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Random walk in a discrete and continuous system with a thin membrane

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Abstract

Random walk in a one-dimensional system with a thin membrane (which is treated as a partially permeable wall with its internal structure being not explicitly involved into our considerations) is discussed for the discrete and continuous time and space variables. The Green's functions of the membrane system for the discrete space variable are obtained using the method of generating function. The Green's functions for the continuous system are obtained from the discrete ones by taking the continuum limit. It is shown that the boundary condition at the membrane, which is commonly used in stationary system (where the flux flowing through the membrane is proportional to the difference of the concentration of the diffusing particle between the membrane surfaces) is appropriate also for the non-stationary system. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

The problem of diffusion in a one-dimensional system with the homogeneity being disturbed at one point (which corresponds to a system with a thin membrane) has many applications in various fields of physics, chemistry and biophysics (see e.g. Refs. [1,2]). The problem is also interesting from the mathematical point of view. Usually, the diffusion in the membrane system is considered within the diffusion

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equation

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} ,$$

where two boundary conditions at a thin membrane are needed (when the membrane is of finite thickness one needs two boundary conditions at each membrane side [3]). Assuming that the flux flowing through the membrane is continuous, we get the first boundary condition but one needs the second one. The internal structure of the membrane is not explicitly involved in our considerations. Instead, we assume that the permeability of the membrane is described by a properly introduced permeability coefficient. For example, it can be defined as a ratio of the total surface of all holes to the membrane surface.

There are two ways to determine the boundary conditions at the membrane from the microscopic models. In the first one, the random walk on the discrete lattice in terms of the discrete Master equations (ME) is considered [4–6], while the second one is based on the diffusion in the phase-space [7]. The boundary conditions for the systems with fully reflecting, fully absorbing or partially absorbing walls have been considered previously [2–7]. In contrast to the membrane system, the diffusion is then investigated only in half-space bounded by the wall. We note that the diffusion in the membrane system is qualitatively different from that in the system with partially absorbing wall, because in the latter one the particle once absorbed has no chance to return to the system.

In our previous paper [8], we have derived the membrane boundary condition applying the second mentioned method i.e. the analysis the phase-space diffusion. Specifically, we modified the procedure based on Fokker–Planck equation earlier developed for the partially absorbing wall [7]. In this paper, we derive the membrane boundary condition using the diffusion discrete model. We start our considerations with the case of membrane system with discrete space and time variables. We note that the discrete diffusion model is interesting by itself as e.g. a model of the systems with the potential barriers of equal height except one. Among others, the model can be applied to the atomic transport across interfaces between the crystals [9,10]. Although the system with a real membrane is rather complicated (due to the membrane internal structure), it can be treated as a finite series of potential barriers [11,12], while the thin membrane can be also modeled by a single one. When the time variable is continuous, the diffusion can be described by the birth–death equations (BDE). To solve these equations, we start our analysis with the ME for the discrete time because the problem of diffusion is often simpler to solve within the ME than BDE. We are able to find the solution of the ME in a simple manner by using the generating function method proposed by Montroll for the defective lattices [13]. In the next step, using the method by Montroll and Weiss [14], we pass from the discrete to continuous time. Then, the ME take the form of BDE and the solutions of former equations become the solutions of the latter ones. In the final step, we obtain the Green's function for the continuous system, from which we deduce the desired boundary condition at the membrane. The derived boundary condition appears to be identical with that found within the phase-space diffusion considerations [8].

2. The discrete time and space variables

At first, we are going to find the ME for the system with a thin membrane. The general form of the equation is

$$P_{n+1}(m) = \sum_{m'} p(m,m')P_n(m') , \tag{1}$$

where $P_n(m)$ is the probability of finding a random walker at site m after n displacements; $p(m,m')$ is the probability to step from m to m' site.

We assume that the walker is permitted to step only to the nearest neighbor sites. Let the membrane be placed between the N and $N+1$ sites and let us assume that the particle, which try to pass from the N to $N+1$ site, do it with probability $(1-q)$, but if it fails (with probability q), it stays at the site N. We call the parameter q as the microscopic reflecting coefficient of the membrane. The particles which appear in the sites different than N and $N+1$ must change their positions in every step. Analogous assumption is taken when the particle moves in opposite direction. Thus, we get the following ME for the considered system

$$P_{n+1}(m) = \frac{1}{2}P_n(m-1) + \frac{1}{2}P_n(m+1) \quad \text{for } m \neq N,\ N+1 , \tag{2a}$$

$$P_{n+1}(N) = \frac{1}{2}P_n(N-1) + \frac{1-q}{2}P_n(N+1) + \frac{q}{2}P_n(N) , \tag{2b}$$

$$P_{n+1}(N+1) = \frac{1-q}{2}P_n(N) + \frac{1}{2}P_n(N+2) + \frac{q}{2}P_n(N+1) . \tag{2c}$$

The solution of the above equations with the initial condition

$$P_0(m, m_0) = \delta_{mm_0}$$

is called the Green's function.

In the standard method of solving of the ME, we first calculate the generating function [13]

$$S(z,m) = \sum_{n=0}^{\infty} z^n P_n(m) ,$$

and then the probability $P_n(m)$ equals

$$P_n(m) = \frac{1}{2\pi i} \oint S(z,m)\frac{\mathrm{d}z}{z^{n+1}} , \tag{3}$$

where the integration path is the circle around $z=0$ in the complex plane. Using the Montroll's method to solve the ME for the defective lattice, we obtain (for details of the calculations see Appendix)

$$P_n^{--}(m,m_0)=\left(\frac{1}{2}\right)^n\left[\binom{n}{\frac{n+m-m_0}{2}}+q\binom{n}{\frac{n+2N-m-m_0+1}{2}}\right.$$
$$-2q(1-q)\sum_{k=0}^{n-2N+m+m_0-2}(2q-1)^k$$
$$\left.\times\binom{n}{\frac{n+2N-m-m_0+k+2}{2}}\right],\quad m,\ m_0\leqslant N\,, \tag{4a}$$

$$P_n^{+-}(m,m_0)=\left(\frac{1}{2}\right)^n(1-q)\left[\binom{n}{\frac{n+m-m_0}{2}}\right.$$
$$\left.+2q\sum_{k=0}^{n-m+m_0-1}(2q-1)^k\binom{n}{\frac{n+m-m_0+k+1}{2}}\right],\quad m\geqslant N+1,\ m_0\leqslant N\,, \tag{4b}$$

where $\binom{n}{p}=n!/p!(n-p)!$ is the binomial; the indices $+$ and $-$ refer to the signs of $(m-N)$ and (m_0-N), respectively. In the above equations, we take into account only those terms where the lower arguments of the binomials are integer. The functions P_n^{++} and P_n^{-+} can be obtained from the functions (4a) and (4b), respectively, by "mirror reflection" of the space axis with respect to the membrane (where the terms $m-m_0$, $2N-m-m_0$ etc. change their sign). So, in this paper, we do not write explicitly the Green's functions for $m_0>N$.

3. The continuous time and discrete space variables

In the limit of the continuous time, one assumes that the moment of time of the random walker hopping is a random variable. Since we are going to find the Green's functions of continuous time from the discrete ones, we use the following relation:

$$P(m,t)=\sum_{n=0}^{\infty}P_n(m)\Psi_n(t)\,, \tag{5}$$

where $P(m,t)$ is the probability of finding the walker in the position m at time t, $\Psi_n(t)$ is the probability that the walker has made exactly n steps during the time t [15]. The last function is related to the probability density $\varphi(t)$ that the walker takes his next step after the time interval t by the relations

$$\Psi_n(t)=\int_0^t\varphi_n(t')\,\mathrm{d}t'\int_{t-t'}^{\infty}\varphi(t'')\,\mathrm{d}t''\,,$$

where

$$\varphi_n(t) = \int_0^t \varphi_{n-1}(t')\varphi(t-t')\,\mathrm{d}t', \qquad \varphi_1(t) = \varphi(t)\,.$$

We can pass from the discrete to continuous time in the ME and in the Green's functions independently from each other.

Let us first consider the ME (1), which gets the following form when the time variable is continuous:

$$\frac{\partial P(m,t)}{\partial t} = \sum_{m'} A_{m,m'} P(m',t)\,.$$

The rates $A_{m,m'}$ are connected with the probabilities $p(m,m')$ from (1) by the relation

$$A_{m,m'} = \frac{p(m,m') - \delta_{m,m'}}{\tau}\,,$$

where τ is the average time between the steps,

$$\tau = \int_0^\infty t\varphi(t)\,\mathrm{d}t\,.$$

Starting from the ME in the form (2a)–(2c) and applying the standard procedure [15], one finds the BDE:

$$\frac{\partial P(m,t)}{\partial t} = a[P(m-1,t) + P(m+1,t) - 2P(m,t)], \quad m \neq N,\ N+1\,, \tag{6a}$$

$$\frac{\partial P(N,t)}{\partial t} = bP(N+1,t) + aP(N-1,t) - (a+b)P(N,t)\,, \tag{6b}$$

$$\frac{\partial P(N+1,t)}{\partial t} = aP(N+2,t) + bP(N,t) - (a+b)P(N+1,t)\,, \tag{6c}$$

where $a = 1/2\tau,\ b = (1-q)a$.

We do not solve the BDE (6) directly but we derive the Green's functions for the continuous time from the Eqs. (4a) and (4b). As shown in the Ref. [15], the Green's functions for the continuous time obtained from the Green's functions for the discrete time (4a) and (4b) are identical with the solutions of the BDE (6) only when the function φ is taken in the form

$$\varphi(t) = \frac{1}{\tau}\exp\left(-\frac{t}{\tau}\right)\,,$$

which corresponds to $\Psi_n(t)$ being the Poisson distribution

$$\Psi_n(t) = \frac{1}{n!}\left(\frac{t}{\tau}\right)^n \exp\left(-\frac{t}{\tau}\right)\,. \tag{7}$$

Using the following formula for the Bessel function with an imaginary argument I_n

$$I_m(u) = \sum_{k=0}^{\infty} \frac{1}{(m+2k)!}\binom{m+2k}{k}\left(\frac{u}{2}\right)^{m+2k}\,,$$

we obtain from (4), (5) and (7), the Green's functions for the case of the continuous time:

$$P_{--}(m,t;m_0) = \exp(-2at)\Bigg[I_{|m-m_0|}(2at) + qI_{2N-m-m_0+1}(2at) \\ -2q(1-q)\sum_{k=0}^{\infty}(2q-1)^k I_{2N-m-m_0+k+2}(2at)\Bigg], \quad m,\ m_0 \leqslant N\,, \tag{8a}$$

$$P_{+-}(m,t;m_0) = \exp(-2at)(1-q)\Bigg[I_{m-m_0}(2at) \\ + 2q\sum_{k=0}^{\infty}(2q-1)^k I_{m-m_0+k+1}(2at)\Bigg], \quad m \geqslant N+1,\ m_0 \leqslant N\,. \tag{8b}$$

Let us note here that when we are going from (4) to (8) the summation is performed over the infinite number of terms because the number of jumps in the finite time interval $[0,t]$ can be arbitrarily large with the probability (7). It is easy to check that the functions (8a) and (8b) solve the Eqs. (6a)–(6c) with the initial condition

$$P(m,0;m_0) = \delta_{m,m_0}\,.$$

4. From the discrete to continuous space variable

To pass from discrete to continuous space variable we apply the standard procedure, where the following relations are assumed [2,6]:

$$x = \varepsilon n, \qquad D = a\varepsilon^2\,,$$

the parameter ε is interpreted as a distance between discrete sites, D is the diffusion coefficient. The Green's functions for the continuous system are obtained in the limit of small ε

$$\frac{1}{\varepsilon}P(m,t;m_0) \underset{\varepsilon\to 0}{\longrightarrow} G(x,t;x_0)\,. \tag{9}$$

For the homogeneous system the Green's function is

$$P_0(m,t;m_0) = \exp(-2at)I_{|m-m_0|}(2at)\,.$$

For small ε (with fixed x) the variables m and $u = 2at$ take very large values. The above function fulfills well-known relation [2]

$$\frac{1}{\varepsilon}\exp(-2at)I_{|m-m_0|}(2at) \underset{\varepsilon\to 0}{\longrightarrow} G_0(x,t;x_0)\,, \tag{10}$$

where G_0 denotes the Green's function for the continuous system without the membrane

$$G_0(x,t;x_0) = \frac{1}{2\sqrt{\pi D t}} \exp\left(-\frac{(x-x_0)^2}{4Dt}\right) . \tag{11}$$

To obtain the limit of small ε for the functions (8a) and (8b), we need to consider the following problem: does the reflecting coefficient q depend on the parameter ε (and what is the function $q = q(\varepsilon)$)? To get the answer, we keep our attention for a moment on the system with the partially absorbing wall, where the Green's functions and boundary conditions at the wall are well-known for discrete [16] as well as for continuous system [7].

For the discrete system the Green's function, which fulfills the following microscopic boundary condition at the partially absorbing wall placed between N and $N+1$ sites (here $m_0 \leqslant N$) [16]:

$$P(N+1,t;m_0) = \alpha P(N,t;m_0) ,$$

is

$$P(m,t;m_0) = \exp(-2at) \Bigg[I_{|m-m_0|}(2at) + \alpha I_{2N-m-m_0+1}(2at) - (1-\alpha^2)\sum_{k=0}^{\infty} \alpha^k I_{2N-m-m_0+k+2}(2at) \Bigg] , \tag{12}$$

where α is the absorbing parameter of the wall, $\alpha \in [0,1]$. The boundary condition at the wall for the continuous system (which is derived from the consideration performed in the phase-space [7]) reads

$$J(x_N,t;x_0) = \lambda G(x_N,t;x_0) ,$$

where J is the diffusive flux $J = -D\partial G/\partial x$. Using the method of Laplace transform we find the Green's function, which obeys the diffusion equation and above boundary condition, in the form

$$G_{--}(x,t;x_0) = \frac{1}{2\sqrt{\pi D t}} \left\{ \exp\left[-\frac{(x-x_0)^2}{4Dt}\right] + \exp\left[-\frac{(2x_N-x-x_0)^2}{4Dt}\right] \right\} - \frac{\lambda}{D} \exp\left[\frac{\lambda(2x_N-x-x_0+\lambda t)}{D}\right] \operatorname{erfc}\left[\frac{2x_N-x-x_0+2\lambda t}{2\sqrt{Dt}}\right] . \tag{13}$$

The function (12) becomes the function (13) in the limit (9) if the following relation is adopted:

$$\alpha(\varepsilon) = \exp\left(-\frac{\varepsilon\lambda}{D}\right) . \tag{14}$$

Now, we return to the membrane system. Guided by the relation (14) we take

$$q(\varepsilon) = \exp\left(-\frac{\varepsilon\lambda}{D}\right) . \tag{15}$$

Since we assume $q(\varepsilon)\in(0,1]$, then $\lambda\geqslant 0$. The parameter λ controls the permeability of the membrane. To sum up the series occurring in the Eqs. (8a) and (8b), we apply the Euler–MacLaurin formula

$$\sum_{k=0}^{\infty} f(k\varepsilon)=\frac{1}{\varepsilon}\int_0^{\infty} f(u)\,\mathrm{d}u-\sum_{k=0}^{\infty}\varepsilon^k\frac{B_{k+1}}{(k+1)!}f^{(k)}(0)\,,$$

where B_k are the Bernoulli numbers. When the limit of small parameter ε is taken, the function f becomes (see (8) and (10))

$$f(u)=\frac{\varepsilon(2q-1)^{u/\varepsilon}}{2\sqrt{\pi Dt}}\exp\left(-\frac{(2x_N-x-x_0+u)^2}{4Dt}\right)\,,$$

where $x_N=\varepsilon N$, $x_0=\varepsilon m_0$, $u=\varepsilon k$.

Applying the formulas

$$\int_0^{\infty}\exp(-\alpha u^2-\beta u)\,\mathrm{d}u=\frac{1}{2}\sqrt{\frac{\pi}{\alpha}}\exp\left(\frac{\beta^2}{4\alpha}\right)\,\mathrm{erfc}\left(\frac{\beta}{2\sqrt{\alpha}}\right)\,, \tag{16}$$

$$\frac{\mathrm{d}^k}{\mathrm{d}u^k}\exp(-\alpha u^2-\beta u)=(-\sqrt{\alpha})^k\exp(-\alpha u^2-\beta u)H_k\left[\sqrt{\alpha}\left(u+\frac{\beta}{2\alpha}\right)\right]\,,$$

where $\alpha>0$, H_k denotes the Hermite polynomial

$$H_k(u)=(2u)^k-2^{k-1}\binom{k}{2}u^{k-2}+2^{k-2}(2k-1)!!\binom{k}{4}u^{k-4}-\cdots\,,$$

and erfc is the complementary error function

$$\mathrm{erfc}(u)=\frac{2}{\sqrt{\pi}}\int_u^{\infty}\mathrm{d}\eta\exp(-\eta^2)\,,$$

we obtain

$$\sum_{k=0}^{\infty} f(k\varepsilon)=\exp\left[-\frac{(2x_N-x-x_0+2\varepsilon)^2}{4Dt}\right]\left[\frac{1}{2}\exp(z^2)\,\mathrm{erfc}(z)\right.$$
$$\left.+\frac{1}{\sqrt{\pi}}\sum_{k=0}^{\infty}\left(-\frac{\varepsilon}{2\sqrt{Dt}}\right)^{k+1}\frac{B_{k+1}}{(k+1)!}H_k(z)\right]\,, \tag{17}$$

with

$$z\cong\frac{2x_N-x-x_0+2\lambda t}{2\sqrt{Dt}}\,. \tag{18}$$

It is easy to see that in the series (17) with the argument (18) only the first term remains non-vanishing in the limit $\varepsilon\to 0$. Using the formulas (16) and

$$2q(\varepsilon)-1\cong 1-\frac{2\varepsilon\lambda}{D}+\Theta(\varepsilon^2)\cong\exp\left(-\frac{2\varepsilon\lambda}{D}\right)\,,$$

we obtain from the relation (9) the Green's functions for the continuous membrane system

$$G_{--}(x,t;x_0) = \frac{1}{2\sqrt{\pi D t}}\left\{\exp\left[-\frac{(x-x_0)^2}{4Dt}\right] + \exp\left[-\frac{(2x_N-x-x_0)^2}{4Dt}\right]\right\}$$

$$-\frac{\lambda}{D}\exp\left[\frac{2\lambda(2x_N-x-x_0+2\lambda t)}{D}\right]\operatorname{erfc}\left[\frac{2x_N-x-x_0+4\lambda t}{2\sqrt{Dt}}\right], \tag{19a}$$

$$G_{+-}(x,t;x_0) = \frac{\lambda}{D}\exp\left[\frac{2\lambda(x-x_0+2\lambda t)}{D}\right]\operatorname{erfc}\left[\frac{x-x_0+4\lambda t}{2\sqrt{Dt}}\right]. \tag{19b}$$

These functions fulfill the boundary conditions

$$J_{--}(x_N^-,t;x_0) = \lambda[G(x_N^-,t;x_0) - G(x_N^+,t;x_0)], \tag{20a}$$

$$J_{+-}(x_N^+,t;x_0) = \lambda[G(x_N^-,t;x_0) - G(x_N^+,t;x_0)], \tag{20b}$$

where $J_{ij}(x,t;x_0)$, $i,j=+,-$ denotes the diffusive flux defined by the formula

$$J_{ij}(x,t;x_0) = -D\frac{\partial G_{ij}(x,t;x_0)}{\partial x}.$$

From (20a) and (20b), and we immediately obtain that the flux is continuous at the membrane

$$J_{--}(x_N^-,t;x_0) = J_{+-}(x_N^+,t;x_0). \tag{21}$$

The boundary conditions (20) and (21) coincide with those derived in Ref. [8] by means of the Fokker–Planck equation. Let us note that due to the integral formula

$$C(x,t) = \int G(x,t;x')C(x',0)\,\mathrm{d}x'$$

where $C(x,t)$ is the concentration of the diffusing substance, the boundary conditions can be written as

$$J(x_N,t) = \lambda[C(x_N^-,t) - C(x_N^+,t)], \tag{22}$$

$$J(x_N^-,t) = J(x_N^+,t),$$

where $J(x,t)$ is the flux generated by any initial concentration of the diffusing particles.

5. Final remarks

Although our considerations concern the case of thin membrane, it is easy to see that the Green's functions (19a) and (19b) can be applied also for the system with a "thick" membrane when the relation $J(x_1^-,t)=J(x_2^+,t)$ is assumed (here x_1 and x_2 are the location of the left and right surface of the membrane, respectively). In this case, the function G_{--} is given by (19a), where $x_N=x_1$, while the function G_{+-} equals (19b), with $x_N=x_2$.

The behavior of the membrane permeability coefficient λ, which is measured in the velocity units (m/s) is somewhat unexpected. The Green's function for the system with removed membrane is obtained from the Green's functions (19) in the limit of infinite λ

$$G_{ij}(x,t;x_0) \underset{\lambda\to\infty}{\longrightarrow} G_0(x,t;x_0)\,.$$

Consequently, for the weak selectively membranes, this coefficient takes very large values.

The boundary condition (22) is often used in the stationary transport in the membrane system (e.g. Ref. [17]). This relation is then derived within the phenomenological models based on the non-equilibrium linear thermodynamics as, for example, the Kedem–Katchalsky model [18]. Here, we have shown that this boundary condition can be used also for the non-stationary transport.

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Appendix

We present here details of the calculations leading to Eqs. (4). As shown in the Ref. [13], the generating function for a defective lattice is

$$S(m,z) = U(m,z) + z\sum_{m',m''} U(m-m',z)r(m',m'')S(m'',z)\,, \tag{A.1}$$

where U being the generating function for the homogeneous lattice is

$$U(m,z) = \frac{x^{|m|}}{\sqrt{1-z^2}}, \qquad x = \frac{1-\sqrt{1-z^2}}{z}\,,$$

and $r(m',m'')$ is defined as

$$r(m',m'') \equiv p(m',m'') - p_0(m',m'')\,. \tag{A.2}$$

The probability r fulfills the following condition for all m''

$$\sum_{m'} r(m',m'') = 0\,,$$

which follows from the conservation of the probabilities p and p_0. In the considered case, m' and m'' take values N and $N+1$ only. From (2a)–(2c) and (A.2) it is easy to see that

$$r(N,N+1) = r(N+1,N) = -\frac{q}{2}, \qquad r(N,N) = r(N+1,N+1) = \frac{q}{2}\,,$$

with all remaining values of $r(m',m'')$ equal zero.

By means of these relations, we calculate $S(N,z)$ and $S(N+1,z)$ from (A.1). Then, we obtain after simple calculations

$$\begin{aligned} S_{--}(m,z;m_0) = {} & U(m-m_0,z) + qU(2N-m-m_0+1,z) \\ & -2q(1-q)\sum_{k=0}^{\infty}(2q-1)^k U(2N-m-m_0+k+2,z)\,, \\ & m,m_0 \leqslant N\,, \end{aligned} \tag{A.3}$$

$$\begin{aligned} S_{+-}(m,z;m_0) = {} & (1-q)U(m-m_0,z) + 2q(1-q) \\ & \sum_{k=0}^{\infty}(2q-1)^k U(m-m_0+k+1,z), \quad m_0 \leqslant N,\ m \geqslant N+1\,. \end{aligned} \tag{A.4}$$

Using (3), (A.3), (A.4) and the identity

$$\frac{1}{2\pi i}\oint U(m,z)\frac{dz}{z^{n+1}} = \begin{cases} \left(\frac{1}{2}\right)^n \begin{pmatrix} n \\ \frac{n+m}{2} \end{pmatrix}, & \text{when } |m| \leqslant n \text{ and } (n+m)/2 \text{ is integer}\,, \\ 0, & \text{otherwise}\,, \end{cases}$$

(here m is an integer and n is a natural number) we obtain the Green's functions (4a) and (4b).

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