Average volume of the domain visited by randomly injected spherical Brownian particles in d dimensions

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In order to extend the greatly simplified Smoluchowski model for chemical reaction rates it is necessary to incorporate many-body effects. A generalization with this feature is the so-called trapping model in which random walkers move among a uniformly distributed set of traps. The solution of this model requires consideration of the distinct number of sites visited by a single n-step random walk. A recent analysis [H. Larralde *et al.*, Phys. Rev. A **45**, 1728 (1992)] has considered a generalized version of this problem by calculating the average number of distinct sites visited by N n-step random walks. A related continuum analysis is given in [A. M. Berezhkovskii, J. Stat. Phys. **76**, 1089 (1994)]. We consider a slightly different version of the general problem by calculating the average volume of the Wiener sausage generated by Brownian particles generated randomly in time. The analysis shows that two types of behavior are possible: one in which there is strong overlap between the Wiener sausages of the particles, and the second in which the particles are mainly independent of one another. Either one or both of these regimes occur, depending on the dimension. [S1063-651X(96)04706-X]

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I. INTRODUCTION

Perhaps the earliest microscopic model for the kinetics in diffusion-limited reactions is that due to Smoluchowski [1,2]. This is formulated in terms of a concentration of diffusing point particles which move in the presence of a single stationary spherical trapping particle. In the picture suggested by Smoluchowski the chemical reaction is calculated in terms of the rate of encounter of initially uniformly distributed Brownian point particles moving in the presence of a single trapping particle. In the present paper we consider a related problem using a slightly different but essentially identical formulation in which spherical particles of radius b move by Brownian motion in the presence of a single stationary point absorber.

Many generalizations of the considerably oversimplified Smoluchowski model have appeared in the literature of both chemistry and physics. One class of these generalizations attempts to incorporate many-body effects by allowing for a concentration of trapping particles rather than the single trapping particle envisioned by Smoluchowski. In the original Smoluchowski picture reaction was identified with the absorption by a sphere of a diffusing point particles. The consideration of this problem leads, quite naturally, to the study of time-dependent properties of the Wiener sausage which is just the volume swept out by a particle with a fixed point, say the center of the particle, which executes Brownian motion, and in doing so follows a random trajectory W_t. Some mathematical properties of the Wiener sausage in two dimensions were derived as early as 1933 by Leontovich and Kolmogorov [3]. A relation between the kinetics of diffusion-limited reactions and properties of the Wiener sausage is discussed in some detail in Refs. [4] and [5].

A calculation of the complete distribution of the volume of a Wiener sausage poses rather formidable mathematical problems except in one dimension where all calculations can be carried out exactly because the volume can be identified as the span of a diffusion process [6,7]. Calculations of the first moment for all values of the time as well as the asymptotic behavior of the second moment and variance of this random variable is given in [8].

A second related mathematical model that includes many-body effects is the so-called trapping model for a random walk on a lattice which was originally formulated by Dvoretsky and Erdös [9]. A large body of literature on this problem is summarized in [10]. In this class of models a random walker is placed at an arbitrary site on a translationally invariant lattice and one generally attempts to calculate the survival probability of the random walker in the presence of randomly distributed traps. To calculate this probability it is necessary to determine statistical properties of the number of distinct sites visited by the random walker in n steps, since survival, in this model, requires that each of the sites visited during the course of the walk must not have been a trap. The difficulties inherent in mathematical analyses required for the analysis of the volume of the Wiener sausage and for the solution of the trapping problem are essentially identical.

A recent generalization of the trapping problem calculates the expected number of distinct sites visited by N independent n-step random walks, all initially at the same site [11,12] $\langle S_N(n)\rangle$. A continuous version of this problem was analyzed in [13]. The results of that analysis showed a surprisingly rich behavior when considered as functions of both N and n. It was shown that the behavior of $\langle S_N(n)\rangle$ could be characterized as being either collective, in which case there is a considerable overlap of trajectories, or disjoint, in which the overlap effects are negligible. A phenomenon of this sort is to be expected in three or more dimensions where random

walks are transient rather than recurrent [14]. An application of some of these ideas as a model for the kinetics of diffusion-limited reactions is presented in [15]. Some of the results in [11] have recently been applied to develop models of animal foraging [16].

In the present paper we consider the problem analogous to that studied in [11] for a set of Brownian particles injected at the same site at random times t_1, t_1+t_2, $t_1+t_2+t_3,\ldots$. Thus no limit is imposed on the number of particles at any given time. It will prove convenient to introduce the notation

$$\Delta_m=\sum_{j=1}^{m} t_i, \tag{1}$$

so that the time at which particle i is born is Δ_i. We will determine properties of the average volume of a Wiener sausage in any number of dimensions. For simplicity, the probability density for each of the t_i is taken to be the negative exponential

$$\psi(t)=\lambda e^{-\lambda t}, \tag{2}$$

with the corresponding cumulative probability $\Psi(t)=\int_t^\infty \psi(\tau)d\tau=e^{-\lambda t}$. Since, from the point of view of mathematical formalism, the problem of calculating statistical properties of the number of distinct sites visited is nearly identical to that of calculating those for the volume of the Wiener sausage we will use language appropriate to the latter case, understanding that the results apply to both problems. The motivation behind our analysis is that of delineating the regimes in which the behavior can be characterized as either being disjoint or collective.

II. GENERAL FORMALISM

The Brownian particle will be modeled as a sphere with radius b. The volume of the Wiener sausage can be written formally in terms of a single-particle indicator function defined with reference to the Wiener trajectory generated by the center of the sphere W_t. The indicator function will be denoted by $I(\mathbf{r}|W_t)$ defined as

$$I(\mathbf{r}|W_t)=\begin{cases}1 & \text{if } |\mathbf{r}-\mathbf{r}_{W_t}|\leq b\\ 0 & \text{otherwise,}\end{cases} \tag{3}$$

in which case the volume of the Wiener sausage corresponding to the trajectory W_t can be represented as the integral

$$v(W_t)=\int I(\mathbf{r}|W_t)d\mathbf{r}. \tag{4}$$

The average volume of the Wiener sausage is then generated by averaging $v(W_t)$ with respect to all Wiener trajectories. If we denote this average by a set of brackets, $\langle\cdots\rangle$, then the average of the volume can be written as an integral

$$\langle v(W_t)\rangle=\int \langle I(\mathbf{r}|W_t)\rangle d\mathbf{r}=\int q(\mathbf{r}|t)d\mathbf{r}. \tag{5}$$

The function $q(\mathbf{r}|t)$ is the probability that the Wiener sausage has been in contact with the trapping point $\mathbf{r}$ for a total time of t. It is also the fraction of Wiener trajectories that have come within a distance b of the point $\mathbf{r}$ at least once during that time. An equivalent way of phrasing this is to say that it is the probability that a point Brownian particle is trapped by time t by a spherical absorber of radius b centered at the point $\mathbf{r}$.

To calculate this trapping probability one needs to solve the diffusion equation with a single sink term, leading to the formal expression

$$q(\mathbf{r}|t)=H(b-r)+f(\mathbf{r}|t)H(r-b), \tag{6}$$

in which $H(z)$ is the Heaviside step function and in which the function $f(\mathbf{r}|t)$ takes into account particles that remain untrapped by time t by the spherical trap at $\mathbf{r}$. An expression for the function $f(\mathbf{r}|t)$ has been derived in [8] where it is shown that in d dimensions, with $\nu=(d/2)-1$

$$f(\mathbf{r}|t)=\frac{2}{\pi}\left(\frac{b}{r}\right)^{\nu}\int_0^\infty (1-e^{-Dt/b^2z^2}) \times\frac{J_\nu(z)Y_\nu\left(\frac{r}{b}z\right)-J_\nu\left(\frac{r}{b}z\right)Y_\nu(z)}{J_\nu^2(z)+Y_\nu^2(z)}\frac{dz}{z}, \tag{7}$$

where D is the diffusion coefficient, and $J_\nu(z)$ and $Y_\nu(z)$ are Bessel functions of the first and second kinds of order ν. These functions can be reduced to simpler forms in $d=1$ and $d=3$ dimensions:

$$f_1(x|t)=\operatorname{erfc}\left(\frac{|x|-b}{2\sqrt{Dt}}\right),\quad f_3(\mathbf{r}|t)=\frac{b}{r}\operatorname{erfc}\left(\frac{r-b}{2\sqrt{Dt}}\right). \tag{8}$$

On substituting Eq. (7) into Eq. (5) and concurrently making use of Eq. (6) we can write an explicit expression for $\langle v(W_t)\rangle$ for a single particle as

$$\langle v(W_t)\rangle=v_b\left\{1+d\left[(d-2)\frac{Dt}{b^2}H(d-2) +\frac{4}{\pi^2}\int_0^\infty \frac{1-e^{-(Dt/b^2)z^2}}{J_\nu^2(z)+Y_\nu^2(z)}\frac{dz}{z^3}\right]\right\}, \tag{9}$$

where v_b is the volume of a d-dimensional sphere. In one and three dimensions this formula produces the relatively simple results

$$\langle v(W_t)\rangle=\begin{cases}2b+\frac{4}{\sqrt{\pi}}\sqrt{Dt}, & d=1\\ \frac{4\pi}{3}b^3+8b^2\sqrt{\pi Dt}+4\pi bDt, & d=3.\end{cases} \tag{10}$$

The calculation of the average volume for multiple particles can be generalized by introducing a hierarchy of indicator functions [12]. These, in turn, generalize the single indicator function in Eq. (3) but can nevertheless be expressed in terms of such functions. As an example of the simplest such generalization we define a function $I_2[\mathbf{r}|W_t(1),W_t(2)]$ which is equal to 1 if $\mathbf{r}$ is intersected by

or covered by one or both of the Wiener sausages $W_t(1)$ or $W_t(2)$ by time t. In the present context we can, for example, write

$$I_2[\mathbf{r}|W_t(1),W_t(2)]=1-\{1-I[\mathbf{r}|W_t(1)]\}\times\{1-I[\mathbf{r}|W_t(2)]\}, \quad (11)$$

since the two Wiener trajectories are assumed to be independent of one another. The volume of the union of two Wiener sausages can be expressed as the integral

$$v[W_t(1),W_t(2)]=\int I_2[\mathbf{r}|W_t(1),W_t(2)]d\mathbf{r}. \quad (12)$$

Thus the average volume is

$$\langle v[W_t(1),W_t(2)]\rangle=v_b+\int_{r\geq b}\{1-[1-f(\mathbf{r}|t)]^2\}d\mathbf{r} \equiv v_b+\int_{r\geq b}[1-g^2(\mathbf{r}|t)]d\mathbf{r}, \quad (13)$$

where $f(\mathbf{r}|t)$ is the function defined implicitly in Eq. (6) $r=(\mathbf{r}\cdot\mathbf{r})^{1/2}$ and we have introduced the notation $g(\mathbf{r}|t)=1-f(\mathbf{r}|t)$ for the probability that the point $\mathbf{r}$ has *not* been in contact with a single Wiener sausage by time t. The generalization of the preceding definition of the function $I_2[\mathbf{r}|W_t(1),W_t(2)]$ to allow for k trajectories, $I_k[\mathbf{r}|W_t(1),W_t(2),\ldots,W_t(k)]$ follows along the same lines.

In the context of our problem there will be a random number n of Brownian particles at any given time. Hence in the calculation of the average volume we must take an average of $I_n[\mathbf{r}|W_{t-\Delta_1}(1),W_{t-\Delta_2}(2),\ldots,W_{t-\Delta_n}]$ with respect to n.

In general the average Wiener volume can be written as

$$V(t)=v_b(1-e^{-\lambda t})+\int_{r\geq b}[1-\langle g(\mathbf{r}|t-\Delta_1)\times g(\mathbf{r}|t-\Delta_2)\ldots\rangle_{\{\Delta_i\}}]d\mathbf{r}, \quad (14)$$

in which we have denoted the average volume by $V(t)$, v_b represents the volume of each of the particles, and the brackets on the right-hand side indicate an average over the set of all birth times and numbers of particles born before t. Since the number of Brownian particles at any time is a random variable it is necessary to decompose Eq. (14) into a sum of contributions from cases in which there are 1,2,3,... particles in the system at time t. We therefore rewrite Eq. (14) in the form

$$V(t)=v_b(1-e^{-\lambda t})+\int_{r\geq b}\left[1-\sum_{n=0}^{\infty}C_n(\mathbf{r}|t)\right]d\mathbf{r}, \quad (15)$$

in which a formal definition of $C_n(\mathbf{r}|t)$ will be given in Eq. (17) below.

The joint probability that the number of particles at time t is exactly equal to n and that the interbirth times lie in the time intervals (t_1,t_1+dt_1), (t_2,t_2+dt_2),..., (t_n,t_n+dt_n) is equal to

$$\psi(t_1)\psi(t_2)\ldots\psi(t_n)\Psi\left(t-\sum_{i=1}^{n}t_i\right)dt_1dt_2\ldots dt_n =\lambda^n e^{-\lambda t}dt_1dt_2\ldots dt_n. \quad (16)$$

The specification of this probability allows us to express the function $C_n(\mathbf{r}|t)$ that appears in Eq. (15) as

$$C_n(\mathbf{r}|t)=\lambda^n e^{-\lambda t}\int_0^t dt_1\int_0^{t-\Delta_1}dt_2\ldots\int_0^{t-\Delta_{n-1}}\times g(\mathbf{r}|t-\Delta_1)g(\mathbf{r}|t-\Delta_2)\ldots g(\mathbf{r}|t-\Delta_n)dt_n. \quad (17)$$

But this has the form of an n-fold convolution integral which can be evaluated by induction, leading to the result

$$C_n(\mathbf{r}|t)=\frac{\lambda^n}{n!}e^{-\lambda t}\left\{\int_0^t g(\mathbf{r}|\tau)d\tau\right\}^n, \quad (18)$$

so that

$$\begin{aligned}V(t)&=v_b(1-e^{-\lambda t})+\int_{r>b}\left[1-\left\{\exp\left(-\lambda t+\lambda\int_0^t g(\mathbf{r}|\tau)d\tau\right)\right\}\right]d\mathbf{r}\\&=v_b(1-e^{-\lambda t})+\int_{r>b}\left[1-\left\{\exp\left(-\lambda\int_0^t f(\mathbf{r}|\tau)d\tau\right)\right\}\right]d\mathbf{r}\\&=v_b(1-e^{-\lambda t})+\int_{r>b}\{1-\exp[-\lambda F(\mathbf{r}|t)]\}d\mathbf{r}\\&=v_b(1-e^{-\lambda t})+J(t),\end{aligned} \quad (19)$$

where $r=(\mathbf{r}\cdot\mathbf{r})^{1/2}$. In writing Eq. (19) we have used the notation

$$F(\mathbf{r}|t)=\int_0^t f(\mathbf{r}|\tau)d\tau=t\int_0^1 f(\mathbf{r}|t\theta)d\theta=t\mathscr{F}(\mathbf{r}|t). \quad (20)$$

Since $f(\mathbf{r}|t)$ is a probability, the function $F(\mathbf{r}|t)$ increases monotonically to infinity with t. We have therefore decomposed $F(\mathbf{r}|t)$ as indicated, where the function $\mathscr{F}(\mathbf{r}|t)\leq 1$. The function $J(t)$ in the last line of Eq. (19) represents the integral appearing in that equation.

The form of Eqs. (19) and (20) indicates that the calculation of $V(t)$ requires only the solution of a diffusion problem for a single particle and a single trap since this is all that is needed for the calculation of $f(\mathbf{r}|t)$. The solution to that problem is known for Brownian motion in a space of arbitrary number of dimensions and is given in Eq. (9). Equation (20) then indicates that in d dimensions

$$\mathscr{F}(\mathbf{r}|t)=\frac{2}{\pi}\left(\frac{b}{r}\right)^{\nu}\int_0^{\infty}\left[1-\frac{1}{\xi z^2}\,(1-e^{-\xi z^2})\right]\times\frac{J_\nu(z)Y_\nu\left(\frac{r}{b}\,z\right)-J_\nu\left(\frac{r}{b}\,z\right)Y_\nu(z)}{J_\nu^2(z)+Y_\nu^2(z)}\,\frac{dz}{z},\quad(21)$$

in which ξ is a dimensionless time defined as $\xi=Dt/b^2$ and, as before, $\nu=(d/2)-1$. In contrast to the just completed analysis for the calculation of $V(t)$, a calculation of the kth moment of the volume, which will not be discussed in the present paper, requires the solution of a k-trap problem [8]. When d is odd the form of the integrand in Eq. (21) can be simplified because the Bessel functions are then expressible in terms of more elementary functions.

The one- and three-dimensional results can all be expressed in terms of a single function which we will denote by $h(s)$. In one dimension the integral defining $\mathscr{F}(x|t)$ can be evaluated exactly, yielding

$$\mathscr{F}(x|t)=h\left[\sqrt{\frac{1}{4\xi}\left(\frac{|x|}{b}-1\right)^2}\right],\quad(22)$$

where

$$h(s)=(1+2s^2)\mathrm{erfc}(s)-\frac{2se^{-s^2}}{\sqrt{\pi}}\quad(23)$$

is a function that satisfies $h(0)=1$ and decreases monotonically to zero as $s\to\infty$. After performing the integration in three dimensions one finds

$$\mathscr{F}(\mathbf{r}|t)=\frac{b}{r}\,h\left[\sqrt{\frac{1}{4\xi}\left(\frac{r}{b}-1\right)^2}\right]\quad(24)$$

in terms of the same $h(s)$.

A. The short-time regime in all dimensions

When the product $\lambda t\mathscr{F}(\mathbf{r}|t)$ is small $V(t)$ can be approximated as

$$V(t)\approx v_b(1-e^{-\lambda t})+\lambda\int_{r\geq b}d\mathbf{r}\int_0^t f(\mathbf{r}|\tau)\,d\tau.\quad(25)$$

However, the function

$$\int_{r\geq b}d\mathbf{r}\int_0^t f(\mathbf{r}|\tau)\,d\tau=\int_0^t d\tau\int_{r\geq b}f(\mathbf{r}|\tau)\,d\mathbf{r}\quad(26)$$

is equal to $\langle v(W_{t-\tau})\rangle-v_b$ so that in the short-time regime

$$V(t)\approx v_b(1-e^{-\lambda t})+\lambda\int_0^t[\langle v(W_{t-\tau})\rangle-v_b]\,d\tau,\quad(27)$$

which corresponds to a situation in which the Wiener sausages of different particles overlap each other only in a sphere of radius b around the origin. Note that $\lambda d\tau$ is the average number of particles produced in a time τ and $\langle v(W_{t-\tau})\rangle$ is the average volume visited by a single particle in time $t-\tau$. Hence the last term on the right-hand side of Eq. (27) gives the total volume visited at short times. We will see that in high dimensions Eq. (27) will essentially be correct at all times since in such spaces the degree of overlap is negligible.

In the following sections we illustrate the use of the formalism developed in this chapter by deriving explicit results for $V(t)$ in one and three dimensions and examining the long-time behavior of this function.

B. Asymptotics in $d=1$

In $d=1$ $v_b=2b$ and

$$J(t)=2\sqrt{Dt}\int_b^{\infty}[1-\exp(-\lambda t h(y))]dy,\quad(28)$$

where $h(s)$ is given in Eq. (23). The functional form of the behavior of $J(t)$ in the long-time regime defined by $\lambda t\gg 1$ can be found by a simple argument. At such times the integrand of Eq. (28), considered as a function of y, is essentially equal to 1 up to the value of y at which the exponent becomes $O(1)$. Denote the approximate value at which this occurs by $y_*(t)$. The integrand in Eq. (28) goes to 0 over a range of y that is much smaller than $y_*(t)$. These considerations suggest that, to a good approximation,

$$V(t)\approx v_b+2\sqrt{Dt}\,y_*(t).\quad(29)$$

An estimate of $y_*(t)$ can be found by observing that when λt is large $h[y_*(t)]$ must be small in order that the exponent be $O(1)$. This is equivalent to the requirement that $y_*(t)\gg 1$ which allows us to use the approximation

$$h(y)\approx\frac{e^{-y^2}}{2\sqrt{\pi}y^3}.\quad(30)$$

It therefore follows that

$$y_*(t)\approx\sqrt{\ln(\lambda t)}.\quad(31)$$

Hence at long times

$$J(t)\approx v_b+2\sqrt{Dt\,\ln(\lambda t)}.\quad(32)$$

Since λt is the average number of particles injected in time t this functional form is identical to that for the case in which N random walkers are injected simultaneously [11,12]. It is, in principal possible to find the complete probability density function for the volume in one dimension. A full analysis of this is rather complicated, but it can be shown that in the limit of long times this density will tend towards a δ function.

The dependence shown in Eq. (32) defines what will be termed ''collective behavior,'' since a particle will spend most of its time visiting the region already visited by other particles. The term ''individual behavior'' will refer to a regime in which $V(t)$ is approximately equal to a sum of contributions from single particles. We see that in one dimension the individual behavior that occurs at short enough times changes to overlapping behavior at longer times. This is clearly a consequence of the restricted geometry in one

dimension which causes the Wiener sausage to be of one shape only. One might expect on intuitive grounds that individual behavior occurs at all values of the time in sufficiently high dimensions. In the remainder of the paper we examine how the occurrence of individual and overlapping behavior depends on the dimension.

III. ASYMPTOTIC RESULTS FOR DIMENSIONS GREATER THAN ONE

A. General theory

In this section we consider the conditions for individual and overlapping behavior in $d>1$ dimensions, showing that when $d\geqslant 5$ only individual behavior will be observed. When $d\leqslant 4$ both individual and overlapping behavior can occur; in $d=4$ dimensions the possibility of finding both types of behavior will be shown to depend on the rate at which particles are produced. The argument will be seen to hinge on the qualitative behavior of the integral defining $V(t)$ given in Eq. (19).

In d dimensions the long-time behavior of $V(t)$ can be expressed as

$$V(t)=dv_b K_d(t), \tag{33}$$

in which $K_d(t)$ is the integral

$$K_d(t)=\int_0^\infty [1-\exp\{-\lambda t\mathscr{F}_d(\rho|t)\}]\rho^{d-1}d\rho, \tag{34}$$

where the isotropy has allowed us to introduce spherical coordinates and $\rho=r/b$. An exact expression for the function $\mathscr{F}_d(\rho|t)$ can be found as an integral as in Eq. (21). To find the long-time limit of this integral we will adopt the same strategy as in the treatment of $d=1$ by defining the function $\rho_*(t)$ as the solution to $\lambda t\mathscr{F}_d[\rho_*(t)|t]=1$. The value of $K_d(t)$ can then be approximated by

$$K_d(t)\approx\frac{1}{d}[\rho_*(t)]^d. \tag{35}$$

This, however, corresponds to overlapping behavior because Eq. (35) is the result obtained for an expanding d-dimensional sphere. Notice, however, that our argument implies that Eq. (35) is valid provided that the function in square brackets in the integrand in Eq. (34) makes a sharp transition from 1 to 0 for some value of ρ. We will show that in a sufficiently high number of dimensions such behavior cannot occur and in consequence only disjoint behavior is possible.

To examine this question it is necessary to determine the behavior of Eq. (21) as a function of ρ and consider the behavior of Eq. (21) in the limit $\rho\to\infty$. In that limit the principal contribution to the integral will come from $z\ll 1$ since when z is large the oscillatory behavior of the Bessel functions sends the integrand to zero at a rate greater than $1/z$. But when z is small $Y_\nu^2(z)\gg J_\nu^2(z)$ which allows the dropping of terms and a consequent simplification of Eq. (35) to

$$\mathscr{F}(\mathbf{r}|t)\approx\frac{2}{\pi\rho^\nu}\int_0^\infty\left[\frac{1}{\xi z^2}(1-e^{-\xi z^2})-1\right]\frac{J_\nu(\rho z)}{Y_\nu(z)}\frac{dz}{z},$$

$$\rho\to\infty. \tag{36}$$

We further restrict our attention to $d\geqslant 3$ which are the only candidates for the possibility of individual behavior over the whole time regime because of the transience of Brownian motion. Remembering that the principal contribution to the value of $\mathscr{F}(\mathbf{r}|t)$ comes from small z we use the approximation

$$Y_\nu(z)\approx-\frac{\Gamma(\nu)}{\pi}\left(\frac{2}{z}\right)^\nu,\quad \nu>0, \tag{37}$$

which further simplifies Eq. (36) to

$$\mathscr{F}(\mathbf{r}|t)\approx\frac{1}{2^{\nu-1}\Gamma(\nu)\rho^\nu}\int_0^\infty\left[1-\frac{1}{\xi z^2}\times(1-e^{-\xi z^2})\right]z^{\nu-1}J_\nu(\rho z)dz. \tag{38}$$

As shown in the Appendix, the integral can be transformed into a somewhat simpler form, allowing us to write

$$\mathscr{F}(\mathbf{r}|t)\approx\frac{1}{\Gamma(\nu)\rho^{2\nu}}\exp\left(-\frac{\rho^2}{4\xi}\right)\int_0^\infty xe^{-x}\left(x+\frac{\rho^2}{4\xi}\right)^{\nu-2}dx, \tag{39}$$

which will be the basis of the analysis to follow.

In analyzing the implications of Eq. (39) we consider two limits $\rho^2\gg$ or $\ll\xi$ which is equivalent to $r^2\gg$ or $\ll Dt$. In the first of these cases the term in parentheses in the integrand of Eq. (39) is dominated by $\rho^2/(4\xi)$ so that the equation $\lambda t\mathscr{F}(\rho|\xi)=1$ is

$$\frac{\lambda t}{\Gamma(\nu)(4\xi)^{\nu-2}\rho^4}\exp\left(-\frac{\rho^2}{4\xi}\right)=1, \tag{40}$$

which may be solved by iteration. In lowest order we find

$$\rho_*(t)\approx\left\{4\xi\ln\left[\frac{\lambda t}{\Gamma(\nu)(4\xi)^\nu}\right]\right\}^{1/2}. \tag{41}$$

In $d=3$ dimensions $\nu=1/2$ and it is clear that $\rho_*^2(t)\gg 4\xi$ consistent with the assumptions required for the analysis. When $d\geqslant 5$ the parameter ν is greater than 1 so that the approximate solution of the form shown in Eq. (41) no longer conforms to the basic assumption. Thus $d=4$ can be regarded as the critical dimension which agrees with properties of the intersections of random walk trajectories [21]. When $d=4$ the quantity $\rho_*^2(t)/4\xi$ is approximately proportional to $\ln[\lambda b^2/D]$, leading to the conclusion that $\rho_*^2(t)/4\xi$ cannot be large unless the dimensionless production rate of particles is large.

Consistency of the argument leading to Eq. (41) also depends on the transition in the integrand of Eq. (34) being a sharp one. We therefore examine the slope of the integrand of that equation at $\rho=\rho_*(t)$ by determining the characteristic length in ρ over which the transition in Eq. (34) is made

and requiring that it be much less than $\rho_*(t)$. Let $\rho_{\rm trans}(t)$ denote the transition length. To a good approximation this is given by

$$\rho_{\rm trans}(t)\approx\left[-\lambda t\left.\frac{d\mathscr{F}}{d\rho}\right|_{\rho=\rho_*(t)}\right]^{-1}\approx\frac{4\xi}{\rho_*(t)}. \tag{42}$$

We see that the requirement that $\rho_{\rm trans}\ll\rho_*(t)$ coincides with the condition for the validity of Eq. (41). Hence the only condition required for the consistency of the analysis just given is that $\rho_*^2(t)\gg 4\xi$.

B. $d=3$

In three dimensions one finds explicitly that at long times

$$V(t)\approx\frac{32\pi}{3}\left[Dt\,\ln\left(\frac{\lambda t}{\sqrt{4\pi Dt/b^2}}\right)\right]^{3/2}$$
$$=\frac{32\pi}{3}\left[\frac{Dt}{2}\ln\left(\frac{\lambda^2 b^2 t}{4\pi D}\right)\right]^{3/2}. \tag{43}$$

This relation for $V(t)$ describes overlapping behavior when the domain visited by the particles can roughly be described as a sphere of radius $\{4Dt\,\ln[\lambda t/\sqrt{4\pi Dt/b^2}]\}^{1/2}$.

The early time behavior of $V(t)$ can be found from Eq. (27) and is explicitly

$$V(t)\approx v_b(1-e^{-\lambda t})+\tfrac{16}{3}b^2\lambda(\pi Dt^3)^{1/2}+2\pi bD\lambda t^2. \tag{44}$$

This implies that the approximate crossover time at which individual behavior is replaced by overlapping behavior is $D/(\lambda b)^2$. If $\lambda b^2/D\gg 1$ (i.e., the average number of particles injected during the time b^2/D is greater than one) then there are a great number of particles in the system at early times and overlapping behavior will be observed, more or less, over the entire range in time. When this condition is not satisfied one can expect both individual and overlapping behavior. In this case the number $D/(b^2\lambda)$ of injected particles before the crossover time is much greater than 1. This is in contrast with our results for $d=1$ in which the regime in which individual behavior occurs is associated with the condition $\lambda t<1$, i.e., the average number of injected particles is less than 1. Such behavior is obvious on the consideration that the geometry is restricted in one dimension.

As in one dimension, the analytic form of Eq. (43) is the same as is found when N particles are injected initially, no further ones being added [11,12] provided that N is replaced by λt. However, in the random walk problem treated in Refs. [11] and [12] the overlapping behavior is manifested at the earliest times, after which the behavior becomes disjoint. In contrast, in the present model in which particles are injected at different times the early time behavior is disjoint and a transition is made to overlapping behavior at larger t. This is to be expected on the grounds that at early times there will be few particles in the system, while implicit in the work reported on in Refs. [11] and [12] was the assumption $N\gg 1$.

C. $d\geq 4$

In the context of the present problem we have pointed out that there is a kind of phase transition in $d=4$ dimensions as also occurs in the behavior of random walk trajectories. A manifestation of this change is the fact that t drops out of the logarithm in Eq. (41). In this case, in the regime of overlapping behavior we have

$$V_{\rm coll}(t)\propto\left[Dt\,\ln\left(\frac{\lambda b^2}{8D}\right)\right]^2 \tag{45}$$

and when the behavior is disjoint

$$V_{\rm ind}(t)\propto b^2 D\lambda t^2. \tag{46}$$

Hence collective behavior dominates when the condition

$$\frac{\lambda b^2}{D}\gg\left[\ln\left(\frac{\lambda b^2}{D}\right)\right]^2 \tag{47}$$

is fulfilled. Again, in agreement with intuition, collective behavior occurs from the beginning of the process when the injection rates are large. If the condition in Eq. (47) does not hold then only disjoint behavior will be observed over the entire time span.

If $d\geq 5$, or equivalently $\nu\geq 3/2$, we can no longer use the approximate solution in Eq. (41) since Eq. (40) requires that $\rho^2\gg 4\xi$. When $\rho^2\ll 4\xi$ we can drop the exponential term in Eq. (39) as well as the factor $\rho^2/(4\xi)$. The resulting equation is readily solved, leading to the estimate

$$\rho_*(t)\approx(\lambda t)^{1/(2\nu)}. \tag{48}$$

However, it is also easy to verify that the transition layer of the integrand in Eq. (34) is also of the order of $\rho_*(t)$ which implies, in turn, that overlapping behavior attributable to the occurrence of such a transition layer cannot occur in dimensions greater than five.

IV. CONCLUDING REMARKS

We have considered qualitative characteristics of the time-dependent behavior of the volume of the union of Wiener sausages associated with Brownian particles injected at random times into a translationally invariant medium at a single site. This is similar to the problem treated in Refs. [11] and [12], in which all of the particles appear in the system at the same time, but differs from it in that at any given time there can be an arbitrary number of particles performing Brownian motion. As in the mathematical development of those references, the resulting system exhibits a complex behavior when considered as a function of time. The results in both analyses can be described in terms of a progression from individual to overlapping behavior or vice versa. However, the critical dimension which defines a boundary between these two types of behavior differs in the two model types. In Refs. [11] and [12] individual behavior first occurred at long times in $d=3$ dimensions while in the present case $d=4$ is a critical dimension, and for $d\geq 5$ only individual behavior is shown to occur over the entire range in time. We expect that the same qualitative behavior will occur when the injection sites are different, but localized in an

appropriate sense, but this generalization has not been explored by us. A further intriguing, and as yet wholly unexplored area, relates to many-particle diffusion in noninteger dimensions, e.g., in fractals.

A fundamental difference between the problem treated in the present paper and the model in which all of the particles are injected simultaneously is the occurrence of different time scales that are defined by the injection rate $T_i=\lambda^{-1}$ and the radius of the sphere defining the Wiener sausage $T_b=b^2/D$. In one and two dimensions only the first of these time scales plays any role because in low dimensions Brownian motion is recurrent. Our detailed analysis shows that in one dimension disjoint behavior occurs when $t<T_i$ and overlap effects manifest themselves when $t>T_i$. This occurs also when $d=2$ because of transience although we have not presented a detailed analysis of that case.

The second time scale becomes important in determining the crossover time between the two qualitative behavior types in three dimensions. In three dimensions when $T_b \gg T_i$ or equivalently when particles are injected very quickly, the system will exhibit overlapping behavior for all times except for a brief initial period during which the behavior is disjoint. When $T_i \gg T_b$ the behavior is disjoint initially, while overlapping behavior occurs after a crossover time $T_x=D/(b\lambda)^2$, which is large compared to T_i. In consequence the number of particles injected by T_x, $D/(b^2\lambda)$ is much greater than 1 and the average volume visited $2\pi b^3[D/(b^2\lambda)]^3$ is much greater than v_b. Thus in three dimensions overlapping behavior always occurs at sufficiently long times. When the particle source is weak, disjoint behavior will be observed for a substantial amount of time.

Individual behavior becomes even more pronounced in four dimensions. When $T_i \gg T_b$ individual behavior can be expected to occur over the entire time span. In the contrary case of an intensive injection rate $T_b \gg T_i$ the regime of overlapping behavior occurs over the entire range in time.

Our analysis incorporates the use of a specific form for the probability density of the times between successive injections, i.e., $\psi(t)=\lambda e^{-\lambda t}$ which leads to the general relation given in Eq. (19). We conjecture, but have so far been unable to prove that at sufficiently long times, Eq. (19) remains valid whenever $\psi(t)$ has a finite moment $\langle t\rangle$ with the rate λ replaced by $\langle t\rangle^{-1}$. The situation in the case in which the interinjection times are infinite may also be of some theoretical interest. Other extensions of this work that suggest themselves are to diffusion on fractals and to a calculation of the second moment of the volume as would be useful for more direct applications to the trapping problem [17,18]. A further extension of interest is one in which an external field is applied which bias the particle motion [19].

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APPENDIX

Details of the transformation of Eq. (38) to (39). We transform the integral in Eq. (38) by making use of the integral [20],

$$\int_0^\infty z^{\nu-1}J_\nu(z)dz=2^{\nu-1}\Gamma(\nu) \tag{A1}$$

to put it into the form

$$\mathscr{F}(\mathbf{r}|t)\approx\frac{1}{\rho^{2\nu}}\left[1-\frac{1}{\beta}\,Q(\beta)\right], \tag{A2}$$

where $\beta=\xi/\rho^2=r^2/(Dt)$.

$$Q(\beta)=\frac{1}{2^{\nu-1}\Gamma(\nu)}\int_0^\infty(1-e^{-\beta z^2})z^{\nu-3}J_\nu(z)dz. \tag{A3}$$

This integral is evaluated by first taking the second derivative with respect to β, thereby transforming it into an integral whose value is given in [20]. The second derivative of $Q(\beta)$ is

$$\begin{aligned}Q''(\beta)&=-\frac{1}{2^{\nu-1}\Gamma(\nu)}\int_0^\infty z^{\nu+1}J_\nu(z)e^{-\beta z^2}dz\\&=-\frac{4}{(4\beta)^{\nu+1}}\,e^{-1/4\beta}.\end{aligned} \tag{A4}$$

The value of $Q(\beta)$ will be found by integrating this relation twice noting that $Q(0)=0$ and $Q'(0)=1$ which follows from Eq. (A3). In this way we find that $Q(\beta)$ is

$$\begin{aligned}Q(\beta)=\beta&-\frac{\beta}{\Gamma(\nu)}\int_{1/(4\beta)}^\infty e^{-x}x^{\nu-1}dx\\&+\frac{1}{4\Gamma(\nu)}\int_{1/(4\beta)}^\infty e^{-x}x^{\nu-2}dx.\end{aligned} \tag{A5}$$

When this is substituted into Eq. (A2) and some simple transformations are made, one finds Eq. (39) as asserted.

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