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Some Properties of a Fractal-Time Continuous-Time Random Walk in the Presence of Traps

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The CTRW has often been applied to problems related to transport in a statistically homogeneous disordered medium, which means that there are no traps or reflecting boundaries to be found in the medium. Two physical applications, one to the migration of photons in a turbid medium and the second to the theory of diffusion-controlled reactions in a random medium, suggest that it might be useful to study properties of the CTRW, particularly as they refer to survival probability in the presence of a trap or a trapping surface. We calculate a number of these properties when the pausing-time density is asymptotically proportional to a stable law, i.e., $\psi(t) \sim T^{\alpha}/t^{\alpha+1}$ as $(t/T) \to \infty$, where $0<\alpha<1$. A forthcoming paper will establish the correspondence between properties of the CTRW and properties of random walkers on a fractal with trapping boundaries.

KEY WORDS: Random walk; surface trapping; laser radiation.

1. INTRODUCTION

The continuous-time random walk[(1)] (CTRW) can be used to furnish approximate solutions to a number of problems relating to transport in a variety of disordered media. A number of analyses of properties of the CTRW, particularly those in which the pausing-time density has no finite moments, have appeared in the literature, motivated by such applications.[(2-7)] In most of these analyses one assumes that the random walk takes place in an unbounded medium. We have recently studied a number of problems related to the diffusion of photons in a turbid medium, suggested by medical applications[(8-11)] of Doppler shift lasers. One of our

This paper is dedicated to Jerry Percus on the occasion of his 65th birthday. May he enjoy many more happy and productive years.

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current investigations relates to the response of fractal media to laser beams to ascertain the effects of anomalous diffusion. A second set of problems motivating the present analysis relates to an elucidation of the kinetics of the reaction $A + B \to B$ with a single B. This has been used to characterize self-segregation effects that may occur in chemical reactions in restricted geometries.[12–14]

As will be shown in a forthcoming article based on scaling arguments and simulated data, one can also find useful approximations for physically interesting quantities for transport on a fractal in the presence of a trap, in terms of the solution to a CTRW on a lattice which is characterized by a pausing-time distribution without finite integer moments (except for the zeroth). These require the solution of a number of problems relating to a CTRW in the presence either of an absorbing point or an absorbing boundary. When there are traps or absorbing boundaries required by the formulation of a physical model, quantities exemplified by the survival probability and the flux into the boundary are of some interest, as well as the concentration profile, which is a useful quantity with or without a boundary. In this paper we present a number of results relating to CTRWs in the presence of specific trapping boundaries. Our analysis is specific for those CTRWs characterized by a pausing-time density of stable law form, i.e., in which the asymptotic form of $\psi(t)$ is $\psi(t) \sim T^{\alpha}/t^{\alpha+1}$, where T is a constant with the dimensions of time, and $0 < \alpha < 1$.

2. THE SINGLE TRAP

We first consider a CTRW in the presence of a single trap which we locate at $\mathbf{r} = \mathbf{0}$ and assume that the random walker is initially at site $\mathbf{r}_0$. We calculate the asymptotic time dependence of the survival probability of the random walker, i.e., the probability that the random walker has not been trapped by time t. This problem is a generalization of classical calculations found in the literature of diffusion theory. Our analysis is motivated by studies of self-segregation phenomena in restricted geometries.[12–14]

In order to carry out the indicated calculation for the CTRW, we first calculate the generating function for the probability that the random walker has not been trapped by step n for a random walk in discrete time. The solution to this problem can be used to make the transition to the survival probability in continuous time. Accordingly, let $f_n(\mathbf{r}_0)$ be the probability that the first passage time from $\mathbf{r}_0$ to the trap is equal to n in discrete time. The survival probability can be expressed in terms of this function as

$$S_n(\mathbf{r}_0) = \sum_{j=n+1}^{\infty} f_n(\mathbf{r}_0) \tag{1}$$

Let $f(\mathbf{r}_0; z)$ be the generating function of the $f_n(\mathbf{r}_0)$ with respect to n. It then follows from Eq. (1) that the generating function for the $S_n(\mathbf{r}_0)$ is

$$S(\mathbf{r}_0; z) = \frac{1 - f(\mathbf{r}_0; z)}{1 - z} = \frac{P(\mathbf{0}; z) - P(\mathbf{r}_0; z)}{(1 - z)\, P(\mathbf{0}; z)} \tag{2}$$

where $P(\mathbf{r}; z)$ is the generating function for the Green's function. If $p(\mathbf{j})$ is the probability to be displaced by $\mathbf{j}$ in a single step, and the $\lambda(\boldsymbol{\theta})$ is the so-called structure function defined by $\lambda(\boldsymbol{\theta}) = \sum_{\mathbf{j}}^{\infty} p(\mathbf{j}) \exp(i\boldsymbol{\theta} \cdot \mathbf{j})$, then it is well known[1] that $P(\mathbf{r}; z)$ can be represented as the multiple integral

$$P(\mathbf{r}; z) = \frac{1}{(2\pi)^D} \int_{-\pi}^{\pi} \cdots \int_{-\pi}^{\pi} \frac{\exp(-i\boldsymbol{\theta} \cdot \mathbf{r})}{1 - z\lambda(\boldsymbol{\theta})}\, d^D\boldsymbol{\theta} \tag{3}$$

Let $\hat{\psi}(s)$ denote the Laplace transform of $\psi(t)$. We may convert $S(\mathbf{r}_0; z)$ into a Laplace transform of the survival probability in continuous time[1] by replacing z by $\hat{\psi}(s)$ in Eq. (3) and multiplying by the factor $[1 - \hat{\psi}(s)]/s$. Denoting this Laplace transform by $\hat{S}(\mathbf{r}_0; s)$ and making the appropriate substitution into Eq. (2), we find

$$\hat{S}(\mathbf{r}_0; s) = \frac{P(\mathbf{0}; \hat{\psi}(s)) - P(\mathbf{r}_0; \hat{\psi}(s))}{sP(\mathbf{0}; \hat{\psi}(s))} \tag{4}$$

This function, together with a Tauberian theorem for Laplace transforms,[15] will be used to find asymptotic properties of the survival probability.

To determine the asymptotic behavior of $S(\mathbf{r}_0, t)$, we must find the behavior of $\hat{S}(\mathbf{r}_0; s)$ in the limit $s \to 0$. We restrict our considerations to symmetric lattice random walks, in which the variance of the single step displacements, denoted by σ^2, is finite. In one dimension it is known[1] that in the small-s limit

$$P(x; s) \sim \frac{\exp\{-(x/\sigma)[2(sT)^\alpha]^{1/2}\}}{\sigma[2(sT)^\alpha]^{1/2}} \tag{5}$$

which implies that for a fixed value of x

$$\hat{S}(x_0; s) \sim \frac{x_0 \sqrt{2}\, T^{\alpha/2}}{\sigma s^{1-\alpha/2}} \tag{6}$$

This result, when converted to the time domain, is equivalent to the asymptotic behavior

$$S(x_0; t) \sim \frac{x_0 \sqrt{2}}{\sigma \Gamma(1 - \alpha/2)} \left(\frac{T}{t}\right)^{\alpha/2} \tag{7}$$

If d_w denotes the anomalous diffusion exponent, so that $\langle x^2 \rangle \sim t^{2/d_w}$ for sufficiently large times, then we know that, in translating from the CTRW to the picture of anomalous diffusion, we have $d_w = 2/\alpha$, which implies that the survival probability decays asymptotically to 0 as $(T/t)^{1/d_w}$. When the parameter α exceeds 1 so that the pausing-time density does have a finite first moment, the survival probability goes asymptotically to 0 as $(T/t)^{1/2}$. The formula in Eq. (7) approaches this type of behavior as $\alpha \to 1$. When $\alpha = 1$ there is a logarithmic correction which will not be calculated here.

The asymptotic formula in Eq. (7) indicates that the parameter α determines the functional form of the asymptotic time dependence of $S(x_0; t)$. In two dimensions α does not play the same crucial role. To see this, we note that $\lim_{s \to 0} P(\mathbf{0}; \hat{\psi}(s)) - P(\mathbf{r}_0; \hat{\psi}(s))$ converges to a function of $\mathbf{r}_0$ (i.e., when the difference is taken, the singularities that appear in the two functions disappear). Further, it is known that

$$P(\mathbf{0}; \hat{\psi}(s)) \sim -\frac{\alpha}{2\pi\sigma^2} \ln(sT) \tag{8}$$

The combination of these results suffices to show that the survival probability in the time domain is asymptotically equal to

$$S(\mathbf{r}_0, t) \sim \frac{2\pi\sigma^2 [P(\mathbf{0}; 1) - P(\mathbf{r}_0; 1)]}{\alpha \ln(t/T)} \tag{9}$$

Thus, in the two-dimensional case, in contrast to one dimension, the functional form of the survival probability is independent of the parameter α, which appears only as a coefficient in Eq. (9). In three or more dimensions, since $P(\mathbf{r}_0; 1)$ is finite independent of $\mathbf{r}_0$, it follows that $S(\mathbf{r}_0, t)$ is asymptotically proportional to a constant, independent of α. This is due to random walkers being able to escape to infinity.

The analysis presented to this point can also be applied to solve a somewhat different problem arising from models of the phenomenon of self-segregation in low-dimensional reacting systems.[12,13] A simplified model that reproduces some qualitative features of such systems can be framed in terms of a single trap surrounded by an initially uniform concentration of diffusing particles. We here argue that because of the symmetry of the assumed system the solution of the backward equation satisfied by $S(\mathbf{r}_0, t)$ is equivalent to the solution of the forward equation for the probability $p(\mathbf{r}, t)$ subject to the initial condition $p(\mathbf{r}, 0) = 1$. In other words, we can replace the initial point $\mathbf{r}_0$ by $\mathbf{r}$ in Eq. (4), in which case we have an expression for the Laplace transform of the probability density $\hat{p}(\mathbf{r}, s)$ for the location of the particle.

This identification of the probability density of the location of the random walker allows us to conclude that when

$$\frac{x}{\sigma} \ll \left(\frac{t}{T}\right)^{\alpha/2} \tag{10}$$

the concentration profile is proportional to $(x/\sigma)(T/t)^{\alpha/2}$ or, equivalently, to $x/t^{1/d_w}$ in a neighborhood of the origin. It is straightforward to calculate corrections to this result taking the exponential form in Eq. (5) into account, but the result itself suffices to allow us to look at another interesting quantity. In some earlier work we derived an expression for the probability density of the distance from the trap to the nearest untrapped particle.[14] When the particles move by ordinary Brownian motion it was shown that the average value of this distance increases at sufficiently long times as $t^{1/4}$. We may use the earlier analysis to show that at short distances the probability density for the nearest neighbor distance L is approximately given by

$$f(L, t) \sim \frac{L}{\sigma}\left(\frac{T}{t}\right)^{\alpha/2} \tag{11}$$

which suggests that the nearest neighbor distance goes like

$$\langle L \rangle \sim \sigma \left(\frac{t}{T}\right)^{\alpha/4} \tag{12}$$

This again approaches the known results in the limits $\alpha \to 0$ and $\alpha \to 1$.

In two dimensions, similar considerations show that the point $\mathbf{r}$ enters the results of the calculation only in the combination $P(\mathbf{0}; 1) - P(\mathbf{r}; 1)$ appearing in Eq. (9), while the time dependence is proportional to $[\ln(t/T)]^{-1}$. We therefore estimate the form of the profile by considering the behavior of the spatially-dependent combination referred to above. Long times in the physical domain correspond to large step numbers in the discrete random walk. The asymptotic probability that the random walker is at $\mathbf{r}$ at step n can be found from the central limit theorem to have the form

$$p_n(r) \sim \frac{1}{2\pi\sigma^2 n} \exp\left(-\frac{r^2}{2n\sigma^2}\right) \tag{13}$$

which, because of cylindrical symmetry, depends only on the radial vector $\mathbf{r}$. A knowledge of the large-n form of the probability allows us to estimate the function $P(\mathbf{0}; 1) - P(\mathbf{r}; 1)$ in the limit of large r $[=(x^2 + y^2)^{1/2}]$ as

$$P(\mathbf{0}; 1) - P(\mathbf{r}; 1) \sim \frac{1}{2\pi\sigma^2} \int_1^\infty \frac{1 - \exp(-r^2/2n\sigma^2)}{n} \, dn$$
$$\sim \frac{1}{2\pi\sigma^2} \int_0^{r^2/(2\sigma^2)} \frac{1 - e^{-v}}{v} \, dv$$
$$\sim \frac{1}{2\pi\sigma^2} \ln\left(\frac{r^2}{2\sigma^2}\right) \sim \frac{1}{\pi\sigma^2} \ln\left(\frac{r}{\sigma}\right) \tag{14}$$

Thus, the approximate form of the concentration profile is

$$p(r, t) \sim \frac{2 \ln(r/\sigma)}{\alpha \ln(t/T)} \tag{15}$$

for $t \gg T$. Notice that the logarithmic profile in Eq. (15) can only strictly be correct for $r \gg \sigma$. It is readily shown that for r/σ very small $p(r, t)$ must be proportional to r^2. The asymptotic form of the profile in Eq. (15) agrees with simulated results. To calculate $\langle L(t) \rangle$, we follow the analysis in ref. 14 to write

$$\langle L(t) \rangle \sim \int_\sigma^\infty \exp\left[-\frac{4\pi}{\alpha \ln(t/T)} \int_0^L r \ln\left(\frac{r}{\sigma}\right) dr \right] dL$$
$$\sim \int_\sigma^\infty \exp\left[-\frac{4\pi L^2 \ln(L/\sigma)}{\alpha \ln(t/T)} \right] dL \tag{16}$$

Notice that the exponent appearing in Eq. (16) should contain a concentration in it to render it dimensionless. This has been arbitrarily set equal to 1. We are interested in the behavior of the integral when $t \gg T$, or correspondingly, when the coefficient of $L^2 \ln(L/\sigma)$ in the exponent in Eq. (16) is small. Let us therefore define the dimensionless coefficient $\varepsilon(t)$ by

$$\varepsilon(t) = 4\pi/[\alpha \ln(t/T)] \tag{17}$$

where, when $t \gg T$, $\varepsilon(t) \ll 1$. The major contribution to the integral in Eq. (16) will come from values of L in which the exponent is approximately $O(1)$. This is equivalent to the regime specified by $L^2 = O[\ln(t/T)]$, or $L \gg \sigma$ when $t \gg T$. To calculate a lowest order approximation to $\langle L(t) \rangle$, we make the substitution $L^2 \ln(L/\sigma) = v^2$ in Eq. (16). This allows us to express L in the approximate form

$$L \sim v/[\ln(v/\sigma)]^{1/2} \tag{18}$$

A more accurate inversion of the equation for L as a function of v includes a correction that is $O[(\ln\ln v)/(\ln v)^{3/2}]$, which goes to 0 in the limit $v \to \infty$. Consequently, we find that to lowest order

$$\langle L(t)\rangle \sim \int_0^\infty \frac{e^{-\varepsilon v^2}}{(\ln v)^{1/2}}\, dv = \frac{1}{[\varepsilon(t)]^{1/2}} \int_0^\infty \frac{e^{-u^2}}{[\ln u - (1/2)\ln \varepsilon(t)]^{1/2}}\, du$$
$$\sim \frac{\sqrt{\pi}}{\{2\varepsilon(t)\ln[1/\varepsilon(t)]\}^{1/2}} \sim \frac{1}{2}\left[\frac{\alpha \ln(t/T)}{\ln\ln(t/T)}\right]^{1/2} \tag{19}$$

Thus, to a good approximation, the nearest neighbor distance from a trap to the nearest untrapped particle will increase like $[\ln(t/T)]^{1/2}$ with a correction that would be difficult to measure without access to data over a very wide range of values of t/T. The exponent α appears only as a coefficient in the result in Eq. (19), playing no role in determining the time dependence of $\langle L(t)\rangle$.[16]

A much simpler problem than the ones discussed requires a calculation of the flux into the trapping point. To find the time dependence of this flux, we note that for the discrete-time random walk the flux into a point due to a random walker originally at $\mathbf{r}_0$ can be identified with the first passage time probability which we have denoted by $f_n(\mathbf{r}_0)$ with the associated generating function[1] $f(\mathbf{r}_0; z) = P(\mathbf{r}_0; z)/P(\mathbf{0}; z)$. It therefore follows that the generating function for the flux from random walkers that are initially uniformly distributed on the lattice is

$$J(z) \equiv \sum_{\mathbf{r}_0}{}' f(\mathbf{r}_0; z) = \frac{1}{P(\mathbf{0}; z)}\left[\frac{1}{1-z} - P(\mathbf{0}; z)\right] \tag{20}$$

where the prime on the summation indicates that there is no contribution from $\mathbf{r}_0 = \mathbf{0}$. One converts this discrete generating function into a Laplace transform for the flux as a function of time by replacing z by $\hat{\psi}(s)$. On considering the stable-law form for $\psi(t)$ as we have earlier, and applying a standard asymptotic analysis, we find that the flux in one and two dimensions behaves at long times as

$$\begin{aligned} &\text{1D:} \quad J(t) \sim \frac{\sigma\sqrt{2}}{\Gamma(\alpha/2)}\left(\frac{T}{t}\right)^{1-\alpha/2} \\ &\text{2D:} \quad J(t) \sim \frac{2\pi\sigma^2}{\Gamma(1+\alpha)}\left(\frac{T}{t}\right)^{1-\alpha} \frac{1}{\ln(t/T)} \end{aligned} \tag{21}$$

When the parameter α is set equal to 1 in the one-dimensional case the expression for the flux has the known asymptotic value $(T/t)^{1/2}$, and in two dimensions it goes to 0 as $1/\ln(t/T)$ in the same limit. In contrast to the result obtained from Eq. (15), the order of the stable law now does appear in a significant way in the expression for the time dependence of the two-dimensional flux. In three or more dimensions the flux is asymptotically proportional to $(T/t)^{1-\alpha}$ for $\alpha < 1$. In contrast, when $\alpha \geqslant 1$, the flux will be independent of time at sufficiently large values of t/T.

3. TRAPPING AT A SURFACE

Our original motivation for studying the problem discussed in this paper was that of developing a model for photon migration in a fractal medium, thereby generalizing the theory used to interpret data from a variety of laser techniques used in both medical(9) and industrial(17) applications. In such applications a laser beam is used to inject photons into a tissue for the purpose of determining physical properties of the tissue of medical interest from measurements of light transmitted through the surface. A much used model is one in which a point beam is inserted into the tissue, and the surface intensity of photons that diffuse through the tissue and subsequently reach the surface is measured either as a function of the distance from the injection point when the beam is continuous(18) or, keeping the measurement point fixed and injecting a pulse of light, as a function of time.(10) Both of these experiments are theoretically capable of providing information about underlying tissue characteristics, provided that the tissue is sufficiently homogeneous. Our original motivation for this investigation as well as one based on scaling arguments applied to simulated data was to anticipate possible differences in observed data that might be attributed to tissue inhomogeneity. One starting point for the study of photon migration in a random medium has been based on the statistical properties of a lattice random walk.(18) The approximations used in solving such a model are basically equivalent to taking a diffusion limit, but the lattice picture gets around some difficulties in specifying the proper boundary conditions. Results obtained from the analysis of the random walk model have been shown to be in good agreement with experimental data obtained at NIH.(18) It is therefore natural to analyze the properties of a CTRW in which the pausing-time densities have a stable-law form in order to understand the effects of photon migration in a medium with significant heterogeneities.

The simplest random walk model of a tissue assumes a flat surface that serves as an interface between a tissue of infinite extent and the air. Let the coordinates of such a system (in three dimensions) be (x, y, z) where $z = 0$ represents the surface of the tissue and $z > 0$ represents the interior. These coordinates will be measured in terms of a scattering length which in the reduction of experimental data is found by curve-fitting to observed results.[17,18] In the following analysis $\mathbf{r} = (x, y, z)$ will be a dimensionless space coordinate. We will further assume that the tissue is of infinite extent, so that the coordinates x and y are allowed to take on all possible values. Retaining the simplest formulation of a model for photon migration, we will assume that the interface at $z = 0$ consists entirely of trapping points. The random walk itself will be assumed to be characterized by symmetric transition probabilities. That this is a useful model for the migration of laser-injected light into tissue is not obvious *a priori*, since it is reasonable to suppose that the scattering of photons is preferentially in the forward direction. Nevertheless, it is justifiable on the basis of the central limit theorem provided that there are a sufficiently large number of scattering events. The agreement between theoretical predictions and experimental data suggests that this is indeed the case, and we will henceforth use the assumption of symmetry.

The structure of the tissue will be represented by a simple cubic lattice, and the migrating photon will be modeled in terms of a single random walker initially at the point $(x, y, z) = (0, 0, z_0)$, where z_0 is a single lattice spacing. Before introducing the CTRW we examine the properties of a random walk in discrete time on such a lattice, subject to the stated initial condition. Since $z = 0$ specifies the absorbing surface, we can find the propagator of such a random walk by the method of images. Let $p_n(\mathbf{r} \mid \mathbf{r}_0)$ be the probability for a lattice random walker in free space to be at $\mathbf{r}$ at step n, given that it was initially at $\mathbf{r}_0$. We will assume that the variance of the displacement in a single step is finite. The probability that the random walker is at $\mathbf{r}$ after making n steps in the presence of the absorbing boundary at $z = 0$ is found by the method of images to be

$$Q_n(x, y, z \mid 0, 0, z_0) = p_n(x, y, z - z_0 \mid 0, 0, z_0) - p_n(x, y, z + z_0 \mid 0, 0, z_0) \tag{22}$$

The free space propagators appearing on the right-hand side of this formula will be approximated by the asymptotic (in the time) form

$$p_n(\mathbf{r} \mid \mathbf{r}_0) = \left(\frac{3}{2\pi n}\right)^{3/2} \exp\left[-\frac{3}{2n}(\mathbf{r} - \mathbf{r}_0)^2\right] \tag{23}$$

which is essentially the diffusion, or central-limit theorem approximation to the propagator of the random walk in discrete time.

Let us first calculate the asymptotic form of the survival probability for a single random walker. Since z_0 is generally small in comparison with physically interesting distances (e.g., in a number of human tissues[20] it is of the order of 0.05 mm, in comparison to distances along the interface that are of the order of 1–2 mm), we may calculate the survival probability as a function of n as

$$S_n \sim \int_{-\infty}^{\infty} dx \int_{-\infty}^{\infty} dy \int_0^{\infty} dz\, Q_n(x, y, z \mid 0, 0, z_0)$$
$$= \left(\frac{6}{\pi n}\right)^{1/2} \int_0^{z_0} \exp\left(-\frac{3z^2}{2n}\right) dz \sim \left(\frac{6}{\pi n}\right)^{1/2} z_0 \exp\left(-\frac{3z_0^2}{2n}\right) \tag{24}$$

This expression for S_n can be used to find the corresponding expression for the Laplace transform of the survival probability through the following transformation:

$$\hat{S}(s) = \frac{1-\hat{\psi}(s)}{s} \sum_{n=0}^{\infty} S_n \hat{\psi}^n(s) \sim \frac{1-\hat{\psi}(s)}{s} \int_0^{\infty} S_n \hat{\psi}^n(s)\, dn$$
$$= z_0 \frac{1-\hat{\psi}(s)}{s} \left[\frac{6}{\ln(1/\hat{\psi})}\right]^{1/2} \exp\left\{-z_0 \left[6 \ln\left(\frac{1}{\hat{\psi}}\right)\right]^{1/2}\right\} \tag{25}$$

Our interest is survival probability at long times, which requires an expansion of $\hat{\psi}(s)$ to lowest nonvanishing order in s, followed by the application of a Tauberian theorem for Laplace transforms.[15] The sequence of these manipulations leads to the asymptotic estimate

$$S(t) \sim \frac{z_0\sqrt{6}}{\Gamma(1-\alpha/2)} \left(\frac{T}{t}\right)^{\alpha/2} \sim \frac{z_0\sqrt{6}}{\Gamma(1-\alpha/2)} \left(\frac{T}{t}\right)^{1/d_w}, \qquad t \gg T \tag{26}$$

and the amount trapped per unit time by the surface at long times is therefore proportional to $(T/t)^{1+\alpha/2} = (T/t)^{1+1/d_w}$. Although the asymptotic behavior of this probability at long times has been computed in the case of a three-dimensional system with a two-dimensional plane interface, it is also readily shown to be valid for a D-dimensional system with a $D-1$ interface.

One further quantity whose properties are important in the analysis of data from laser experiments will be denoted by $\Gamma(\rho, t)$. This function is defined by

$\Gamma(\rho, t)\, d\rho = \text{Prob}\{$The random walker surfaces at a distance between ρ and $\rho + d\rho$ from the entrance point of the beam at time $t\}$

where, in translating from a lattice picture to a continuum, we define ρ to be the distance from the injection point of the beam, i.e., $\rho^2 = x^2 + y^2$. In the interpretation of experimental data $\Gamma(\rho, t)$ can be interpreted as the observed intensity measured on the surface at time t at distance ρ from the injection point, given a pulse injection of light at $t=0$ at $(0, 0, z_0)$. Again, a calculation of this function can be based on the solution of the analogous problem in discrete time. To find the probability that we will denote by $\Gamma_n(\rho)$, we observe that it can be expressed as

$$\Gamma_n(\rho) = \tfrac{1}{6} Q_{n-1}(x, y, z_0 \mid 0, 0, z_0) \tag{27}$$

since, for simplicity, the random walk is chosen to be a nearest-neighbor random walk on a simple cubic lattice. This allows us to identify the probability of making a transition $(x, y, z_0) \to (x, y, 0)$ as being equal to 1/6. To avoid some messy algebra, we will choose a specific form for $\hat{\psi}(s)$ having the desired small-s behavior, i.e.,

$$\hat{\psi}(s) = 1/[1 + (sT)^\alpha] \tag{28}$$

We further denote the structure function of the random walk by $\lambda(\boldsymbol{\theta})$, which for the case of a nearest-neighbor random walk on a simple cubic lattice is $(\cos\theta_1 + \cos\theta_2 + \cos\theta_3)/3$.

The Laplace transform of the probability density for absorption at $(x, y, 0)$ at time t can be found exactly as

$$\begin{aligned}\hat{\Gamma}(\rho; s) &\equiv \sum_{n=1}^{\infty} \Gamma_n(\rho)\, \hat{\psi}^n(s) \\ &= \frac{1}{6(2\pi)^3} \int_{-\pi}^{\pi}\int_{-\pi}^{\pi}\int_{-\pi}^{\pi} \frac{e^{-i(x\theta_1 + y\theta_2)}(1 - e^{-2iz_0\theta_3})}{(sT)^\alpha + 1 - \lambda(\boldsymbol{\theta})}\, d^3\boldsymbol{\theta}\end{aligned} \tag{29}$$

The asymptotic form of $\hat{\Gamma}(\rho; s)$ will be found by first exponentiating the denominator of the integrand using the identity $u^{-1} = \int_0^\infty \exp(-\xi u)\, d\xi$. This step leads to the representation

$$\begin{aligned}\hat{\Gamma}(\rho; s) = \frac{1}{6(2\pi)^3} \int_0^\infty e^{-\xi(sT)^\alpha - \xi}\, d\xi \int_{-\pi}^{\pi}\int_{-\pi}^{\pi}\int_{-\pi}^{\pi} e^{-i(x\theta_1 + y\theta_2)}(1 - e^{-2iz_0\theta_3}) \\ \times \exp\left[\frac{\xi}{3}(\cos\theta_1 + \cos\theta_2 + \cos\theta_3)\right] d^3\theta\end{aligned} \tag{30}$$

The integrals with respect to the θ's can be evaluated in terms of Bessel functions of imaginary argument, leaving us with the final expression

$$\hat{\Gamma}(\rho; s) = \frac{1}{6}\int_0^\infty e^{-\xi(sT)^\alpha - \xi} I_x\left(\frac{\xi}{3}\right) I_y\left(\frac{\xi}{3}\right)\left[I_0\left(\frac{\xi}{3}\right) - I_{2z_0}\left(\frac{\xi}{3}\right)\right] d\xi \tag{31}$$

The asymptotic time dependence of $\Gamma(\rho; t)$ will be determined by the singular terms in the expansion of this integral in the limit $sT \to 0$. Since the integral in Eq. (31) can be regarded as a Laplace transform [the term $(sT)^\alpha$ representing the transform parameter], we can make use of an Abelian theorem[15] to relate the small-sT behavior to the large-ξ behavior of the integrand. In the limit $\xi \to \infty$ we can write

$$e^{-\xi} I_x\left(\frac{\xi}{3}\right) I_y\left(\frac{\xi}{3}\right)\left[I_0\left(\frac{\xi}{3}\right) - I_{2z_0}\left(\frac{\xi}{3}\right)\right] \sim 6\left(\frac{3}{2\pi}\right)^{3/2} \frac{z_0^2}{\xi^{5/2}}\left\{1 - \frac{3f(\rho, z_0)}{8\xi} + \cdots\right\} \tag{32}$$

in which the function $f(\rho, z_0)$ is

$$f(\rho, z_0) = 4\rho^2 + 8z_0^2 - 7 \tag{33}$$

from which it follows that as $sT \to 0$

$$\hat{\Gamma}(\rho; s) \sim \frac{\sqrt{6}}{\pi} z_0^2 \left\{(sT)^{3\alpha/2} + \frac{3f(\rho, z_0)}{20}(sT)^{5\alpha/2} + \cdots\right\} \tag{34}$$

But this implies that in the limit $t/T \to \infty$

$$\begin{aligned} \Gamma(\rho; t) &\sim \frac{\sqrt{6}}{\pi} z_0^2 \left\{\frac{T^{3\alpha/2}}{|\Gamma(-3\alpha/2)|\, t^{3\alpha/2+1}} - \frac{3f(\rho, z_0)}{20} \frac{T^{5\alpha/2}}{|\Gamma(-5\alpha/2)|\, t^{5\alpha/2+1}} \cdots\right\} \\ &\sim \frac{\sqrt{6}}{\pi} z_0^2 \frac{T^{3\alpha/2}}{|\Gamma(-3\alpha/2)|\, t^{3\alpha/2+1}} \left\{1 - \frac{3f(\rho, z_0)}{20} \frac{|\Gamma(-3\alpha/2)|}{|\Gamma(-5\alpha/2)|}\left(\frac{T}{t}\right)^\alpha + \cdots\right\} \\ &\sim \frac{\sqrt{6}}{\pi} z_0^2 \frac{T^{3\alpha/2}}{|\Gamma(-3\alpha/2)|\, t^{3\alpha/2+1}} \exp\left[-\frac{3f(\rho, z_0)}{20} \frac{|\Gamma(-3\alpha/2)|}{|\Gamma(-5\alpha/2)|}\left(\frac{T}{t}\right)^\alpha\right] \end{aligned} \tag{35}$$

which shows that the dependence on ρ is Gaussian to the present order of approximation. Figure 1 shows data generated for $\Gamma(\rho; t)$ as a function of ρ, obtained by using the exact enumeration method[5] for a random walk on a comb structure[21,22] (equivalent to $\alpha = 1.5$). The data points, which obviously fall very close to a straight line, indicate an excellent fit to the Gaussian form in ρ. We have derived Eq. (35) by using the method of images for the discrete-time random walk, followed by a conversion of the results to continuous time. Alternatively, we could have reversed these steps, again arriving at the result in Eq. (35). The point of this remark is that only the second option will be relevant in our discussion of the case of a fractal medium.

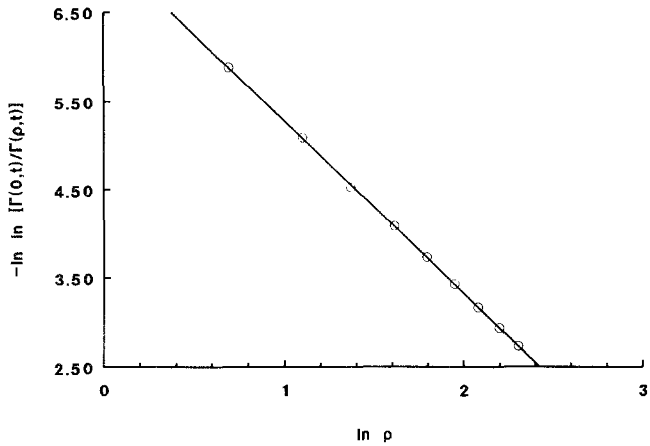

Fig. 1. Data points taken from an exact enumeration calculation of $-\ln\ln[\Gamma(0, t)/\Gamma(\rho; t)]$ for a comb with a trap at one end, plotted as a function of ρ for $t = 5 \times 10^4$ steps. The calculated slope is -1.96, in excellent agreement with the predicted value of -2.

One of the results shown in ref. 18 is that the density of transmitted light through the surface at distance ρ from the entry point of the beam in a steady-state experiment goes like ρ^{-3} for $\rho \gg 1$. This result is also found by integrating the approximation in Eq. (35) over all values of t, and agrees with one's intuition. Although the random walkers which model the beam may spend a long time in the medium, they all eventually exit the surface, and must do so in the same spatial pattern as if they had spent a short time there. One further result that follows from Eq. (35) is an equation for the amount of photons trapped per unit time on the surface. This is defined by

$$\mathscr{T}(t) \equiv 2\pi \int_0^\infty \Gamma(\rho; t)\, \rho\, d\rho \tag{36}$$

The asymptotic time dependence is readily found to be

$$\mathscr{T}(t) \propto t^{-(1+1/d_w)} \tag{37}$$

independent of spatial dimension. In a companion paper we will show how these results are modified for transport in a fractal structure in the presence of an absorbing surface.

In applications of the theory to the analysis of experimental data it is necessary to take into account the effects of internal absorption or scavenging. In the literature related to multiple scattering in human tissue this is usually included phenomenologically in terms of a Beer's law term of the form $\exp(-\mu t)$, which is equivalent to a constant scavenging rate. In the Laplace transform domain the inclusion of Beer's law absorption changes $\hat{\psi}(s)$ to $\hat{\psi}(s+\mu)$ wherever $\hat{\psi}(s)$ appears. Such a change is quite important for the asymptotic analysis. Without absorption, i.e., when $\mu=0$, we can expand $\hat{\psi}(s)$, for small $|s|$, as $\hat{\psi}(s) \sim 1-(sT)^{\alpha}$, whereas when $\mu \neq 0$ the analytic behavior in the neighborhood of $s=0$ is changed to

$$\hat{\psi}(s+\mu) \sim \hat{\psi}(\mu) + s\hat{\psi}'(\mu) \tag{38}$$

in the same limit. Because the transform parameter s only appears to first order in this expression, the principal factor determining the kinetics of survival is the constant absorption. For example, the results derived for the survival probability in Eqs. (7) and (9) or for the flux in Eqs. (21) and (35) are to be multiplied by $e^{-\mu t}$, so that the fractal-time behavior will be effectively masked by the exponential decay.

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