

Closed-Form Solutions for Continuous Time Random Walks on Finite Chains

Ophir Flomenbom and Joseph Klafter

School of Chemistry, Raymond & Beverly Sackler Faculty of Exact Sciences, Tel Aviv University, Ramat Aviv, Tel Aviv 69978, Israel
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Continuous time random walks (CTRWs) on finite arbitrarily inhomogeneous chains are studied. By introducing a technique of counting all possible trajectories, we derive closed-form solutions in Laplace space for the Green's function (propagator) and for the first passage time probability density function (PDF) for nearest neighbor CTRWs in terms of the input waiting time PDFs. These solutions are also the Laplace space solutions of the generalized master equation. Moreover, based on our counting technique, we introduce the adaptor function for expressing higher order propagators (joint PDFs of time-position variables) for CTRWs in terms of Green's functions. Using the derived formula, an escape problem from a biased chain is considered.

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One-dimensional stochastic processes [1–18] are widely used to describe dynamics in biological, chemical, and physical systems [19–33]. Quite ubiquitous among these are nearest neighbor hopping processes on finite discrete lattices (chains). Yet, closed-form solutions for the dynamics along arbitrarily inhomogeneous chains have been missing. To fill this gap, we consider here a continuous time random walk (CTRW) on a chain of l distinct states governed by state- and direction-dependent waiting time probability density functions (PDFs): $\psi_{n\pm 1n}(t) = \omega_{n\pm 1n} \varphi_{n\pm 1n}(t)$ for transitions between state n to state $n \pm 1$, and $\psi_{Nn}(t) = \omega_{Nn} \varphi_{Nn}(t)$ ($N \equiv l + n$) for irreversible trapping from state n , with the normalization conditions $\int_0^\infty \varphi_{jn}(t) dt = 1, \forall n, j = n \pm 1, N$, and $\sum_j \omega_{jn} = 1, \forall n$ [Fig. 1(a)]. CTRWs with direction-independent exponential waiting time PDFs are Markovian, and those with nonexponential or direction-dependent waiting time PDFs are semi-Markovian [1–3,14] (see comment below). One can get semi-Markovian processes from Markovian ones by considering branched Markovian processes, where the states along a branch have the same observable value as the backbone state they have originated from [6], or by averaging over disorder [9].

The dynamics of the CTRW considered here is presented by a stochastic trajectory of l distinct states [Fig. 1(b)] with the renewal property of having no correlations between waiting times [1–3,14]. This property relies on two factors: (i) the waiting times are drawn from the $\varphi_{jn}(t)$ randomly and independently of the global time and of the past transitions [1–3,14], and (ii) each state has a distinct observable value [14]. Semi-Markovian processes generate uncorrelated nonexponential waiting time trajectories [2,14]. Trajectories similar to those shown in Fig. 1(b) are obtained, for instance, from single molecule measurements [14–32], thus allowing the construction of the waiting time PDFs directly from the trajectory. In many cases, the trajectories consist of two observable states [14–18,20,21,25–27,29–32]. In some of these examples [14,29,30(a),31], correlations between waiting times are

observed. This nonrenewal property of the trajectory can appear when assigning the same observable value for different states of the underlying multistate chain in a specific way [14]. Here we are interested in renewal processes, although, for some cases, the statistical properties of nonrenewal ones can also be constructed from the results given in this Letter. The Green's function $G_{nm}(t)$,

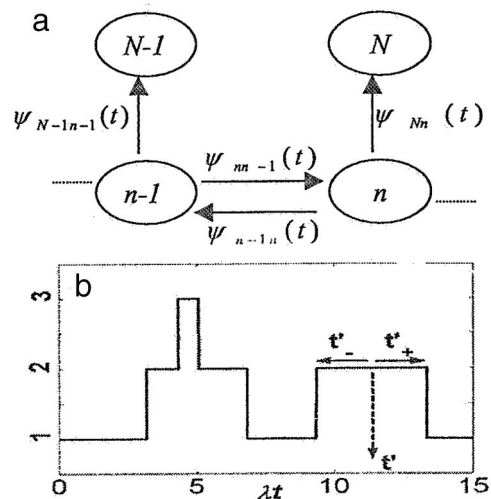


FIG. 1. (a) Part of an l -state semi-Markov chain with nearest neighbor transitions and trapping. The dynamics are governed by the directional waiting time PDFs, $\psi_{n\pm 1n}(t) = \omega_{n\pm 1n} \varphi_{n\pm 1n}(t)$ and $\psi_{Nn}(t) = \omega_{Nn} \varphi_{Nn}(t)$. A way to simulate such a system is to first draw a random number out of a uniform distribution, which determines the direction according to the transition probabilities. Once the direction is set, say, from state n to state j , a time is drawn out of $\varphi_{jn}(t)$, randomly and independently. This Gillespie kind of algorithm produces an uncorrelated waiting time l -state trajectory. (b) Part of a stochastic trajectory, generated from the algorithm above, with $l = 3$, $\omega_{21} = \omega_{23} = 1$, $\omega_{12} = \omega_{32} = 1/2$, and $\varphi_{jn} = \lambda^2 t e^{-\lambda t}$. t'_- and t'_+ are, respectively, the backward and forward recurrence times when occupying state 2 at time t' and the next transition is to state 1. We call the corresponding waiting time a “junction waiting time.”

which is the PDF to occupy state n at time t when starting from state m at time 0 and $(n, m) \in 1, \dots, l$, obeys for semi-Markovian processes with nearest neighbor transitions the integro-differential generalized master equation (GME),

$$\begin{aligned} \partial G_{nm}(t)/\partial t = & - \int_0^t [K_{n+\ln}(t-t') + K_{n-\ln}(t-t') \\ & + K_{Nn}(t-t')] G_{nm}(t') dt' \\ & + \int_0^t K_{nn-1}(t-t') G_{n-1m}(t') dt' \\ & + \int_0^t K_{nn+1}(t-t') G_{n+1m}(t') dt', \\ n = & 1, \dots, l, \end{aligned} \quad (1)$$

with $K_{10}(t) = K_{ll+1}(t) = 0$, and a delta initial condition ($t \rightarrow 0$), $G_{nm}(t) = \delta_{nm} \delta(t)$. Here, $K_{jn}(t)$ is the so-called memory kernel from state n to state j , and $K_{Nn}(t)$ is the trapping kernel of state n . The relationship between the kernels and the waiting time PDFs is given in Laplace space $[\bar{g}(s) = \int_0^\infty g(t) e^{st} dt]$ by [9,10,22], $\bar{K}_{jn}(s) = \bar{\psi}_{jn}(s)/\bar{\Psi}_n(s)$, where $\bar{\Psi}_n(s) = [1 - \sum_i \bar{\psi}_{in}(s)]/s$. Two special cases of the GME are obtained for (a) exponential and direction-independent $\varphi_{jn}(t)$, $\psi_{jn}(t) = a_{jn} e^{-\lambda_n t}$ (a_{jn} is the transition rate from state n to state j , and $\lambda_n = \sum_j a_{jn}$), and (b) power law and state- and direction-independent $\varphi_{jn}(t)$, $\psi_{jn}(t) \sim a_{jn} t^{-1-\gamma}$; $0 < \gamma < 1$. For case (a), Eq. (1) reduces to the master equation with a sink [11], while for case (b), Eq. (1) becomes a fractional master equation with a sink. When taking the continuum limit of case (b), one gets a fractional Fokker-Planck equation [13] with a sink.

In this Letter, we derive closed-form solutions in Laplace space for the Green's function that obeys Eq. (1), and for the first passage time PDF, given in terms of the state- and direction-dependent waiting time PDFs [34]. The solutions are obtained by counting all possible trajectories, and are applied here to study an escape problem from a biased chain. Moreover, based on our counting technique, we introduce the adaptor function for expressing higher order propagators for CTRWs in terms of the Green's functions. For the particular case of exponential waiting time PDFs, the factorization of higher order propagators (joint PDFs of time-position variables) into a product of Green's functions, known for Markovian processes, is recovered.

To derive a closed-form expression for the Laplace transform of $G_{nm}(t)$, we start from the integral representation of $G_{nm}(t)$, $G_{nm}(t) = \int_0^t W_{nm}(t-\tau) \Psi_n(\tau) d\tau$. Here, $W_{nm}(t)$ is the PDF to reach state n exactly at time t when starting from state m exactly at time 0, and $\Psi_n(t) = \sum_j \Psi_{jn}(t)$, where $\Psi_{jn}(t) = \int_0^t \psi_{jn}(\tau) d\tau$, is the sticking probability, that is, the probability to arrive at state n before time t and stay there. Using the convolution theorem, we obtain the Laplace space relationship,

$$\bar{G}_{nm}(s) = \bar{W}_{nm}(s) \bar{\Psi}_n(s). \quad (2)$$

To obtain $\bar{G}_{nm}(s)$ we need to calculate only $\bar{W}_{nm}(s)$, since $\bar{\Psi}_n(s)$ is an input function. We first focus on $\bar{W}_{1l}(s)$. The main idea of our calculation is to express $\bar{W}_{1l}(s)$ as a sum over all possible trajectories (again we use the convenient property of a convolution),

$$\bar{W}_{1l}(s) = \bar{\Gamma}_{1l}(s) \sum_{j=0}^{\infty} \bar{\alpha}_l(s, j). \quad (3)$$

Here, $\bar{\Gamma}_{1l}(s)$ is the Laplace transform of the waiting time PDF of the path of direct transitions from state l to state 1. In general, $\bar{\Gamma}_{nm}(s)$ is defined by

$$\bar{\Gamma}_{nm}(s) = \prod_{i=m}^{n\mp 1} \bar{\psi}_{i\pm 1i}(s), \quad n \neq m; \quad \bar{\Gamma}_{mm}(s) = 1, \quad (4)$$

where the upper (lower) sign corresponds to the case $n > m$ ($n < m$). $\bar{\Gamma}_{1l}(s) \bar{\alpha}_l(s, j)$ is the Laplace transform of the waiting time PDF of making j nearest neighbor closed loops when starting at state l and ending at state 1 exactly at time t , and the index l stands for the system size. By nearest neighbor closed loops we mean, for example, a loop made of a transition from state n to state $n+1$ and a back transition from state $n+1$ to state n (not necessarily consecutive transitions). $\bar{\alpha}_l(s, j)$ is obtained by counting all possible continuous trajectories with j nearest neighbor closed loops that started at state l and ended at state 1. Our main objective is to find a general expression for $\bar{\Phi}_l(s)$, which is defined by

$$\bar{\Phi}_l(s) = \left(\sum_{j=0}^{\infty} \bar{\alpha}_l(s, j) \right)^{-1}. \quad (5)$$

We first consider a two-state chain for which $\alpha_2(t, 0) = \delta(t)$, and for $j \geq 1$, $\alpha_2(t, j) = \int_0^t \psi_{21}(t-z) \times [\int_0^z \psi_{12}(z-y) \alpha_2(y, j-1) dy] dz$. This leads to the recursion relation in Laplace space $\bar{\alpha}_2(s, j) = [\bar{\psi}_{12}(s) \bar{\psi}_{21}(s)]^j = \bar{\alpha}_2^j(s, 1)$, and thus to

$$\bar{\Phi}_2(s) = 1 - \bar{\alpha}_2(s, 1).$$

A recursion relation similar to that of $\bar{\alpha}_2(s, j)$ is found for $\bar{\alpha}_3(s, j)$, for a three-state chain, $\bar{\alpha}_3(s, j) = [\bar{\psi}_{21}(s) \bar{\psi}_{12}(s) + \bar{\psi}_{32}(s) \bar{\psi}_{23}(s)]^j = \bar{\alpha}_3^j(s, 1)$, which results in

$$\bar{\Phi}_3(s) = 1 - \bar{\alpha}_3(s, 1).$$

For a four-state chain, the recursion relation for $\bar{\alpha}_4(s, j)$ is more involved. For the first two values of j ($= 0, 1$), $\bar{\alpha}_4(s, 0) = 1$, and $\bar{\alpha}_4(s, 1) = \sum_{i=1}^3 \bar{\psi}_{ii+1}(s) \bar{\psi}_{i+1i}(s)$, as usual. However, $\bar{\alpha}_4(s, 2)$ does not obey the relation $\bar{\alpha}_4(s, 2) = \bar{\alpha}_4^2(s, 1)$, but rather reads $\bar{\alpha}_4(s, 2) = \bar{\alpha}_4^2(s, 1) - \bar{\beta}_4(s)$, where $\bar{\beta}_4(s) = \bar{\psi}_{21}(s) \bar{\psi}_{12}(s) \bar{\psi}_{43}(s) \bar{\psi}_{34}(s)$. The term

$\bar{\Gamma}_{14}(s)\bar{\beta}_4(s)$ is a broken (discontinuous) trajectory. It is the only broken trajectory in $\bar{\Gamma}_{14}(s)\bar{\alpha}_4^2(s, 1)$. This explains the form of $\bar{\alpha}_4(s, 2)$. For $j > 1$ we find $\bar{\alpha}_4(s, j) = \bar{\alpha}_4(s, 1)\bar{\alpha}_4(s, j-1) - \bar{\beta}_4(s)\bar{\alpha}_4(s, j-2)$, which, after some algebra, leads to

$$\bar{\Phi}_4(s) = 1 - [\bar{\alpha}_4(s, 1) - \bar{\beta}_4(s)].$$

By performing similar calculations for longer chains, we find that $\bar{\Phi}_l(s)$ is given by

$$\bar{\Phi}_l(s) = 1 + \sum_{i=1}^{[(l-1)/2, l/2]} (-1)^i \bar{h}_i(s), \quad (6)$$

where $[(l-1)/2, l/2] = l/2$ if l is even and $(l-1)/2$ otherwise [$\bar{\Phi}_l(s)|_{l=1} \equiv 1$], and,

$$\bar{h}_i(s) = \prod_{j=1}^i \sum_{k_j=k_{j-1}+2}^{l-1-2(i-j)} \bar{\psi}_{k_j, k_j+1}(s) \bar{\psi}_{k_j+1, k_j}(s), \quad (7)$$

with $k_0 = -1$. Equations (3)–(7) give $\bar{W}_{1l}(s)$. For the case $(m, n) \neq (1, l)$, $\bar{W}_{nm}(s) = \bar{\Gamma}_{nm}(s)\bar{\Phi}_l^{nm}(s)/\bar{\Phi}_l(s)$. $\bar{\Phi}_l^{nm}(s)$ is given by Eq. (6) but for a chain in which the states n and m and those between them are *excluded*, and if after this “step” a state is left alone at one of the “edges” of the reduced chain, it is excluded as well [with $\bar{\Phi}_l^{ij}(s) = \bar{\Phi}_l^{ji}(s) = 1$ for $i = 2, j = l-1$]. Note that by using $\bar{\Phi}_l^{nm}(s)$, one can define a corresponding $\bar{W}_{1l}^{nm}(s)$. $\bar{W}_{1l}^{nm}(s)$ can then be used to express $\bar{W}_{nm}(s)$. We omit further discussion about this point here. Combining our results, we find that $\bar{G}_{nm}(s)$, which is the Laplace space solution of the GME for any choice of state- and direction-dependent kernels, is given by

$$\bar{G}_{nm}(s) = \bar{\Gamma}_{nm}(s) \frac{\bar{\Phi}_l^{nm}(s)}{\bar{\Phi}_l(s)} \bar{\Psi}_n(s). \quad (8)$$

Equations (4) and (6)–(8) are the main results of this Letter. From Eq. (8) one can obtain the first passage time PDF, $F_{Nm}(t)$, which is the PDF of the trapping times to the trap state N when starting at state m , and contains in it the probability to be trapped at state N . $\bar{F}_{Nm}(s)$ is obtained when replacing in Eq. (8) $\bar{\Psi}_n(s)$ by $\bar{\psi}_{Nn}(s)$,

$$\bar{F}_{Nm}(s) = \bar{\Gamma}_{nm}(s) \frac{\bar{\Phi}_l^{nm}(s)}{\bar{\Phi}_l(s)} \bar{\psi}_{Nn}(s). \quad (9)$$

We turn now to study, as an example, an escape problem from an l -state biased chain with irreversible trapping at each state. The system is characterized by the state- and direction-independent waiting time PDFs, $\psi_{n+\ln}(t) = \psi(t)q$, $\psi_{n-\ln}(t) = \psi(t)p$, $\psi_{Nn}(t) = \psi(t)w$, and $p + q + w = 1$. To calculate the statistical properties of the process, we need to calculate $\bar{\Phi}_l(s)$. $\bar{\Phi}_l(s)$ is found from Eqs. (6) and (7) to be

$$\bar{\Phi}_l(s) = 2(1/2)^l \bar{Z}_{l+1}(s)/\bar{Z}_1(s), \quad (10)$$

where $\bar{Z}_j(s) = [\bar{Q}_+^j(s) - \bar{Q}_-^j(s)]$, and $\bar{Q}_\pm(s) = 1 \pm \sqrt{1 - 4pq\bar{\psi}^2(s)}$. Equation (10), when substituted in Eq. (9), generalizes previous results in Ref. [6]. Using Eq. (10), we calculate the mean first passage time, T , to reach state 0 when starting at state l . We assume that when an irreversible trapping to some state N occurs, the process starts again from state l after some characteristic time t_d . T is defined by

$$T = \vartheta \left(\tau_{l+1} + \tau_0 + t_d \sum_{i=1}^l \tau_{l+i} \right), \quad (11)$$

where $\vartheta = 1/\bar{F}_{0l}(0)$ is the average number of events that occurred until an event that terminated at state 0 occurred, and $\tau_j = \int_0^\infty t F_{jl}(t) dt = -[\partial \bar{F}_{jl}(s)/\partial s]_{s=0}$. Substituting Eq. (10) and $\bar{\Phi}_l^{nl}(s) = \bar{\Phi}_{n-1}(s)$ ($n > 1$) in Eq. (9), we get in the limit of $q \rightarrow 0$,

$$T = t_\psi (cp^{-l} - 1)/w; \quad t_\psi = \int_0^\infty t \psi(t) dt, \quad (12)$$

where $c = 1 + wt_d/t_\psi$. The dependence of T on the chain length l changes from a *linear* one to an *exponential* one as w changes from $w \rightarrow 0$ to $w \rightarrow 1$, meaning that by tuning w one can drastically change the system’s behavior. This simple model can be used to describe the biological activity of RNA polymerase, which has recently been studied on the single molecule level [33].

Finally, we note that by simple combinations of terms given by Eq. (8) one can obtain the Green’s function for a process with a special first event waiting time PDF [2], which is used, for example, when the process has been taking place before the observation started, and the Green’s function for a circular (closed-one-dimensional) chain. Moreover, our technique of counting all possible trajectories makes it clear that to get (in Laplace space of appropriate dimensions) higher order propagators for any semi-Markovian hopping process, the properties of only the “junction waiting time” [Fig. 1(b)] should be specially considered. Thus, we introduce the “adaptor” function

$$A_{n'n}(u, s) = \frac{\bar{\psi}_{n'n}(s) - \bar{\psi}_{n'n}(u)}{u - s},$$

for a transition between state n to state n' . $A_{n'n}(u, s)$ is the double Laplace transform of the joint backward and forward recurrence times PDF [Fig. 1(b)]. Only for the Markovian case, $A_{n'n}(u, s) = \bar{\Psi}_n(s)\bar{\psi}_{n'n}(u)$, leading to the well-known factorization of higher order propagators into a product of Green’s functions. For example, by using the $A_{n\pm ln}(u, s)$, the double Laplace transform of $G_{knm}(\tau, t) \equiv G_{knm}(t + \tau, t|0)$ ($\tau \rightarrow u$ and $t \rightarrow s$), which is the PDF to occupy state k at time $\tau + t$ and state n on time t when starting from state m on time 0, is given for a chain with nearest neighbor transitions by

$$\begin{aligned} \bar{G}_{knm}(u, s) = & \bar{W}_{nm}(s)[A_{n-\ln}(u, s)\bar{G}_{kn-1}(u) \\ & + A_{n+\ln}(u, s)\bar{G}_{kn+1}(u)]. \end{aligned} \quad (13)$$

Higher order propagators are useful, for example, in discriminating between different models describing activities of single molecules [14,18,23,24]. In particular, $G_{knm}(\tau, t)$ for the semi-Markovian process characterized by $\varphi_{jn}(t) \sim t^{-1-\gamma}$, $0 < \gamma < 1$, was given, in the continuum limit, in Ref. [23]. Equation (13), however, can be applied for any choice of the waiting time PDFs, and gives with Eqs. (2)–(9) a detailed characterization of any one-dimensional semi-Markovian hopping process with nearest neighbor transitions.

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- [34] Note that by utilizing the matrix representation of the GME, one can immediately write down a formal expression for the Green's function. However, a closed-form formula for the Green's function in terms of the state- and direction-dependent kernels was not given before using this approach.