

## Distribution of Time-Averaged Observables for Weak Ergodicity Breaking

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We find a general formula for the distribution of time-averaged observables for systems modeled according to the subdiffusive continuous time random walk. For Gaussian random walks coupled to a thermal bath we recover ergodicity and Boltzmann's statistics, while for the anomalous subdiffusive case a weakly nonergodic statistical mechanical framework is constructed, which is based on Lévy's generalized central limit theorem. As an example we calculate the distribution of  $\bar{X}$ , the time average of the position of the particle, for unbiased and uniformly biased particles, and show that  $\bar{X}$  exhibits large fluctuations compared with the ensemble average  $\langle X \rangle$ .

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A central pillar of statistical mechanics is the ergodic hypothesis, which yields the equivalence of time and ensemble averages in the limit of long measurement time  $t$ . The Deborah number  $D_e = t_p/t$  is the ratio of the time scale of relaxation of the physical phenomenon under observation  $t_p$  and the time of observation [1]. For a system to exhibit ergodic behavior,  $D_e$  must be small. Recently there is much interest in weak ergodicity breaking [2], where the Deborah number diverges [3–7]. Weakly nonergodic behavior is found in systems whose dynamics are characterized by power law distributed sojourn times in microstates of the system, in such a way that the averaged waiting time is infinite (i.e., scale-free dynamics). Weak ergodicity breaking was investigated for blinking quantum dots [3], intermittent nonlinear maps generating subdiffusion deterministically [5], numerical simulations of fractional transport in a washboard potential [6], and *in vivo* gene regulation by DNA-binding proteins [7]. On the stochastic level, all these systems are modeled using the well-known continuous time random walk (CTRW) approach or the corresponding fractional Fokker-Planck equation [8–11]. Previously, nontrivial statistics of occupation times for the CTRW model were found, and it is well established that time averages remain random variables even in the limit of long measurement time [4]. The main open theoretical challenge is to find the distribution of time averages of physical observables. Such a general theory, presented in this Letter, gives analytical estimates for the statistical deviations of time averages from ensemble averages. The theory replaces standard ergodic statistical mechanics and is applicable for a wide class of systems modeled using the CTRW or the related fractional Fokker-Planck equation.

We consider the one-dimensional CTRW on a lattice, with lattice points  $x = 1, \dots, L$ . After waiting, the particle can jump to one of its nearest neighbors; with probability  $q_x$  it jumps to its left, and with probability  $1 - q_x$  to its right. The waiting times on lattice cells are independent identically distributed random variables with a common probability density function (PDF)  $\psi(\tau)$ . We consider the widely applicable case [8–12], where the PDF of the wait-

ing times behaves like  $\psi(\tau) \sim A_\alpha \tau^{-1-\alpha}/|\Gamma(-\alpha)|$  with  $0 < \alpha < 1$ ,  $A_\alpha > 0$  when  $\tau \rightarrow \infty$ . In this case the average waiting time is infinite and the Deborah number diverges. Such waiting times yield anomalous subdiffusion and are well investigated [8–12], in the context of chaotic dynamics [13], geophysics [14], subdiffusive chemical reactions which are important in biological applications [15], and charge transport in amorphous semiconductors [16], to name a few examples. The vast literature on the CTRW deals mainly with ensemble averages of physical observables; for example, the behavior of the ensemble average of the coordinate  $\langle X \rangle$  was thoroughly investigated in many physical situations. Here we investigate the time averages; for example, we will find the distribution of  $\bar{X}$ .

Two types of CTRWs are considered. Thermal random walks describe a physical situation where the particle is coupled to a thermal heat bath with a temperature  $T$  [9,10]. In this case the jump probabilities  $q_x$  satisfy usual detailed balance conditions which relate  $q_x$  with an external force field  $F(x)$  acting on the system and temperature  $T$  [4,9,10]. When these conditions are imposed on the dynamics an ensemble of noninteracting particles attains Boltzmann equilibrium. A second class of random walks is nonthermal and this situation may describe a system far from thermal equilibrium. In this case the ensemble reaches an equilibrium which depends, of course, on the transition probabilities  $q_x$  (see details below). We will treat the nonergodicity for both cases.

We introduce two types of measurements which we identify with two different types of ensembles. In the first the time average of a physical observable is made for a fixed time  $t$  and  $t \rightarrow \infty$ . Repeating the experiment many times, on an ensemble of trajectories, the distribution of the time average is constructed. In the second approach the number of jumps  $n$  the particle makes is fixed and  $n \rightarrow \infty$ . So in the first ensemble, time is fixed and  $n$  fluctuates, while the opposite situation describes the second case. The fixed  $n$  ensemble is very convenient for calculations and yields the same results as the fixed time approach.

We begin the analysis by considering the random walk where  $n$  is the operational time. The probability of occupy-

ing lattice site  $x$  after  $n$  jumps is given by the discrete time master equation

$$P_x(n+1) = q_{x+1}P_{x+1}(n) + (1 - q_{x-1})P_{x-1}(n). \quad (1)$$

After many jumps  $n \rightarrow \infty$  an equilibrium  $P_x^{\text{eq}}(n+1) = P_x^{\text{eq}}(n)$  is obtained, which satisfies

$$P_x^{\text{eq}} = q_{x+1}P_{x+1}^{\text{eq}} + (1 - q_{x-1})P_{x-1}^{\text{eq}}. \quad (2)$$

Such an equilibrium does not depend on the initial condition of the system [17] and is reached provided that the system is finite, and that  $q_x \neq 1$   $q_x \neq 0$  besides on the boundaries.

We consider the number ensemble where  $n$  is fixed. The time  $t_x$  spent by the particle in lattice cell  $x$  is called the occupation time. The total measurement time is  $t = \sum_{x=1}^L t_x$ . According to the CTRW model the time  $t_x$  is a sum of independent identically distributed sojourn times with the common power law tailed PDF  $\psi(\tau)$ . Let  $n_x$  be the number of sojourn times in cell  $x$ , which is clearly large when  $n \rightarrow \infty$ . For the discrete time random walk described by Eq. (1) we have  $n_x/n = P_x^{\text{eq}}$ . Hence according to generalized central limit theorem [18] the random variable  $t_x$  obeys Lévy statistics. Namely, the Laplace transform of the random variable  $t_x > 0$  equals

$$\langle \exp(-u_x t_x) \rangle = \exp[-A_\alpha P_x^{\text{eq}} n (u_x)^\alpha] \quad (3)$$

and the PDF of  $t_x$  is the inverse Laplace transform  $u_x \rightarrow t_x$  of Eq. (3) [18], denoted with  $l_{\alpha, A_\alpha P_x^{\text{eq}} n}(t_x)$ . For the ergodic case,  $\alpha = 1$  in Eq. (3), the PDF of  $t_x$  is  $\delta(t_x - P_x^{\text{eq}} \langle \tau \rangle n)$ , where  $\langle \tau \rangle = A_1$  is the averaged waiting time, and since  $n \langle \tau \rangle \rightarrow t$  the PDF of  $t_x$  is  $\delta(t_x - P_x^{\text{eq}} t)$ , as expected.

The time average of a physical observable  $\bar{O}$  is

$$\bar{O} = \sum_{x=1}^L \bar{p}_x \mathcal{O}_x, \quad (4)$$

where  $\bar{p}_x = t_x/t$  is the occupation fraction and  $\mathcal{O}_x$  is the value of the physical observable when the particle is in state  $x$ . For example, if the observable  $\mathcal{O}$  is the position  $X$  of the particle we have  $\bar{X} = \sum_{x=1}^L x \bar{p}_x$ . For ergodic systems and in the long time limit  $\bar{p}_x = P_x^{\text{eq}}$  and then the time average is equal to the ensemble average  $\bar{O} = \langle \mathcal{O} \rangle =$

$\sum_{x=1}^L P_x^{\text{eq}} \mathcal{O}_x$ . When  $\alpha < 1$  the dynamics is nonergodic and  $\bar{O}$  is a random variable, even in the long time limit.

To obtain the distribution of  $\bar{O}$  we find now the  $L$  dimensional joint PDF of the occupation fractions  $P_L(\bar{p}_1, \dots, \bar{p}_x, \dots, \bar{p}_L)$  [19]. First note that the  $L$  occupation fractions  $\bar{p}_x$  are constrained according to the condition  $\sum_{x=1}^L \bar{p}_x = 1$ , hence,

$$P_L(\bar{p}_1, \dots, \bar{p}_L) = \delta\left(1 - \sum_{x=1}^L \bar{p}_x\right) \int_0^\infty g(\bar{p}_1, \dots, \bar{p}_{L-1}, t) dt, \quad (5)$$

where  $g(\bar{p}_1, \dots, \bar{p}_{L-1}, t)$  is the  $L$  dimensional joint PDF of the random variables in its parenthesis. Since the occupation times  $t_x$  are all independent we have

$$g(\bar{p}_1, \dots, \bar{p}_{L-1}, t) = \frac{\partial(t_1, \dots, t_{L-1}, t)}{\partial(\bar{p}_1, \dots, \bar{p}_{L-1}, t)} [\prod_{x=1}^{L-1} l_{\alpha, A_\alpha P_x^{\text{eq}} n}(t_x)] \times l_{\alpha, A_\alpha P_L^{\text{eq}} n}\left(t - \sum_{x=1}^{L-1} t_x\right). \quad (6)$$

Calculating the Jacobian, using Eq. (5) and

$$l_{\alpha, A_\alpha P_x^{\text{eq}} n}(t_x) = \frac{1}{(A_\alpha n)^{1/\alpha}} l_{\alpha, P_x^{\text{eq}}}\left(\frac{t_x}{(A_\alpha n)^{1/\alpha}}\right) \quad (7)$$

we find

$$P_L(\bar{p}_1, \dots, \bar{p}_L) = \delta\left(1 - \sum_{x=1}^L \bar{p}_x\right) \times \int_0^\infty dy y^{L-1} \prod_{x=1}^L l_{\alpha, P_x^{\text{eq}}}(y \bar{p}_x). \quad (8)$$

This equation is the key for the calculation of the distribution of the time average  $\bar{O}$ , as we will soon show. The multidimensional PDF of the occupation fractions Eq. (8) is independent of the number of steps  $n$  and  $A_\alpha$ . A derivation of Eq. (8) using the fixed time ensemble will be presented in a longer publication.

To proceed, we investigate the characteristic function  $\langle e^{-u \sum_{x=1}^L \mathcal{O}_x t_x} \rangle_t$  of the random variable  $\sum_{x=1}^L \mathcal{O}_x t_x$  in Laplace  $t \rightarrow s$  space

$$\langle e^{-u \sum_{x=1}^L \mathcal{O}_x t_x} \rangle_s = \int_0^\infty e^{-st} \langle e^{-u \sum_{x=1}^L \mathcal{O}_x t_x} \rangle_t dt. \quad (9)$$

Using Eq. (8), we obtain

$$\begin{aligned} \langle e^{-u \sum_{x=1}^L \mathcal{O}_x t_x} \rangle_s &= \int_0^\infty dt \int_0^\infty dy \int_0^\infty dt_1 \cdots \int_0^\infty dt_L t \delta\left(t - \sum_{x=1}^L t_x\right) y^{L-1} e^{-st-u \sum_{x=1}^L \mathcal{O}_x t_x} \prod_{x=1}^L l_{\alpha, P_x^{\text{eq}}}(y t_x) \\ &= -\frac{d}{ds} \int_0^\infty dy y^{L-1} \int_0^\infty dt_1 \cdots \int_0^\infty dt_L e^{-s \sum_{x=1}^L t_x - u \sum_{x=1}^L \mathcal{O}_x t_x} \prod_{x=1}^L l_{\alpha, P_x^{\text{eq}}}(y t_x) \\ &= -\frac{d}{ds} \int_0^\infty dy y^{L-1} \prod_{x=1}^L \left\{ \frac{\exp[-P_x^{\text{eq}} (\frac{s+\mathcal{O}_x u}{y})^\alpha]}{y} \right\}. \end{aligned} \quad (10)$$

Solving the last integral we find the characteristic function

$$\langle e^{-u \sum_{x=1}^L \mathcal{O}_x t_x} \rangle_s = \frac{\sum_{x=1}^L P_x^{\text{eq}} (s + \mathcal{O}_x u)^{\alpha-1}}{\sum_{x=1}^L P_x^{\text{eq}} (s + \mathcal{O}_x u)^\alpha}. \quad (11)$$

Using inversion technique found in [20], we transform Eq. (11), and we find the PDF of the time average  $\bar{\mathcal{O}}$

$$f_\alpha(\bar{\mathcal{O}}) = -\frac{1}{\pi} \lim_{\epsilon \rightarrow 0} \text{Im} \frac{\sum_{x=1}^L P_x^{\text{eq}} (\bar{\mathcal{O}} - \mathcal{O}_x + i\epsilon)^{\alpha-1}}{\sum_{x=1}^L P_x^{\text{eq}} (\bar{\mathcal{O}} - \mathcal{O}_x + i\epsilon)^\alpha}. \quad (12)$$

This is our main result; it is a very general formula for the distribution of time-averaged observables. When  $\alpha \rightarrow 1$

$$f_{\alpha=1}(\bar{\mathcal{O}}) = \delta(\bar{\mathcal{O}} - \langle \mathcal{O} \rangle) \quad (13)$$

which is the expected ergodic behavior. The opposite limit of  $\alpha \rightarrow 0$ , using  $\sum_{x=1}^L P_x^{\text{eq}} = 1$ , gives

$$\lim_{\alpha \rightarrow 0} f_\alpha(\bar{\mathcal{O}}) = \sum_{x=1}^L P_x^{\text{eq}} \delta(\bar{\mathcal{O}} - \mathcal{O}_x). \quad (14)$$

This makes perfect physical sense, since when  $\alpha \rightarrow 0$  the particle is localized for the whole duration of measurement in a single cell. Note that our results can be easily generalized to dimensions higher than one.

In many applications the continuum behavior of the CTRW is important [10]. Taking the continuum limit of Eq. (12) we find

$$f_\alpha(\bar{\mathcal{O}}) = -\frac{1}{\pi} \lim_{\epsilon \rightarrow 0} \text{Im} \frac{\int_0^L dx P^{\text{eq}}(x) [\bar{\mathcal{O}} - \mathcal{O}(x) + i\epsilon]^{\alpha-1}}{\int_0^L dx P^{\text{eq}}(x) [\bar{\mathcal{O}} - \mathcal{O}(x) + i\epsilon]^\alpha}. \quad (15)$$

Here  $P^{\text{eq}}(x)dx$  is the equilibrium probability (in ensemble sense) of finding the particle in  $(x, x + dx)$  and  $0 < x < L$ . When the random walk is coupled to a thermal heat bath

with temperature  $T$ , in the presence of an external force field  $F(x)$ , the equilibrium of the ensemble is described by Boltzmann's statistics [9,10]

$$P^{\text{eq}}(x) = \frac{\exp[-\frac{V(x)}{k_b T}]}{Z}, \quad (16)$$

where  $Z$  is the partition function and  $F(x) = -dV(x)/dx$ . As mentioned, such an equilibrium is found for the CTRW model when thermal detailed balance conditions are imposed on  $q_x$ . Solving Eq. (15), we have

$$f_\alpha(\bar{\mathcal{O}}) = \frac{\sin \pi \alpha}{\pi} \times \frac{I_{\alpha-1}^<(\bar{\mathcal{O}}) I_{\alpha-1}^>(\bar{\mathcal{O}}) + I_{\alpha-1}^>(\bar{\mathcal{O}}) I_{\alpha-1}^<(\bar{\mathcal{O}})}{[I_{\alpha-1}^>(\bar{\mathcal{O}})]^2 + [I_{\alpha-1}^<(\bar{\mathcal{O}})]^2 + 2 \cos \pi \alpha I_{\alpha-1}^>(\bar{\mathcal{O}}) I_{\alpha-1}^<(\bar{\mathcal{O}})}, \quad (17)$$

where

$$I_{\alpha-1}^<(\bar{\mathcal{O}}) = \int_{\bar{\mathcal{O}} < \mathcal{O}(x)} dx P^{\text{eq}}(x) |\bar{\mathcal{O}} - \mathcal{O}(x)|^\alpha \quad (18)$$

and similarly for  $I_{\alpha-1}^>(\bar{\mathcal{O}})$ ,  $I_{\alpha-1}^<(\bar{\mathcal{O}})$ , and  $I_{\alpha-1}^>(\bar{\mathcal{O}})$ . The integration domain in Eq. (18) is for  $x$  satisfying the condition  $\bar{\mathcal{O}} < \mathcal{O}(x)$ .

As an example, consider a particle in a domain  $-L/2 < x < L/2$  undergoing an unbiased random walk. This is a free particle in the sense that no external field is acting on it. The time average of the particle's position  $\bar{X}$  is considered, and obviously for this case  $P^{\text{eq}}(x) = 1/L$  for  $-L/2 < x < L/2$ . Using Eq. (17) we find the PDF of the time-averaged position

$$f_\alpha(\bar{X}) = \frac{1}{L} \frac{N_\alpha \left(\frac{1}{4} - \frac{\bar{X}^2}{L^2}\right)^\alpha}{\left|\frac{1}{2} - \frac{\bar{X}}{L}\right|^{2(1+\alpha)} + \left|\frac{1}{2} + \frac{\bar{X}}{L}\right|^{2(1+\alpha)} + 2\left|\frac{1}{4} - \left(\frac{\bar{X}}{L}\right)^2\right|^{1+\alpha} \cos \pi \alpha}, \quad (19)$$

where  $N_\alpha = (1 + \alpha) \sin \pi \alpha / (\pi \alpha)$ . When  $\alpha \rightarrow 1$  we have the ergodic behavior  $\bar{X} = \langle X \rangle = 0$  while  $f_{\alpha \rightarrow 0}(\bar{X}) = 1/L$  for  $|\bar{X}| < L/2$  which is the uniform distribution, reflecting the mentioned localization of the particle in space when  $\alpha \rightarrow 0$ . In Fig. 1 comparisons between our analytical results and numerical simulations [21] of the CTRW process with a fixed measurement time  $t$  show excellent agreement without fitting.

As a second example, consider a biased particle in the domain  $0 < x < \infty$  and in a constant force field  $-F < 0$ . Assuming the particle is in contact with a heat bath, with temperature  $T$ , Boltzmann's equilibrium is reached for an ensemble of particles,  $P^{\text{eq}}(x) = \exp(-Fx/k_b T)/Z$ . The PDF of  $\bar{X}$  is found using Eq. (17)

$$f_\alpha(\bar{X}) = \frac{\sin \pi \alpha}{\pi} \frac{F}{k_b T} \times \frac{\Gamma(\alpha) e^{\bar{X}} \bar{X}^\alpha}{\left(\int_0^{\bar{X}} dy e^{y^2} |y|^\alpha\right)^2 + \Gamma^2(1 + \alpha) + 2\Gamma(1 + \alpha) \int_0^{\bar{X}} dy e^{y^2} |y|^\alpha \cos \pi \alpha}, \quad (20)$$

where  $\bar{x} = F\bar{X}/k_b T$ . When  $\alpha \rightarrow 1$  we find ergodicity  $f_1(\bar{X}) = \delta(\bar{X} - \langle X \rangle)$  with  $\langle X \rangle = k_b T/F$  while in the opposite limit  $\alpha \rightarrow 0$ , an exponential decay of the PDF of  $\bar{X}$  is found:  $\lim_{\alpha \rightarrow 0} f_\alpha(\bar{X}) = \exp(-F\bar{X}/k_b T)/Z$  reflecting localization with a profile determined by the equilibrium density of many noninteracting particles. These behaviors are demonstrated in Fig. 2.

We now discuss briefly the meaning of weak ergodicity breaking. In many situations in physics a system is non-

ergodic since its phase space is decomposed into regions of phase space where the system once starting in one region cannot explore the others. In this case time averages depend strongly on the initial condition of the system and there is no full exploration of phase space. In contrast, for weak ergodicity breaking, the particle will visit each lattice cell many times, no matter what its initial condition is. Hence exploration of phase space is possible, and for this reason we were able to construct in this Letter a general

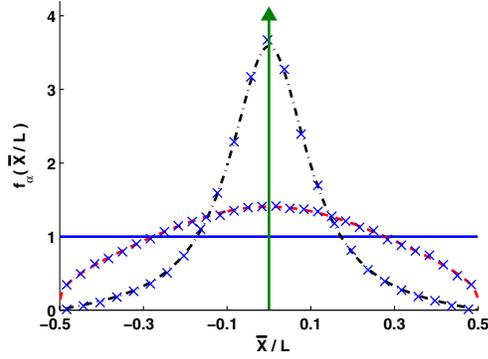


FIG. 1 (color online). The PDF of  $\bar{X}/L$  for unbiased CTRW, simulations (crosses) versus theory (curves) Eq. (19). When  $\alpha = 1$  we find ergodic behavior and  $\bar{X} = \langle X \rangle = 0$  (i.e., the arrow symbolizing a delta function). For  $\alpha = 0.7$  (the dotted dashed curve) and  $\alpha = 0.3$  (the dashed curve) large fluctuations of time averages are observed. When  $\alpha \rightarrow 0$  the PDF of  $\bar{X}$  is uniform reflecting localization of the particle (solid line).

theory of nonergodic statistical mechanics which is not sensitive to the initial conditions of the system. This has several implications; for example, the joint PDF of occupation fractions Eq. (8) and the PDF of time averages Eqs. (15) and (17) are related to the population density  $P^{\text{eq}}(x)$ . Therefore, we may find a general relation between fluctuations of time averages and fluctuations of ensemble averages: using Eq. (11) the average of  $\bar{O}$  is  $\langle \bar{O} \rangle = \langle O \rangle = \int_0^L O(x)P^{\text{eq}}(x)dx$  and

$$\langle \bar{O}^2 \rangle - \langle \bar{O} \rangle^2 = (1 - \alpha)(\langle O^2 \rangle - \langle O \rangle^2) \quad (21)$$

with  $\langle O^2 \rangle = \int_0^L O(x)^2 P^{\text{eq}}(x)dx$ . For the example of a particle in a uniform force field  $F$ , when the physical observable is the position, we have  $\langle \bar{X} \rangle = k_b T/F$  and  $\langle \bar{X}^2 \rangle - \langle \bar{X} \rangle^2 = (1 - \alpha)(k_b T/F)^2$ .

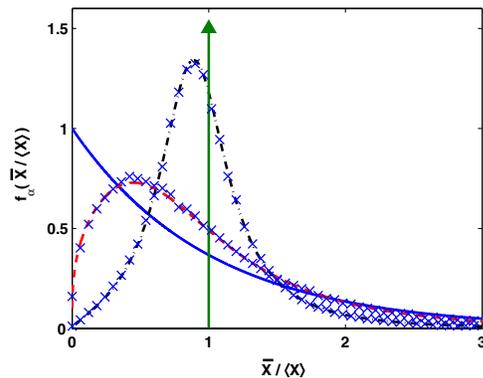


FIG. 2 (color online). Same as Fig. 1 for biased CTRW: now we show the PDF of  $\bar{X}/\langle X \rangle$ . The theoretical curves based on Eq. (20) perfectly agree with simulations (crosses) without fitting. A transition between ergodic behavior for  $\alpha = 1$  (the delta function) to localization behavior (solid curve  $\alpha \rightarrow 0$ ) where the PDF of  $\bar{X}$  decays exponentially is found.

To summarize, we have obtained very general distributions of time averages of physical observables of weakly nonergodic systems Eqs. (12) and (15). Unlike usual ergodic statistical mechanics where the time averages are equal to the ensemble averages, we find large fluctuations of time averages. Because of the large number of applications of the CTRW model, and the recent interest in weak ergodicity breaking in dynamics of single particles, our theory is likely to find its applications in many systems. The deep relations between the stochastic CTRW and other models of anomalous diffusion, e.g., the quenched trap model, and deterministic dynamics, indicate that our nonergodic theory might find further profound justification.

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