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Brownian motion and diffusion: From stochastic processes to chaos and beyond

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One century after Einstein's work, Brownian motion still remains both a fundamental open issue and a continuous source of inspiration for many areas of natural sciences. We first present a discussion about stochastic and deterministic approaches proposed in the literature to model the Brownian motion and more general diffusive behaviors. Then, we focus on the problems concerning the determination of the microscopic nature of diffusion by means of data analysis. Finally, we discuss the general conditions required for the onset of large scale diffusive motion. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1832773]

Brownian motion (BM) played a fundamental role in the development of molecular theory of matter, statistical mechanics and stochastic processes. Remarkably, one century after Einstein's work, BM is still at the origin of scientific discussions as testified by a recent experiment performed to detect a trace of deterministic chaotic sources on macroscopic diffusion. Several authors, which discussed the results of such an experiment, argued that the possibility to discern experimentally between a deterministic chaotic and noisy dynamics, at the microscopic level, is severely limited by subtle technical and conceptual points. However, the remarks raised by the scientific community have gone over the criticism and have led to a deeper understanding of the role of chaos in the diffusion. After a short historical introduction to BM, we focus on the dynamical conditions to observe macroscopic diffusion. In particular, we discuss the technical and conceptual limits in distinguishing, by means of data analysis, the deterministic or stochastic nature of diffusion. A main tool for that is the ϵ-entropy. Part of the discussion is devoted to the problem of macroscopic diffusion in deterministic nonchaotic dynamics.

I. INTRODUCTION

At the beginning of the twentieth century, the atomistic theory of matter was not yet fully accepted by the scientific community. While searching for phenomena that would prove, beyond any doubt, the existence of atoms, Einstein realized that "… *according to the molecular-kinetic theory of heat, bodies of microscopically-visible size suspended in a liquid will perform movements of such magnitude that they can be easily observed in a microscope* …," as he wrote in his celebrated paper in 1905.[1] In this work, devoted to explain the irregular motion of Brownian particles on theoretical grounds, Einstein argued that the motion of these small bodies has a diffusive character. Moreover, he discovered an important relation involving the diffusion coefficient D, the fluid viscosity η, the particles radius a (having assumed spherical particles), Avogadro's number N_A, the temperature T and the gas constant R:

$$D = \frac{1}{N_A}\frac{RT}{6\pi\eta a}. \tag{1}$$

This relation can be employed, and actually had been, to determine experimentally the Avogadro's number.[2] Indeed, the diffusion coefficient can be measured by monitoring the growth, with time t, of the particle displacement $\Delta x = x(t) - x(0)$, which is expected to behave as

$$\langle(\Delta x)^2\rangle \simeq 2Dt. \tag{2}$$

Einstein relation (1), that may be seen as the first example of the fluctuation–dissipation theorem,[3] allowed for the determination of Avogadro's number and gave one of the ultimate evidences of the existence of atoms.

Einstein's theoretical explanation of BM is based on the intuition that the irregular motion of a Brownian particle is a consequence of the huge number of collisions per unit time with the surrounding fluid molecules. Since Einstein's approach, diffusion and irregular phenomena were commonly associated to the presence of many degrees of freedom. The effects of the disregarded degrees of freedom on an observed small part of a system can be either studied directly (as initiated by Smoluchowski[4]) or modeled by means of stochastic dynamics (as proposed by Langevin[5]). From the latter point of view, BM provided the first and main stimulus to the building of the modern theory of stochastic processes.

After the (re)discovery of deterministic chaos,[6,7] it was clear that also fully deterministic and low dimensional systems can give rise to erratic seemingly random motions, practically indistinguishable from those produced by a stochastic process. This implied an affective unpredictability of chaotic systems and the need for a probabilistic description also of a strictly deterministic world. The success in understanding the basic mechanisms for the onset of chaos, and the wealth of interesting phenomena occurring in low dimensional systems hinted at optimistic expectations about the possibility of a systematic deterministic approach to irregular

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natural phenomena. This raised a rapid development of time series analysis with the idea to demonstrate the deterministic character of many irregular phenomena.

Nowadays we are aware of the limits of this optimistic program,[8,9] and we know that a definite answer on the deterministic or stochastic character of experimental signals is impossible. However, some tools of time series analysis, such as the entropy analysis at varying the scale of resolution, are very useful to characterize important features of complex systems. Among the recent developments in this context, we can mention the experiment by Gaspard *et al.*[10] on the motion of a Brownian particle. The debate[11,12] around the possible theoretical interpretation of the experiment is a clear indication of how, one century after the seminal Einstein's work, BM continues to be a subject of intricate and fascinating discussions.

Beyond its undoubted importance for applications in many natural phenomena, deterministic chaos also enforces us to reconsider some basic problems standing at the foundations of statistical mechanics such as, for instance, the applicability of a statistical description to low dimensional systems.

In addition, the combined effects of noise and deterministic evolution can generate highly nontrivial and rather intriguing behaviors. As an example, we just mention the stochastic resonance[13,14] and the role of colored noise in dynamical systems.[15]

The aim of this paper is a discussion on the viable approaches to characterize and understand the dynamical (microscopic) character of BM (Sec. II). In particular, we shall focus on the distinction between chaos and noise from a data analysis and on conceptual aspects of the modeling problem (Sec. III). Moreover, we shall investigate and discuss about the basic microscopic ingredients necessary for BM as, for instance, the possibility of genuine BM in nonchaotic deterministic systems (Sec. IV). Finally, we conclude (Sec. V) with a discussion on the role of chaos in statistical mechanics.

II. THE ORIGIN OF DIFFUSION

Einstein's work on BM is based on statistical mechanics and thermodynamical considerations applied to suspended particles, with the assumption of velocity decorrelation (molecular chaos).

Instead, one of the first attempts to develop a purely dynamical theory of BM dates back to Langevin[5] that, as he writes, gave "… *a demonstration* [of Einstein results] *that is infinitely more simple by means of a method that is entirely different.*" Langevin considers the Newton equation for a small spherical particle in a fluid, taking into account that the Stokes viscous force it experiences is only a mean force. In one direction, say, e.g., the x-direction, one has

$$m\,\frac{d^2x}{dt^2} = -\,6\pi\eta a\,\frac{dx}{dt} + F, \qquad (3)$$

where m is the mass of the particle. In the right-hand side (r.h.s.) the first term is the Stokes viscous force. F is a fluctuating random force which models the effects of the huge number of impacts with the surrounding fluid molecules, responsible for the thermal agitation of the particle. In statistical mechanics terms, this corresponds to molecular chaos.

With the assumption that the force F is a Gaussian, time uncorrelated random variable, the probability distribution functions (pdf) for the position and velocity of the Brownian particle can be exactly derived.[16] In particular, the pdf of the position, at long times, reduces to the Gaussian distribution in agreement with Einstein's result.

Langevin's work along with that of Ornstein and Uhlenbeck[16] are at the foundation of the theory of stochastic differential equations. The stochastic approach is, however, unsatisfactory being at the level of a phenomenological description.

The next theoretical challenge toward the building of a dynamical theory of Brownian motion is to understand its microscopic origin from first principles. A very early attempt was made in 1906 by Smoluchowski, who tried to derive the large scale diffusion of Brownian particles starting from the microscopic description of their collisions with the fluid molecules.[4] A renewed interest on the subject appeared some years later, when it was realized that even purely deterministic systems composed of a large number of particles produce macroscopic diffusion, at least on finite time scales. These models had an important impact in the justification of Brownian motion theory and, more in general, in deriving a consistent microscopic theory of irreversibility.

Some of these works considered chains of harmonic oscillators of equal masses,[17–20] while others[21–23] analyzed the motion of a heavy impurity linearly coupled to a chain of equal mass oscillators. For an infinite number of oscillators, the momentum of the heavy particle behaves as a genuine stochastic process described by the Langevin equation (3). When their number is finite, diffusion remains an effective phenomenon lasting for a (long but) limited time.

Soon after the discovery of dynamical chaos, it was realized that also simple low dimensional deterministic systems may exhibit a diffusive behavior. In this framework, the two-dimensional Lorentz gas,[24] describing the motion of a free particle through a lattice of hard round obstacles, provided the most valuable example. Particle trajectories can be ballistic (with very few collisions in the case of infinite horizon) or chaotic as a consequence of the convexity of the obstacles. In the latter case, at large times, the mean square displacement from the particle initial condition grows linearly with time. Lorentz system is closely related to the Sinai billiard,[25,26] which can be obtained from the Lorentz gas by folding the trajectories into the unitary lattice cell. The extensive study on billiards has shown that chaotic behavior might usually be associated to diffusion in simple low dimensional models, supporting the idea that chaos was at the very origin of diffusion. However, more recently (see, e.g., Ref. 27) it has been shown that even nonchaotic deterministic systems, such as a bouncing particle in a two-dimensional billiard with polygonal but randomly distributed obstacles (wind-tree Ehrenfest model), may exhibit a diffusionlike behavior (see Sec. IV).

Deterministic diffusion is a generic phenomenon present also in simple chaotic maps on the line. Among the many contributions we mention the work by Fujisaka, Grossmann,[28,29] and Geisel.[30,31] A typical example is the 1d discrete-time dynamical system:

$$x(t+1) = [x(t)] + F(x(t) - [x(t)]), \tag{4}$$

where $x(t)$ (the position of a pointlike particle) performs diffusion in the real axis. The bracket $[\dots]$ denotes the integer part of the argument. $F(u)$ is a map defined on the interval $[0, 1]$ that fulfills the following properties:

(i) The map, $u(t+1)=F(u(t))$ (mod 1) is chaotic;

(ii) $F(u)$ must be larger than 1 and smaller than 0 for some values of u, so there exists a nonvanishing probability to escape from each unit cell (a unit cell of real axis is every interval $C_\ell \equiv [\ell, \ell+1]$, with $\ell \in \mathbf{Z}$);

(iii) $F_r(u)=1-F_l(1-u)$, where F_l and F_r define the map in $u \in [0,1/2[$and $u \in [1/2,1]$, respectively. This antisymmetry condition with respect to $u=1/2$ is introduced to avoid a net drift.

A very simple and much studied example of F is

$$F(u) = \begin{cases} 2(1+a)u & \text{if } u \in [0,1/2[, \\ 2(1+a)(u-1)+1 & \text{if } u \in [1/2,1], \end{cases} \tag{5}$$

where $a > 0$ is the control parameter. It is useful to remind the link between diffusion and velocity correlation, i.e., the Taylor–Kubo formula, that helps in understanding how diffusion can be realized in different ways. Defining $C(\tau) = \langle v(\tau)v(0)\rangle$ as the velocity correlation function, where $v(t)$ is the velocity of the particle at time t. It is easy to see that for continuous time systems [e.g., Eq. (3)]

$$\langle (x(t)-x(0))^2\rangle \simeq 2t\int_0^t d\tau C(\tau). \tag{6}$$

Standard diffusion, with $D=\int_0^\infty d\tau C(\tau)$, is always obtained whenever the hypotheses for the validity of the central limit theorem are verified.

(I) The variance of the velocity must be finite: $\langle v^2\rangle < \infty$.

(II) The decay to zero of the velocity correlation function $C(\tau)$ at large times should be faster than τ^{-1}.

In discrete-time systems, the velocity $v(t)$ and the integration of $C(t)$ are replaced by the finite difference $x(t+1) - x(t)$ and by the quantity $\langle v(0)^2\rangle/2+\Sigma_\tau C(\tau)$, respectively.

Condition (I) is justified by the fact that having an infinite variance for the velocity is rather unphysical. It should be noted that this requirement is independent of the microscopic dynamics under consideration: Langevin, deterministic chaotic, or regular dynamics.

Condition (II), corresponding to the request of molecular chaos, is surely verified for the Langevin dynamics where the presence of the stochastic force entails a rapid decay of $C(\tau)$. In deterministic regular systems, such as the many oscillator model, the velocity decorrelation comes from the huge number of degrees of freedom that act as a heat bath on a single oscillator. While in the (nonchaotic) Ehrenfest windtree model decorrelation originates from the disorder in the obstacle positions, the situation is more subtle for deterministic chaotic systems. In fact, even if nonlinear instabilities generically lead to a memory loss and, henceforth, to the validity of the molecular-chaos hypothesis, slow decay of correlation, e.g., $C(\tau) \sim \tau^{-\beta}$ with $\beta < 1$, may appear in very intermittent systems.[32] When this happens, condition (II) is violated, and superdiffusion, $\langle x^2(t)\rangle \sim t^{2-\beta}$, is observed. Though interesting, superdiffusion is a quite rare phenomenon. Moreover, usually, small changes of the control parameters of the dynamics restore standard diffusion. Therefore, also for chaotic systems we can state that the "rule" is the standard diffusion and the "exception" is the superdiffusion.[33]

We end this section by asking whether is it possible to determine, by the analysis of a Brownian particle, if the microscopic dynamics underlying the observed macroscopic diffusion is stochastic, deterministic chaotic, or regular?

III. DISTINCTION BETWEEN CHAOS AND NOISE

Inferring the microscopic deterministic character of Brownian motion on an experimental basis would be attractive from a fundamental viewpoint. Moreover it could provide further evidence to some recent theoretical and numerical studies.[34,35] Before discussing a recent experiment[10] in this direction, we must open the "Pandora box" of the longstanding and controversial problem of distinguishing chaos from noise in signal analysis.[36]

The first observation is that, very often in the analysis of experimental time series, there is not a unique model of the "system" that produced the data. Moreover, even the knowledge of the "true" model might not be an adequate answer about the character of the signal. From this point of view, BM is a paradigmatic example: In fact it can be modeled by a stochastic as well as by a deterministic chaotic or regular process.

In principle a definite answer exists. If we were able to determine the maximum Lyapunov exponent (λ) or the Kolmogorov–Sinai (KS) entropy (h_{KS}) of a data sequence, we would know without uncertainty whether the sequence was generated by a deterministic law ($\lambda, h_{KS} < \infty$) or by a stochastic one ($\lambda, h_{KS} \to \infty$). Nevertheless, there are unavoidable practical limitations in computing such quantities. Those are indeed defined as infinite time averages taken in the limit of arbitrary fine resolution. But, in experiments, we have access only to a finite, and often very limited, range of scales and times.

However, there are measurable quantities that are appropriate for extracting information on the signal character. In particular, we shall consider the (ϵ, τ)-entropy per unit time[37–39] $h(\epsilon, \tau)$ that generalizes the Kolmogorov–Sinai entropy. In a nutshell, while for evaluating h_{KS} one has to detect the properties of a system with infinite resolution, for $h(\epsilon, \tau)$ a finite scale (resolution) ϵ is requested. The Kolmogorov–Sinai entropy is recovered in the limit $\epsilon \to 0$, i.e., $h(\epsilon, \tau) \to h_{KS}$. This means that if we had access to arbitrarily small scales, we could answer the original question about the character of the law that generated the recorded signal. Even if this limit is unattainable, still the behavior of $h(\epsilon, \tau)$ provides a very useful scale-dependent description of the nature of a signal.

A. ϵ-entropy

The ϵ-entropy was originally introduced in the context of information theory by Shannon[38] and, later, by Kolmogorov[37] in the theory of stochastic processes. An operative definition is as follows.

One considers a continuous variable $\mathbf{x}(t) \in \Re^d$, that represents the state of a d-dimensional system, and one introduces the vector

$$\mathbf{X}^{(m)}(t) = (\mathbf{x}(t), \ldots, \mathbf{x}(t + m\tau - \tau)), \tag{7}$$

which lives in $\Re^{md}$ and is a portion of the trajectory discretized in time with step τ. Then the phase space $\Re^d$ is partitioned using hyper-cubic cells of side ϵ. The vector $\mathbf{X}^{(m)}(t)$ is coded into the word, of length m,

$$W^m(\epsilon, t) = (i(\epsilon, t), \ldots, i(\epsilon, t + m\tau - \tau)), \tag{8}$$

where $i(\epsilon, t+j\tau)$ labels the cell in $\Re^d$ containing $\mathbf{x}(t+j\tau)$. For bounded motions, the number of visited cells (i.e., the alphabet) is finite. Under the hypothesis of stationarity, the probabilities $P(W^m(\epsilon))$ of the admissible words $\{W^m(\epsilon)\}$ are obtained from the time evolution of $\mathbf{X}^{(m)}(t)$. The (ϵ, τ)-entropy per unit time, $h(\epsilon, \tau)$ is then defined by[38]

$$h(\epsilon, \tau) = \lim_{m\to\infty} h_m(\epsilon, \tau) = \frac{1}{\tau} \lim_{m\to\infty} \frac{1}{m} H_m(\epsilon, \tau), \tag{9}$$

where H_m is the m-block entropy:

$$H_m(\epsilon, \tau) = - \sum_{\{W^m(\epsilon)\}} P(W^m(\epsilon)) \ln P(W^m(\epsilon)), \tag{10}$$

and $h_m(\epsilon, \tau) = [H_{m+1}(\epsilon, \tau) - H_m(\epsilon, \tau)]/\tau$.

It is worth remarking on a few important points. A rigorous mathematical procedure[39] would require to take the infimum over all possible partitions with elements of size smaller than ϵ. The Kolmogorov–Sinai entropy is obtained in the limit of small ϵ

$$h_{KS} = \lim_{\epsilon\to 0} h(\epsilon, \tau). \tag{11}$$

Note that in deterministic systems $h(\epsilon)$ and henceforth h_{KS} do not depend on the sampling time so that (11) can be in principle used with any choice of τ.[40] However, in practical computations, where the rigorous definition is not applicable, the specific value of τ is important and $h(\epsilon)$ may also depend on the used norm. For very small ϵ, no matter of the norm, the correct value of the Kolmogorov–Sinai entropy is usually recovered. Indeed, when the partition is very fine, usually, it well approximates a generating partition. It is worth reminding that the Kolmogorov–Sinai entropy is a dynamical invariant, i.e., independent of the used state representation (7).

In deterministic systems the following chain of inequalities holds:[40]

$$h(\epsilon, \tau) \leqslant h_{KS} \leqslant \sum_i^+ \lambda_i, \tag{12}$$

where the summation is over all positive Lyapunov exponents. A system is chaotic when $0 < h_{KS} < \infty$ and regular when $h_{KS} = 0$. Typically, one observes that $h(\epsilon, \tau)$ attains a plateau h_{KS}, below a resolution threshold, ϵ_c, associated to the smallest characteristic length scale of the system. Instead, for $\epsilon > \epsilon_c$ due to (12) $h(\epsilon, \tau) < h_{KS}$, in this range the details of the ϵ-dependence may be informative on the large scale (slow) dynamics of the system (see, e.g., Refs. 36 and 41).

In stochastic signals $h_{KS} = \infty$, but for any $\epsilon > 0$, $h(\epsilon, \tau)$ is finite and a well defined function of ϵ and τ. The nature of the dependence of $h(\epsilon, \tau)$ on ϵ and τ provide a characterization of the underlying stochastic process (see Refs. 37, 39, and 41). For an important and wide class of stochastic processes[39] an explicit expression for $h(\epsilon, \tau)$ can be given in the limit $\tau \to 0$. This is the case of stationary Gaussian processes characterized by a power spectrum $S(\omega) \propto \omega^{-(2\alpha+1)}$, with $0 < \alpha < 1$, for which[37]

$$\lim_{\tau\to 0} h(\epsilon, \tau) \sim \epsilon^{-1/\alpha}. \tag{13}$$

The case $\alpha = 1/2$, corresponding to the power spectrum of a Brownian signal, would give $h(\epsilon) \sim \epsilon^{-2}$. Some stochastic processes, such as, e.g., time uncorrelated and bounded ones, are characterized by a logarithmic divergence below a critical ϵ_c, which may depend on τ.

B. Numerical determination of the ϵ-entropy

In experiments, usually, only a scalar variable $u(t)$ can be measured and moreover the dimensionality of the phase space is not known. In these cases it is reconstructed by delay embedding technique,[8,9] where the vector $\mathbf{X}^{(m)}(t)$ is built as $(u(t), u(t+\tau), \ldots, u(t+m\tau-\tau))$, now in $\Re^m$. This is a special instance of (7).

Then to determine the entropies $H_m(\epsilon)$, very efficient numerical methods are available[42,43] (the reader may find an exhaustive review in Refs. 8 and 9). Here, avoiding technicalities, we just mention some subtle points which should be taken into account in data analysis.

First, if the information dimension of the attractor for a given system is d_1 then, to have a meaningful measure of the entropy, the embedding dimension m has to be larger than d_1. Second, as mentioned above, the plateau $h_m(\epsilon) \approx h_{KS}$ appears only below a critical ϵ_c, meaning that it is possible to distinguish a deterministic signal from a random one only for $\epsilon < \epsilon_c$. However, one should be aware of the fact that the finiteness of the data set imposes a lower cut-off scale ϵ_d below which no information can be extracted from the data (see Ref. 44). Also in stochastic signals, there exists a lower critical cut-off ϵ_d due to the finiteness of the data set, and often, as mentioned at the end of the previous subsection, one has logarithmic divergences below ϵ_c. Since, this is also what happens in general for $\epsilon < \epsilon_d$, the interpretation of the results requires much attention.[8,36,44]

Therefore, if m is not large enough and/or ϵ is not small enough (or in the lack of a good estimation of the important range of scales for the different behaviors) one may obtain misleading results.

Another important problem concerns the choice of τ. If τ is much larger or much shorter than the characteristic timescale of the system at the scale ϵ, then the correct behavior of the ϵ-entropy[36] cannot be properly recovered.

To exemplify the above difficulties let us consider the map

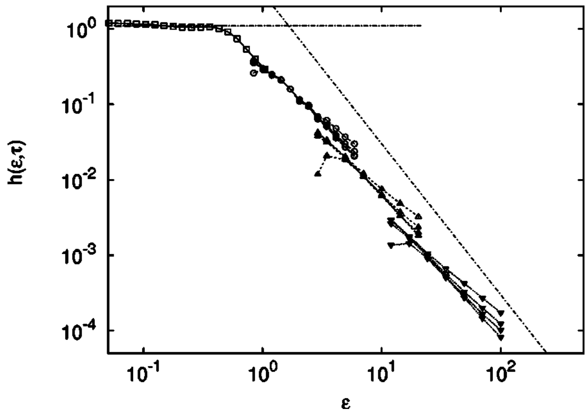

FIG. 1. Numerically evaluated (ϵ,τ)-entropy for the map (14) with $p=0.8$ computed by the standard techniques (Ref. 6) at $\tau=1$ (○), $\tau=10$ (△), and $\tau=100$ (▽) and different block length ($m=4$, 8, 12, 20). The boxes give the entropy computed with $\tau=1$ by using periodic boundary condition over 40 cells. The straight lines correspond to the two asymptotic behaviors, $h(\epsilon)=h_{\rm KS}\simeq 1.15$ and $h(\epsilon)\sim\epsilon^{-2}$.

$$x(t+1)=x(t)+p\sin(2\pi x(t)), \tag{14}$$

which, for $p>0.7326\ldots$, is chaotic [similarly to (4)] and displays large scale diffusion. On the basis of the previous discussion, the ϵ-entropy is expected to behave as

$$h(\epsilon)\simeq\begin{cases}\lambda \text{ for } \epsilon\ll 1,\\ D/\epsilon^2 \text{ for } \epsilon\gg 1,\end{cases} \tag{15}$$

where λ is the Lyapunov exponent and D is the diffusion coefficient. The typical problems encountered in numerically computing $h(\epsilon)$ can be appreciated in Fig. 1. First notice that the threshold $\epsilon_c\approx 1$. As for the importance of the choice of τ, note that the diffusive behavior $h(\epsilon)\sim\epsilon^{-2}$ is roughly obtained only by considering the envelope of $h_m(\epsilon,\tau)$ evaluated at different values of τ. The reason for this is as follows. The natural sampling interval would be $\tau=1$, however, this choice requires considering larger and larger embedding dimensions m at increasing ϵ. Indeed, a simple dimensional argument suggests that the characteristic time of the system is determined by its diffusive behavior $T_\epsilon\approx\epsilon^2/D$. If we consider, for example, $\epsilon=10$ and the typical values of the diffusion coefficient $D\simeq 10^{-1}$, the characteristic time, T_ϵ, is much larger than the elementary sampling time $\tau=1$. On the other hand, the plateau at the value $h_{\rm KS}$ can be recovered only for $\tau\approx 1$, even if, in principle, any value of τ could be used.

The above difficulties can be partially overcome by means of a recently introduced method based on exit times.[45] The main advantage of this approach is that it is not needed to fix *a priori* τ, because the "correct" τ is automatically selected.

C. Does Brownian motion arise from chaos, noise, or regular dynamics?

We are now ready to discuss the experiment and its results reported in Ref. 10. In this experiment, a long time record (about 1.5×10^5 data points) of the motion of a small colloidal particle in water was sampled at regular time intervals ($\Delta t=1/60$ s) with a remarkable high spatial resolution (25 nm). To our knowledge, this is the most accurate measurement of a BM. The data were then processed by means of standard nonlinear time-series analysis tools, i.e., the Cohen–Procaccia method,[42] to compute the ϵ-entropy. This computation shows a power-law dependence $h(\epsilon)\sim\epsilon^{-2}$. Actually, similarly to what displayed in Fig. 1, this behavior is recovered only by considering the envelope of the $h(\epsilon,\tau)$-curves, for different τ's. However, unlike Fig. 1, no saturation $h(\epsilon,\tau)\approx$ const is observed in the small ϵ-region because of the finiteness of data set and resolution as well. From the previous discussion, this can be understood as the fact that ϵ_c of the observed system is much smaller than the smallest detectable scale extracted from data and, therefore, the KS entropy cannot be properly estimated. Nevertheless, from the chain of inequalities (12) and by *assuming* from the outset that the system dynamics is deterministic, the authors deduce, from the positivity of $h(\epsilon)$, the existence of positive Lyapunov exponents in the system. Their conclusion is thus that microscopic chaos is at the origin of the macroscopic diffusive behavior.

As pointed out by several works, a few points need to be considered in the data analysis of the aforementioned experiment, namely: The huge amount of involved degrees of freedom (Brownian particle and the fluid molecules); the impossibility to reach high enough (spatial and temporal) resolution; the limited amount of data points.

The limitation induced by the finite resolution is particularly relevant to the experiment. Even if one assumes that the number of data points and the embedding dimension are large enough, the impossibility to see a saturation to a constant value, $h(\epsilon)\approx$ const, prevents any conclusion about the character of the analyzed signal. For example, whenever the the analysis of Fig. 1 would be restricted to the region with $\epsilon>1$, then discerning whether the data were originated by a chaotic system or by a stochastic process would be impossible. In fact in both cases the behavior $h(\epsilon)\sim\epsilon^{-2}$ would have been observed.

As for the number of degrees of freedom, we recall that, for a correct evaluation of the entropy of the microscopic dynamics, a very high embedding dimension should be used, in practice $m>d_1$. For a fluid [with $O(10^{23})$ molecules] the necessary number of points is of course prohibitive. Moreover, as stressed by Grassberger and Schreiber,[12] when the number of degrees of freedom is so high that it can be considered practically infinite there is an additional difficulty related to the definition of entropy and Lyapunov exponents, which become norm dependent.

Furthermore, the limited amount of data severely affects even our ability to recognize if the signal is deterministic but of zero entropy (i.e., regular). This has been pointed out by Dettmann *et al.*,[11,27] who have shown that the same entropic analysis of Ref. 10, applied to the Ehrenfest wind-tree model (see next section) reproduced results very similar to those extracted in the Brownian experiment. This model is deterministic and nonchaotic. In fact if the time records were long enough to see the periodic nature of the signal, and the embedding dimension high enough to resolve the system manifold, the measured entropy would have been zero.

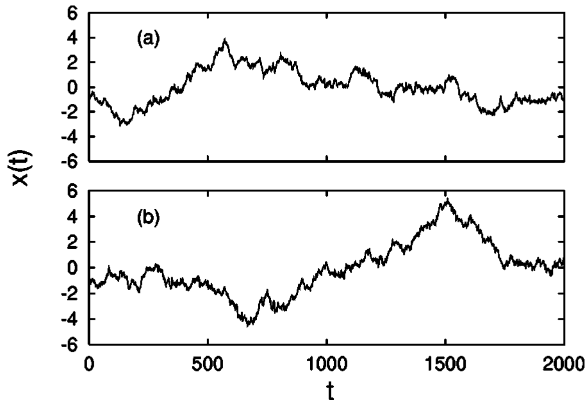

FIG. 2. (a) Signals obtained from Eq. (17) with $M=10^4$ and random phases uniformly distributed in $[0,2\pi]$. The numerically computed diffusion constant is $D\approx 0.007$. (b) Time record obtained with a continuous random walk (16) with the same value of the diffusion constant as in (a). In both cases data are sampled with $\tau=0.02$, i.e., 10^5 data points.

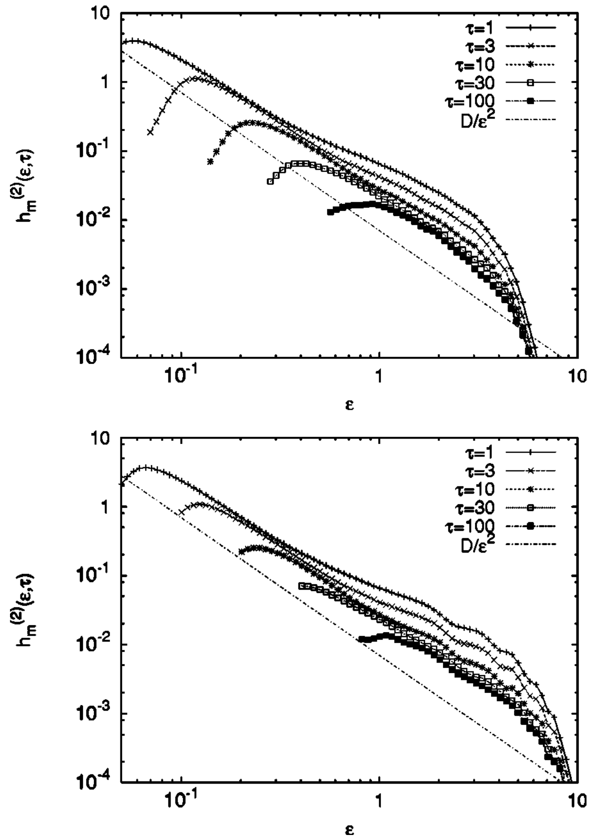

FIG. 3. $h(\epsilon)$ computed with the Grassberger–Procaccia algorithm using 10^5 points from the time series of Fig. 2. We show the results for embedding dimension $m=50$. The straight lines show the D/ϵ^2 behavior.

The following example serves as a clue to better understand these points. Let us consider two signals, the first generated by a continuous random walk:

$$\dot{x}(t) = \sqrt{2D}\,\eta(t), \tag{16}$$

where η is a zero mean Gaussian variable with $\langle \eta(t)\eta(t')\rangle = \delta(t-t')$, and the second obtained as a superpositions of Fourier modes:

$$x(t) = \sum_{i=1}^{M} X_{0i}\sin(\Omega_i t + \phi_i). \tag{17}$$

The coordinate $x(t)$ in Eq. (17), upon properly choosing the frequencies[23,36] and the amplitudes (e.g., $X_{0i}\propto\Omega_i^{-1}$), describes the motion of a heavy impurity in a chain of M linearly coupled harmonic oscillators. We know[23] that $x(t)$ performs a genuine BM in the limit $M\to\infty$. For $M<\infty$ the motion is periodic and regular, nevertheless for large but finite times it is impossible to distinguish signals obtained by (16) and (17) (see Fig. 2). This is even more striking looking at the computed ϵ-entropy of both signals (see Fig. 3).

The results of Fig. 3 along with those by Dettman *et al.*[11] suggest that, also by assuming the deterministic character of the system, we are in the practical impossibility of discerning chaotic from regular motion.

From the above discussion, one may have reached a very pessimistic view on the possibility to detect the "true" nature of a signal by means of data analysis only. However, the scenario is different when the question about the character of a signal remains restricted only to a certain interval of scales. In this case, in fact, it is possible to give an unambiguous classification of the signal character based solely on the entropy analysis and free from any prior knowledge on the system–model that generated the data. Indeed, we can define stochastic–deterministic behavior of a time series on the basis of the absence–presence of a saturation plateau $h(\epsilon)\approx$ const in the observed range of scales. Moreover the behavior of $h(\epsilon,\tau)$ as a function of (ϵ,τ) provides a very useful "dynamical" classification of stochastic processes.[39,45] One has then a practical tool to classify the character of a signal as deterministic or stochastic without referring to a specific model, and is no longer obliged to answer the metaphysical question, whether the system that produced the data was a deterministic or a stochastic[36,46] one.

This is not a mere way to escape the original question. Indeed, it is now clear that the maximum Lyapunov exponent and the Kolmogorov–Sinai entropy are not completely satisfactory for a proper characterization of the many faces of complexity and predictability of nontrivial systems, such as intermittent systems or with many degrees of freedom (e.g., turbulence).[41]

In the literature the reader may find several methods developed to distinguish chaos from noise. They are based on the difference in the predictability using prediction algorithms rather than estimating the entropy[47,48] or they relate determinism to the smoothness of the signal.[49,50] All these methods have in common the necessity to choose a certain length scale ϵ and a particular embedding dimension m, therefore they suffer the same limitations of the entropy analysis presented here.

IV. DIFFUSION IN NONCHAOTIC SYSTEMS

With all the provisos concerning its interpretation, Gaspard's and co-workers'[10] experiment had a very positive role not only in stimulating the discussion about the chaos–noise distinction but also in focusing the attention on deep conceptual aspects of diffusion. In this context, from a theoretical point of view, the study of chaotic models exhibiting diffusion and their nonchaotic counterpart contributed to a better understanding of the role of chaos on macroscopic diffusion.

In Lorentz gases, the diffusion coefficient is related, by means of periodic orbits expansion methods,[51–53] to chaotic indicators such as the Lyapunov exponents. This suggested, for certain time, that chaos was or might have been the basic ingredient for diffusion. However, as argued by Dettmann and Cohen,[27] even an accurate numerical analysis based on the ϵ-entropy has no chance to detect differences in the diffusive behavior between a chaotic Lorentz gas and its nonchaotic counterpart, such as the wind-tree Ehrenfest's model. In the latter model, particles (wind) scatter against square obstacles (trees) randomly distributed in the plane but with fixed orientation. Since the reflection by the flat edges of the obstacles cannot produce exponential separation of trajectories, the maximal Lyapunov exponent is zero and the system is not chaotic. In this case the relation between the diffusion coefficient and the Lyapunov exponents is of course nullified.

The result of Ref. 27 implies thus that chaos may be not indispensable for having deterministic diffusion. The question may be now posed on what are the necessary microscopic ingredients to observe deterministic diffusion at large scales. In the wind-tree Ehrenfest's model, most likely, the disorder in the distribution of the obstacles is crucial. In particular, one may conjecture that a finite spatial entropy density, h_S, is necessary to the diffusion. So that deterministic diffusion may be a consequence either of a nonzero "dynamical" entropy ($h_{\mathrm{KS}}>0$) in chaotic systems or of a nonzero "static" entropy ($h_S>0$) for nonchaotic systems. This is key point, because someone can argue that a deterministic infinite system with spatial randomness can be interpreted as an effective stochastic system, but this is probably a "matter of taste." With the aim of clarifying this point, we consider now a spatially disordered nonchaotic model,[54] which is the one-dimensional analog of a two-dimensional nonchaotic Lorentz system with polygonal obstacles. It has the advantage that both the case with finite and zero spatial entropy density can be investigated. Let us start with the map defined by Eqs. (4) and (5), and introduce some modifications to make it nonchaotic. One can proceed as exemplified by Fig. 4, that is by replacing the function (5) on each unit cell by its step-wise approximation that is generated as follows. The first half of C_ℓ is partitioned in N micro-intervals $[\ell+\xi_{n-1}, \ell+\xi_n[$, $n=1,\ldots,N$, with $\xi_0=0<\xi_1<\xi_2<\ldots<\xi_{N-1}<\xi_N=1/2$. In each interval the map is defined by its linear approximation

$$F_\Delta(u) = u - \xi_n + F(\xi_n) \quad \text{if } u \in [\xi_{n-1},\xi_n[, \tag{18}$$

where $F(\xi_n)$ is (5) evaluated at ξ_n. The map in the second half of the unit cell is then determined by the antisymmetry condition with respect to the middle of the cell. The quenched random variables $\{\xi_k\}_{k=1}^{N-1}$ are uniformly distributed in the interval $[0, 1/2]$, i.e., the micro-intervals have a *random* extension. Further they are chosen independently in each cell C_ℓ (so one should properly write $\xi_n^{(\ell)}$). All cells are partitioned into the same number N of randomly chosen micro-intervals (of mean size $\Delta=1/N$). The modification of

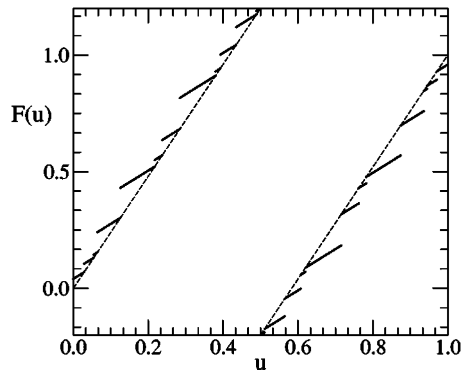

FIG. 4. Sketch of the random staircase map in the unitary cell. The parameter a defining the macroscopic slope is set to 0.23. Half domain $[0, 1/2]$ is divided into $N=12$ micro-intervals of random size. The map on $[1/2, 1]$ is obtained by applying the antisymmetric transformation with respect to the center of the cell $(1/2, 1/2)$.

the continuous chaotic system is conceptually equivalent to replacing circular by polygonal obstacles in the Lorentz system.[27] The steps with unitary slope are indeed the analogous of the flat boundaries of the polygon. While the discontinuities in F_Δ, allowing for a moderate dispersion of trajectories, play a role similar to the vertex of the polygon that splits a narrow beam of particles hitting on it. Since F_Δ has slope 1 almost everywhere, the map is no longer chaotic, violating the condition (i) (see Sec. II). For $\Delta\to 0$ (i.e., $N\to\infty$) the continuous chaotic map (4) is recovered. However, this limit is singular and as soon as the number of intervals is finite, even if extremely large, chaos is absent. It has been found[54] that this model still exhibits diffusion in the presence of both quenched disorder and a quasi-periodic external perturbation

$$x(t+1) = [x(t)] + F_\Delta(x(t) - [x(t)]) + \varepsilon \cos(\alpha t). \tag{19}$$

The strength of the external forcing is controlled by ε and α defines its frequency, while Δ indicates a specific quenched disorder realization.

The diffusion coefficient D has been numerically computed from the linear asymptotic behavior of the mean square displacement. The results, summarized in Fig. 5, show that D is significantly different from zero only for values $\varepsilon>\varepsilon_c$. For $\varepsilon>\varepsilon_c$, D exhibits a saturation close to the value of the chaotic system (horizontal line) defined by Eqs. (4) and (5). The existence of a threshold ε_c is not surprising. Due to the staircase nature of the system, the perturbation has to exceed the typical discontinuity of F_Δ to activate the "macroscopic" instability which is the first step toward the diffusion. Data collapsing, obtained by plotting D vs εN, in Fig. 5 confirms this argument. These findings are robust and do not depend on the details of forcing. Therefore, we have an example of a nonchaotic model in the Lyapunov sense by construction, which performs diffusion. Now the question concerns the possibility that the diffusive behavior arises from the presence of a quenched randomness with nonzero spatial entropy per unit length. To clarify this point, similarly to Ref. 27, the model can be modified in such a way that the spatial entropy per unit cell is forced to be zero, and see if the diffusion still persists.

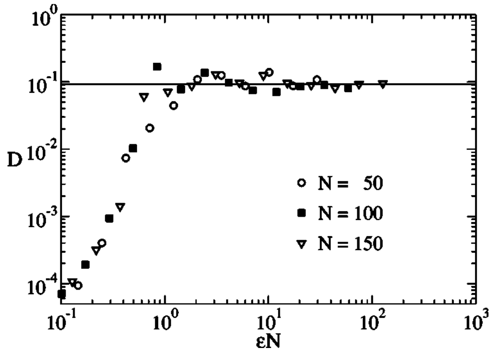

FIG. 5. Log–log plot of the dependence of the diffusion coefficient D on the external forcing strength ϵ. Different data relative to a number of cell micro-intervals $N=50$, 100, and 150 are plotted vs the natural scaling variable ϵN to obtain a collapse of the curves. Horizontal line represents the result for chaotic system (4) and (5).

This program can be accomplished by repeating the same disorder configuration every M cells (i.e., $\xi_n^{(\ell)}=\xi_n^{(\ell+M)}$), ensuring a zero entropy for unit length. Looking at the diffusion of an ensemble of walkers it was observed that diffusion is still present with D very close to the expected value (as in Fig. 5). However, a close analysis reveals the presence of weak average drift V, that vanishes approximately as $V \sim 1/M$ for large M. This suggests that, at large times, $\langle (x(t))^2 \rangle \simeq (Vt)^2+2Dt$ and the ballistic motion should overcome diffusion. However, the crossover time τ_c, at which the motion switches from diffusive to ballistic, diverges with M as $\tau_c \sim DM^2$, so for very large but finite M, the ballistic regime is not observed in simulations. Finally, it should be considered that the value of V depends on the realization of the randomness, and after averaging over the disorder the drift becomes zero. Indeed the behavior $V\sim 1/M$ indicates a self-averaging property for large M. Therefore, we can conclude that the system displays genuine diffusion for a very long time even with a vanishing (spatial) entropy density, at least for sufficiently large M.

These results along with those by Dettmann and Cohen[27] allow us to draw some conclusions on the fundamental ingredients for observing deterministic diffusion (both in chaotic and nonchaotic systems).

- An instability mechanism is necessary to ensure particle dispersion at small scales (here small means inside the cells). In chaotic systems this is realized by the sensitivity to the initial condition. In nonchaotic systems this may be induced by finite size instability mechanisms. Also with zero maximal Lyapunov exponent one can have a fast increasing of the distance between two trajectories initially close.[55] In the wind-tree Ehrenfest model this stems from the edges of the obstacles, in "stepwise" system (5), (18), and (19) from the jumps.
- Mechanisms able to suppress periodic orbits and, therefore, to allow for a diffusion at large scale.

It is clear that the first requirement is not very strong while the the second is more subtle. In systems with "strong chaos," all periodic orbits are unstable and, so, it is automatically fulfilled. In nonchaotic systems, such as the nonchaotic billiards studied by Dettmann and Cohen and the map (5), (18), and (19), the stable periodic orbits seem to be suppressed or, at least, strongly depressed, by the quenched randomness (also in the limit of zero spatial entropy). We note that, unlike the two-dimensional nonchaotic billiards, in the one-dimensional (1D) system (5), (18), and (19), the periodic orbits may survive to the presence of disorder, so we need the aid of a quasiperiodic perturbation to obtain their destruction and the consequent diffusion.

V. DISCUSSIONS AND CONCLUSIONS

Before summarizing the results of this article, we believe that it is conceptually important to comment about the relevance of chaos in statistical mechanics approaches.

The statistical mechanics[56] had been funded by Maxwell, Boltzmann, and Gibbs for systems with a very large number of degrees of freedom without any precise requirement on the microscopic dynamics, apart from the assumption of ergodicity. After the discovery of deterministic chaos it becomes clear that also in systems with few degrees of freedom statistical approaches are necessary. But, even after many years, the experts do not agree yet on the fundamental ingredients which should ensure the validity of the statistical mechanics.

The spectrum of points of view is very wide, ranging from the Landau (and Khinchin[57]) belief on the main role of the many degrees of freedom and the (almost) complete irrelevance of ergodicity, to the opinion of who, as Prigogine and his school,[58,59] considers chaos as the basic ingredient. We strongly recommend the reading of Ref. 60 for a detailed discussion of irreversibility. This work discusses the "orthodox" point of view (based on the role of the large number of degrees of freedom, as stressed by Boltzmann[61]) re-proposed by Lebowitz[62] and the following debate on the role of deterministic chaos.[58,63] Here we focus only on the aspects related to diffusion problems (and some related aspects, e.g., conduction).

By means of the powerful method of periodic orbits expansion, in systems with very strong chaos (namely hyperbolic systems), it has been shown that there exists a close relation between transport properties (e.g., viscosity, thermal and electrical conductivity and diffusion coefficients) and indicators of chaos (Lyapunov exponents, KS entropy, escape rate). These aspects are, e.g., discussed in Refs. 51 and 52. At a first glance, the existence of such relations seem to give evidence against the "anti dynamical" point of view of Landau and Khinchin. However, it may be incorrect to employ those results to obtain definite answers valid for generic systems. In fact it seems to us that there are rather clear evidences that chaos is not a necessary condition for the validity of some statistical behavior.[64–66] Beyond the problem of diffusion in nonchaotic systems, it is worth mentioning the interesting results of Lepri *et al.*[67] showing that the Gallavotti–Cohen formula,[68] originally proposed for chaotic systems, holds also in some nonchaotic model. Moreover, recently Li and co-workers[69,70] studied the transport properties in quasi-one-dimensional channels with triangular scatters. In such systems, the maximal Lyapunov exponent is zero because of the flatness of triangle sides. However, numerical simulations show that, when the scatterers are placed at random (or their height is random), the Fourier heat law remains valid. Another interesting nonchaotic model exhibiting the Fourier heat conduction is the simple one-dimensional hard-particle system with alternating masses.[71] For a recent review on heat conduction in one dimension see Ref. 72.

These and many other examples prove that the heat conduction is present also in system without microscopic chaos. This is a further indication that microscopic chaos is not the unique possible source of a macroscopic transport in a given dynamical system.

Finally let us briefly summarize the main items of this article. The problem of distinguishing chaos from noise cannot receive an absolute answer in the framework of time series analysis. This is due to the finiteness of the observational data set and the impossibility to reach an arbitrary fine resolution and high embedding dimension. However, this restriction is not necessarily negative, and we can classify the signal behavior, without referring to any specific model, as stochastic or deterministic on a certain range of scales.

Diffusion may be realized in stochastic and deterministic systems. In particular, in the latter case, chaos is not a prerequisite for observing diffusion and, more in general, nontrivial statistical behaviors.

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