

Analytic Solutions for Some Reaction-Diffusion Scenarios

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ABSTRACT: Motivated currently by the problem of coalescence of receptor clusters in mast cells in the general subject of immune reactions, and formerly by the investigation of exciton trapping and sensitized luminescence in molecular systems and aggregates, we present analytic expressions for survival probabilities of moving entities undergoing diffusion and reaction on encounter. Results we provide cover several novel situations in simple 1-d systems as well as higher-dimensional counterparts along with a useful compendium of such expressions in chemical physics and allied fields. We also emphasize the importance of the relationship of discrete sink term analysis to continuum boundary condition studies.

General Prescription for Survival Probability

$$\tilde{Q}(\varepsilon) = \frac{1}{\varepsilon} \left[1 - \frac{\sum_r \tilde{\eta}(x_r, \varepsilon)}{1/\mathcal{C}_d + \sum_r \tilde{\Pi}(x_r, x_r, \varepsilon)} \right]$$

1. INTRODUCTION

Many physical situations are characterized by entities that move from one location to another and then, when they are within a radius of influence of either one another or some other entity of a different kind, undergo a process such as capture, annihilation, or chemical reaction. The moving entities can be variously an electron, an excitation, an atom, a molecule, a biological object such as a receptor or receptor cluster, a cell, or even an animal such as a mouse carrying an epidemic. The nature of the motion may be quantum mechanical, wavelike or coherent, ballistic, incoherent or diffusive, super diffusive, or a superposition of several of these. The combined phenomenon is termed reaction-diffusion.^{1–62} Because such situations are ubiquitous in areas of research ranging from chemistry^{1–12} and physics^{13–33,35–43,46} to biology,^{27,56–58,60–64} an enormous literature has accumulated on this subject. Understandably, theoretical results have been discovered and rediscovered and published often without reference to one another. The lack of communication across widely differing disciplines and communities is natural. However, it appears that this lack of communication has also resulted in incorrect claims. One example is the assertion that, while perfect capture situations can be solved, generalization to include finite reaction rates makes the problem of reaction-diffusion impossible to solve analytically even in principle.^{34,35} A desire to remove such misunderstandings, and to avoid waste of effort that duplication of analysis involves, provides our motivation for including a reference to older results in addition to presenting new ones in the following.

Our own work in this field began in the subject area of exciton dynamics where one of the authors and his collaborators undertook²¹ a systematic analysis of exciton trapping and sensitized luminescence in molecular systems and aggregates against the backdrop of experiments by Fayer,^{8–10,13} Zewail,^{11,12} Wolf,⁴⁷ Schmid,^{48–51} and others, that targeted in some cases the magnitude of the diffusion constant of Frenkel excitons and in others the degree of their coherence. After

decades, our interest in the general subject resurfaced in a collaborated effort by the two present authors in the quite different (biophysics) area of coalescence of receptor clusters in mast cells,⁶² as well as other immune cells: T cells⁶³ and B cells.⁶⁴ New results we have obtained should find applicability not only in the specific areas of our own research but also in much wider contexts.

The outline of the paper is as follows. Given that many authors (see especially refs 2, 16, 21, and 53) have written about reaction diffusion analysis from a variety of viewpoints in diverse notation, it is necessary to minimize confusion arising from terminology. For this reason only, our own approach and methodology^{4,21–26} are put forward in section 2, along with an illustrative application to the well-known and simple system of a single trap in a one-dimensional discrete system. During this application, we look into the generalization of an older (perfect absorber) finding. Passage to the continuum within a 1-d system and some new results including those for trapping in a potential are discussed in section 3. Section 4 is dedicated to higher-dimensional systems of radial symmetry, 2-d and 3-d, with practically useful scenarios such as those involving infinite and finite lines of traps, infinite sheets of traps, spherical capture regions, and trapping rings. In section 5, we point out the importance of discrete sink term analysis vis-a-vis continuum boundary condition studies. Concluding remarks form section 6.

2. FORMALISM, TERMINOLOGY, AND A SIMPLE APPLICATION

Consider a particle that may occupy a site m in a discrete space of arbitrary dimensions with probability $P_m(t)$ at time t and

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move in some way, e.g., with or without translational invariance. A standard Master equation for $P_m(t)$ is

$$\frac{dP_m(t)}{dt} = \text{motion terms} - C \sum_r \delta_{m,r} P_m(t) \quad (1)$$

where the motion terms are linear in probabilities, C is the capture rate, and sites r denote the reaction locations at which the particle disappears. Here $\delta_{m,r}$ represents the Kronecker delta function and the prime denotes a sum over all reaction locations at which one produces trapping or coalescence or similar effects. If the particle hops via nearest neighbor rates F in 1-d, the motion terms could be of the form $F(P_{m+1} + P_{m-1} - 2P_m)$. More generally, r , m , etc., would be vectors in the appropriate dimensions.

Equation 1, when transformed into the Laplace domain, becomes

$$\tilde{P}_m(\varepsilon) = \tilde{\eta}_m(\varepsilon) - C \sum_r \tilde{\Psi}_{m,r}(\varepsilon) \tilde{P}_r(\varepsilon) \quad (2)$$

where tildes denote Laplace transforms and ε is the Laplace variable. Ψ is the probability propagator of the homogeneous part of eq 1, and $\eta_m(t)$ is the homogeneous solution $\sum_n \Psi_{m,n}(t) P_n(0)$ in the absence of the trap, i.e., $C = 0$. While many derived quantities such as the yield and the effective rate of reaction may be of interest in specific contexts, the common focus is to calculate the total survival probability $Q(t) = \sum_m P_m(t)$. To obtain this quantity, one notes that the sum over all m over both $\tilde{\eta}_m$ and $\tilde{\Psi}_{m,r}$ gives $1/\varepsilon$, since the probability sum over all sites of the homogeneous solution is always one in the time domain. Therefore, in the Laplace domain, the survival probability is given as

$$\tilde{Q}(\varepsilon) = \frac{1}{\varepsilon} [1 - C \sum_r \tilde{P}_r(\varepsilon)] \quad (3)$$

One can also compute the rate of disappearance by summing over m in eq 1. Thus,

$$\frac{dQ(t)}{dt} = -C \sum_r P_r(t) \quad (4)$$

The total probability at the starting site given in eq 3 has to be calculated in the *presence* of the trapping sites. Let us define its counterpart in the absence of the trapping sites, i.e., $C = 0$, and the same initial conditions

$$\left(\sum_r P_r(t) \right)_0$$

and rewrite eq 4 as

$$\frac{dQ(t)}{dt} = - \int_0^t dt' \mathcal{M}(t-t') \left(\sum_r P_r(t') \right)_0 \quad (5)$$

The important quantity in eq 5 is the function $\mathcal{M}(t)$. In the limit of small C , it is simply $C\delta(t)$ so that we have a simplified form of eq 4 with $(\sum_r P_r(t'))_0$ in place of $\sum_r P_r(t')$. This is the capture-limited case. In the opposite limit of small motion (large C), the function $\mathcal{M}(t)$ turns out to be more complicated and is determined by motion parameters. That is the motion-limited case.

In order to understand these last statements accurately, we substitute $m = s$, where s is the trap site into eq 2 and sum over trap locations s

$$\sum_s \tilde{P}_s(\varepsilon) = \sum_s \tilde{\eta}_s(\varepsilon) - C \sum_s \sum_r \tilde{\Psi}_{s,r} \tilde{P}_r(\varepsilon) \quad (6)$$

The quantity

$$\nu_r = \sum_s \Psi_{s,r} \quad (7)$$

is the sum of the probability propagators from one trap site r to all trap sites s . While this expression actually does depend on the site r , that dependence will disappear in highly symmetrical situations or in an averaging sense. We are now going to assume that the r -dependence has been removed either exactly or in an averaging sense.⁴ One can for instance calculate an average over all trap sites r of ν_r and call it ν (independent of r). After such an assumption/approximation, we can write the actual $\sum_r \tilde{P}_r$ in terms of the homogeneous counterpart $(\sum_r \tilde{P}_r)_0$, which is precisely $\sum_r \tilde{\eta}_r$

$$\sum_r \tilde{P}_r(\varepsilon) = \frac{\sum_r \tilde{\eta}_r}{1 + C\tilde{\nu}(\varepsilon)} \quad (8)$$

At once, we get the above-mentioned eq 5 where $\mathcal{M}(t)$ is precisely given by

$$\tilde{\mathcal{M}}(\varepsilon) = \frac{1}{1/C + \tilde{\nu}(\varepsilon)} \quad (9)$$

We see here that generally this form is compatible with the concept of a sum of the capture time and a motion time. This result is in contradiction to Kashchiev's form of the reaction rate, which involves a mere product of probabilities.²⁸ Much discussion has occurred in the exciton field in molecular crystals about the consequence of the form of eq 9.^{21,26} In the motion limit, C is large enough relative to the motion term and $1/C$ can be neglected to give $\tilde{\mathcal{M}}(\varepsilon) = 1/\tilde{\nu}(\varepsilon)$, whereas in the capture limit $\tilde{\mathcal{M}}(\varepsilon) = C$.

Finally, a general prescription for the total survival probability in Laplace domain and discrete space is therefore

$$\tilde{Q}(\varepsilon) = \frac{1}{\varepsilon} \left[1 - \frac{\sum_r \tilde{\eta}_r}{1/C + \tilde{\nu}(\varepsilon)} \right] \quad (10)$$

The key quantity to calculate is the ν -function, which is the (ensemble average of the) sum of propagators of the homogeneous system (in absence of traps) from one trap location to all others. The idea of the ν -function was first put forward⁴ to generalize the one-trap analysis to arbitrary trap concentrations, exactly for periodic traps and approximately for any placement. It was used in luminescence calculations for molecular crystals²⁵ for an arbitrary kind of motion and different placements of trap sites and in other contexts such as of cellular membranes.²⁷

Discrete systems are interesting in their own right in that they appear in nature in the form of crystal lattices. A great deal of experimental work on exciton transport in molecular crystals^{8-13,47-51} necessitated their study. Additionally, discrete lattice results produce continuum counterparts when appropriate limits are taken and then become useful to problems in many areas including biophysics. Correspondingly, we now display the continuum limit of the above discrete formalism. If, for simplicity, we start on a 1-d discrete lattice with a single stationary trap at site r and a particle hopping via nearest

neighbor rates F between lattice sites, the starting equation, given in eq 1, would have the explicit form

$$\frac{dP_m}{dt} = F(P_{m+1} + P_{m-1} - 2P_m) - \delta_{m,r}CP_m \quad (11)$$

where $\delta_{m,r}$ is the Kronecker delta function and C is the capture rate. The continuum prescription is obtained by dividing eq 11 by the lattice constant a , taking the limit (see, e.g., refs 23 and 27) $a \rightarrow 0$, and correspondingly $ma \rightarrow x$, $a \rightarrow dx$, $Fa^2 \rightarrow D$, $P_m/a \rightarrow P(x, t)$, $aC \rightarrow C_1$, and $\delta_{m,r}/a \rightarrow \delta(x - x_r)$ to obtain

$$\frac{\partial P(x, t)}{\partial t} = D \frac{\partial^2 P(x, t)}{\partial x^2} - C_1 \delta(x - x_r) P(x, t) \quad (12)$$

Here x represents the 1-d space coordinate, D the particle diffusion constant, C_1 the capture parameter in units m/s, and $\delta(x - x_r)$ a Dirac delta function at location x_r . Equation 12 is the 1-d diffusion equation with the addition of a trapping term. This result is obvious also when one applies the continuum limit directly to the discrete space Master equation^{36–38} and the simultaneous limit $F \rightarrow \infty$, $C \rightarrow \infty$, where F tends as $1/a^2$ and C as $1/a$, is essential to make the continuum prescription viable.^{23,27} Following this procedure, we can divide also

$$\tilde{P}_m(\varepsilon) = \tilde{\eta}_m(\varepsilon) - \tilde{\Psi}_{mr}(\varepsilon) \frac{\tilde{\eta}_r(\varepsilon)}{1/C + \tilde{\Psi}_{rr}(\varepsilon)} \quad (13)$$

by the lattice constant a and take $a \rightarrow 0$ to obtain

$$\tilde{P}(x, \varepsilon) = \tilde{\eta}(x, \varepsilon) - \tilde{\Pi}(x, x_r, \varepsilon) \frac{\tilde{\eta}(x_r, \varepsilon)}{1/C_1 + \tilde{\Pi}(x_r, x_r, \varepsilon)} \quad (14)$$

Upon generalization to many traps at sites x_r , this result leads, in the Laplace domain, to the continuum result for the survival probability

$$\tilde{Q}(\varepsilon) = \frac{1}{\varepsilon} \left[1 - \frac{\sum_r' \tilde{\eta}(x_r, \varepsilon)}{1/C_d + \sum_r' \tilde{\Pi}(x_r, x_r, \varepsilon)} \right] \quad (15)$$

Here $\tilde{\Pi}(x, x_r, \varepsilon)$ is the self-propagator and its sum \sum_r' thus corresponds to the ν -function. $\tilde{\eta}(x_r, \varepsilon)$ is the homogeneous solution at the trap site in the absence of the trap, \sum_r' represents the sum over all trap sites which typically becomes an integral in the continuum limit, x_r denote vectors in the appropriate number of dimensions, and C_d is the d -dimensional capture parameter. In the continuum, C_d has the units of length raised to d divided by time. The passage from the discrete formula to its continuum version has introduced Π propagators in place of the discrete Ψ 's.

Let us now illustrate the terminology and formalism in the context of the simplest of systems, viz., a particle moving on a 1-d chain with nearest-neighbor interactions with a single trap, derive a result given earlier by Spouge⁴¹ in the limit of infinite capture rate, and study its counterpart for arbitrary capture rate.

2.1. Generalization of Spouge's Infinite Capture Rate Result to Finite Rates. Suppose that a particle is initially placed at site n on the 1-d lattice characterized by trapping at an infinite rate (perfect absorption) at a single trap at the origin. What is the dependence on time t and trap location n of the survival fraction $Q(t)$? This simple but physically important question was posed by Spouge and answered as⁴¹

$$Q(t) = 1 - e^{-2Ft} I_n(2Ft) - 2e^{-2Ft} \sum_{k=0}^{\infty} (-1)^k [I_{1+n+2k}(2Ft) + I_{2+n+2k}(2Ft)] \quad (16)$$

What are the precise problems one encounters in obtaining a usable counterpart of this result for a finite capture rate (imperfect absorption)? Let us apply the discrete prescription given in eq 10 to this case, where there is only a single trap. The prescription becomes

$$\tilde{Q}(\varepsilon) = \frac{1}{\varepsilon} \left[1 - \frac{\tilde{\eta}_n}{1/C + \tilde{\nu}(\varepsilon)} \right] \quad (17)$$

where $\eta_n = \Psi_{n,0}$ and $\nu = \Psi_{0,0}$, the homogeneous solution starting at site n in the absence of the trap and the self-propagator, respectively. The discrete space self-propagator for this problem is well-known:

$$\Psi_m(t) = I_m(2Ft) \exp(-2Ft) \quad (18)$$

where I_m is the modified Bessel function of the first kind. The Laplace transform of both ν and η can be obtained⁶⁵

$$\tilde{\nu}(\varepsilon) = \frac{1}{\sqrt{(\varepsilon + 2F)^2 - 4F^2}}, \quad \tilde{\eta}_n(\varepsilon) = \frac{(2F)^n}{\sqrt{(\varepsilon + 2F)^2 - (2F)^2} [\varepsilon + 2F + \sqrt{(\varepsilon + 2F)^2 - (2F)^2}]^n}$$

Using the substitution⁴ $\cosh \zeta = 1 + \varepsilon/(2F)$, one may rewrite eq 17 as

$$\tilde{Q}(\varepsilon) = \frac{1}{\varepsilon} \left[1 - \frac{\exp(-\zeta n)}{1 + 2F \sinh \zeta / C} \right] \quad (19)$$

where $\sinh \zeta = (4F\varepsilon + \varepsilon^2)^{1/2}/(2F)$. Equation 19 is the exact solution in the Laplace domain.

The rate of disappearance dQ/dt is given by

$$-\frac{dQ(t)}{dt} = L_\varepsilon^{-1} \left\{ \frac{\exp(-\zeta n)}{1 + 2F \sinh(\zeta)/C} \right\} \quad (20)$$

where L_ε^{-1} denotes the inverse Laplace transform. Re-expressing the exponential in terms of $\sinh \zeta$ and $\cosh \zeta$ leads to

$$-\frac{e^{2Ft}}{Cn} \frac{dQ(t)}{dt} = \int_0^t \frac{I_n[2F(t-t')]}{(t-t')e^{Ct'}} dt' + 2F \int_0^t \int_0^{t'} \frac{I_n[2F(t-t')]}{(t-t')e^{C(t^2-t'^2)^{1/2}}} I_1(t'') dt'' dt' \quad (21)$$

and shows that inclusion of a finite reaction rate does add considerable complexity to the explicit solution in that it involves two quadratures. Simplification occurs for a large capture rate with respect to motion rate. In this case, the smallness of F/C and consequently the use⁶⁷ of the exponential approximation $[1 + (2F/C) \sinh \zeta]^{-1} \approx e^{-2F\zeta/C}$ yield

$$-\frac{dQ(t)}{dt} = L_\varepsilon^{-1} \{ e^{-\zeta(\varepsilon + 2F/C)} \} \quad (22)$$

For instantaneous reaction, $C \rightarrow \infty$ and eq 19 becomes

$$\tilde{Q}(\varepsilon) = \frac{1}{\varepsilon} [1 - e^{-\zeta n}] \quad (23)$$

Rewriting the exponential term

$$e^{-\zeta^n} = \frac{(2F)^n}{(2F + \varepsilon + \sqrt{4F\varepsilon + \varepsilon^2})^n}$$

and inverting the Laplace transform, one gets

$$Q(t) = 1 - e^{-2Ft} I_n(2Ft) - 2Fe^{-2Ft} \int_0^t [I_0(2Ft - 2Ft') + I_1(2Ft - 2Ft')] I_n(2Ft') dt' \quad (24)$$

The integral in eq 24 can be expressed in terms of an infinite sum of Bessel functions of the first kind, J_m , by using summation identities of Bessel functions. We get

$$Q(t) = 1 - e^{-2Ft} \left[i^{-n} J_n(i2Ft) + \frac{2}{i^{n+1}} \sum_{k=0}^{\infty} (-1)^k J_{1+n+2k}(i2Ft) + \frac{2}{i^{n+2}} \sum_{k=0}^{\infty} (-1)^k J_{2+n+2k}(i2Ft) \right] \quad (25)$$

This expression is equivalent to Spouge's expression given in eq 16, since $J_\nu(x) = i^\nu I_\nu(-ix)$. We now show that eq 25 or eq 16 can be rewritten in terms of a *finite sum* of Bessel functions, resulting in a practically more useful expression for the perfect absorber case. Using the standard expansion of sine and cosine in terms of Bessel functions, and de Moivre identity between trigonometric and exponential functions, one can write $Q(t)$ for $n = 1, 2, 3$

$$\begin{aligned} Q(t)_{n=1} &= e^{-2Ft} [I_0(2Ft) + I_1(2Ft)] \\ Q(t)_{n=2} &= e^{-2Ft} [I_0(2Ft) + 2I_1(2Ft) + I_2(2Ft)] \\ Q(t)_{n=3} &= e^{-2Ft} [I_0(2Ft) + 2I_1(2Ft) + 2I_2(2Ft) \\ &\quad + e^{-2Ft} I_3(2Ft)] \end{aligned}$$

Using the pattern that emerges, eq 25 can be recast in the form of a finite sum of modified Bessel functions of the first kind

$$Q(t) = e^{-2Ft} (I_0(2Ft) - I_n(2Ft) + 2 \sum_{m=1}^n I_m(2Ft)) \quad (26)$$

The practical advantage of eq 26 over Spouge's original result, eq 16, is that computation of $Q(t)$ is highly simplified particularly for initial placement of the excitation not too far from the origin because our formula does not necessitate a summation of an infinite number of terms. For the special case of a particle initially starting at lattice site $n = 1$, our expression reduces to

$$Q(t) = e^{-2Ft} [I_0(2Ft) + I_1(2Ft)] \quad (27)$$

and agrees with ref 41.

3. RESULTS IN 1-D CONTINUUM

A problem that is simple but important because of its recurrence in many physical contexts is that of calculating the survival probability of a diffusing particle in a 1-d continuum, where the particle is initially a distance x_0 away from a trap located at the origin. The well-known result for perfect absorption is^{16,41,42,44,68}

$$Q(t) = \operatorname{erf}\left(\frac{1}{2} \sqrt{\frac{\tau_1}{t}}\right) \quad (28)$$

where $C_1 \rightarrow \infty$ and $\tau_1 = x_0^2/D$ is the diffusion time in one dimension, i.e., the time taken by the diffusing particle in the trapless system to arrive from its initial location to the trap.

The counterpart of this result for arbitrary capture (imperfect absorption) is not known as well (this is one of the reasons misleading statements that such results do not exist have been made in the literature) but deserves to be known. It has been derived many times independently,^{16,36,54,68} and is

$$Q(t) = \operatorname{erf}\left(\frac{1}{2} \sqrt{\frac{\tau_1}{t}}\right) + e^{(1/\xi_1) + (1/\xi_1^2)(t/\tau_1)} \operatorname{erfc}\left(\frac{1}{2} \sqrt{\frac{\tau_1}{t}} + \frac{1}{\xi_1} \sqrt{\frac{t}{\tau_1}}\right) \quad (29)$$

The fact that absorption in this case can be imperfect is represented by the arbitrary nonvanishing value of the parameter $\xi_1 = 2D/(C_1 x_0)$.

We now show how easily these results can be obtained from our prescription in eq 15. The 1-d free-space diffusion propagator

$$\Pi(x, x_0, t) = \frac{1}{\sqrt{4\pi Dt}} \exp\left[-\frac{(x - x_0)^2}{4Dt}\right] \quad (30)$$

leads to the self-propagator

$$\Pi(0, 0, t) = \frac{1}{\sqrt{4\pi Dt}}$$

and to the homogeneous solution at the trap site

$$\eta(t) = \Pi(0, x_0, t) = \frac{1}{\sqrt{4\pi Dt}} \exp\left[-\frac{x_0^2}{4Dt}\right] \quad (31)$$

The Laplace transforms of these two key quantities, when inserted in eq 15, immediately yield the survival probability in the Laplace domain:

$$\tilde{Q}(\varepsilon) = \frac{1}{\varepsilon} \left[1 - \frac{\exp(-\sqrt{\varepsilon \tau_1})}{1 + \xi_1 \sqrt{\varepsilon \tau_1}} \right] \quad (32)$$

Laplace inversion of this expression is trivial upon the use of the scaling property of transforms and leads to the general result eq 29. For perfect absorption, ξ_1 vanishes and we have eq 28.

As another interesting application, we mention trapping while the particle moves *in a potential* so that translational invariance is destroyed and the free-space diffusion propagator no longer applies. The governing equation in the absence of traps is the Fokker–Planck equation

$$\frac{\partial P(x, t)}{\partial t} = \frac{\partial}{\partial x} \left[\frac{dU(x)}{dx} P(x, t) + D \frac{\partial P(x, t)}{\partial x} \right] \quad (33)$$

with a quadratic potential of the form $U(x) = \gamma x^2/2$ and a localized initial condition, $P(x, 0) = \delta(x - x_0)$. We have shown elsewhere⁶⁶ that, using the Ornstein–Zernike solution⁶⁹ of this equation, it is straightforward to calculate the survival probability if the trap location is at the center of the potential. We have found explicitly that the Laplace transform of the survival probability is

$$\tilde{Q}(\varepsilon) = \frac{1}{\varepsilon} \frac{\frac{1}{2\gamma\sqrt{\pi}} \left(\frac{\gamma\tau_1}{2}\right)^{-1/4} e^{\gamma\tau_1/4} W_{(1/4),-(\varepsilon/2\gamma), (1/4)} \left(\frac{\gamma\tau_1}{2}\right) \Gamma\left(\frac{\varepsilon}{2\gamma}\right)}{\frac{\xi_1 \varepsilon \sqrt{\tau_1}}{\sqrt{2\gamma}} + \Gamma\left(\frac{\varepsilon}{2\gamma} + 1\right) \Gamma\left(\frac{\varepsilon}{2\gamma} + \frac{1}{2}\right)} \quad (34)$$

where γ describes the strength of the potential and W is the Whittaker W -function defined in ref 70 as

$$W_{\kappa,\mu}(z) = e^{-z/2} z^{(1/2)+\mu} U\left(\frac{1}{2} + \mu - \kappa, 1 + 2\mu, z\right),$$

larg $|z| < \pi$

in terms of the confluent hypergeometric functions

$$U(a, b, c) = \frac{1}{\Gamma(a)} \int_0^\infty e^{-ct} t^{a-1} (1+t)^{b-a-1} dt$$

Equation 34 is a new result. We have shown that it reduces in the infinite capture rate limit to the simpler version obtained by Bagchi, Fleming, and Oxtoby⁵ but contains explicit novel information in the general case.

3.1. General Prescription for Any Initial Distributions.

Since the problem is linear, the superposition principle can be applied to obtain a solution for any initial distribution of point particles. In the special case of perfect absorption, the principle of superposition states that

$$Q(t) = \int_0^\infty \rho(x_0) \operatorname{erf}\left(\frac{x_0}{\sqrt{4Dt}}\right) dx_0 \quad (35)$$

where $\rho(x_0)$ is the initial distribution of point particles.

Among results published in the literature for specific distributions (and perfect absorption) are the case of an initial random distribution for which $\rho(x_0)$ is of Poisson form $\rho(x_0) = c \exp(-cx_0)$, where c is an arbitrary constant,

$$Q(t) = \exp(c^2 Dt) \operatorname{erfc}(c\sqrt{Dt}) \quad (36)$$

calculated by Torney and McConnel⁷ and by Sancho et al.,³¹ and the case of the Rayleigh distribution, also known as a biased Gaussian, $\rho(x_0) = x_0 \exp(-x_0^2/(2\sigma^2))/\sigma^2$, where σ describes the width of the distribution

$$Q(t) = \frac{\sigma}{\sqrt{2Dt + \sigma^2}} \quad (37)$$

reported by Doering and ben-Avraham.²⁹ We present in the following a generalized formula usable for arbitrary capture rates (imperfect absorption) and arbitrary initial distributions.

Although the principle of superposition is obviously not restricted to perfect absorption, multiplying the general expression given in eq 29 by an initial distribution and integrating the product over the position x_0 becomes algebraically tedious. To simplify this calculation, one can apply the superposition principle in the Laplace domain and attempt to derive a prescription, which does not require the computation of Laplace inversions. We derive such a prescription below.

We start with $\tilde{Q}(\varepsilon)$ given in eq 32, showing the explicit dependence on the initial location x_0 :

$$\tilde{Q}(\varepsilon) = \frac{1}{\varepsilon} \left[1 - \frac{e^{-(\varepsilon/D)^{1/2} x_0}}{\frac{\sqrt{4\varepsilon D}}{C_1} + 1} \right]$$

On averaging this result over the initial distribution $\rho(x_0)$, i.e., by performing an integral such as in eq 35, we see that the Laplace transform of dQ/dt equals

$$\left(\frac{p}{p + \sqrt{\varepsilon/D}} \right) \int_0^\infty \rho(x_0) e^{-x_0(\varepsilon/D)^{1/2}} dx_0$$

where $p = C_1/2D$. This expression can be interpreted as the product of two Laplace transforms each with $(\varepsilon/D)^{1/2}$ as the Laplace variable. The first is the transform of an exponential, the second of the distribution ρ . We now recall two identities.⁶⁵ The first is the standard scaling property of Laplace transforms; the second is that, if $f(t)$ is the Laplace inverse of $\tilde{f}(\varepsilon)$, the inverse of $\tilde{f}(\varepsilon^{1/2})$ is

$$\frac{1}{2\sqrt{\pi}} t^{-3/2} \int_0^\infty u e^{-u^2/4t} f(u) du$$

Combining them, we get our final result for the rate of disappearance of the survival probability:

$$\frac{dQ(t)}{dt} = -\frac{C_1}{4\sqrt{\pi}} (Dt)^{-3/2} \int_0^\infty x e^{-x^2/4Dt} H(x) dx \quad (38)$$

where

$$H(x) = \int_0^x \rho(x_0) e^{-p(x-x_0)} dx_0 \quad (39)$$

and $p = C_1/(2D)$.

This prescription allows us to calculate the particle survival probability for any initial distribution of noninteracting particles which diffuse in the presence of a single stationary trap. The two examples given at the beginning of this subsection can be recovered as particular cases. We sketch below the manner in which our general prescription can be used to compute $Q(t)$ for an initial distribution localized at an arbitrary site x_0 and for arbitrary capture rate. Since ρ is a δ -function,

$$H(x) = e^{-px} \int_0^x \rho(y) e^{py} dy = e^{-px} e^{px_0} \Theta(x - x_0) \quad (40)$$

where Θ is the Heaviside step function. Substituting eq 40 into eq 38, we are left to evaluate

$$\frac{dQ(t)}{dt} = -\frac{C_1}{4\sqrt{\pi}} (Dt)^{-3/2} \int_{x_0}^\infty x e^{-x^2/4Dt} e^{-p(x-x_0)} dx \quad (41)$$

Straightforward algebra leads to

$$\frac{dQ(t)}{dt} = \frac{1}{\xi_1^2 \tau_1} e^{(1/\xi_1) + (1/\xi_1^2)(t/\tau_1)} \operatorname{erfc}\left(\frac{\sqrt{\tau_1}}{2\sqrt{t}} + \frac{1}{\xi_1} \sqrt{\frac{t}{\tau_1}}\right) - \frac{1}{\sqrt{\pi} \xi_1 \sqrt{\tau_1 t}} e^{-\tau_1/4t} \quad (42)$$

This expression for the rate of disappearance is directly equivalent to eq 29 and is valid for arbitrary capture rate.

4. HIGHER-DIMENSIONAL SYMMETRICAL SYSTEMS

We present results in this section for 2-d and 3-d systems of high symmetry in the continua.

The stationary trapping prescription given in eq 15 can be obviously applied to higher dimensions where a trap in 1-d becomes an absorbing surface in higher dimensions. The second term in the denominator of eq 15 is the ensemble average of the sum of propagators of the homogeneous system (in absence of traps) from one trap location to all others—the ν -function formalism of ref 4 thus finds a natural use in these higher-dimensional calculations. Numerous real-world applications involve situations characterized by high symmetry wherein only the radial coordinate enters into consideration. For such, we develop analytic results in the following.

4.1. Results for 2-d Systems. Let us consider a radially symmetric situation in which the trapping region is the circumference of a circle (disk) of radius R and the origin as its center. In Cartesian coordinates, the 2-d propagators for isotropic diffusion are simply products of Gaussian 1-d propagators (isotropy assumed) and therefore independent of the angular polar coordinate θ . In calculating ν in an expression such as eq 15, we can take one of the angles to be 0. The Cartesian coordinates of the two points are $R \cos \theta$, $R \sin \theta$ and R , 0, respectively. We are therefore led to the evaluation of integrals such as

$$\int_0^{2\pi} \frac{\exp\left[-\frac{R^2[(1-\cos\theta)^2 + \sin^2\theta]}{4Dt}\right]}{4\pi Dt} d\theta$$

which can be evaluated easily in terms of the I_0 Bessel function of argument $R^2/2Dt$. In 1-d, there would be no integral and the denominator would have a square root of t .

Inspection of eq 15 shows that we must calculate two quantities that correspond to the ν -function and η . The former is the average self-propagator in the trap region which is a circumference of radius R . The latter is the homogeneous (trapless) solution for the given initial conditions that the circumference of radius R_0 is occupied uniformly. Both these quantities are obtainable by evaluating

$$\int_0^{2\pi} \int_0^\infty G(\vec{r}, \vec{r}', t) P(\vec{r}', 0) r' dr' d\theta'$$

given the Green's function

$$G(\vec{r}, \vec{r}', t) = \frac{1}{4\pi Dt} e^{-|\vec{r}-\vec{r}'|^2/4Dt} \quad (43)$$

where $|\vec{r}-\vec{r}'|^2 = (x-x')^2 + (y-y')^2$, and the rotationally symmetric initial condition $P(\vec{r}, 0) = \delta(r-R_0)/(2\pi r)$. Carrying out the integration over r' , and realizing that the left integral is the I_0 Bessel function, we get the important result⁶⁸

$$P(R, r, t) = \frac{1}{4\pi Dt} e^{-(r^2+R^2)/4Dt} I_0\left(\frac{rR}{2Dt}\right) \quad (44)$$

as the probability density for the particle to occupy the circumference of radius R given that it initially occupied that of radius r .

4.1.1. Circular Trap of Finite Radius. We now analyze an initial circular symmetric distribution of diffusing noninteracting point particles initially at R_0 . The circular trap is centered at the origin and has a radius R , where $R < R_0$ (Figure 1).

In our result derived above, we put $r = R$ to get

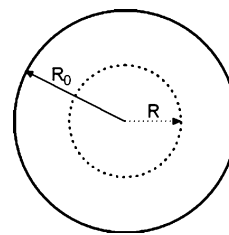


Figure 1. The high-symmetry situation in 2-d and 3-d systems illustrated by showing an inner region (dotted) which is the trapping region (of radius R) and an outer region (solid) which is the region where the moving particles are initially placed uniformly (of radius R_0). These regions are circumferences of circles in the 2-d case and surfaces of spheres in the 3-d case. There is perfect radial symmetry which means that there is no angular dependence in the problems considered.

$$P(R, R, t) = \frac{1}{4\pi Dt} e^{-R^2/2Dt} I_0\left(\frac{R^2}{2Dt}\right)$$

and $r = R_0$ to get

$$P(R, R_0, t) = \frac{1}{4\pi Dt} e^{-(R^2+R_0^2)/4Dt} I_0\left(\frac{RR_0}{2Dt}\right)$$

and Laplace transform both expressions and substitute in eq 15. This yields

$$\tilde{Q}(\varepsilon) = \frac{1}{\varepsilon} \left[1 - \frac{\frac{1}{2\pi D} K_0\left(\sqrt{\frac{\varepsilon}{\gamma_0}}\right) I_0\left(\sqrt{\frac{\varepsilon}{\varepsilon_0}}\right)}{\frac{1}{C_2} + \frac{1}{2\pi D} K_0\left(\sqrt{\frac{\varepsilon}{\varepsilon_0}}\right) I_0\left(\sqrt{\frac{\varepsilon}{\varepsilon_0}}\right)} \right] \quad (45)$$

where $\varepsilon_0 = D/R^2$ and $\gamma_0 = D/(R_0^2)$. For perfect absorption, $C_2 \rightarrow \infty$ in eq 45. This gives

$$\tilde{Q}(\varepsilon) = \frac{1}{\varepsilon} \left[1 - \frac{K_0\left(\sqrt{\frac{\varepsilon}{\gamma_0}}\right)}{K_0\left(\sqrt{\frac{\varepsilon}{\varepsilon_0}}\right)} \right] \quad (46)$$

Here I_0 and K_0 are the zero-order modified Bessel functions of the first and second kind, respectively. Our arbitrary capture rate (imperfect absorption) result is new, whereas the perfect absorption result is known⁶⁸ and can be trivially obtained as a limit. Both require numerical inversion.

Let us now obtain limiting expressions for long and short times. For short time, $t \rightarrow 0$, $\varepsilon \rightarrow \infty$, and the zero-order modified Bessel functions of the first and second kind can be approximated⁷⁰ as $K_0(z) \sim (\pi/(2z))^{1/2} \exp(-z)$ and $I_0(z) \sim (1/(2\pi z))^{1/2} \exp(-z)$. This results, for the finite reaction case, in

$$\tilde{Q}(\varepsilon) = \frac{1}{\varepsilon} \left[1 - \sqrt{\frac{R}{R_0}} \frac{e^{-(\varepsilon\tau_2)^{1/2}}}{\xi_2 \sqrt{\varepsilon\tau_2} + 1} \right] \quad (47)$$

where $\tau_2 = (R_0 - R)^2/D$ and $\xi_2 = 4\pi DR/[C_2(R_0 - R)]$ are 2-d quantities associated with motion and capture, respectively. Equation 47 can be inverted exactly giving

$$Q(t) = 1 - \sqrt{\frac{R}{R_0}} \left[\operatorname{erfc} \left(\frac{1}{2} \sqrt{\frac{\tau_2}{t}} \right) - e^{(1/\xi_2) + (1/\xi_2^2)(t/\tau_2)} \operatorname{erfc} \left(\frac{1}{2} \sqrt{\frac{\tau_2}{t}} + \frac{1}{\xi_2} \sqrt{\frac{t}{\tau_2}} \right) \right] \quad (48)$$

This result appears to be new but is similar to eq 29 which is the 1-d result for a point trap and initial delta-function condition. It is straightforward to recover from our analysis other known results such as a short time approximation for perfect absorption.⁴³

$$Q(t) = 1 - \sqrt{\frac{R}{R_0}} \operatorname{erfc} \left(\frac{1}{2} \sqrt{\frac{\tau_2}{t}} \right) \quad (49)$$

To obtain expressions for the long time limit, one can follow Ritchie and Sakakura⁷¹ and Taitelbaim⁴³ to obtain for imperfect absorption⁴³

$$Q(t) = 2 \left[\ln \left(\frac{R_0}{R} \right) + \frac{2\pi D}{C_2} \right] \frac{1}{\ln \left(\frac{4Dt}{R^2} \right) + \frac{4\pi D}{C_2} - 2E_\gamma} \quad (50)$$

and for instantaneous reaction⁷¹

$$Q(t) = 2 \ln \left(\frac{R_0}{R} \right) \frac{1}{\ln \left(\frac{4Dt}{R^2} \right) - 2E_\gamma} \quad (51)$$

Here $E_\gamma = 0.57722\dots$ is Euler's constant. We note that, for a partially absorbing finite circular or spherical trap with highly symmetric initial conditions, it is natural to recover known results obtained from the radiative boundary conditions in the long time limit.

4.1.2. Infinite Line Trap. Consider now an infinite line of traps along the y -axis from $-\infty$ to ∞ through $x = 0$ and initial point particles placed on an infinite line from $-\infty < y < \infty$ through $x = x_0$. This problem can be solved by using the expression for a trapping ring of radius R and an initial radially symmetric distribution of point particles given in eq 45 by replacing R_0 with at $R_0 = R + x_0$

$$\tilde{Q}(\varepsilon) = \frac{1}{\varepsilon} \left[1 - \frac{\frac{1}{2\pi D} K_0 \left([R + x_0] \sqrt{\frac{\varepsilon}{D}} \right) I_0 \left(R \sqrt{\frac{\varepsilon}{D}} \right)}{\frac{1}{C_2} + \frac{1}{2\pi D} K_0 \left(R \sqrt{\frac{\varepsilon}{D}} \right) I_0 \left(R \sqrt{\frac{\varepsilon}{D}} \right)} \right] \quad (52)$$

As $R \rightarrow \infty$, the arguments of both $K_0(z)$ and $I_0(z)$ tend to infinity

$$I_0 \left(R \sqrt{\frac{\varepsilon}{D}} \right) K_0 \left(R \sqrt{\frac{\varepsilon}{D}} \right) \simeq \frac{\sqrt{D}}{2R\sqrt{\varepsilon}}$$

$$I_0 \left(R \sqrt{\frac{\varepsilon}{D}} \right) K_0 \left((R + x_0) \sqrt{\frac{\varepsilon}{D}} \right) \simeq \frac{\sqrt{D} e^{-x_0(\varepsilon/D)^{1/2}}}{2R \left(1 + \frac{x_0}{R} \right) \sqrt{\varepsilon}}$$

$$\tilde{Q}(\varepsilon) = \frac{1}{\varepsilon} \left[1 - \frac{\int_0^\infty e^{-(\tau_1/4t) - \varepsilon t} \operatorname{erfc} \left(\sqrt{\frac{\chi}{4t}} \right) \frac{dt}{\sqrt{t}} - \frac{4}{\sqrt{\pi\chi\varepsilon}} \left[\frac{\sqrt{\tau_1}}{2} K_1(\sqrt{\tau_1\varepsilon}) - \frac{\sqrt{\tau_1 + \chi}}{2} K_1(\sqrt{(\tau_1 + \chi)\varepsilon}) \right]}{\sqrt{\pi\tau_1}\xi_1 + \int_0^\infty e^{-\varepsilon t} \operatorname{erfc} \left(\frac{1}{2} \sqrt{\frac{\chi}{t}} \right) \frac{dt}{\sqrt{t}} - \frac{2}{\varepsilon\sqrt{\pi\chi}} + \frac{2}{\sqrt{\varepsilon\pi}} K_1(\sqrt{\varepsilon\chi})} \right] \quad (54)$$

where $K_1(z)$ is the first order modified Bessel function of the second kind and $\chi = 4l^2/D$. Looking at eq 54, a result we have

Substituting these expressions into eq 52 with $C_2 = \pi RC_1$ and taking the limit as $R \rightarrow \infty$, we obtain

$$Q(t) = e^{(1/\xi_1) + (1/\xi_1^2)(t/\tau_1)} \operatorname{erfc} \left(\frac{1}{2} \sqrt{\frac{\tau_1}{t}} + \frac{1}{\xi_1} \sqrt{\frac{t}{\tau_1}} \right) + \operatorname{erf} \left(\frac{1}{2} \sqrt{\frac{\tau_1}{t}} \right) \quad (53)$$

As one might expect from the physics of the limiting process, this result is identical to the expression obtained for a single trap at the origin and an initial particle placed at $x = x_0$, eq 29. Ben-Naim et al.⁵⁴ have previously pointed out that the infinite 1-d trapping system with an imperfect trap is equivalent to a semiinfinite 1-d diffusion system. This equivalence has been used by Park et al.⁶ to explain results in a photobleaching experiment resulting from an infinite line trap.

4.1.3. Finite Line Trap: Open Trapping Surface. Thus far, we have restricted our discussion to closed trapping surfaces. In this section, we investigate the possibility of applying the trapping prescription to open trapping surfaces like a finite trap segment extending from $-l \leq y \leq l$ and passing through $x = 0$. The self-propagator is computed from $e^{-(y-y_0)^2/(4Dt)}/(4\pi Dt)$ by integrating y and y_0 from $-l$ to l and dividing by $2l$ for appropriate normalization

$$P(0, 0, t) = \frac{1}{4\pi Dt} \int_{-l}^l \int_{-l}^l e^{-(y-y')^2/4Dt} dy' dy = \frac{1}{\sqrt{4\pi Dt}} \operatorname{erf} \left(\frac{l}{\sqrt{Dt}} \right) - \frac{1}{\pi l} e^{-l^2/2Dt} \sinh \left(\frac{l^2}{2Dt} \right)$$

We see that if we take $l \rightarrow \infty$ we obtain the same self-propagator as for an infinite line of traps. The first term in the above expression cannot be transformed exactly. However, for the second term, the exact Laplace transform can be found in ref 65. If the particles are initially placed on a line from $-l$ to l through $x = x_0$ to the right of the trapping line, we can compute the homogeneous solution at the trap site:

$$P(0, x_0, t) = \frac{e^{-x_0^2/4Dt}}{8\pi l Dt} \int_{-l}^l \int_{-l}^l e^{-(y-y')^2/4Dt} dy' dy = \frac{1}{\sqrt{4\pi Dt}} e^{-x_0^2/4Dt} \operatorname{erf} \left(\frac{l}{\sqrt{Dt}} \right) - \frac{1}{\pi l} e^{-(2l^2 + x_0^2)/4Dt} \sinh \left(\frac{l^2}{2Dt} \right)$$

The result for the survival probability then is

not encountered in earlier literature, it becomes obvious that solutions for open trapping surfaces become more complex due

to missing symmetry in the problem. However, we reiterate that, as long as we obtain an expression in the Laplace domain, it is possible to invert the solution numerically.

4.2. Results in 3-d Systems. To apply the trapping prescription to 3-d problems described by simple diffusion, we need to compute the 3-d free-space propagator. The Green's function in three dimensions is a product of three 1-d Gaussian propagators and written in spherical polar coordinates as

$$G(\vec{r}, \vec{r}', t) = \frac{1}{(4\pi Dt)^{3/2}} e^{-|\vec{r}-\vec{r}'|^2/4Dt} \quad (55)$$

where $|\vec{r}-\vec{r}'|^2 = (x-x_0)^2 + (y-y_0)^2 + (z-z_0)^2$. For a spherically symmetric initial condition, $P(\vec{r}, 0) = \delta(r-R_0)/(4\pi r^2)$, the two quantities that correspond to the ν -function and the η are obtained by evaluating

$$\int_0^\pi \int_0^{2\pi} \int_0^\infty G(\vec{r}, \vec{r}', t) P(\vec{r}, 0) r'^2 \sin \theta dr' d\theta d\phi$$

resulting in

$$P(R, r; t) = \frac{1}{8\pi Rr\sqrt{\pi Dt}} (e^{-(r-R)^2/4Dt} - e^{-(r+R)^2/4Dt}) \quad (56)$$

which is in agreement with earlier results.⁶⁸

4.2.1. Spherical Trap of Finite Extent. In this subsection, the trapping problem of a spherical trapping shell and an initial spherical distribution of point particles is investigated (Figure 1). This problem seems to have important biological applications in the process of passive diffusion. In this process, small molecules may diffuse across a cell membrane. For example, in the process of photosynthesis, oxygen molecules may be absorbed by oxygen-evolving complexes embedded in the thylakoid membrane⁷² while undergoing passive diffusion through the membrane. In such a system, one might be interested in the total amount of unbound oxygen, which is a measure of energy production in this process.

To solve this problem, we follow our previous methodology. The probability densities on the spherical surfaces of radius R and R_0 are calculated as

$$P(R, R, t) = \frac{1}{8\pi R^2\sqrt{\pi Dt}} (1 - e^{-R^2/Dt})$$

$$P(R, R_0, t) = \frac{1}{8\pi RR_0\sqrt{\pi Dt}} (e^{-(R_0-R)^2/4Dt} - e^{-(R_0+R)^2/4Dt})$$

Both expressions can be Laplace transformed exactly,⁶⁵ and the particle survival probability in Laplace domain for finite reaction is

$$\tilde{Q}(\varepsilon) = \frac{1}{\varepsilon} \left[1 - \frac{\frac{1}{4\pi RR_0\sqrt{D\varepsilon}} e^{-(\varepsilon/\gamma_0)^{1/2}} \sinh \sqrt{\frac{\varepsilon}{\varepsilon_0}}}{\frac{1}{C_3} + \frac{1}{4\pi R^2\sqrt{D\varepsilon}} e^{-(\varepsilon/\varepsilon_0)^{1/2}} \sinh \sqrt{\frac{\varepsilon}{\varepsilon_0}}} \right] \quad (57)$$

where $\varepsilon_0 = D/R^2$ and $\gamma_0 = D/R_0^2$. This expression we have derived cannot be inverted directly as far as we know.

The instantaneous reaction limit, for $C_3 \rightarrow \infty$, is

$$\tilde{Q}(\varepsilon) = \frac{1}{\varepsilon} \left(1 - \frac{R}{R_0} e^{-\varepsilon^{1/2}((1/\gamma_0)^{1/2} - (1/\varepsilon_0)^{1/2})} \right) \quad (58)$$

and can be inverted exactly, giving in the time domain

$$Q(t) = 1 - \frac{R}{R_0} \operatorname{erfc} \left(\frac{1}{2} \sqrt{\frac{\tau_3}{t}} \right) \quad (59)$$

where $\tau_3 = \tau_2 = (R_0 - R)^2/D$ in agreement with refs 1, 65, and 68.

A straightforward expansion of eq 57 about $\varepsilon = 0$, followed by Laplace inversion after retaining a few terms,⁷³ gives

$$Q(t) = 1 - \frac{RC_3}{R_0(4\pi DR + C_3)} + \frac{1}{\sqrt{t}} \left(\frac{RC_3}{\sqrt{D}(4\pi DR + C_3)} - \frac{C_3^2 R}{\sqrt{D}(4\pi D + C_3)^2} \right) \quad (60)$$

As $t \rightarrow \infty$, this result reduces to a known expression

$$Q(t) = 1 - \frac{RC_3}{R_0(4\pi DR + C_3)}$$

given by Rice.¹ For perfect absorption, the asymptotic expression given in eq 60 becomes $Q(t) = 1 - R/R_0$.¹ Therefore, for finite and instantaneous reaction in 3-d, the survival probability will never reach zero. This result is expected, since, in 3-d, the probability of a diffusing particle reaching any specific point (including the starting point) as time approaches infinity is less than 1.

4.2.2. Infinite Sheet of Traps. Similarly, one can obtain also an expression for an infinite sheet in 3-d. We again start with the solution for a spherical trap given in eq 57 and replace $R_0 = R + x_0$ to obtain

$$\tilde{Q}(\varepsilon) = \frac{1}{\varepsilon} \left[1 - \frac{\frac{1}{4\pi R^2(1+x_0/R)\sqrt{D\varepsilon}} e^{-x_0(\varepsilon/D)^{1/2}}}{\frac{1}{2\pi R^2 C_1} + \frac{1}{4\pi R^2\sqrt{D\varepsilon}}} \right] \quad (61)$$

with $2\pi R^2 C_1 = C_3$. In the limit as $R \rightarrow \infty$, it is trivial to show that eq 61 becomes the 1-d result given in eq 32 after Laplace inversion.

4.2.3. Trapping Ring in 3-d. Consider, finally, a trapping ring of radius R centered at the origin in the x, y plane ($z = 0$ or $\phi = \pi/2$) and an initial point particle at $(0, 0, z)$ above the ring on the z -axis. Tedious but straightforward calculations allow us to obtain a new expression for the survival probability in the Laplace domain

$$\tilde{Q}(\varepsilon) = \frac{1}{\varepsilon} - \frac{e^{-((R^2+z^2)\varepsilon/D)^{1/2}}}{\varepsilon 4\pi D \sqrt{R^2+z^2}} \left\{ \frac{1}{C_3} + \frac{1}{(4\pi D)^{3/2}} \left[\frac{4R\varepsilon}{\sqrt{\pi D}} {}_2F_3(A) - \frac{\sqrt{2D}}{R} \left[2E_\gamma + \ln \left(\frac{16R^2}{D} \right) \right] - 2\sqrt{\pi\varepsilon} {}_1F_2(B) \right] \right\}^{-1} \quad (62)$$

where ${}_pF_q$ is the generalized hypergeometric function defined in ref 70 with $A = 1, 1; 1, 3/2, 3/2, 2; R^2\varepsilon/D$ and $B = 1/2; 1, 3/2; R^2\varepsilon/D$. This expression cannot be inverted directly and must be evaluated through numerical procedures.

5. RELATIONSHIP OF DISCRETE SINK TERM ANALYSIS TO CONTINUUM BOUNDARY CONDITION STUDIES

While most of the analysis presented so far in this and similar papers^{1,14–16,21–23,25,43,44,74} has relied on an approach to reaction-diffusion phenomena involving discrete lattice analysis as reflected in eq 10, and then its continuum limit, eq 15, when appropriate, there have been many investigations of such problems carried out by solving the diffusion equation with constant-density initial conditions and radiative or absorbing boundary conditions. A collection of the latter kind of expressions can be found in the book by Carslaw and Jaeger⁶⁸ on the subject of heat conduction. An important undertaking therefore is an investigation of the relationship of these two kinds of analysis.

Several thorough discussions of the relationship between discrete sink term analysis and continuum boundary condition studies exist in the literature. We particularly refer the reader to early discussions by Fixman,² Weiss,⁵³ Redner,¹⁶ and their collaborators, as well as to a very recent study.⁴⁶ The subject is subtle and deserving of careful comment and investigation. We report our own considerations below and postpone to a future publication⁶⁶ an analysis of some peculiarities that we have found in higher dimensions.

To study the relationship, we return to the standard elementary problem of a particle diffusing in 1-d in the presence of a single stationary trap located at the origin. The absorption process at the trap site occurs at a finite rate. A closely related problem is the study of the 1-d diffusion equation obeyed by the concentration profile $c(x, t)$, viz., $\partial c/\partial t = D(\partial^2 c/\partial x^2)$, with an initial condition that is not localized but constant density, i.e., $c(x, 0) = c_0$. The boundary condition at $x = 0$ is what is called “radiative” and employs h , the reaction rate at the boundary, through

$$\left. \frac{\partial c}{\partial x} \right|_{x=0} = hc(x, t)|_{x=0} \quad (63)$$

The solution for $x \geq 0$ is⁶⁸

$$\frac{c(x, t)}{c_0} = \operatorname{erf}\left(\frac{x}{\sqrt{4Dt}}\right) + e^{hx+h^2Dt} \operatorname{erfc}\left(\frac{x}{\sqrt{4Dt}} + h\sqrt{Dt}\right) \quad (64)$$

This is the ratio of the time-dependent density to its initial constant value.

Let us compare this result with eq 29 which is a consequence of the trapping formalism:

$$Q(t) = \operatorname{erf}\left(\frac{x_0}{\sqrt{4Dt}}\right) + e^{(C_1x_0/2D)+(C_1^2/4D)t} \operatorname{erfc}\left(\frac{x_0}{\sqrt{4Dt}} + \frac{C_1}{2}\sqrt{\frac{t}{D}}\right) \quad (65)$$

If we choose the parameters h and C_1 , each of which is particular to the approach that uses it, to be interrelated^{16,36,46,53} through

$$h = C_1/(2D)$$

where C_1 is the 1-d capture rate at the trap site in units of velocity, and replace x in the first expression by x_0 in the second, eqs 64 and 65 are found to be completely equivalent to each other!

This should be surprising at first glance, since the two quantities calculated are quite *different* from each other although both involve a trap at the origin. One is the time-dependent profile of an initially constant density. The other is the time-dependent survival fraction expressed as a function of the initial location of the particle. Is their close relationship accidental? Is it universally valid? Surely these questions deserve investigation.

5.1. Requirement on Diffusion Propagator for Equivalence. To investigate why this equivalence occurs, we write

$$c(x, t) = \int \Pi^*(x, x', t)c(x', 0) dx' \quad (66)$$

where $c(x', 0)$ is the initial distribution. For the first kind of calculation discussed in the previous subsection, we put in this result the constant initial condition $c(x', 0) = c_0$ and for the second the localized initial condition $c(x', 0) = \delta(x' - x_0)$. For the first, one obtains

$$\frac{c(x, t)}{c_0} = \int \Pi^*(x, x', t) dx' \quad (67)$$

whereas for the second, after an *additional* integration over x , we get

$$Q(x_0, t) = \Pi^*(x, x_0, t) dx \quad (68)$$

The quantity Π^* appearing in the above equations is the propagator for the problem *with the trap*, consequently, in the present case, the Green's function for the diffusion equation *in the presence of the trap*.

It is clear by an inspection of the above two equations that the equivalence we seek will occur if

$$\Pi^*(x, y, t) = \Pi^*(y, x, t) \quad (69)$$

for any x and y . We will now show that such symmetry in the full propagator will occur if the symmetry is present in the homogeneous (trap-less) propagator, which it does for the diffusion equation. Translational invariance therefore ensures the equivalence of eqs 67 and 68.

We start very generally by investigating these two quantities on a discrete lattice and inspecting eq 2 which does not presume any translational invariance. We consider a single trap at r , introduce the symbol q to denote initial placement of the particle at site q , and write, as a consequence of eq 2,

$$\tilde{P}_{m,q} = \tilde{\Psi}_{m,q} - C\tilde{\Psi}_{m,r}\tilde{P}_{r,q} \quad (70)$$

On solving this equation by putting $m = r$, we get

$$\tilde{P}_{r,q} = \tilde{\Psi}_{r,q} - C\frac{\tilde{\Psi}_{m,r}\tilde{\Psi}_{r,q}}{1 + C\tilde{\Psi}_{r,r}} \quad (71)$$

The probabilities P are really the propagators Ψ^* in the presence of the trap. We thus have the relation between propagators in the presence of the trap and those in the absence of the trap:

$$\tilde{\Psi}_{m,q}^* = \tilde{\Psi}_{m,q} - \frac{\tilde{\Psi}_{m,r}\tilde{\Psi}_{r,q}}{(1/C) + \tilde{\Psi}_{r,r}} \quad (72)$$

Switching indices m and q , we see that translational invariance in the free propagators (i.e., invariance under switching of m and q) ensures $\Psi_{m,q}^* = \Psi_{q,m}^*$. This is of course also true in the

continuum limit. We have therefore recovered the invariance relation 69 and understood why boundary condition and sink term results agree with each other.⁵³ While the invariance we have discussed holds for free diffusion, it breaks down in the presence of a potential.⁶⁶

$$C_d = (\text{Area})Dh \quad (73)$$

Here *Area* corresponds to the surface (hyper)area of the trap. In 1-d, 2-d, and 3-d, it is the number 2, the circumference of a circle of radius *R* and the surface of a sphere of radius *R*, respectively. Accordingly, $C_1 = 2Dh$,^{16,36,46,53} $C_2 = 2\pi R Dh$, and $C_3 = 4\pi R^2 Dh$.²

6. CONCLUDING REMARKS

The analysis of symmetrical systems in continua of dimensions higher than 1, a discussion of the relationship of trapping prescriptions to boundary condition studies, and a demonstration of the ease with which a unified approach leads to all of these different results are the main goals of the present paper. Additionally, the paper should serve as an accessible collection of some exact results for reaction-diffusion systems that we have found useful in understanding phenomena in areas as widely different as exciton transport in molecular crystals, sensitized luminescence, receptor cluster coalescence in cells, and epidemic spread via animal–animal interactions. The discussion focuses on static traps with noninfinite capture rate centered at a given location. Although we have derived all these results from our own unified approach (explained in section 2), some of them have appeared earlier in the literature, derived by various authors in their own contexts. To the best of our knowledge, eqs 26, 34, 45, 48, 54, 57, 60, and 62 are among the new results we presented. In our derivations, we have applied existing formalisms^{21,53} in notation most familiar to ourselves.

We would like to point out that the connection of sink-term trapping analysis to boundary condition studies is important to understand fully. It is known that, for translationally invariant particle motion in 1-d, the expression obtained from the trapping prescription is equivalent to the solution obtained from a diffusion equation.^{16,36} To obtain equivalence, the diffusion equation must satisfy proper radiative boundary conditions subject to a continuous initial condition. In the previous section, it can be seen that the equivalence holds only if the motion propagator is translationally invariant in the absence of the reaction. However, there are many reaction-diffusion systems which cannot be described by boundary value problems. It has been suggested in ref 2 that the boundary condition used to solve boundary value problems may, and perhaps should, be considered only a consequence of a particular choice of sink terms.

The higher-dimensional results we have displayed in section 4, while all for radial symmetry, are encountered quite often in applications. The idea is to convert 2-d and 3-d analysis in such cases to an effectively 1-d analysis (the radial coordinate *r* being the single variable forming the 1-d description) but with additional features not present in real 1-d situations.

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Notes

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