The effective temperature for the thermal fluctuations in hot Brownian motion

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We revisit the effective parameter description of hot Brownian motion – a scenario where a colloidal particle is kept at an elevated temperature than the ambient fluid. Due to the time scale separation between heat diffusion and particle motion, a stationary halo of hot fluid is carried along with the particle, resulting in a spatially varying comoving temperature and viscosity profile. The resultant Brownian motion in the overdamped limit can be well described by a Langevin equation with effective parameters such as effective temperature $T_{\text{HBM}}$ and friction coefficient $\zeta_{\text{HBM}}$ that quantifies the thermal fluctuations and the diffusivity of the particle. These parameters can exactly be calculated using the framework of fluctuating hydrodynamics and requires the knowledge of the complete flow field and the temperature field around the particle. Additionally, it was also observed that configurational and the kinetic degrees of freedom admits to different effective temperatures, $T_{\text{HBM}}^x$ and $T_{\text{HBM}}^v$, respectively, with the former predicted accurately from fluctuating hydrodynamics. A more rigorous calculation by Falasco et. al. Physical Review E, 90, 032131(2014) extends the overdamped description to a generalized Langevin equation where the effective temperature becomes frequency dependent and consequently, for any temperature measurement from a Brownian trajectory requires the knowledge of this frequency dependence. We use this framework to expand on this earlier work and look at the first order correction to the limiting values in the hydrodynamic limit and the kinetic limit. We use the linearized Stokes equation and a constant viscosity approximation to calculate the dissipation function in the fluid. The effective temperature is calculated from the weighted average of the temperature field with the dissipation function. Further, we provide a closed form analytical result for effective temperature in the small as well high frequency limit. Since hot Brownian motion can be used to probe the local environment in complex systems, we have also calculated the effective diffusivity of the particle in the small frequency limit. To look into the kinetic temperature, the velocity autocorrelation function is computed from the generalized Langevin equation and the Wiener-Khinchine theorem and numerically integrated to evaluate $T_{\text{HBM}}^v$ as a function of the ratio of particle density and fluid density $\rho_P/\rho_0$. The two limiting cases of $\rho_P/\rho_0 \rightarrow 0$ and $\rho_P/\rho_0 \rightarrow \infty$ is also discussed.

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I. INTRODUCTION

Nonequilibrium scenarios of an equilibrium physical phenomena often serve as a testing field for the equilibrium theories and their limits of validity. Often, the description of such a nonequilibrium counterpart is done within the framework of the equilibrium description using renormalized parameters for the equilibrium theory. Perhaps, the most popular system which has garnered an unfettered attention from the scientific community is that of Brownian motion, due to its ubiquitous existence in the mesoscopic world and its nonequilibrium generalization. One such scenario is that of hot Brownian motion\textsuperscript{2, 3}. In hot Brownian motion a colloid is kept at a higher temperature than the ambient fluid. Since heat diffusion happens at faster time scale as compared to the particle diffusion, the heating of the colloid results in a stationary temperature profile which is comoving with the colloid itself. In a fluid, the resulting spatially varying temperature profile leads to a spatially varying viscosity profile. The ensuing motion is still diffusive, but with an enhanced diffusion coefficient. An attempt to describe this enhanced diffusive motion using the coarse grained Langevin picture requires the use of renormalized parameters to quantify the thermal fluctuations in the system. Even though these parameters can be viewed as phenomenological quantities that is used to simplify the underlying dynamics, nevertheless they can be rigorously estimated from the hydrodynamical description of the motion\textsuperscript{2, 3}.

The cost that we pay in having an effective description is that there is not one single temperature that quantifies the thermal fluctuations. The different degrees of motion, as well as different modes of motion have different effective temperature. Consequently, a set of these effective temperature $T_{\text{HBM}}$ is used for describing the rotational and translational motion of a heated colloid, as well as for the configurational and kinetic degrees of freedom for the two modes of motion. Consequently, $T_{\text{HBM}}^x$ enters the generalized Stokes Einstein relation and the Boltzmann distribution in the presence of an external potential $U(x)$:\textsuperscript{4}

\[ D_{\text{HBM}} = k_B T_{\text{HBM}}^x / \zeta_{\text{HBM}} \quad \text{and} \quad P(x) \sim e^{-U(x)/k_B T_{\text{HBM}}^x} \]

(1)

and $T_{\text{HBM}}^v$ defines the width of the velocity fluctuations $V$ of the heated colloid with mass $M$,

\[ P(V) \sim e^{-MV^2/2k_B T_{\text{HBM}}^v} \quad \text{and} \quad \langle V^2 \rangle = \frac{3k_B T_{\text{HBM}}^v}{M} \]

(2)

The more general approach is to interpret this effective temperature as a frequency dependent quantity $T'(\omega)$ that together with Basset-Boussinesq force quantifies the thermal fluctuations acting in the colloid.

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In this article we take a look at this effective parameter $T(\omega)$ and using the framework of fluctuating hydrodynamics developed earlier we derive the limiting behavior of the quantity $T(\omega)$ for an incompressible solvent. In the earlier work by Falsaco et. al., the overdamped description of hot Brownian motion was formally extended to its generalized form

$$\mathbf{P} = -\frac{1}{M} \int_0^t \zeta(t-t') \mathbf{P}(t') + \xi$$

with the effective parameters inheriting the frequency dependence. The non-equilibrium thermal fluctuations for the Brownian particle can now be written as:

$$\langle \xi_i(\omega) \xi_j(\omega') \rangle = 2k_B T(\omega) \Re \epsilon(\omega) \delta_{ij} \delta(\omega + \omega')$$

where $\zeta(\omega)$ is the effective friction coefficient which can be determined from the solution to the Stokes equation.

The current work is motivated by the fact that the limiting behavior of the frequency dependent temperature can be used in nonequilibrium thermometry. Furthermore, in experiments as well as in computer simulations the finite time resolution puts an upper cutoff on the frequency of sampling and therefore the correct limiting values are not accessible. Hence, it becomes essential to have a closed form analytic expression for the effective temperature in the limit of $\omega \to 0$ and $\omega \to \infty$. Using the framework of hot Brownian motion, we calculate the frequency dependent dissipation function in a closed form and derive the expression of $T(\omega)$ and the frequency dependent diffusivity $D(\omega)$. The expressions for these effective parameters, in particular the first order correction to $T(\omega)$ and $D(\omega)$ near $\omega \to 0$ are important in validating the coarse grained dynamics of the heated particle and its application as a spectroscopic tool. We note that the limit of $\omega \to \infty$ has the physical interpretation of frequencies larger than the inverse of the vorticity diffusion time.

We summarize the main results of our work in the following lines. The expression for the effective temperature quantifying the thermal fluctuations is given by the expression

$$\mathcal{T}(\omega) = T_0 + \frac{5}{12} \Delta T - \frac{T(\omega)}{(1 + a \sqrt{\frac{\rho_0 \omega}{2 \eta}})}$$

which in the limit of $\omega \to 0$ yields

$$\mathcal{T}(\omega) \approx T_0 + \frac{5 \Delta T}{12} \left[ 1 + a \sqrt{\frac{\rho_0 \omega}{2 \eta}} \right].$$

On the opposite limit of $\omega \to \infty$, $T(\omega)$ take the form

$$\mathcal{T}(\omega) \approx T_0 + \Delta T \left[ 1 - \frac{1}{3a} \sqrt{\frac{2 \eta}{\rho_0 \omega}} \right].$$

The frequency dependent diffusion coefficient is then evaluated near $\omega \to 0$ to yield

$$D(\omega) \approx \frac{k_B T_0}{\zeta_0} \left[ \left( 1 + \frac{5 \Delta T}{12 T_0} \right) - a \sqrt{\frac{\rho_0 \omega}{2 \eta}} \left[ 1 + \frac{5 \Delta T}{6 T_0} a \sqrt{\frac{\rho_0 \omega}{2 \eta}} \right] \right].$$

The rest of the article is organized in the following sections. In Section II, we briefly describe the route to calculate the effective temperature $T(\omega)$. In Section III, we evaluate the complete frequency dependent dissipation function for an incompressible solvent. Using the form of this dissipation function, we calculate the effective temperature for the thermal fluctuations in Section IV and the first order correction to its limiting value as $\omega \to 0$ in Section IV A. In order to make our results more transparent in the limit of $\omega \to \infty$, we approximate the flow field and the dissipation function in the high frequency regime and estimate the asymptotic behavior of the effective temperature as $\omega \to \infty$ in Section IV B. In Section V we evaluate the kinetic temperature $\mathcal{T}_{BBM}$ as a function of the ratio of the density of the particle to that of the fluid and the effective frequency dependent diffusivity in Section VI. Finally, we briefly discuss the significance of our results in Section VII.

II. HYDRODYNAMICS

In a hydrodynamic model of Brownian motion, the fluid is usually treated as a continuum that obeys the linearized Stokes equation

$$\rho \partial_t \mathbf{u} = \nabla \cdot \Pi$$

where the isotropic part of the stress tensor $\Pi$ is given by the pressure and the deviatoric part is given by the strain

$$\Pi_{ij} = -\rho \delta_{ij} + \tau_{ij} \quad \text{with} \quad \tau_{ij} = 2\eta \epsilon_{ij},$$

where $\epsilon_{ij} = (\partial_i u_j + \partial_j u_i)/2$. The notation $\partial_i$ and $\partial_t$ denotes a partial derivative with respect to the spatial coordinate $x_i$ and time $t$, respectively. The dynamics of the colloid is coupled to the fluid via the boundary condition on its surface. In what follows, we assume this to be a perfect no-slip boundary condition. Far away from the colloid the fluid is assumed stationary. The linearized Stokes equation is usually further augmented by the incompressibility condition $\nabla \cdot \mathbf{u} = 0$, and is typically valid in the limit of $\omega \to 0$. Eq. (9) can be exactly solved for the systematic part of the flow field for any arbitrary velocity $U(t)$ prescribed on the surface $S$ of the colloid. In order to include thermal fluctuations in the system, the hydrodynamical fields are split into a systematic and a fluctuating part, $\mathbf{u} = \mathbf{u}_s + \tilde{\mathbf{u}}$, $p = p_s + \tilde{p}$ and $\Pi = \Pi_s + \tilde{\Pi}$. The linearity of Eq. (9) ensures that the systematic and the fluctuating parts decouple. The fluctuating flow field $\mathbf{u}(\mathbf{r}, t)$ is driven by the fluctuating stresses $\tilde{\tau}_{ij}$. The strength of the fluctuations is determined by the viscosity of the solvent and the temperature via the fluctuation–dissipation relation (FDT). In an incompressible solvent the FDT reads as

$$\langle \tilde{\tau}_{ij}(\mathbf{r}, t) \tilde{\tau}_{kl}(\mathbf{r}', t') \rangle = 2k_B T \delta(\mathbf{r} - \mathbf{r}') \delta(t - t') \left[ \delta_{ij} \delta_{kl} + \delta_{il} \delta_{kj} \right].$$

In hot Brownian motion, even though there exists a radial temperature and viscosity field due to the heating of the colloid, the local thermal equilibrium in the fluid allows the modification of the FDT to include the local temperature $T(\mathbf{r})$ and the viscosity $\eta(\mathbf{r})$ to quantify the strength of the fluctuating stresses.
The solvent degrees of freedom can now be contracted to yield the equation of motion for the colloid. A rigorous calculation then yields the expression for the effective parameters of the coarse grained theory. In particular, the effective temperature that quantifies the thermal fluctuations for the coarse grained Langevin dynamics is given by weighted average of $T(r, t)$

$$
T(\omega) = \frac{\int T(r, \omega) \phi(r, \omega) d\mathbf{r}}{\int \phi(r, \omega) d\mathbf{r}}
$$

(12)

where $\phi(r, t)$ is the dissipation function in the fluid, which for an incompressible solvent reads:

$$
\phi(r) = \frac{\eta}{2} [\nabla \mathbf{u}(r) + (\nabla \mathbf{u}(r))^\top] : [\nabla \mathbf{u}^*(r) + (\nabla \mathbf{u}^*(r))^\top],
$$

(13)

where $u^*$ denotes the complex conjugate of $u$ and $(\nabla \mathbf{u}(r))^\top$ is the transpose of $\nabla \mathbf{u}(r)$. The flow field that enters the expression for the dissipation function, is the systematic part of the flow and obeys the linearized Stokes Eq. (9).

III. DISSIPATION FUNCTION

We first turn our attention to calculate the frequency dependent dissipation function $\phi(r, \omega)$. We consider a spherical colloid of radius $a$ suspended in a solvent of density $\rho_0$, viscosity $\eta$ and at a temperature $T_0$. The colloid is now heated which results in a radially varying temperature and viscosity field in the solvent. In a comoving frame of the particle, the steady state temperature profile $T(r)$ follows the stationary heat equation

$$
\nabla^2 T(r) = 0
$$

(14)

together with the boundary conditions

$$
T(r = a) = T_0 + \Delta T \quad \text{and} \quad T(r \to \infty) = T_0.
$$

(15)

The solution of above equation is the radial field:

$$
T(r) = T_0 + \Delta T \frac{a}{r}
$$

(16)

Since the heating of the colloids results in a radially symmetric temperature field, the viscosity in the fluid no longer remains constant throughout. This spatially varying viscosity can be obtained from the equation of state $\eta(T, \rho)$. To simplify the calculations, we consider the situation when the heating of the colloid is small so that the spatial variation of the viscosity can be ignored in the Stokes equation.

Using the continuity equation $\partial \rho + \nabla \cdot \rho \mathbf{u} = 0$, it is easy to deduce that in the limit of $\omega \to 0$, the solvent can be treated as incompressible. The perturbations due to sound propagation has a characteristic frequency $c/a$ and only comes into play in the high frequency limit. Writing down the Stokes equation explicitly in the frequency space

$$
-\mathbf{i} \omega \rho_0 \mathbf{u} = -\nabla p + \eta \nabla^2 \mathbf{u}
$$

$$
\nabla \cdot \mathbf{u} = 0,
$$

(17)

we seek the frequency dependent solution to the above equation subject to the boundary conditions:

$$
\mathbf{u}(r, t) = \mathbf{U}(r, t) + \Omega(r, t) \times \mathbf{r} \quad \text{on} \quad S
$$

$$
\lim_{r \to \infty} \mathbf{u}(r, t) = 0.
$$

(18)

Without loss of generality, exploiting the spherical symmetry of the problem, we choose $\mathbf{U}(t)$ along the direction of the $z$-axis, so that $\mathbf{U}(t) = U(t) \mathbf{z}$. The solution to Eq. (17) with the boundary conditions Eq. (18) can be derived by writing down $\mathbf{u}(r, \omega)$ as

$$
\mathbf{u}(r, \omega) = \nabla \times \nabla_r (h(r) U_{\omega}).
$$

(19)

The scalar function $h(r)$ satisfies the differential equation

$$
\nabla^2 h(r) + \alpha_{\omega}^2 \nabla^2 h(r) = 0
$$

(20)

Solving for $h(r)$ and using the boundary conditions from (18) the flow field takes the form

$$
\mathbf{u}(r, \omega) = -\frac{2U_{\omega}}{r} \frac{\partial h}{\partial r} \cos \theta \mathbf{\hat{r}} + \frac{U_{\omega}}{r} \left( \frac{\partial h}{\partial r} + \frac{\partial^2 h}{\partial r^2} \right) \sin \theta \mathbf{\hat{\theta}}
$$

(21)

with $\frac{\partial h}{\partial r}$ given by

$$
\frac{\partial h}{\partial r} = \frac{3a}{2\alpha_{\omega}^2 r^2} \left[ (i \alpha_{\omega} r - 1) e^{i \alpha_{\omega} (r - a)} - (1 + i \alpha_{\omega} a + \frac{1}{3} \alpha_{\omega}^2 a^2) \right]
$$

(22)

The final form of the flow field reads:

$$
\mathbf{u}_r(\omega) = \left[ 2A_{\omega} \left( -\frac{1}{r^3} + \frac{i \alpha_{\omega}}{r^2} \right) e^{i \alpha_{\omega} r} - B_{\omega} \frac{2}{r^3} \right] U_{\omega} \cos \theta
$$

$$
\mathbf{u}_\theta(\omega) = \left[ A_{\omega} \left( -\frac{1}{r^3} + \frac{i \alpha_{\omega}}{r^2} + \frac{\alpha_{\omega}^2}{r} \right) e^{i \alpha_{\omega} r} - B_{\omega} \frac{1}{r^3} \right] U_{\omega} \sin \theta,
$$

(23)

where the frequency dependence of the quantities have been denoted by the subscript. $\alpha_{\omega}$ is given by

$$
\alpha_{\omega}^2 = \frac{i \omega \rho_0}{\eta}.
$$

(24)

In order to satisfy the boundary condition at infinity, $\alpha_{\omega}$ is chosen so that $\text{Im}(\alpha_{\omega}) > 0$,

$$
\text{Re}(\alpha_{\omega}) = \text{Im}(\alpha_{\omega}) = \sqrt{\frac{\omega \rho_0}{2 \eta}}
$$

(25)

$A_{\omega}$ and $B_{\omega}$ are determined from the no-slip boundary condition on the surface: $u_r(r = a) = U_{\omega} \cos \theta$, $u_\theta(r = a) = -U_{\omega} \sin \theta$ and reads

$$
A_{\omega} = -\frac{3}{2} \left( \frac{a}{\alpha_{\omega}} \right) e^{-\frac{1}{2} \alpha_{\omega} a}
$$

$$
B_{\omega} = \frac{1}{2} \left( \frac{a}{\alpha_{\omega}} \right) (3 - 3 i \alpha_{\omega} a - \alpha_{\omega}^2 a^2)
$$

(26)
Using Eq. (13), we write down the dissipation function explicitly in spherical polar coordinates,

\[ \phi(r, \omega) = 2\eta \left[ \left( \partial_r u_r \right) \left( \partial_r u_r \right)^* + \left( \frac{1}{r} \partial_\theta u_\theta + \frac{u_r}{r} \right) \left( \frac{1}{r} \partial_\theta u_\theta + \frac{u_r}{r} \right)^* \right] + \left( \frac{u_r}{r} + \frac{u_\theta \cot \theta}{r} \right) \left( \frac{u_r}{r} + \frac{u_\theta \cot \theta}{r} \right)^* + \eta \left[ r \partial_\theta \left( \frac{u_\theta}{r} \right) + \frac{\partial_\theta u_r}{r} \right] \left[ r \partial_\theta \left( \frac{u_\theta}{r} \right) + \frac{\partial_\theta u_r}{r} \right]^* . \]

Plugging in the flow field from Eq. (23), the dissipation function can be written down in a closed analytical form. To simplify this expression, we define a frequency dependent length scale \( \lambda_\omega \) as,

\[ \lambda_\omega = k_\omega^{-1} = \sqrt{\frac{2\eta}{\nu_0 \omega}} \]

Using this definition for \( k_\omega \), the dissipation function is given by:

\[ \phi(r, \omega) = 2\eta |U_\omega|^2 \left[ \frac{3}{4} \left( \frac{9a^2}{k_\omega^2} \right) \frac{1}{r^8} \mathcal{A}' \cos^2 \theta + \frac{1}{4} \left( \frac{9a^2}{k_\omega^2} \right) \frac{1}{r^3} \mathcal{B}' \sin^2 \theta \right] \]

where \( \mathcal{A}' \) and \( \mathcal{B}' \) are the complex conjugate of \( \mathcal{A} \) and \( \mathcal{B} \), respectively. The terms \( \mathcal{A} \) and \( \mathcal{B} \) are functions of \( r \) and \( \omega \) and a detailed expression for them are given in Appendix A.

### IV. EFFECTIVE NOISE TEMPERATURE

Finally, we are now in a position to calculate the effective frequency dependent temperature \( T(\omega) \) using Eq. (12). The denominator, which is the integral of the dissipation function over the whole volume yields,

\[ \int dV(\mathbf{r}, \omega) = 2\eta |U_\omega|^2 \int_0^\infty dr \int_0^{\pi} d\sin \theta \int_0^{2\pi} d\phi \left[ \frac{3}{4} \mathcal{A}' \cos^2 \theta + \frac{1}{4} \mathcal{B}' \sin^2 \theta \right] \]

\[ = 4\pi \eta |U_\omega|^2 \left[ \frac{9a^2}{k_\omega^2} \right] \int_0^\infty r^2 dr \left[ \frac{1}{2} \mathcal{A}' + \frac{1}{3} \mathcal{B}' \right] \]

\[ = 4\pi \eta |U_\omega|^2 \left[ \frac{9a^2}{k_\omega^2} \right] \left( \frac{k_\omega^4}{27a} \right) \left( 1 + \alpha_k \right) . \]

Using the final form of the above equation, a little algebra gives the result

\[ \int dV(\mathbf{r}, \omega) = 4\pi \eta |U_\omega|^2 3a \left( 1 + \alpha_k \right) . \]

Note that in the limit of \( \omega \to 0 \) the integral evaluates to

\[ \lim_{\omega \to 0} \int dV(\mathbf{r}, \omega) = 4\pi \eta |U_\omega|^2 3a . \]

Using the form of the temperature profile from Eq. (16), the numerator in Eq. (12) takes the form

\[ \int dV \left( T_0 + \Delta T \right) \phi(\mathbf{r}, \omega) \]

The first term in the above equation is trivial and is given by the spatial integral of the dissipation function rescaled by a factor \( T_0 \)

\[ \int dV T_0 \phi(\mathbf{r}, \omega) = 4\pi \eta T_0 |U_\omega|^2 3a \left( 1 + \alpha_k \right) . \]

The second term which integrates the spatially varying part of the temperature field weighted by the dissipation function yields a slightly complicated expression,

\[ \Delta T \int dV \frac{\alpha_k}{r} \phi(\mathbf{r}, \omega) = 4\pi \eta |U_\omega|^2 \Delta T \frac{5a}{4} \mathcal{F}(\omega) \]

with \( \mathcal{F}(\omega) \) given by

\[ \mathcal{F}(\omega) = \left[ \left( 1 + 2\alpha_k \right) \frac{6}{5} a^2 k_\omega^2 + \frac{2}{5} a^3 k_\omega^3 \right] + a^3 k_\omega^3 e^{2ak} \left( -6\pi(1 + \alpha_k) \cos ak \right) + 6ak \left( 1 + \frac{2}{3} \alpha_k \right) \sin ak \left. + \left( 3i(1 + \alpha_k) \right) + 3ak \left( 1 + \frac{2}{3} \alpha_k \right) \right] U(\omega) \]

where \( U(\omega) \) is the second term which integrates the spatially varying part of the temperature field weighted by the dissipation function yields a slightly complicated expression.

### A. The Low Frequency Limit of \( T(\omega) \)

In this section, we estimate the effective temperature \( T(\omega) \) in the limit of \( \omega \to 0 \) and derive the first order correction in \( \omega \) to its limiting value of \( T_{\text{HBM}} \). To this end we expand the function \( \mathcal{F}(\omega) \) in Taylor series as \( \omega \to 0 \). For small values of \( \omega \) one can very easily read off from Eq. (35)

\[ \mathcal{F}(\omega) \approx 1 + 2\alpha_k \] for \( \omega \to 0 \).

Hence, the frequency dependence of the effective temperature take the form

\[ T(\omega) \approx T_0 + \frac{5\Delta T}{12} \left( 1 + \alpha_k \right) = T_0 + \frac{5\Delta T}{12} \left( 1 + a \sqrt{\frac{\rho_0}{2\eta}} \right) \]

Note that \( \tau_r = a^2 / \nu = a^2 / (\eta / \rho_0) \) is the typical vorticity diffusion timescale in the fluid.
\[
\frac{T(\omega) - T_0}{\Delta T} = \frac{\omega}{T} \left[ \frac{1}{\omega^2 + \theta^2} \right] \]

The dissipation function calculated from the above expressions for the flow field yields:

\[
\phi(r, \omega) = 2\eta |U_\omega|^2 \left( \frac{81 \alpha^2 \eta^2}{r^6} \right) \frac{1}{r^3} \left[ \mathcal{A}_1 \mathcal{R}_1^3 \cos^2 \theta + \mathcal{B}_1 \mathcal{B}_1^3 \sin^2 \theta \right]
\]

B. The High Frequency Limit of \( T(\omega) \)

The general expression for the effective temperature as given in Eq. (36) is not very transparent and the high frequency behavior is difficult to extract from this expression, in particular spurious oscillations appear due to the behavior of the integrals \( Ei(x) \). It should be pointed out here, that the limit of \( \omega \to \infty \) implies that we look at large frequencies with \( \omega >> \sqrt{2a^2} \) but \( \omega < c/a \), where \( c \) is the adiabatic sound speed in the solvent. This allows us to treat the solvent incompressible and use the Stokes solution as given in Eq. (23).

In order to have a gainful insight into the effective temperature at large frequencies, instead of working with Eq. (36), we look at the flow field given in Eq. (23) in the high frequency limit. Since \( \alpha \omega \sim \sqrt{\omega} \), the terms with coefficient \( A_{\omega} \) in \( u_r \) and \( u_\theta \) are exponentially damped in the expression of dissipation function. A careful consideration gives the approximate flow field in the high frequency limit as

\[
\begin{align*}
  u_r(\omega) &= \left[ 2A_{\omega} \left( \frac{i\alpha \omega}{r^2} \right) e^{i\alpha \omega r} - B_\omega \frac{2}{r^3} \right] U_\omega \cos \theta \\
  u_\theta(\omega) &= \left[ A_\omega \left( \frac{\alpha^2}{r^2} \right) e^{i\alpha \omega r} - B_\omega \frac{1}{r^3} \right] U_\omega \sin \theta,
\end{align*}
\]

(39)

The dissipation function calculated from the above expressions for the flow field yields:

\[
\phi(r, \omega) = 2\eta |U_\omega|^2 \left( \frac{81 \alpha^2 \eta^2}{r^6} \right) \frac{1}{r^3} \left[ \mathcal{A}_1 \mathcal{R}_1^3 \cos^2 \theta + \mathcal{B}_1 \mathcal{B}_1^3 \sin^2 \theta \right]
\]

Figure 1. Plot of \( (T(\omega) - T_0)/\Delta T \) using Eq. (36) as a function of the dimensionless quantity \( ak_\omega \) (the solid line). The low frequency approximation in (38) is depicted by the dot-dashed line while the horizontal dashed line denotes the value of the ratio \( (T(\omega) - T_0)/\Delta T \) at zero frequency.

Figure 2. The plots depict the actual flow fields in the solvent as a function of \( k_\omega \) from Eq. (23) and its comparison to the high frequency approximation in Eq. (39). Plot of the real and imaginary parts of the flow field \( u_r(\omega) \cos \theta \) (real part: figure (a), imaginary part: figure (b)) and \( u_\theta(\omega) \sin \theta \) (real part: figure (c), imaginary part: figure (d)) using the complete solution of the Stokes equation Eq. (23) for two different values of \( k_\omega \): \( k_\omega = 1 \) (the dot-dashed line) and 10 (the dotted line). The corresponding high frequency approximations given in Eq. (39) is also plotted for the same values of \( k_\omega \): \( k_\omega = 1 \) (the solid line) and 10 (the dashed line). In the high frequency limit the approximation of given in Eq. (39) matches with the flow field for all values of \( r \) and consequently the dotted line and the dashed line overlap to give an appearance of a dot-dashed line. The radius of the nanoparticle \( a \) is unity.

with

\[
\mathcal{A}_1 = 1 + \frac{2i}{3} a^2 k_\omega^2 \left( 1 - \frac{r^2}{a^2} e^{-(r-a)(1+i)k_\omega} \right)
\]

\[
+ (1 + i)ak_\omega \left( 1 - \frac{r}{3a} e^{-(r-a)(1+i)k_\omega} \right)
\]

(41)

and

\[
\mathcal{B}_1 = 1 + \frac{2i}{3} a^2 k_\omega^2 \left( 1 - \frac{r^2}{a^2} e^{-(r-a)(1+i)k_\omega} \right)
\]

\[
+ (1 + i)ak_\omega \left( 1 - \frac{1}{3a} e^{-(r-a)(1+i)k_\omega} \right)
\]

\[
- i(1 - i)r^3 k_\omega^3 e^{-(r-a)k_\omega}
\]

(42)

Since \( k_\omega \) diverges as \( \sqrt{\omega} \), the exponentials are damped in the dissipation function and rapidly approach zero as \( \omega \to \infty \). Hence, we approximate \( \mathcal{A}_1 \) as

\[
\mathcal{A}_1 = 1 + \frac{2i}{3} a^2 k_\omega^2 + (1 + i)ak_\omega
\]

(43)

and \( \mathcal{B}_1 \) as

\[
\mathcal{B}_1 = 1 + \frac{2i}{3} a^2 k_\omega^2 + (1 + i)ak_\omega - i(1 - i)r^3 k_\omega^3 e^{-(r-a)k_\omega}
\]

(44)
As a consequence, the dotted line and the dashed line overlap to give

To get the asymptotic correction to \( \mathcal{T} \), known to us, we can use the generalized Langevin equation

following expression for \( \mathcal{T} \) reads as

where the Green’s function \( G(\omega) \) is given by

In order to evaluate the equipartition value \( \langle V^2 \rangle \) we use the Wiener-Khinchine theorem and write down the velocity auto-correlation function as:

From this the equal time correlation reads as

The kinetic temperature that governs the width of the distribution of the velocities of the heated particle follows from the equipartition theorem Eq. (2), with \( M \) replaced by the effective mass \( M_{\text{eff}} \) which equals the mass of the particle \( M \) augmented by half the mass of the displaced fluid – an effect that is solely the consequence of the incompressible nature of the solvent and is made apparent later. To proceed further, we need the explicit form for \( \zeta(\omega) \) and we use the result for the friction coefficient on a spherical colloidal particle in an incompressible solvent:

where \( \zeta_0 = 6\pi \eta a \) is the Stokes friction coefficient. The last term in the equation is responsible for the effective mass of the particle. This becomes more evident once we write down the Green’s function as

where \( \rho_p \) is the density of the colloid. It is apparent that the above equation can simply be written down as

with

but with \( \zeta(\omega) = \zeta_0[1 + a k_\omega(1 - i)] \). Plugging in the expression for \( G(\omega) \) from Eq. (53) in Eq. (51) and using the variable \( x = a k_\omega \) the kinetic temperature reads as

V. THE KINETIC TEMPERATURE \( T^*_{\text{HBM}} \)

Once the complete spectrum of the effective temperature is known to us, we can use the generalized Langevin equation in Eq. (3) to evaluate the velocity autocorrelation function \( V(t) V(0) \) for the colloid. The convolution in Eq. (3) is easier to handle in the frequency domain and the expression for the velocity of the particle reads as

where \( \mathcal{V}(\omega) = G(\omega) \xi(\omega) \)
with $\chi = \frac{3}{2}(2\rho_P/\rho_0 + 1)$. Substituting for $\mathcal{T}(x)$ from (36), we rewrite the above equation as
\[
\mathcal{T}_{\text{HBM}}^x = T_0 + \frac{5}{12} \Delta T \int_0^{\infty} \frac{4\lambda x(1+x)\mathcal{F}(x)}{(1+x)^2 + x^2(1+\chi x)^2} \, dx \tag{57}
\]
The integral is numerically evaluated for different values of $\chi$ and the quantity $\mathcal{T}_{\text{HBM}}^x - T_0$ is plotted against the density ratio $\rho_P/\rho_0$ for a better insight. We note that for $\rho_P/\rho_0 \rightarrow 0$ such that the particle density $\rho_P$ remains finite but the fluid density $\rho_0 \rightarrow \infty$ leads to an infinite time for the vorticities to diffuse in the fluid and the frequency dependent length scale $\lambda_\omega \rightarrow 0$. Consequently, the particle equilibrates with the region of the fluid close to the surface and the kinetic temperature $\mathcal{T}_{\text{HBM}}^x(\rho_P/\rho_0 \rightarrow 0) \approx T_0 + 0.87\Delta T$. The opposite limit of $\rho_P/\rho_0 \rightarrow \infty$, with $\rho_0$ finite and $\rho_P \rightarrow \infty$ is the Brownian limit where the particle is infinitely massive. Due to this infinite inertia of the particle, the kinetic degrees of freedom of the particle equilibrates with the whole solvent and the kinetic temperature equals the configurational temperature $\mathcal{T}_{\text{HBM}}^x = \mathcal{T}_{\text{HBM}}^2 = T_0 + \frac{5}{12} \Delta T$.

VI. FREQUENCY DEPENDENT DIFFUSIVITY

One of the promising applications of hot Brownian motion is its use in Photothermal Correlation Spectroscopy (PhoCs) which has the advantage of replacing the conventional fluorescence spectroscopic techniques. Using hot Brownian motion to investigate the local microscopic environment of complex fluids and extract useful information about the viscoelastic response of such systems would warrant an a-priori knowledge of the dependence of the the diffusivity on the frequency arising from the inhomogeneous temperature profile. By definition the frequency dependent diffusivity is given by
\[
D(\omega) = \frac{1}{3} \int_0^{\infty} \langle \mathbf{V}(t) \cdot \mathbf{V}(0) \rangle e^{i\omega t} \, dt \tag{58}
\]
Using the form $\langle \mathbf{V}(t) \cdot \mathbf{V}(0) \rangle$ from Eq. (50) the frequency dependent diffusion coefficient becomes
\[
D(\omega) = |G(\omega)|^2 k_B T(\omega) \Re e[\zeta(\omega)] \tag{59}
\]
For $\omega = 0$ the equation reduces to the value reported by Falasco et al.:
\[
D(0) = k_B T(0) \frac{\zeta_0}{\zeta_0} = k_B T_0 \left(1 + \frac{5}{12} \frac{\Delta T}{T_0} \right) \tag{60}
\]
indicating that the heated colloid moves with an enhanced diffusivity. For frequencies close to $\omega = 0$, the first order correction to this value comes by expanding (59) near $\omega = 0$ and leads to the expression:
\[
D(\omega) \approx k_B T_0 \frac{\zeta_0}{\zeta_0} \left[1 + \frac{5}{12} \frac{\Delta T}{T_0} \right] - a_k \omega \left[1 + \frac{5}{6} \frac{\Delta T}{T_0} a_k \right] \tag{61}
\]

VII. CONCLUSION

In conclusion, the framework of fluctuating hydrodynamics is used to derive an expression for the frequency dependent effective temperature $\mathcal{T}(\omega)$ of a hot Brownian particle. This effective parameter is particularly useful in describing the dynamics of the heated colloid using a generalized Langevin equation. We consider the colloidal particle to be suspended in an incompressible solvent and the subsequent heating of the colloid to be small. Consequently, the resulting spatial variation of the viscosity profile can be neglected in the linearized Stokes equation. Our work focuses on two specific limits of this effective temperature - the low frequency limit corresponding to $\omega \rightarrow 0$ and the high frequency limit of $\omega \gg 2\eta a^2 / \rho_0$. In the low frequency limit, the first order correction to $\mathcal{T}_{\text{HBM}}^x$ is calculated and is shown to scale as $\sqrt{\omega}$. The analytical form $\mathcal{T}(\omega \rightarrow 0)$ and is further used in evaluating the effective frequency dependent diffusion coefficient $D(\omega)$ and its approximation in the small frequency limit. This is particularly useful in applications of hot Brownian motion in experiments. Further, a careful treatment of the corresponding high frequency limit of $\mathcal{T}(\omega)$ is done by a suitable approximation of the flow field for $\omega \gg 2\eta a^2 / \rho_0$. Finally, using the generalized Langevin equation and $\mathcal{T}(\omega)$, we estimate the kinetic temperature $\mathcal{T}_{\text{HBM}}^x$ that governs the distribution of the velocities of the heated particle. A numerical evaluation of the equal time correlation $\langle \mathbf{V}^2 \rangle$ and using the equipartition theorem yields $\mathcal{T}_{\text{HBM}}^x$ as a function of ratio of the densities of the particle and the fluid $\rho_P/\rho_0$. A consistency check is done by looking at the two limiting cases $\rho_P/\rho_0 \rightarrow 0$ and $\rho_P/\rho_0 \rightarrow \infty$.

Appendix A: Calculation of the dissipation function

The terms $\mathcal{A}(r, \omega)$ and $\mathcal{B}(r, \omega)$ can be further written down as $\mathcal{A}(r, \omega) = \sum_{i=1}^3 a_i(r, \omega)$ and $\mathcal{B}(r, \omega) = \sum_{i=1}^3 b_i(r, \omega)$, with

\[
\begin{align*}
\mathcal{a}_j(r, \omega) & = \int_0^{\infty} e^{i\omega t} \, \left(\frac{1}{\chi_j^2} - 1\right) \mathcal{F}_j(\chi_j) \, d\chi_j \\
\mathcal{b}_j(r, \omega) & = \int_0^{\infty} e^{i\omega t} \, \left(\frac{1}{\chi_j^2} - 1\right) \mathcal{G}_j(\chi_j) \, d\chi_j
\end{align*}
\]
where the coefficients $a_i(r, \omega)$ given by

\begin{align}
a_1(r, \omega) &= (1 - e^{-k_o(r-a)(1+i)}) \\
a_2(r, \omega) &= \frac{2}{3} i a^2 k_o^2 \left(1 - \frac{r^2}{a^2} e^{-k_o(r-a)(1+i)}\right) \\
a_3(r, \omega) &= (1 + i) a k_o \left(1 - \frac{r}{a} e^{-k_o(r-a)(1+i)}\right)
\end{align}

and $b_i(r, \omega)$

\begin{align}
b_1(r, \omega) &= (1 - e^{-k_o(r-a)(1+i)}) \\
b_2(r, \omega) &= \frac{2}{3} i a^2 k_o^2 \left(1 - \frac{3 r^2}{2 a^2} e^{-k_o(r-a)(1+i)}\right) \\
b_3(r, \omega) &= (1 + i) a k_o \left(1 - \frac{r}{a} e^{-k_o(r-a)(1+i)}\right) \\
b_4(r, \omega) &= -i (1 + i) a^3 k_o^3 \left(1 - \frac{r}{a} e^{-k_o(r-a)(1+i)}\right).
\end{align}

Using the above equations, the expressions for $\mathcal{A}\mathcal{A}^*$ and $\mathcal{B}\mathcal{B}^*$ can be recast in the form

\begin{align}
\mathcal{A}\mathcal{A}^* &= g_1(k_o) + g_2(r, k_o) e^{-k_o(r-a)} \sin k_o(r-a) + \\
&\quad g_3(r, k_o) e^{-k_o(r-a)} \cos k_o(r-a) + \\
&\quad g_4(r, k_o) e^{-2k_o(r-a)}
\end{align}

and

\begin{align}
\mathcal{B}\mathcal{B}^* &= f_1(k_o) + f_2(r, k_o) e^{-k_o(r-a)} \sin k_o(r-a) + \\
&\quad f_3(r, k_o) e^{-k_o(r-a)} \cos k_o(r-a) + \\
&\quad f_4(r, k_o) e^{-2k_o(r-a)},
\end{align}

where the coefficients $g_i$ reads:

\begin{align}
g_1(k_o) &= 1 + 2 a k_o + 2 a^2 k_o^2 + \frac{4}{3} a^3 k_o^3 + \frac{4}{9} a^4 k_o^4 \\
g_2(r, k_o) &= \left(1 + 2 k_o r + 2 a k_o^2 r + \frac{4}{3} a^3 k_o^3 + \frac{4}{9} a^4 k_o^4 r^4\right) \\
g_3(r, k_o) &= -2 \left(1 + a k_o + r k_o + 2 a^2 k_o^2 r + \frac{2}{3} a^3 k_o^3 r + \frac{2}{3} a^4 k_o^4 r^2\right) \\
g_4(r, k_o) &= 2 a k_o \left(1 + \frac{2}{3} a k_o - \frac{r}{a} + \frac{2}{3} a^2 k_o r - \frac{2}{3} a^3 k_o^2 r + \frac{2}{3} a^4 k_o^3 r^2\right)
\end{align}

and the coefficients $f_i$ are given by

\begin{align}
f_1(k_o) &= 1 + 2 a k_o + 2 a^2 k_o^2 + \frac{4}{3} a^3 k_o^3 + \frac{4}{9} a^4 k_o^4 \\
f_2(r, k_o) &= \left(1 + 2 k_o r + 2 a k_o^2 r + \frac{4}{3} a^3 k_o^3 + \frac{4}{9} a^4 k_o^4 r^4\right) \\
f_3(r, k_o) &= -2 \left(1 + a k_o + r k_o + 2 a^2 k_o^2 r + \frac{2}{3} a^3 k_o^3 r + \frac{2}{3} a^4 k_o^4 r^2\right) \\
f_4(r, k_o) &= 2 a k_o \left(1 + \frac{2}{3} a k_o - \frac{r}{a} + \frac{2}{3} a^2 k_o r - \frac{2}{3} a^3 k_o^2 r + \frac{2}{3} a^4 k_o^3 r^2\right)
\end{align}

Appendix B: Calculation of the effective temperature in the high frequency limit

Using the expression for $\mathcal{A}_1$ and $\mathcal{B}_1$ from Eqs. (43) and (44), we rewrite the dissipation function as

\begin{align}
\phi(r, \omega) &= 2 \eta |U_o|^2 \left(\frac{81 a^2 r^2}{5 f_0^3 \omega^2}\right) \frac{1}{r^8} \left[3 f_1(k_o) \cos^2 \theta + \tilde{g}(r, k_o) \sin^2 \theta\right]
\end{align}

where $f_1(k_o)$ is defined in Eq. (A6) and $\tilde{g}(r, k_o)$ is given by

\begin{align}
\tilde{g}(r, k_o) &= 1 + 2 a k_o + 2 a^2 k_o^2 + \frac{4}{3} a^3 k_o^3 + \frac{4}{9} a^4 k_o^4 \\
&\quad + 2 k_o r e^{-k_o(r-a)} - \frac{4}{3} a^2 r^2 k_o^3 e^{-k_o(r-a)} + \\
&\quad 2 k_o^5 r^4 e^{-2k_o(r-a)}
\end{align}

The spatial integral of the dissipation function yields

\begin{align}
\int dr \phi(r, \omega) &= 2 \eta |U_o|^2 \left(\frac{81 a^2 r^2}{5 f_0^3 \omega^2}\right) \left(\frac{1}{a^8}\right) G(\omega)
\end{align}

with

\begin{align}
G(k_o) &= \left[2 f_1(k_o) + \frac{4}{27} \tilde{g}_1(k_o)\right]
\end{align}

where $\tilde{g}_1(k_o)$ is given by

\begin{align}
\tilde{g}_1(k_o) &= \frac{1}{45 a^5} \left(9 + 18 a k_o + 18 a^2 k_o^2 + 57 a^3 k_o^3 - 41 a^4 k_o^4 + 15 a^5 k_o^5 + 30 a^6 k_o^6\right) \\
&\quad + 15 a^5 e^{a k_o} k_o^5 \left(-3 + 2 a^2 k_o^2\right) Ei(-a k_o)
\end{align}
The numerator in the expression for the effective temperature evaluates to

\[ \int dr T(r) \phi(r, \omega) = 2T_0 \eta U_0 \left( \frac{81a^2\eta^2}{540\omega^2} \right) \left( \frac{1}{a^8} \right) \mathcal{G}(\omega) + 2\eta U_0 \left( \frac{81a^2\eta^2}{540\omega^2} \right) \left( \frac{1}{a^6} \right) \mathcal{H}(\omega) \]  

(B6)

with \( \mathcal{H}(\omega) \) given by

\[ \mathcal{H}(\omega) = \left[ 2f_1(k) + \frac{4}{27} \tilde{g}_2(k) \right] \]  

(B7)

where \( \tilde{g}_2(k) \) has the form

\[ \mathcal{H}(\omega) = \left[ 9 + 18ak_\omega + 18a^2k_\omega^2 + 48a^3k_\omega^3 - 14a^4k_\omega^4 
- 6a^5k_\omega^5 + 12a^6k_\omega^6 - 12a^7k_\omega^7 
- 6a^6k_\omega^6e^{ak_\omega} \left( -3 + 2a^2k_\omega^2 \right) \text{Ei}(-ak_\omega) 
+ 108a^6k_\omega^6e^{2ak_\omega} \Gamma(0, 2ak_\omega) \right] \]  

(B8)

where \( \Gamma(0, x) = \int_0^\infty s^{-1} e^{-sx} ds \).

REFERENCES

1. G. Falasco, M. V. Gnann, D. Rings, and K. Kroy, “Effective temperatures of hot Brownian motion,” Physical Review E 90, 032131–10 (2014).
2. D. Rings, R. Schachoff, M. Selmke, F. Cichos, and K. Kroy, “Hot Brownian Motion,” Physical Review Letters 105 (2010), 10.1103/PhysRevLett.105.090604.
3. D. Rings, M. Selmke, F. Cichos, and K. Kroy, “Theory of Hot Brownian Motion,” Soft Matter 7, 3441 (2011).
4. D. Chakraborty, M. V. Gnann, D. Rings, J. Glaser, F. Otto, F. Cichos, and K. Kroy, “Generalised Einstein relation for hot Brownian motion,” EPL (Europhysics Letters) 96, 60009 (2011).
5. R. Wulfert, M. Oechsle, T. Speck, and U. Seifert, “Driven Brownian particle as a paradigm for a nonequilibrium heat bath: Effective temperature and cyclic work extraction,” Physical Review E : Rapid Communications 95, 050103(R) (2017).
6. T. Chow and J. Hermans, “Brownian motion of a spherical particle in a compressible fluid,” Physica 65, 156–162 (1973).
7. R. Radünz, D. Rings, K. Kroy, and F. Cichos, “Hot Brownian Particles and Photothermal Correlation Spectroscopy,” The Journal of Physical Chemistry A 113, 1674–1677 (2009).