The colour of thermal noise in classical Brownian motion: a feasibility study of direct experimental observation

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Abstract. One hundred years after Einstein modelled Brownian motion, a central aspect of this motion in incompressible fluids has not been verified experimentally: the thermal noise that drives the Brownian particle, is not white, as in Einstein’s simple theory. It is slightly coloured, due to hydrodynamics and the fluctuation–dissipation theorem. This theoretical result from the 1970s was prompted by computer simulation results in apparent violation of Einstein’s theory. We discuss how a direct experimental observation of this colour might be carried out by using optical tweezers to separate the thermal noise from the particle’s dynamic response to it. Since the thermal noise is almost white, very good statistics is necessary to resolve its colour. That requires stable equipment and long recording times, possibly making this experiment one for the future only. We give results for experimental requirements and for stochastic errors as functions of experimental window and measurement time, and discuss some potential sources of systematic errors.

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1. Introduction: Einstein’s theory

It is well known that Einstein did not know about Brownian motion when he first formulated what is known as his theory for it [1]. He was looking for observable consequences of what was then called the molecular-kinetic theory of heat. So he was not concerned about the finer details of specific situations. In fact, apart from dated mathematical language, his papers on Brownian motion [2] remain paradigms for how to model the essence of a phenomenon with ease and transparency by leaving out everything that can possibly be left out.

The simplest version of his theory,

\[ \dot{x}(t) = (2D)^{\frac{1}{2}} \eta(t), \]  

for the trajectory \( x(t) \) of a Brownian particle, here in one dimension, works so well also for real experimental situations that its extreme simplicity may be overlooked: there is no way to simplify this theory further. The white-noise process \( \eta(t) \) is the simplest possible,

\[ \langle \eta(t) \rangle = 0; \quad \langle \eta(t) \eta(t') \rangle = \delta(t - t'), \]  

and when normalized as here, a constant with dimension like a diffusion coefficient must appear where it does. The factor 2 in front of \( D \) in equation (1) follows from the mathematical equivalence between this Langevin equation and the diffusion equation in which \( D \) was already defined before Einstein’s 1905 work. The new physics was in Einstein’s relation

\[ D = k_B T/\gamma_0 \]  

between diffusion constant, Boltzmann energy, and Stokes friction coefficient \( \gamma_0 \) for a spherical particle,

\[ \gamma_0 = 6\pi \rho \nu R. \]  

Here \( \rho \) is the density of the fluid, \( \nu \) its kinematic viscosity, and \( R \) is the sphere’s radius.
2. The Einstein–Ornstein–Uhlenbeck theory

Details left out in the model described in equations (1)–(4) will be found missing, of course, if one looks in the right places. For example, the length of the trajectory $x(t)$ is infinite for any finite time interval considered. Ornstein and Uhlenbeck showed that this mathematical infinity does not appear in an equation introduced by de Haas-Lorentz in her thesis [3, 4], Newton’s second law

$$m \ddot{x} = -\gamma_0 \dot{x} + F_{\text{thermal}},$$

(5)

where $m$ is the inertial mass of the Brownian particle, and the force from the surrounding medium is written as a sum of two terms: Stokes friction, $-\gamma_0 \dot{x}$, and a random thermal force $F_{\text{thermal}} = \sqrt{2k_BT\gamma_0} \eta(t)$ with ‘white noise’ statistical properties following from equation (2). The random motion resulting from equation (5) is known as the Ornstein–Uhlenbeck process. In the limit of vanishing $m$, Einstein’s theory is recovered. Together, they make up the Einstein–Ornstein–Uhlenbeck theory of Brownian motion.

The OU-process improves Einstein’s simple model for Brownian motion by accounting for inertia, but, as pointed out by Lorentz [5], only when the particle’s density is much larger than the fluid’s. When particle and fluid densities are comparable, as in the motion Brown observed, hydrodynamical effects that the OU-process ignores, are more important than the inertial effect of the particle’s mass. These effects are the frequency-dependence of friction and the inertia of entrained fluid. Stokes obtained the friction coefficient, equation (4), for motion with constant velocity [6]. Brownian motion is anything but that. Also, mass and momentum of the fluid entrained by a sphere doing rectilinear motion with constant velocity is infinite according to Stokes solution to Navier–Stokes equation [6, 7]. This gives a clue that entrained fluid matters, and the pattern of motion too.

3. Stokes friction for a sphere in harmonic rectilinear motion in an incompressible fluid

The friction coefficient that is relevant for a more correct description of Brownian motion, differs from the friction coefficient that is usually associated with Stokes’ name, equation (4), but is also the subject of [6]. Stokes was not addressing the hydrodynamics of Brownian motion in 1851, but the hydrodynamics of an incompressible fluid surrounding a sphere that does rectilinear harmonic motion with no-slip boundary condition, at vanishing Reynolds number, and with the fluid at rest at infinity. However, since this hydrodynamics is linear, any trajectory of a spherical particle can be written as a linear superposition of Stokes periodic solutions by virtue of Fourier analysis [8], as long as the condition of vanishing Reynolds number is satisfied by the trajectory. For a Brownian particle’s trajectory it is, so the following result is fundamental for Brownian motion.

In general, when a rigid body moves through a dense fluid like water, the friction between body and fluid depends on the body’s past motion, since that determines the fluid’s present motion. For a sphere performing rectilinear harmonic motion $x(t; f)$ with cyclic frequency $\omega = 2\pi f$ in an incompressible fluid and at vanishing Reynold’s number, Stokes found the ‘frictional’
force [6, 9]

\[ F_{\text{fric}}(t; f) = -\gamma_0 \left( 1 + \frac{R}{\delta(f)} \right) \dot{x}(t; f) - \left( 3\pi \rho R^2 \delta(f) + \frac{2}{3} \pi \rho R^3 \right) \ddot{x}(t; f) \]

\[ = -\gamma_{\text{Stokes}}(f) \dot{x}(t; f); \quad (6) \]

\[ \gamma_{\text{Stokes}}(f) \equiv \gamma_0 \left( 1 + (1 - i) \frac{R}{\delta(f)} - i \frac{2R^2}{9\delta(f)^2} \right), \quad (7) \]

where only the term containing \( \dot{x}(t; f) = -i2\pi f x(t; f) \) dissipates energy, while the term containing \( \ddot{x}(t; f) = -(2\pi f)^2 x(t; f) \) is an inertial force from entrained fluid. The notation is the same as above: \( \gamma_0 \) is the friction coefficient of Stokes’ law for rectilinear motion with constant velocity, equation (4). The penetration depth \( \delta \) characterizes the exponential decrease of the fluid’s velocity field as a function of distance from the oscillating sphere. It is frequency dependent,

\[ \delta(f) \equiv (\nu/\pi f)^{1/2} = R(f_{\nu}/f)^{1/2}, \quad (8) \]

and large compared to \( R \) for the frequencies we shall consider. For a sphere with diameter \( 2R = 1.0 \mu m \) in water at room temperature where \( \nu = 1.0 \mu m^2 s^{-1}, f_{\nu} \equiv \nu/(\pi R^2) = 1.4 \text{ MHz}. \)

Note that the mass of the entrained fluid, the coefficient to \( \dddot{x} \) in equation (6), becomes infinite in the limit of vanishing frequency \( f \), i.e., the flow pattern around a sphere moving with constant velocity has infinite momentum, according to Stokes’ steady-state solution to Navier–Stokes’ equations.

**4. Beyond Einstein: Brownian motion in an incompressible fluid**

The friction on a sphere that, without rotating, follows an arbitrary trajectory \( x(t) \) with vanishing Reynolds number in an incompressible fluid that is at rest at infinity, is found by Fourier decomposition of \( x(t) \) to a superposition of rectilinear oscillatory motions \( \tilde{x}(f) \). Using equation (6) on these, gives

\[ \tilde{F}_{\text{fric}}(f) = -\gamma_{\text{Stokes}}(f)(-i2\pi f)\tilde{x}(f), \quad (9) \]

which Fourier transforms back to [8],

\[ F_{\text{fric}}(t) = -\gamma_0 \dddot{x} - \frac{2}{3} \pi \rho R^3 \ddot{x}(t) - 6\pi \rho R^3 f_{\nu}^{1/2} \int_{-\infty}^{t} dt' (t - t')^{-1/2} \dddot{x}(t'). \quad (10) \]

So the Langevin equation for Brownian motion of a micro-sphere in an incompressible fluid reads [10, 11]

\[ m\dddot{x}(t) = F_{\text{fric}}(t) + F_{\text{ext}}(t) + F_{\text{therm}}(t), \quad (11) \]

where \( F_{\text{ext}} \) denotes all external forces on the sphere, such as gravity or optical tweezers, and \( F_{\text{therm}} \) denotes the random thermal force on the sphere from the surrounding fluid.
Several authors have derived expressions for the thermal force using different arguments and finding the same result

\[ \tilde{F}_{\text{therm}}(f) = (2k_B T \text{Re} \gamma_{\text{Stokes}}(f))^{1/2} \tilde{\eta}(f); \]  

(12)

see overviews in [12, 13]. Briefly, Brownian motion in a fluid is the result of fluctuations in the fluid described by fluctuating hydrodynamics [7]. In this theory one assumes that the random currents split up into systematic and random parts, the latter obeying a fluctuation–dissipation theorem. From this theory one derives the expression of the thermal force on a sphere in the fluid.

Equations (10)–(12) constitute the accepted hydrodynamically correct theory for classical Brownian motion, i.e., in an incompressible fluid. The form of the thermal force remains a theoretical result, however, a consequence of fluctuating hydrodynamics. It is not a controversial result, it is not questioned. But because it is a small effect, it has not been demonstrated experimentally.

This theory was prompted into existence by computer simulations by Alder and Wainwright [14, 15]. They simulated a fluid of hard-sphere molecules, and observed power-law tails in the velocity auto-correlation function of individual spheres, apparently in conceptual contradiction with Einstein’s theory.

5. Power-law tails

In the absence of external forces, the position power spectrum of Brownian motion following from equations (10)–(12) is

\[ P(f) \propto \langle |\dddot{\mathbf{x}}|^2 \rangle \propto \frac{2k_B T \text{Re} \gamma_{\text{Stokes}}(f)}{|m(2\pi f)^2 + i2\pi f \gamma_{\text{Stokes}}(f)|^2}. \]  

(13)

Here, the frequency-dependent numerator is the power spectrum of the thermal force in equation (11), while the denominator is given by the other terms in equation (11). The frequency-dependent friction coefficient, \( \gamma_{\text{Stokes}}(f) \), appears both in numerator and denominator, and both appearances contribute, with opposite signs, to the \( t^{-3/2} \) power-law tail in the velocity auto-correlation function.

By Wiener–Khintchine’s theorem, the velocity auto-correlation function is

\[ \phi(t) = \langle \dot{\mathbf{x}}(t) \dot{\mathbf{x}}(0) \rangle \propto \int_{-\infty}^{\infty} df e^{-i2\pi f (2\pi f)^2 P(f)}. \]  

(14)

At asymptotically large values of \( t \), \( \phi(t) \) is given by \( P(f) \)'s behaviour at small values of \( f \),

\[ (2\pi f)^2 P(f) = 2D(1 - \sqrt{f/f_v} + \mathcal{O}(f/f_v)). \]  

(15)

Hence

\[ \phi(t) = \frac{D}{2\pi f_v^{1/2}} t^{-3/2} + \mathcal{O}(t^{-5/2}) \quad \text{for} \quad t \to \infty, \]  

(16)
quite different from the exponential decrease following from Einstein’s simple theory, but not conceptually different from it [10, 11, 16].

Experimental evidence for this power-law tail remained sparse for years. Dynamic light scattering offered promise of its observation, but only Boon and Boullier [17, 18] reported an experimental result of the magnitude predicted theoretically, with statistical errors about half the size of the signal. Paul and Pursey used photon correlation dynamic laser light scattering to measure the time dependence of the mean-squared displacement of polystyrene spheres with radius \( R \sim 1.7 \mu m \) [19]. They found clear evidence for the expected \( \tau^{-3/2} \) -behaviour (\( \tau^{1/2} \) in the mean-squared displacement), but with an amplitude of only 74 \( \pm \) 3% of that predicted theoretically. They never found the reason why 26% of the theoretically expected amplitude was missing [20]. Ohbayashi et al [21] also used photon correlation spectroscopy, on a suspension of polystyrene spheres with radius 0.80 \( \mu m \), and found agreement between the theoretical amplitude of the \( \tau^{-3/2} \) tail and their experimental results which has 9–10% error bars. Their results also agree with the predicted significant temperature dependence. This convincing experiment thus demonstrates the validity of the theory, equation (11). This is the current experimental status of the power-law tail of the velocity auto-correlation function of classical Brownian motion.

The amplitude that was measured, albeit indirectly, in these experiments, is the coefficient to the first-order term in the expansion above of \( P(f) \) in powers of \( (f/f_c)^{1/2} \). This coefficient has a contribution from the denominator, from Stokes frequency-dependent friction coefficient, and another, half-as-large contribution with opposite sign from the numerator’s, the noise term’s, frequency dependence.

Instead of measuring a photon correlation function for laser light scattered off a suspension of microspheres, developments in instrumentation [22] and data analysis [23] for optical tweezers have made it possible to measure this amplitude directly for a single microsphere [24]. Here, we point out that the issue of the ‘colour’ of the noise, the frequency dependence of its non-white power spectrum, never has been addressed separately, but now may be within reach with a rather challenging single-particle experiment with optical tweezers.

6. How to isolate the noise term experimentally

When a microsphere is held in an optical trap with trapping force \( F_{ext}(x) = -\kappa x \) while its position \( x(t) \) is recorded for a long, but finite time \( T_{msr} \), the resulting experimental power spectrum of its Brownian motion is [23]

\[
P_k^{(ex)} = \frac{|\tilde{x}_k|^2}{T_{msr}} = \frac{2k_BT \Re \gamma_{Stokes}(f_k)|\tilde{\eta}_k|^2}{|\kappa - i2\pi f_k\gamma_{Stokes}(f_k) - m(2\pi f_k)^2|^2}, \tag{17}
\]

Here, \( \tilde{x}_k \) and \( \tilde{\eta}_k \) are the Fourier transforms of \( x(t) \) and \( \eta(t) \) on the finite time interval of recording.

\[
\tilde{x}_k = \int_{-T_{msr}/2}^{T_{msr}/2} dte^{i2\pi f_k t}x(t), \quad f_k \equiv k/T_{msr}, \quad k \text{ integer.} \tag{18}
\]

From equation (2) it then follows that

\[
\langle \tilde{\eta}_k \rangle = 0; \quad \langle \tilde{\eta}_k^* \tilde{\eta}_\ell \rangle = T_{msr}\delta_{k,\ell}; \quad \langle |\tilde{\eta}_k|^4 \rangle = 2T_{msr}^2, \tag{19}
\]
and, since the real and imaginary parts of $\tilde{\eta}_k$ are uncorrelated, Gaussian distributed, real random variables, $(|\tilde{\eta}_k|^2)_{k=1,2,...}$ are uncorrelated non-negative random variables with exponential distribution. Consequently, $(P^{(\text{ex})}_k)_{k=1,2,...}$ are uncorrelated non-negative exponentially distributed random variables, each of which consequently has RMSD equal to its expectation value. This expectation value is

$$P_{\text{hydro}}(f) = \langle P^{(\text{ex})}_k \rangle = \frac{D/(2\pi^2)(1 + (f/f_\nu)^{1/2})}{(f - f^3/2)^2/f^2 - f^2/f_{m^*})^2 + (f + f^{3/2}/f_{m^*})^2},$$

where the corner frequency $f_c = \kappa/(2\pi\gamma_0)$ characterizes the strength of the trapping force, and $f_{m^*} \equiv \gamma_0/(2\pi m^*) \simeq 3f_c/2 = 1.9$ MHz as $m^* \equiv m + 2\pi\rho R^3/3 \simeq 3m/2$ for a polystyrene bead with diameter 1 $\mu$m.

The low-frequency limit of this power spectrum is

$$P_{\text{hydro}}(f) = \frac{2D}{(2\pi f_c)^2} \frac{1 + (f/f_c)^{1/2}}{1 + O(f^{3/2}/(f_{m^*}^{3/2}f_c)).}$$

Here, the frequency-dependent numerator is the power spectrum of the thermal force, the coloured thermal noise, in the Langevin equation (11). It is a first-degree polynomial in $f^{1/2}$. The numerator, on the other hand, is just a constant, $f_c^2$, to leading order in $f^{1/2}$, and to first and second order in $f^{1/2}$. In other words, in the low-frequency limit considered here, the dynamics of the microsphere is not seen when it is held in a strong trap. Only the thermal force that bounces it about in the trap, is seen in the power spectrum. The sphere itself moves adiabatically with the force so that it always is at a position in the trap where the trapping force exactly balances the ‘instantaneous’ value of the thermal force. We write ‘instantaneous’ in quotes because we are discussing a low-frequency approximation, so ‘instantaneous’ means ‘instantaneous with the limited time resolution we have at this low frequency.’ This qualitative explanation has a quantitative underpinning in equation (11). The physical meaning of the approximation in equation (21) that leaves the denominator constant, equal to $f_c^2$, is: it corresponds to neglecting the inertial force on the left-hand side of equation (21) relatively to the other terms—e.g., the friction force—entirely as usual, and to neglecting also this friction force relatively to the trapping force. This gives, with $F_{\text{ext}}(t) = -\kappa x(t)$, the approximate equation

$$\kappa x(t) = F_{\text{therm}}(t),$$

which shows that once we have calibrated the trap, found $\kappa$, e.g., as in [23], then we can record the thermal force simply by recording $x(t)$: the microsphere in the trap has become a force-meter, with the microsphere playing the role of the pointer arrow, and its displacement from the trap’s centre is a calibrated linear measure of the ‘instantaneous’ thermal force. The time resolution for which this scenario holds, corresponds to $f \ll f_c$, as seen from equation (21).

Popular speaking, we have used optical tweezers to separate the thermal forces acting on the microsphere from the dynamic response of the sphere. In practice, this might be used to demonstrate the coloured noise experimentally as follows.

The constant $P_{\text{hydro}}(0) = 2D/(2\pi f_c)^2$ can be determined experimentally with high precision by fitting equation (20) to the experimental power spectrum [23]. That done, the value of this constant can be divided into the experimental spectral values. After subtraction of 1, one has left
Figure 1. Double-log plot of $P_{\text{hydro}}(f)/P_{\text{hydro}}(0) - 1$ for the optimal realistic case of microsphere diameter $2R = 1 \mu m$ and $f_c = 5$ kHz. Dotted line, leading-order approximation $(f/f_\nu)^{1/2}$; full line, exact expression from equation (20).

a residue with expectation value

$$\frac{P_{\text{hydro}}(f)}{P_{\text{hydro}}(0)} - 1 = (f/f_\nu)^{1/2} + \mathcal{O}(f^{3/2}/(f_\nu^{1/2} f_c)),$$  \hspace{1cm} (23)

i.e., a simple power-law relationship to leading order. Thus, when plotted with double-log axis, this re-scaled and subtracted version of the experimental power spectrum should fall on a straight line with slope $1/2$ and known intercept given by $f_\nu$, as illustrated in figure 1.

Whether this behaviour is demonstrated convincingly or not, depends on the theory being correct, of course, but also very much on the signal-to-noise ratio in the experimental power spectrum, one has recorded. Figure 1 shows that in order to observe one decade of power law behaviour where it is most easy, in the interval (20 Hz, 200 Hz), one must be able to distinguish $P(f)/P(0)$ from 1 to within 1 per mil. This precision is achievable in a power spectrum of the kind discussed here [23], but it has not been demonstrated that it can be obtained at the very low frequencies of interest here, where low-frequency noise external to the experiment contributes significantly to the power spectrum (see figure 7B in [23]) unless extreme care is taken to suppress it.

Also, the lower the frequency at which one wants the spectral value with precision, the longer one must measure, when the goal, as here, is a set of spectral values equidistantly spaced on the logarithmic frequency axis (see figures 5A and 6A in [23]). If, e.g., one wants to achieve 1 per mil precision on a data point at 20 Hz by ‘blocking’ experimental power spectral values in the interval (10 Hz, 40 Hz), then one needs $10^6$ power spectral values in that interval, since non-averaged experimental power spectral values scatter exponentially about their expectation value, i.e., with RMSD equal to the expectation value. If one records $x(t)$ for a time $T_{\text{inst}}$, Fourier transformation as in equation (18) yields an experimental power spectral value at a discrete set of frequencies $f_k$ equidistantly separated by $\Delta f = 1/T_{\text{inst}}$. So to get $10^6$ such frequencies in an interval of length 30 Hz, one must measure for a time $T_{\text{inst}} = 10^6 (30 \text{ Hz})^{-1} = 9$ h. Alternatively,
one may use ‘windowing’ and average many independent experimental power spectra to achieve a similar noise reduction, or a combination of both methods [23]. Either way, or combining ‘blocking’ and ‘windowing,’ one must measure for 9 h. That done, one has also, from the same recording, data points at higher frequencies, and with better precision, if they are equidistantly spaced on the logarithmic frequency axis, as one wants them here (see figures 5A and 6A in [23]).

This long measurement is by necessity and with advantage broken into many much, much shorter windows of measurement. But the experiment is not allowed to drift much in the course of those 9 h, not w.r.t. quantities affecting the spectrum, i.e., quantities affecting \( f_\nu \). These are \( R \), the radius of the sphere, and \( \nu \), the kinematic viscosity of the fluid used. So changing sphere during the experiment is not a good idea, and holding on to one sphere for 9 h is a challenge. Temperature control is also a must, as \( \nu \) is quite dependent on temperature in the case of water. Temperature control may also be a challenge as a strong trap is needed, and it heats the fluid near the sphere [25].

Figure 2 shows the same function as figure 1, but in a log-linear plot, together with the function one would get if the thermal noise were white in equation (11). If one sets the less ambitious goal of distinguishing experimentally between white and predicted coloured noise, less precision is needed, say 1%, and one needs only record 5 min worth of Brownian motion.

7. Conclusion

We have pointed out how one may measure the power spectrum of the coloured thermal noise of classical Brownian motion, and we have described the requirements on data with regards to
the amount of data necessary in order to get the proper signal-to-noise ratio. The experiment is demanding and a challenge for the best optical tweezers labs, if it is possible at all, and there are probably other practical experimental problems than those described here, problems that are better left for experimenters to describe and, hopefully, tackle.

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