

Occupation times and ergodicity breaking in biased continuous time random walks

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Abstract

Continuous time random walk (CTRW) models are widely used to model diffusion in condensed matter. There are two classes of such models, distinguished by the convergence or divergence of the mean waiting time. Systems with finite average sojourn time are ergodic and thus Boltzmann–Gibbs statistics can be applied. We investigate the statistical properties of CTRW models with infinite average sojourn time; in particular, the occupation time probability density function is obtained. It is shown that in the non-ergodic phase the distribution of the occupation time of the particle on a given lattice point exhibits bimodal U or trimodal W shape, related to the arcsine law. The key points are as follows. (a) In a CTRW with finite or infinite mean waiting time, the distribution of the number of visits on a lattice point is determined by the probability that a member of an ensemble of particles in equilibrium occupies the lattice point. (b) The asymmetry parameter of the probability distribution function of occupation times is related to the Boltzmann probability and to the partition function. (c) The ensemble average is given by Boltzmann–Gibbs statistics for either finite or infinite mean sojourn time, when detailed balance conditions hold. (d) A non-ergodic generalization of the Boltzmann–Gibbs statistical mechanics for systems with infinite mean sojourn time is found.

1. Introduction

In Nature, one encounters many phenomena in which some quantity varies with time in a very complicated way. There is no hope of determining this variation in detail, but it may be true that certain averaged features vary in a way that can be described by simple laws. The averaging over a suitable time interval is an awkward procedure; one therefore replaces the irregularly varying function of time by an ensemble of functions. All averages are redefined as averages over the ensemble rather than over some time interval of the single realization of the

time varying quantity. Van Kampen [1] clarifies the concept described above, by considering a bounded Brownian motion. One may actually observe a large number of particles and average the result; that means that one has a physical realization of the ensemble. One might also observe one and the same particle on a long time interval; the results from these two averaging procedures will be the same if one assumes that sections of the single-particle trajectory are statistically independent. In a single-molecule experiment one usually performs an observation on one particle during some time interval; thus the time average is the observed quantity rather than the ensemble average [2]. The condition for ergodicity of the system, namely that time averages and ensemble averages coincide, is that the behaviour of the measured signal during one interval does not affect the behaviour during the next interval. The time average of any physical quantity is defined as

$$\bar{f} = \frac{1}{t} \int_0^t f(t') dt'. \quad (1)$$

In order for the condition of ergodicity to be fulfilled it should be possible to represent it also as

$$\bar{f} = \frac{1}{N} \sum_{i=1}^N f_i, \quad (2)$$

where f_i is a short time average

$$f_i = \frac{1}{\tau} \int_{(i-1)\tau}^{i\tau} f(t') dt' \quad (3)$$

and $\tau = t/N$. As usual we take the limit where $t \rightarrow \infty$ and $N \rightarrow \infty$ such that τ remains finite. Then ergodicity requires that the f_i s are statistically independent, which means that there exists some time interval τ which is longer than the microscopic timescale of the system but short compared to the total measurement time. The basic assumption is that such an intermediate interval exists; if that is not so, ergodicity is broken and different methods are needed. If the microscopical timescale for the dynamics diverges, we expect a non-ergodic behaviour, since the microscopical timescale is always of the order of the macroscopic total measurement time and no intermediate time interval τ exists.

Ergodicity is a key concept for Boltzmann equilibrium, where Boltzmann's probability

$$p_B(\sigma) = e^{-\frac{H(\sigma)}{k_B T}} / \sum_{\sigma'} e^{-\frac{H(\sigma')}{k_B T}} \quad (4)$$

is the probability (in the ensemble sense) of finding the system in state σ . Another important quantity is the fraction of time for which the system occupies a given state σ during a measurement,

$$\bar{p}_t(\sigma) = T_\sigma / t, \quad (5)$$

where T_σ is the time of occupation of state σ , and t is the total measurement time. The time average then can be written as

$$\bar{f} = \sum_{\sigma} \bar{p}_t(\sigma) f(\sigma), \quad (6)$$

while the ensemble average of the same quantity is given by

$$\langle f \rangle = \sum_{\sigma} p_B(\sigma) f(\sigma). \quad (7)$$

For a canonical ergodic system in equilibrium, $\bar{p}_t(\sigma) = p_B(\sigma)$, and thus the two averages coincide. For non-ergodic systems, knowledge of the occupation time of each state is needed

in order to predict statistical behaviour of the system, since even in equilibrium the average is determined by \bar{p}_t and not by p_B .

Following Bouchaud [3] we distinguish between two different types of non-ergodicity. Strong non-ergodicity is the case where the system phase space is divided into unconnected regions, which make it impossible for the system to visit all of its phase space. The more interesting case to our mind is weak non-ergodicity. In this case the phase space is fully connected; moreover the system does visit all of its phase space but the fraction of time of occupation of a given volume in phase space is not equal to the fraction of phase space volume occupied by it. In what follows we refer to weak ergodicity breaking as non-ergodicity.

In this paper we show that the continuous time random walk (CTRW) [4] with a probability density function (PDF) of waiting times which decays as a power law, i.e., $\psi(\tau) \propto \frac{1}{\tau^{\alpha+1}}$ as $\tau \rightarrow \infty$ and $0 < \alpha < 1$, displays a non-ergodic behaviour. The CTRW is widely used for describing anomalous diffusion [5, 6]. In a CTRW, unlike in a discrete random walk, the sojourn time at each site is a random variable drawn from a PDF, $\psi(\tau)$. If the mean sojourn time is finite, we can find an intermediate time interval such that the time average and the ensemble average coincide. On the other hand, if the PDF is broad, such that the mean sojourn time is infinite, there is no characteristic timescale in the system, and thus the system is not ergodic. For non-ergodic systems the Boltzmann–Gibbs statistics fails, and the question which arises is, how we can describe equilibrium and near equilibrium states of such a system?

This question is timely, as experiments on single-molecule dynamics are performed—for example, the experiment on the motion of particles in a complex actin network (i.e., a network of polymers) [7]. It was found that the particle displays a subdiffusive motion, i.e., $\langle \Delta x^2 \rangle \propto t^\alpha$ with $0 < \alpha < 1$ (α depends only on the ratio of the particle size to the network mesh size). It was also found that the PDF of the sojourn time has a power law asymptotic behaviour $\psi(\tau) \propto \tau^{-(1+\alpha)}$, a CTRW behaviour. There are many other systems which show a similar behaviour and are believed to be describable using CTRW models [5, 6]; examples of such systems are the laser cooling of atoms [8], blinking nanocrystals [9, 10], and virus motion in a living cell [11, 12]. In all these systems the origin of the non-ergodic behaviour is the very broad (power law) distribution of the relevant sojourn times (see [13] and references therein for a brief review of systems with broad waiting times, and for a discussion on the relation between power law distributions and non-ergodicity see [14]).

In this paper we study a CTRW model in the non-ergodic phase; we find the PDF of the fraction of occupation time \bar{p}_t (see equation (5)). A brief summary of our results was published recently [15]. The outline of the paper is as follows. In section 2, the model is introduced, and the importance of the first-passage time for analysis of the occupation time distribution is emphasized. In section 3, the PDF of the first-passage time in the biased CTRW is obtained (previous work considered the unbiased case in detail [16]). The relation between the fraction of occupation time PDF and the equilibrium occupation probability is discussed in section 4; in this context section 5 presents the visitation fraction distribution and shows how it leads to the generalized arcsine PDF of occupation times. In the last section we present results of numerical simulations and discuss the relation to Boltzmann–Gibbs statistical mechanics.

2. The model and relation between the discrete time RW and CTRW

We consider a particle motion in a one-dimensional lattice; each step is independent of the previous steps, and the step length is equal to the lattice spacing. The particle stays at each site for a random time τ , with a PDF

$$\psi(\tau) \sim a\tau^{-(1+\alpha)}/|\Gamma(-\alpha)| \quad (\text{for } \tau \rightarrow \infty), \quad (8)$$

where a is a parameter with units $[\tau^\alpha]$, and $0 < \alpha < 1$, leading to a diverging mean sojourn time. In Laplace $\tau \rightarrow u$ space, the small u behaviour of the PDF is

$$\hat{\psi}(u) \sim 1 - au^\alpha + \dots, \quad (9)$$

where $\hat{\psi}(u)$ is the Laplace transform of $\psi(\tau)$. The lattice is finite, of length $N + 1$, and the boundaries on $x = 0$ and $x = N$ are reflecting. The probability of jumping left from site x is $q_l(x)$, and that of jumping right is $1 - q_l(x)$. This probability may change from site to site. This kind of motion is a generalized CTRW. Our aim is to calculate the PDF of the fraction of occupation time for each site in the lattice $f_i(\bar{p}_i(x))$ (where $\bar{p}_i(x) = T_x/t$, T_x is the total occupation time of site x , and t is the total measurement time), in order to study and quantify the ergodic properties of the model.

We consider a two-state process. One state is when the particle is at the site of interest (say x) and the other one is when the particle is at any other site of the lattice. Accordingly there are two different sojourn time PDFs; for the sojourn time at the site, the PDF is $\psi_+(\tau) = \psi(\tau)$, while for the sojourn time outside the observed cell, the PDF is denoted by $\psi_-(\tau)$. $\psi_-(\tau)$ is a combination of the first-passage-time (FPT) PDF when the particle starts at $x - 1$ and reaches x , $\psi_{Lx}^{\text{fpt}}(\tau)$, and the FPT PDF when the particle goes from $x + 1$ to x , $\psi_{Rx}^{\text{fpt}}(\tau)$, where each of them is multiplied by the probability of jumping from x to the left or to the right respectively, i.e.,

$$\psi_-(\tau) = q_l(x)\psi_{Lx}^{\text{fpt}}(\tau) + [1 - q_l(x)]\psi_{Rx}^{\text{fpt}}(\tau). \quad (10)$$

In order to calculate the FPT PDF in the CTRW model we introduce a general relation between the survival probability of a CTRW particle and the survival probability in a discrete time random walk (RW) [17]. Let $S_{\text{CT}}(t)$ be the survival probability of a CTRW particle, i.e., the probability that the particle did not arrive at site x , up to time t . One can express $S_{\text{CT}}(t)$ as the sum over n of the survival probabilities after n jumps in a discrete time random walk, $S_{\text{Dis}}(n)$, where each term has a weight given by $w(n, t)$ —the probability that n jumps occurred in the interval $(0, t)$. Namely

$$S_{\text{CT}}(t) = \sum_{n=0}^{\infty} w(n, t) S_{\text{Dis}}(n). \quad (11)$$

In Laplace space it takes the form

$$\begin{aligned} \hat{S}_{\text{CT}}(u) &= \frac{1 - \hat{\psi}(u)}{u} \sum_{n=0}^{\infty} \hat{\psi}^n(u) S_{\text{Dis}}(n) \\ &= \frac{1 - \hat{\psi}(u)}{u} \tilde{S}_{\text{Dis}}[\hat{\psi}(u)], \end{aligned} \quad (12)$$

where we used

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

and $\tilde{}$ denotes the z transform defined as

$$\tilde{S}[z] = \sum_{n=0}^{\infty} z^n S(n). \quad (13)$$

The first-passage-time PDF is given by minus the derivative of the survival probability with respect to the time t ; in Laplace space it reads

$$\begin{aligned} \hat{\psi}^{\text{fpt}}(u) &= -u \hat{S}_{\text{CT}}(u) + S_{\text{CT}}(t=0) \\ &= -\tilde{S}_{\text{Dis}}[\hat{\psi}(u)](1 - \hat{\psi}(u)) + 1. \end{aligned} \quad (14)$$

Following equation (14), we first find the FPT probability in the discrete time RW model and later derive from it the FPT PDF for the CTRW.

3. The first-passage-time PDF for the biased CTRW

We consider a biased CTRW, i.e., $q_l(x) = q_l$ for any x (excluding the boundaries). The calculation for the case $q_l = 1/2$ was given in [16]. To calculate the first-passage-time PDF we start with a discrete time RW model. The particle starts at $x - 1$ and we calculate the PDF for the first time it arrives at x (i.e., $\psi_{Lx}^{\text{fpt}}(\tau)$). The boundary conditions are: (a) site 0 is a reflecting boundary; (b) site x is an absorbing site. The master equations describing this model are

$$\begin{aligned} p_0(n) &= q_l p_1(n-1) \\ p_1(n) &= q_l p_2(n-1) + p_0(n-1) \\ p_y(n) &= [1 - q_l] p_{y-1}(n-1) + q_l p_{y+1}(n-1) \\ p_{x-1}(n) &= [1 - q_l] p_{x-2}(n-1) \\ p_x(n) &= [1 - q_l] p_{x-1}(n-1) + p_x(n-1) \\ F_x(n) &= [1 - q_l] p_{x-1}(n-1), \end{aligned} \quad (15)$$

where y is limited to the range $2 \leq y \leq x - 2$. $F_x(n)$ is the probability of first arriving at site x after the n th step, and $p_y(n)$ is the probability of being at site y after the n th step. In this case the survival probability $S_{Lx}(t)$ (i.e., the probability of not arriving at site x up to time t starting at $x - 1$) is given by

$$\begin{aligned} S_{Lx}(t) &= 1 - p_x(t) = \sum_{n=0}^{\infty} w(n, t) [1 - p_x(n)] \\ &= \sum_{n=0}^{\infty} w(n, t) [1 - p_x(n-1) - (1 - q_l) p_{x-1}(n-1)], \end{aligned} \quad (16)$$

where $p_x(t)$ is the probability that the particle is at site x at time t . In Laplace space

$$\hat{S}_{Lx}(u) = \frac{1 - \hat{\psi}(u)}{u} \sum_{n=0}^{\infty} \hat{\psi}^n(u) [1 - p_x(n-1) - (1 - q_l) p_{x-1}(n-1)]. \quad (17)$$

Combining equations (17) with (15) and performing some algebra, we can rewrite equation (17) as

$$\hat{S}_{Lx}(u) = \frac{1}{u} (1 - \tilde{F}_x[\hat{\psi}(u)]). \quad (18)$$

Using equations (14) and (18) we write for the FPT PDF

$$\hat{\psi}_{Lx}^{\text{fpt}}(u) = \tilde{F}_x[\hat{\psi}(u)]. \quad (19)$$

Now we turn to solving the discrete time model in order to obtain $\tilde{F}_x[\hat{\psi}(u)]$. We use a method presented by Redner [18]; we start by performing a z transform (defined in equation (13)) of the master equation (15), which is then rewritten as

$$\begin{aligned} \tilde{p}_0(z) &= z q_l \tilde{p}_1(z) \\ \tilde{p}_1(z) &= z q_l \tilde{p}_2(z) + z \tilde{p}_0(z) \\ \tilde{p}_y(z) &= q_l z \tilde{p}_{y+1}(z) + [1 - q_l] z \tilde{p}_{y-1}(z) \\ \tilde{p}_{x-1}(z) &= [1 - q_l] z \tilde{p}_{x-2}(z) + 1 \\ \tilde{p}_x(z) &= [1 - q_l] z \tilde{p}_{x-1}(z) + z \tilde{p}_x(z) \\ \tilde{F}(z) &= [1 - q_l] z \tilde{p}_{x-1}(z). \end{aligned} \quad (20)$$

By substituting the first equation into the second one we get a relation between $\tilde{p}_1(z)$ and $\tilde{p}_2(z)$, and so on, and equation (20) can be rewritten as

$$\begin{aligned}\tilde{p}_0(z) &= zq_l \tilde{p}_1(z) \\ \tilde{p}_1(z) &= \frac{zq_l}{1 - z^2q_l} \tilde{p}_2(z) \\ \tilde{p}_2(z) &= \frac{zq_l}{1 - [1 - q_l]z \frac{zq_l}{1 - z^2q_l}} \tilde{p}_3(z) \\ &\vdots \\ \tilde{p}_{x-1}(z) &= [1 - q_l]z\varphi_{x-2} \tilde{p}_{x-1}(z) + 1 \\ \tilde{p}_x(z) &= [1 - q_l]z \tilde{p}_{x-1}(z) + z \tilde{p}_x(z) \\ \tilde{F}(z) &= \frac{[1 - q_l]z}{1 - [1 - q_l]z\varphi_{x-2}} \tilde{p}_{x-1}(z).\end{aligned}\tag{21}$$

The general relation is

$$\tilde{p}_x(z) = \varphi_x(z, q_l) \tilde{p}_{x+1}(z).\tag{22}$$

As seen from equation (21), the recursion relation for φ_y reads

$$\varphi_y = \frac{zq_l}{1 - z[1 - q_l]\varphi_{y-1}},\tag{23}$$

and the seed of the recursion relation is

$$\begin{aligned}\varphi_0 &= zq_l \\ \varphi_1 &= \frac{zq_l}{1 - z^2q_l}.\end{aligned}\tag{24}$$

The recursion relation is solved in appendix A using a method introduced by Goldhirsch and Gefen [19]. Substituting the solution into equation (21) yields

$$\tilde{F}(z) = \frac{[1 - q_l]z}{1 - z^2q_l[1 - q_l] \frac{A_+\lambda_+^{L-3} + A_-\lambda_-^{L-3}}{A_+\lambda_+^{L-2} + A_-\lambda_-^{L-2}}},\tag{25}$$

where

$$\lambda_{\pm} = \frac{1 \pm \sqrt{1 - 4z^2q_l[1 - q_l]}}{2}\tag{26}$$

and

$$A_- = \frac{(1 - z^2q_l) - \lambda_+}{[\lambda_- - \lambda_+]}; \quad A_+ = \frac{-(1 - q_lz^2) + \lambda_-}{[\lambda_- - \lambda_+]}. \tag{27}$$

The asymptotic behaviour of the first-passage time is obtained by substituting into $\tilde{F}[\hat{\psi}(u)]$ the asymptotic behaviour of $\hat{\psi}(u)$ as $u \rightarrow 0$ (equation (9)); we find

$$\hat{\psi}_{Lx}^{\text{fpt}}(u) = 1 - a \frac{1 - 2[1 - q_l]^{1-x}q_l}{1 - 2q_l} u^\alpha + \dots.\tag{28}$$

The FPT PDF for the case where the particle starts at $x + 1$ and reaches x , $\hat{\psi}_{Rx}^{\text{fpt}}(u)$, is obtained by replacing x by $N - x$ and q_l by $1 - q_l$ in the expression for $\hat{\psi}_{Lx}^{\text{fpt}}(u)$. Writing explicitly $\hat{\psi}_{Lx}^{\text{fpt}}(u)$ and $\hat{\psi}_{Rx}^{\text{fpt}}(u)$ in equation (10), we find

$$\hat{\psi}_-(x, u) \simeq 1 - A_x^* u^\alpha + \dots,\tag{29}$$

where

$$A_x^* \equiv a \left[\frac{2}{2q_l - 1} \left(q_l^2 \left[\frac{q_l}{1 - q_l} \right]^{x-1} - [1 - q_l]^2 \left[\frac{1 - q_l}{q_l} \right]^{N-x-1} \right) - 1 \right]. \tag{30}$$

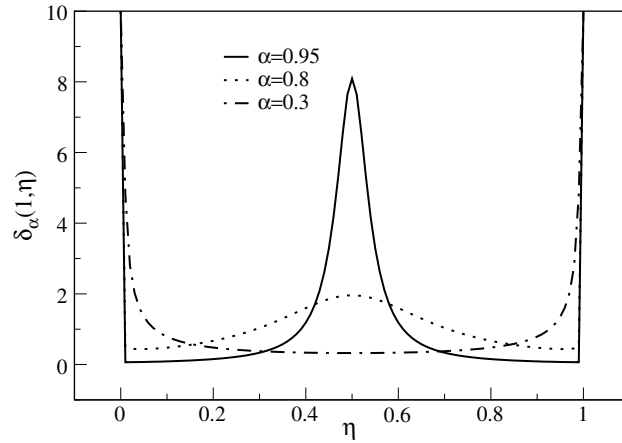


Figure 1. Lamperti's PDF equation (32) with $R = 1$. The dash-dotted curve corresponds to $\alpha = 0.3$. The PDF attains its maxima around 1 and 0, while the probability of obtaining the ergodic expectation value ($1/2$ in this case) is almost zero. The dotted curve and the solid curve correspond to $\alpha = 0.8, 0.95$ respectively. As α approaches 1, the ergodic behaviour is recovered. When $\alpha = 1$ the PDF becomes a delta function centred at the ensemble average $\eta = 1/2$.

As mentioned, the process of occupying site x , then disengaging from it, reoccupying x , etc, can be thought of as a two-state process with waiting times given by ψ_{\pm} respectively, and the PDF of the fraction of occupation time $\bar{p}_t(x)$ is found on the basis of Lamperti's limit theorem [20]:

$$f_t(\bar{p}_t(x)) = \delta_{\alpha}(a/A_x^*, \bar{p}_t(x)), \quad (31)$$

where

$$\delta_{\alpha}(R, \eta) = \frac{\sin(\alpha\pi)}{\pi} \frac{R\eta^{\alpha-1}(1-\eta)^{\alpha-1}}{R^2(1-\eta)^{2\alpha} + \eta^{2\alpha} + 2R(1-\eta)^{\alpha}\eta^{\alpha}\cos(\alpha\pi)}. \quad (32)$$

In appendix B, equations (31), (32) are derived. R is called the asymmetry parameter; the case $R = 1$ corresponds to a symmetric PDF (see figure 1). For the unbiased CTRW where $q_l = 1/2$, equation (30) simplifies to

$$A_x^* \equiv a[N-1]. \quad (33)$$

In figures 1, 2 we show some plots of the PDF equation (31). Note that previously non-trivial occupation time PDFs were investigated in the context of a diffusing particle in a random medium [21] and for a class of stochastic processes [22, 23].

4. Equilibrium distribution and its relation to the occupation time PDF

We express the result of equation (31) in terms of the equilibrium probability of being at site x , $p_{\text{eq}}(x)$. $p_{\text{eq}}(x)$ is the probability of finding a single member of a large ensemble of non-interacting particles in equilibrium, on the lattice cell x . Note that for a finite system, $p_{\text{eq}}(x)$ is independent of the sojourn time PDF. One can verify easily that for the biased CTRW

$$\begin{aligned} p_{\text{eq}}(x) &= \Omega \left(\frac{1-q_l}{q_l} \right)^x & 0 < x < N \\ p_{\text{eq}}(0) &= \Omega(1-q_l) \\ p_{\text{eq}}(N) &= \Omega(1-q_l) \left(\frac{1-q_l}{q_l} \right)^{N-1}, \end{aligned} \quad (34)$$

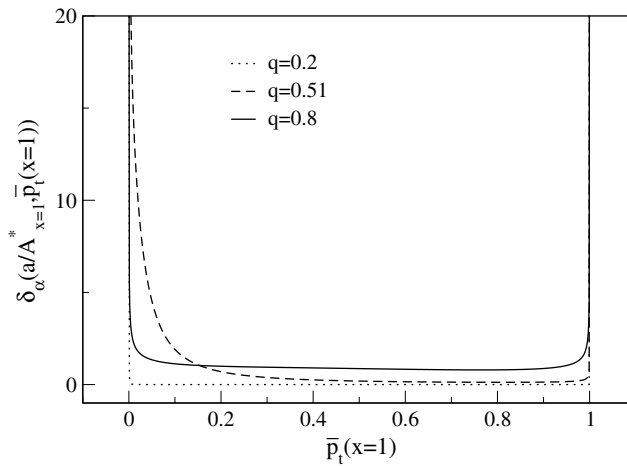


Figure 2. The PDF of the fraction of time of occupation of the site $x = 1$ ($\bar{p}_t(x = 1)$), during a CTRW in the presence of constant bias; $q_l = 0.2, 0.51, 0.8$ respectively. We used $N = 9$ and $\alpha = 0.3$. For $q_l = 0.2$ the particle is found mostly in the vicinity of the boundary on $x = 9$; hence we most probably find $\bar{p}_t(x = 1) \simeq 0$. For $\alpha = 0.3$ the PDF of the fraction of occupation times exhibits bimodal behaviour; the average is not likely to be observed in a measurement.

where Ω is the normalization factor. Using equations (30) and (34) one can express the asymmetry parameter as

$$R_x \equiv a/A_x^* = \frac{p_{\text{eq}}(x)}{1 - p_{\text{eq}}(x)}. \quad (35)$$

Assume that the RW obeys the detailed balance relation between the probabilities of jumping right and left. This means that $\frac{q_l}{1 - q_l} = e^{-\frac{\Lambda\xi}{k_B T}}$, where Λ is the constant force, ξ is the lattice spacing, k_B is the Boltzmann constant, and T is the temperature. In this case $p_{\text{eq}}(x)$ is just the well known Boltzmann probability and equation (35) reads

$$R_x = \frac{p_B(x)}{1 - p_B(x)}. \quad (36)$$

It is worth mentioning that, as shown here, the detailed balance relation does not necessarily imply ergodicity of the system, but an equilibrium distribution in an ensemble sense which is given by the Boltzmann probability.

5. Visitation fraction and the generalized arcsine PDF

To generalize our results beyond the uniformly biased CTRW, we study the distribution of the number of visits in the x th cell n_x , given that during the measurement time there were n visits (i.e., jumps between cells). The master equation describing the evolution of the probability of occupying site y after n jumps in the CTRW is identical to the master equation describing the discrete time RW, namely

$$p_x(n) = q_l(x+1)p_{x+1}(n-1) + (1 - q_l(x-1))p_{x-1}(n-1) \quad (37)$$

(excluding the reflecting boundaries). For the discrete time RW we assume ergodicity of the process, implying

$$n_x/n = p_{\text{eq}}(x), \quad (38)$$

where $p_{\text{eq}}(x)$ is the probability of occupying site x in equilibrium, in the ensemble sense. $p_{\text{eq}}(x)$ is defined by the condition that $p_x(n) = p_x(n-1) = p_{\text{eq}}(x)$ in the limit $n \rightarrow \infty$. $p_{\text{eq}}(x)$ and the visitation fraction n_x/n are related to the master equation, equation (37), and to the lattice properties, but not to the waiting time PDF. Thus equation (38) holds for both discrete and continuous time RW. For a discussion on the transformation between the discrete time RW and the CTRW (subordination) in the context of the fractional Fokker–Planck equation, see [24–26]. In the case of finite mean waiting time $\langle \tau \rangle$, the fraction of occupation time may be written as

$$T_x/t \simeq n_x \langle \tau \rangle / n \langle \tau \rangle = n_x/n, \quad (39)$$

which together with equation (38) implies ergodicity of the system. For a CTRW with diverging mean waiting time the PDF of the occupation time is derived as follows.

We denote by $f_{n,t}^0(T_x)$ the PDF of T_x in the case where the particle is not at x at the end of the measurement. $f_{n,t}^0(T_x)$ is written as

$$f_{n,t}^0(T_x) = \left\langle \delta \left(T_x - \sum_{i \in x} \tau_i \right) I(t_n < t < t_{n+1}) \right\rangle, \quad (40)$$

t_i is the time at which the i th jump occurs, and τ_i is the i th sojourn time in x (see figure B.1 in appendix B). $I(t_n < t < t_{n+1}) = 1$ if the statement in parentheses is true, and 0 otherwise. The brackets $\langle \rangle$ denote the average over all τ 's. The summation is over all sojourn times in x . Performing a double Laplace transform of equation (40) yields

$$\begin{aligned} \hat{f}_{n,s}^0(u) &= \left\langle \int_0^\infty \int_0^\infty e^{-uT_x} e^{-st} \delta \left(T_x - \sum_{i \in x} \tau_i \right) I(t_n < t < t_{n+1}) dT_x dt \right\rangle \\ &= \frac{\hat{\psi}^{n_x}(u+s) \hat{\psi}^{n-n_x}(s) (1 - \hat{\psi}(s))}{s}, \end{aligned} \quad (41)$$

where we assume n_x visits in x . Like in equation (40), the PDF of T_x in the case where the particle is within the x th cell at the end of the measurement is denoted by $f_{n,t}^1(T_x)$; that is,

$$f_{n,t}^1(T_x) = \left\langle \delta \left(T_x - \left[\sum_{i \in x} \tau_i + \tau^* \right] \right) I(t_n < t < t_{n+1}) \right\rangle, \quad (42)$$

where $\tau^* \equiv t - t_n$ is the time between the last jump and the end of the measurement (see figure B.1), and in double Laplace space

$$\hat{f}_{n,s}^1(u) = \hat{\psi}^{n_x}(u+s) \hat{\psi}^{n-n_x}(s) \frac{1 - \hat{\psi}^{n_x+1}(u+s)}{(s+u)}. \quad (43)$$

The probability for the particle to be within the x th cell at the end of the measurement is given by $p_{\text{eq}}(x)$. Thus the double Laplace transform of the PDF of the occupation time of the x th cell, given that there were n jumps between cells during a measurement of time t , is given by

$$\hat{f}_{n,s}(u) = p_{\text{eq}}(x) \hat{f}_{n,s}^1(u) + (1 - p_{\text{eq}}(x)) \hat{f}_{n,s}^0(u). \quad (44)$$

Substituting equations (41) and (43) in (44), and using equation (38) we rewrite $\hat{f}_{n,s}(u)$ as

$$\begin{aligned} \hat{f}_{n,s}(u) &\simeq \left[p_{\text{eq}}(x) \frac{1 - \hat{\psi}(u+s)}{(s+u)} + (1 - p_{\text{eq}}(x)) \frac{1 - \hat{\psi}(s)}{s} \right] \\ &\times \left(\hat{\psi}^{p_{\text{eq}}(x)}(u+s) \hat{\psi}^{1-p_{\text{eq}}(x)}(s) \right)^n, \end{aligned} \quad (45)$$

where we replaced n_x with $p_{\text{eq}}(x)n$ (equation (38)). Summing over the number of events during the measurement, n , one obtains

$$\hat{f}_s(u) = \sum_n \hat{f}_{n,s}(u) = \left[p_{\text{eq}}(x) \frac{1 - \hat{\psi}(u+s)}{(s+u)} + (1 - p_{\text{eq}}(x)) \frac{1 - \hat{\psi}(s)}{s} \right] \times \frac{1}{1 - \hat{\psi}^{p_{\text{eq}}(x)}(u+s) \hat{\psi}^{1-p_{\text{eq}}(x)}(s)}, \tag{46}$$

an equation valid only in the scaling limit. Taking the limit $s, u \rightarrow 0$ (using equation (9)), which corresponds to the long measurement and occupation time limit, one finds the asymptotic behaviour of $\hat{f}_s(u)$ as

$$\hat{f}_s(u)_{s,u \rightarrow 0} \sim \frac{p_{\text{eq}}(x)(u+s)^{\alpha-1} + (1 - p_{\text{eq}}(x))s^{\alpha-1}}{p_{\text{eq}}(x)(u+s)^\alpha + (1 - p_{\text{eq}}(x))s^\alpha}. \tag{47}$$

Inverting the double Laplace transform [27] yields the PDF of the fraction of occupation time:

$$f\left(\frac{T_x}{t}\right) = \delta_\alpha\left(R_x, \frac{T_x}{t}\right) \quad \text{where } R_x = \frac{p_{\text{eq}}(x)}{1 - p_{\text{eq}}(x)}. \tag{48}$$

This equation is the main result of this section; it recovers the special case of a uniformly biased CTRW of equation (31).

Remark. The concept of the visitation fraction discussed in this section allows us to define more precisely the different types of ergodicity breaking. A system is said to be ergodic if the visitation fraction of each site is equal to the fraction of occupation time, which in turn is equal to the equilibrium probability of occupying the site in the ensemble sense, equation (39). If the visitation fraction obeys equation (38), but is not equal to the fraction of occupation time, the system is said to be weakly non-ergodic. If the process does not obey equation (38) for the visitation fraction, and the fraction of occupation time is not equal to the ensemble equilibrium probability, the process is said to be strongly non-ergodic. Note that since weakly non-ergodic systems obey the relation (38), a statistical mechanical description is still possible, unlike in strongly non-ergodic systems [29].

6. Discussion

If we imply the detailed balance condition, the equilibrium probability is given by Boltzmann probability,

$$p_B(x) = e^{-\frac{U(x)}{T}} / \sum_y e^{-\frac{U(y)}{T}} \tag{49}$$

where $U(x)$ is the potential at the x th cell and T is the temperature. The asymmetry parameter in this case is $R_x = \frac{p_B(x)}{1 - p_B(x)}$. Then equation (48) yields a relation between the non-ergodic dynamics and the partition function.

The asymmetry parameter R_x can be written as $R_x = Ze^{\frac{U(x)}{T}} - 1$, where Z is the partition function. It can also be written as the partition function of the system excluding the site x , where the energies are measured relative to the energy of site x , i.e.,

$$R_x = Z' = Ze^{\frac{U(x)}{T}} - 1 = e^{\frac{U(x)}{T}} \sum_y e^{-\frac{U(y)}{T}} - 1 = \sum_{y \neq x} e^{-\frac{[U(y)-U(x)]}{T}}. \tag{50}$$

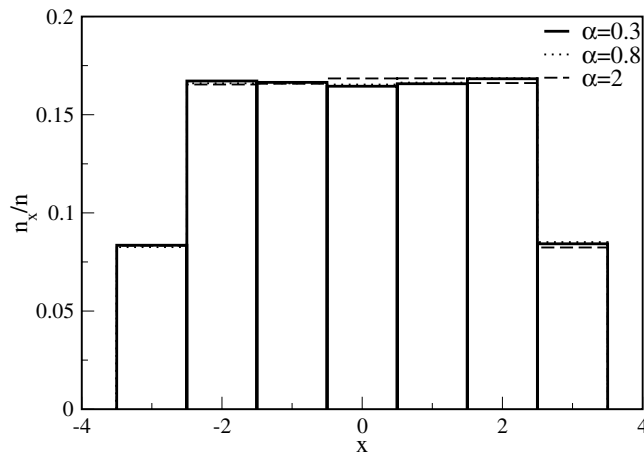


Figure 3. The visitation fraction in each cell for different values of α in an unbiased CTRW. $n_x/n = p_{\text{eq}}(x) = 1/N$ (beside x on the boundary) both for the ergodic case $\alpha > 1$ and for the non-ergodic phase $\alpha < 1$. The visitation fraction is obtained from a single trajectory.

The distribution of the visitation fraction in each cell was verified using numerical simulations, which give an estimate of the time needed to converge to the long time limit. The waiting time PDF that we used in all the numerical simulation is

$$\psi(\tau) = \alpha\tau^{-(1+\alpha)} \quad \text{for } \tau > 1. \quad (51)$$

In figure 3 we show the distribution of the visitation fraction in an unbiased CTRW ($q_l = 1/2$) for ergodic and non-ergodic cases ($\alpha > 1$ and $\alpha < 1$ respectively). In both cases, the visitation fraction in each cell (excluding the reflecting boundaries) is equal; however, as was found above (equation (32)), the occupation time is a random variable in the non-ergodic case.

The visitation fraction, equation (38), and the fraction of occupation time PDF, equation (48), allow us to generalize our results to the question of the fraction of occupation time of $M < N$ cells and not necessarily one. This is important for coarse grained description of CTRW systems. In figure 4 we show the PDF of the fraction of occupation time of M cells, for the case of an unbiased CTRW (reflecting boundary condition), and a system of size $N + 1$. In this case (see equation (50)) $R_x = M/(N + 1 - M)$ for any M lattice points excluding the boundary points.

In order to verify the validity of the visitation fraction rule, equation (38), in a thermal model, we performed a simulation of a motion in a harmonic potential with finite temperature. The motion is similar to the motion presented in section 2; the sojourn time at each site is randomly distributed from the PDF equation (51). For each site there is a detailed balance relation between the probability of jumping to the left and the probability of jumping to the right (see the details in [15]). The distribution of the visitation fraction for different values of α is shown in figure 5; as expected, it coincides with the ensemble average, which in turn coincides with Boltzmann distribution, i.e., $n_x/n = p_B(x)$. The time it took for the system to converge to the distribution presented is 10^5 , 10^8 , 10^{16} for $\alpha = 2$, 0.8, 0.3 respectively. Note that this time is not a machine time, since the number of operations is determined by the number of jumps (visits) which is the same for all cases (we used $n = 10^5$). During the motion we also measured the time that the particle stays at a specific site; we repeated the procedure many times and then obtained the statistics for the occupation time of the site. It

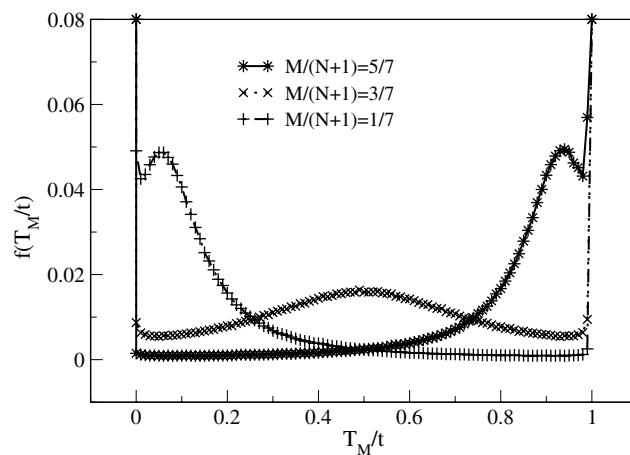


Figure 4. The PDF of the fraction of occupation time of M cells (T_M/t) for different values of $M/(N+1)$ for the unbiased CTRW. We used $\alpha = 0.8$. The dash-dotted, dotted, and solid curves correspond to $M/(N+1) = 1/7, 3/7, 5/7$ respectively. In all cases we do not consider cells on the boundary. The symbols correspond to simulation results while the curves correspond to analytic results, equation (48), without fitting.

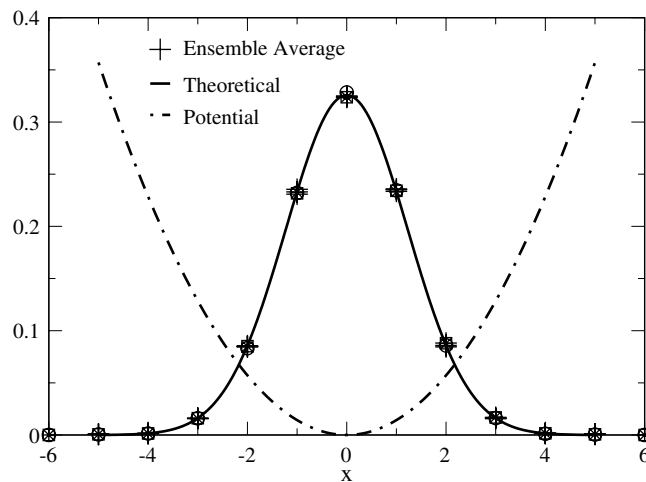


Figure 5. A CTRW in a harmonic potential $U(x) = x^2$, with temperature $T = 3$. The dot-dashed curve shows the scaled potential; the solid curve shows the Boltzmann probability. The plus signs show the distribution of the location of many particles after long walking time, i.e., the ensemble average, which as expected coincides with the Boltzmann probability. The circles, squares, and stars show the fraction of visits number for $\alpha = 2, 0.8, 0.3$ respectively. For all cases it coincides with the Boltzmann probability. The figure illustrates that the visitation fraction rule of equation (38) holds.

was found that the results are in excellent agreement with the prediction of the analytic theory, equation (48). The results for different values of α are presented in figure 6.

In conclusion, there are few key points in our theory:

- (a) In the CTRW with power law behaviour the distribution of the visitation fraction is the same as in a CTRW with finite mean sojourn time, i.e., $n_x/n = p_{\text{eq}}$.

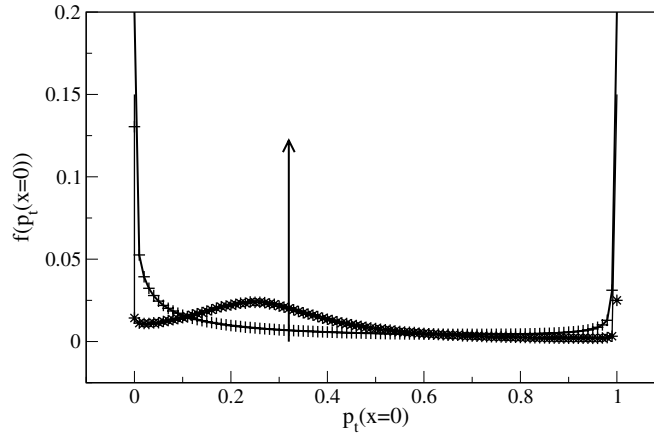


Figure 6. The PDF of the fraction of occupation time of the site $x = 0$ during a CTRW in a harmonic potential well $U(x) = x^2$ and at temperature $T = 3$. The plus signs and stars show the simulation results with $\alpha = 0.3, 0.8$ respectively, while the solid lines show the analytic results without fitting. The arrow shows the value expected from Boltzmann statistics. In this simulation we used 10^6 paths each of duration 10^6 to produce the PDF.

- (b) The PDF of the fraction of occupation time is a delta function around the Boltzmann probability in the case of finite mean sojourn time (ergodic system), while in the case of diverging mean waiting time the PDF of the occupation time is given by $\delta_\alpha(R_x, T_x/t)$, equation (32).
- (c) The exponent α is the same as the exponent describing the subdiffusion $\langle x^2 \rangle \propto t^\alpha$.
- (d) The asymmetry parameter R_x is related to the Boltzmann probability and to the partition function according to equation (50).
- (e) The ensemble average is given by Boltzmann–Gibbs statistics when the detailed balance condition is applied.
- (f) A generalization of Boltzmann–Gibbs statistical mechanics for systems with infinite mean sojourn time is possible once the fraction of occupation time PDF, $\delta_\alpha(R_x, T_x/t)$, is known.
- (g) If thermal detailed balance condition does not hold, our main result, equation (32) which yields the PDF of occupation fraction, still holds. However, now the asymmetry parameter R_x is found from the non-Boltzmann ensemble equilibrium equation (35).

Recently, the dynamical foundation of weak ergodicity breaking was investigated in [29], for deterministic (non-stochastic) maps.

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Appendix A. Solving the recursion relation for φ_x in the biased case

In order to solve the recursion relation of φ_x we define

$$\varphi_x = \frac{g_x}{h_x} = \frac{zq_l}{1 - z(1 - q_l)\frac{g_{x-1}}{h_{x-1}}} = \frac{zq_l h_{x-1}}{h_{x-1} - z(1 - q_l)g_{x-1}}, \quad (52)$$

and hence we may write out the iteration rules

$$g_x = zq_l h_{x-1} \quad (53)$$

$$h_x = h_{x-1} - z(1 - q_l)g_{x-1}. \quad (54)$$

Substituting equation (53) into (54) one obtains

$$h_x = h_{x-1} - z^2 q_l (1 - q_l) h_{x-2}. \quad (55)$$

We look for a solution of the form

$$h_x = A_+ \lambda_+^x + A_- \lambda_-^x, \quad (56)$$

which, together with equation (55), yields

$$\lambda_{\pm} (1 - \lambda_{\pm}) - z^2 q_l (1 - q_l) = 0. \quad (57)$$

Solving the equation leads to

$$\lambda_{\pm} = \frac{1 \pm \sqrt{1 - 4z^2 q_l [1 - q_l]}}{2}. \quad (58)$$

The seeds of the recursion are

$$\begin{aligned} h_0 &= 1; & g_0 &= zq_l \\ h_1 &= 1 - z^2 q_l; & g_1 &= zq_l, \end{aligned} \quad (59)$$

and the equations for A_{\pm} (defined in equation (56)) are

$$\begin{aligned} A_+ + A_- &= 1 \\ A_+ \lambda_+ + A_- \lambda_- &= 1 - q_l z^2. \end{aligned} \quad (60)$$

The solution of these equations is

$$A_- = \frac{(1 - z^2 q_l) - \lambda_+}{[\lambda_- - \lambda_+]}; \quad A_+ = \frac{-(1 - q_l z^2) + \lambda_-}{[\lambda_- - \lambda_+]}. \quad (61)$$

First we find $\tilde{p}_{L-1}(z)$ according to equations (21), (56), as

$$\tilde{p}_{L-1}(z) = \frac{\lambda_+^{L-2} + A_- [\lambda_-^{L-2} - \lambda_+^{L-2}]}{\lambda_+^{L-2} + A_- [\lambda_-^{L-2} - \lambda_+^{L-2}] - z^2 q_l [1 - q_l] [\lambda_+^{L-3} + A_- [\lambda_-^{L-3} - \lambda_+^{L-3}]]}$$

and then $\tilde{F}(z)$ is given by

$$\tilde{F}(z) = \frac{[1 - q_l] z}{1 - z^2 q_l [1 - q_l] \frac{A_+ \lambda_+^{L-3} + A_- \lambda_-^{L-3}}{A_+ \lambda_+^{L-2} + A_- \lambda_-^{L-2}}}. \quad (62)$$

Appendix B. Generalized arcsine distribution

Consider a stochastic process, defined as follows. Events occur at random times t_1, t_2, \dots measured from some time origin $t = 0$ (see figure B.1). We take the origin to be at an event occurrence time. The intervals between events, $\tau_1 = t_1, \tau_2 = t_2 - t_1, \dots$, are independent random variables. The even intervals are identically distributed random variables with PDF $\psi_-(\tau)$, and the odd intervals are identically distributed random variables with PDF $\psi_+(\tau)$. One can look at the process as a set of transitions between two states (+ and -), characterized by the sequence $\{\tau_1^+, \tau_2^-, \tau_3^+, \tau_4^-, \tau_5^+, \dots\}$ of \pm sojourn times. This is a renewal process with two waiting time PDFs ψ_+ and ψ_- . Let us introduce some quantities that we use in our calculations. First the number of events in the interval $(0, t)$ is denoted by n , and is the random variable for

the largest i for which $t_i < t$ (see figure B.1). The time of occurrence of the last event before t , that is of the n th event, is therefore

$$t_n = \tau_1 + \tau_2 + \cdots + \tau_n. \quad (63)$$

The backward recurrence time, τ^* , is defined as the time measured backward from t to the last event before t , i.e.,

$$\tau^* = t - t_n. \quad (64)$$

A schematic diagram of the process with all these times is shown in figure B.1. The occupation times T_+ and T_- , i.e., the times spent by the process in + and - states respectively up to time t , can be expressed as the sums of the sojourn times in each state. Assume that at $t = 0$ the particle was in the + state; then

$$T_+ = \tau_1 + \tau_3 + \cdots + \tau_n, \quad (65)$$

$$T_- = \tau_2 + \tau_4 + \cdots + \tau_{n-1} + \tau^*, \quad (66)$$

if $n = 2k + 1$, and

$$T_+ = \tau_1 + \tau_3 + \cdots + \tau_{n-1} + \tau^*, \quad (67)$$

$$T_- = \tau_2 + \tau_4 + \cdots + \tau_n, \quad (68)$$

if $n = 2k$. We denote by $f_t^\pm(T_+)$ the PDF of T_+ in a measurement of length t , given that at $t = 0$ the process starts at a \pm state. $f_{t,n}^\pm(T_+)$ will denote the PDF of T_+ in a measurement of length t , given that at $t = 0$ the process starts at a \pm state and n events occur in the interval $(0, t)$. $f_t^\pm(T_+)$ can be expressed as the sum over n of $f_{n,t}^\pm(T_+)$, i.e.,

$$f_t^\pm(T_+) = \sum_{n=0}^{\infty} f_{n,t}^\pm(T_+). \quad (69)$$

We introduce $f_{n,t}^+(T_+)$ as

$$f_{n,t}^+(T_+) = \left\langle \delta \left(T_+ - \sum_{\substack{i=1 \\ (\text{odd}'s)}}^n \tau_i \right) I(t_n < t < t_{n+1}) \right\rangle \quad (70)$$

for $n = 2k + 1$, and

$$f_{n,t}^+(T_+) = \left\langle \delta \left(T_+ - \sum_{\substack{i=1 \\ (\text{odd}'s)}}^{n-1} \tau_i - \tau^* \right) I(t_n < t < t_{n+1}) \right\rangle \quad (71)$$

for $n = 2k$. $I(t_n < t < t_{n+1}) = 1$ if the statement in the parentheses is true, and 0 otherwise. The brackets $\langle \rangle$ denote the average over all τ s.

Performing Laplace transformation (LT) as follows:

$$\hat{f}_{n,s}(u) \equiv \int_0^\infty e^{-uT_+} \int_0^\infty e^{-st} f_{n,t}(T_+) dt \quad (72)$$

and using the definition of the LT of the sojourn times PDF

$$\hat{\psi}_\pm(s) = \int_0^\infty e^{-s\tau} \psi_\pm(\tau) d\tau, \quad (73)$$

one obtains

$$\hat{f}_{2k,s}^+(u) = \hat{\psi}_+^k(s+u) \hat{\psi}_-^k(s) \frac{1 - \hat{\psi}_+(s+u)}{s+u}, \quad (74)$$

$$\hat{f}_{2k+1,s}^+(u) = \hat{\psi}_+^{k+1}(s+u) \hat{\psi}_-^k(s) \frac{1 - \hat{\psi}_-(s)}{s}, \quad (75)$$

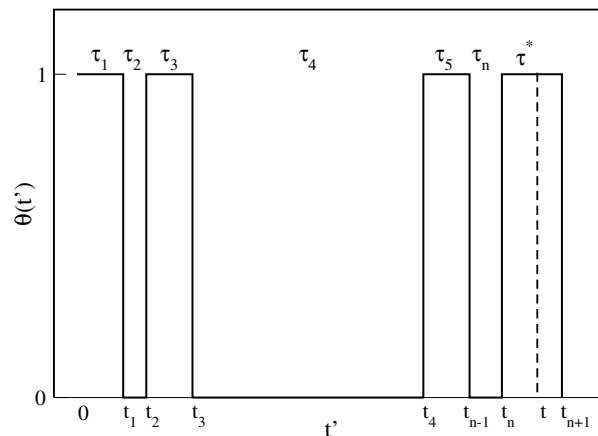


Figure B.1. An example of a two-state renewal process. The function θ is equal to one if the process is in the + state and zero if it is in the - state.

for odd and even n respectively. Hence, summing over k we find

$$\hat{f}_s^+(u) = \left[\hat{\psi}_+(s+u) \frac{1 - \hat{\psi}_-(s)}{s} + \frac{1 - \hat{\psi}_+(s+u)}{s+u} \right] \frac{1}{1 - \hat{\psi}_+(s+u) \hat{\psi}_-(s)}. \quad (76)$$

Following the same methods as were used above, one can easily obtain $\tilde{f}_s^-(u)$ as

$$\hat{f}_s^-(u) = \left[\hat{\psi}_-(s) \frac{1 - \hat{\psi}_+(s+u)}{s+u} + \frac{1 - \hat{\psi}_-(s)}{s} \right] \frac{1}{1 - \hat{\psi}_+(s+u) \hat{\psi}_-(s)}. \quad (77)$$

In the case where the PDFs for sojourn times in the + and - states (namely $\psi_{\pm}(\tau)$) have different asymptotic time dependences (i.e., ψ_+ exponential and ψ_- a power law with a diverging first moment), the PDF of the occupation time will be dominated by the PDF which decays more slowly. The most interesting case is when the $\psi_{\pm}(\tau)$ have the same α , but not necessarily the same constant of proportionality. We consider

$$\psi_{\pm}(\tau) \sim a_{\pm} \tau^{-(1+\alpha)} / |\Gamma(-\alpha)| \quad (\text{for } \tau \rightarrow \infty), \quad (78)$$

where a_{\pm} is a parameter with units $[\tau^{\alpha}]$, and $0 < \alpha < 1$, leading to a diverging mean sojourn time. According to the Tauberian theorem [28] the large t , T_+ behaviour corresponds to the behaviour for small u , s in Laplace space. For the above-mentioned sojourn time PDF, the small u expansion is

$$\hat{\psi}_{\pm}(u) \sim 1 - a_{\pm} u^{\alpha} + \dots \quad (79)$$

Substituting the expansion of $\hat{\psi}_{\pm}(u)$ into the general results obtained in equations (76), (77) and taking the limit as u and s go to zero, we find the asymptotic behaviour of the PDF of occupation times:

$$\hat{f}_s^{\pm}(u) \sim \frac{1}{s} \frac{R \left(1 + \frac{u}{s}\right)^{\alpha-1} + 1}{R \left(1 + \frac{u}{s}\right)^{\alpha} + 1} \quad u, s \rightarrow 0 \quad (80)$$

where

$$R \equiv a_+ / a_-. \quad (81)$$

Note that the asymptotic behaviour of $\hat{f}_s^\pm(u)$ is independent of the initial state (\pm); thus we omit it hereafter. One can verify the normalization of $f_t(T_+)$ by checking that $\hat{f}_s(0) = \frac{1}{s}$; moreover one can see that in the case $\alpha = 1$, i.e., when the mean sojourn times are finite, ergodicity is recovered, or, in other words, the PDF converges to a single value which is equal to the ensemble average

$$f_t(T_+) |_{\alpha=1} = \delta\left(T_+ - t \frac{\langle \tau_+ \rangle}{\langle \tau_+ \rangle + \langle \tau_- \rangle}\right). \quad (82)$$

In the last equation we used the fact that in the case $\alpha = 1$, $a_\pm = \langle \tau_\pm \rangle$, where $\langle \tau_\pm \rangle$ is the average waiting time in the \pm state respectively. Note that the average occupation time of each state, for any α , is equal to $\langle T_\pm \rangle = t \frac{a_\pm}{a_+ + a_-}$ which is independent of α , i.e., in the ergodic case, the PDF converges to a delta function on its mean value.

In order to invert the double LT of the PDF in equation (80), we use the following technique introduced by Godreche and Luck [27]. If for the double LT the following scaling:

$$\hat{f}_s(u) = \frac{1}{s} g\left(\frac{u}{s}\right) \quad (83)$$

as $u, s \rightarrow 0$ is valid, then the asymptotic behaviour of $f_t(\bar{p}_t^+ = \frac{T_+}{t})$ as $t, T_+ \rightarrow \infty$ is given by

$$f_t(\bar{p}_t^+) = -\frac{1}{\pi x} \lim_{\varepsilon \rightarrow 0} \text{Im} \left[g\left(-\frac{1}{x + i\varepsilon}\right) \right] \Big|_{x=\bar{p}_t^+}. \quad (84)$$

In our case, according to equation (80),

$$g\left(\frac{u}{s}\right) = \frac{R \left(1 + \frac{u}{s}\right)^{\alpha-1} + 1}{R \left(1 + \frac{u}{s}\right)^\alpha + 1}, \quad (85)$$

and the PDF of the fractional occupation time is

$$f_t(\bar{p}_t^+) = \delta_\alpha(R, \bar{p}_t^+) \quad (86)$$

where

$$\delta_\alpha(R, x) = \frac{\sin[\alpha\pi]}{\pi} \frac{Rx^{\alpha-1} [1-x]^{\alpha-1}}{R^2 [1-x]^{2\alpha} + x^{2\alpha} + 2R [1-x]^\alpha x^\alpha \cos(\alpha\pi)}. \quad (87)$$

This PDF was obtained by Lamperti [20] using different methods, and the case for $R = 1$ was obtained recently by GL [27].

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