume $\mathcal{V}$ outside the particle and the no-slip boundary condition on the particle surface $\mathcal{S}$ is imposed by Eq. (2c). The stress tensor $\boldsymbol{\sigma}$ has components

$$\sigma_{ij}(\boldsymbol{r},t) = -p(\boldsymbol{r},t)\delta_{ij} + 2\eta(\boldsymbol{r},t)\Gamma_{ij}(\boldsymbol{r},t), \tag{3}$$

where p is the pressure and $\Gamma_{ij} = (\partial_i v_j + \partial_j v_i)/2$ the shear rate tensor, with the dynamic viscosity η. The incompressibility condition Eq. (2b) can be eliminated by expressing p (and thus $\boldsymbol{\sigma}$) as a functional of the flow field $\boldsymbol{v}$. Finally, the thermal noise is represented by a zero-mean Gaussian random stress tensor $\boldsymbol{\tau}$ that vanishes on the particle surface [12] and otherwise obeys the fluctuation-dissipation relation

$$\langle\tau_{ij}(\boldsymbol{r},t)\tau_{kl}(\boldsymbol{r}',t')\rangle = 2\eta(\boldsymbol{r},t)k_{\mathrm{B}}T(\boldsymbol{r},t)\delta(\boldsymbol{r}-\boldsymbol{r}') \times\delta(t-t')(\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}) \tag{4}$$

corresponding to a local equilibrium with the prescribed deterministic temperature field $T(\boldsymbol{r},t)$ [14]. In general, the dynamical viscosity $\eta(\boldsymbol{r},t)$ inherits some spatiotemporal dependence of $T(\boldsymbol{r},t)$. The vectors $\boldsymbol{V}(t)$ and $\boldsymbol{\Omega}(t)$, denoting the translational and rotational velocity of the Brownian particle, couple to the solvent dynamics via the boundary condition (2c) on the particle surface $\mathcal{S}$. They evolve themselves according to Newton's equations of motion

$$m\dot{\boldsymbol{V}}(t) = \boldsymbol{F}(t) + \boldsymbol{F}_e(t), \tag{5a}$$

$$I\dot{\boldsymbol{\Omega}}(t) = \boldsymbol{T}(t) + \boldsymbol{T}_e(t), \tag{5b}$$

where m is the mass of the particle, I the moment of inertia, $\boldsymbol{F}_e$ and $\boldsymbol{T}_e$ are the external force and torque, and $\boldsymbol{F}$ and $\boldsymbol{T}$ are the hydrodynamic force and torque exerted by the fluid, defined by

$$\boldsymbol{F}(t) = -\int_{\mathcal{S}} \boldsymbol{\sigma}(\boldsymbol{r},t)\cdot\boldsymbol{n}\, d^2r, \tag{6a}$$

$$\boldsymbol{T}(t) = -\int_{\mathcal{S}} \boldsymbol{r}\times(\boldsymbol{\sigma}(\boldsymbol{r},t)\cdot\boldsymbol{n})\, d^2r, \tag{6b}$$

with $\boldsymbol{n}$ the inner radial unit vector. Note that we have suppressed the time dependence of $\mathcal{S}$ in Eqs. (2c) and (6) in order to make the above set of equations linear not only in the flow field but also in the particle velocity. See Refs. [15,16] for a discussion of the validity of linear hydrodynamics in relation to Brownian motion. We also suppress the corresponding time-dependent thermal advection, by requiring the deterministic part of the temperature field to obey the stationary heat equation in the comoving frame,

$$\nabla^2 T(\boldsymbol{r}) = 0, \quad T(\boldsymbol{r}) = T_0 + \Delta T \;\text{ on } \mathcal{S}, \quad T(r\to\infty) = T_0. \tag{7}$$

This technical simplification and other implicit idealizations, such as neglecting the viscous heating due to the particle motion with respect to the housekeeping heat and taking the heat conductivity of the solvent to be constant, can be justified for common experimental conditions, such as those realized for laser-heated nanoparticles in water [1,17]. Together with the prescription Eq. (7), the system (2)–(4) then entirely describes the time evolution of the fluid and the heated Brownian particle. Notice that the fluctuations in the fluid temperature are irrelevant in our hydrodynamic description as they do not couple with momentum fluctuations thanks to the incompressibility assumption. Consequently, only the deterministic fluid temperature $T(\boldsymbol{r})$ is considered. The solution of Eq. (7) is the radial field:

$$T(r) = T_0 + \Delta T R/r. \tag{8}$$

While the following derivation does not strictly depend on the specific form of $T(\boldsymbol{r})$ (as long as it does not depend on the particle velocity), and even an explicit externally imposed dependence on time could be included, we restrict the discussion in the following sections to this paradigmatic case.

We now proceed to contract the description of fluid plus particle into an equation for the particle alone. We rewrite the hydrodynamic forces introduced in Eq. (5) in the form

$$\boldsymbol{F} \equiv \boldsymbol{F}_d + \boldsymbol{\xi}^T, \tag{9a}$$

$$\boldsymbol{T} \equiv \boldsymbol{T}_d + \boldsymbol{\xi}^R \tag{9b}$$

to account for contributions $\boldsymbol{\xi}$ independent of the particle velocity that are expected to arise due to the inhomogeneity of Eq. (2a). By Eq. (2), $\boldsymbol{v}(\boldsymbol{r},t)$ is a linear functional of $\boldsymbol{V}(t')$ and $\boldsymbol{\Omega}(t')$ with $-\infty < t' < t$, so in view of Eq. (6) this implies that the systematic components $\boldsymbol{F}_d$ and $\boldsymbol{T}_d$ are linear functionals of $\boldsymbol{V}(t')$ and $\boldsymbol{\Omega}(t')$, respectively, with $-\infty < t' < t$. Hence, we can write

$$\boldsymbol{F}_d(t) = -\int_{-\infty}^{t} \zeta(t-t')\boldsymbol{V}(t')\,dt', \tag{10}$$

$$\boldsymbol{T}_d(t) = -\int_{-\infty}^{t} \gamma(t-t')\boldsymbol{\Omega}(t')\,dt', \tag{11}$$

where $\zeta(t)$ and $\gamma(t)$ are positive, time-symmetric memory kernels accounting for the time-dependent drag on the particle [12]. Equations (5) then take the GLE form

$$M\dot{\boldsymbol{V}}(t) = -\int_{-\infty}^{t} \zeta(t-t')\boldsymbol{V}(t')\,dt' + \boldsymbol{\xi}^T(t) + \boldsymbol{F}_e(t), \tag{12}$$

$$I\dot{\boldsymbol{\Omega}}(t) = -\int_{-\infty}^{t} \gamma(t-t')\boldsymbol{\Omega}(t')\,dt' + \boldsymbol{\xi}^R(t) + \boldsymbol{T}_e(t), \tag{13}$$

once we identify $\boldsymbol{\xi}^{T,R}$ as the Langevin noise, whose statistical properties have to be derived from those of the random stress tensor $\boldsymbol{\tau}$.

For better readability, the actual calculation is detailed in Appendix A, and only the main results and their physical interpretations are given in the main text. We focus mostly on the translational motion, but the rotational case is very analogous. It is moreover convenient to switch to the frequency representation defining, for a generic function $g(\omega)$, the Fourier transform $g(\omega) \equiv \int_{-\infty}^{\infty} e^{i\omega t} g(t)\, dt$ and the half-Fourier transform $g^{+}(\omega) \equiv \int_{0}^{\infty} e^{i\omega t} g(t)\, dt$.

To complete the contraction, we compare the energy dissipated by the fluid friction acting on the particle at a mean velocity $\langle \boldsymbol{V}(\omega)\rangle$

$$\zeta(\omega)\delta_{ij}\langle V_i(\omega)\rangle\langle V_j^*(\omega)\rangle = 2\int_{\mathcal{V}} \phi^T(\boldsymbol{r},\omega)\, d^3r, \tag{14}$$

with the correlation function of the energy supplied by the random force at frequencies ω and ω':

$$\begin{aligned}&\left\langle \xi_i^T(\omega)\xi_j^{T*}(\omega')\right\rangle\langle V_i(\omega)\rangle\langle V_j^*(\omega')\rangle\\ &\quad = 2k_{\mathrm{B}}\delta(\omega-\omega')\int_{\mathcal{V}} \phi^T(\boldsymbol{r},\omega)T(\boldsymbol{r})\, d^3r\,. \end{aligned}\tag{15}$$

From Appendix A, we have quoted the representation in terms of the dissipation function,

$$\phi^T(\boldsymbol{r},\omega) \equiv \eta(\partial_i u_j \partial_i u_j^* + \partial_i u_j \partial_j u_i^*), \tag{16}$$

which gives the energy dissipated by the fluid at position $\boldsymbol{r}$ and frequency ω in terms of the average flow field $\boldsymbol{u}(\boldsymbol{r},\omega)$. Setting the arbitrary average velocity of the particle to unity, Eq. (14) allows one to calculate the memory kernel $\zeta(\omega)$ in terms of the spatial integral of the dissipation function $\phi^T(\boldsymbol{r},\omega)$.

From Eqs. (14) and (15) we then find the relation

$$\begin{aligned}&\left\langle \xi_i^T(\omega)\xi_j^{T*}(\omega')\right\rangle\langle V_i(\omega)\rangle\langle V_j^*(\omega')\rangle\\ &\quad = k_{\mathrm{B}}\mathcal{T}^T(\omega)\zeta(\omega)\delta_{ij}\delta(\omega-\omega')\langle V_i(\omega)\rangle\langle V_j^*(\omega')\rangle \end{aligned}\tag{17}$$

with

$$\mathcal{T}^T(\omega) \equiv \frac{\int_{\mathcal{V}} \phi^T(\boldsymbol{r},\omega)T(\boldsymbol{r})\, d^3r}{\int_{\mathcal{V}} \phi^T(\boldsymbol{r},\omega)\, d^3r}. \tag{18}$$

Since $\phi^T(\boldsymbol{r},\omega)$ is a quadratic function of $\langle \boldsymbol{V}(\omega)\rangle$ (see Appendix B) the ratio in Eq. (18) is independent of $\langle \boldsymbol{V}(\omega)\rangle$. Moreover, as the particle velocity $\langle \boldsymbol{V}(\omega)\rangle$ is arbitrary it can be deleted in Eq. (17), which renders Eq. (17) in the form of a generalized fluctuation-dissipation relation:

$$\left\langle \xi_i^T(\omega)\xi_j^{T*}(\omega')\right\rangle = k_{\mathrm{B}}\mathcal{T}^T(\omega)\zeta(\omega)\delta_{ij}\delta(\omega-\omega')\,. \tag{19}$$

According to Eq. (A9), the Langevin noise $\boldsymbol{\xi}^T$ is Gaussian distributed with mean zero. Therefore Eq. (19) fully characterizes the noise statistics. Analogous results hold for the rotational motion. They are obtained by substituting $\zeta \to \gamma$ in Eq. (19) and $\phi^T \to \phi^R$ in the definition (18).

III. THE NOISE TEMPERATURE $\mathcal{T}(\omega)$

Equation (18) defines the frequency-dependent noise temperature that is the central quantity for the Brownian motion under nonisothermal conditions. Its nonlocal nature manifests itself in the weighted average over the temperature field $T(\boldsymbol{r})$, with the dissipation function determining how strongly the diverse local temperatures in the surroundings affect the Brownian motion of the particle at the origin.

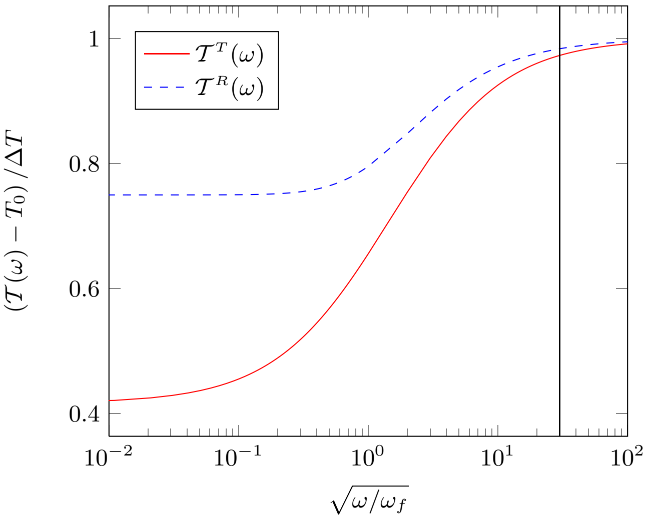

FIG. 1. (Color online) The universal frequency-dependent noise temperatures for the motion of a Brownian particle, obtained by the definitions (16) and (18) assuming a temperature-independent solvent viscosity η. The rotational noise temperature (blue/top line) is given by the exact expression Eq. (B3), while the translational one (red/bottom line) is obtained by numerical integration of Eqs. (18) and (B1). The vertical solid line indicates the characteristic frequency beyond which the finite compressibility of water would matter for a solid particle of radius $R \simeq 100$ nm.

Clearly, the noise autocorrelation can always be cast in such a form by defining a suitable function $\mathcal{T}(\omega)$ that measures the violation of the equilibrium fluctuation-dissipation relation. Here the nontrivial statement is that $\mathcal{T}(\omega)$ is explicitly derived from an underlying hydrodynamic description. Moreover, in the next sections, we will show that $\mathcal{T}(\omega)$ plays the role of a frequency-dependent effective temperature, in the sense that dynamical isothermal relations can directly be extended to the nonisothermal case if the temperature T_0 is replaced by $\mathcal{T}(\omega)$.

Contenting ourselves with explicit evaluations to leading order in the temperature heterogeneity $T(\boldsymbol{r}) - T_0$, we can in the following neglect a possible temperature dependence of the viscosity, which would affect our results to subleading order, only. Figure 1 shows the frequency-dependent temperatures $\mathcal{T}(\omega)$ for the translational and the rotational motion of a sphere, which are derived in Appendix B assuming constant heat conductivity and viscosity, i.e., Eq. (7) and $\eta(\boldsymbol{r}) = \eta$. As a consequence, η cancels in Eq. (18), and the obtained noise temperatures are universal functions independent of the solvent properties. All the subsequent results are derived under the latter approximation.

To gain a physical understanding of the functional form of $\mathcal{T}(\omega)$, consider its origin from the hydrodynamic coupling between the particle and distant solvent volume elements that are locally equilibrated at different temperatures $T(\boldsymbol{r})$. In our low-Reynolds number approximation, the exchange of momentum is dominated by vorticity diffusion [18] with the diffusivity given by the kinematic viscosity $\nu \equiv \eta/\varrho$. This defines the inverse characteristic time scale $\omega_f \equiv 2\nu/R^2$ for fluid transport over distances on the order of the particle radius, as introduced in Eq. (1).

Low-frequency fluctuations are those with $\omega \ll \omega_f$, during which the vorticity spreads out considerably from the particle. Since the translational field is more long-range than the rotational one ($\boldsymbol{u}^T \sim 1/r$ versus $\boldsymbol{u}^R \sim 1/r^2$), the translational noise is effectively cooler, as it involves an average over farther, i.e., cooler, regions of fluid. Ultimately, in the limit $\omega \to 0$, we find that the noise temperatures reduce to the effective temperatures $T_{\text{HBM}}^{X,\Theta}$ known to characterize the overdamped hot Brownian motion of the positions and angles, respectively [6,7], for which we employ the shorthand notation

$$T^X \equiv \mathcal{T}^T(0) = T_0 + \tfrac{5}{12}\Delta T,$$
$$T^\Theta \equiv \mathcal{T}^R(0) = T_0 + \tfrac{3}{4}\Delta T\,.$$

In contrast, during *high-frequency fluctuations* with $\omega \gg \omega_f$, fluid momentum cannot diffuse significantly from the particle surface. The vorticity emanating from a particle oscillating at frequency ω cannot penetrate the fluid beyond the skin depth $k_0^{-1} \equiv (2\nu/\omega)^{1/2} \ll R$, resulting in an exponential decay $\phi(r,\omega) \propto e^{-k_0(r-R)}$ of the dissipation function; see Eqs. (B1) and (B2). Therefore, the average in Eq. (18) is essentially restricted to a thin skin of solvent around the particle surface, and the noise temperatures tend towards the surface temperature $T_s = T_0 + \Delta T$ for large ω. However, note that the finite compressibility becomes relevant at very high frequencies. As a consequence, the noise temperature may deviate significantly from our predictions for frequencies larger than the inverse of the time it takes a sound wave in the solvent to traverse a distance R [15,19].

It is worth mentioning another peculiarity implied by the incompressibility assumption. To accelerate a particle in an incompressible fluid, the displaced fluid has to be moved from the front to the back of the particle. Therefore, the layer of fluid that is set into motion never collapses completely onto the particle surface, as it does for rotation at high frequencies. While the noise temperature is not affected, since the bulk dissipation turns out to be subdominant (see Appendix B), incompressibility results in a renormalized particle mass [10,12]:

$$M = m + m\varrho/(2\varrho_p). \tag{20}$$

The added mass in Eq. (20), owing to the inertia of the displaced fluid, becomes relevant in the following. In the next sections we analyze some immediate implications of the above results for the dynamics of a hot Brownian sphere that is either freely diffusing or trapped in a confining potential.

IV. THE KINETIC TEMPERATURE

The GLEs (5a) and (5b) both contain a Gaussian noise satisfying a fluctuation-dissipation relation with constant effective temperatures in the high-frequency limit. Therefore, one may expect to find Maxwell-Boltzmann distributions of translational and angular velocities under stationary conditions, which is corroborated by molecular dynamics simulations [6,7]. We thus define the kinetic temperatures such that the stationary averages of the velocities satisfy

$$\tfrac{3}{2}k_B T^V \equiv \tfrac{1}{2}M\langle \boldsymbol{V}^2\rangle, \quad \tfrac{3}{2}k_B T^\Omega \equiv \tfrac{1}{2}I\langle \boldsymbol{\Omega}^2\rangle, \tag{21}$$

which reduce to the equipartition theorem with $T^\Omega = T^V = T_0$ in case of a constant fluid temperature $T(\boldsymbol{r}) \equiv T_0$. For simplicity, we concentrate on the translational motion, in the following, but the same procedure applies also to the rotational motion.

From the Fourier transform of Eq. (12) in the absence of an external force,

$$-i\omega m \boldsymbol{V}(\omega) = -\zeta^+(\omega)\boldsymbol{V}(\omega) + \boldsymbol{\xi}^T(\omega), \tag{22}$$

we derive the velocity spectral density

$$C_V(\omega) \equiv \langle \boldsymbol{V}(\omega)\cdot\boldsymbol{V}(-\omega)\rangle = |R_V(\omega)|^2 C_\xi^T(\omega)\,. \tag{23}$$

Here

$$C_\xi^T(\omega) = 3k_B\mathcal{T}^T(\omega)\zeta(\omega) \tag{24}$$

is the noise spectral density and $R_V(\omega)$ is the velocity response defined as

$$R_V(\omega) = \frac{1}{\zeta^+(\omega) - i\omega M}\,. \tag{25}$$

The Wiener-Khinchine theorem then gives the velocity autocorrelation function

$$\langle \boldsymbol{V}(t)\cdot\boldsymbol{V}(0)\rangle = \frac{1}{2\pi}\int_{-\infty}^{\infty} |R_V(\omega)|^2 C_\xi^T(\omega)e^{-i\omega t}\,d\omega, \tag{26}$$

from which the translational kinetic temperature T^V, defined in Eq. (21), follows as

$$T^V = \frac{M}{\pi}\int_0^\infty |R_V(\omega)|^2\mathcal{T}^T(\omega)\zeta(\omega)\,d\omega, \tag{27}$$

since the integrand is an even function of ω.

To further evaluate this result, we introduce into the response R_V the explicit expression for the memory kernel of a sphere translating in an incompressible fluid with no-slip boundary conditions [12]:

$$\zeta^+(\omega) = 6\pi\eta R\left[1 + (1-i)\sqrt{\frac{R^2\omega}{2\nu}} - iR^2\omega/9\nu\right]. \tag{28}$$

The first term in the brackets is the usual Stokes friction $\zeta^+(\omega = 0) \equiv \zeta$, the second describes the vorticity diffusion and gives rise to the long-time tails [20,21]. The third term accounts for the mentioned mass renormalization [Eq. (20)]. With the notation $x^2 = \omega/\omega_f$, Eq. (27) now reads

$$T^V = \frac{1}{\pi}\int_0^\infty \frac{4\alpha x(x+1)\mathcal{T}^T(x)}{(1+x)^2 + x^2(1+\alpha x)^2}\,dx, \tag{29}$$

which depends on the particle-to-fluid density ratio via the parameter $\alpha \equiv 2(2\varrho_p/\varrho + 1)/9$. The same procedure gives the rotational kinetic temperature

$$T^\Omega = \frac{1}{\pi}\int_0^\infty \frac{12\beta x(1+2x+2x^2)(3+6x+6x^2+2x^3)\mathcal{T}^R(x)}{\{(3+6x+6x^2+2x^3)^2 + x^4[2(1+x)+3\beta(1+2x+2x^2)]^2\}}\,dx \tag{30}$$

with $\beta \equiv 2\varrho_p/(15\varrho)$.

Equations (29) and (30) can be integrated numerically using the translational and rotational noise temperatures $\mathcal{T}(\omega)$ introduced in Sec. III. The results are shown in Fig. 2. The kinetic temperatures are seen to depend on the density ratio ϱ_p/ϱ. To understand this, consider a translating sphere. In the Markov limit, its velocity relaxes within the Stokes time, corresponding to the relaxation rate

$$\omega_p \equiv \frac{\zeta}{M} = \frac{6\pi\eta R}{M} = \frac{9\varrho}{4\varrho_p}\omega_f, \tag{31}$$

introduced in Eq. (1). The density ratio thus relates the characteristic time for the kinematic equilibration of the particle with the fluid, i.e., the time it takes to spread the particle momentum to a fluid mass comparable to the particle mass, to the time it takes to spread its momentum to a fluid volume comparable to the particle volume. Accordingly, the kinematic equilibration affects either a small or large fluid volume compared to the particle size, suggesting a kinetic temperature close to the temperature T_s at the particle surface or close to the stationary effective temperature T^X, respectively.

Indeed, if $\varrho_p/\varrho \ll 1$, only the upper part of the spectrum $\mathcal{T}(\omega)$ contributes to the kinetic temperatures, as seen from Eqs. (29) and (30), where the integrand contributes significantly only for $x \gg 1$. Hence, the rotational kinetic temperature T^Ω approaches the surface temperature:

$$T^\Omega \sim \mathcal{T}^R(\infty) = T_0 + \Delta T = T_s \quad \text{for } \varrho_p/\varrho \to 0. \tag{32}$$

Due to the mass renormalization [Eq. (20)] the translational kinetic temperature T^V always remains somewhat below this limit, though. Although the noise temperature attempts to shake the particle with a strength proportional to the surface temperature T_s, the particle cannot move without exciting a long-range flow field that ultimately increases its own inertia. This effect limits the velocity fluctuations of the particle to a nonuniversal apparent "equipartition" temperature T^V that depends on the density ratio ϱ_p/ϱ and attains the limit

$$T^V \simeq T_0 + 0.86\,\Delta T < T_s \quad \text{for } \varrho_p/\varrho \to 0. \tag{33}$$

As a consequence, the translational particle velocity never thermalizes to the fluid temperature at the particle surface.

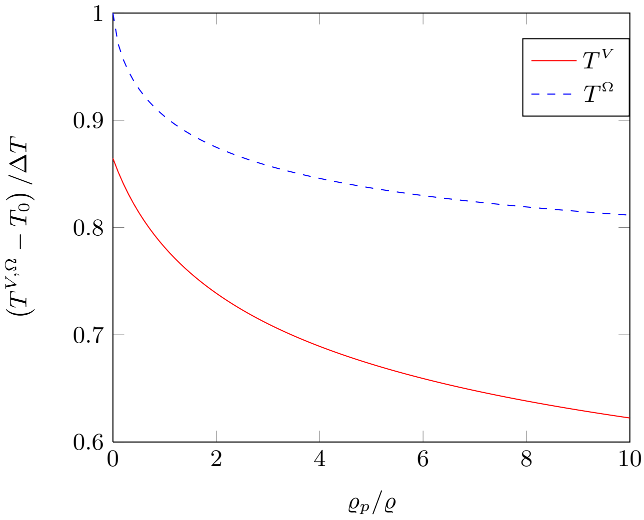

FIG. 2. (Color online) Rotational (blue/top line) and translational (red/bottom line) kinetic temperature as function of the density ratio ϱ_p/ϱ.

In the opposite limit, $\varrho_p/\varrho \gg 1$, the frequency-dependent terms in Eq. (28), which are proportional to $R^2\omega_p/\nu = 2\omega_p/\omega_f \ll 1$, become small. In this limit, the kinetic temperature approaches the stationary values of the respective effective noise temperatures $\mathcal{T}(0)$, which coincide with the known temperatures for the configurational degrees of freedom, represented by the positional and orientational coordinates X and Θ [6,7] (see Sec. III). They determine the translational and rotational diffusion coefficient of the hot Brownian particle, e.g., for translation,

$$\begin{aligned} D &= \lim_{t\to\infty} \frac{1}{6}\frac{d}{dt}\langle[\boldsymbol{X}(t) - \boldsymbol{X}(0)]^2\rangle \\ &= \frac{1}{2}\int_{-\infty}^{\infty} \langle \boldsymbol{V}(t)\cdot\boldsymbol{V}(0)\rangle\,dt = \frac{1}{2}C_V(\omega = 0). \end{aligned} \tag{34}$$

Using Eqs. (23)–(25) and (28), we recover (to leading order in the temperature increment ΔT, i.e., not accounting for the temperature-induced spatial variations in the viscosity) the generalized Einstein relation [6]

$$D_{\mathrm{HBM}} = \left.\frac{k_B\mathcal{T}^T(\omega)}{\zeta^+(\omega)}\right|_{\omega=0} = \frac{k_B(T_0 + \frac{5}{12}\Delta T)}{6\pi\eta R}. \tag{35}$$

The same reasoning applies to the orientation Θ. Hence, we see that for a hot Brownian particle that is much denser than the solvent, the kinetic temperatures reduce to the effective configurational temperatures,

$$T^{V,\Omega} \sim \mathcal{T}^{T,R}(0) = T^{X,\Theta} \quad \text{for } \varrho_p/\varrho \to \infty. \tag{36}$$

Moreover, in any case, both the translational and rotational velocities of a hot spherical particle can be statistically characterized by a (nonuniversal) Maxwell-Boltzmann distribution

$$P(\boldsymbol{V},\boldsymbol{\Omega}) \propto \exp\left(-\frac{M\boldsymbol{V}^2}{2k_B T^{V}} - \frac{I\boldsymbol{\Omega}^2}{2k_B T^{\Omega}}\right) \tag{37}$$

with effective temperatures that depend on the density ratio ϱ_p/ϱ, in agreement with the fact that probability distributions of nonequilibrium ensembles explicitly depend on the dynamics of the system.

V. PARTICLE IN A HARMONIC POTENTIAL

The discussion of the previous section can be repeated for a particle trapped in a harmonic potential. While the kinetic temperature of a free particle is determined by the competition between the vorticity diffusion time ω_f^{-1} and the Stokes relaxation time ω_p^{-1} introduced in Eq. (1), a sufficiently narrow confining potential introduces an additional interfering time scale. In the following, we examine more closely the case of translational diffusion in confinement, but qualitatively similar results can be derived for the rotational case. The parabolic confinement potential $\mathcal{U}(\boldsymbol{X}) = K\boldsymbol{X}^2/2$ gives rise to the trap relaxation time

$$\omega_t^{-1} = 6\pi\eta R/K = \omega_p/\omega_0^2, \tag{38}$$

where $\omega_0^2 = K/m$ is the undamped oscillation frequency. With $\boldsymbol{F}_e = -K\boldsymbol{X}$, the Fourier-transformed Eq. (12),

$$-m\omega^2\boldsymbol{X}(\omega) = i\omega\zeta^+(\omega)\boldsymbol{X}(\omega) - K\boldsymbol{X}(\omega) + \boldsymbol{\xi}(\omega),$$

yields the spectral density

$$C_x(\omega) \equiv \langle \boldsymbol{X}(\omega)\cdot\boldsymbol{X}(-\omega)\rangle = |R_x(\omega)|^2 C_\xi(\omega), \tag{39}$$

where the positional response function is defined by

$$R_x(\omega) = \frac{1}{m\left(\omega_0^2-\omega^2\right) - i\omega\zeta^+(\omega)}. \tag{40}$$

We use the relation

$$\langle \boldsymbol{V}(t)\cdot\boldsymbol{V}(0)\rangle = -\frac{d^2}{dt^2}\langle \boldsymbol{X}(t)\cdot\boldsymbol{X}(0)\rangle$$

between the stationary correlation functions for position and velocity in frequency space, $C_V(\omega) = \omega^2 C_X(\omega)$. The kinetic temperature, as defined in Eq. (21), follows as

$$\begin{aligned} T^{V} &= \frac{M}{\pi}\int_0^\infty \omega^2|R_x(\omega)|^2\mathcal{T}^T(\omega)\zeta(\omega)\,d\omega \\ &= \frac{1}{\pi}\int_0^\infty \frac{4\alpha x^5(x+1)\mathcal{T}^T(x)}{x^4(1+x)^2+(x^3+\alpha x^4-\omega_t/\omega_f)^2}\,dx. \end{aligned} \tag{41}$$

The result is again integrated numerically and depicted in Fig. 3. Clearly, if $\omega_t \ll \omega_f$, which means that the potential is not effective while the velocity is relaxing, we recover the result for free diffusion [Eq. (29)]. This should be the case for an optically trapped nanoparticle in water under standard experimental conditions. Indeed, for a gold particle with $R \simeq 100$ nm, assuming a trap stiffness $K = 10^{-6}$ Nm^{-1} [22], we estimate $\omega_t/\omega_f \simeq 10^{-2}$. The velocity relaxation time decreases as we increase the ratio ω_t/ω_f, resulting in a higher kinetic temperature. When $\omega_t \gg \omega_f$ the narrow confinement eventually overrides the inertia of the particle motion due to its effective mass M, so that the kinetic temperature T^{V} approaches the surface temperature T_s. Similarly as for the particle velocity, we define the positional temperature of a hot Brownian particle in a harmonic potential via the generalized equipartition theorem,

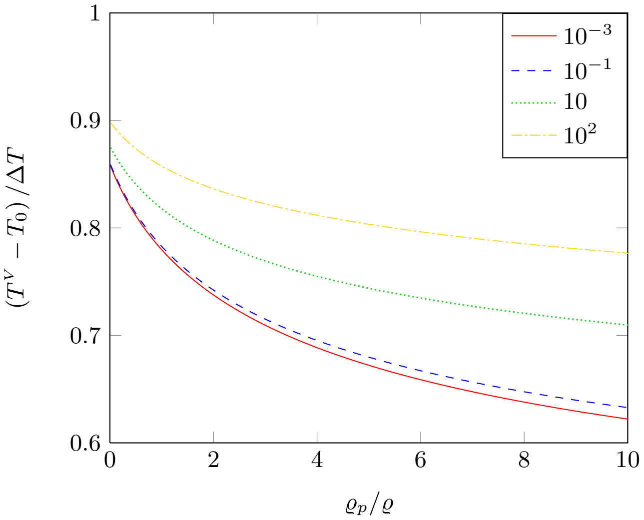

FIG. 3. (Color online) Kinetic temperature T^{V} of a particle in harmonic confinement, as given by Eq. (41) for various $\omega_t/\omega_f = 10^{-3},\ldots,10^2$. For $\omega_t/\omega_f \lesssim 10^{-1}$, T^{V} is hardly distinguishable from the kinetic temperature of a free particle.

$$\tfrac{3}{2}k_B T^{X} \equiv \tfrac{1}{2}\omega_0^2 m\langle \boldsymbol{X}^2\rangle, \tag{42}$$

where the average is taken with respect to the stationary distribution. Using Eqs. (39) and (40) we straightforwardly obtain

$$\begin{aligned} T^{X} &= \frac{\omega_0^2 m}{\pi}\int_0^\infty |R_x(\omega)|^2\mathcal{T}^T(\omega)\zeta(\omega)\,d\omega \\ &= \frac{1}{\pi}\int_0^\infty \frac{4(\omega_t/\omega_f)x(x+1)\mathcal{T}^T(x)}{x^4(1+x)^2+(x^3+\alpha x^4-\omega_t/\omega_f)^2}\,dx. \end{aligned} \tag{43}$$

This result is integrated numerically and plotted in Fig. 4. Again, if $\omega_t \ll \omega_f$, we recover the configurational temperature of a free particle, since the integrand in Eq. (43) is sharply peaked at $x \ll 1$, corresponding to $\omega \ll \omega_f$. Physically, the relaxation in the potential takes place quasistatically with respect to the free hot Brownian motion, which can then be represented in the Markov approximation, in perfect analogy to the equilibrium case. The corresponding Langevin equation is

$$\zeta\dot{\boldsymbol{X}} = -\nabla\mathcal{U} + \boldsymbol{\xi}, \qquad \langle \xi_i(t)\xi_j(t')\rangle = 2D_{\mathrm{HBM}}\delta(t-t')\delta_{ij},$$

and its stationary solution is the generalized Boltzmann distribution

$$P(\boldsymbol{X}) \propto \exp\left(-\frac{\mathcal{U}(\boldsymbol{X})}{k_B T^{X}}\right).$$

with the effective temperature

$$T^{X} = \zeta D_{\mathrm{HBM}} = T_0 + \tfrac{5}{12}\Delta T \tag{44}$$

of free hot Brownian motion [6] (originally denoted by T_{HBM}).

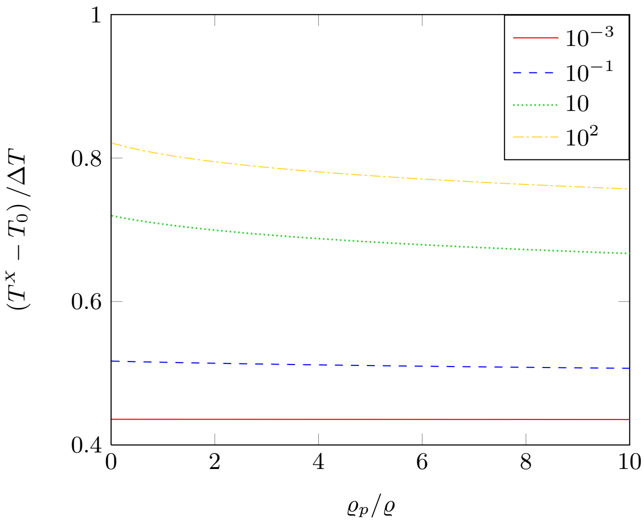

FIG. 4. (Color online) Positional temperature T^x of a particle in harmonic confinement, as given by Eq. (43) for various $\omega_t/\omega_f = 10^{-3},\ldots,10^2$. At small ω_t/ω_f the temperature is independent of ϱ_p/ϱ.

In contrast, if $\omega_t \approx \omega_p$ the potential interferes with the relaxation of the particle, resulting in a higher T^x than in the free case. Eventually, in the extreme limit $\omega_t \gg \omega_p$, the integral peaks near ω_t, and T^x approaches the kinetic temperature T^v (nonuniformly in ϱ_p/ϱ). It is moreover worth noting that the stationary probability distribution can in any case still be written in the form of Eq. (37), albeit with nonuniversal temperatures T^v and T^x that generally depend on the density ratio ϱ_p/ϱ and on the stiffness K of the potential. Analogous conclusions hold for the rotational degrees of freedom.

VI. CONCLUSION

Starting from the fluctuating hydrodynamic description of the solvent, which we required to be in local thermal equilibrium with an inhomogeneous temperature field $T(\boldsymbol{r})$, we have derived a generalized Langevin equation for the motion of a Brownian suspended particle. While the discussion was limited to the important case of hot Brownian motion, where $T(\boldsymbol{r})$ decays radially around the particle, essentially the same reasoning applies to more general temperature profiles [9]. As a consequence of the nonisothermal conditions, the noise temperature $\mathcal{T}(\omega)$ characterizing the strength of the stochastic Langevin forces becomes frequency dependent and differs between different degrees of freedom, which couple to different hydrodynamic modes. We remark that the frequency-dependent noise temperature is not merely defined, as it is customary, through the violation of equilibrium ensemble properties of the particle position and velocity, e.g., the broken proportionality between response and equilibrium correlation functions expressed by the (first-kind) fluctuation-dissipation theorem [23,24]. Rather, $\mathcal{T}(\omega)$ is derived upon contraction of the underlying stochastic description and its physical origin is fully understood in terms of nonequilibrium hydrodynamic fluctuations of the viscous solvent.

From the noise temperature we derived approximate expressions for the effective temperatures at which the rotational and translational degrees of freedom of a spherical particle appear to thermalize. Explicit numerical results have been limited to first order in the temperature increment ΔT, so that the temperature dependence of the fluid viscosity could be neglected. We found the (angular) velocities to be Maxwell-Boltzmann distributed with nonuniversal, but explicitly known, effective temperatures. The violation of the equilibrium (second-kind) fluctuation-dissipation theorem given by Eq. (19), i.e., the broken proportionality between noise correlation and memory kernel, is ultimately the reason why effective temperatures appear in the equilibrium-like distributions of the particle position and velocity [9]. In the long-time limit we regained previous results for the configurational temperatures governing free and weakly confined hot Brownian motion.

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APPENDIX A: DERIVATION OF THE GLE's NOISE AUTOCORRELATION FUNCTION

Extending the calculation presented in Ref. [13] to a nonisothermal solvent, we derive expressions (14) and (15) in Sec. II for the translational motion. The same procedure can be applied separately to rotational motion bearing in mind that, due to linearity and spherical symmetry, the flow field $\boldsymbol{v}$ can be divided into the two independent fields $\boldsymbol{v}^T$ and $\boldsymbol{v}^R$ generated, respectively, by the particle translation and rotation, and satisfying the boundary conditions:

$$\boldsymbol{v}^T(\boldsymbol{r},t) = \boldsymbol{V}(t) \quad \text{on } \mathcal{S},$$
$$\boldsymbol{v}^R(\boldsymbol{r},t) = \boldsymbol{\Omega}(t)\times\boldsymbol{r} \quad \text{on } \mathcal{S}.$$

Since we focus on the translational motion only, we omit the superscript T. Using Eq. (10), the Fourier transform of the generalized Langevin equation (12) reads

$$-i\omega m\boldsymbol{V}(\omega) = -\zeta^+(\omega)\boldsymbol{V}(\omega) + \boldsymbol{\xi}(\omega) + \boldsymbol{F}_e(\omega)$$

and may be rewritten as

$$-i\omega m\boldsymbol{V}(\omega) = \boldsymbol{f}(\omega) + \tilde{\boldsymbol{f}}(\omega) + \boldsymbol{F}_e(\omega), \tag{A1}$$

where we have divided the force exerted by the fluid into deterministic $\boldsymbol{f}(\omega)$ and random $\tilde{\boldsymbol{f}}(\omega)$ components:

$$\boldsymbol{f}(\omega) \equiv -\zeta^+(\omega)\langle\boldsymbol{V}(\omega)\rangle, \tag{A2a}$$
$$\tilde{\boldsymbol{f}}(\omega) \equiv -\zeta^+(\omega)\tilde{\boldsymbol{V}}(\omega) + \boldsymbol{\xi}(\omega)\ , \tag{A2b}$$

with $\boldsymbol{V} \equiv \langle\boldsymbol{V}\rangle + \tilde{\boldsymbol{V}}$. It is easy to see that $\boldsymbol{f}(\omega)$ is the force exerted by the deterministic flow field $\boldsymbol{u} \equiv \langle\boldsymbol{v}\rangle$, the solution of

$$i\omega\varrho\boldsymbol{u}(\boldsymbol{r},\omega)+\nabla\cdot\boldsymbol{\sigma}(\boldsymbol{r},\omega)=0, \tag{A3a}$$

$$\nabla\cdot\boldsymbol{u}(\boldsymbol{r},\omega)=0, \tag{A3b}$$

$$\boldsymbol{u}(\boldsymbol{r},\omega)=\langle\boldsymbol{V}(\omega)\rangle \quad \text{on } \mathcal{S}, \tag{A3c}$$

while $\tilde{\boldsymbol{f}}(\omega)$ is the force exerted by the stochastic flow field $\tilde{\boldsymbol{u}}\equiv\boldsymbol{v}-\langle\boldsymbol{v}\rangle$, the solution of

$$i\omega\varrho\tilde{\boldsymbol{u}}(\boldsymbol{r},\omega)+\nabla\cdot\tilde{\boldsymbol{\sigma}}(\boldsymbol{r},\omega)=-\nabla\cdot\boldsymbol{\tau}(\boldsymbol{r},\omega), \tag{A4a}$$

$$\nabla\cdot\tilde{\boldsymbol{u}}(\boldsymbol{r},\omega)=0, \tag{A4b}$$

$$\tilde{\boldsymbol{u}}(\boldsymbol{r},\omega)=\tilde{\boldsymbol{V}}(\omega) \quad \text{on } \mathcal{S}. \tag{A4c}$$

This splitting of equations and boundary conditions is again allowed by the linearity of the problem. In the following calculation, in order to ease the notation, we omit the arguments $\boldsymbol{r}$ and ω of the hydrodynamic fields where there is no possibility of confusion.

We start by calculating twice the energy dissipated by the particle moving at velocity $\langle\boldsymbol{V}(\omega)\rangle$:

$$\begin{aligned}
\langle V_i(\omega)\rangle[\zeta^+(\omega)+\zeta^{+*}(\omega)]\langle V_i^*(\omega)\rangle &\overset{(\mathrm{A2a})}{=} -[f_i(\omega)\langle V_i^*(\omega)\rangle+f_i^*(\omega)\langle V_i(\omega)\rangle]\\
&\overset{(6)}{=} \langle V_i^*(\omega)\rangle\int_{\mathcal{S}}\sigma_{ij}n_j\,d^2r+\langle V_i(\omega)\rangle\int_{\mathcal{S}}\sigma_{ij}^*n_j\,d^2r\\
&\overset{(\mathrm{A3c})}{=} \int_{\mathcal{S}}u_i^*\sigma_{ij}n_j\,d^2r+\int_{\mathcal{S}}u_i\sigma_{ij}^*n_j\,d^2r \qquad\qquad (\mathrm{A5})\\
&= \int_{\mathcal{V}}\partial_j(u_i^*\sigma_{ij})\,d^3r+\int_{\mathcal{V}}\partial_j(u_i\sigma_{ij}^*)\,d^3r\\
&\overset{(\mathrm{A3a})}{=} \int_{\mathcal{V}}(\sigma_{ij}\partial_ju_i^*+\sigma_{ij}^*\partial_ju_i)\,d^3r\\
&\overset{(\mathrm{A3b})}{=} \int_{\mathcal{V}}2\eta(\Gamma_{ij}\partial_ju_i^*+\Gamma_{ij}^*\partial_ju_i)\,d^3r\\
&= 2\int_{\mathcal{V}}\phi(\boldsymbol{r},\omega)\,d^3r, \qquad\qquad (\mathrm{A6})
\end{aligned}$$

where in Eq. (A5) we employed the divergence theorem and in Eq. (A6) we defined the dissipation function:

$$\phi(\boldsymbol{r},\omega)\equiv\eta(\boldsymbol{r})\left(\partial_iu_j\partial_iu_j^*+\partial_iu_j\partial_ju_i^*\right)(\boldsymbol{r},\omega).$$

Since $\zeta(\omega)=2\Re e\zeta^+(\omega)=\zeta^+(\omega)+\zeta^{+*}(\omega)$, being $\zeta(t)$ real and time symmetric, we can rewrite Eq. (A6) in the following form:

$$\zeta(\omega)\delta_{ij}\langle V_i(\omega)\rangle\langle V_j^*(\omega)\rangle=2\int_{\mathcal{V}}\phi(\boldsymbol{r},\omega)\,d^3r. \tag{A7}$$

This proves Eq. (14). We proceed with the evaluation of the energy supplied by the random force $\boldsymbol{\xi}(\omega)$:

$$\begin{aligned}
\xi_i(\omega)\langle V_i(\omega)\rangle &\overset{(\mathrm{A2b})}{=} [\tilde{f}_i(\omega)+\zeta^+(\omega)\tilde{V}_i(\omega)]\langle V_i(\omega)\rangle\\
&\overset{(\mathrm{A2a})}{=} \tilde{f}_i(\omega)\langle V_i(\omega)\rangle-f_i(\omega)\tilde{V}_i(\omega)\\
&\overset{(6)}{=} -\langle V_i(\omega)\rangle\int_{\mathcal{S}}\tilde{\sigma}_{ij}n_j\,d^2r+\tilde{V}_i(\omega)\int_{\mathcal{S}}\sigma_{ij}n_j\,d^2r\\
&\overset{(\mathrm{A3c})\,(\mathrm{A4c})}{=} -\int_{\mathcal{S}}u_i\tilde{\sigma}_{ij}n_j\,d^2r+\int_{\mathcal{S}}\tilde{u}_i\sigma_{ij}n_j\,d^2r \qquad\qquad (\mathrm{A8})\\
&= -\int_{\mathcal{V}}\partial_j(u_i\tilde{\sigma}_{ij})\,d^3r+\int_{\mathcal{V}}\partial_j(\tilde{u}_i\sigma_{ij})\,d^3r\\
&\overset{(\mathrm{A3a})\,(\mathrm{A4a})}{=} -\int_{\mathcal{V}}\tilde{\sigma}_{ij}\partial_ju_i\,d^3r+\int_{\mathcal{V}}\sigma_{ij}\partial_j\tilde{u}_i\,d^3r+\int_{\mathcal{V}}u_i\partial_j\tau_{ij}\,d^3r\\
&\overset{(\mathrm{A3b})\,(\mathrm{A4b})}{=} \int_{\mathcal{V}}u_i\partial_j\tau_{ij}\,d^3r=-\int_{\mathcal{V}}\tau_{ij}\partial_ju_i\,d^3r
\end{aligned}$$

In Eq. (A8) we made use again of the divergence theorem. Summing up,

$$\xi_i(\omega)\langle V_i(\omega)\rangle=-\int_{\mathcal{V}}\tau_{ij}(\boldsymbol{r},\omega)\partial_ju_i(\boldsymbol{r},\omega)\,d^3r, \tag{A9}$$

which asserts that $\boldsymbol{\xi}(\omega)$ is Gaussian with vanishing mean, being the integral of the deterministic quantity $\partial_j u_i$ times the zero-mean Gaussian field τ_{ij}. Using Eq. (A9) we evaluate the noise correlation function:

$$\begin{aligned}\langle \xi_i(\omega)\xi_j^*(\omega')\rangle\langle V_i(\omega)\rangle\langle V_j^*(\omega')\rangle &= \int_{\mathcal{V}} d^3r' \int_{\mathcal{V}} d^3r\, \partial_j u_i(\boldsymbol{r},\omega)\langle \tau_{ij}(\boldsymbol{r},\omega)\tau_{kl}^*(\boldsymbol{r}',\omega')\rangle \partial_l u_k^*(\boldsymbol{r}',\omega')\\ &= 2k_{\mathrm{B}}\delta(\omega-\omega')\int_{\mathcal{V}} \eta\left(\partial_i u_j \partial_i u_j^* + \partial_i u_j \partial_j u_i^*\right) T\, d^3r\\ &= 2k_{\mathrm{B}}\delta(\omega-\omega')\int_{\mathcal{V}} \phi(\boldsymbol{r},\omega)T(\boldsymbol{r})\, d^3r. \end{aligned} \tag{A10}$$

In Eq. (A10) we used the Fourier transform of Eq. (4) together with $\tau_{kl}^*(\boldsymbol{r}',\omega') = \tau_{kl}(\boldsymbol{r}',-\omega')$, since $\boldsymbol{\tau}$ is real. This proves Eq. (15).

APPENDIX B: HYDRODYNAMICS OF A TRANSLATION AND ROTATING SPHERE

1. Translational motion

The Fourier transform of the flow field generated by a sphere translating with velocity $\langle V(\omega)\rangle \boldsymbol{e}_z$ reads in polar coordinates (r,φ,θ) [11, p. 623]:

$$\boldsymbol{u}^T(r,\theta,\omega) = \frac{1}{r}\left[\sin\theta\left(g + r\frac{dg}{dr}\right)\boldsymbol{e}_\theta - 2g\cos\theta\,\boldsymbol{e}_r\right],$$

with

$$g(r,\omega) = \frac{3\langle V(\omega)\rangle R}{2(kr)^2}\left\{(ikr-1)\,e^{ik(r-R)} - \left[1 + ikR - \frac{1}{3}(kR)^2\right]\right\},$$

where $k = (1+i)k_0$, and $k_0 = \sqrt{\omega/2\nu}$ is the inverse of the characteristic fluid diffusion length. The associated dissipation function is

$$\phi^T = \eta\left(\frac{12}{r^4}\cos^2\theta\left|g - r\frac{dg}{dr}\right|^2 + \sin^2\theta\left|\frac{d^2g}{dr^2}\right|^2\right),$$

that becomes after integration over θ:

$$\begin{aligned}&\int_0^\pi \phi^{\mathrm{T}}(r,\theta,\omega)\sin\theta\, d\theta\\ &= \frac{3\eta|\langle V(\omega)\rangle|^2R^2}{2k_0^4r^8}\{5[9 + 2k_0R(9 + k_0R(9 + 2k_0R(3 + k_0R)))]\\ &\quad + e^{-2k_0(r-R)}[45 + 2k_0r[45 + k_0r(45 + k_0r(30 + k_0r(15 + 2k_0r(3 + k_0r))))]]\\ &\quad - 2e^{-k_0(r-R)}\big[\big(45 + 45k_0R + 15k_0(3 + 2k_0R(3 + k_0R))r + 12k_0^3R(3 + 2k_0R)r^2 + 2k_0^3(-3 + 2k_0^2R^2)r^3\big)\cos[k_0(R-r)]\\ &\quad - k_0\big(-15R(3 + 2k_0R) + 15\big(3 - 2k_0^2R^2\big)r + 36k_0(1 + k_0R)r^2 + 2k_0^2(3 + 2k_0R(3 + k_0R))r^3\big)\sin[k_0(R-r)]\big]\big\}. \end{aligned} \tag{B1}$$

Notice that Eq. (B1) displays a term which does not decay with an exponential cutoff but only algebraically as $1/r^8$. But its contribution to $\mathcal{T}(\omega)$ actually diminishes at high frequencies, $k_0 \to \infty$. In order to obtain $\mathcal{T}(\omega)$ we numerically integrate Eq. (18) together with Eq. (B1). The result is shown in Fig. 1.

2. Rotational motion

The Fourier transform of the flow field generated by a sphere rotating with angular velocity $\langle \Omega(\omega)\rangle \boldsymbol{e}_z$ reads [10, p. 91]

$$\boldsymbol{u}^R(r,\theta,\omega) = \frac{\langle\Omega(\omega)\rangle R^3}{r^2}\sin\vartheta\,\frac{1-ikr}{1-ikR}e^{i[k(r-R)]}\boldsymbol{e}_\varphi \equiv f(r,\vartheta,\omega)\boldsymbol{e}_\varphi.$$

The associated dissipation function $\phi^R(\boldsymbol{r},\omega)$ is

$$\begin{aligned}\phi^R &= \frac{\eta}{r^2}(|r\partial_r f - f|^2 + |\partial_\vartheta f - \cot\vartheta f|^2)\\ &= \frac{\eta|\langle\Omega(\omega)\rangle|^2R^6[9 + 18k_0r + 18(k_0r)^2 + 12(k_0r)^3 + 4(k_0r)^4]\sin^2\vartheta\, e^{-2k_0(r-R)}}{r^6[1 + 2k_0R + 2(k_0R)^2]}. \end{aligned} \tag{B2}$$

Notice in Eq. (B2) the exponential cutoff where the fluid's diffusion characteristic length k_0^{-1} appears. Using Eqs. (18) and (B2) we obtain the first-order approximation in ΔT for the noise temperature of rotational motion:

$$\frac{\mathcal{T}^R(\omega) - T_0}{\Delta T} = \frac{9 + 18k_0R + 18(k_0R)^2 + 12(k_0R)^3 - 8(k_0R)^4 E_1(2k_0R)e^{2k_0R}}{4[3 + 6k_0R + 6(k_0R)^2 + 2(k_0R)^3]}, \tag{B3}$$

where $E_1(x) = \int_1^\infty dy\, e^{-xy}/y$ is the exponential integral. The result is plotted in Fig. 1.

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