# Random walk to a nonergodic equilibrium concept

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(Received 19 June 2005; published 20 January 2006)

Random walk models, such as the trap model, continuous time random walks, and comb models, exhibit weak ergodicity breaking, when the average waiting time is infinite. The open question is, what statistical mechanical theory replaces the canonical Boltzmann-Gibbs theory for such systems? In this paper a nonergodic equilibrium concept is investigated, for a continuous time random walk model in a potential field. In particular we show that in the nonergodic phase the distribution of the occupation time of the particle in a finite region of space approaches U- or W-shaped distributions related to the arcsine law. We show that when conditions of detailed balance are applied, these distributions depend on the partition function of the problem, thus establishing a relation between the nonergodic dynamics and canonical statistical mechanics. In the ergodic phase the distribution function of the occupation times approaches a $\delta$ function centered on the value predicted based on standard Boltzmann-Gibbs statistics. The relation of our work to single-molecule experiments is briefly discussed.

DOI: 10.1103/PhysRevE.73.016125 PACS number(s): 02.50.−r, 05.20.−y, 05.40.−a, 05.90.+m

## I. INTRODUCTION

There is growing interest in nonergodicity of systems whose dynamics is governed by power law waiting times, in such a way that a state of the system is occupied with a sojourn time whose average is infinite. Such nonergodicity, called weak ergodicity breaking [1], was first introduced in the context of glassy dynamics. It has found several applications in physics: phenomenological models of glassy dynamics [1], laser cooling [2], blinking quantum dots [3,4], and models of atomic transport in optical lattices [5]. For example, single blinking quantum dots, when interacting with a continuous wave laser field, turn at random times from a bright state in which many fluorescent photons are emitted to a dark state. It is found that the distribution of dark and bright times follows power law behavior. Somewhat similar statistical behavior is found also for laser cooling of atoms, where the atom is found in two states in momentum space, a cold trapped state and a free state; the sojourn time probability density function has a power law behavior $\psi(\tau) \propto \tau^{-(1+\alpha)}$ with $\alpha < 1$. For such systems the time average of physical observables, for example the time average of fluorescence intensity of single quantum dots, is nonidentical to the ensemble average even in the long time limit. From a stochastic point of view such ergodicity breaking is expected, since the condition to obtain ergodicity is that the measurement time $t$ be much longer than the microscopical time scale of the problem. However, the microscopic time scale in our examples is infinite, namely, the mean trapping times or the mean dark and bright times diverge. When these characteristic time scales are infinite, namely, $\alpha < 1$, we can never make time averages for long enough times to obtain ergodicity.

It is important to note that the concept of a waiting (i.e., trapping) time probability density function (PDF) $\psi(\tau)$, with diverging first moment, is widespread and found in many fields of physics [2,6–10]. It was introduced into the theory of transport of charge carriers in disordered materials [11] in the context of the continuous time random walk (CTRW). The CTRW describes a random walk on a lattice with a waiting time PDF of times between jump events $\psi(\tau)$. The model exhibits anomalous diffusion [11] and aging behaviors [12–15], when $\alpha < 1$, which are related to ergodicity breaking. The CTRW found many applications in the context of chaotic dynamics [14,16,17], tracer diffusion in complex flows [18,19], financial time series [20], and diffusion of beads in a polymer network [21], to name a few [6,7]. Other models with dynamics similar to that of the CTRW are the comb model [8] and the annealed version of the trap model [12]. In turn the trap model is related to the random energy model [22]. All these systems and models can be at least suspected of exhibiting nonergodic behavior, and hence constructing a general theory of nonergodicity for such systems is in our opinion a worthy goal.

Systems and models exhibiting anomalous diffusion, and CTRW behaviors can be divided into two categories: systems where the random walk is close to thermal equilibrium, where the temperature of the system is well defined at least from an experimental point of view, and nonthermal systems. The ergodicity breaking of thermal CTRW models is in conflict with the Boltzmann-Gibbs ergodic assumption. As far as we know, there is no theory characterizing the nonergodic properties of the CTRW for either the thermal or nonthermal type of random walk.

Hence one goal of this paper is to obtain the nonergodic properties of the well known CTRW model on a lattice. Second, we investigate ergodicity breaking and its relation to Boltzmann-Gibbs statistics. Using rather general arguments and using a CTRW model we investigate the distribution of the total occupation times of a lattice point or a state of the system (in Sec. VI we consider the more general case of occupation times in connected and disconnected domains). We show that in the limit of long measurement time and in

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the ergodic phase the occupation times are obtained using the Boltzmann-Gibbs canonical ensemble, provided that detailed balance conditions are satisfied. In the nonergodic phase we obtain nontrivial distributions of the occupation times, which are related to the arcsine law. These limiting distributions are unique in the sense that they do not depend on all the dynamical details of the underlying model. Further, the distributions we obtain depend on Boltzmann's probability, namely, on the temperature $T$ and the partition function $Z$. Thus a connection is established between nonergodic dynamics and the basic tool of statistical mechanics.

The study of occupation times in the context of classical Brownian motion was considered by Lévy. Consider a Brownian path generated with $\dot{x}(t)=\eta(t)$, where $\eta(t)$ is Gaussian white noise, in the time interval $(0,t)$, and with free boundary conditions. The total time $t_+$ the particles spend on the half space $x>0$ is called the occupation time of the positive half space. The fraction of occupation time $p^+=t_+/t$ is distributed according to the celebrated arcsine law [23]

$$\lim_{t\to\infty} f(p^+) = \frac{1}{\pi\sqrt{p^+(1-p^+)}}, \tag{1}$$

where $0\leq p^+\leq 1$. In contrast to naive expectation, it is unlikely to find $p^+=1/2$, which would mean that the particle remains half of the time in $x>0$. Instead $f(p^+)$ diverges at $p^+=0$ and $p^-=1$, indicating that the Brownian particle tends to stay either in $x>0$ or in $x<0$ for long times of the order of the measurement time $t$. Hence $f(p^+)$ has a U shape. Such behavior is related to the survival probability of the Brownian particle. The probability of a Brownian particle starting at $x>0$ remaining in $x>0$ without crossing $x=0$ decays like a power law $t^{-1/2}$. The average time the particle remains in $x>0$, before the first crossing of $x=0$, is infinite. Similar U-shape distributions, in far less trivial examples, were investigated more recently in the context of random walks in random environments [24], renewal processes [25], stochastic processes [26], zero-temperature Glauber spin dynamics [27], diffusion equations [28], two-dimensional Ising model [30], and growing interfaces [29].

The study of nonergodicity within the CTRW framework is timely due to recent single-molecule [31] types of experiments. In many experiments anomalous diffusion and power law behavior was observed using single-particle tracking techniques [3,21,32–34] (e.g., single quantum dots [4]). An interesting example is the diffusive motion of magnetic beads in an actin network [21]. The latter exhibit a CTRW type of behavior while the system has a well defined temperature $T$, namely, the random walk seems close to thermal equilibrium and the particle is coupled to a thermal heat bath. In particular, long tailed $t^{-(1+\alpha)}$ waiting time distributions were recorded and anomalous subdiffusion $\langle r^2\rangle\sim t^{\alpha}$ with $\alpha<1$ was observed. While clearly ensemble average classifications of the anomalous process, e.g., the mean square displacement, are important, it is the time averages of single-particle trajectories that distinguish the single-particle measurement from standard ensemble average type of measurement. Thus stochastic theories of nonergodicity can help with the fundamental question in single-molecule experiments: Are time averages recorded in such experiment identical to the corresponding ensemble averages? and if not how do we classify the nonergodic phase?

This paper is organized as follows. In Sec. II we discuss a possible generalization of Boltzmann-Gibbs statistics for nonergodic dynamics. In Sec. III we introduce the CTRW model, which yields the nonergodic dynamics. Section IV is the main technical part of the paper, in which we obtain first passage time properties of the CTRW. The relation of these properties to the nonergodic behavior is shown. In Sec. V we give the main results and compare between the nonergodic framework and standard Boltzmann-Gibbs statistics. In Sec. VI occupation times on a finite domain (coarse graining) are considered, as well as detailed numerical simulations. A brief summary of some of our results was published recently [35].

## II. FROM BOLTZMANN STATISTICS TO NONERGODICITY

In this section we discuss a possible nonergodic generalization of Boltzmann-Gibbs theory, without attempting to prove its validity.

The basic tool in statistical mechanics is Boltzmann's probability $P_x^B$ of finding a system in a state with energy $E_x$,

$$P_x^B = \frac{\exp(-E_x/T)}{Z}, \tag{2}$$

where $T$ is the temperature and $Z=\sum_x \exp(-E_x/T)$. In Eq. (2) we use the canonical ensemble and assume a classical system, with discrete energy states $\{0\leq E_1\leq E_2\cdots\}$. To obtain the average energy of the system, we use

$$\langle E\rangle = \sum_x E_x P_x^B, \tag{3}$$

and similarly for other physical observables like entropy, free energy, etc. Equation (3) is an ensemble average. When measurement of a single system is made, a time average of a physical observable is recorded. Consider a system randomly changing between its energy states $\{E_x\}$. At a given time the system occupies one energy state. Let $t_x$ be the total time spent by the system in energy state $E_x$, within the total observation period $(0,t)$. The system may visit state $E_x$ many times during the evolution; hence $t_x$ is composed in principle from many sojourn times. We define the occupation fraction

$$\bar{p}_x = \frac{t_x}{t}, \tag{4}$$

and the time average energy is

$$\bar{E} = \sum_x E_x \bar{p}_x. \tag{5}$$

According to statistical mechanics, once the ergodic hypothesis is satisfied, and within the canonical formalism $\bar{p}_x=P_x^B$ and then $\bar{E}=\langle E\rangle$, and similarly for other physical observables. More generally, the occupation fraction $\bar{p}_x$ is a random variable, whose statistical properties depend on the underlying dynamics. If Boltzmann's conditions hold the probability density function of $\bar{p}_x$ is

$$f(\bar{p}_x) = \delta(\bar{p}_x - P_x^B) \tag{6}$$

in the thermodynamic limit. The last equation is a restatement of the ergodic hypothesis.

In this paper we discuss a possible generalization of the ergodic hypothesis. Our proposal is that the PDF of $\bar{p}_x$, for certain models described by CTRW type of dynamics, is described by a $\delta_\alpha$ function

$$f(\bar{p}_x) = \delta_\alpha(\mathcal{R}_x, \bar{p}_x) = \frac{\sin \alpha\pi}{\pi} \frac{\mathcal{R}_x \bar{p}_x^{\alpha-1}(1-\bar{p}_x)^{\alpha-1}}{\mathcal{R}_x^2(1-\bar{p}_x)^{2\alpha} + \bar{p}_x^{2\alpha} + 2\mathcal{R}_x(1-\bar{p}_x)^\alpha \bar{p}_x^\alpha \cos \pi\alpha}, \tag{7}$$

where $0 < \alpha \leq 1$. This PDF was obtained by Lamperti [36] in the context of the mathematical theory of occupation times (see Appendix A for details). For $\mathcal{R}_x = 1$, $\alpha = 1/2$ we have the arcsine law. Here we claim that when local detailed balance condition is satisfied

$$\mathcal{R}_x = \frac{P_x^B}{1 - P_x^B}. \tag{8}$$

When $\alpha = 1$ we get the usual ergodic behavior defined in Eq. (6). Equation (7) is valid only in the limit of long measurement time. In the nonergodic phase $\alpha < 1$, Eqs. (7) and (8) establish a relation between the ergodicity breaking and Boltzmann-Gibbs statistics. The exponent $\alpha$ is the anomalous diffusion exponent in the relation $\langle x^2 \rangle \propto t^\alpha$.

For CTRWs not satisfying detailed balance condition a more general rule holds. We will show that the PDF of the fraction of time spent on lattice point $x$, $\bar{p}_x$, is still given by Eq. (7). However, now

$$\mathcal{R}_x = \frac{P_x^{\mathrm{eq}}}{1 - P_x^{\mathrm{eq}}}, \tag{9}$$

where $P_x^{\mathrm{eq}}$ is the probability that a particle occupies lattice point $x$ in equilibrium (an equilibrium is obtained for a system of finite size). Here $P_x^{\mathrm{eq}}$ and $P_x^B$ are probabilities in the ensemble sense, namely, if we consider an ensemble of $N$ noninteracting particles (or systems) satisfying some dynamical rule, $P_x^{\mathrm{eq}}$ and $P_x^B$ yield in principle the probability that a member of the ensemble occupies state $x$ in equilibrium, which is not identical to $\bar{p}_x$ for nonergodic systems.

Let us give some general arguments for the validity of Eqs. (7) and (8). Consider a particular energy state of the system and call it $E_x$. At a given time the system is either in energy state $E_x$ or in any of the other energy states. When the system does not occupy state $x$ we will say that the system is in state $nx$ (not $x$). Assume that sojourn times in states $x$ and $nx$ are

$$\psi_x(\tau) \sim \frac{A_x}{|\Gamma(-\alpha)|\tau^{1+\alpha}}, \quad \psi_{nx}(\tau) \sim \frac{A_{nx}}{|\Gamma(-\alpha)|\tau^{1+\alpha}} \tag{10}$$

when $\tau$ is large. Also assume that sojourn times in states $x$ and $nx$ are not correlated. Thus we imagine the system occupying state $x$, then occupying state $nx$, then again state $x$, etc. The amplitudes $A_x$ and $A_{nx}$ will generally depend on the particular dynamics of the system. We show in Appendix A that Eq. (7) holds with $\mathcal{R}_x = A_x / A_{nx}$. Generally it seems a hopeless mission to calculate the ratio $A_x/A_{nx}$ from any microscopical model. However, a simple physical argument yields the ratio $\mathcal{R}_x$. Assume that for an ensemble of systems Boltzmann-Gibbs statistical mechanics holds. Such an assumption means that on average we must have

$$\langle \bar{p}_x \rangle = P_x^B, \tag{11}$$

where $P_x^B$ is Boltzmann's probability of finding a member of an ensemble of systems in state $x$. On the other hand, Eq. (7) yields

$$\langle \bar{p}_x \rangle = \int_0^1 \bar{p}_x f(\bar{p}_x) d\bar{p}_x = \frac{\mathcal{R}_x}{1 + \mathcal{R}_x}. \tag{12}$$

Using Eqs. (11) and (12) we obtain Eq. (8).

Our work is related to the concept of weak ergodicity breaking, suggested by Bouchaud [1]. In standard statistical mechanics, one divides the phase space of the system into equally sized cells, and the system is supposed to visit these cells with equal probability under certain constraints (e.g., the energy of the system is constant for the microcanonical ensemble). Strong ergodicity breaking means that in order to leave one phase space cell to go to another, one has to cross a barrier (e.g., an energy barrier) which becomes infinite in the thermodynamic limit. In this case the time it takes for the system to move from one state to the other is infinite. It is worth thinking of such a process in terms of a distribution of escape times, $\psi(\tau) = R \exp(-R\tau)$, where $Rt$ is small even in the thermodynamic limit of long measurement time, $t$. In that case the particle or system simply remains in a certain domain of phase space for the whole period of observation, and the system does not explore its entire phase space available for ergodic systems. A very different scenario was suggested by Bouchaud, in the context of glassy dynamics and the trap model. If the distribution of sticking times follows power law behavior, the average escape time diverges,

$$\langle \tau \rangle \propto \int_0^\infty \tau \tau^{-1-\alpha} d\tau \to \infty, \tag{13}$$

when $\alpha < 1$. Note that also for the strong nonergodicity case we may have an infinite waiting time $\langle \tau \rangle = 1/R$ when $R \to 0$. However, for power law waiting times the system or particle may still explore its phase space. Or, in other words, exponential waiting times and power law waiting times yield very different types of dynamics, even if for both the average waiting time is infinite. Thus, roughly speaking, for weak nonergodicity and for ensemble of particles we may still get Boltzmann-Gibbs statistics, since from any initial condition the phase space is totally covered. However, the system remains weakly nonergodic, since during its evolution, the system will randomly pick one state, which it will occupy for a very long period (but it still visits all the other states) and then time averages are not equal to ensemble averages. The goal of this paper is to show that the strong assumptions we used are correct within a specific model, the well known CTRW model.

## III. CTRW IN A FORCE FIELD

We consider a one-dimensional CTRW on a lattice. The lattice points are labeled with index $x$ and $x=-L,-L+1,\ldots,0,\ldots,L$; hence the system size is $2L+1$. On each lattice point we define a probability $0<Q_R(x)<1$ for jumping right, and a probability for jumping left $Q_L(x)=1-Q_R(x)$. Let $\psi(\tau)$ be the PDF of waiting times at the sites; this PDF does not depend on the position of the particle. If the particle starts at site $x=0$, it will wait there for a period $\tau_1$ determined from $\psi(\tau)$; it will then jump with probability $Q_L(0)$ to the left, and with probability $Q_R(0)$ to the right. After the jump, say to lattice point 1, the particle will pause for a period $\tau_2$, whose statistical properties are determined by $\psi(\tau)$. It will then jump either back to point 0 or to $x=2$, according to the probability law $Q_R(1)$. Then the process is renewed. We consider reflecting boundary conditions, namely, $Q_L(L)=Q_R(-L)=1$.

The case of a long tailed waiting time PDF, where $\psi(\tau)\propto\tau^{-(1+\alpha)}$ when $\tau\to\infty$ and $0<\alpha<1$, yields a nonergodic behavior. In this case the average waiting time is infinite. The Laplace transform of $\psi(\tau)$ is

$$\hat{\psi}(u)=\int_0^\infty e^{-u\tau}\psi(\tau)d\tau. \tag{14}$$

As usual, according to the Tauberian theorem [23], the small $u$ behavior is

$$\hat{\psi}(u)\sim 1-Au^\alpha+\cdots \tag{15}$$

and $A>0$ is a constant.

Choose a specific lattice point $x$ and then define $\theta_x(t)=1$ if the particle is on $x$; otherwise it is zero. We define the occupation fraction as the time average of $\theta_x(t)$,

$$\bar{p}_x=\frac{\int_0^t\theta_x(t')dt'}{t}, \tag{16}$$

namely, $\bar{p}_x=t_x/t$, where $t_x$ is the total time spent on lattice point $x$ (i.e., the occupation time of site $x$). We will later calculate the PDF of $\bar{p}_x$.

Two special cases are the unbiased CTRW, where $Q_L(x)=Q_R(x)=1/2$, and the uniformly biased CTRW with $Q_L(x)=q$. In these cases no transition probabilities depend on the position of the random walker $x$, except on the boundaries of course. In the language of random walks these cases describe a symmetric diffusion process and diffusion with a drift. Note that in our model $Q_L(x)$ are not random variables; rather they are included in the model to mimic a deterministic potential field acting on the system. For detailed discussion of CTRW models see [6,7].

The case of diffusion with a constant drift, i.e., $q\neq 1/2$, is used many times to model diffusion under the influence of a constant external driving force $\mathcal{F}$. If the physical process is close to thermal equilibrium the condition of detailed balance is imposed on the dynamics, in order that for an ensemble of particles Boltzmann equilibrium is reached [see further discussion after Eq. (19)]. The potential energy at each point $x$ due to the interaction with the external driving force is $E(x)=-\mathcal{F}ax$, where $a$ is the lattice spacing. The condition of detailed balance then reads

$$\frac{Q_L(x)}{Q_R(x)}=\exp\left(-\frac{\mathcal{F}a}{T}\right), \tag{17}$$

note that the right hand side of Eq. (17) is independent of the lattice coordinate. Since $Q_L(x)=q$ is independent of $x$ we have

$$q=\frac{1}{1+\exp(\mathcal{F}a/T)}. \tag{18}$$

More generally we define an energy profile for the system $\{E_{-L},E_{-L+1},\ldots,E_i,\ldots\}$. The general detailed balance condition is then

$$\frac{Q_L(x)}{1-Q_L(x-1)}=\exp\left(-\frac{E_{x-1}-E_x}{T}\right). \tag{19}$$

Certain restrictions on the detailed balance condition for nonlinear forces are discussed in Sec. VI. The choice of the detailed balance condition means that for an *ensemble* of particles standard Boltzmann-Gibbs statistics holds. Thus, for example, if we observe many independent particles, and look at their density profile in equilibrium, we will see a profile that is determined by Boltzmann equilibrium. On the other hand if we consider a trajectory of a single particle and from it find $\bar{p}_x$ we are not likely to find the value of $\bar{p}_x$ close to Boltzmann's probability, when $\alpha<1$. Thus ergodicity breaking is found on the level of a single particle. Note that there is an interesting transition between one-particle information and many-particle behavior; however, this is not the subject of our work [28].

## IV. FIRST PASSAGE TIMES

The problem of ergodicity breaking is related in this section to the problem of first passage times.

The process $\theta_x(t)$ is a two-state process, with state $x$ denoting a particle on lattice point $x$ and state $nx$ indicating that the particle is not on $x$. Obviously the waiting times in state $x$ are given by $\psi_x(\tau)=\psi(\tau)$. To obtain the PDF of waiting times in state $nx$, $\psi_{nx}(\tau)$, we must calculate the statistical properties of first passage times. After the particle leaves point $x$ it is located on either $x+1$ or $x-1$ with probabilities $Q_R(x)$ and $Q_L(x)$, respectively. Let $t_L$ denote the time it will take the particle to return to $x$ starting at point $x-1$, i.e., the first passage time from $x-1$ to $x$. Let $t_R$ be the first passage time to reach $x$ starting from $x+1$. Let $f_R(t_R)$ $[f_L(t_L)]$ be the PDF of the first passage time $t_R$ $(t_L)$, respectively. Then the PDF of times in state $nx$ is given by

$$\psi_{nx}(\tau)=Q_R(x)f_R(\tau)+Q_L(x)f_L(\tau). \tag{20}$$

In principle once the long time behavior of the PDFs of first passage times is obtained, we have $\psi_{nx}(\tau)$ and $\psi_x(\tau)$, and then we may use the formalism developed in Appendix A to obtain the PDF of the occupation fraction $\bar{p}_x$. We now investigate the first passage time PDFs for biased and unbiased

CTRWs, using an analytical approach. The reader not interested in mathematical details may skip to Sec. V.

### A. Relation between discrete time and continuous time RWs

For convenience we define a new lattice. We consider the CTRW in one dimension, on lattice points $x=0,1,2,\ldots,\tilde{L}$. Point $x=0$ is a "sticky" absorbing boundary, namely, once the particle reaches point $x=0$ it remains there forever. Point $\tilde{L}$ is a reflecting boundary, and initially at time $t=0$ the particle is on $x=1$. Let $S_{\mathrm{CT}}(t)$ be the survival probability of the CTRW particle, where the subscript "CT" indicates CTRW. The object of interest is the PDF of first passage times $f_{\mathrm{CT}}(t)$, which is minus the time derivative of $S_{\mathrm{CT}}(t)$. The solution is possible due to an important relation [37] between the CTRW first passage time problem and that of discrete time random walks. In Ref. [37] the first passage time problem with CTRW dynamics with exponential waiting times was considered.

The point 0 of the new lattice is point $x$ in the original problem and $\tilde{L}=L-x$ and similarly for the other $\tilde{L}-1$ points of the new lattice. Hence the calculation of the first passage PDF on the new lattice $x=0,1,2,\ldots,\tilde{L}$ yields $f_R(t_R)$. With straightforward change of notation we may consider also $f_L(t_L)$.

Let $S_{\mathrm{CT}}(t)$ be the survival probability of the CTRW particle in the interval $x=1,\ldots,x=\tilde{L}$. Let $S_{\mathrm{dis}}(N)$ be the probability of survival after $N$ jump events, for a particle starting at $x=1$ (the subscript "dis" stands for discrete). Then

$$S_{\mathrm{CT}}(t)=\sum_{N=0}^{\infty}S_{\mathrm{dis}}(N)w(N,t) \tag{21}$$

where $w(N,t)$ is the probability for $N$ steps, within time $t$, in a CTRW process. In Laplace, $t\to u$ space it is easy to show using the convolution theorem of the Laplace transform that

$$\hat{w}(N,u)=\frac{1-\hat{\psi}(u)}{u}\hat{\psi}^N(u) \tag{22}$$

where $\hat{\psi}(u)$ is the Laplace transform of $\psi(\tau)$. In this work the discrete Laplace transform of an arbitrary function $G(N)$, also called the $z$ transform, is defined as

$$\tilde{G}(z)=\sum_{N=0}^{\infty}z^N G(N). \tag{23}$$

Using Eqs. (21) and (22) we find

$$\hat{S}_{\mathrm{CT}}(u)=\frac{1-\hat{\psi}(u)}{u}\tilde{S}_{\mathrm{dis}}[\hat{\psi}(u)]. \tag{24}$$

This equation establishes the relation between the discrete and continuous time problems.

Let $P_x(N)$ be the probability of occupying site $x$ after $N$ jumps and $P_x(0)=\delta_{x1}$. The master equation describing the discrete time problem is given by

$$P_0(N+1)=Q_L(1)P_1(N)+P_0(N)$$

(since the origin 0 is absorbing),

$$P_1(N+1)=Q_L(2)P_2(N),$$

$$P_2(N+1)=Q_R(1)P_1(N)+Q_L(3)P_3(N),$$

$$P_x(N+1)=Q_R(x-1)P_{x-1}(N)+Q_L(x+1)P_{x+1}(N),$$

$$P_{\tilde{L}-1}(N+1)=P_{\tilde{L}}(N)+Q_R(\tilde{L}-2)P_{\tilde{L}-2}(N),$$

$$P_{\tilde{L}}(N+1)=Q_R(\tilde{L}-1)P_{\tilde{L}-1}(N). \tag{25}$$

The probability to be absorbed for the first time at $x=0$ after $N+1$ jumps (the discrete time) is

$$F_{\mathrm{dis}}(N+1)=Q_L(1)P_1(N). \tag{26}$$

The discrete survival probability is given by

$$S_{\mathrm{dis}}(N)=1-P_0(N). \tag{27}$$

Using Eq. (25)

$$S_{\mathrm{dis}}(N)=1-[Q_L(1)P_1(N-1)+P_0(N-1)], \tag{28}$$

and from Eq. (26)

$$S_{\mathrm{dis}}(N)=1-[F_{\mathrm{dis}}(N)+P_0(N-1)]. \tag{29}$$

Using Eq. (27) we have

$$S_{\mathrm{dis}}(N)-S_{\mathrm{dis}}(N-1)=-F_{\mathrm{dis}}(N), \tag{30}$$

which simply means that the change in the survival probability at step $N$ is equal to minus the probability of first passage. The $z$ transform [defined in Eq. (23)] of Eq. (30) yields

$$\tilde{S}_{\mathrm{dis}}(z)=\frac{1-\tilde{F}_{\mathrm{dis}}(z)}{1-z}. \tag{31}$$

Hence from Eq. (24)

$$\hat{S}_{\mathrm{CT}}(u)=\frac{1}{u}\{1-\tilde{F}_{\mathrm{dis}}[\hat{\psi}(u)]\}. \tag{32}$$

Let $f_{\mathrm{CT}}(t)$ be the first passage time PDF of the CTRW problem. As usual

$$f_{\mathrm{CT}}(t)=-\frac{d}{dt}S_{\mathrm{CT}}(t), \tag{33}$$

which is the continuous pair of Eq. (30). If the random walker always returns to the origin, then $\int_0^\infty f_{\mathrm{CT}}(t)dt=1$, and Eq. (33) yields

$$\hat{f}_{\mathrm{CT}}(u)=-u\hat{S}_{\mathrm{CT}}(u)+1 \tag{34}$$

and using Eq. (32)

$$\hat{f}_{\mathrm{CT}}(u)=\tilde{F}_{\mathrm{dis}}[\hat{\psi}(u)]. \tag{35}$$

This is the most important equation of this subsection. At least in some cases the solution of the discrete time first passage time problem in $z$ space is possible, and then we can transform the solution to Laplace $u$ space of the seemingly more difficult case of continuous time. Note that our assumption that the random walk is recurrent is valid only when the system size is finite, and $Q_L(x)>0$ for any $x$ except on the boundary.

### B. First passage time for unbiased and uniformly biased CTRW

We start with finding the first passage time distribution for the unbiased CTRW in Laplace space. For the unbiased random walk we have $Q_L(x)=Q_R(x)=1/2$, for $x\neq 0$, $x\neq \tilde{L}$. And as mentioned $x=0$ is the absorbing boundary condition, while $\tilde{L}$ is a reflecting wall. As shown we may consider the first passage time for the discrete time random walk Eq. (25) and then use the transformation Eq. (35) to obtain the corresponding CTRW first passage time. In Appendix B we solved the discrete time model to obtain

$$\tilde{F}_{\mathrm{dis}}(z)=\frac{z/2}{1-\frac{z^2}{4}\frac{B_+\Lambda_+^{\tilde{L}-3}+B_-\Lambda_-^{\tilde{L}-3}}{B_+\Lambda_+^{\tilde{L}-2}+\Lambda_-^{\tilde{L}-2}}}, \tag{36}$$

where

$$\Lambda_\pm=\frac{1\pm\sqrt{1-z^2}}{2}, \tag{37}$$

$$B_-=\frac{1-z^2/2-\Lambda_+}{\Lambda_--\Lambda_+}, \tag{38}$$

and $B_+=1-B_-$. Using the Laplace transform of the waiting time PDF Eq. (15) and Eqs. (35) and (36) we obtain the small $u$ behavior of the first passage time PDF,

$$\hat{f}_{\mathrm{CT}}(u)\sim 1-(2\tilde{L}-1)Au^\alpha+\cdots. \tag{39}$$

Equation (39) yields the Laplace transform of the first passage times of the unbiased CTRW with reflecting boundary condition on $\tilde{L}$, absorbing on the origin, and initial location of the particle on $x=1$.

We now find the first passage time distribution for the biased CTRW in Laplace space. Now the probability to jump left is $Q_L(x)=q$ and hence the probability to jump to the right is $Q_R(x)=1-q$, for $x\neq 0$, $x\neq\tilde{L}$. The two boundary conditions are the same as in the unbiased case. Like the unbiased case we treat the problem of the discrete time random walk (for details see Appendix B) and then use the transformation Eq. (35) to obtain the corresponding CTRW first passage time distribution.

In this case

$$\tilde{F}(z)=\frac{qz}{1-q(1-q)z^2\frac{B_+\lambda_+^{\tilde{L}-3}+B_-\lambda_-^{\tilde{L}-3}}{B_+\lambda_+^{\tilde{L}-2}+B_-\lambda_-^{\tilde{L}-2}}}, \tag{40}$$

where

$$\lambda_\pm(z)=\frac{1\pm\sqrt{1-4qz^2(1-q)}}{2}, \tag{41}$$

$B_++B_-=1$, and

$$B_+(z)=\frac{1-\lambda_--z^2(1-q)}{\lambda_+-\lambda_-}. \tag{42}$$

We use the relation Eq. (35) and insert in Eq. (40) the small $u$ behavior of the Laplace transform of the waiting time PDF Eq. (15). In the limit $u\to 0$ we find the Laplace transform of the PDF of the first passage time of the CTRW particle

$$\hat{f}_{\mathrm{CT}}(u)\sim 1-\frac{Au^\alpha}{2q-1}\left[1-2(1-q)\left(\frac{1-q}{q}\right)^{\tilde{L}-1}\right]+\cdots. \tag{43}$$

It can be easily seen that for $q=1$ or $L=1$, $\hat{f}_{\mathrm{CT}}(u)\sim 1-Au^\alpha$ as expected since then $\hat{f}_{\mathrm{CT}}(u)=\hat{\psi}(u)$. The second term on the right hand side of Eq. (43) will diverge when $q<1/2$ and $L\to\infty$, as expected for an infinite system, and for a random walker moving against the average drift. We see from Eq. (43) that the PDF of first passage times $f_{\mathrm{CT}}(t)\propto t^{-(1+\alpha)}$ in the limit of long times, when $\alpha<1$. In the limit $q\to 1/2$ the solution for the biased case Eq. (43) reduces to the unbiased solution Eq. (39).

## V. MAIN RESULTS

### A. Nonthermal random walks

First consider the unbiased one-dimensional CTRW on a lattice $x=-L,\ldots,L$. The PDF of the fraction of occupation time $\bar{p}_x=t_x/t$ on a lattice point $x$, excluding the boundary points, is obtained using Eqs. (20), (39), (A15), and (A18). The general idea of the proof is to note that $\hat{\psi}_x(u)=\hat{\psi}(u)$,

$$\hat{\psi}_{nx}(u)\sim 1-A(2L-1)u^\alpha, \tag{44}$$

for $u\to 0$ and hence using Appendix A we find

$$\lim_{t\to\infty} f(\bar{p}_x)=\delta_\alpha((2L-1)^{-1},\bar{p}_x), \tag{45}$$

where the $\delta_\alpha$ function was defined in Eq. (7). Equation (45) does not depend on the position $x$ of the observation point, reflecting the symmetry of the problem. From Eq. (45) we see that the amplitude ratio satisfies $\mathcal{R}_x=1/(2L-1)<1$ when $L>1$. This inequality means that we are less likely to find the particle on the particular lattice point $x$ under observation (state $x$), if compared with the probability of finding the particle on any of the other lattice points (state $nx$).

For the biased random walk, when the probability of jumping left is $q$, we consider the PDF of fraction of time $\bar{p}_x$ on a lattice point $x$. Now clearly different locations have different distributions of the fraction of occupation time, reflecting the fact that the system is biased. The Laplace transform of the sojourn times on $x$ is simply

$$\hat{\psi}_x(u)=\hat{\psi}(u)\sim 1-Au^\alpha \tag{46}$$

and using Eq. (20) the sojourn times in all other states ($nx$) is

$$\hat{\psi}_{nx}(u) = (1-q)\hat{f}_R(L-x,u) + q f_L(x+L,u). \quad (47)$$

Here $\hat{f}_R(L-x,u)$ is the Laplace transform of the first passage time PDF, for a system of size $L-x+1$, obtained in Eq. (43); similarly for $\hat{f}_L(L-x,u)$, but now replace $q$ with $1-q$ in Eq. (43). Using Eq. (47) we find the small $u$ behavior of $\hat{\psi}_{nx}(u)$, and then using Eq. (A15) we find

$$\hat{\psi}_{nx}(u) \sim 1 - \frac{A}{\mathcal{R}_x}u^{\alpha} + \cdots, \quad (48)$$

$$\mathcal{R}_x = \left\{ \frac{2}{2q-1}\left[ q^2\left(\frac{q}{1-q}\right)^{L+x-1} - (1-q)^2\left(\frac{1-q}{q}\right)^{L-x-1}\right] - 1\right\}^{-1}. \quad (49)$$

Equations (48) and (49) and the results obtained in Appendix A indicate that the PDF of fraction of occupation time is

$$f(\bar{p}_x) = \delta_{\alpha}(\mathcal{R}_x, \bar{p}_x) \quad (50)$$

with $\mathcal{R}_x$ given in Eq. (49).

As expected the PDF of the fraction of occupation time for the biased CTRW depends on the location of the site under consideration. As usual if $q<1/2$ the particle prefers to stick to the right wall. In our case this behavior implies that if $q<1/2$ and $x\simeq -L$ ($L$ is large) then $\mathcal{R}_x \to 0$, which means that the lattice point $x$ is never occupied, as expected.

### B. Equilibrium–ergodicity breaking relationship

Equations (45), (50), and (49) describe the nonergodic properties of the CTRW for biased and unbiased cases. We will now consider the relation of the problem of nonergodicity with the equilibrium of the process. Consider an ensemble of independent random walkers performing the CTRW process in the finite domain. After a long period of time an equilibrium will be reached, for which the density of particles is found in a steady state profile. Such an equilibrium is obtained after each individual member of the ensemble made many jump events (one can easily prove that such an equilibrium is reached). We denote the probability of finding such a random walker on point $x$ with $P_x^{\mathrm{eq}}$. It is straightforward to obtain $P_x^{\mathrm{eq}}$, though some care must be used when we take into consideration the boundary conditions of the problem. In equilibrium

$$P_x^{\mathrm{eq}} = \frac{\left(\frac{1-q}{q}\right)^x}{Z} \quad (51)$$

and on the boundaries

$$P_L^{\mathrm{eq}} = \frac{(1-q)\left(\frac{1-q}{q}\right)^{L-1}}{Z},$$

$$P_{-L}^{\mathrm{eq}} = \frac{q\left(\frac{1-q}{q}\right)^{-L+1}}{Z}. \quad (52)$$

$Z$ is then obtained from the normalization condition $\Sigma_{x=-L}^{L} P_x^{\mathrm{eq}}=1$. Here $Z$ is not necessarily related to Boltzmann-Gibbs statistics.

Using the equilibrium properties of the system, after a short calculation of the normalization constant and some algebra, we find that Eqs. (45), (50), and (49) may be written in a more elegant form as

$$f(\bar{p}_x) = \delta_{\alpha}\left(\frac{P_x^{\mathrm{eq}}}{1-P_x^{\mathrm{eq}}}, \bar{p}_x\right). \quad (53)$$

Note that $P_x^{\mathrm{eq}}$ yields the equilibrium properties of many noninteracting random walkers, or the density profile of a large number of particles. Hence the single-particle nonergodicity is related to statistical properties of the equilibrium of many particles. The fact that we find such a relation should be anticipated, since if we average $\bar{p}_x$, namely, consider $\langle \bar{p}_x\rangle = \int_0^{\infty} \bar{p}_x f(\bar{p}_x) d\bar{p}_x$, we must obtain $P_x^{\mathrm{eq}}$; hence $f(\bar{p}_x)$ must be clearly related to $P_x^{\mathrm{eq}}$. And the requirement $\langle \bar{p}_x\rangle = P_x^{\mathrm{eq}}$ implies that $\mathcal{R}_x = P_x^{\mathrm{eq}}/(1-P_x^{\mathrm{eq}})$ as we indeed found (and similar to our discussion in Sec. II). For the unbiased case, $q=1/2$ we have $P_x^{\mathrm{eq}}=1/2L$, which leads to Eq. (45). Note that the equilibrium population on the boundaries $x=\pm L$ is half the value of that found on $x\neq \pm L$, and hence $Z=2L$ even though we have $2L+1$ lattice points.

### C. Thermal unbiased and uniformly biased random walks

If the CTRW particle is interacting with a thermal heat bath, we can relate the nonergodicity to Boltzmann-Gibbs statistics. The equilibrium probability of occupying a given cell in the ensemble sense is given by the Boltzmann probability if the system is in thermal equilibrium. Hence, rewriting Eq. (53),

$$f(\bar{p}_x) = \delta_{\alpha}\left(\frac{P_x^B}{1-P_x^B}, \bar{p}_x\right). \quad (54)$$

The factor $P_x^B/(1-P_x^B)$ means that with probability $P_x^B$ the particle is in state $x$, and with probability $1-P_x^B$ the particle is in state $nx$, i.e., the rest of the system (here we mean probability in the ensemble sense). For the free particle we recall that the Boltzmann probability of occupying a lattice point is simply

$$P_x^B = \frac{1}{Z} \quad (55)$$

and as mentioned $Z=2L$ is the normalization condition, or the partition function of the problem.

For a biased CTRW when detailed balance condition Eq. (18) holds, the equilibrium probability is

$$P_x^B = \frac{\exp(-\mathcal{F}ax/T)}{Z}, \quad (56)$$

where $x$ is the lattice site under observation, $\mathcal{F}$ is the constant force, and $a$ is the lattice spacing. Here the partition function is

$$Z = \frac{2\left[q^2\left(\frac{1-q}{q}\right)^{-L+1} - (1-q)\left(\frac{1-q}{q}\right)^{L-1}\right]}{2q-1} \quad (57)$$

which is easily verified once proper reflecting boundary conditions are applied, and using Eq. (18).

## VI. COARSE GRAINING AND NUMERICAL SIMULATION

So far we considered the occupation time on one lattice cell. We now consider the more general case where we observe occupation times on a finite number of cells. Such coarse graining is important for systems where many cells exist, since then the statistics of the occupation time of one cell is poor, unless the measurement time is extremely long.

We also wish to investigate the generality of our results for a nonlinear force field. In Ref. [35] we considered the harmonic potential as an example while here we consider a double-well potential (see details below).

Consider a CTRW in a potential field $V(x)$. The system is divided into two regions; we observe the occupation times in a connected interval $X$ given by $x_a < x < x_b$. The remaining part of the space is denoted $nX$ (not $X$). We assume a bounded motion in the potential field and that equilibrium state is obtained for the ensemble. The arguments in Sec. II may be used to generalize our results. Since waiting time PDFs of a CTRW follow a power law behavior, we expect $\psi_X \sim A_X t^{-(1+\alpha)}/\Gamma(-\alpha)$ and $\psi_{nX} \sim A_{nX} t^{-(1+\alpha)}/\Gamma(-\alpha)$. This is a generalization of Eqs. (7) and (8), where we considered a single state. Following the results of Appendix A and the arguments given in Sec. II the PDF of the fraction of occupation time in $X$ is given by $\delta_\alpha(\mathcal{R}_X, \bar{p}_X)$, where $\mathcal{R}_X = p_X^{\text{eq}}/(1-p_X^{\text{eq}})$. If the system is in thermal equilibrium

$$p_X^{\text{eq}} = \sum_{x \in X} p^B(x).$$

### A. Numerical demonstration

To demonstrate this behavior we performed a numerical simulation of the CTRW process in a potential field of the form $V(x) = x^4 - x^2$. This potential has two minima at $x = \pm 1/\sqrt{2}$, and the barrier between them is of height 0.25. We (i) used the condition of detailed balance Eq. (19), and (ii) at the symmetry point of the potential $x=0$, set $Q_L(0) = Q_R(0) = 1/2$. These two conditions yield $Q_L(x)$. Note that the detailed balance condition as presented in Eq. (19) is valid only in the continuum limit. For example, using the conditions mentioned above and discretization of the lattice in units equal to 1 yields values which are greater than 1 for $Q_L(x)$. In order to use the detailed balance condition the discretization should be made with small values of $\Delta x$ such that in all the accessible region of space (where the Boltzmann probability is finite) $0 < Q_L(x) < 1$. In the simulation presented here we used $\Delta x = 0.05$ and the temperature was of the order of the barrier between the minima in order for the CTRW particle to visit all the accessible region within reasonable time. The random waiting times were generated according to the normalized power law waiting time PDF $\psi(\tau) = \alpha \tau^{-(1+\alpha)}$, for $\tau > 1$.

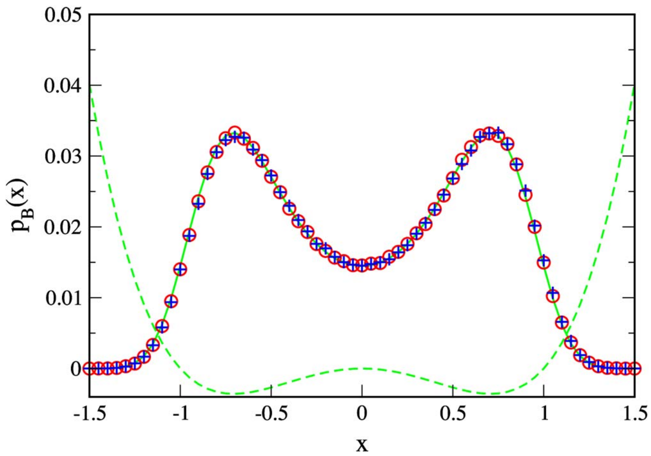


FIG. 1. (Color online) Boltzmann's equilibrium for an ensemble of CTRW particles in a double-well potential field, and fixed temperature. In simulations (crosses and circles) the CTRW with $\alpha = 0.5$ and 0.7 is considered. The figure illustrates that for an ensemble of particles, standard equilibrium is obtained; ergodicity breaking is found only when long time averages of single particle trajectories are analyzed. The scaled potential (dashed curve) is the double-well potential field, and the solid curve is the Boltzmann equilibrium distribution. To construct the histogram we measured the position of $N = 10^6$ particles, after each is evolved for a time $t = 10^8$. The temperature is $T = 0.3$. Note that all quantities are dimensionless.

We first checked that the Boltzmann equilibrium is reached for an ensemble of particles. In these simulations we build histograms of the position of $N = 10^6$ particles, after each particle evolves for a time $t = 10^8$ (several values of $\alpha$ were used). In Fig. 1 we find good agreement between our simulations and Boltzmann statistics when many particles are considered. The figure illustrates that an observer of a large number of particles cannot detect ergodicity breaking, and the single-particle limit is essential for our discussion. We then consider one trajectory at a time. We obtain from the simulations the total time spent by the particle on lattice region $x_a < x < x_b$. This time is $t_X$ and the fraction of occupation time $\bar{p}_X = t_X/t$. In the ergodic phase and long time limit $\bar{p}_X$ will approach the value predicted by Boltzmann statistics. While in the nonergodic phase we test if our prediction Eqs. (7) and (8) hold.

In Figs. 2 and 3 we consider two values of $\alpha$, $\alpha = 0.5, 0.7$, and fix the temperature $T$. All figures show an excellent agreement between our theoretical predictions Eqs. (7) and (8) and numerical simulations. It is more important, however, to understand the meaning of the figures.

For $\alpha \ll 1$ we expect that the particle will get stuck on one lattice point during a very long period, which is of the order

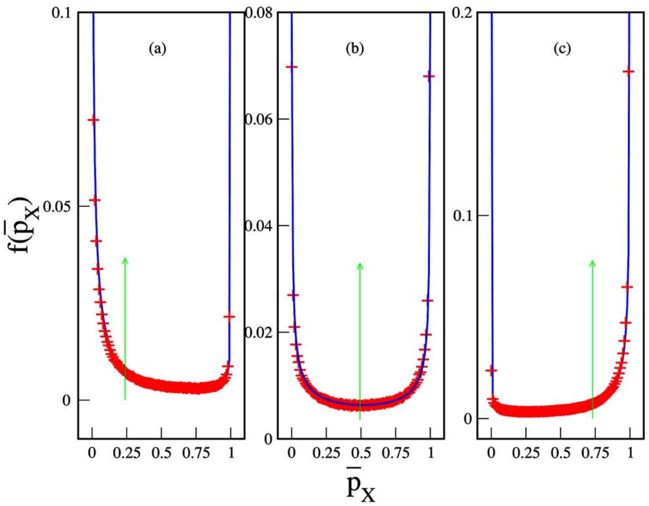


FIG. 2. (Color online) The PDF of occupation times $\bar{p}_X = t_X/t$ where $t_X$ is the total time spent on a finite domain, and $t$ is the measurement time. Here we used $\alpha=0.5$. The domains we considered are $x>0.6$, $x>0$, and $x>-0.6$, corresponding to (a), (b), and (c) in the figure. For an ergodic process satisfying detailed balance, the PDF $f(\bar{p}_X)$ would be a $\delta$ function centered around the value predicted by Boltzmann which is given by the arrows. In a given numerical experiment, it is unlikely to obtain the value of $\bar{p}_X$ predicted by Boltzmann, though Boltzmann statistics does yield the average of $\bar{p}_X$ over many particles. To construct histograms we used $10^6$ trajectories, measurement time $t=10^8$, and temperature $T=0.3$. The solid curves correspond to the analytical formula Eqs. (7) and (8) used without any fitting parameters, and the pluses show the simulation results.

of the measurement time $t$. This trapping point can be either within the region of observation or outside this region. In these cases we expect to find $\bar{p}_X \simeq 1$ or $\bar{p}_X \simeq 0$, respectively. Hence the PDF of $\bar{p}_X$ has a U shape. This case exhibits large

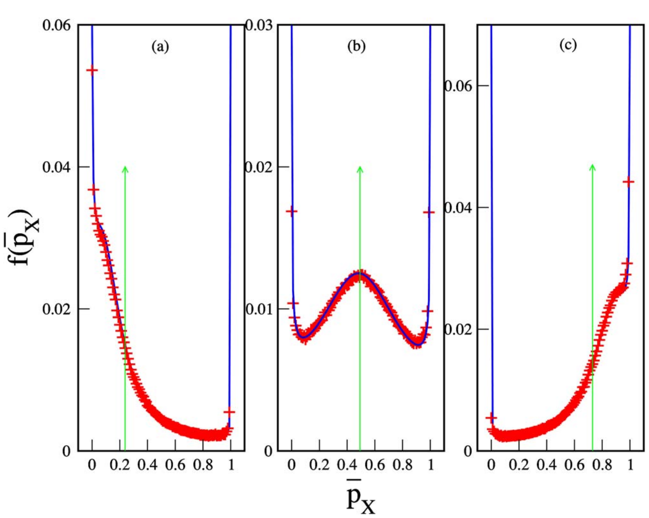


FIG. 3. (Color online) Same as Fig. 2 but now $\alpha=0.7$. In this case the probability of obtaining the ergodic value (arrow) in a single measurement is higher than for the case $\alpha=0.5$ (see Fig. 2). Note that the larger the region we observe the more the PDF is tilted toward 1 since it is more probable that the particle is within the region of observation.

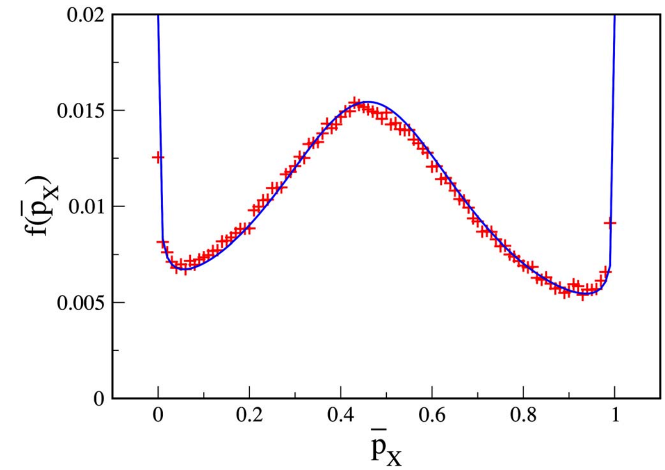


FIG. 4. (Color online) The PDF of the fraction of occupation time in the disconnected regions $x<-0.6$ and $x>0.6$, for $\alpha=0.75$ and temperature $T=0.3$. The theoretical line (solid curve) is determined by summing the equilibrium probabilities of occupying all the cells within the regions, and substituting it into the constant $\mathcal{R}_X$. The good agreement between the theory and the numerical simulation (pluses) illustrates that our theory is not limited to connected regions.

deviations from ergodic behavior, in the sense that we have a very small probability for finding the occupation fraction close to the value predicted based on Boltzmann's ergodic theory. As shown in Fig. 2 such U-shape behavior is found for the case $\alpha=0.5$. We also plotted the prediction made using the ergodic assumption (the arrows in the figure) to demonstrate the fact that a measurement is not likely to yield the average, which is located on $P_X^B$.

When we increase $\alpha$ we anticipate a more ergodic behavior, in particular in the limit $\alpha \to 1$. An ergodic behavior means that the PDF of the occupation fraction $\bar{p}_X$ is centered on the Boltzmann's probability. In Fig. 3 where $\alpha=0.7$ we start seeing a peak in the PDF of $\bar{p}_X$ centered in the vicinity of the ensemble average value. Note, however, that the PDF $f(\bar{p}_X)$ still attains its maxima at $\bar{p}_X=0$ and $\bar{p}_X=1$. Hence we find a weaker nonergodic behavior if compared with the case $\alpha=0.5$.

In all previous simulations we considered only connected regions of space, but our theory is not limited to that case. In order to illustrate that the occupation time of unconnected regions is also given by Eqs. (7) and (8), a simulation in which the occupation time of the regions $x>0.6$ and $x<-0.6$ was performed. The CTRW considered is the same as the one considered in the previous simulations, i.e., a CTRW in a double-well potential. The temperature is $T=0.3$, and $\alpha=0.75$. In Fig. 4 it is shown that there is a good agreement between the theoretical predicted PDF (solid curve) and the simulation results (pluses), without fitting.

In Figure 5 we consider the occupation time of the regions near the double-well potential minima, $-0.9<x<-0.5$ and $0.5<x<0.9$. We fix $\alpha=0.8$ and vary the temperature, using Eqs. (7) and (8). At temperature $T\simeq 0.4$ (solid line) we see that the PDF of $\bar{p}_X$ is symmetric. This happens when $P_X^B=1/2$, namely, for a case that there is probability one-half of occupying the observation region, and probability one-

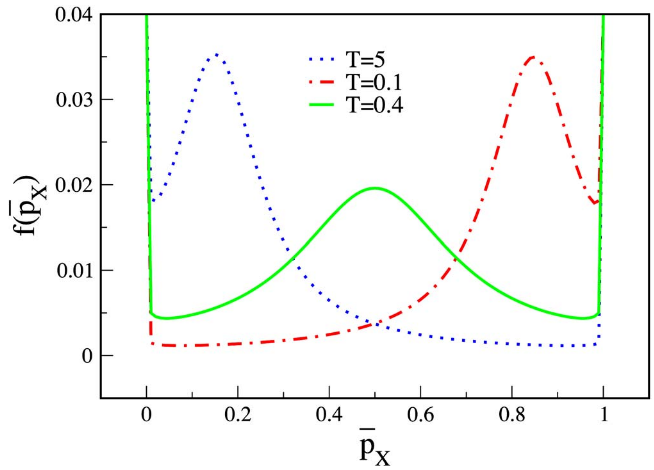


FIG. 5. (Color online) The PDF of the fraction of occupation time of the regions near the potential minima, $-0.9<x<-0.5$ and $0.5<x<0.9$, for $\alpha=0.8$ when temperature $T$ is varied. When the temperature is such that the probability of occupying the minima regions is equal to $1/2$ the PDF is symmetric; for our parameters this temperature is $T_s=0.4$. For $T\ll T_s$ the particle tends to be located all the time in the vicinity of the minima; hence for $T=0.1<T_s$ (dot-dashed curve) the PDF has more weight on values $\bar{p}_X>1/2$. For $T\gg T_s$ the particle is hardly found in the minima regions; hence for $T=5>T_s$ (dotted curve) the PDF has more weight on values $\bar{p}_X<1/2$.

half to be out of this interval. When the temperature is very low, we expect to find the particle in the ground state, namely, near the minima. Hence the PDF of $\bar{p}_X$ is tilted toward $\bar{p}_X\simeq 1$ when temperature is lowered (see Fig. 5 when $T=0.1$). In contrast, when the temperature is high, we expect the probability of occupying the observation region to be reduced (as usual entropy wins at high temperature). And indeed we observe that when $T=5$ the PDF of $\bar{p}_X$ is more tilted toward the left, namely, to $\bar{p}_X\simeq 0$.

### B. Validity of main Eqs. (7) and (8)

Our numerical work as well as our analytical solutions for the biased and nonbiased CTRWs show the validity of Eqs. (7) and (8). What happens for more general types of potential fields? Can we claim that Eq. (7) has a wider applicability? Consider the CTRW with potential profile $\{\ldots,E_x,\ldots\}$, with the dynamics satisfying the detailed balance condition (i.e., the mentioned continuum limit condition holds). We claim that if for $\psi(\tau)$ with finite moments, the system is ergodic, then for the same energy profile but when the waiting time PDF has a long tail, Eqs. (7) and (8) hold. Our reasoning is that we can think of $\alpha$ as a control parameter, which we can vary between $0<\alpha\leq 1$. And since for the case $\alpha=1$ we have $\mathcal{R}_x=P_x^B/(1-P_x^B)$ also for $0<\alpha<1$ this relation must hold (since $P_B$ does not depend on $\alpha$). Further, the transformation in Laplace space $u\langle\tau\rangle\to Au^{\alpha}$ in the small $u$ limit of the waiting time PDF, seems to indicate that the behavior we found has a general validity.

A way to understand the ergodicity breaking laws Eqs. (7) and (8) is to consider the number of times $n_x$ the particle visits lattice point $x$ during a long measurement time. In that case the particle visits $x$ many times, and we assume that the fraction of number of visits satisfies

$$\frac{n_x}{n}=\exp\left(-\frac{V(x)}{T}\right), \tag{58}$$

where $n$ is the total number of jumps made by the particle. If the first moment of the waiting time distribution is finite, we have $t_x/t=n_x/n$, since the average time spent on $x$ is $n_x$ times the mean waiting time. When the average waiting time is infinite, $\alpha<1$, one can show that the PDF of $t_x/t$ is given by Eq. (7) if condition Eq. (58) holds. Equation (58) should be tested in more detail, for example using numerical simulations.

## VII. SUMMARY AND DISCUSSION

We obtained the nonergodic properties of biased and unbiased continuous time random walks. In particular the distribution of the occupation fraction $\bar{p}_x$ was found. Our results are valid for both thermal and nonthermal cases. In both cases the nonergodicity is described using the $\delta_\alpha(\mathcal{R}_x,\bar{p}_x)$ PDF Eq. (7), where $\alpha$ is the anomalous diffusion exponent $\langle x^2\rangle\sim t^{\alpha}$. For both thermal and nonthermal random walks the parameter $\mathcal{R}_x$ is related to the ensemble averaged equilibrium properties of the system, Eqs. (53) and (54), respectively. If the system is in the vicinity of thermal equilibrium, the equilibrium of the system is the Boltzmann-Gibbs equilibrium, in the ensemble sense. Such behavior is found when detailed balance conditions are applied properly. In this case the characterization of the nonergodic properties of the occupation times is related to the partition function and temperature. The nonergodicity manifests itself when the time average of single-particle observables is considered; in particular, the occupation time in a given energy state or on a particular lattice point. Hence the nonergodicity might reveal itself in single-particle experiments.

Models and systems describing anomalous diffusion are widespread. In most cases ensemble average properties of such processes are investigated, both in theory and in experiment. For single-particle experiments, where the problem of ensemble averaging is removed, we may either (i) reconstruct the ensemble averages, by repeating the single-molecule experiment many times, or (ii) investigate the ergodic properties of the system, by considering the fraction of occupation time in a particular state and obtaining its distribution. It is the second type of measurement that is considered here, which yields insight into single-particle properties that differ from the standard ensemble measurement, provided that a nonergodic phase is investigated. While the theory of anomalous diffusion processes is now vast, the nonergodic properties of such processes are still not well understood. Investigation of this topic beyond the CTRW approach is left for future work.

## ACKNOWLEDGMENTS

E.B. was supported by the National Science Foundation Award No. CHE-0344930, the Center for Complexity Science Jerusalem, and the Israel Science Foundation. E.B. thanks J. P. Bouchaud, A. Comtet, S. N. Majumdar, and G. Margolin for comments and discussion.

### APPENDIX A: THE $\delta_\alpha(\mathcal{R}_X,\bar{P}_X)$ FUNCTION

In this appendix we rederive the limit theorem Eq. (7). While this goal was accomplished by Lamperti a long time ago [36], we believe that it is worth rederiving this result using a method similar to what is used today in the statistical physics community. In particular Godreche and Luck [25] in their analysis of power law renewal processes rederived the symmetric $\delta_\alpha(\mathcal{R}_x,\bar{p}_x)$ with $\mathcal{R}_x=1$ for a two-state process, where sojourn times in both states are identically distributed. Here, we consider the case of interest in this paper, where the sojourn times in the two states are not statistically identical. We also derive an exact distribution for the occupation fraction of a two-state process in Laplace space Eqs. (A12) and (A13).

Consider a system evolving between two states + and − corresponding to states $x$ and $nx$, respectively. Let $\theta(t)=1$ when the system is in state +; otherwise $\theta(t)=0$ and the system is in state −. A schematic diagram of $\theta(t)$ is shown in Fig. 6. Let $\{t_i\}$ denote dots on the time axis at which transition events between state + to state − or vice versa occur. Let $\{\tau_i\}$ be sojourn times in either state + or state −. If the process starts with state +, then $\tau_i$ is a + state if $i$ is odd. We also denote the total number of jumps in the measurement time interval $(0,t)$ as $n$. We assume that the sojourn times are independent identically distributed random variables. The PDF of sojourn times is $\psi_+(\tau)$ and $\psi_-(\tau)$ for states + and −, respectively. Such a simple process is called a two-state renewal process. As usual it is convenient to analyze such a stochastic process using the Laplace transforms

$$\hat{\psi}_\pm(s)=\int_0^\infty e^{-s\tau}\psi_\pm(\tau)d\tau. \tag{A1}$$

While we will consider general properties of the stochastic process, we will eventually focus on two main cases. First

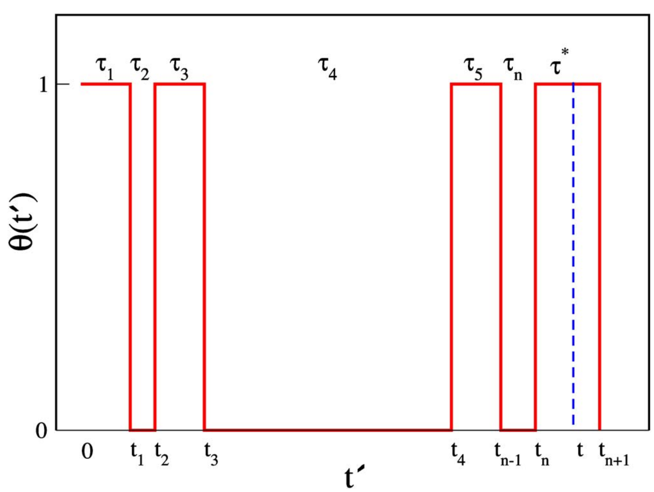


FIG. 6. (Color online) A schematic diagram of the two-state process.

consider the case where all moments of $\psi_\pm(\tau)$ are finite, e.g., exponential PDFs belong to this category. Then the following small $s$ expansion holds:

$$\hat{\psi}_\pm(s)\sim 1-s\langle\tau_\pm\rangle+\cdots. \tag{A2}$$

Here $\langle\tau_\pm\rangle$ are the mean sojourn times in states ±. A second generic case is

$$\hat{\psi}_\pm(s)\sim 1-s^\alpha A_\pm+\cdots \tag{A3}$$

with $0<\alpha<1$. For example the one-sided Lévy PDF $\hat{\psi}_\pm(s)=\exp(-A_\pm s^\alpha)$ belongs to this class. In the time domain these PDFs behave like

$$\psi_\pm(t)\sim\frac{A_\pm}{|\Gamma(-\alpha)|t^{1+\alpha}} \tag{A4}$$

when $t$ is large; namely, for this family of PDFs the average waiting times in both states diverge.

Let $t_+$ be the total time spent in state +, within the time period $(0,t)$. Then the occupation fraction in + is

$$\bar{p}_+=\frac{t_+}{t}=\frac{\int_0^t\theta(t')dt'}{t}. \tag{A5}$$

We now consider statistical properties of $t_+$ focusing on the scaling limit $t\to\infty$. Let $f^+_{t,n}(t_+)$ be the PDF of $t_+$ where $n$ renewal (i.e., jump) events occurred in the time interval $(0,t)$, and the start of the process is in state +.

Consider the case of $n$ odd, $n=2k+1$ with $k=0,1,\ldots$ . Then since we start with state +, $t_+=\sum_{i=1,\text{odd}}^n\tau_i$ where the summation is only over odd $i$'s. Also we have $t_n<t<t_{n+1}$, where $t_{n+1}$ is the occurrence time of a renewal event which occurs after the end of the measurement (see Fig. 6). Hence

$$f^+_{t,n}(t_+)=\left\langle\delta\left(t_+-\sum_{i=1,\text{odd}}^n\tau_i\right)I(t_n\le t\le t_{n+1})\right\rangle, \tag{A6}$$

where $\delta(x)$ is the Dirac delta function, and

$$I(t_n\le t\le t_{n+1})=\begin{cases}1 & \text{If condition in parentheses is true,}\\ 0 & \text{otherwise.}\end{cases} \tag{A7}$$

In Eq. (A6) $\langle\cdots\rangle$ denotes an average over the stochastic process, soon to be specified in more detail. Later we will consider the case $n$ even, and then sum over $n$ to obtain the PDF of $t_+$.

Let $s$ be the Laplace pair of $t$, and $u$ of $t_+$. It is convenient to consider the double Laplace transform $\hat{f}^+_{n,s}(u)$ of $f^+_{n,t}(t_+)$:

$$\hat{f}^{+}_{n,s}(u) = \int_0^\infty e^{-t_+u}\int_0^\infty e^{-st}f^{+}_{n,t}(t_+)dt_+dt$$
$$= \left\langle \int_0^\infty \int_0^\infty e^{-ut_+-st}\delta\left(t_+ - \sum_{i=1,\text{odd}}^{n}\tau_i\right) \times I(t_n \leqslant t \leqslant t_{n+1})dt_+dt \right\rangle$$
$$= \left\langle \frac{e^{-st_n}-e^{-st_{n+1}}}{s}\exp\left(-u\sum_{i=1,\text{odd}}^{n}\tau_i\right)\right\rangle, \quad \text{(A8)}$$

where we made use of Eq. (A6). Now we may consider the average $\langle\cdots\rangle$, using $t_n=\sum_{i=1,\text{odd}}^{n}\tau_i+\sum_{i=2,\text{even}}^{n-1}\tau_i$ and similarly for $t_{n+1}$. Recalling that $\tau_i$ with odd (even) $i$ are + (−) states, respectively, we find after averaging over the $\{\tau_i\}$'s

$$\hat{f}^{+}_{n,s}(u) = \hat{\psi}_+^{k+1}(s+u)\hat{\psi}_-^{k}(s)\frac{1-\hat{\psi}_-(s)}{s} \quad \text{(A9)}$$

where $n=2k+1$. For even $n$ such that $n=2k$, $k=1,2,\ldots$, we obtain

$$\hat{f}^{+}_{n,s}(u) = \hat{\psi}_+^{k}(s+u)\hat{\psi}_-^{k}(s)\frac{1-\hat{\psi}_+(s+u)}{s+u}. \quad \text{(A10)}$$

Note that for even $n$ the last + interval falls on $t$, hence we must investigate the statistical properties of $\tau^*=t-t_n$, the time difference between the end of the measurement and the last jump in the sequence (see Fig. 6). We are ready to obtain the double Laplace transform of $f_t^+(t_+)$, i.e., the PDF of $t_+$ when the process starts in the + state,

$$\hat{f}^{+}_{s}(u) = \sum_{k=0}^{\infty}[\hat{f}^{+}_{2k,s}(u)+\hat{f}^{+}_{2k+1,s}(u)]. \quad \text{(A11)}$$

Using Eqs. (A9)–(A11) we obtain the exact solution to the problem in Laplace $s$, $u$ space:

$$\hat{f}^{+}_{s}(u) = \left(\frac{1-\hat{\psi}_+(s+u)}{s+u}+\hat{\psi}_+(s+u)\frac{1-\hat{\psi}_-(s)}{s}\right) \times \frac{1}{1-\hat{\psi}_+(s+u)\hat{\psi}_-(s)}. \quad \text{(A12)}$$

It is easy to check the normalization condition $f_s^+(u=0)=1/s$ provided of course that $\psi_\pm(\tau)$ are normalized PDFs. In a similar way one can show that if we start the process in state − the double Laplace transform of the PDF of $t_+$ denoted with $f_t^-(t_+)$ is

$$\hat{f}^{-}_{s}(u) = \left(\hat{\psi}_-(s)\frac{1-\hat{\psi}_+(s+u)}{s+u}+\frac{1-\hat{\psi}_-(s)}{s}\right)\frac{1}{1-\hat{\psi}_+(s+u)\hat{\psi}_-(s)}. \quad \text{(A13)}$$

Equations (A12) and (A13) yield in principle the exact expression for the occupation fraction, which might be useful in determining the preasymptotic behavior, for example, using numerical inverse Laplace transform.

For the generic case Eq. (A3), in the limit of $s\to 0$ and $u\to 0$, their ratio remaining arbitrary, Eqs. (A12) and (A13) yield

$$\hat{f}^{\pm}_{s}(u) \sim \frac{\mathcal{R}(s+u)^{\alpha-1}+s^{\alpha-1}}{\mathcal{R}(s+u)^{\alpha}+s^{\alpha}} \quad \text{(A14)}$$

with

$$\mathcal{R} = \frac{A_+}{A_-}. \quad \text{(A15)}$$

The amplitude ratio $\mathcal{R}$ determines the degree of symmetry in the problem. Note that in this scaling limit the initial state of the process, i.e., the process being in state + or − at the initial time, is not important.

The small $(s,u)$ limit considered in Eq. (A14) corresponds to large measurement time $t$ and the occupation time $t_+$ limit. We invert Eq. (A4) using a method given in Ref. [25]. The method states that if in the limit $s,u\to 0$ a double Laplace transform behaves like

$$\hat{f}_s(u) = \frac{1}{s}g\left(\frac{s}{u}\right) \quad \text{(A16)}$$

then the PDF of the scaled variable $\bar{p}_+=t_+/t$ is in the long time $t$ limit

$$f(\bar{p}_+) = -\frac{1}{\pi x}\lim_{\epsilon\to 0}\text{Im}\left[\left. g\left(-\frac{1}{x+i\epsilon}\right)\right|_{x=\bar{p}_+}\right]. \quad \text{(A17)}$$

Using Eq. (A14) we find the PDF of the fraction of occupation time $\bar{p}_+=t_+/t$,

$$f(\bar{p}_+) = \delta_\alpha(\mathcal{R},\bar{p}_+) = \frac{\sin\pi\alpha}{\pi}\frac{\mathcal{R}\bar{p}_+^{\alpha-1}(1-\bar{p}_+)^{\alpha-1}}{\mathcal{R}^2(1-\bar{p}_+)^{2\alpha}+\bar{p}_+^{2\alpha}+2\mathcal{R}(1-\bar{p}_+)^{\alpha}\bar{p}_+^{\alpha}\cos\pi\alpha}. \quad \text{(A18)}$$

The PDF is normalized according to $\int_0^1 f(\bar{p}_+)d\bar{p}_+=1$, it is valid only in the long time $t$ limit, and is independent of it. In this sense an equilibrium is obtained. In particular when $A_+=A_-$ and $\alpha=1/2$ we find the arcsine distribution. It is easy to show that the average is

$$\langle\bar{p}_+\rangle = \frac{\langle t_+\rangle}{t} = \frac{A_+}{A_++A_-}. \quad \text{(A19)}$$

In the limit $\alpha\to 1$ we obtain

$$f(\bar{p}_+) = \delta\left(\bar{p}_+ - \frac{\langle\tau\rangle_+}{\langle\tau\rangle_+ + \langle\tau\rangle_-}\right), \tag{A20}$$

where $\langle\tau\rangle_\pm$ are the average waiting times when the waiting time PDFs have finite moments. We identify this behavior with an ergodic behavior, since according to Eq. (A5) $\bar{p}_+$ is a time average of $\theta(t)$, which is equal to the ensemble average value when moments of $\psi_\pm(\tau)$ are finite.

## APPENDIX B: FIRST PASSAGE TIME

### 1. First passage time for unbiased CTRW

For the unbiased random walk we have $Q_L(x)=Q_R(x)=1/2$, for $x\neq 0$, $x\neq\tilde{L}$. And as mentioned $x=0$ is the absorbing boundary condition, while $\tilde{L}$ is a reflecting wall. Using Eq. (23) the $z$ transform of Eq. (25) is

$$\tilde{P}_0(z) = \frac{z}{2}\tilde{P}_1(z) + z\tilde{P}_0(z)$$

using the initial conditions $P_1(0)=1$,

$$\tilde{P}_1(z) - 1 = \frac{z}{2}\tilde{P}_2(z),$$

for $x=2,\ldots,L-2$,

$$\tilde{P}_x(z) = \frac{z}{2}[\tilde{P}_{x-1}(z) + \tilde{P}_{x+1}(z)],$$

$$\tilde{P}_{\tilde{L}-1}(z) = z\tilde{P}_{\tilde{L}}(z) + \frac{z}{2}\tilde{P}_{\tilde{L}-2}(z),$$

$$\tilde{P}_{\tilde{L}}(z) = \frac{z}{2}\tilde{P}_{\tilde{L}-1}(z), \tag{B1}$$

and using Eq. (26)

$$\tilde{F}_{\text{dis}}(z) = \frac{z}{2}\tilde{P}_1(z). \tag{B2}$$

To solve these equations we use a recursive solution method [38,39]. We define $\phi_x(z)$ using the relation

$$\tilde{P}_x(z) = \phi_x(z)\tilde{P}_{x-1}(z), \tag{B3}$$

and it is easy to show using Eqs. (B1) and (B3) that

$$\phi_{\tilde{L}}(z) = z/2 \quad \phi_{\tilde{L}-1}(z) = (z/2)/(1 - z^2/2). \tag{B4}$$

The function $\phi_x(z)$ also satisfies the recursion relation

$$\phi_{x-1}(z) = \frac{(z/2)}{1 - z\phi_x(z)/2} \tag{B5}$$

which is easy to obtain from Eq. (B1). Let

$$\phi_x(z) = \frac{g_x(z)}{h_x(z)} \tag{B6}$$

and using Eq. (A5)

$$\begin{pmatrix} g_{x-1}(z) \\ h_{x-1}(z) \end{pmatrix} = \begin{pmatrix} 0 & \frac{z}{2} \\ -\frac{z}{2} & 1 \end{pmatrix} \begin{pmatrix} g_x(z) \\ h_x(z) \end{pmatrix}. \tag{B7}$$

Since we are interested only in the ratio $g_x(z)/h_x(z)$ we may set $h_{\tilde{L}}(z)=1$ and $g_{\tilde{L}}(z)=z/2$ using Eq. (B4). Equation (B4) gives the seeds for the iteration rule Eq. (B7): $h_{\tilde{L}-1}(z)=1-z^2/2$ and $g_{\tilde{L}-1}(z)=z/2$, which yield $h_{\tilde{L}-2}(z), g_{\tilde{L}-2}(z)$, etc. Let

$$h_x(z) = B_+(\Lambda_+)^{\tilde{L}-x} + B_-(\Lambda_-)^{\tilde{L}-x}, \tag{B8}$$

then from $h_{\tilde{L}}(z)=1$ we have $B_+ + B_- = 1$. $\Lambda_\pm$ are eigenvalues of the matrix in Eq. (B7):

$$\Lambda_\pm = \frac{1 \pm \sqrt{1 - z^2}}{2}. \tag{B9}$$

Using $h_{\tilde{L}-1}(z)=1-z^2/2$ it is easy to obtain Eq. (38). The relations

$$\tilde{P}_1(z) = \frac{1}{1 - z\phi_2(z)/2}, \tag{B10}$$

$\phi_2(z)=zh_3(z)/2h_2(z)$, and Eqs. (B2) and (B8) lead to Eq. (36).

### 2. First passage time for biased CTRW

For the uniformly biased CTRW the probability to jump left is $Q_L(x)=q$ and hence the probability to jump to the right is $Q_R(x)=1-q$, for $x\neq 0$, $x\neq\tilde{L}$. The two boundary conditions are that $x=0$ is absorbing, while $\tilde{L}$ is a reflecting wall. In this case the $z$ transform of the master equation (25) is

$$\tilde{P}_0(z) = zq\tilde{P}_1(z) + z\tilde{P}_0(z),$$

$$\tilde{P}_1(z) - 1 = zq\tilde{P}_2(z),$$

$$\tilde{P}_x(z) = z(1-q)\tilde{P}_{x-1}(z) + zq\tilde{P}_{x+1}(z),$$

$$\tilde{P}_{\tilde{L}-1}(z) = z\tilde{P}_{\tilde{L}}(z) + z(1-q)\tilde{P}_{\tilde{L}-2}(z),$$

$$\tilde{P}_{\tilde{L}}(z) = z(1-q)\tilde{P}_{\tilde{L}-1}(z), \tag{B11}$$

and using Eq. (26)

$$\tilde{F}_{\text{dis}}(z) = zq\tilde{P}_1(z). \tag{B12}$$

The solution of the biased master equation (B11) follows the same procedure as for the unbiased and yields Eqs. (40)–(42). Using Eq. (B12) one can show that $\tilde{F}_{\text{dis}}(z=1)=1$, for any finite $\tilde{L}$ and $q\neq 0$, namely, if we wait long enough the particle always reaches the sticky boundary at $x=0$.

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