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The Langevin equation

L'équation de Langevin

Yves Pomeau [a,*], Jarosław Piasecki [b]

[a] *University of Arizona, Department of Mathematics, Tucson, USA*
[b] *Faculty of Physics, University of Warsaw, Poland*

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ABSTRACT

The existence of atoms has been long predicted by philosophers and scientists. The development of thermodynamics and of the statistical interpretation of its concepts at the end of the nineteenth century and in the early years of the twentieth century made it possible to bridge the gap of scales between the macroscopic world and the world of atoms. Einstein and Smoluchowski showed in 1905 and 1906 that the Brownian motion of particles of measurable size is a manifestation of the motion of atoms in fluids. Their derivation was completely different from each other. Langevin showed in 1908 how to put in a coherent framework the subtle effect of the randomness of the atomic world, responsible for the fluctuating force driving the motion of the Brownian particle and the viscosity of the "macroscopic" flow taking place around the same Brownian particle. Whereas viscous forces were already well understood at this time, the "Langevin" force appears there for the first time: it represents the fluctuating part of the interaction between the Brownian particle and the surrounding fluid. We discuss the derivation by Einstein and Smoluchowski as well as a previous paper by Sutherland on the diffusion coefficient of large spheres. Next we present Langevin's short note and explain the fundamental splitting into a random force and a macroscopic viscous force. This brings us to discuss various points, like the kind of constraints on Langevin-like equations. We insist in particular on the one arising from the time-reversal symmetry of the equilibrium fluctuations. Moreover, we discuss another constraint, raised first by Lorentz, which implies that, if the Brownian particle is not very heavy, the viscous force cannot be taken as the standard Stokes drag on an object moving at uniform speed. Lastly, we examine the so-called Langevin–Heisenberg and/or Langevin–Schrödinger equation used in quantum mechanics.

RÉSUMÉ

L'existence des atomes a été prédite depuis longtemps par savants et philosophes. Le développement de la thermodynamique et son interprétation par la mécanique statistique, à la fin du XIX^{e} et au début du XX^{e} siècle, ont rendu possible le comblement de l'écart entre l'échelle spatiale du monde macroscopique et celle des atomes. En 1905 et 1906, Einstein et Smoluchowski montrent, selon deux approches complètement différentes, que

* Corresponding author.
E-mail address: pomeau@lps.ens.fr (Y. Pomeau).

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le mouvement brownien de particules de taille mésoscopique mesurable directement est une manifestation du mouvement incessant des atomes dans le fluide environnant. Peu après, en 1908, Langevin montre comment mettre dans un cadre cohérent l'effet des fluctuations aléatoires du monde atomique, qui font mouvoir la particule brownienne, et la viscosité du fluide régissant les mouvements macroscopiques de cette même particule brownienne pour les ralentir. Nous examinons les méthodes de déduction d'Einstein et de Smoluchowski ainsi qu'un article antérieur de Sutherland sur la diffusion d'une solution de sphéres mésoscopiques dans un liquide. Nous présentons ensuite la note de Langevin aux *Comptes rendus de l'Académie des sciences*, en insistant sur la division fondamentale entre force aléatoire et force visqueuse. Ceci nous amène à différentes questions, telles que les contraintes à satisfaire par la force de Langevin et les généralisations de l'équation de Langevin. Nous insistons sur les contraintes issues de la réversibilité en temps des fluctuations d'équilibre. Nous discutons aussi une remarque de Lorentz montrant que, si la particule brownienne n'est pas très dense, on ne peut utiliser la formule de la traînée de Stokes pour une vitesse constante dans l'équation de Langevin. Finalement, nous examinons ce qu'on appelle la théorie de Schrödinger–Langevin (ou de Heisenberg–Langevin) en mécanique quantique.

1. Introduction

The note by Langevin [1] on Brownian motion had a lasting impact on scientific research and is still often cited because of two things. First it puts in a mathematically coherent frame the effect of thermal fluctuations on macroscopic dynamics, then it opens the way to many extensions to situations other than the one of Brownian particles diffusing in fluids at equilibrium. In his note, Langevin refers to two papers on the same topic (Brownian motion) by Einstein [3] and by Smoluchowski [4]. Both works made a connection between equilibrium fluctuations and viscous friction to find the diffusion coefficient of a "large" Brownian particle in a fluid. A former paper by Sutherland (seemingly unknown to Langevin) gives also an explicit and correct expression for the diffusion coefficient of large particles in a fluid at equilibrium, without referring to Brownian motion. It is not without interest to explain how Einstein and Smoluchowski derived their result, obviously without using the, then non-existent, Langevin equation. For reasons explained below, we begin with Sutherland derivation. As a side remark, we notice that Einstein does use the words "Brownian motion" only once in the introduction of his original publication, whereas the words are in the title of Langevin's note. Smoluchowski is more precise and refers to the movements observed by the naturalist Robert Brown in 1827 and continues by saying that Einstein's theory is in agreement with the observation by Brown. The first reference to the phenomenon of Brownian motion and its link with the existence of atoms is much older (and amazingly prescient). It goes back to Lucretius in *De rerum natura* (*On the Nature of things*) written in about 60 BC. Lucretius said rightly (in latin verses...) that the random motion of dust particles observed in a beam of light is evidence of the perpetual motion existing in the world of atoms. The observations by the Scottish naturalist Robert Brown were published in 1827. Contrary to Lucretius, they are reported without reference to the atoms. Jan Ingenhousz is also credited for having observed in 1785 the irregular motion of particles of coal on the surface of alcohol. From the historical point of view, *De rerum natura* played a major role in inspiring Pierre Gassendi (first half of the seventeenth century) in his discussion of the atomic structure of matter, which inspired Newton and Laplace. Recently, Kahane [7] wrote a comment, published online, on the note by Langevin. The point of view of this comment is different from ours. It focuses on the mathematics initiated by Langevin's paper. It is also of interest to mention another publication in the *Comptes rendus* on Brownian motion by Gouy [5] as well as the well-known book by Perrin[1] on atoms.

1.1. Sutherland

Sutherland, an Australian scientist at the University of Melbourne, published in 1904 and 1905 [2] two papers on the diffusion coefficient of large biomolecules in a fluid. He used both Stokes' law of drag for a sphere in a viscous fluid and the concept of osmotic pressure to derive the diffusion coefficient of such large biomolecules. His result is consistent with what was found slightly later by Einstein and Smoluchowski. Even though Sutherland derived his result first, he can hardly be seen as a forerunner of Einstein, Smoluchowski, and Langevin, because he never refers to Brownian motion. This derivation is of interest because of its shortness, because it uses somehow a fluctuation–dissipation approach of this problem and because it is rather close to but far less detailed than Einstein's derivation. It is based on the (non-trivial) idea that osmotic

[1] In the reference J. Perrin, C. R. hebd. Séanc. Acad. Sci. Paris CXLVII (1908) 530 & 532, Perrin explains how he measured the Avogadro number N by measuring the diffusion of Brownian particles. His result is $N = 5.4{\cdot}10^{23}$. In another paper, J. Perrin, C. R. hebd. Séanc. Acad. Sci. Paris CXLVII (1908) 475 & 476, he shows the experimental validity of Stokes' drag law for the particles he uses. The matter of both papers as well as many other things are in his classical book: J. Perrin, Les atomes, 1913, http://gallica.bnf.fr/ark:/12148/bpt6k373955h.

pressure is a force field exerted on the foreign molecules diffusing in the medium, if they are not distributed uniformly in space. This balance of forces (gradient of osmotic pressure and viscous drag on the diffusing molecules) yields the average velocity of the particles, a velocity making the diffusive flux of concentration.

Sutherland uses a slightly extended version of Stokes' formula taking into account the so-called Navier boundary condition on the surface of the spheres, namely a partial sliding of the fluid on this surface. We shall not keep this possibility and take the standard Stokes formula with the no-slip boundary condition. He then considers a concentration gradient of identical spheres ∇c. At low concentration, this yields a gradient of osmotic pressure $RT\nabla c$ where R is the perfect gas constant and T the absolute temperature. This osmotic pressure yields a force per unit volume acting on the large molecules. Sutherland assumes that this force is balanced by the Stokes friction (per unit volume) between the spheres and the surrounding fluid. This yields the following condition for the balance of forces per unit volume:

$$RT\nabla c = -6\pi a\eta \mathbf{V} N \tag{1}$$

where N (same notation as Sutherland) is the number density of the large spheres of radius a, $\mathbf{V}$ is their velocity with respect to the solvent and η the shear viscosity of this solvent. He compares now this expression with the familiar expression for the flux of particles by diffusion in a medium with a non-uniform concentration. This flux is $\mathbf{V}N$. It is also proportional to the diffusion coefficient D of the spheres times minus the gradient of concentration. Therefore (in modern notations, namely with k_B instead of a ratio R/C with C Avogadro number):

$$D = \frac{k_B T}{6\pi a\eta} \tag{2}$$

The result of this elegant derivation is compared by Sutherland with experimental data. He introduces a quantity denoted as B, which is the volume occupied by the large molecules, but this volume is not known otherwise and so this formula does not give access to the Avogadro number, contrary to Perrin's experiments where the diffusion coefficient is measured for small spheres of known radius and compared with Einstein and Smoluchowski formulae.

As had been noted by Smoluchowski [4], the derivations of the diffusion coefficient by Sutherland and by Einstein (given below) assume the balance of two forces, the gradient of osmotic pressure, and the viscous drag of particles drifting in the solvent. Without saying it, this mixes two kind of "forces", the osmotic pressure derived from a condition of stationarity of equilibrium and another force, the Stokes drag, derived for a dynamical process, with no obvious connection with an equilibrium condition. Such a mixing of equilibrium and non-equilibrium forces appears elsewhere in the mechanics of continuous media: to take an example, one can think to the moving contact line problem [8], where capillary forces (equilibrium forces) and viscous stress (a "dynamical" force) act together in a rather complex (and still disputed) way. In this respect, the microscopic approach of Smoluchowski is a priori free of this kind of difficulty and yields at the end results consistent with what was found by Sutherland and Einstein.

1.2. Einstein

The basic idea of Einstein [3] for computing the diffusion coefficient of a dilute suspension is quite similar to the one of Sutherland. Unlike Sutherland, he derives explicitly various results for the osmotic pressure of dilute solutions, essentially the one stating that the gradient of osmotic pressure of a dilute suspension is $RT\nabla c$, a result taken as given by Sutherland. Then Einstein, like Sutherland, writes that at the steady state the gradient of the osmotic pressure is equal to the number density of the molecules times the drag force exerted on each particle due to its velocity relative to the solvent. This yields the above Eq. (1) again – Einstein's equation number (18) and (19). From this, Einstein derives the same expression of the diffusion coefficient given in Eq. (2) above. At this point, the papers of Einstein and of Sutherland become different. As already said, Sutherland derives the order of magnitude of the diffusion coefficients of solutions of large molecules and compares them with experimental data. On the contrary, Einstein spends the rest of his paper making a connection between the diffusion coefficient he has found and the behavior of a *single* diffusing object, namely a Brownian particle. He emphasizes the meaning of this point: although thermodynamics and the mechanics of continuous media deal with assemblies of many molecules, Brownian motion concerns a single object and, because it is observable, its observation makes a bridge between the microscopic and the macroscopic worlds.

Einstein derives first the diffusion coefficient of a single object making small random jumps (he restricts himself to what happens along one coordinate). He shows that the dynamics of this particle follows a diffusion law if the average jump is zero. The order of magnitude of the distance run during time t grows like $(2Dt)^{1/2}$. Using now the diffusion coefficient for the suspension of spheres he has computed, Einstein gives an order of magnitude estimate for $(2Dt)^{1/2}$. For a sphere of radius of 0.8 micrometers in water at 17°C, he obtains a displacement of about 6 micrometers after one minute, measurable with the technologies of the time.

Einstein ends up his paper by saying rightly that the result of this measurement would be important for the theory of heat, referring presumably to the still ongoing discussions on the nature of heat as a possible manifestation of the energy stored in the motion of molecules.

1.3. Smoluchowski

The Smoluchowski approach is based on the assumption that the observed motion of a Brownian particle of mass M results from collisions with molecules of the fluid with much smaller mass $m \ll M$. His description of the phenomenon of Brownian motion starts thus at a purely microscopic level involving the elastic collision law between spherically symmetric particles. Smoluchowski estimates the typical value of the Brownian particle velocity v using the equipartition law

$$v = c\sqrt{\frac{m}{M}}$$

where c is the thermal velocity of fluid particles.

In order to perform effectively the analysis, simplifying approximations are adopted based on order of magnitude considerations. For instance, the velocity of the Brownian particle, $v = |\mathbf{v}|$, is supposed not to be affected by collisions, their only effect showing up in sudden changes of the direction of motion.

Smoluchowski's microscopic approach permits to analyze separately two distinct cases, depending on whether the radius of the Brownian particle a is small or large with respect to the mean free path λ of the fluid particles.

- When $a \ll \lambda$, one can consider consecutive collisions of the Brownian sphere M as statistically independent events, and neglect their influence on the velocity distribution within the fluid. Smoluchowski assumes then that the length of the free trajectories of mass M between collisions is always the same, equal to its mean free path l (another simplifying approximation). His aim is to evaluate the mean square distance covered by mass M after $N = \nu t$ collisions (ν denotes the collision frequency). So, he has essentially to solve a well-defined random walk problem where the Brownian particle performs N steps of length l, each making some small angle ϵ with the previous step. His estimation of the value of ϵ on the basis of the elastic collision law yields

  $$\epsilon = \frac{3mc}{4Mv} = \frac{3v}{4c}$$

  It is also supposed that all directions corresponding to the angle ϵ are equally probable.
  After laborious calculations, Smoluchowski successfully solves the problem and arrives at the formula

  $$< (\Delta\mathbf{r}(t))^2 >= \frac{64}{9}c^2\frac{t}{\nu}$$

  which is valid for sufficiently long times. The root-mean-square displacement of the Brownian particle turns out to be independent of its mass, and proportional to the square root of time in the long-time limit ($N = \nu t \to \infty$). This is the main message of Smoluchowski's analysis when $a \ll \lambda$, whereas the value of the numerical factor in the above formula clearly results from a number of simplifying approximations.
- When the radius a of the Brownian particle is large compared to the mean free path λ of the surrounding fluid, collisions cannot be treated as sequences of independent events. The relatively simple random walk picture is not applicable any more. In order to make the analysis tractable, Smoluchowski considers the exponential decay law of the velocity of a sphere put in a viscous fluid. If the initial velocity is $\mathbf{v_0}$, the component along the direction of $\mathbf{v}_0$ after time t is reduced to

  $$v_0 \exp\left(-\frac{t\zeta}{M}\right)$$

  where ζ is the friction coefficient. If $a \gg \lambda$ and the velocity is not too high, one can use the Stokes formula:

  $$\zeta = 6\pi\eta a$$

  The law above should be looked upon as describing the evolution of the initial velocity towards the small value v given by the equipartition law.
  Smoluchowski supposes that during the relaxation time $\tau = M/\zeta$, the motion of mass M can be approximately treated as rectilinear. Finally, he uses the important hypothesis that the motion of the massive Brownian particle is like that of a gas particle whose mean free path equals $v\tau$, and which follows a zigzag trajectory composed of segments of fixed length oriented in random directions. Such a problem has been solved by Smoluchowski in a paper published in French in 1906, yielding for the mean square displacement the formula:

  $$< (\Delta\mathbf{r}(t))^2 >= 2\frac{l^2}{\tau}t$$

  Inserting here for the mean free path $l = v\tau$, one finds:

  $$< (\Delta\mathbf{r}(t))^2 >= 2v^2\tau t = c^2\frac{2m}{\zeta}t = c^2\frac{m}{3\pi\eta a}t$$

In order to relate this formula to the result derived for $a \ll \lambda$, Smoluchowski multiplies the right-hand side by (64/27), arriving finally at the formula:

$$< (\Delta \mathbf{r}(t))^2 >= \frac{64}{81} c^2 \frac{m}{\pi \eta a} t$$

He remarks that the numerical factor here is in fact unimportant, as it reflects many simplifying assumptions made in the course of the analysis. However, the fundamental message is qualitatively correct, the same as that derived by Einstein: both approaches predict the diffusive character of the Brownian motion. Smoluchowski's theory permits to relate it to the microscopic dynamics by using effectively probabilistic methods.

1.4. Lorentz criticism of Einstein's derivation and other questions

Before to comment Langevin's article, we note a point made by Lorentz, which is relevant for all early papers on the subject, namely the use of the Stokes formula for steady motion. We shall return to this question after having introduced and discussed Langevin's note.

In the many papers on the theory of Brownian motion, it is rarely pointed out that its validity (unnoticed by Einstein, Smoluchowski, and Langevin) requires that the mass density of the "large" Brownian sphere is much larger than the one of the surrounding fluid, although in most real situations the two mass densities are quite close to avoid sedimentation in the field of gravity. Lorentz [6] was the first one to notice this point in lectures published in 1921.

This can be explained as follows. The decay of the velocity due to the viscosity of a sphere with initial velocity V_0, radius a and mass M in a fluid of mass density ρ_f and shear viscosity η is ruled by the equation

$$M \frac{\mathrm{d}V}{\mathrm{d}t} = -\zeta V$$

where $\zeta = 6\pi a\eta$ is still the Stokes friction coefficient. The decay time for V derived from this equation is $\tau \sim \frac{\rho_s a^2}{\eta} = \frac{\rho_s}{\rho_f}\tau_v$, where τ_v is the decay time of the vortex of size a under the action of viscosity and ρ_s the mass density of the Brownian sphere, supposed to be homogeneous. The (implicit) assumption behind the choice of a constant (as opposed to a time convolution, see below) friction implies $\tau >> \tau_v$, equivalent to $\rho_s >> \rho_f$. If this inequality is not satisfied and if one substitutes for the instantaneous friction a time convolution, the end result for the diffusion coefficient remains the one by Einstein and Sutherland, but the decay of the velocity fluctuation is not given by Langevin's equation with a constant friction. In particular, it is not exponential with respect to time and decays at large times with a time power $t^{-3/2}$ in 3D and $1/t$ in 2D, so that the Einstein integral for the self diffusion coefficient does not converge there [9]. Notice that this is also consistent with the fact that there is no Stokes coefficient for a disc in 2D, because there the drag is proportional to the velocity of the particle times the logarithm of the Reynolds number.

The diffusion coefficient of a single particle is related to the time correlations of the velocity. This is shown as follows. The displacement of the particle between time $t = 0$ and t is related to its velocity $\mathbf{v}(t)$

$$\Delta \mathbf{r}(t) = \int_0^t \mathrm{d}t' \mathbf{v}(t') \tag{3}$$

Compute now the average value of the squared displacement between time 0 and t:

$$< (\Delta \mathbf{r}(t))^2 >= \int_0^t \int_0^t \mathrm{d}t' \mathrm{d}t'' < \mathbf{v}(t') \cdot \mathbf{v}(t'') >$$

where the dot indicates a scalar product. Because of the invariance under time translation of the time-dependent fluctuations, the average $< \mathbf{v}(t') \cdot \mathbf{v}(t'') >$ depends on the time difference $(t' - t'')$ only. Changing the integration variable from (t', t'') to $(t', t_0) = (t', t' - t'')$, one finds

$$< (\Delta \mathbf{r}(t))^2 >= 2t \int_0^t \mathrm{d}t_0 < \mathbf{v}(0) \cdot \mathbf{v}(t_0) >$$

$< (\Delta \mathbf{r}(t))^2 > /6t$ in the long time limit tends to the diffusion coefficient of the particle. This yields the expression of the diffusion coefficient due to Einstein:

$$D = \frac{1}{d} \int_0^\infty \mathrm{d}t_0 < \mathbf{v}(0) \cdot \mathbf{v}(t_0) >$$

where d is the dimension of space. This integral diverges in 2D, because the velocity correlation decays like $1/t$ at large values of t.

The divergence of the (more general) Green–Kubo integrals for the transport coefficients is even stronger in 1D. By an extension of the fluctuation integrals, it is predicted to be like the integral of $t^{-1/2}$. A more detailed discussion shows [9] that the Green–Kubo integrand decays like $t^{-2/3}$ instead, as observed numerically.

2. Langevin's note

Langevin's paper is short and clear. He considers, as did Einstein, the law of motion of a single "large" sphere in a fluid made of atoms at equilibrium. That the sphere is "large" means that it is much bigger than any microscopic length scale. Langevin splits the force felt by the Brownian particle in two: one part is the Stokes drag, assumed to be proportional to the instantaneous speed. The other is a random force $\mathbf{X}(t)$, representing the effect of the random collisions of the atoms in the fluid, keeping the energy of the particle equal on average to its thermal value. Langevin does not characterize the Langevin force very accurately, except for this constraint of maintaining the average kinetic energy so that this force has zero mean value and is uncorrelated to the position of the particle. Indeed there was no Dirac delta function at the time of this publication, and it was not even possible to write the now standard formula

$$< X_i(t)X_j(t') >= 2\zeta\delta_{ij}\delta(t-t')k_{\mathrm{B}}T \tag{4}$$

where δ_{ij} is the Kronecker delta, whereas i and j stand for the Cartesian indices, and $\delta(t-t')$ is Dirac delta. Nowadays, one imposes to the Langevin force that $\mathbf{X}(t)$ has Gaussian statistics. The only constraint, although quite indirect, is that the random force $\mathbf{X}$ must be such that it keeps the average thermal energy of the Brownian sphere and that the fluctuations of the force are statistically independent of the velocity of the particle. We comment this (non-trivial) point below.

The Langevin equation as written by Langevin (for one Cartesian component of the velocity and of the Langevin force) is:

$$M\frac{\mathrm{d}V}{\mathrm{d}t} = -\zeta V + X(t) \tag{5}$$

It is of interest to show how Langevin derives the diffusion coefficient of his equation. As did Einstein, he considers what happens along one coordinate only, the direction x, and writes $x(t)$ for the position of the Brownian particle in this direction. The equation becomes:

$$M\frac{\mathrm{d}^2x}{\mathrm{d}t^2} + \zeta\frac{\mathrm{d}x}{\mathrm{d}t} = X(t) \tag{6}$$

Multiplying both sides by $x(t)$ and taking the average, he rewrites it as:

$$\frac{M}{2} < \frac{\mathrm{d}^2x^2}{\mathrm{d}t^2} > -M < V^2 >= -\frac{\zeta}{2} < \frac{\mathrm{d}x^2}{\mathrm{d}t} > + < xX(t) > \tag{7}$$

where x is the coordinate along the x-axis. Langevin assumes now that the Langevin force is uncorrelated with the position (namely that $< xX >= 0$), he writes that "obviously" (*evidemment* in French), $< xX(t) >= 0$ because of the "irregularity of the complementary actions", this referring to the force X which has been defined before as "complementary" of the viscous force, meaning that it is a fluctuation existing whatever is this force. Taking now $z =< \frac{\mathrm{d}x^2}{\mathrm{d}t} >$, he obtains a simple ordinary differential equation for z:

$$\frac{M}{2}\frac{\mathrm{d}z}{\mathrm{d}t} + \frac{\zeta}{2}z = k_{\mathrm{B}}T \tag{8}$$

This uses the equipartition theorem $M < V^2 >= k_{\mathrm{B}}T$ (in modern terms) written for each Cartesian component of the speed. Solving now Eq. (8), Langevin finds that for times much longer than $\frac{M}{\zeta}$, the solution z tends to a constant, which is equivalent to $< x^2(t) >$ growing linearly with time:

$$< x^2(t) > - < x^2(0) > \approx \frac{2k_{\mathrm{B}}T}{\zeta}t$$

In this way, Einstein's result is recovered. To a modern reader, this derivation, however brilliant it is, does not tell clearly the reason behind the splitting of the force on the Brownian particle into a viscous force and a random force. To say, as done often (but not by Langevin), that the random force takes into account the random collisions of the fluid particles is not fully convincing, we believe: in a hard spheres fluid, for instance, the interaction of the fluid with the Brownian particle will be all in collisions of the fluid particles with the Brownian sphere, including therefore the viscous friction *and* the Langevin random force. A way of escaping this difficulty is to assume that the statistical properties of the random Langevin force can, in principle, be derived by assuming a *fixed* sphere at zero speed, compute all statistical properties of the force exerted on it and assume that the fluctuations of this force are exactly the one of the Langevin force $X(t)$. Such a force is

statistically independent of the velocity *before* it is observed. Therefore, as assumed by Langevin, this force is uncorrelated with the position, which is the time integral of the velocity before the observation. Indeed, after this force had an effect on the motion of the Brownian sphere, the velocity of this sphere is correlated with the Langevin force before the velocity is observed. Somehow, as often in non-equilibrium statistical physics, one has to introduce a kind of asymmetry in the time direction: there this asymmetry arises from an assumption about the correlation between the velocity and the Langevin force. In Boltzmann's kinetic theory, it is introduced by the so-called Stosszahlansatz, which assumes that particle velocities are uncorrelated *before* a binary collision.

In non-equilibrium systems, this asymmetry has measurable consequences, but at equilibrium this does not lead to an observable asymmetry under time reversal. Such a symmetry under time reversal is a fundamental element of Onsager's proof of the symmetry of transport coefficient. On average and at equilibrium, the velocity fluctuations described by Langevin's equation are symmetrical under time reversal, and no observation on them can allow one to distinguish between the two possible directions of time. This makes hopefully more convincing the assumption of statistical independence of the Langevin force at a given time and of the velocity before. In subsection 2.1 below, we discuss a simple model where the splitting between a random force and a macroscopic drag force follows from a analysis of the microscopic dynamics.

How to reconcile this analysis by Langevin with the criticism by Lorentz? A first remark is that, if the Brownian particle is much larger than any microscopic length (mean freepath in a dilute gas, interparticle distance in a dense fluid), the random force is not due to random impacts of particles on the Brownian sphere: if it was so, the statistics of this random force would not be linked to the shear viscosity of the fluid. It would be made of random hits, a case which could be studied within Smoluchowski's theory. This link with viscosity shows that the random force is due to fluctuations of the fluid surrounding the sphere, fluctuations having a collective behavior linked to the fluids' equations. Indeed, those fluctuations ultimately result from the discreteness of the particles making this fluid. This is what is represented by the fluctuating hydrodynamics of Landau and Lifshitz [10], which is inspired by Langevin's theory and amounts to add a random force to the Stokes equations to take into account the existence of particles at micro scale. Once this is done one can, in principle, solve the (linear) fluid equations including such random forces and compute the Langevin force on a macroscopic object, a random force that will include the viscosity because it is derived from the fluids' equations.

Of course, a question remains: will the random Langevin force computed this way have the statistical properties given by the correlation functions in equation (4) above?

2.1. Origin of the dissipative and fluctuating forces acting on the Brownian particle: a microscopic approach

Here we present the derivation from the microscopic dynamics of the splitting of the force acting on the Brownian particle into dissipative and fluctuating parts [11]. The model system is composed of a flat wall of surface area L^2 and mass $M = \sigma L^2$ that separates an infinite cylindrical volume into two infinite parts filled with a gas of non-interacting particles of mass $m \ll M$. At the initial moment $t = 0$, the wall is at rest, and the semi-infinite volumes on its right and on its left are at thermal equilibrium with the same pressures, but different temperatures. The further frictionless motion of the wall for $t > 0$ is entirely induced by elastic collisions with surrounding gas particles. The wall is oriented perpendicularly to the symmetry axis of the cylinder, and has only one translational degree of freedom. The stochastic process followed by mass M can be shown to correspond exactly to the Brownian motion and its analysis shows well the splitting between the Langevin force and a "viscous" friction, both coming from the random collisions with the gas particles.

Suppose that at time $t_n = n\Delta$, the velocity of the wall is V_n (Δ is a time interval). In order to calculate the value of V_{n+1}, we use the binary collision law

$$V' = (1-\epsilon)V + \epsilon v \tag{9}$$

where V and v are precollisional velocities of the wall and the gas particle. There appears here a small parameter

$$\epsilon = \frac{2m}{m+M} = \frac{2m}{m+\sigma L^2} \ll 1$$

Let us introduce the stochastic variable $k_n =$, which is the number of collisions within the time interval (t_n, t_{n+1}), and let us denote by v_j the precollisional velocity of the gas particle at the jth encounter. The iterative use of the collision law yields then the formula

$$V_{n+1} = (1-\epsilon)^{k_n} V_n + \epsilon \sum_{j=1}^{k_n} (1-\epsilon)^{k_n - j} v_j$$

When $\epsilon \ll 1$, the above formula takes the form:

$$V_{n+1} = \exp(-\epsilon k_n) V_n + \epsilon \sum_{j=1}^{k_n} v_j \exp[-\epsilon(k_n - j)]$$

The further analysis of the stochastic process governing the evolution of the velocity V can be rigorously performed assuming the scaling of the time interval $\Delta \sim L^{-\gamma}, 0 < \gamma < 2$. For instance, the asymptotic evaluation of the term $\exp(-\epsilon k_n)V_n$ in the limit $L \to \infty$ yields the formula

$$V_n \exp(-\epsilon k_n) \sim V_n \exp(-\frac{2m}{\sigma}\alpha\Delta)$$

where

$$\alpha = \frac{1}{2}[\rho^- < |v| >^- + \rho^+ < |v| >^+]$$

and ρ^-, ρ^+ and $< |v| >^-, < |v| >^+$ are the initial densities and mean speeds of the gas particles to the left and to the right of the wall, respectively. We find here the source of the dissipative contribution describing for long times the exponential decay of the velocity of the moving piston.

The final result of a rather long derivation of the normalized probability density $\chi(V;t)$ for finding the massive Brownian wall with velocity V at time t, which takes additionally into account thermal fluctuations of the gas precollisional velocities, takes the form

$$\chi(V;t) = N \exp\left\{-\frac{2\alpha V^2}{\delta\epsilon[1-\exp(-4\alpha\epsilon L^2 t)]}\right\}$$

N is the normalizing factor, and

$$\delta = \frac{1}{2}[\rho^- < |v|^3 >^- + \rho^+ < |v|^3 >^+]$$

The role of the dynamical friction coefficient is played by the quantity

$$2\alpha\epsilon L^2 = \frac{2m}{\sigma}[\rho^- < |v| >]^- + \rho^+ < |v| >]^+]$$

The distribution $\chi(V;t)$ approaches, in the long time limit, a Maxwell distribution with temperature T_∞. In the case of the initial thermal equilibria on both sides of the Brownian wall with possibly different temperatures T_- and T_+, respectively, one finds

$$T_\infty = \sqrt{T_- T_+}$$

The connection with the Langevin splitting into a random force and friction force shows up in Eq. (9) for the collision law: at each such collisions, a little bit of the memory of the velocity of the piston is erased (this is the friction term), whereas a random contribution to its speed is added, which depends on the velocity of the incoming gas particle. In the case of a gas of interacting particles (instead of the present model of non-interacting particles), one must substitute to this analysis a more complex one. This is based upon the Landau–Lifshitz equations adding thermal fluctuations to the linear Stokes equations. The drag force computed by solving the Stokes equation without the fluctuations yields the viscous force proportional to the velocity of the particle, whereas the force due to the fluctuating part of the Landau–Lifshitz equations yields the Langevin fluctuating force. Of course, this makes a derivation more complex than the one of the model of this subsection, but it keeps the same splitting of the force into a viscous-like part and a random Langevin force.

3. A first physical constraint on Langevin's equation

Langevin initiated many developments, in part because he introduced a rather straightforward method to take into account fluctuations in dynamical problems, fluctuations having various sources, not always in the discreteness of matter. It is out of question to review all those developments, or even part of them. We shall comment below some points in this field, which we believe deserve attention.

We shall focus mostly on questions related to the modeling of physical phenomena where the discreteness of matter is the source of fluctuations. A good example of questions posed by the extension of the Langevin theory is the case where the friction is not viscous friction, namely not proportional to the velocity. This arises when one considers the Brownian motion of a particle of mesoscopic size diffusing on a solid surface whilst remaining close to it. Then the viscous drag proportional to the velocity is usually replaced by a friction law where the drag is not proportional to the velocity. This leads to major theoretical problems. A model of solid friction often used in this field amounts to add to the viscous drag $-\zeta V$ a Coulomb friction, which is a constant times a discontinuous and strongly nonlinear function of the velocity, something like $(-\nu\frac{\mathbf{V}}{V})$. As shown [13] by Goldenfeld and collaborators (see also references [14] and [15] for the Langevin equation with Coulombic solid friction), the equilibrium distribution function of the velocity fluctuations for such a friction law with the usual white noise term is not Maxwellian. This forbids us to consider this model as a fair description of the equilibrium Brownian motion of a particle on a solid surface, but does not exclude at all that it is relevant for other situations. The requirement that the equilibrium fluctuations of the velocity have a Maxwellian distribution is central to a picture of equilibrium. Similar

questions (yet unsolved) arise when dealing with the thermal fluctuations in electric circuits including a diode leaving the current to flow in one direction only. In such cases, presumably, the splitting into a viscous "friction" and a random Langevin-like force statistically independent does not hold anymore. It should also be stressed that adding a Langevin-like force with vanishing mean value to non-linear equations is not consistent in the sense that taking then the average, one does not recover the original equations because of the induced correlations.

3.1. Langevin's equation in an external potential and anomalous diffusion

Even though the Langevin equation in its original form is linear, it is possible, however, without violating the constraint of reversibility, to extend it to a nonlinear equation. This was shown in a general form by Kolmogorov [21] and particularized later [19], when it was extended to the Brownian motion in an external potential. This subsection explains how the Langevin equation can be rightfully extended to "nonlinear" situations that could explain the observed anomalous Brownian motion. This amounts to add to the right-hand side of the Langevin equation a force equal to minus the gradient $\nabla\Phi$ of a potential $\Phi(\mathbf{r})$, where $\mathbf{r}$ is the position of the Brownian particle. This changes Langevin equation into a pair of equations:

$$M\frac{\mathrm{d}\mathbf{V}}{\mathrm{d}t} = -\zeta\mathbf{V}(t) - \nabla\Phi + \mathbf{X}(t) \tag{10}$$

and

$$\mathbf{V} = \frac{\mathrm{d}\mathbf{r}}{\mathrm{d}t} \tag{11}$$

This remark could be of interest because it has been observed recently [20] that Brownian motion in a rather complex setting may yield a squared displacement that increases linearly with time, like in all standard diffusion processes, but with a non-Gaussian probability distribution of the position (reference [17] reports careful recent observations of the regular Brownian motion). The results reported in [20] could perhaps be explained by using the Langevin equation written above with a potential $\Phi(\mathbf{r})$, consistent with the constraint of reversibility: the complex surrounding of the Brownian particle could be represented by a random landscape of this potential $\Phi(\mathbf{r})$ with randomly distributed wells of depth bigger than $k_{\mathrm{B}}T$. If this happens the Brownian motion is dominated by the time spent near the bottom of the potential wells. Of course, the particle gets out of those traps after a long escape time proportional to an Arrhenius factor. Supposing that the probability distribution of the random depth of the potential wells is not limited from below, there is a competition between the time spent in the wells and the time devoted to the exploration of the landscape outside of the wells. The more time is spent doing this exploration, the bigger is the probability of getting trapped in a deeper well. Over a certain length of time, it is not obvious to decide what amount of time is spent outside of the deepest well met during this exploration and inside. The balance between the two is found by comparing the escape time of the deepest well and the time spent in the regular exploration. If one assumes that there is still a regular diffusion-like process, this tells that a volume $(Dt)^{3/2}$ is explored in time t. This is to be compared with the escape time of the deepest well in this volume, depending itself for t large on the tail of the distribution of depth of the wells. If this escape time is much longer than t, this implies that the exploration of the landscape is not by diffusion, and the opposite on the contrary. It could be that the experiments reported in [20] concern an intermediate regime where the two time scales (escape and regular diffusion) are of the same order of magnitude. This could be in agreement with the fact that the probability distribution of the positions decays more slowly than a Gaussian at large values, because the large distances are more sensitive to the exploration of the landscape outside of the potential wells and so contribute more to the probability distribution than the small ones and so contribute positively to the probability. Said otherwise, those large distances outside of the deep potential wells are explored with a diffusion coefficient much larger than the true diffusion coefficient, because this one includes long stays inside the deep wells.

3.2. Langevin equation with non instantaneous friction

Returning to the standard Brownian motion of a sphere of mesoscopic size in a fluid, let us consider the case where the mass densities of the sphere and of the surrounding fluid are of the same order of magnitude (the relevant literature can be found in the reference [12]). By reproducing Lorentz' argument, we have shown that if the two mass densities are of the same order, the viscous force cannot be replaced anymore by its standard Stokes expression. One must take into account that the friction force is history dependent. This is done by replacing in the Langevin equation the instantaneous friction by a convolution term:

$$M\frac{\mathrm{d}\mathbf{V}}{\mathrm{d}t} = -\int_0^t \mathrm{d}t'\,\Gamma(t-t')\mathbf{V}(t') + \mathbf{X}(t) \tag{12}$$

In this equation, the kernel $\Gamma(t')$ is to be derived by solving Stokes' equation. This was done by Stokes himself and is explained by Landau in *Fluid Mechanics* at the end of section 24 (*Oscillations in a viscous fluid*) as problem number 5. A convenient version of the "non-local" Langevin equation (12) for a sphere can be written by doing part of the integral on the

right-hand side. This splits into three terms. One of the terms is just the regular Stokes friction, another one represents the inertia of the mass of fluid $M_f = \frac{2}{3}\pi a^3 \rho_f$ carried by the motion of the sphere, ρ_f being the mass density of the fluid. Only the first one on the right-hand side (called sometimes the Basset term) remains as a time integral. The result is:

$$M\frac{d\mathbf{V}}{dt} = -\zeta\left(a^2\frac{\rho_f}{\pi\eta}\right)^{1/2}\int_0^t dt'\frac{(\mathbf{V}(t')-\mathbf{V}(t))}{(t-t')^{3/2}} - \zeta\mathbf{V} - M_f\frac{d\mathbf{V}}{dt} + \mathbf{X}(t) \tag{13}$$

It shows that the time integral $\int_0^\infty dt'\Gamma(t')$ is the friction coefficient ζ. For an arbitrary $X(t)$ and initial condition $V(0)$, this can be solved to give [16]:

$$V(t) = A(t)V(0) + \int_0^t dt' A(t')X(t-t') \tag{14}$$

The function $A(t)$ is given as a function of the kernel $\Gamma(t)$ by using the Laplace transform. Let $f_L(s) = \int_0^\infty dt e^{-st} f(t)$ be the Laplace transform of the function of time $f(t)$. The Laplace transform of $A(.)$ is given by:

$$A_L(s) = \frac{1}{s+\Gamma_L(s)}$$

Multiplying equation (14) by $V(0)$ and taking the average on the Langevin force, which is independent of the velocity $V(0)$, one readily finds that

$$< V(t)V(0) >= A(t) < V(0)^2) >$$

Because the Langevin force is independent of the velocity $V(0)$, the average square of $V(t)$ is the sum of two terms, one proportional to the square of $V(0)$, the other to the square of the Langevin force:

$$< V^2(t) >= A^2(t) < V^2(0) > + \int_0^t dt' \int_0^t dt'' A(t')A(t'') < X(t-t')X(t-t'') > \tag{15}$$

This yields a condition relating the correlation $< X(t-t')X(t-t'') >=< X(0)X(t'-t'') >$ and the functions $A(.)$ and $B(.)$, both being derived a priori from the solution of the Stokes equation for a non-uniform velocity. This condition expresses that the equality $< V^2(t) >=< V^2(0) >$ holds for any time t.

A derivation of the Langevin equation for a sphere in a fluid described by the linearized Navier–Stokes (or Stokes) equation is given by Hauge and Martin-Lof [22], including the effect of the Landau–Lifshitz fluctuating part. This paper shows the validity of the Langevin equation with a friction term given by a convolution. We refer the interested reader to this paper, which includes also a discussion of the rotational Brownian motion, possibly coupled with the translation for particles of arbitrary shape.

4. Another physical constraint on Langevin's equation: the micro reversibility

The solution of the Langevin equation with a Gaussian delta correlated noise and constant friction coefficient is readily found and can be expressed in a number of ways. Because the noise is Gaussian and because the Langevin equation is linear, the probability distributions of the velocity fluctuations are Gaussian. This is true, in particular, of the two-time probability

$$< \delta(V_0 - V(0))\delta(V_1 - V(t) >= P(V_0, V_1; t)$$

Because $V(t)$ is Gaussian, this joint probability is also Gaussian. It is the exponential of a negative quadratic form $-(\alpha V_0^2 + \beta V_1^2 + 2\gamma V_0 V_1)$, where the coefficients are a priori functions of t. This quadratic form is constrained by the fact that $< V_0^2 >=< V_1^2 >= \frac{3k_BT}{M}$ and that the pair correlation is an even function of time. In the case of the original Langevin equation, it takes the form:

$$< \mathbf{V}(0)\cdot\mathbf{V}(t) >= \frac{3k_BT}{M}e^{-\zeta|t|/M}$$

When a convolution on time replaces the constant friction, it is a non-trivial result that the condition that the equality $< V^2(t) >=< V^2(0) >$ for any time t is still satisfied by a Gaussian white noise for the Langevin force. As shown in reference [16], the result expressed with the coefficients (α, β, γ) is that

$$\alpha = \beta = \frac{1}{1-A^2(t)}$$

and

$$\gamma = -\frac{A(t)}{1 - A^2(t)}$$

In the general case of friction given by an integral kernel, the function $A(t)$ can be found only numerically and it depends on the ratio $\frac{\rho_f}{\rho_s}$ and of the dimensionless time $|\frac{t\zeta}{M}|$. In the case of a constant friction (instead of a convolution), one recovers the standard result for the original Langevin equation, $A(t)$ being a simple exponential.

We emphasize that the pair correlation $< \mathbf{V}(0) \cdot \mathbf{V}(t) >$ is a function of the modulus of t because it is a pair correlation of the same fluctuations for a process that is stationary on average. Therefore, the conditions defining the coefficients (α, β, γ) are symmetrical with respect to permutations between V_0 and V_1. This symmetry is nothing but the symmetry under time reversal. It extends by the same type of argument to all situations where the fluctuations are Gaussian and steady on average, for instance whenever the viscous friction is linear, but not local in time. This symmetry under time reversal is a fundamental property of systems at thermal equilibrium. It does not hold in general outside of equilibrium, for instance in fluctuations of a turbulent flow [18], [19]. This symmetry breaking has deep consequences on the theoretical picture of non-equilibrium phenomena like turbulence.

The constraint of reversibility should not be confused with the property that, on average, the system is in a stationary state with fluctuations. If one considers the correlations of the same fluctuating scalar quantity, say $Q(t)$, at different times, the time correlation $< Q(t)Q(t') >$ is an even function of $(t - t')$, that is a function of $|(t - t')|$. This ls shown by translating the argument of $Q(t)$ and $Q(t')$ either by $(-t)$ or by $(-t')$, which does not change the result by the assumed invariance under time translation (or of steadiness of the statistical properties). This shows that $< Q(t)Q(t') >$ does not make any difference between the two possible directions of time. Of course, things become different in general if one considers time correlations between two different functions of time, like $< Q(t)R(t') >$. This is still a function of $(t - t')$, but may include parts that are odd functions of $(t - t')$, as observed in signals of turbulent velocities for instance. However, at equilibrium (and without magnetic field or spontaneous magnetization), correlations like $< Q(t)R(t') >$ are even functions of $(t - t')$ if $Q(.)$ and $R(.)$ have the same symmetry under time reversal.

5. The quantum Langevin equation

The Langevin equation was derived by Langevin for classical (= non quantum) systems. In order to include the randomness linked to quantum phenomena, several works have been devoted to a so-called Langevin–Heisenberg or Langevin–Schrödinger equation. Such equations are formulated in a number of different ways, but they all amount to take into account the randomness of the quantum jumps by adding a random Langevin-like "force" to the deterministic quantum equations (either the Schrödinger equation itself or the equation for the density matrix). Formally, this kind of extension reminds one of what has been done by Langevin, starting from Newton's equation of the heavy particle: one adds two effects, the viscous damping that is balanced on average by a force fluctuating randomly in time. Usually, the quantum jumps under consideration correspond physically to the emission or absorption of photons, which induces a fast transition from one eigenstate of the quantum system to another one. Seemingly, such abrupt change of state have a qualitative similarity with the collisions suffered by a classical Brownian particle from the fluid around it.

However, this approach suffers from a defect because any random term added (including as multiplicative) to evolution equations of quantum mechanics (either in their Schrödinger version or for the density matrix) has a fair chance to break the constraint of unitarity of the evolution, necessary for the conservation of the total probability in the statistical interpretation of quantum mechanics. This is not the case, however, of the "Langevin–Schrödinger" equation derived by van Kampen [24] in the case of a rapidly varying external *classical* random potential in the one-particle Schrödinger equation. In this case, one obtains a bona-fide Langevin-like equation including the randomness of the external classical field. However, this does not correspond at all to situations where one would like to have a Langevin–Schrödinger equation, namely a situation where the randomness of the external field is of purely quantum origin and represents, as just said, the random emission and absorption of photons.

As noticed, for instance by Plenio and Knight [26], the introduction of Langevin-like terms in theories to represent the emission or absorption of photons leads to equations that do not conserve the total probability.The conservation of the total probability is equivalent to the constraint that the trace of the density matrix remains exactly equal to one. According to those authors, the trace of the density matrix is conserved "in the mean" only, something that is physically unclear, to say the least: a prerequisite of any probability theory is that the total probability is *exactly* equal to one, not "on average" or "in the mean". Suppose for instance that the average total probability is one. It means that added fluctuations will make it sometimes larger than one. Even by stretching mathematics to their extreme, a probability larger than one does not make any sense.

The decay of excited atomic states with emission of photons has been fully described in an early paper by Dirac [23]. It made a highly non-trivial application of perturbation theory to the then nascent quantum mechanics. Somehow it describes the "inner" dynamics of the emission process, occurring on a very short time scale but rarely enough to be considered as instantaneous at the time scale of any other process. In particular, the duration of the transition is much shorter than the inverse frequency of those transitions. As shown in [25], this leads quite naturally to a Kolmogorov-like theory for the changes of the atomic states, which assumes fundamentally that the emission is a Markov process with very short duration without attempting to resolve this short time scale, namely by redoing, one way or another, the tricky calculation by Dirac giving explicitly the rate of transition. Somehow this is a bit like Boltzmann kinetic theory: this kinetic theory relies on the

solution of the two-body problem, but does not need a detailed solution (in particular the dynamics during the scattering process is irrelevant and it cannot be known analytically in the case of non-spherical potential, but the scattering cross section can be used to write Boltzmann's equation). This one enters in the scattering cross section computed elsewhere. In this sense, Langevin-inspired theories of quantum jumps cannot yield fair results because they attempt to solve both the "inner" dynamics of the emission of photons and the much slower one of secular changes of the quantum state of the atom.

The theory introduced in [25] is consistent with the requirement of unitarity based on a Kolmogorov-like equation for the fluctuations of the density matrix describing the atomic state. Moreover, this theory takes into account correctly the physics of the so-called quantum jumps: such jumps take place in a very short time, and, although unlikely on average, induce a finite change of the state of the system (practically an atom or an ion in an excited state: the quantum jump is linked to the small probability of emission of a photon). The problem considered in [28] and in [25] is not the one of a population of atoms, but of a single atom submitted to a resonant electromagnetic field, a situation corresponding to real experiments where the state of the atom is a linear superposition of the ground and excited states because of the optical Rabi oscillations. Without going into details that are outside of this presentation, it is clear that a single atom shows relatively far larger fluctuations of radiation than the population of many atoms considered by Einstein. Nevertheless, the idea of quantum jump remains pertinent for a single atom and leads quite naturally to extend, as done in [25], the statistical approach of Einstein [29] to this single atom situation. The statistics is done over the set of "Everett Universes", a new "Universe" appearing at every emission of photon by the atom decaying from an excited state to the ground one. This problem is made more complex than usual because of the optical Rabi oscillations between the excited and the ground state, oscillations due to the pumping at the resonant frequency. Details can be found in reference [25] and in a forthcoming paper by the same authors.

To conclude, this shows, in a particular case, that adding a Langevin noise to a formally deterministic theory is not the "universal" way of deriving statistical equations for dynamical processes. The foremost example of this is Boltzmann's kinetic theory, which has a built-in statistical interpretation without any added Langevin-like noise term.

6. Brownian motion of non-spherical particles

A rather obvious question is the one of the Brownian motion of non-spherical particles. Indeed, one of the first experiments by Perrin concerned the angular diffusion of non-spherical particles due to thermal fluctuations. The extension of Langevin's equation to this kind of situation is non-trivial. It induces rather complex questions because of the relationship between mobility and orientation of the Brownian particle. We refer to the paper by Cichocki et al. on the subject [27]. This paper explains how to compute the diffusion coefficient of Brownian non-spherical particles by explicit formulae based on the mobility tensor of the particle, a tensor relating the drag with the torque exerted on the particle for given (constant) linear and angular velocities. Being formulated by the diffusion equation, this approach satisfies the requirement for time-reversal symmetry.

7. Summary and conclusions

The legacy of Langevin's note is enormous. It introduced, in a single paper, extremely innovative mathematical techniques and was also able to explain a conspicuous physical phenomenon at "mesoscopic" scales, small enough to be sensitive to thermal fluctuation and big enough to be of measurable size. Indeed, much progress has been done since the original note, but by looking at the literature one can see that it is still a subject of inspiration and of new questions. Among them, we may single out the case of non-linear friction, like that felt by a tiny object sliding on the surface of a solid. The constraint of a theory for this Brownian motion consistent with the fundamental properties of reversibility of equilibrium and independence of the position and velocities in classical statistical mechanics makes this problem very challenging!

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