Anomalous diffusion: Testing ergodicity breaking in experimental data

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Recent advances in single-molecule experiments show that various complex systems display nonergodic behavior. In this paper, we show how to test ergodicity and ergodicity breaking in experimental data. Exploiting the so-called dynamical functional, we introduce a simple test which allows us to verify ergodic properties of a real-life process. The test can be applied to a large family of stationary infinitely divisible processes. We check the performance of the test for various simulated processes and apply it to experimental data describing the motion of mRNA molecules inside live *Escherichia coli* cells. We show that the data satisfy necessary conditions for mixing and ergodicity. The detailed analysis is presented in the supplementary material.

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I. INTRODUCTION

Several techniques including fluorescence correlation spectroscopy, single-particle tracking, and fluorescence recovery after photobleaching have been used to uncover anomalous diffusion in crowded fluids, for example, in the cytoplasm of living cells [1,2]. Ergodicity and the related Boltzmann ergodic hypothesis are the fundamental concepts in statistical physics [3]. Their importance stems from the fact that for ergodic systems the phase average of an observable quantity can be compared with its infinite-time average. This implies that observing one long trajectory of an ergodic process is equivalent to observing a large number of its independent realizations. This property is crucial in the context of conducting physical experiments. The substantial progress in single-particle tracking experiments [1,4–9] was accompanied by theoretical studies on the ergodic properties of systems displaying anomalous behavior [10–24]. Experimental measurements confirmed ergodicity breaking in blinking quantum dots systems [5,6] as well as in the lipid granules in living fission yeast cells [9]. Theoretical studies on weak ergodicity breaking for continuous-time random walks were introduced in [13]. These concepts were extended to the case of fractional Fokker-Planck equations describing subdiffusive dynamics in the presence of an external potential [15,19]. Ergodicity of anomalous dynamics following the generalized Langevin equations was studied in [12,17]; see also [10,11,14]. The relationship between ergodicity and irreversibility was investigated in [16]. A detailed analysis of the ensemble and time average mean-square displacement of fractional Brownian motion and its extensions can be found in [18,20,24]. The generalized Khinchin theorem for Lévy flights [21] and ergodic properties of infinitely divisible (ID) processes recently were fully characterized in [22] in terms of correlation cascades [25].

In this paper, we solve the challenging problem of how to verify ergodic properties (ergodicity and mixing) from empirical data. Exploring the concept of dynamical functional, we introduce a test which allows us to test ergodicity and ergodicity breaking in experimental data. We also show how to verify mixing—an important property of chaotic systems which is stronger than ergodicity. We illustrate the strength of the introduced test by applying it to simulated data (Ornstein-Uhlenbeck process, harmonizable process) and to the real-life data describing the motion of mRNA molecules inside live *Escherichia coli* cells [1].

II. ERGODICITY, MIXING, AND DYNAMICAL FUNCTIONAL

First of all, we emphasize that all the results and methods presented below apply to the general class of stationary ID processes. The physical interpretation of stationarity is that the system is in its thermal equilibrium [26]. The class of ID processes considered here plays a central role in the theory of stochastic processes and their applications [27,28]. Prominent examples of ID distributions are Gaussian, α-stable, Pareto, exponential, γ, Linnik, Mittag-Leffler, and tempered α-stable distributions.

Let us consider a stationary ID stochastic process $Y(n)$, $n \in \mathbb{N}$. $Y(n)$ can be represented as a probability measure $\mathbb{P}$ on the space $(\Omega,\mathcal{B})$. Here, Ω is the phase space of all the functions $f : \mathbb{N} \to \mathbb{R}$ and $\mathcal{B}$ is the σ algebra of events [28]. The probability space $(\Omega,\mathcal{B},\mathbb{P})$ together with the usual shift transformation $S : \Omega \to \Omega$, $S[f(n)] = f(n+1)$, is a standard dynamical system that fully describes the evolution in time of the process $Y(n)$.

The dynamical system $(\Omega,\mathcal{B},\mathbb{P},S)$ is ergodic [or equivalently, the process $Y(n)$ is ergodic] if for every invariant set $A \in \mathcal{B}$ we have that $\mathbb{P}(A) = 0$ or $\mathbb{P}(A) = 1$ [26]. Recall that the set A is invariant if $\mathbb{P}(A) = \mathbb{P}[S^{-1}(A)]$. More intuitively, ergodicity means that the phase space Ω cannot be divided into two nontrivial sets such that a point starting in one set will never get to the second set. It should be emphasized that for every stationary and ergodic process the Boltzmann ergodic hypothesis is satisfied—the temporal and ensemble averages coincide [29,30],

$$\lim_{n\to\infty} \frac{1}{n} \sum_{k=0}^{n-1} g[Y(k)] = \langle g[Y(0)] \rangle, \tag{1}$$

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provided that $\langle g[Y(0)]\rangle$ is well defined. Here, $g(\cdot)$ is a deterministic function, and by $\langle\cdot\rangle$ we denote the ensemble average.

Another fundamental property investigated in this paper is mixing. We say that the dynamical system $(\Omega,\mathcal{B},\mathbb{P},S)$ is mixing [or equivalently, the process $Y(n)$ is mixing] if

$$\lim_{n\to\infty}\mathbb{P}[A\cap S^n(B)]=\mathbb{P}(A)\mathbb{P}(B) \tag{2}$$

for all $A,B\in\mathcal{B}$. Here, by S^n we denote n-fold superposition of S. Thus, mixing can be viewed as the asymptotic independence of the sets A and B under the transformation S. It is well known that mixing is a stronger property than ergodicity [26]. Thus, to show ergodicity, it is enough to prove mixing, which is easier in many cases [22].

A solution to the problem of ergodicity and mixing of Gaussian stationary processes was found by Maruyama, Grenander, Fomin, and Itô [31–34]. The first three authors proved that the stationary Gaussian process $Y(t)$ is ergodic if and only if its spectral measure has no atoms. Itô proved that $Y(t)$ is mixing if and only if its autocorrelation function vanishes at infinity. For the α-stable case, see [35].

In recent papers [21,22,36], ergodic properties of ID processes were described using the concept of correlation cascades. However, in the context of empirical data analysis, a mathematical tool called *dynamical functional* gives very satisfactory results [28,37].

The dynamical functional $D(n)$ corresponding to the process $Y(n)$ is defined as

$$D(n)=\langle\exp\{i[Y(n)-Y(0)]\}\rangle\,. \tag{3}$$

Thus, $D(n)$ is actually a Fourier transform of $Y(n)-Y(0)$ evaluated for the Fourier-space variable $k=1$. The following result illustrates the strength of the dynamical functional [37,38]. The stationary ID process $Y(n)$ is mixing if and only if

$$\lim_{n\to\infty}D(n)=|\langle\exp\{iY(0)\}\rangle|^2. \tag{4}$$

The above condition should be viewed as the asymptotic independence of $Y(n)$ and $Y(0)$ as $n\to\infty$. Moreover, if $Y(n)$ is Gaussian, then the dynamical functional is equal to $D(n)=\exp\{\sigma^2[r(n)-1]\}$, where $r(n)$ is the autocorrelation function of Y and σ^2 is the variance of $Y(0)$. Thus, in the Gaussian case, condition (4) is equivalent to the fact that $r(n)\to 0$ as $n\to\infty$ (cf. [34]).

The above condition (4) can be written in the equivalent form

$$\lim_{n\to\infty}E(n)=0, \tag{5}$$

where

$$E(n)=D(n)-|\langle\exp\{iY(0)\}\rangle|^2. \tag{6}$$

Consequently, using formula (4) and the result of Koopman and von Neumann [39], we get that the stationary ID process $Y(n)$ is ergodic if and only if

$$\lim_{n\to\infty}\frac{1}{n}\sum_{k=0}^{n-1}D(k)=|\langle\exp\{iY(0)\}\rangle|^2. \tag{7}$$

Equivalently, $Y(n)$ is ergodic if and only if

$$\lim_{n\to\infty}\frac{1}{n}\sum_{k=0}^{n-1}E(k)=0. \tag{8}$$

For more details on the origins of conditions (4) and (7), see [37], Lemma 3. As we show in the next section, the above results can be successfully applied to verify ergodicity and mixing in experimental data.

III. TESTING ERGODICITY AND MIXING

Suppose now that we have at our disposal experimental measurements of some random process $Y(n)$. Our goal is to check whether the process is ergodic and mixing. If the ID process is stationary, then the results of previous section allow us to verify its ergodic properties very efficiently. In the case when the number of experimental realizations of $Y(n)$ is large enough to calculate ensemble averages, the procedure is the following:

One calculates the ensemble averages on the right side of (3) and on the right side of (4). If the convergence in (5) holds for large n, then the process is mixing, otherwise $Y(n)$ displays mixing breaking.

We have applied the above test to the simulated trajectories of two different processes. The first one, the classical Ornstein-Uhlenbeck process, is given by the Langevin equation

$$dY(t)=-0.05Y(t)dt+dB(t), \tag{9}$$

where $B(t)$ is the standard Brownian motion. The results are presented in Fig. 1. They confirm that the Ornstein-Uhlenbeck process is mixing.

We have also tested the nonmixing and nonergodic Gaussian process of the form

$$Y(n)=\sqrt{T}\cos(0.5n+\theta), \tag{10}$$

where T is an exponentially distributed random variable with parameter 0.5 and θ is uniformly distributed on $[0,2\pi]$. The result of the test is shown in Fig. 2 (left panel). It confirms that the process is not mixing.

Analogous procedure can be applied to verify ergodicity of experimentally observed process:

If the convergence in (8) holds for large n, then the process is ergodic; otherwise $Y(n)$ displays ergodicity breaking.

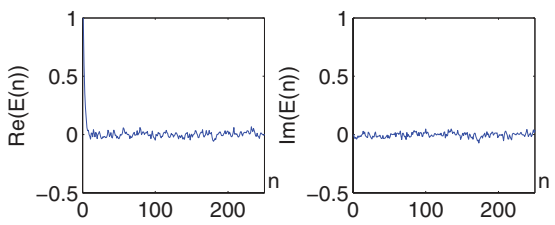

FIG. 1. (Color online) Verification of the mixing property for the Ornstein-Uhlenbeck process. The real and imaginary parts of the function $E(n)$ decay to zero. Thus, condition (5) is satisfied and the process is mixing. The ensemble averages were calculated on the basis of 1000 simulated trajectories.

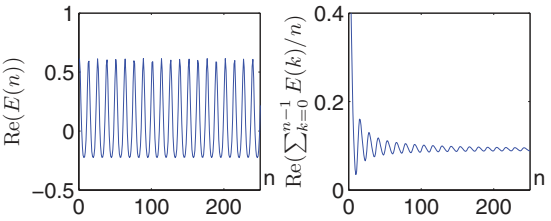

FIG. 2. (Color online) Left panel: testing of mixing property for the Gaussian process defined in (10). Clearly, condition (5) is not satisfied, and thus the process is not mixing. Right panel: real part of the function $\frac{1}{n}\sum_{k=0}^{n-1} E(k)$ corresponding to the process (10). Since condition (8) is violated, the process displays ergodicity breaking. The ensemble averages were calculated on the basis of 1000 simulated trajectories.

In Fig. 2 (right panel), we see the result of the test for the Gaussian process (10). Clearly, the process displays ergodicity breaking. Figure 3 depicts the results of the test for the Ornstein-Uhlenbeck process. Since condition (8) is satisfied, the process is ergodic.

We have also applied the test to the stationary stable harmonizable process [28] of the form

$$Y(t) = A^{1/2}[G_1 \cos(t) + G_2 \sin(t)]. \tag{11}$$

Here, $A > 0$ is the one-sided α-stable random variable, and G_1 and G_2 are standard normal random variables. Moreover, A, G_1, and G_2 are independent. $Y(t)$ is known to be nonergodic [35]. Results in Fig. 4 confirm this fact. Clearly, the real part of $\frac{1}{n}\sum_{k=0}^{n-1} E(k)$ does not converge to zero.

Since the dynamical functional is a Fourier transform of $Y(n) - Y(0)$, it takes values in the interval $[-1,1]$. Therefore, this interval determines the order of magnitude of the y axes in the figures.

IV. ONE-TRAJECTORY CASE

It gets much more complicated when there are not enough trajectories to calculate ensemble averages. Suppose that we have only one realization of the process $Y(n)$, $n = 0,1,\ldots,N$, where N is an appropriately large integer. If we assume that $Y(n)$ is mixing, then Boltzmann ergodic hypothesis is satisfied—the temporal and ensemble averages coincide. Thus, the dynamical functional $D(n)$ in (3) can be approximated by

$$\widehat{D}(n) = \frac{1}{N-n+1}\sum_{k=0}^{N-n} \exp\{i[Y(n+k) - Y(k)]\},$$

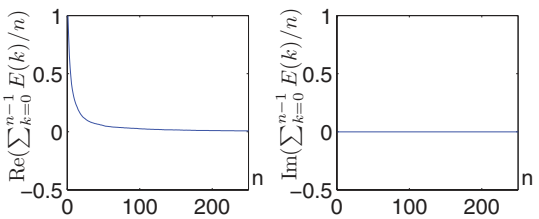

FIG. 3. (Color online) Verification of ergodicity for the Ornstein-Uhlenbeck process. Clearly, condition (8) is satisfied. This confirms that the process is ergodic. The ensemble averages were calculated on the basis of 1000 simulated trajectories.

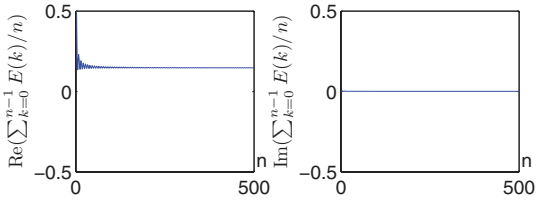

FIG. 4. (Color online) Verification of ergodicity breaking for harmonizable process (11). Since condition (8) is not satisfied, the process displays ergodicity breaking. The ensemble averages were calculated on the basis of 1000 simulated trajectories.

whereas the ensemble average on the right side of (4) can be approximated by

$$\widehat{a} = \left|\frac{1}{N+1}\sum_{k=0}^{N} \exp\{iY(k)\}\right|^2.$$

Then, the modified condition (5) takes the form

$$\widehat{E}(n) \approx 0 \tag{12}$$

for large n. Here

$$\widehat{E}(n) = \widehat{D}(n) - \widehat{a}.$$

Condition (12) is necessary for mixing. Therefore, violation of (12) implies that $Y(n)$ does not have the mixing property. It should be emphasized that (12) is by no means sufficient for mixing. This means that we can only prove lack of mixing if we have one trajectory of a random process. To show that the process is mixing, ensemble averages need to be calculated.

In Fig. 5 we observe the behavior of the function $\widehat{E}(n)$ for one trajectory of the harmonizable process (11). This process is known to be nonergodic and nonmixing [35].

Analogous considerations can be conducted for ergodicity. Having one realization of a process, we can check the following condition:

$$\frac{1}{n}\sum_{k=0}^{n-1} \widehat{E}(k) \approx 0 \tag{13}$$

for large n. The above condition is necessary for ergodicity. This means that its violation implies ergodicity breaking. Similarly as for mixing, more than one trajectory are needed to check the sufficient condition for ergodicity (8).

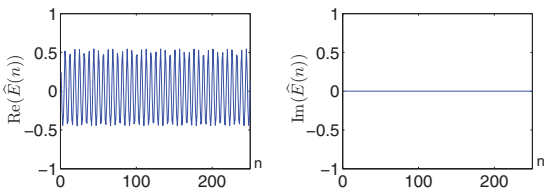

FIG. 5. (Color online) Real and imaginary parts of the function $\widehat{E}(n)$ corresponding to one simulated trajectory of the stable harmonizable process. Since condition (12) is not satisfied, the process is not mixing. Length of the simulated trajectory $N = 2^{12} + 1$.

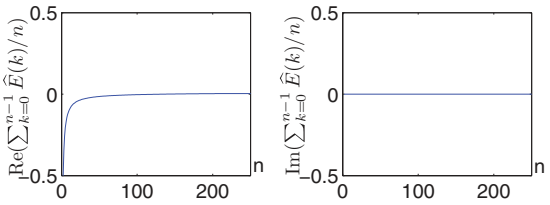

FIG. 6. (Color online) The real and imaginary parts of the function $\sum_{k=0}^{n-1}\widehat{E}(k)/n$ corresponding to the longest trajectory of Golding-Cox data (x coordinate). The necessary condition for ergodicity (13) is clearly satisfied.

V. EXPERIMENTAL DATA

Next, we applied our test to Golding-Cox data [1], describing the motion of mRNA molecules inside live *E. coli* cells. In the whole data set, there were not enough trajectories to calculate ensemble averages. Consequently, we were able to check only the necessary condition for mixing (12) and ergodicity (13), which requires only one appropriately long trajectory. In Fig. 6 we see the result of the test for the longest Golding-Cox trajectory. The necessary condition (13) is clearly satisfied. However, to make sure that the process is ergodic, one needs to verify the sufficient condition (8). This requires more than one trajectory for analysis. We have also analyzed all the other Golding-Cox trajectories that were longer than $2^9 = 512$ points. All these sample paths satisfied necessary conditions for mixing (12) and ergodicity (13). The detailed analysis is presented in the supplementary material [40].

VI. CONCLUDING REMARKS

In this paper, we have discussed necessary and sufficient conditions for mixing (5) and ergodicity (8) in the language of the dynamical functional (3). The main finding is a simple test, which can be applied to verify mixing and ergodicity in experimental data. The test can be applied to the whole family of ID stationary processes. The reasonable length of each analyzed trajectory should not be shorter than 500 points; see [40].

We have also analyzed the case when there is only one experimentally recorded realization of the process at disposal. In this case, the necessary conditions for mixing (12) and ergodicity (13) can be verified. This means that violation of (12) or (13) implies mixing breaking or ergodicity breaking, respectively. To make a definite statement about mixing and ergodicity, more than one trajectory are needed in order to calculate ensemble averages.

The number of trajectories needed to calculate ensemble averages depends strongly on the underlying distribution. For the Gaussian case it is enough to have about 100 trajectories, but it is not enough for a heavy-tailed α-stable law. Every distribution needs to be analyzed separately. Therefore, we encourage experimentalists to make measurements with more trajectories and with higher resolution. Then, the proposed here methodology will allow to rigorously verify ergodicity and mixing; see Figs. 1–3.

The introduced test can be applied to anomalous diffusion processes measured experimentally; see Ref. [40]. We hope that these results, in conjunction with our earlier studies [21,22], can be used to identify the source of anomalous diffusion [41,42].

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