

## Introduction: 100 years of Brownian motion

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### I. INTRODUCTION

In the year 1905 Albert Einstein published four papers that raised him to a giant in the history of science. These works encompass the photo-electric effect (for which he obtained the Nobel prize in 1921), his first two papers on (special) relativity theory, and his first paper on Brownian motion, entitled “Über die von der molekularkinetischen Theorie der Wärme geforderte Bewegung von in ruhenden Flüssigkeiten suspendierten Teilchen” (submitted on 11 May 1905).<sup>1</sup> Thanks to Einstein’s intuition, the phenomenon observed by the Scottish botanist Robert Brown<sup>2</sup> in 1827—a little more than a naturalist’s curiosity—becomes the keystone of a fully probabilistic formulation of statistical mechanics and a well-established subject of physical investigation which we celebrate in this Focus Issue entitled—for this reason—“100 Years of Brownian Motion.”

Although written in a dated language, Einstein’s first paper on Brownian motion already contains the cornerstones of the modern theory of stochastic processes. The author starts out using arguments of thermodynamics and the concept of osmotic pressure of suspended particles to evaluate a particle diffusion constant by balancing a diffusion current with a drift current (through Stokes’ law). In doing so, he obtains a relation between two transport coefficients: the particle diffusion constant and the fluid viscosity, or friction. This relation, known as the Einstein relation,<sup>3</sup> was later generalized in terms of the famous fluctuation-dissipation theorem by Callen and Welton<sup>4</sup> and by the linear response theory of Kubo.<sup>5</sup> A much clearer discussion of Einstein’s arguments can be found in his thesis work, accepted by the University of Zurich in July 1905, which he submitted for publication<sup>6</sup> on 19 August 1905.

The second part of his 1905 paper contains a heuristic derivation of the (overdamped) diffusion equation, from which he deduces his famous prediction that the root mean square displacement of suspended particles is proportional to the square root of time. Moreover, the trajectories of a Brownian particle can be regarded as *memory-less* and *nondifferentiable*,<sup>7</sup> so that its motion is *not* ballistic (a bold statement that troubled mathematicians for half a century!). The latter also explained why earlier attempts to measure the

velocity of Brownian particles yielded puzzling results and indeed were doomed to fail.

A crucial consequence of Einstein’s theory is that from a measurement of the diffusion constant, i.e., by measuring the *distance* traveled rather than the velocity, it would be possible to extract an independent estimate of the important, and much debated Avogadro–Loschmidt number,  $N$ . Notably, the earliest determination of this number dates back to 1865 (!) when Johann Josef Loschmidt tried first to measure the size of molecules.<sup>8</sup> His data for mercury were compatible with a “best” value of  $4.4 \times 10^{23}$  molecules per mole. Inspired by Einstein’s work, an ingenious “reality check” on the role of fluctuations was performed through a series of experiments<sup>9</sup> by Perrin and his students in 1908–1911; Einstein’s predictions could be beautifully verified by setting the Avogadro–Loschmidt number in the range  $(6.4–6.9) \times 10^{23}/[\text{mol}]$ ; by 1914 the first three digits of the actual figure of  $6.022\,141\,5 \times 10^{23}/[\text{mol}]$  with a standard uncertainty of  $0.000\,001\,0 \times 10^{23}/[\text{mol}]$ , were finally accepted.<sup>10</sup>

The publication of Einstein’s papers provided further strong evidence for the atomistic hypothesis of matter. The immediate validation of his theory finally vindicated the arguments of the “discontinuists;” the remaining “continuists,” such as Wilhelm Ostwald, and in particular Ernst Mach [the latter being famous for his cynical remark to all “discontinuists:” “haben wir’s denn gesehen?” (die Atome/Moleküle), meaning “have we actually seen it?” (the atoms/molecules)] had thus no choice left but to concede.

We will not belabor any further the history of Brownian motion and the pioneering developments of its theory by Einstein’s contemporaries like Marian von Smoluchowski<sup>11</sup> (who worked on the molecular kinetic approach to Brownian motion since 1900, but did not publish until 1906), Paul Langevin,<sup>12</sup> and Norbert Wiener.<sup>13</sup> Beautiful accounts have been given in the literature by several authors. We mention here in particular the intriguing and most insightful introductory chapter by Mazo,<sup>14</sup> the short histories by Haw<sup>15</sup> and Powles,<sup>16</sup> or the notes presented by Nelson.<sup>17</sup>

### II. THE IMPACT OF BROWNIAN MOTION THEORY UP TO PRESENT

Without any doubt, the problem of Brownian motion has played a central role in the development of both the foundations of thermodynamics and the dynamical interpretation of statistical physics. A theory of Brownian motion based on the

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molecular-kinetic theory of heat, as that proposed by Einstein in 1905, does provide the link between an elementary underlying microscopic dynamics and macroscopic observable phenomena, such as the ubiquitous fluctuations of extended systems in natural and social sciences.

The early theories of Brownian motion inspired many prominent developments in various areas of physics, still subject of active research. In the following we briefly mention some of those addressed in the present Focus Issue.

Among the first to dwell on the ramifications of the fluctuation-dissipation relation were, as mentioned already, Callen and Welton.<sup>4</sup> These authors generalized the relations by Einstein, and subsequently by Nyquist and Johnson for the voltage fluctuations, to include quantum effects. In their fundamental work, they established a generally valid connection between the response function and the associated equilibrium quantum fluctuations, i.e., the quantum fluctuation-dissipation theorem.

Another key development must be credited to Lars Onsager: via his *regression hypothesis*, he linked the relaxation of an observable in the presence of weak external perturbations to the decay of correlations between associated microscopic variables.<sup>18</sup> This all culminated in the relations commonly known as the Green–Kubo relations.<sup>5,19,20</sup> This notion of “Linear Response” which in turn is related to the fluctuation properties of the corresponding variables (response-fluctuation theorems) can as well be extended to arbitrary (dynamical and nondynamical) systems that operate far from equilibrium.<sup>21</sup> The corresponding fluctuation-theorem relations (where the imaginary part of response function generally is no longer related to the mechanism of physical energy-dissipation<sup>21</sup>) provide most valuable information on the role of *nonequilibrium* fluctuations.

These “fluctuation theorems,” which describe the linear response to external perturbations in arbitrary statistical systems far away from thermal equilibrium, should not be confused with the recent beautiful *nonequilibrium work relations*, often also termed *fluctuation theorems*. This latter branch of fluctuation research was initiated by Evans *et al.*<sup>22</sup> and then formalized in the *chaotic hypothesis* of Galavotti and Cohen.<sup>22</sup> Independently, Jarzynski<sup>23</sup> proposed an interesting *equality*, being valid for both closed and open *classical* statistical systems: It relates—*a priori* surprisingly—the difference of two *equilibrium* free energies to the expectation of a particularly designed, stylized *nonequilibrium* work functional.

There is also an ongoing debate on the true origin of irregularity that causes the stochastic, random character of Brownian trajectories. In particular, is a chaotic microscopic dynamics sufficient, or is it more the role played by the extreme high dimensionality of the phase space that, on reduction, causes the jittery motion of the individual Brownian particles? The present Focus Issue contains an elucidative contribution by Vulpiani and collaborators,<sup>24</sup> who address precisely this and related issues. Answering this basic question becomes even more difficult when we attempt to include quantum mechanics. The description of Brownian motion for general quantum systems still presents true challenges, see the discussion herein by Hänggi and Ingold<sup>25</sup> and Ankerhold

*et al.*<sup>26</sup> For example, little is known for the modeling from first principles of quantum fluctuations in stationary nonequilibrium systems, or on the connection between the complexity obtained upon phase-space reduction and the microscopic quantum chaos.

The theory of Brownian motion also had a substantial impact on the theory of quantum mechanics itself. The formulation of quantum mechanics as a sum over paths<sup>27,28</sup> has its roots in the diffusive nature of the trajectories of a Brownian walker in continuous time: The Feynman–Kac propagator is nothing but a Schrödinger equation in imaginary time. In diffusion theory this idea had been utilized as early as in 1953 by Onsager and Machlup<sup>29</sup> for Gauss–Markov processes with linear coefficients. Its nontrivial extensions to the case with nonlinear drifts and nonlinear diffusion coefficients<sup>30</sup> and to colored noise driven nonlinear dynamics<sup>31</sup> have been mastered only 15–30 years ago.

The debate on Brownian motion also inspired mathematicians like Cauchy, Khintchine, Lévy, Mandelbrot, and many physicists and engineers to go beyond Einstein’s formulation. Nondifferentiable Brownian trajectories in modern language are called “fractal” and statistically self-similar on all scales. These extensions carry names such as fractal Brownian motion, Lévy noise, Lévy flights, Lévy walks, continuous time random walks, fractal diffusion, etc.<sup>32,33</sup> This topic is presently of wide interest and is being used to describe a variety of complex physical phenomena exhibiting, e.g., the anomalous diffusive behaviors reviewed here by Sokolov and Klafter,<sup>34</sup> or the diffusion limited growth and aggregation mechanisms discussed by Sander and Somfai.<sup>35</sup>

The quest for a mathematical description of the Brownian trajectories led to a new class of differential equations, namely the so-called stochastic differential equations. Such equations can be regarded as generalizations—pioneered by Paul Langevin—of Newtonian mechanical equations that are driven by independent, stochastic increments obeying either a Gaussian (white Gaussian noise) or a Poisson (white Poisson noise) statistics. This yields a formulation of the Fokker–Planck equations (master equations) in terms of a nonlinear Langevin equations generally driven by multiplicative, white Gaussian (Poisson) noise(s). As the aforementioned independent increments correspond to unbounded trajectory variations, the integration of such differential equations must be given a more general meaning: This led to the stochastic integration calculus of either the Ito type, the Stratonovich type, or generalizations thereof.<sup>21,36,37</sup> In recent years, this method of modeling the statistical mechanics of generally nonlinear systems driven by random forces has been developed further to account for physically more realistic noise sources possessing a finite or even infinite noise-correlation time (colored noise), i.e., noise sources that are non-Markovian.<sup>38</sup> In this Focus Issue Luczka<sup>39</sup> provides a timely overview of this recent progress together with the newest findings.

A powerful scheme to describe and characterize a statistical nonlinear dynamics from microscopic first principles is given by the methodology of non-Markovian, generalized Langevin equations or the associated generalized master equations. This strategy is by now well developed and un-

derstood only for *thermal equilibrium systems*. The projector operator approach,<sup>40,41</sup> which is used to eliminate the irrelevant (phase space) degrees of freedom, yields a clear-cut method to obtain the formal equations for either the rate of change of the probability or the reduced density operator, i.e., the generalized (quantum) master equation or the nonlinear generalized (quantum) Langevin equation.<sup>42</sup> The latter approach proved very useful to characterize the complex relaxation dynamics in glasses and related systems.<sup>43</sup>

There exists an abundance of processes in physics, chemistry (chemical kinetics), biology and engineering, where the dynamics involves activated barrier crossings and/or quantum tunneling-assisted processes through barriers. In all these processes the time scale for escape events is governed by fluctuations that typically are of Brownian motion origin. The first attempts to characterize escape dynamics date back to the early 1930s with contributions by Farkas, Wigner, Eyring, Kramers, to name a few prominent ones. This topic was extended in the late 1970s early 1980s to account also for (non-Markovian) memory effects, solvent effects, quantum tunneling, nonequilibrium fluctuations, correlated noises (i.e., colored noises),<sup>38</sup> nonlinear bath degrees of freedom and time-dependent forcing. The interested reader is directed to a comprehensive review<sup>44</sup> and is further referred to the up-to-date accounts given by Pollak and Talkner<sup>45</sup> and Hänggi and Ingold<sup>25</sup> in this issue.

The combined action of external driving and noise has given rise to new phenomena, where the constructive role of Brownian motion provides a rich scenario of far-from equilibrium effects. The most popular such novel feature is the phenomenon of *stochastic resonance*.<sup>46</sup> It refers to the fact that an optimal level of applied or intrinsic noise can dramatically boost the response (or, more generally the transport) to typically weak, time-dependent input signals in nonlinear stochastic systems. This theme naturally plays a crucial role in biology with its variety of threshold-like systems that are subjected to noise influences.<sup>47</sup>

A more recent but increasingly popular example of the constructive role of fluctuations (intrinsic and external, alike) is the noise-assisted transport in periodic systems, namely the so-called *Brownian motors*.<sup>48</sup>

Researchers are still very much active in these two topics: This Focus Issue contains both an experimental and a theoretical contribution to stochastic resonance by Bechinger *et al.*<sup>49</sup> and Casado-Pascual *et al.*<sup>50</sup> The theme of noise-assisted transport is multifaceted and very rich; this is corroborated with several appealing contributions by Linke *et al.*,<sup>51</sup> Borromeo and Marchesoni,<sup>52</sup> Savel'ev and Nori,<sup>53</sup> and Eichhorn *et al.*<sup>54</sup>

### III. RESUME

This Focus Issue on “100 Years of Brownian Motion” is not only timely but also confirms that research in this area is very much alive and still harbors plenty of surprises that only wait to be unravelled by future researchers. The original ideas that Einstein put forward in 1905 are very modern and still find their way to applications in such diverse areas as soft matter physics,<sup>55</sup> including the granular systems investigated here by Brilliantov and Pöschel<sup>56</sup> and the soliton dif-

fusion in linear defects,<sup>52,57</sup> solid state physics, chemical physics, computational physics, and beyond. In recent years, ideas and tools developed within the context of the Brownian motion theory are gaining increasing impact in life sciences (the contribution by Zaks *et al.*<sup>58</sup> provides a timely example) and even in human studies, where econophysics is becoming a lively crossroad of interdisciplinary research, as shown with the study by Bouchaud<sup>59</sup> in this issue.

We Guest Editors share the confident belief that the contributions in this Focus Issue by leading practitioners from a diverse range of backgrounds will together provide a fair and accurate snapshot of the current state of this rich and interdisciplinary research field. Last but not least, we hope that this collection of articles will stimulate readers into pursuing future research of their own.

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