BROWNIAN MOTION AT ABSOLUTE ZERO*

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Abstract

We derive a general quantum formula giving the mean-square displacement of a diffusing particle as a function of time. Near 0 ^{o}K we find a new universal logarithmic behavior (valid for times longer than the relaxation time), and deviations from classical behavior can also be significant at larger values of time and temperature. Our derivation depends neither on the specific composition of the "heat-bath" nor on the strength of the coupling between the bath and the particle. An experimental regime of micro-seconds and micro-degrees Kelvin would elicit the pure logarithmic diffusion.

The so-called "fluctuation-dissipation" theorem—which relates the thermal fluctuations of a variable x to the response of that variable to a weak external force—is usually described as generalizing the Smoluchowski-Einstein relation for Brownian motion, $D = kT\mu$; but it is not easy to find in the literature any explicit derivation of this relation as a direct corollary of the theorem. In this paper we will provide such a derivation under the assumption that the times involved are

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long compared to the relaxation time τ, as defined below. But, because the fluctuation-dissipation theorem is really a quantum-mechanical relationship, it will tell us something more than just the laws of classical diffusion, which will emerge only in the limit $\hbar \to 0$, or equivalently in the limit of long times and high temperatures. In the opposite limit where $kT\,\Delta t \ll \hbar$, the usual linear dependence $\Delta x^2 \sim \Delta t$ will turn out to give way to a new universal behavior $\Delta x^2 \sim \ln \Delta t$, which probably should be interpreted as a diffusion driven by quantal zero-point motions rather than by thermal kinetic energy. The logarithmic behavior will follow from a general formula (12) for $\langle \Delta x^2 \rangle$, which will hold for all times long compared to τ, given the assumption of constant mobility μ. In what follows we will derive this general formula, discuss the limiting cases just alluded to, and show that some deviations from classical behavior may be observable on the basis of current experimental technique.

In recent years there have been several efforts [1-4] to understand the dynamics of a quantum particle coupled to a heat bath. Insofar as our work overlaps those efforts, our results appear to agree. The main difference is that the cited papers make far-reaching assumptions about the nature of the medium ("heat-bath") in which the particle moves, and require the coupling between particle and bath to be linear (meaning in effect that the coupling is weak). In contrast, we only use that the *response* to a weak *external* perturbation is linear, allowing the coupling of the particle to the bath/environment itself to be strong, as it will in fact be in most situations. On the other hand we will predict only the mean-square displacement, whereas the more special treatments can in principle yield the full density operator as a function of Δt.

The Fluctuation-Dissipation Theorem in the Time-Domain

The fluctuation-dissipation theorem, as usually stated, refers to the fourier transforms of the auto-correlation and response functions. Let $x(t)$ be some dynamical variable (q-number) in the Heisenberg picture, and let $f(t)$ be an infinitely weak external force applied to x at time t. (We will not need the more general form of the theorem in which the external coupling is to a different variable y.) The response function $R(t)$ is defined by the relation

$$\langle x(t)\rangle_f - \langle x\rangle_0 = \int R(t-s)f(s)ds, \tag{1}$$

where $\langle\cdot\rangle_f$ denotes expectation-value in the presence of the force, assuming the system of which x is a variable to have been in thermal equilibrium with temperature T at early times; and $\langle\cdot\rangle_0$ is the same expectation-value for zero force. Also let

$$C(t) = \frac{1}{2}\langle x(t)x(0) + x(0)x(t)\rangle$$

be the "autocorrelation" or "two-point" function in equilibrium at temperature T. (Or, if you prefer, you can subtract off $\langle x(t)\rangle\langle x(0)\rangle = \langle x(0)\rangle^2$ from this definition without invalidating what follows. This would be equivalent to working with $x-\langle x\rangle$ in place of x.) Then the fluctuation-dissipation theorem [5] stated in the frequency domain is (with $\beta = 1/kT$)

$$\mathrm{Im}\,\widetilde{R}(\nu) = \hbar^{-1}\tanh(\pi\beta\hbar\nu)\widetilde{C}(\nu). \tag{2}$$

[We are using the following definition of fourier transform $\mathcal{F} \equiv \widetilde{(\cdot)}$:

$$\widetilde{\phi}(\nu) = \int dt\, 1^{\nu t}\,\phi(t)^*,$$

where $1^x \equiv e^{2\pi i x}$.]

Our first job is to transform this relation to the time-domain. To that end, let us introduce in place of $R(t)$ (which vanishes for $t < 0$ by virtue of causality) the equivalent odd function

$$\check{R}(t) = \mathrm{sgn}(t) R(|t|).$$

It is then easy to check that $2i \,\mathrm{Im}\, \mathcal{F}(R) = \mathcal{F}(\check{R})$, whence (2) can be written in the equivalent form:

$$\mathcal{F}(\check{R}) = \frac{2i}{\hbar} \tanh(\pi\beta\hbar\nu) \mathcal{F}(C). \tag{3}$$

(In fact it is actually this form, rather than (2) that comes out initially in the most straightforward derivation of the fluctuation-dissipation theorem; it is thus more appropriate to view (2) as a consequence of (3) than vice versa.) By taking the fourier transform of (3) we could now express $R(t)$ as a convolution of $C(t)$, but our main interest here is to do the opposite. Let us therefore solve (3) for $\widetilde{C}$, obtaining

$$\widetilde{C}(\nu) = (-i\hbar/2) \coth(\pi\beta\hbar\nu) [\mathcal{F}(\check{R})](\nu) + c\delta(\nu), \tag{4}$$

where c is a constant and where, for definiteness, the principal part of coth may be taken. [The ambiguity in $1/\tanh(\pi\beta\hbar\nu)$ is just a term proportional to $\delta(\nu)$, which would drop out of (4) anyway, since it would be multiplying the odd function $\mathcal{F}(\check{R})$.] The fourier transform of (4) reads

$$C = \frac{i\hbar}{2} \mathcal{F}(\coth(\pi\beta\hbar\nu)) * \check{R} + c, \tag{5}$$

determining C, up to an additive constant, in terms of the fourier transform

$$\mathcal{F}(\coth \pi\beta\hbar\nu) = (i/\beta\hbar) \coth(\pi t/\beta\hbar). \tag{6}$$

In equation (6), the coth on the right-hand-side is also to be understood as a principal part, but unlike before, this choice is forced on us, because the addition of any $\delta(t)$-piece to $\coth \pi t/\beta\hbar$ would spoil its oddness, in disagreement with the oddness of the left-hand-side of (6). Understanding all coth's to be principal parts, then, we have finally (in view also of the definition of $\check{R}$), the following explicit formula for $C(t)$ in terms of $R(t)$:

$$C(t) = \frac{1}{2\beta} \int_{-\infty}^{\infty} dt' \mathrm{sgn}(t'-t) R(|t'-t|) \coth(\pi t'/\beta\hbar) + c. \tag{7}$$

[The appearance of the undetermined constant c is due to the possibility of redefining the zero of x without affecting (1). By working with the alternative definition of $C(t)$ mentioned just before equation (2), we would remove this ambiguity, and correspondingly could set $c = 0$, given some assumptions on the asymptotic behavior of x and R.]

The mean-square displacement $< \Delta x^2 >$

Now the mean-square displacement of x due to equilibrium fluctuations in time Δt is $< \Delta x^2 >$, where $\Delta x := x(t+\Delta t) - x(t)$. Taking $t = 0$ for convenience, we have (since the equilibrium state is time-independent) $< \Delta x^2 > = < (x(\Delta t) - x(0))^2 >$ $= < x(\Delta t)^2 > + < x(0)^2 > - < \{x(\Delta t), x(0)\} > = 2C(0) - 2C(\Delta t)$, or

$$\frac{1}{2} < \Delta x^2 >= C(0) - C(\Delta t). \tag{8}$$

Combining this result with (7) gives us a general equation for $\langle \Delta x^2 \rangle$ in terms of the response function R:

$$\frac{1}{2} < \Delta x^2 >= \frac{1}{2\beta} \int_0^{\infty} dt' R(t') \left[2 \coth \Omega t' - \coth \Omega(t'+t) - \coth \Omega(t'-t)\right], \tag{9}$$

where for brevity we have set $\Omega = \pi/\beta\hbar$. Notice that the undetermined constant c in (7) has dropped out of this result.

Quantum Brownian motion

At this stage, let us specialize x to be a cartesian coordinate of an otherwise free particle immersed in a homogeneous medium with temperature T. For an idealized inertia-less Brownian particle, the response to a weak external force would be immediate motion at velocity $v = \mu f$, μ being the "mobility"; in other words, R would be the step function $R(t) = \mu\theta(t)$. However this idealization is plainly too unrealistic, because it leads to a divergent result in (9). [In this sense we might say that the fluctuation-dissipation theorem knows that particles have inertia!] A more reasonable Ansatz for R must incorporate a "relaxation time" or "rise time" τ representing the time it takes the particle to accommodate itself to any sudden change in $f(t)$. Such an Ansatz is, for example,

$$R(t) = \mu(1 - e^{-t/\tau})\theta(t), \tag{10}$$

which describes the classical motion of a particle subject to viscous friction. Without making so specific a choice however, we will employ a cruder cutoff which should be adequate for times much greater than τ:

$$R(t) = \mu\theta(t - \tau). \tag{11}$$

With this R, (9) can be integrated exactly (using the distributional identity, PP(coth x) $= d/dx \ln\sinh|x|$) to produce the following fundamental equation of quantum Brownian motion:

$$\frac{1}{2} < \Delta x^2 >= \frac{\mu\hbar}{\pi} \ln \frac{\sqrt{\sinh\Omega|t-\tau|\sinh\Omega|t+\tau|}}{\sinh\Omega\tau} \qquad (\Delta t \gg \tau), \tag{12}$$

where again $\Omega := \pi/\beta\hbar$.

Now strictly speaking, there is the inconsistency in our derivation of (12) that $C(t)$ is ill-defined for a particle moving in an unbounded space, because $\langle x^2 \rangle$ in equilibrium would be infinite, and (8) would therefore assume the indeterminate form $\langle \Delta x^2 \rangle = \infty - \infty$. To overcome this problem, one could confine the particle in a very long "box" (confining potential), it being intuitively clear that this could alter neither $\langle \Delta x^2 \rangle$ nor $R(t)$ in the limit of an infinitely large such box.

Three limiting cases of the general formula (12)

The possible limiting cases of (12) are determined by the relative magnitudes of the three times τ, $\beta\hbar$ and Δt, which we may call respectively the relaxation time, the "quantum time", and the "diffusion time". A priori, there would be essentially 3!=6 distinct cases, but since we must have $\Delta t \gg \tau$ in order to apply (12), we will limit ourselves to only three of them. [It is nonetheless instructive to notice that (12) becomes self-contradictory for Δt near τ since it then equates an intrinsically positive expression to a negative right-hand-side. This implies that (11) could not be the exact response function for any system, even in principle. More generally, one can derive from (7) and the definition of $C(t)$, a positivity criterion which any putative response function must fulfill in order to be physically viable. We do not know how restrictive this criterion is in practice, but we have checked that the R of (10) yields a mean-square displacement which is non-negative for all times, as one might have expected.]

Case 1 $\beta\hbar \ll \tau \ll \Delta t$

This is the classical limit, and (12) reduces to the classical relation

$$\frac{1}{2} < \Delta x^2 >= (\mu/\beta)\Delta t = \mu k T \Delta t, \tag{13}$$

or $\mu k T(\Delta t - \tau)$ if the leading correction is retained.

Case 2 $\tau \ll \Delta t \ll \beta\hbar$

This is the extreme quantum limit, in which (time)(energy)$\ll \hbar$ for the time-scale set by the diffusion time Δt and the energy-scale set by the thermal energy kT. In this limit (12) reduces to

$$\frac{1}{2} < \Delta x^2 >= \frac{\mu\hbar}{\pi} \ln \frac{\Delta t}{\tau}, \tag{14}$$

or

$$\frac{\mu\hbar}{\pi} \ln \sqrt{(\frac{\Delta t}{\tau})^2 - 1}$$

if somewhat more precision is desired. It is noteworthy that the temperature has disappeared entirely from this expression (except insofar as it influences μ and τ), suggesting a quantum Brownian motion due entirely to "zero-point" fluctuations, which are present even at absolute zero. Indeed, the striking logarithmic dependence in (14) could also have been derived by first taking the zero-temperature limit of the fluctuation-dissipation theorem itself, and only then applying it to the R of a diffusing particle.

Case 3 $\tau \ll \beta\hbar \ll \Delta t$

Intermediate between cases 1 and 2, this situation might be described as one in which the relaxation occurs on quantum time-scales, although the diffusion-time itself is already classically long. [A suggestive way to rewrite the inequality $\tau \ll \beta\hbar$ is as the relation between diffusion constants, $D_{\mathrm{classical}} \ll D_{\mathrm{quantum}}$, where $D_{\mathrm{classical}} = \mu/\beta$, and $D_{\mathrm{quantum}} = \hbar/m$, with m taken from the "viscous damping" relation $\tau = \mu m$ envisaged in (10).] In this case (12) reduces to

$$\frac{< \Delta x^2 >}{2} = \frac{\mu \Delta t}{\beta} + \frac{\mu\hbar}{\pi} \ln \frac{\beta\hbar}{2\pi\tau}, \tag{15}$$

which one can interpret as the result of a two-stage spreading which follows the quantum law (14) up to the time $t_Q := \beta\hbar/2\pi$, and thereafter continues according to the classical law (13), with the second term in (15) remaining forever as a kind of residue of the quantum era. In order for this residue to be significant, we need $\mu\Delta t/\beta \lesssim (\mu\hbar/\pi)\ln(\beta\hbar/2\pi\tau)$, or

$$\Delta t \lesssim \frac{\beta\hbar}{\pi} \ln \frac{\beta\hbar/\pi}{2\tau},$$

which can occur non-trivially (i.e. without reducing to case 2) only if $\beta\hbar/\tau$ is exponentially big, so that $\ln(\beta\hbar/\tau) \gtrsim \Delta t/\beta\hbar \gg 1$. Taking $m = \tau/\mu$ as earlier, this amounts to a requirement that the particle be extremely light:

$$m \lesssim (\beta\hbar/\mu)e^{-\frac{\pi\Delta t}{\beta\hbar}}. \tag{16}$$

Remarks and numerical estimates

Equations (13), (14) and (15) are all special cases of the more general relation (12), which should be valid whenever $\tau \ll \Delta t$. In other situations, or for more general response-functions $R(t)$, one must refer back to (9) itself, from which the spreading can always be computed as long as $R(t)$ is known. A particularly interesting response function to treat would be (10), and another interesting case might be a particle moving in a superfluid.

In the zero-temperature limit, i.e. in case 2 above, our formula (14) may be compared with a result of Ambegaokar [4], who used a path-integral formalism, and assumed a linear coupling between the particle and an environment comprising an infinite collection of harmonic oscillators. He obtained an expression for the mean-square displacement of a Brownian particle in the quantum regime which corresponds to our result given in (14), if we make certain identifications. According to Ambegaokar (with a presumed misprint corrected),

$$< (\Delta x^2) >= ((h/\pi^2)/\gamma m)\, ln|(t\sqrt{\omega_c\gamma})| \,+\, const., \tag{17}$$

for $(1/\gamma) < t < (\beta\hbar)$. Here, $\langle \Delta x^2 \rangle$ is given by a density operator ρ which reduces to a delta-function at $t = 0$, and ω_c defines an upper frequency cutoff beyond which the linear relationship between particle velocity and environmental friction breaks down. Also, judging from equation (5.3) of [4], it appears natural to identify $1/\gamma$ with our τ, and therefore $1/m\gamma$ with our μ. If we do so, and also equate ω_c to τ^{-1}, then we recover (14) from (17) with the constant set to zero.

In connection with (14) one can ask the following question: classically, what kind of response function would lead to a logarithmic law of diffusion? If we take the $\hbar \to 0$ limit of (9), we find that the relevant response function should be proportional to $1/t$, which is physically impossible. This implies that the effect described by (14) is of purely quantum mechanical origin.

Finally, let us estimate the thresholds of time and temperature at which significant deviations from classical behavior should appear. In order to be in the "pure quantum regime", we need $\Delta t \ll \beta\hbar$, which can also be written in the time-energy form, $kT\Delta t \ll \hbar$. Taking $T \sim 10^{-6}$ deg (cf. [6]) and $\Delta t \sim 10^{-6}$sec yields $kT\Delta t/\hbar \sim 0.1$, which ought to be well within the "pure quantum regime", meaning that (14) should apply if the relaxation time is short enough (and the "reservoir" in thermal equilibrium). For higher temperatures or longer times, deviations of the sort described by (15) might be observable if τ is small enough and a condition like (16) is satisfied.

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