

First Passage Time Problems in Time-Dependent Fields

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This paper discusses the simplest first passage time problems for random walks and diffusion processes on a line segment. When a diffusing particle moves in a time-varying field, use of the adjoint equation does not lead to any simplification in the calculation of moments of the first passage time as is the case for diffusion in a time-invariant field. We show that for a discrete random walk in the presence of a sinusoidally varying field there is a resonant frequency ω^* for which the mean residence time on the line segment is a minimum. It is shown that for a random walk on a line segment of length L the mean residence time goes like L^2 for large L when $\omega \neq \omega^*$, but when $\omega = \omega^*$ the dependence is proportional to L . The results of our simulation are numerical, but can be regarded as exact. Qualitatively similar results are shown to hold for diffusion processes by a perturbation expansion in powers of a dimensionless velocity. These results are extended to higher values of this parameter by a numerical solution of the forward equation.

KEY WORDS: Random walks; diffusion processes; first passage times; residence times; diffusive coherence.

1. INTRODUCTION

There has been considerable recent interest in pulsed-field gel electrophoresis, in particular for the separation of large DNA molecules.⁽¹⁻³⁾ Similar ideas have also arisen with respect to other types of chromatographic processes.⁽⁴⁾ The analysis of such kinds of experiments requires an understanding of both ordinary diffusion and reptation in a time-dependent field. It is relatively straightforward to calculate transport

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properties for such fields provided that the underlying substrate is known to be both infinite and spatially homogeneous. A property closely related to diffusive transport in a medium is the first passage time of a particle to a barrier, which is known as the elution time in the terminology of chromatographic systems. The theory of first passage times is now a classical one whose simplest properties have been worked out in great detail for time-homogeneous Markov processes.^(5,6) A major result in this theory is that the equation adjoint to the evolution equation simplifies the calculation of moments of the first passage time. In one dimension, use of the adjoint equation leads to a solution in closed form for moments of the first passage time for both diffusion processes and nearest neighbor random walks.

In the present paper we calculate some properties of the simplest first passage time problem for both a random walk on a lattice and a diffusion process in a time-dependent field. The analysis will be restricted to the case of a one-dimensional, sinusoidally oscillating field and we will focus on statistical properties of the residence time on a line segment. Our results indicate that the oscillating field can create a form of coherent motion capable of reducing the mean residence time by a significant amount. This coherence effect is accompanied by a corresponding reduction of the variance. A way to quantify the amount of oscillation-induced coherence is through the dependence of the mean residence time on the length L of the interval. We will show that the dependence of the mean residence time is proportional to L^2 for large L when the sinusoidal field is not at a particular frequency, ω^* , that will be termed the resonant frequency. At that frequency the dependence changes to a proportionality to the first power of L , a dependence that is characteristic of a random walk in the presence of a uniform field.

Three techniques for analyzing time-dependent first passage times will be used. The lattice random walk will be studied by the method of exact enumeration,⁽⁷⁾ which allows a simple calculation of many properties of random walks with steps to nearest neighbors only. This method, however, is available only for discrete random walks. In the case of diffusion processes one can convert the partial differential equation together with the appropriate set of boundary conditions into an initial-value problem for an infinite set of ordinary differential equations with time-dependent coefficients. When the amplitude of the oscillatory field is sufficiently small an approximate solution to these equations can be found by means of a perturbation expansion. Finally, we use a numerical solution of the time-inhomogeneous diffusion equation to check qualitative conclusions of our analyses both for the discrete and continuum versions of the problem.

2. THE LATTICE RANDOM WALK

The system to be studied consists of a discrete-time random walk on a line segment $(0, L)$, the points $r=0$ and $r=L$ being absorbing. Our analysis will make use of the forward equations for $p_n(r|r_0)$, which is the probability that the random walker is at lattice site r at step n , given the initial position r_0 . The equations satisfied by these probabilities are given by

$$p_{n+1}(r) = \frac{1}{2}[1 + \varepsilon(n)] p_n(r-1) + \frac{1}{2}[1 - \varepsilon(n)] p_n(r+1) \quad (1)$$

where, for convenience, we have suppressed the dependence on r_0 . These equations are to be solved subject to the boundary conditions $p_n(0) = p_n(L) = 0$. In most of the following analysis we will analyze results for the particular initial condition

$$p_0(r) = 1/(L-1) \quad (2)$$

The qualitative conclusions following from this choice will be seen to agree, with only minor modifications, with those from the more specific initial condition $p_0(r|r_0) = \delta_{r,r_0}$. We will mainly be interested in the survival probability $S(n|r_0)$, defined by

$$S(n|r_0) = \sum_{r=1}^{L-1} p_n(r|r_0) \quad (3)$$

We denote the survival probability by $S(n)$ when it corresponds to the particular initial condition in Eq. (2). The j th moment of the residence time can be expressed in terms of $S(n)$ as

$$\langle n^j \rangle = j \sum_{n=1}^{\infty} n^{j-1} S(n) \quad (4)$$

The function $S(n)$ is easy to compute numerically from the results of the exact enumeration calculation, and, when found, is given exactly within the precision of the computer numerics. Briefly stated, the exact enumeration method associates a register with each site of the lattice. The initial state is one in which the registers corresponding to sites $1, 2, \dots, L-1$ are assigned a value 1 and the registers for sites 0 and L have a 0. The registers for these absorbing points remain unchanged during the course of the random walk. At the first step the 1 in register j is divided into two parts, $\frac{1}{2}[1 - \varepsilon(n)]$ and $\frac{1}{2}[1 + \varepsilon(n)]$, which are assigned, respectively, to registers $j+1$ and $j-1$ for the following step when site j is not surrounded by two absorbing sites. When $j=1$, so that one of the adjacent sites is a trap, $\frac{1}{2}[1 - \varepsilon(n)]$ is added to register 2 (provided that $L > 2$) and the remaining amount is lost. A similar computation is made for register $L-1$.

After the first step the procedure is iterated in exactly the same way, except that one takes the contents of counter j at step n , $C_j(n)$, and adds $\frac{1}{2}[1 + \varepsilon(n)] C_j(n)$ to $C_{j+1}(n+1)$ and $\frac{1}{2}[1 - \varepsilon(n)] C_j(n)$ to $C_{j-1}(n+1)$ for j a strictly interior point. Finally, the function $S(n)$ is written in terms of the $C_j(n)$ as

$$S(n) = C_j(n) \left/ \sum_{j=1}^{L-1} C_j(0) \right. \quad (5)$$

Equation (1) is the evolution equation corresponding to a specific biasing field $\varepsilon(n)$. In what follows we will concentrate on the specific choice

$$\varepsilon(n) = \varepsilon \sin(\omega n) \quad (6)$$

where the parameter ω will be restricted to the interval $(0, \pi)$ to avoid aliasing. The first point addressed in our study is the behavior of the mean residence time, $\langle n(\omega) \rangle$, considered as a function of frequency ω . Typical results for this function for $L = 50, 100,$ and 200 are shown in Fig. 1. The

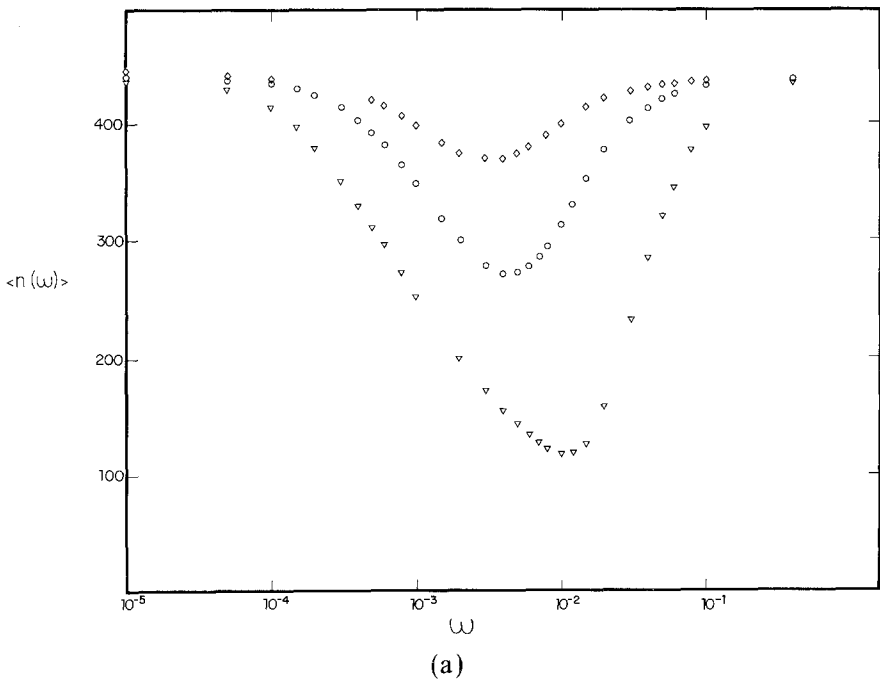
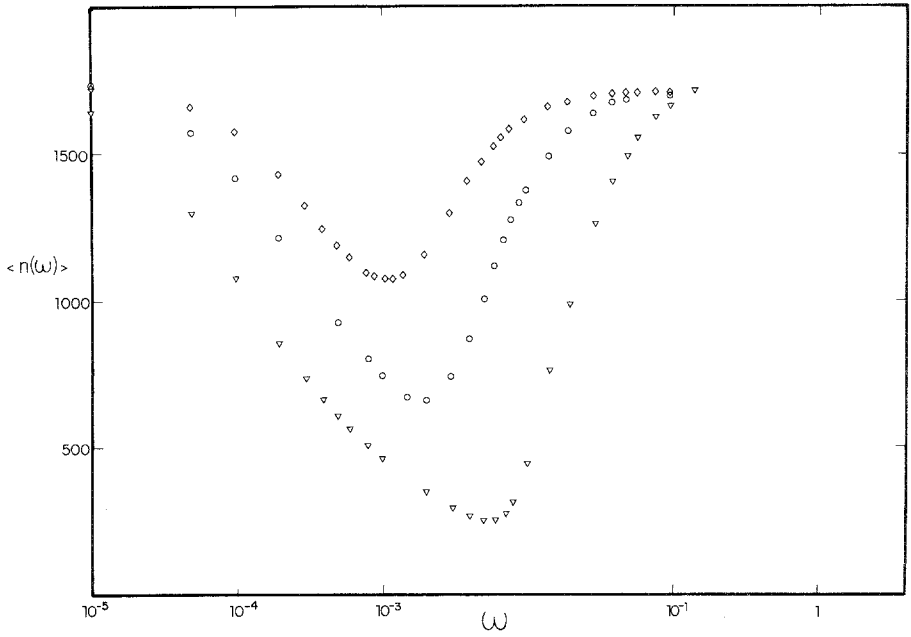
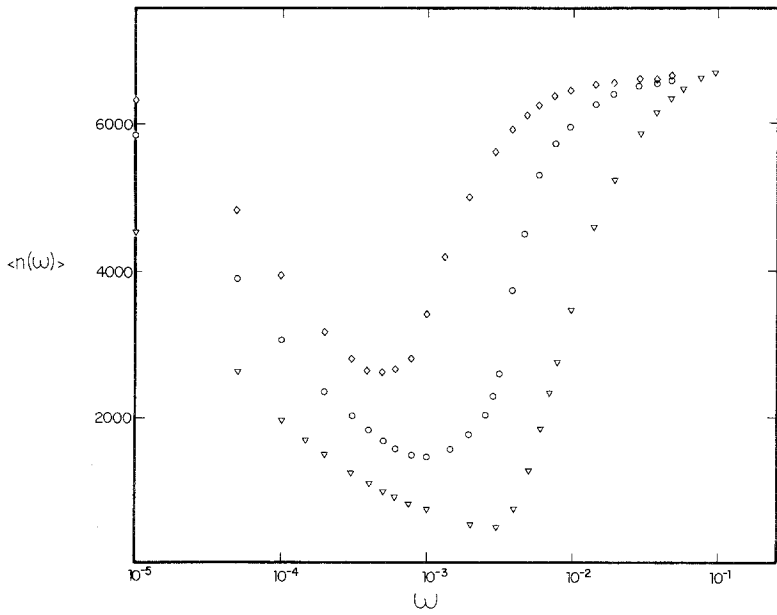


Fig. 1. The mean first passage time, $\langle n(\omega) \rangle$, plotted as a function of ω for a sinusoidal field acting on a random walk on a lattice segment in one dimension. The curves shown are for several values of the amplitude ε : $\varepsilon = 0.05$ (\diamond), $\varepsilon = 0.1$ (\circ), $\varepsilon = 0.3$ (∇). The segment lengths are (a) $L = 50$, (b) $L = 100$, (c) $L = 200$.



(b)



(c)

Fig. 1 (continued)

striking feature of these results is the appearance of a minimum at the resonant frequency ω^* , which depends on L and ε . It is evident that by operating the field at the frequency ω^* one can obtain a substantial reduction in the mean residence time. The reason for the minimum residence time lies in coherent motion induced by the sine function in Eq. (6). A random walker will initially tend to move in the $+r$ direction because $\sin(\omega n)$ is initially positive, thereby enhancing the early absorption at $r=L$ over the unbiased case. When the field reverses, the absorption is enhanced at $r=0$. The process continues in this fashion, but since random walkers near the edges have been depleted, the diffusion mechanism takes on increased importance at later times. When ω is very small, the absorption enhancement must also be small, which leaves the diffusive absorption as a dominant effect over the entire time span. When ω is large, there is an initial spurt of absorption, but after random walkers near the ends of the

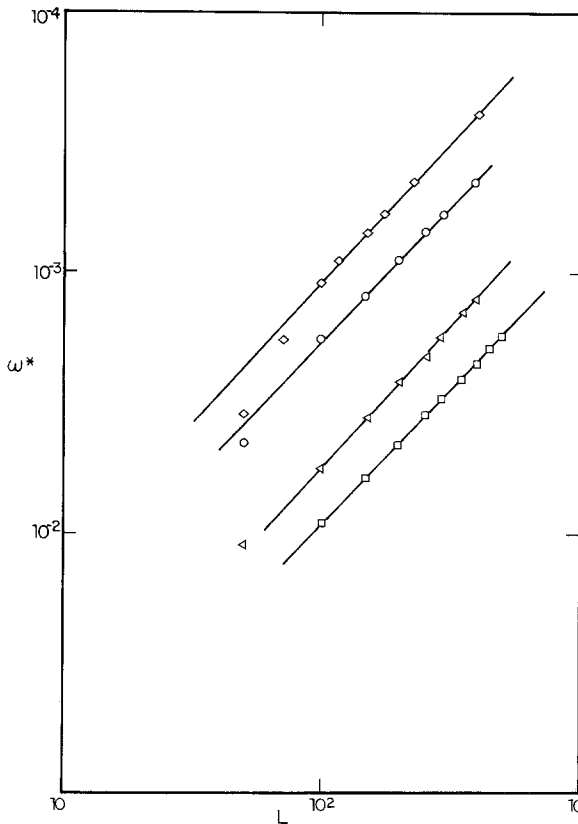


Fig. 2. The resonant frequency, ω^* , plotted as a function of the size of the lattice segment for several values of the amplitude ε : $\varepsilon = 0.05$ (\diamond), 0.1 (\circ), 0.3 (∇), and 0.5 (\square).

interval have been absorbed, the kinetics of the absorption process is again governed by diffusion. It is instructive to examine the behavior of the variance $\sigma^2(n(\omega)) = \langle n^2(\omega) \rangle - \langle n(\omega) \rangle^2$ as a function of ω . This is also seen to have a minimum near ω^* . We have also examined the behavior of the resonant frequency ω^* as a function of L , finding that

$$\omega^*(L) \sim 1/L \tag{7}$$

for large L , as shown in Fig. 2. When $\varepsilon < 0.5$, ω^* is very close to being proportional to ε .

Somewhat more interesting than Eq. (7) is the dependence of $\langle n(L|\omega) \rangle$ as a function of the interval length for fixed frequency. When $\omega = \omega^*$ one finds that

$$\langle n(L|\omega) \rangle \sim L^2 \tag{8}$$

but when $\omega = \omega^*$ the relation changes to a first power dependence:

$$\langle n(L|\omega^*) \rangle \sim L \tag{9}$$

This dependence is illustrated in Fig. 3. Since the decrease in mean residence time is principally due to the time dependence of the average displacement, one expects that an increase in the amplitude of the bias

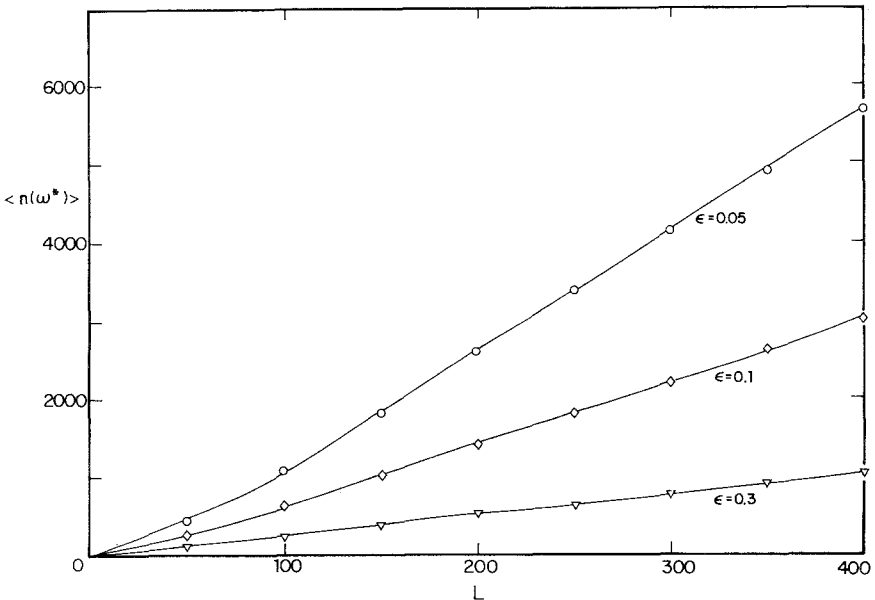


Fig. 3. Dependence of the minimum average residence time $\langle n(\omega^*) \rangle$ on the lattice size, L , for several values of ε ; $\varepsilon = 0.05$ (\circ), 0.1 (\diamond), and 0.3 (∇). The figures indicate that $\langle n(\omega^*) \rangle$ varies as the first power of L when $L \gg 1$.

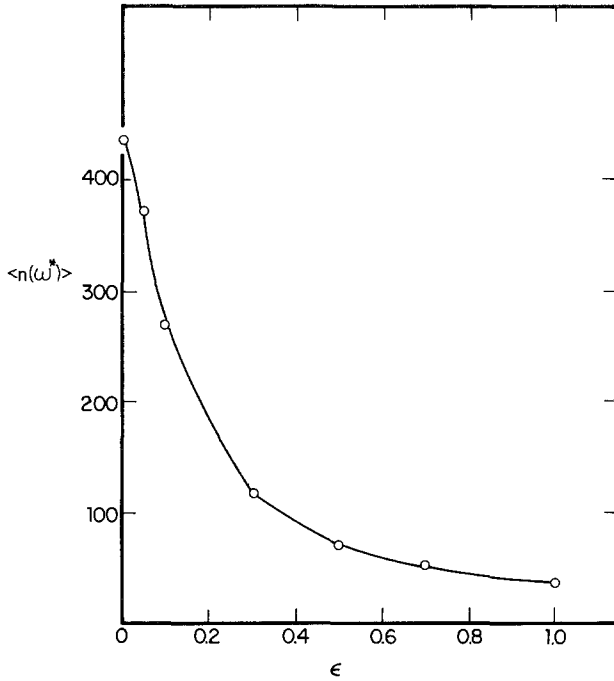


Fig. 4. The mean residence time, $\langle n(\omega^*) \rangle$, as a function of the bias parameter ϵ for a system size, $L = 50$.

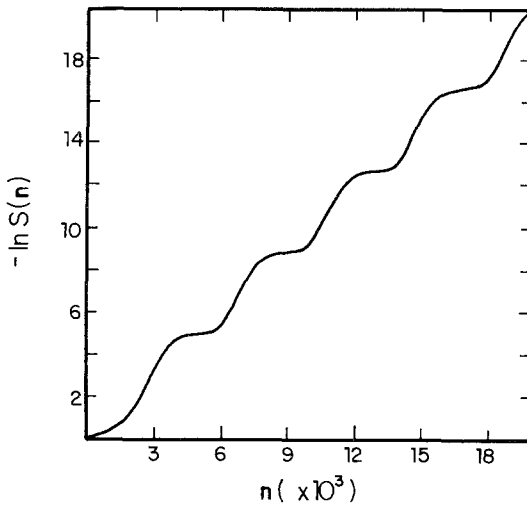


Fig. 5. A typical plot of the survival probability as a function of step number for a lattice random walk, for $\epsilon = 0.3$, $\omega = 8 \times 10^{-4}$, and $L = 200$.

parameter ε will lead to a decrease in the mean residence time. Figure 4 shows the form taken by this function for $L = 50$. A typical plot of $S(n)$ as a function of step number is given in Fig. 5, showing that the oscillatory field is not damped out. Further numerical calculations indicate that the oscillations become increasingly evident as ω approaches ω^* .

A number of points of lesser importance were also investigated. We looked at $\langle n \rangle$ as a function of the initial position of the random walker r_0 , rather than for the uniform initial condition given in Eq. (2). Some results of this are shown in Fig. 6. The curves in Fig. 6a of the mean residence time as a function of initial position demonstrate that in the pure diffusion limit the mean residence time is symmetric with respect to the center of the interval. As the frequency is increased, the location of the maximum is shifted

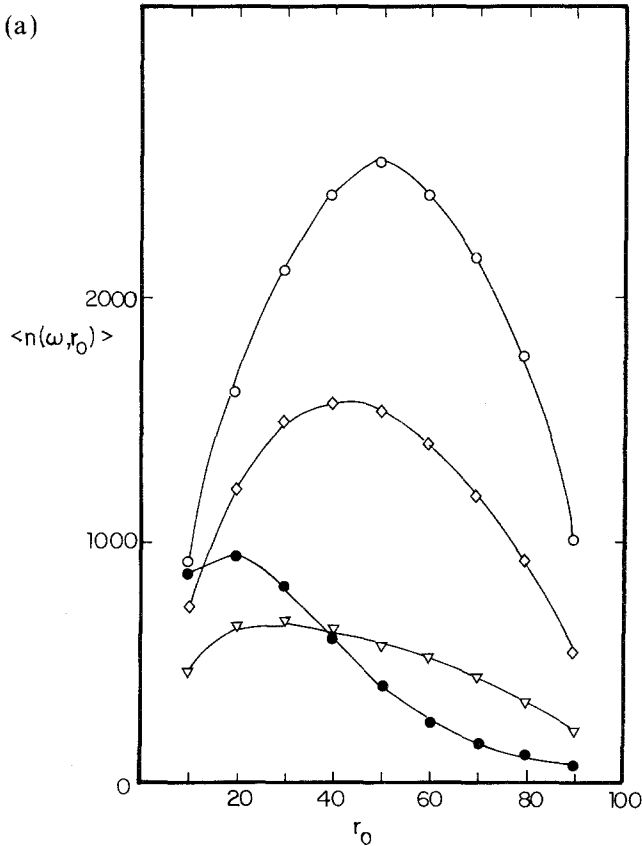


Fig. 6a. A plot of the mean residence time, $\langle n(\omega) \rangle$, as a function of the initial position of a random walker for $\varepsilon = 0.3$, $L = 100$, and several values of ω ; $\omega = 0$ (\circ), 10^{-4} (\diamond), 10^{-3} (∇), and 10^{-2} (\bullet).

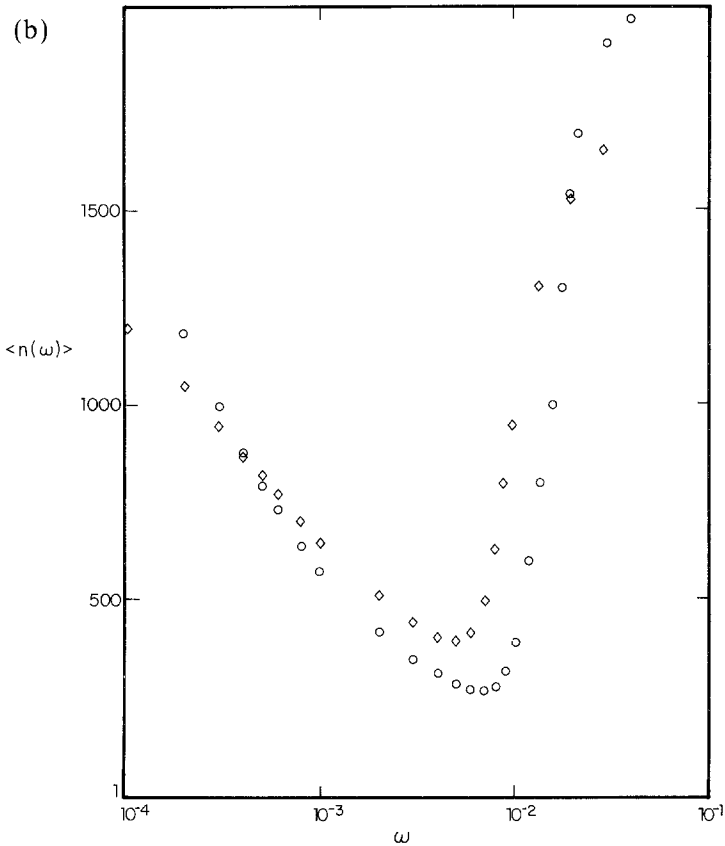


Fig. 6b. The mean residence time plotted as a function of r for two different initial positions; $r_0 = 20$ (\diamond) and 50 (\circ). The remaining parameters are $\varepsilon = 0.3$ and $L = 100$.

further and to the left of the interval. This serves to demonstrate that the initial surge is the most important factor in determining the residence time. Figure 6b shows that the behavior of $\langle n \rangle$ as a function of frequency in the neighborhood of the minimum is only slightly affected by the starting point. The difference becomes more pronounced at higher frequencies.

In order to examine the initial biased motion a little more closely, we considered a generalization of the sinusoid in Eq. (6) by including a phase angle in the sine function,

$$\varepsilon(n) = \sin(n\omega + \varphi) \quad (10)$$

Some results of the calculation are shown in Fig. 7, from which it is evident that the phase is significant in determining the mean residence time at very low frequencies. However, because an increase in frequency switches the transition probabilities at a faster rate, the effect of the initial phase tends

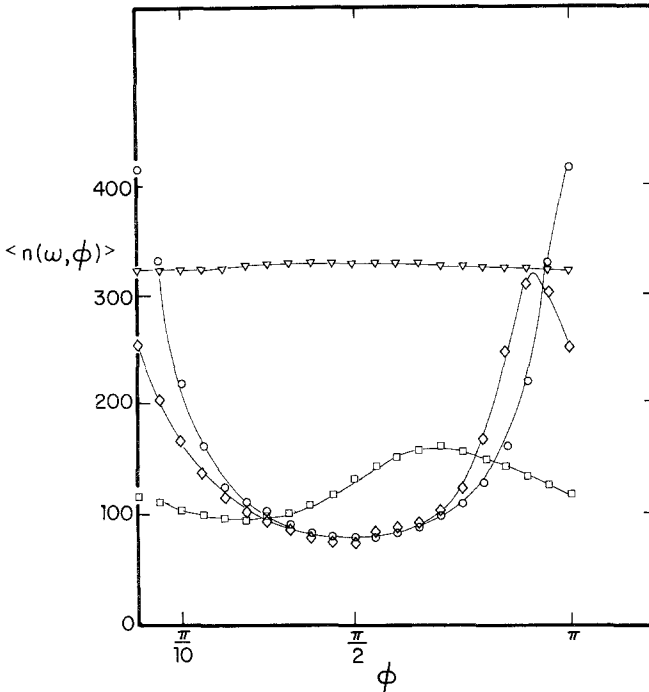


Fig. 7. Graphs of the mean residence time, $\langle n(\omega, \phi) \rangle$, as a function of the phase, ϕ , for different values of ω . The segment length is $L = 50$ and $\varepsilon = 0.3$. The curves shown are for $\omega = 0$ (\circ), $= 10^{-3}$ (\diamond), $= 10^{-2}$ (\square), $= 5 \times 10^{-2}$ (∇). The greatest effects are seen to occur at the lowest values of the frequency, while at high frequencies there is a cancellation effect.

to wash out. It is nevertheless interesting to note that when one averages over ϕ , the coherence effect remains, as is evident from Fig. 8, showing the mean residence time for $\langle \tau(\omega, \phi) \rangle$ averaged over ϕ . The minimum is not nearly as pronounced as in the case $\phi = 0$. Figure 9 shows that when the minimum value of $\langle \tau(\omega^*) \rangle$ is averaged over ϕ it retains its proportionality to the first power of L .

3. DIFFUSION PROCESSES

In this section we analyze some aspects of first passage time problems in a time-dependent field for diffusion in a continuum. Again, we restrict ourselves to one dimension, considering only the case of diffusion on a line segment, whose ends $x = 0$ and $x = L$ are assumed to be absorbing points. The results of this analysis will be found to resemble closely those found in the case of lattice random walks. As in the case of the discrete random walk, we cannot find a readily computable solution to the problem, but it

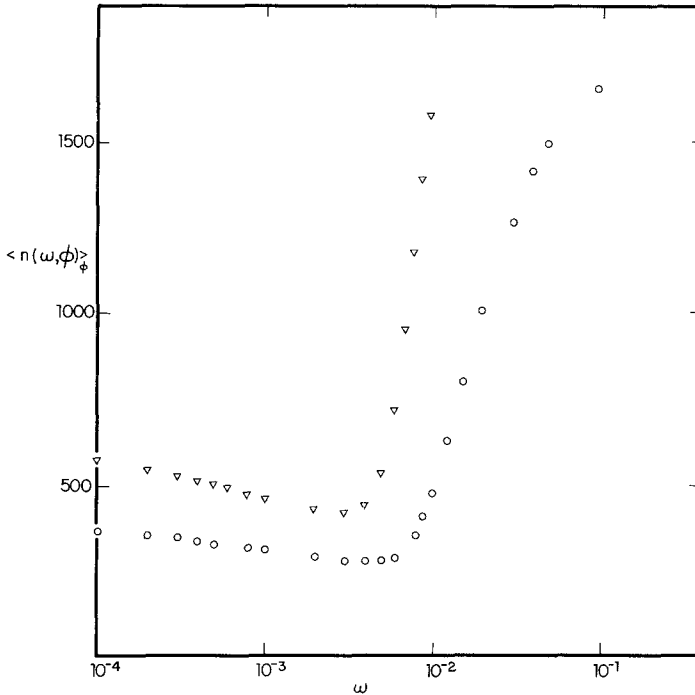


Fig. 8. Curves of the phase averaged mean residence time, $\langle n(\omega, \phi) \rangle_\phi$ plotted as a function of frequency for $L = 100$ (○) and $L = 150$ (▽).

is possible to use perturbation theory when the field is small in an appropriate sense. In the next section we present results based on a numerical solution of the diffusion equation for the case in which perturbation theory is not appropriate.

We denote the probability density for the position of the diffusing particle by $p(x, t)$, which is assumed to satisfy

$$\partial p / \partial t = D \partial^2 p / \partial x^2 - v(t) \partial p / \partial x \quad (11)$$

for a general time-dependent field $v(t)$ and a diffusion constant D . We will convert this partial differential equation into an infinite set of ordinary differential equations, but it is convenient first to transform Eq. (11) into dimensionless form. We do this by setting $v(t) = Vg(t)$, where V is a constant with the dimensions of velocity and $g(t)$ is a dimensionless function that specifies the time-dependent behavior of the field. Equation (11) can be rewritten in dimensionless form by defining the parameters

$$\tau = Dt/L^2, \quad \varepsilon = LV/D, \quad y = x/L \quad (12)$$

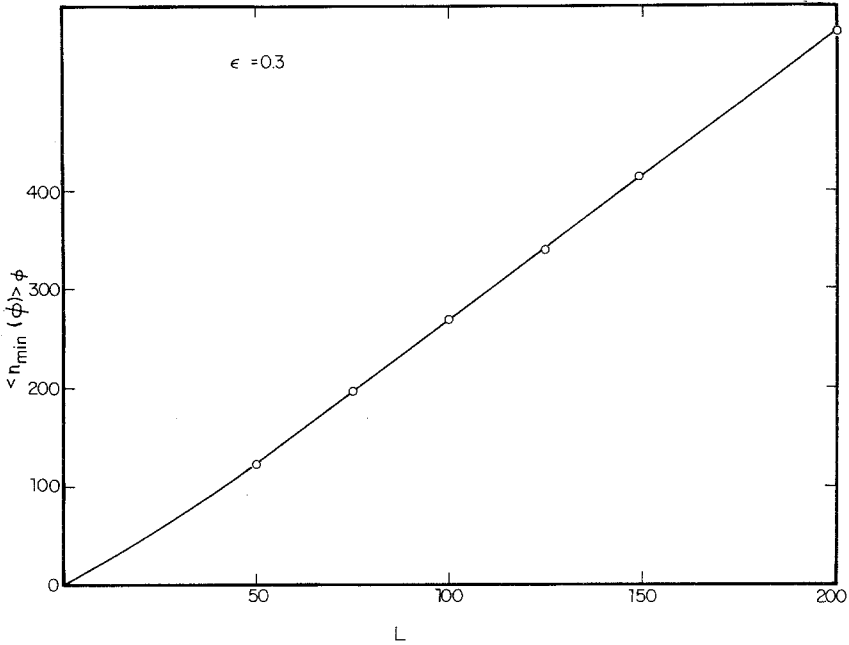


Fig. 9. A plot of the phase averaged mean residence time at the resonant frequency, as a function of L . The fit to a straight line is evident.

so that τ is the dimensionless time, ϵ is a measure of the amplitude of the time-dependent field, and y , the dimensionless length, varies between 0 and 1. These parameters allow us to rewrite Eq. (11) as

$$\partial p / \partial \tau = \partial^2 p / \partial y^2 - \epsilon g(L\tau/V) \partial p / \partial y \tag{13}$$

which is to be solved subject to the absorbing boundary conditions

$$p(0, \tau) = p(1, \tau) = 0 \tag{14}$$

and the initial condition $p(y, 0) = 1$.

In order to transform Eq. (13) into a set of ordinary differential equations, we expand $p(y, \tau)$ in a Fourier series

$$p(y, \tau) = \sum_{n=1}^{\infty} a_n(\tau) \sin(n\pi y) \tag{15}$$

which clearly satisfies the boundary conditions in Eq. (14). On substituting this *ansatz* into Eq. (13), we find that the $a_n(\tau)$ satisfy the set of equations

$$\sum_{n=1}^{\infty} (\dot{a}_n + \pi^2 n^2 a_n) \sin(n\pi y) = -\epsilon h(\tau) \sum_{n=1}^{\infty} n a_n \cos(n\pi y) \tag{16}$$

where, for simplicity, we have set $h(\tau) = 4g(L^2\tau/D)$. However, since the $\{\sin(\pi n y)\}$ form a set of orthogonal functions over $(0, 1)$, we can simplify these equations by multiplying both sides by $\sin(\pi n y)$ and integrating with respect to y . In this way we find that the a_n are solutions to

$$\dot{a}_n + \pi^2 n^2 a_n = -\varepsilon h(\tau) \sum_{m=1}^{\infty} G_{nm} a_m \quad (17)$$

in which the constants G_{nm} are found to be

$$\begin{aligned} G_{nm} &= nm/(n^2 - m^2) & \text{for } n+m \text{ odd} \\ &= 0 & \text{for } n+m \text{ even} \end{aligned} \quad (18)$$

The survival time distribution $S(\tau)$ can be found from the $a_n(\tau)$ in terms of an infinite series

$$S(\tau) = \int_0^1 p(y, \tau) dy = \sum_{n=0}^{\infty} a_{2n+1}(\tau)/(2n+1) \quad (19)$$

It will be convenient to express the mean residence time $\langle \tau_r \rangle$ in terms of the Laplace transforms of $a_n(\tau)$ with respect to τ . The transform of $a_n(\tau)$ will be denoted by $\hat{a}_n(s)$, in which case the expression for $\langle \tau_r \rangle$ can be written as

$$\langle \tau_r \rangle = \int_0^{\infty} S(\tau) d\tau = \sum_{n=0}^{\infty} \hat{a}_{2n+1}(0)/(2n+1) \quad (20)$$

As in the discrete case, we present results for diffusion in a sinusoidally varying field by choosing

$$g(t) = \sin(\omega_0 t) \quad (21)$$

To keep the formulation completely dimensionless, we define a dimensionless frequency ω in terms of ω_0 by

$$\omega = \omega_0 L^2/D \quad (22)$$

We will assume that the amplitude of the field is small in the sense that $\varepsilon \ll 1$ and expand the functions $a_n(\tau)$ in the perturbation series

$$a_n(\tau) = \sum_{j=0}^{\infty} a_n^{(j)}(\tau) \varepsilon^j \quad (23)$$

Successive terms in the perturbation series satisfy the equations

$$\begin{aligned} \dot{a}_n^{(0)} + \pi^2 n^2 a_n^{(0)} &= 0 \\ \dot{a}_n^{(m+1)} + \pi^2 n^2 a_n^{(m+1)} &= -\sin(\omega\tau) \sum_{j=1}^{\infty} G_{nj} a_j^{(m)} \end{aligned} \quad (24)$$

In Eq. (20) we expressed the mean residence time in terms of the Laplace transforms of the $a_n(\tau)$, which makes it attractive to consider this last set of equations in the transform domain. Since the sine function can be written as a difference of exponential functions, we transform Eq. (24) into the algebraic set of equations

$$\hat{a}_n^{(0)} = \frac{2 \sin(n\pi y)}{s + \pi^2 n^2} \tag{25}$$

$$\hat{a}_n^{(m+1)}(s) = \frac{i}{s + \pi^2 n^2} \sum_{l=1}^{\infty} G_{nl} [\hat{a}_l^{(m)}(s - i\omega) - \hat{a}_l^{(m)}(s + i\omega)]$$

The lowest order terms in the perturbation expansion are found to be

$$\hat{a}_n^{(1)}(s) = -\frac{2\omega}{s + \pi^2 n^2} \sum_{l=1}^{\infty} \frac{G_{nl} \sin(\pi l y_0)}{(s + \pi^2 l^2)^2 + \omega^2}$$

$$\hat{a}_n^{(2)}(s) = \frac{2\omega^2}{s + \pi^2 n^2} \sum_{l=1}^{\infty} G_{nl} \sum_{k=1}^{\infty} G_{lk} \frac{\sin(\pi k y_0)}{s + \pi^2 k^2} \tag{26}$$

$$\times \frac{3s + 2\pi^2(2l^2 + k^2)}{[(s + \pi^2 l^2)^2 + \omega^2][(s + \pi^2 k^2)^2 + 4\omega^2]}$$

The mean residence time $\langle \tau_r \rangle$ can also be written in terms of a perturbation series,

$$\langle \tau_r \rangle = \sum_{j=0}^{\infty} \langle \tau_j \rangle \varepsilon^j \tag{27}$$

in which the lowest order term is $\langle \tau_0 \rangle = y_0(1 - y_0)/2$.

Figure 10a shows some curves of $\langle \tau_1 \rangle / \langle \tau_0 \rangle$ as a function of ω for different values of x_0 . It is evident from the form of Eq. (26) that the first-order terms are antisymmetric around $y_0 = 0.5$. Therefore, the equivalent curves for $y_0 = 0.9$ and 0.8 would just be the negative of those for $y_0 = 0.1$ and 0.2 , respectively, and $\langle \tau_1 \rangle$ for $y_0 = 0.5$ is identically equal to zero. The decrease in the mean residence time for $y_0 > 0.5$ is expected because of the initial surge of particles toward the right-hand side of the interval. Figure 10b shows some curves of $\langle \tau_2 \rangle / \langle \tau_0 \rangle$ for different initial positions. These are always negative, which is in qualitative agreement with our lattice calculations. It is interesting to note that the actual magnitudes of the normalized moments are quite small, suggesting that the perturbation expansion is valid for ε values as high as of the order of 1.

Finally, in order to check that qualitative properties derived from our lattice calculations and the perturbation expansion apply more generally,

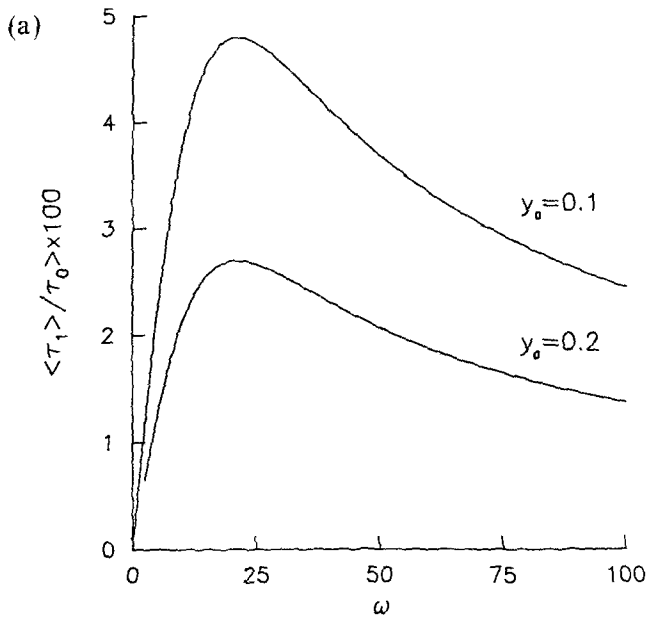


Fig. 10a. Graphs of $\langle \tau_1 \rangle / \langle \tau_0 \rangle$ for diffusion in a sinusoidal field as a function of ω for different values of the starting point y_0 . These curves are drawn for $y_0 < 0.5$. The corresponding curves for $y_0 > 0.5$ are the negatives of those shown here.

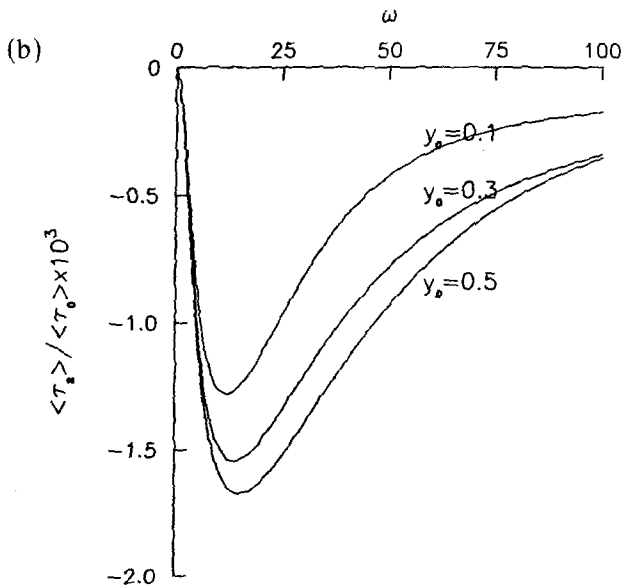


Fig. 10b. Graphs of $\langle \tau_2 \rangle / \langle \tau_0 \rangle$ as a function of ω for the same values of y_0 . These remain negative for all starting points.

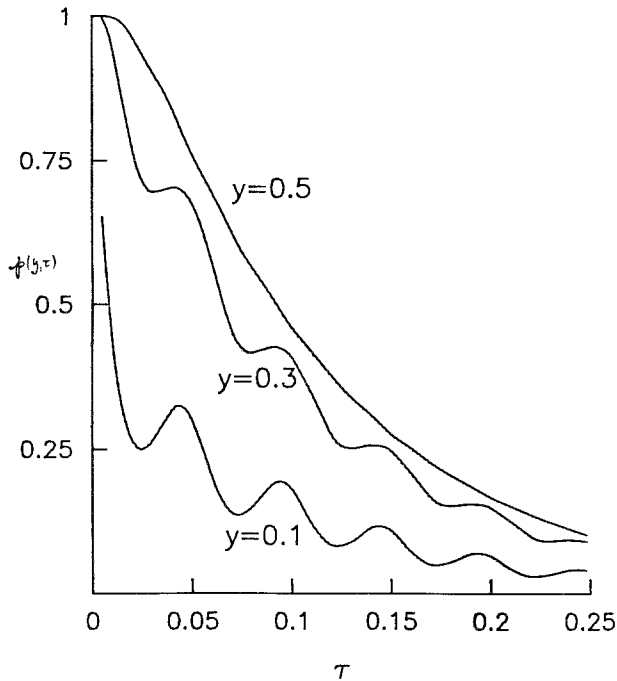


Fig. 11. Typical curves of $p(y, \tau)$ as a function of τ for different values of y . These show that the most noticeable changes in the probability density occur near the trapping points.

we solved the time-dependent diffusion equation using the Crank–Nicholson method for different values of the parameters. Figure 11 shows some curves of $p(y, \tau)$ corresponding to the initial condition $p(y, 0) = 1$. These show that the effects of the field are strongest at the edges, where, of course, the probability is most readily strongly affected by the absorbing points, and tends therefore to most strongly reflect the influence of the oscillating field.

4. DISCUSSION

We have analyzed the simplest examples of random walks and diffusion on a line segment, subjected to a sinusoidal field. Much of our calculations are numerical, since there are no tools analogous to the adjoint equation that proves so useful in the analysis of time-homogeneous Markov processes. We have shown that a sinusoidal field induces coherent motion that tends to reduce the residence time on the line segment. This, however, has not been proved in generality, although our results strongly suggest the truth of the assertion. The phenomenon persists when there is a

constant field in addition to the sinusoidal one. This may be applicable to the improvement of the resolution of electrophoretic systems for the separation of proteins, since the application of a sinusoidal field can amplify differences in the speed of two proteins if the frequency is properly chosen. We have carried out calculations of the properties of diffusive motion in a randomly time-dependent field, but there do not appear to be any significant changes induced by such a field.

It is interesting to speculate on the effects of a sinusoidal field in the so-called trapping problem.⁽⁸⁾ The one firmly established result in this area is the long-time limit of the survival probability

$$\ln S(t) \approx -at^{D/(D+2)} \quad (28)$$

due to Donsker and Varadhan,⁽⁹⁾ where a is a constant. Can an oscillating field change this? Our feeling is that it cannot, because the resonant frequency applies to a single length in one dimension, whereas the classical statement of the trapping problem involves a complete spectrum of possible lengths. On the other hand, a sinusoidal field may have a considerable influence on trapping kinetics at early times. This remains to be explored, as does the effects of time-dependent fields in dimensions greater than 1.

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