Non-Isothermal Fluctuation-Dissipation Relations and Brownian Thermometry

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The classical theory of Brownian motion rests on fundamental laws of statistical mechanics, such as the equipartition theorem and the fluctuation-dissipation theorem, which are not applicable in non-isothermal situations. We derive the generalized fluctuation-dissipation relations and Langevin equations governing such non-isothermal Brownian motion, including explicit results for the frequency-dependent noise temperature and Brownian thermometry far from equilibrium.

The perpetual thermal dance of the molecules of a fluid manifests itself in incessant erratic movements of small suspended particles. If a quantitative characterization of this mesoscopic motion is available, it can serve as a Brownian thermometer. More than a century ago, A. Einstein [1] provided such a mesoscopic stochastic model demonstrating that the 3-dimensional position and velocity fluctuations of isothermal Brownian motion are indeed given by the thermal energy $k_B T$,

$$\lim_{t \to \infty} \langle (X(t) - X(0))^2 \rangle / 6t = 3 k_B T / \zeta,$$

$$\lim_{t \to 0} \langle V(t) \cdot V(0) \rangle = 3 k_B T / M,$$

filtered through the friction $\zeta$ and the renormalized inertial mass $M$ of the particle, respectively. Note that both coefficients characterize the fluid-particle interactions, and that the Brownian thermometer not only measures $T$ but additionally witnesses the atomistic origin of the fluctuations through the appearance of Boltzmann’s constant (originally the gas constant divided by the Loschmidt number). The experimental verification of Eqs. (1) could thus be interpreted as a confirmation of the atomistic structure of matter [2], and their generalization in the form of the fluctuation-dissipation theorem (FDT) became a corner stone of statistical mechanics [3].

A more accurate quantum-mechanical description modifies Eqs. (1), showing that eigenmodes of eigenfrequencies $\omega \gg k_B T / \hbar$ freeze out exponentially [3]. The physical interpretation of the quantum version of the FDT is then that a Brownian thermometer probes a universal energy spectrum $\hbar \omega \coth(\hbar \omega / 2 k_B T)$ rather than a universal thermal energy $k_B T$.

In this Letter, we show that a very similar (though somewhat less universal) situation is encountered far from equilibrium. We generalize Eqs. (1) and the FDT to the case of a classical Brownian particle in a non-isothermal fluid by explicitly calculating the appropriate energy spectrum $k_B T(\omega)$ of the thermal noise. Thereby, we extend Einstein’s theory of a Brownian thermometer to non-isothermal solvents. Similar as the kinetic coefficients $\zeta$ and $M$, and unlike the universal quantum spectrum, $T(\omega)$ is found to be a non-universal function of the particle-fluid interactions. This should not come as a surprise. Due to the lack of a zero’th law, the reading of any thermometer operating far from equilibrium should certainly be expected to “depend on its orientation, shape, surface properties (...) and other variables in the system being observed” [7, 8]. Nevertheless, our derivation, much like its equilibrium counterpart, establishes a comparatively simple, yet precise, mesoscopic model of Brownian motion that is essentially independent of molecular details. Also like in equilibrium, it remains a paradigm for fluctuations in other mesoscopic devices operating under non-isothermal conditions, e.g. in electrical engineering (Johnson–Nyquist noise) and nanophotonics (antenna noise).

General theory—Brownian motion in a thermally inhomogeneous bath is a vivid research subject [8, 12]. Yet, the dynamical equations on which most of this work hinges are often taken for granted. Explicit derivations assume the particle to be sensitive only to local (in space and time) thermal fluctuations, conforming with the principles of non-equilibrium thermodynamics [10]. It is rarely acknowledged that such an assumption disregards the solvent dynamics, namely how the momentum spreads from the particle into the fluid and is returned in the form of colored noise. This issue has recently received considerable attention for isothermal Brownian motion [13, 19] and is our main focus, here.

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exists a frequency-dependent coupling, mediated by the hydrodynamic modes, between the particle and distant volume elements $\Delta V$ of the solvent. In a discretized representation, we enumerate these volume elements by $\alpha$, instead of $r$. The particle-solvent coupling can then be fully encoded in the local response function $\phi_{ij}(X, \omega)$ \cite{20} of the fluid momentum (at position $\alpha$) in the $i$-direction to the particle motion (at position $X$) in the $j$-direction. Since $\phi_{ij}(X, \omega)$ quantifies how kinetic energy is turned into heat at the local reservoir $\alpha$, maintained at temperatures $T^\alpha$, it amounts to a local frequency-dependent friction tensor. The reverse process of turning heat into kinetic energy gives rise to random fluctuations of the particle velocity, which we represent by a stochastic force $\xi_i^\alpha(X,t)$. By virtue of the assumption of local thermal equilibrium, this force is Gaussian with zero mean, and its strength is uniquely determined by the local temperature $T^\alpha$ and the (Fourier-transformed) response $\phi_{ij}(X,t)$ through the local FDT,

$$\langle \xi_i^\alpha(X,t)\xi_j^\beta(X,t') \rangle \approx T^\alpha \phi_{ij}(X,t-t') \Delta \nu \delta_{\alpha\beta}. \quad (2)$$

The total acceleration of the Brownian particle of mass $m$ can accordingly be written as a sum over the contributions from all volume elements,

$$m \ddot{X}_i \approx \sum_{\alpha} \left[ -\int_{-\infty}^{t} \sum_{j=1}^{3} \phi_{ij}^\alpha(X,t-t') \Delta \nu \dddot{X}_j(t') dt' + \xi_i^\alpha \right]. \quad (3)$$

In the continuum limit, $\Delta \nu \to 0$, the terms in the sum can be added up to the friction and noise terms

$$\zeta_{ij}(X,t) \approx \sum_{\alpha} \phi_{ij}^\alpha(X,t) \Delta \nu$$

and $\xi_i(X,t) \approx \sum_{\alpha} \xi_i^\alpha(X,t)$, respectively. Thereby, the equation of motion can be rewritten in the form of the generalized Langevin equation

$$m \ddot{X}(t) = -\int_{-\infty}^{t} \zeta(X,t-t') \cdot \dot{X}(t') dt' + \xi(X,t). \quad (4)$$

(plus an optional external force term). The non-equilibrium Gaussian noise $\xi$ has vanishing mean, and its correlations in the frequency domain read:

$$\langle \xi_i(X,\omega)\xi_j(X,\omega') \rangle = k_B T_{ij}(X,\omega) \zeta_{ij}(X,\omega) \delta(\omega + \omega'). \quad (5)$$

In contrast to the familiar isothermal situation \cite{18}, the color of the Brownian noise is not governed by the frequency-dependence of the friction alone, but also by that of the tensorial noise temperature $T_{ij}(X,\omega)$

$$T_{ij}(X,\omega) = \frac{\int_V \phi_{ij}(X,r,\omega) T(r) dr}{\int_V \phi_{ij}(X,r,\omega) dr}. \quad (6)$$

Equations \cite{21} constitute the central results of our contribution. In particular, the last two equations provide a generalization of the FDT (of the second kind) for a Brownian particle in a non-isothermal solvent. Due to the long-range hydrodynamic interactions between the particle and the fluid the noise temperature is manifestly non-local—Eq. \cite{4} has the form of a spatial average over the local fluid temperature $T(r)$ around the instantaneous particle position $X$. The weighting by the response tensor $\phi_{ij}$ reveals that $T_{ij}$ is not a property of the solvent or the particle, alone, but characterizes their mutual coupling. Via the symmetry of $\phi_{ij}$ in $\omega$ \cite{22}, it inherits the time-reversal invariance of the microscopic dynamics of the bath. And via the spatial dependence of $\phi_{ij}$ it “knows” about the particle radius, boundary conditions, and transport mode (translation or rotation) \cite{23,27}. The tensorial structure of $T_{ij}$ reflects that, for an arbitrary temperature field $T(r)$, hydrodynamic modes may carry different amounts of thermal energy along different spatial directions. If one interprets the colloidal particle as a Brownian thermometer sampling the solvent temperature field $T(r)$ via the mesoscopic intermediate $T_{ij}$ all this should be recognized as an important necessary feature rather than a deficiency. Indeed, the anisotropy and the variation of $T_{ij}(X,\omega)$ with its arguments and the transport mode can serve as measures for the departure from thermal equilibrium \cite{23}.

Intuitively, the meaning of the noise temperature can probably be best understood from the observation that it maps the local equilibrium condition of the hydrodynamic model onto a local equilibrium condition in the contracted system, introducing a quantum-type thermal spectrum. Due to the underlying local-equilibrium assumption, $k_BT_{ii}(X,\omega)$ literally represents the thermal energy that an equilibrium bath would have to supply via the noise component $\xi_i$ in order to mimic the effect of the non-isothermal solvent. That this energy has, in general, a colored spectrum and depends on the current position of the particle, and the direction of its motion, is the price one pays for integrating out the slow non-equilibrium degrees of freedom of the bath. It is worthwhile pointing out, though, that these “imperfections” of the equilibrium analogy are (partially) mended in certain practically important limits to be discussed below.

_Fluctuation-dissipation relations_—The violation of the classical FDT of the second kind in Eqs. \cite{3,4} entails a corresponding violation of the classical FDT of the first kind. To elucidate this point, we now focus on so-called hot Brownian motion \cite{29}, the case where the Brownian particle itself acts as the heat source and generates a co-moving radial temperature field $T(r) = T_h + \Delta TR/r$ in the fluid. Besides its practical relevance \cite{30}, this highly symmetric system may serve as a prototypical example to understand the main implications of a frequency-dependent noise temperature without the additional complications of the tensorial structure and position dependence of the noise temperature. For the hot Brownian particle Eqs. \cite{2,3} imply

$$C_V(\omega) = 2k_B T(\omega) \text{ Re } R_V(\omega), \quad (7)$$

where $C_V(\omega) = \langle V_i(\omega)V_i(-\omega) \rangle$ is the spectral density.
and \( R_v(\omega) = (\zeta^+(\omega) - i\omega m)^{-1} \) the response function of the particle velocity \[25\]. The time-dependencies of the linear response and of the spontaneous fluctuations of the Brownian particle obviously cannot be identified, as in equilibrium. Hence, the classical FDT is broken and Onsager’s regression hypothesis is not applicable. As in quantum mechanics, a single value of the noise temperature is generally not sufficient to fully characterize the stochastic excitations provided by the heat reservoir \[31\]. Instead, the energy carried by a degree of freedom is given as a sum of filtered mode contributions, with the filter bandwidth selected by its own response function. The fluctuation-dissipation ratio

\[ \Theta(t) = D(t)/\mu(t) \]

of the positional fluctuations and the response (to a small constant force) no longer reduces to a universal constant \( T_0 \), as in an isothermal bath. Instead, for the hot particle, the time-dependent diffusion coefficient

\[ D(t) = \int_0^t C_v(t')dt' \]

and the integrated response function

\[ \mu(t) = \int_0^t R_v(t')dt' \]

only become proportional to each other in asymptotic limits (Fig. 1). More precisely, it follows from Eq. \[5\] that \( \Theta(t) \) interpolates between the two effective temperatures \[22\]

\[ T^v = \frac{M}{k_B} \langle V_i^2 \rangle = \frac{M}{\pi} \int d\omega T(\omega) \text{Re} R_v(\omega) \]

at short times, and

\[ T^x = T(0) < T^v \]

at long times, in agreement with what was found in numerical simulations \[32, 33\]. The generalization of Eqs. \[1\] for a hot particle is thus

\[ \lim_{t \to \infty} \langle [X(t) - X(0)]^2 \rangle / 6t = 3k_B T^x / \zeta \]

\[ \lim_{t \to 0} \langle V(t) \cdot V(0) \rangle = 3k_B T^v / M. \]

Along the same lines, one can discuss a hot Brownian particle in harmonic confinement, a scenario of considerable practical relevance to metal nanoparticles controlled by optical tweezers \[31\]. Choosing the positional degree of freedom \( X_i \) as our observable and introducing the external confinement force \(-m\omega_0^2 X_i(t)\) into Eq. \[2\], one obtains the equivalent of the velocity equation \[5\] for the particle position,

\[ \omega C_X(\omega) = 2k_B T(\omega) \text{Im} R_X(\omega). \]

Here, \( C_X(\omega) = \langle X_i(\omega) X_i(-\omega) \rangle \) is now the spectral density and \( R_X(\omega) = (m(\omega^2 - \omega^2) - i\omega \zeta^+(\omega))^{-1} \) the response function of the position coordinate. Since the response functions \( R_v(\omega) \) and \( R_x(\omega) \) for the velocity and the position filter the overall temperature spectrum \( T(\omega) \) differently, energy equipartition between the (harmonic) position and momentum degrees of freedom inevitably breaks down. Velocity and position fluctuations of a hot Brownian harmonic oscillator thus thermalize to different effective temperatures

\[ m\omega_0^2 \langle X_i^2 \rangle = k_B T^x \]

\[ M \langle V_i^2 \rangle = k_B T^v. \]

We emphasize that these depend on the confinement strength for sufficiently strong confinement/weak solvent coupling, so that they generally cannot be identified with the corresponding temperatures denoted by the same symbols in Eq. \[1\].

**Brownian thermospectrometry**—The (at first sight) maybe somewhat disturbing dependence of the apparent equipartition temperatures on the confinement can actually be exploited to restore Onsager regression and the classical FDT, albeit with an effective temperature. If the motion of the Brownian particle is only weakly damped by the solvent, the response function is sharply peaked around the eigenfrequency \( \omega_0 \). This limit can practically be realized for large particle-to-fluid density ratios \( \rho_p/\rho \gg 1 \), e.g. for a Brownian particle suspended in a gas \[35, 37, 38\]. Then almost only the resonant mode \( k_B T(\omega_0) \) of the thermal energy spectrum is on average absorbed (Fig. 2). Thereby, from the Fourier back-transform of Eq. \[10\], the mean energy follows in the familiar equipartition form:

\[ m\omega_0^2 \langle X_i^2 \rangle = M \langle V_i^2 \rangle = k_B T(\omega_0). \]

This shows that by varying the trap stiffness \( m\omega_0^2 \), i.e. by endowing the Brownian thermometer with a tunable fre-
FIG. 2. The effective temperature $T^X$ governing the Boltzmann factor of a hot Brownian particle in harmonic confinement (normalized to the temperature difference $\Delta T$ between the particle surface and the ambient temperature $T_0$). The dashed-dotted and dashed lines correspond to weak ($q_0/\rho = 20$), and strong ($q_0/\rho = 0.5$) coupling to the solvent, or under- and over-damped oscillations, respectively. For large but physically accessible values of the particle/fluid density ratio $q_0/\rho$, $T^X$ coincides with the noise temperature $T(\omega_0)$ (solid line), in agreement with Eq. (12). Under these conditions, the Brownian particle can serve as a thermospectrometer for the noise spectrum $k_BT(\omega)$. Inset: the corresponding position spectral densities evaluated at $\omega_0/\omega_l = 4.$

The notion of a frequency-dependent temperature that quantifies violations of the FDT far from equilibrium is moreover reminiscent of the effective temperature previously suggested to govern the linear response of glasses [42]. This idea has been tested, with mixed success, in several models [43]. We therefore emphasize that our Eq. (11) and the corresponding generalized fluctuation-dissipation relations are not postulated, but analytically derived [24, 25], thereby providing an independent testbed for rigorously analyzing the scope of the notion of effective temperatures, far from equilibrium. Moreover, our theory may serve as a starting point to consistently extend the notions of stochastic thermodynamics [44] to non-isothermal systems.

Discussion.—It seems interesting to note that a quantity completely analogous to our $T(\omega)$, endowed with the very same physical meaning, is commonly used in electronics. In that context, the noise temperature is introduced to account for fluctuations in non-equilibrium conductors, when the Johnson–Nyquist FDT in not satisfied [40]. Exactly as in the equilibrium case, the mapping between the two related phenomena is established by substituting in Eq. (6)—the analogue of Nyquist’s formula [2]—the thermal force with the voltage and the friction coefficient with the resistance. Exploiting the analogy further, one finds that the Brownian noise temperature [4] exhibits the same formal structure as the noise temperature of radio receivers. Just like Eq. (4), the effective antenna temperature, which results from the electromagnetically mediated coupling to a non-isothermal environment, is an average over the temperatures of the surrounding radiation field weighted by the radiation pattern [41].

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Note that our discussion therefore includes “kinematic” thermophoresis due to the inhomogeneous noise strength, while it does not account for “molecular” thermophoresis due to molecular interactions.
Supplemental Material

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We consider a particle of arbitrary shape immersed in a non-isothermal simple fluid in local thermal equilibrium. The dynamical state of the system is given is terms of a few reduced variables, evolving in accord with the linearized fluctuating hydrodynamic equations and Newton’s law. The particle is described by the center of mass $X(t)$, and the translational and rotational velocity, respectively, $V(t)$ and $\Omega(t)$. The fluid occupies the volume $\mathcal{V}$ and is described by the velocity and temperature field, respectively, $\mathbf{v}(r,t)$ and $T(r,t)$. The velocity field obeys the equations

$$\rho \partial_t \mathbf{v}(r,t) - \nabla \cdot \boldsymbol{\sigma}(r,t) = \nabla \cdot \mathbf{\tau}(r,t), \quad (1a)$$
$$\nabla \cdot \mathbf{v}(r,t) = 0. \quad (1b)$$
$$\mathbf{v}(r,t) = \mathbf{V}(t) + \mathbf{\Omega}(t) \times r \quad \text{on } S \quad (1c)$$

The symmetric stress tensor $\boldsymbol{\sigma}$ has components $\sigma_{ij} = -p \delta_{ij} + 2\eta \Gamma_{ij}$, where $p$ is the pressure and $\Gamma_{ij} = (\partial_v v_j + \partial_j v_i)/2$ the shear rate tensor. The dynamic viscosity $\eta$ is in general a function of the temperature. Hence it is space dependent, $\eta = \eta[T(r)]$. For simplicity, the fluid is assumed incompressible. Therefore the mass conservation reduces to Eq. (1b).

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The noise stress tensor $\mathbf{\tau}$ describes thermal fluctuations in the fluid, whose statistical properties follow from the local fluctuation-dissipation theorem.

$$\langle \tau_{ij}(r,t) \tau_{kl}(r',t') \rangle = 2\eta(r,t) k_B T(r,t) \delta(r-r') \delta(t-t') (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}). \quad (2)$$

We assume that $\mathbf{\tau} \equiv 0$ on the boundary $S$. The temperature field entering Eq. (2) is in general the solution of a heat equation with appropriate boundary conditions describing the heat sources in $\mathcal{V}$ and the fluxes across $S$. In the following we assume the temperature field to be a prescribed function, independent of time. This is indeed the case whenever the particle motion causes negligible disturbances to the fluid temperature.

The velocities evolve by Newton’s equations of motion, i.e.

$$m \ddot{\mathbf{V}}(t) = \mathbf{F}(t) + \mathbf{F}_e(t), \quad (3a)$$
$$\mathbf{I} \cdot \ddot{\mathbf{\Omega}}(t) = \mathbf{T}(t) + \mathbf{T}_e(t) \quad (3b)$$

where $m$ is the mass of the particle and $\mathbf{I}$ its tensor of inertia. The force and torque exerted by the fluid are

$$\mathbf{F}(t) = -\int_S \boldsymbol{\sigma}(r,t) \cdot \mathbf{n}(r) \, d^2r, \quad (4a)$$
$$\mathbf{T}(t) = -\int_S r \times (\boldsymbol{\sigma}(r,t) \cdot \mathbf{n}(r)) \, d^2r, \quad (4b)$$

with $\mathbf{n}(r)$ the inner normal vector field of the particle surface $S$. External forces $\mathbf{F}_e(t)$ and torques $\mathbf{T}_e(t)$ may also be present. The system (3)–(4) describes the evolution of the fluid and the Brownian particle entirely.

Our aim is to eliminate the hydrodynamic fields and reduce Eqs. (1)–(4) to a generalized Langevin equation for the particle variables only. Thus we rewrite system (3) in the form

$$\mathbf{L} \cdot \dot{\mathbf{b}}(t) = \mathbf{h}(t) + \mathbf{f}_e(t), \quad (5)$$

where we combine the translational and rotational velocity into the 6-vector $\mathbf{b}(t) \equiv (\mathbf{V}(t), \mathbf{\Omega}(t))$, and we define the generalized tensor of inertia

$$\mathbf{L} \equiv \begin{pmatrix} m & 0 \\ 0 & \mathbf{I} \end{pmatrix},$$

and the generalized forces

$$\mathbf{h}(t) \equiv \begin{pmatrix} \mathbf{F}(t) \\ \mathbf{T}(t) \end{pmatrix}, \quad \mathbf{f}_e(t) \equiv \begin{pmatrix} \mathbf{F}_e(t) \\ \mathbf{T}_e(t) \end{pmatrix}. \quad (6)$$
One should appreciate that the hydrodynamic force splits into two parts

$$\mathbf{h}(t) \equiv \mathbf{h}_r(t) + \mathbf{\xi}(t),$$  

(7)

where $\mathbf{h}_r(t)$ is identified with the resistance force and $\mathbf{\xi}(t)$ with the Langevin noise. Equation (7) is easily justified as follows. By equation (11), $\mathbf{u}(r, t)$ and $p(r, t)$ are linear functionals of $\mathbf{b}(t')$ with $-\infty < t' < t$. Therefore, in view of Eq. (1), the total hydrodynamic force necessarily contains a contribution which is a linear functional of $\mathbf{b}(t')$ with $-\infty < t' < t$, i.e. we can write

$$\mathbf{h}_r(t) = -\int_{-\infty}^{t} \mathbf{Z}(X, t - t') \cdot \mathbf{b}(t') dt'.$$

(8)

Here $\mathbf{Z}(X, t)$ is a $6 \times 6$ time-dependent friction tensor, which also depends on the particle position owing to the non-constant fluid viscosity. We omit to show this dependence in the following. Moreover, since (1a) is a linear but non-homogeneous equation, the term $\mathbf{\xi}(t)$ has to be included in Eq. (7) in order to account for contributions to the hydrodynamic force which are independent of the particle velocity.

In the subsequent derivation we shall derive the statistics of the Langevin noise $\mathbf{\xi}(t)$ and relate it to the dissipative term $\mathbf{h}_r(t)$. The linearity of the problem suggests to operate in the frequency space. Given a generic function of time $g(t)$, we denote its Fourier transform and half-Fourier transform, respectively, by

$$g(\omega) = \int_{-\infty}^{\infty} g(t) e^{-i\omega t} dt, \quad g^+(\omega) = \int_{0}^{\infty} g(t) e^{-i\omega t} dt.$$

The complex conjugate of $g(\omega)$ will be denoted by $g^*(\omega)$.

The Fourier transform of Newton’s equation (5), with the definitions (7) and (8), reads

$$-i\omega \mathbf{L} \cdot \mathbf{b}(\omega) = -\mathbf{Z}^+(\omega) \cdot \mathbf{b}(\omega) + \mathbf{\xi}(\omega) + \mathbf{f}_e(\omega).$$

(9)

If the velocity 6-vector is split into its average and random part, i.e. $\mathbf{b}(\omega) = \bar{\mathbf{b}}(\omega) + \langle \mathbf{b}(\omega) \rangle$ the total hydrodynamic force takes the form $\mathbf{h}(\omega) = \langle \mathbf{h}_r(\omega) \rangle + \bar{\mathbf{h}}(\omega)$, with

$$\langle \mathbf{h}_r(\omega) \rangle = -\mathbf{Z}^+(\omega) \cdot \langle \mathbf{b}(\omega) \rangle,$$

$$\bar{\mathbf{h}}(\omega) = -\mathbf{Z}^+(\omega) \cdot \bar{\mathbf{b}}(\omega) + \mathbf{\xi}(\omega).$$

(10a)

(10b)

The physical meaning of Eqs. (10) is best understood if we cast the Fourier transform of Eqs. (1) into the decoupled set of equations

$$-i\omega \mathbf{u}(r, \omega) - \nabla \cdot \mathbf{\sigma}(r, \omega) = 0,$$

(11a)

$$\nabla \cdot \mathbf{u}(r, \omega) = 0, \quad \mathbf{u}(r, \omega) = \langle \mathbf{U}(\omega) \rangle + \langle \mathbf{\Omega}(\omega) \rangle \times \mathbf{r} \quad \text{on} \; S,$$

(11b)

and

$$-i\omega \mathbf{u}(r, \omega) - \nabla \cdot \mathbf{\sigma}(r, \omega) = \nabla \cdot \mathbf{\tau}(r, \omega),$$

(12a)

$$\nabla \cdot \mathbf{u}(r, \omega) = 0, \quad \mathbf{u}(r, \omega) = \langle \mathbf{U}(\omega) \rangle + \langle \mathbf{\Omega}(\omega) \rangle \times \mathbf{r} \quad \text{on} \; S.$$

(12b)

Clearly, equations (11) describe the deterministic fluid velocity, i.e. $\mathbf{u}(r, \omega) \equiv \langle \mathbf{v}(r, \omega) \rangle$, while equations (12) account for its random thermal fluctuations, i.e $\tilde{\mathbf{u}}(r, \omega) \equiv \mathbf{v}(r, t) - \mathbf{u}(r, \omega)$. In view of the boundary conditions (11c) and (12c), one recognizes (10a) as the friction produced by the deterministic velocity field solution of Eqs. (11), and (10b) as the random force exerted by the velocity field solution of Eqs. (12). Note that the deterministic part of the velocity vector $\langle \mathbf{b}(\omega) \rangle$ can be chosen arbitrarily.

We are now in the position to evaluate the statistics of the Langevin noise $\mathbf{\xi}(\omega)$. We operate in three steps. First, we derive an expression for the friction tensor $\mathbf{Z}(\omega)$. Then we show that $\mathbf{\xi}(\omega)$ is a Gaussian variable with zero mean. Finally we link the noise correlation tensor $\langle \mathbf{\xi}(\omega) \mathbf{\xi}^*(\omega) \rangle$ to the friction tensor. The benefit coming from casting Eqs. (1) into Eqs. (11)–(12) is easily seen. Indeed one can conveniently choose the boundary conditions in Eq. (11) to express the friction tensor and noise statistics in terms of functions of $\mathbf{u}(r, \omega)$, without solving the much more involved problem represented by the stochastic equations (12).
To evaluate the components of the friction tensor $Z_{ij}$ we make use of its properties, hinging only on the symmetry of the stress tensor $\sigma$ \[1\]. It can be showed that in general $Z$ is symmetric and is a real even function of time. From the latter property it immediately follows the relation

$$ Z_{ij}(\omega) = Z_{ij}^* (\omega) + Z_{ij}^+ (\omega). \quad (13) $$

We exploit the freedom of choosing the boundary condition \[12c\] to select velocity vectors whose non-zero one, and denote them by $b_i(\omega)$—the superscript $\alpha$ will also be appended to the corresponding hydrodynamic fields.

We wish to find an expression in terms of the solution to Eqs. \[11\] for the quantity

$$ Z_{ij}(\omega)\langle b_i(\omega) \rangle \langle b_j^*(\omega) \rangle = Z_{\alpha\beta}(\omega)\langle b_\alpha(\omega) \rangle \langle b_\beta^*(\omega) \rangle, \quad (14) $$

where the equality holds by virtue of the choice of $b(\omega)$. In Eq. \[14\] and in the following we apply the Einstein summation convention to latin indices only. Also, we suppress the function arguments where there is no risk of confusion. Equation \[14\] reads

$$ Z_{ij}(\omega)\langle b_i(\omega) \rangle \langle b_j^*(\omega) \rangle = (Z_{ij}^+ + Z_{ij}^*)\langle b_i(\omega) \rangle \langle b_j^*(\omega) \rangle = - (\alpha h_{ij}(\beta b_i) + \langle b_i(\omega) \rangle \langle b_j^*(\omega) \rangle)
$$

\[15\]

\[16\]

$$ = \int_S \langle (\omega V^\star) \rangle \Omega_i \sigma_{ij} n_j d^2 r + \int_S (\sigma \cdot n) \sigma_{ij} n_j d^2 r + \int_S \langle \Omega_i \rangle \int_S (\omega V^\star) \Omega_i \sigma_{ij} n_j d^2 r
$$

\[17\]

$$ \int_S \int_S \langle (\omega V^\star) \rangle \Omega_i \sigma_{ij} n_j d^2 r + \int_S (\sigma \cdot n) \sigma_{ij} n_j d^2 r
$$

\[18\]

where in \[15\] we used the divergence theorem and in \[16\] we defined the response tensor as

$$ \Phi_{\alpha\beta}(X, r, \omega) \equiv \eta(r) \left( \partial_{\beta} u_i (r, \omega) \partial_{\alpha} u_j (r, \omega) + \partial_{\beta} u_j (r, \omega) \partial_{\alpha} u_i (r, \omega) \right) = \Phi_{\beta\alpha}(X, r, \omega), \quad (17) $$

where the $X$ dependence of the hydrodynamic fields is not explicitly showed. Equation \[16\] is valid whatever the magnitude of $b_\alpha$ and $b_\beta$, in particular when they are unit vectors. With this choice we have

$$ Z_{\alpha\beta}(X, \omega) = 2 \int_V \Phi_{\alpha\beta}(X, r, \omega) d^3 r + i \varpi \int_V \langle u_i^\star \beta \omega \rangle (\sigma_{ij} n_j \sigma_{ij} n_j) d^3 r, \quad (18) $$

which has to be invariant under exchange of $\alpha$ and $\beta$, owing to the symmetry of $Z$. Therefore one can eliminate the second term in Eq. \[18\] and obtain for the friction tensor

$$ Z_{\alpha\beta}(X, \omega) = \int_V \Phi_{\alpha\beta}(X, r, \omega) d^3 r + \int_V \Phi_{\beta\alpha}(X, r, \omega) d^3 r = 2 \Re \int_V \Phi_{\alpha\beta}(X, r, \omega) d^3 r, \quad (19) $$

that can be written as

$$ Z_{\alpha\beta}(X, \omega) = \int_V \varphi_{\alpha\beta}(X, r, \omega) d^3 r, \quad (20) $$

where $\varphi_{\alpha\beta}(X, r, \omega) \equiv 2 \Re \Phi_{\alpha\beta}(X, r, \omega)$ is twice the real part of the response tensor \[17\], computed by setting unit vectors in Eq. \[18\].

Then we turn to the random force $\xi(\omega)$

$$ \xi_i(\alpha b_i) = \left( \hat{h}_i + Z_{ij}^\alpha b_j \right) (\alpha b_i) = \hat{h}_i (\alpha b_i) - \alpha h_j \tilde{b}_j $$. 
where $\xi$ is performed with the response tensor $\phi$, which shows that a zero-mean Gaussian field $\tau$ inherits the time reversal invariance of the stress tensor $\tau$. Hence, its correlation matrix suffices to specify the statistics completely. In addition, $\xi$ inherits the time reversal invariance of the stress tensor $\tau$.

Using (23), we determine the noise correlations

$$\langle \xi_i(\omega) \xi_j^*(\omega') \rangle = \langle b_i(\omega) \rangle \langle b_j^*(\omega') \rangle$$

which shows that $\xi$ is Gaussian with vanishing mean, being the integral of the deterministic quantity $\partial_j^n u_i$ times the zero-mean Gaussian field $\tau$. Hence, its correlation matrix suffices to specify the statistics completely. In addition, $\xi$ inherits the time reversal invariance of the stress tensor $\tau$.

In (24) we made use of the divergence theorem, and in (22) of the property $\partial_i \partial_j^n u_i = \partial_j^n \partial_i u_i$, which is a direct consequence of the symmetry of $\sigma$. We have found

$$\xi_i \langle b_i \rangle = \xi_\alpha \langle b_\alpha \rangle = \int_V \tau_{ij} \partial_j^n u_i \, d^3 r,$$

In (25) we used the Fourier transform of $b_\omega$. One gets, setting the magnitude of $\langle b_\alpha \rangle$ and $\langle b_\omega \rangle$ to one

$$\langle \xi_\alpha(X, \omega) \xi_\beta^*(X, \omega') \rangle = 2 k_B \delta(\omega - \omega') \int_V \Phi_{\alpha\beta}(X, r, \omega) T(r) \, d^3 r.$$

Besides, the symmetry under time reversal of $\xi$ implies that Eq. (27) is invariant under complex conjugation. This yields immediately

$$\langle \xi_\alpha(X, \omega) \xi_\beta^*(X, \omega') \rangle = k_B \delta(\omega - \omega') 2 \text{Re} \int_V \Phi_{\alpha\beta}(X, r, \omega) T(r) \, d^3 r.$$

From Eqs. (20) and (28) we can finally obtain the noise correlation tensor in the form

$$\langle \xi_\alpha(X, \omega) \xi_\beta^*(X, \omega') \rangle = k_B T_{\alpha\beta}(X, \omega) Z_{\alpha\beta}(X, \omega) \delta(\omega - \omega'),$$

where $T_{\alpha\beta}$ is the frequency-dependent noise temperature defined by the spatial average of the temperature field $T(r)$ performed with the response tensor $\phi_{\alpha\beta}$

$$T_{\alpha\beta}(X, \omega) = \frac{\int_V \phi_{\alpha\beta}(X, r, \omega) T(r) \, d^3 r}{\int_V \phi_{\alpha\beta}(X, r, \omega) \, d^3 r}.$$

[1] E. H. Hauge and A. Martin-Löf, J. Stat. Phys. 7, 259 (1973).