Spatiotemporal properties of diffusive systems with a mobile imperfect trap

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We study analytically a one-dimensional system initially uniformly filled with diffusing particles A, and a single imperfect mobile trap T initially located at the origin, $x=0$. For arbitrary values of diffusion constants D_A and D_T, and any trapping rate constant V, we calculate exactly the total rate of trapping as well as the asymptotic concentration of A's at $x=0$. For $D_A=D_T$ we also analytically derive the local rate of trapping and the concentration of A's at any point x. Characteristic length scales and extensions to higher dimensions are also discussed. [S1063-651X(98)04701-1]

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I. INTRODUCTION

The standard reaction rate theory for diffusion-limited elementary reactions of type $A+B\rightarrow C$ or $A+B\rightarrow B$ is based on many simplifying assumptions [1–3]. In Smoluchowski's approach one assumes the B species to be so diluted that the system can be considered as consisting of only a *single* particle B surrounded by a swarm of diffusing particles A. Moreover, the B is assumed to be an *immobile* sphere acting on the surrounding, freely diffusing pointlike particles A as a perfect trap (i.e., upon collision of an A with the trap reaction is certain to occur). The reaction rate is computed from the flux of A's into the sphere. The case where the diffusion constants D_A and D_B of A's and B's, respectively, are nonzero is then treated by the concept of the relative diffusion constant $D'=D_A+D_B$ applied to the above-mentioned system with an immobile trap B. Although this theory was later analyzed and improved by many researchers (for reviews see [1–3]), it has not been rigorously solved yet, and confidence in it is based mainly on its agreement with a number of experiments [2]. It is therefore important to examine physical effects associated with various assumptions this simplified theory is founded on, especially after it was demonstrated [4,5] that in restricted (i.e., low-dimensional or disordered) geometries Smoluchowski's theory must be modified to account for the self-segregation of the reactants.

In this paper we investigate a one-dimensional system in which there is a *single diffusing imperfect* trap (henceforth we shall denote it by T) surrounded by many diffusing particles A initially uniformly filling up the whole available space. Such geometry, with both T and A's mobile, was already investigated numerically by Schoonover *et al.* [6], who focused on the problem of determining the asymptotic properties of the distance between the trap and the nearest-neighbor particle A. In the present study, in turn, we concentrate on examining spatiotemporal properties of such systems. We study analytically two problems: what is the time evolution of the mean concentration of A's at any point x, and what is the reaction (or trapping) rate at time t. We determine both the local and total rate of trapping, which we denote by $R(x,t)$ and $R(t)$, respectively. Moreover, defining the trap absorptivity V as a parameter ranging between perfect trapping and no trapping, we also examine the role played by another of Smoluchowski's assumptions that reaction at the trap is inevitable.

Our study extends the well-examined case of an *immobile*, perfect or imperfect trap [5,7–11] or traps [11–13]. However, due to mathematical complexity, only a few results related to the case of *mobile* traps are available, and most of them deal with the problem of determining the survival probability of mobile particles A placed among randomly distributed diffusing traps [14]. An interesting model with a variable number of mobile traps was recently studied by Sánchez *et al.* [15]. In addition to this, the role of the $A+T\rightarrow T$ reactions with mobile traps T in the diffusion-limited fluorescence quenching in liquids was investigated by Lianos and Argyrakis [16].

The paper is organized as follows. In Sec. II, starting from a discrete description, we define the model in the continuous formalism. In Sec. III, using a coordinate system in which the trap is at rest, we calculate rigorously the total rate of trapping $R(t)$ and the concentration of particles A at a distance z from the trap, $a_T(z,t)$. These calculations are carried out for arbitrary values of V, D_T, D_A, z, and t. In Sec. IV we return to the laboratory coordinate system in which both T and A's are mobile. Using it we derive formulas for the concentration $a(x,t)$ of particles A and the local reaction rate $R(x,t)$. The key quantity employed to compute $a(x,t)$ in this limit is $f(x,t|y)$, the conditional concentration of particles A at site x provided that at the same time t the trap is located at y. We calculate its explicit asymptotic form for any values of D_T and D_A. We derive a general expression for $a(x,t)$ for $D_A=D_T$, and find its form at $x=0$, the initial location of the trap, for arbitrary values of diffusion constants. We prove that in the long-time limit $a(x,t)$ converges to a *constant*, *positive* value depending only on D_T and D_A, which means that if both the trap T and particles A are mobile, the trapping becomes inefficient. In Sec. V we present results of our numerical simulations of a discrete model, which turn out to be in excellent agreement with our theory derived for continuous space and time. In Sec. VI we investigate asymptotic properties of the depletion zone formed between the mobile trap T and particles A. Finally, Sec. VII is devoted to conclusions.

II. THE MODEL

Consider a one-dimensional lattice with a lattice constant λ_A. At time $t=0$ a single trap T is placed at $x=0$, and a swarm of diffusing particles A is uniformly distributed at the lattice sites $x_j=j\lambda_A$, $j=\pm 1,\pm 2,\ldots$, so that A's can be found on both sides of the trap. The initial concentration of particles A will be denoted by ρ_0. The evolution of the system is governed by the following rules. At subsequent intervals τ each particle A, as well as the trap T, performs a random jump. The jump length of particles A is equal to the lattice constant λ_A, and so their locations are limited to the lattice sites. However, because we want to be able to consider systems with arbitrary values of the ratio D_T/D_A, and since diffusion constants are directly related to the jump lengths, we assume that the jump length of the trap T, λ_T, can assume *any* nonnegative value. Although the same effect could be achieved by a physically more realistic assumption that the trap T and particles A have different jump rates and the same jump lengths, such modification would lead to unnecessary complications in both analytical and numerical analyses of the problem; we expect the long-time behavior of the system to depend on the values of diffusion constants, but not on the microscopic details of the jumps. We also assume that particles A do not interact among themselves in any way, so it is possible to find many of them at the same site at a time. However, upon contact with the trap, particles A may react with it and be removed from the system. The reaction probability will be denoted by κ ($0\leqslant\kappa\leqslant 1$). The case $\kappa=1$ corresponds to T being a perfect trap, and $\kappa=0$ to the absence of reaction. We assume that the reaction is the only interaction between T and A's. Therefore, for $\kappa<1$, not only can particles and the trap coexist at the same lattice site, but it is possible for them to jump over each other without any interaction. This assumption implies also that the motion of the trap is independent of the locations and velocities of particles A.

To investigate the evolution of this system we introduce the conditional concentration of particles A, $f(x,t|y)$, defined as the expected number of A's that at time t can be found at site x provided that at this time the mobile trap T is located at y. In the absence of reaction, i.e., if $x\neq y$ or $\kappa=0$, the master equation reads

$$f(x,t+\tau|y)=\frac{1}{4}[f(x-\lambda_A,t|y-\lambda_T)+f(x-\lambda_A,t|y+\lambda_T)+f(x+\lambda_A,t|y-\lambda_T)+f(x+\lambda_A,t|y+\lambda_T)]. \tag{1}$$

This equation expresses the fact that on average only half of A's from $x-\lambda_A$ or $x+\lambda_A$ will move in a single time step τ to x, and only in half of the systems in the ensemble will their motion be accompanied by the trap T arriving at y from $y-\lambda_T$ or $y+\lambda_T$.

In the long-time limit Eq. (1) can be approximated by the standard diffusion equation with coefficients $D_T=\lambda_T^2/2\tau$ and $D_A=\lambda_A^2/2\tau$. However, for $x=y$ and $\kappa>0$ one must add to it an appropriate term that will account for the reaction at the trap. It is very difficult to write such a term precisely in the discrete formalism for arbitrary values of λ_T, λ_A, and τ, especially when λ_T and λ_A are incommensurate. Therefore, although discrete formulation of the problem is directly related to many physical phenomena, it is practically intractable analytically. However, as we expect the reaction term to be proportional to f for $x=y$ and vanish at $x\neq y$, we conclude that the equation for $f(x,t|y)$ in the continuous formalism, which is more amenable to rigorous, analytical methods, takes on the form

$$\frac{\partial f}{\partial t}=D_A\frac{\partial^2 f}{\partial x^2}+D_T\frac{\partial^2 f}{\partial y^2}-V\delta(x-y)f, \tag{2}$$

where V is the trapping rate constant ranging from 0 (no trapping) to ∞ (perfect trapping). Once we have computed $f(x,t|y)$, the expected concentration $a(x,t)$ of particles A at (x,t) can be calculated from

$$a(x,t)=\int_{-\infty}^{\infty}f(x,t|y)dy. \tag{3}$$

Note that although we study a one-dimensional system, our mathematical treatment is carried out in a two-dimensional space, with x and y treated as *independent* variables and the trapping occurring along the line $y=x$. As we shall also work in two different coordinate systems, in order to avoid confusion we adopt a convention that x always denotes positions of particles A, y refers to the position of the trap T, and z denotes the relative distance between the trap and particles A; both x and y are used only in a laboratory reference system where both T and A's are mobile, whereas z is used only in the coordinate system in which the trap is at rest.

We expect both formalisms, the continuous one and the discrete one, to give the same results in the long-time limit. As we will show below, in this limit the solutions of Eqs. (2) and (3) become independent of V and converge to those obtained in the limit $V\to\infty$, unless $V=0$. Similarly we expect that the values of $f(x,t|y)$ in the discrete formulation of the problem asymptotically converge to those with $\kappa=1$, unless $\kappa=0$. Presumably there is no direct relation between other values of κ and V.

In the next two sections we shall concentrate on solving the continuous problem with the initial state given by

$$f(x,t=0|y)=\rho_0\delta(y), \tag{4}$$

which corresponds to the uniform distribution of A's, and the trap located at the origin. We will also present results of computer simulations of the discrete model, which will enable us to compare the results obtained for the two different models at short times and to investigate the transition of the discrete system to the long-time limit.

III. COORDINATE SYSTEM WITH THE TRAP AT REST

The reaction rate at the trap can be calculated most easily in the coordinate system in which the trap is at rest. Consider now such a coordinate system with the initial state made up of only *one* particle A located at a distance z_0 from the trap. Let $p(z,t)$ denote the probability of finding it at z at time t. Because in this reference system A performs a random walk with the relative diffusion constant $D'=D_T+D_A$, and can be trapped only upon arriving at $z=0$, the evolution of $p(z,t)$ is governed by an analog of Eq. (2):

$$\frac{\partial p(z,t)}{\partial t}=D'\frac{\partial^2 p(z,t)}{\partial z^2}-V\delta(z)p(z,t). \tag{5}$$

This equation was already used [17,18] to study particles diffusing in a one-dimensional system in the presence of a single, *immobile*, imperfect trap located at the origin of the system $(z=0)$, and the explicit form of $p(z,t)$ subject to the initial condition $p(z,0)=\delta(z-z_0)$ was found to be given by [18]

$$p(z,t)=\frac{1}{\sqrt{4\pi D't}}\exp\left(-\frac{(z-z_0)^2}{4D't}\right)-\frac{h}{2}\exp\left(-\frac{(|z|+|z_0|)^2}{4D't}\right)F\left(h\sqrt{D't}+\frac{|z|+|z_0|}{\sqrt{4D't}}\right), \tag{6}$$

where $F(x)\equiv\exp(x^2)\mathrm{erfc}(x)$, $\mathrm{erfc}(x)\equiv 2\pi^{-1/2}\int_x^\infty \exp(-\eta^2)d\eta$, and $h\equiv\frac{1}{2}V/D'$. Notice now that due to linearity and homogeneity of Eq. (5), its solution for arbitrary initial conditions can be obtained as the superposition of solutions of type (6). After some algebra we thus conclude that if particles A are initially uniformly distributed in the system with some concentration ρ_0, then at time t their concentration $a_T(z,t)$ measured in the reference system of the trap is given by

$$a_T(z,t)=\rho_0\left[\mathrm{erf}\left(\frac{|z|}{\sqrt{4D't}}\right)+\exp\left(\frac{-z^2}{4D't}\right)F\left(h\sqrt{D't}+\frac{|z|}{\sqrt{4D't}}\right)\right]. \tag{7}$$

In the limit $h\rightarrow\infty$, which corresponds to $V\rightarrow\infty$, we thus have

$$a_T(z,t)=\rho_0\mathrm{erf}\left(\frac{|z|}{\sqrt{4(D_T+D_A)t}}\right). \tag{8}$$

An interesting feature of Eq. (7) is that the limit $h\rightarrow\infty$ is mathematically similar to $t\rightarrow\infty$ with $z/\sqrt{t}$ fixed, and both yield the same result (8). Consequently, in the long-time limit, for any finite reaction rate V, the profile of particles A in the reference system of the mobile trap eventually approaches that of the perfect trap.

The immediate consequence of Eq. (7) is that the expected number $M(t)$ of particles A that have reacted by time t is given by

$$\begin{aligned} M(t)&=\int_{-\infty}^{\infty}[a_T(z,0)-a_T(z,t)]dz \\ &=2\rho_0[h^{-1}F(h\sqrt{D't})+2\sqrt{D't/\pi}-h^{-1}]. \end{aligned} \tag{9}$$

For the perfect trap $(h\rightarrow\infty)$ or in the long-time limit we have

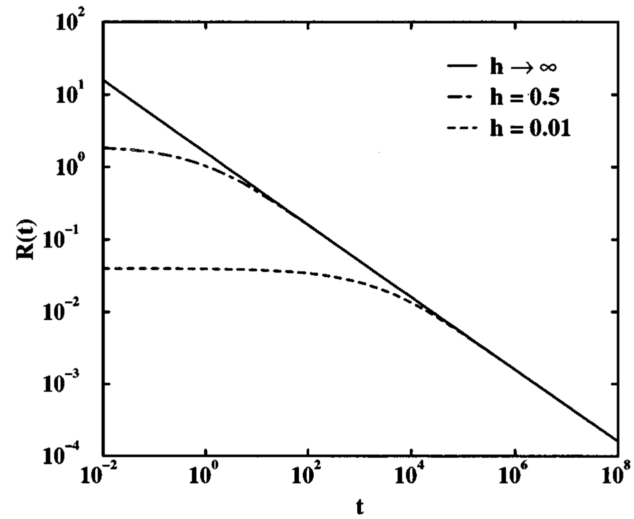

FIG. 1. The total rate of trapping $R(t)$ for $h\rightarrow\infty$ (solid line), $h=0.5$ (dot-dashed line), and $h=0.01$ (dashed line), plotted using Eq. (11). Other parameters are $D_A=D_T=\rho_0=1$.

$$M(t)=4\rho_0\sqrt{\pi^{-1}(D_T+D_A)t}. \tag{10}$$

Hence the total trapping rate at time t, $R(t)=dM(t)/dt$, reads

$$R(t)=2h\rho_0 D'F(h\sqrt{D't})=Va_T(0,t), \tag{11}$$

which asymptotically simplifies to

$$R(t)=2\rho_0\sqrt{\frac{D_T+D_A}{\pi t}}. \tag{12}$$

The above formulas rigorously confirm the conjecture of Schoonover *et al.* [6] that asymptotically $M(t)\sim t^{1/2}$ and $R(t)\sim t^{-1/2}$, and are consistent with the results of Ref. [9] obtained for the particular case $D_T=0$. The plots of $R(t)$ obtained for $D_A=D_T=1$, $\rho_0=1$, and $h=0.01$, 0.5, and $h\rightarrow\infty$ are presented in Fig. 1. Note that initially $R(t)\approx 2h\rho_0 D'$, and $R(t)$ converges to the form given in Eq. (12) for $t\gtrsim 1/(h^2D')$.

IV. LABORATORY COORDINATE SYSTEM

A. Transition to the long-time limit

Although it is possible to write down the rather complicated propagator for Eq. (2), integrals that must then be computed to evaluate $f(x,t|y)$ or $a(x,t)$ cannot be worked out in a closed form except when one of the diffusion constants vanishes.

It is possible, however, to derive in a closed form the long-time asymptotics of $f(x,t|y)$ for any values of V, D_T, and D_A. To this end notice that the ratio $a(x,t)/\rho_0$ is dimensionless and D_T/D_A, $x/\sqrt{D_T t}$, and $V\sqrt{t/D_T}$ are the only mutually independent dimensionless combinations of x, t, D_T, D_A, and V. Therefore, $a(x,t)$ must assume the form

$$a(x,t)=\rho_0\mathcal{F}(D_T/D_A,x/\sqrt{D_T t},V\sqrt{t/D_T}) \tag{13}$$

for which the limits $V\to\infty$ and $t\to\infty$, $x/\sqrt{t}=\text{const}$ are equivalent. Therefore, to investigate the long-time behavior of the system described by Eq. (2) with any nonzero V it suffices to concentrate on the limiting case $V\to\infty$ of the perfect trap, which we shall do henceforth. This scaling form reveals also two important properties of $a(x,t)$ at $x=0$. First, $a(x,t)/a(0,t)\to 1$ as $t\to\infty$, for any x. Second, in the limit of the perfect trap, $V\to\infty$, $a(0,t)$ must be independent of t. Later we shall confirm these conclusions by explicit calculations carried out for the case $D_A=D_T$.

B. Reformulation of the problem

If $V\to\infty$, upon contact with the trap, particles A inevitably react. Therefore, the problem of solving Eq. (2) reduces in this limit to the one of solving the standard diffusion equation

$$\frac{\partial f}{\partial t}=D_A\frac{\partial^2 f}{\partial x^2}+D_T\frac{\partial^2 f}{\partial y^2} \tag{14}$$

with the boundary condition

$$x=y\Rightarrow f(x,t|y)=0 \tag{15}$$

imposed by the instantaneous reaction at $x=y$.

Because the reaction renders the solutions of Eq. (14) nonanalytic along the line $y=x$, it is convenient to solve Eqs. (14) and (15) with the initial state composed of particles A uniformly distributed with concentration ρ_0 only on a half-line $x>0$,

$$f_1(x,t=0|y)=\rho_0 H(x)\delta(y), \tag{16}$$

where $H(x)$ denotes the Heaviside step function, and we added the index ''1'' to f to distinguish it from the solution obtained for the full initial state (4), which will be given below.

Equations (15) and (16) imply

$$x\leqslant y\Rightarrow f_1(x,t|y)=0, \tag{17}$$

so that Eq. (3) can be rewritten as

$$a_1(x,t)=\int_{-\infty}^{x} f_1(x,t|y)dy, \tag{18}$$

where $a_1(x,t)$ denotes the concentration of A's at (x,t) for the initial distribution (16).

C. The form of $f_1(x,t|y)$ for $x>y$

Assuming D_T, $D_A>0$ we can symmetrize the form of Eq. (14) by replacing y with a new variable

$$\tilde{y}\equiv y\sqrt{D_A/D_T}. \tag{19}$$

Now Eq. (14) takes on a simpler form,

$$\frac{\partial \tilde{f_1}}{\partial t}=D_A\left(\frac{\partial^2 \tilde{f_1}}{\partial x^2}+\frac{\partial^2 \tilde{f_1}}{\partial \tilde{y}^2}\right) \tag{20}$$

and the initial and boundary conditions turn into

$$\tilde{f_1}(x,t=0|\tilde{y})=\rho_0\tan(\alpha)H(x)\delta(\tilde{y}) \tag{21}$$

and

$$\tilde{y}=x\tan(\alpha)\Rightarrow \tilde{f_1}(x,t|\tilde{y})=0, \tag{22}$$

respectively, where α is defined through

$$\tan(\alpha)=\sqrt{D_A/D_T}. \tag{23}$$

Equation (20) with the ''half-line'' initial state (21) and ''absorbing'' boundary condition (22) can be solved by the method of images [19]. The solution reads

$$\tilde{f_1}(x,t|\tilde{y})=\tilde{\Phi}(x,\tilde{y},t)-\tilde{\Phi}(c_1x+c_2\tilde{y},c_2x-c_1\tilde{y},t), \tag{24}$$

where

$$\tilde{\Phi}(x,\tilde{y},t)=\frac{\rho_0}{4\sqrt{\pi D_T t}}\exp\left(\frac{-\tilde{y}^2}{4D_A t}\right)\operatorname{erfc}\left(\frac{-x}{\sqrt{4D_A t}}\right) \tag{25}$$

denotes the solution of Eqs. (20) and (21) *without* taking into account the boundary condition (22) imposed by trapping, and c_1 and c_2 are some constants related to D_T and D_A through

$$c_1\equiv\cos(2\alpha)=\frac{D_T-D_A}{D_T+D_A},$$

$$c_2\equiv\sin(2\alpha)=2\frac{\sqrt{D_T D_A}}{D_T+D_A}. \tag{26}$$

Note that in accordance with the method of images, the second term on the right-hand side of Eq. (24) is simply the image of $\tilde{\Phi}(x,\tilde{y},t)$ rotated by the angle 2α around the origin of the x-y plane.

Upon return to the original variables we come to

$$f_1(x,t|y)=\Phi(x,y,t)-\Phi(c_1x+(1-c_1)y,(1+c_1)x-c_1y,t), \tag{27}$$

where

$$\Phi(x,y,t)=\frac{\rho_0}{4\sqrt{\pi D_T t}}\exp\left(\frac{-y^2}{4D_T t}\right)\operatorname{erfc}\left(\frac{-x}{\sqrt{4D_A t}}\right). \tag{28}$$

Equation (27) was derived for nonzero diffusion constants. One can easily verify that for $D_T=0$, $D_A>0$

$$f_1(x,t|y)=\rho_0\delta(y)\operatorname{erf}\left(\frac{x}{\sqrt{4D_A t}}\right)H(x), \tag{29}$$

and for $D_A=0$, $D_T>0$

$$f_1(x,t|y)=\frac{\rho_0 H(x)}{\sqrt{4\pi D_T t}}\left[\exp\left(\frac{-y^2}{4D_T t}\right)-\exp\left(\frac{-(2x-y)^2}{4D_T t}\right)\right]. \tag{30}$$

D. Concentration profiles and the local trapping rate

Having obtained the form of $f_1(x,t|y)$, we can now insert Eq. (27) into Eq. (18), and compute $a_1(x,t)$. However, due to the form of the upper limit in Eq. (18), $a_1(x,t)$ can be investigated analytically only when $D_T = D_A$, which implies $c_1 = 0$. The explicit form of $a_1(x,t)$ for this case reads

$$a_1(x,t) = \frac{1}{4}\rho_0\left[\operatorname{erfc}^2(-\xi) - \frac{2}{\sqrt{\pi}}\exp(-\xi^2)i\operatorname{erfc}(-\xi)\right], \quad (31)$$

where $i\operatorname{erfc}(z) \equiv \int_z^\infty \operatorname{erfc}(\eta)d\eta = \pi^{-1/2}\exp(-z^2) - z\operatorname{erfc}(z)$, and

$$\xi \equiv x/\sqrt{4D_T t}. \quad (32)$$

The mean local rate of trapping, $R_1(x,t)$, is equal to the average number of particles A that are being trapped at (x,t), and is defined through

$$\frac{\partial a_1(x,t)}{\partial t} = D_A\frac{\partial^2 a_1(x,t)}{\partial x^2} - R_1(x,t). \quad (33)$$

Using Eq. (31) its explicit form for $D_T = D_A$ is found to read

$$R_1(x,t) = \frac{1}{2\sqrt{\pi}}\cdot\frac{\rho_0}{t}\exp(-\xi^2)i\operatorname{erfc}(-\xi). \quad (34)$$

The scaling plot of $R_1(x,t)$ will be given in Sec. IV E below.

Another interesting feature of the system is the dependence of $a_1(x,t)$ at the origin $(x=0)$ on the values of the diffusion constants D_T and D_A. Using Eqs. (18), (27), and the integral

$$\int_0^\infty \exp(-\eta^2)\operatorname{erfc}(a\eta)\,d\eta = \frac{1}{\sqrt{\pi}}\arctan(1/a) \quad (35)$$

we find that

$$a_1(0,t) = \frac{\rho_0}{2}\left[\frac{1}{2} - \frac{1}{\pi}\frac{D_T + D_A}{D_T - D_A}\arctan\left(\frac{D_T - D_A}{2\sqrt{D_T D_A}}\right)\right]. \quad (36)$$

We can see that the concentration of particles A at the origin is *independent of time*. Following our dimensional analysis we conclude that this rather surprising effect can be observed only in the limit of the perfect trap. In a more realistic case of systems containing an unperfect trap we expect that $a_1(0,t)$ would depend on time, decreasing from ρ_0 at $t=0$, and approaching the above result as $t\to\infty$. In Sec. V we present simulation data for a discrete model that show that in this case Eq. (36) is valid only in the long-time limit. Another interesting feature of $a_1(x,t)$ given by Eq. (36) is that it drops to 0 only if either D_T or D_A goes to 0. Moreover, it actually depends only on the ratio D_A/D_T of the diffusion constants, attains the maximal value $\frac{1}{2}\rho_0(\frac{1}{2} - 1/\pi) \approx 0.09\rho_0$ for $D_T = D_A$, and is not sensitive to interchanging the values of D_T and D_A. In Fig. 2 we present the semilogarithmic plot of $a_1(0,t)/\rho_0$, computed from Eq. (36), as a function of D_A/D_T.

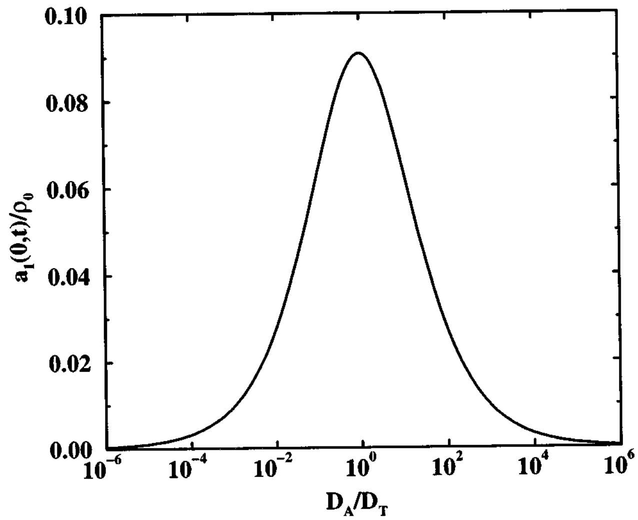

FIG. 2. The average relative concentration of particles A at the origin, $a_1(0,t)/\rho_0$, as a function of D_A/D_T, plotted using Eq. (36). The maximal value is $1/4 - 1/2\pi \approx 0.09$.

E. Particles A on both sides of the trap

We can now generalize our results for the initial condition (4) made up of particles A uniformly distributed on both sides of the trap simply by using the principle of superposition, which implies that for any values of D_A and D_T:

$$f(x,t|y) = f_1(x,t|y) + f_1(-x,t|-y), \quad (37)$$

$$a(x,t) = a_1(x,t) + a_1(-x,t), \quad (38)$$

$$R(x,t) = R_1(x,t) + R_1(-x,t). \quad (39)$$

Explicit forms of $a(x,t)$ and $R(x,t)$ for $D_A = D_T$ can be found using Eqs. (31) and (34), respectively. In Fig. 3 we present a scaling plot of $R(x,t)$ and $R_1(x,t)$ for $D_A = D_T$. $R_1(x,t)$ reflects the asymmetry of the initial condition (16).

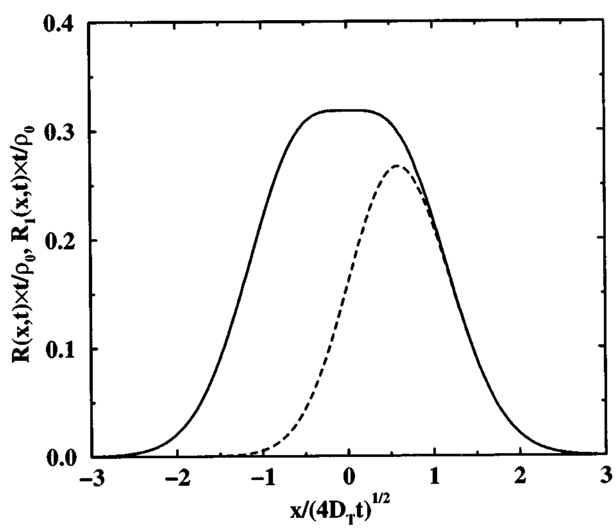

FIG. 3. The scaling plot of the local trapping rate as a function of $\xi = x/\sqrt{4D_T t}$ for $D_A = D_T$ and two different initial conditions. The solid line represents $R(x,t)t/\rho_0$ [full initial condition (4)] and the dashed one shows $R_1(x,t)t/\rho_0$ [half-line initial condition (16)]. $x=0$ is the initial location of the trap.

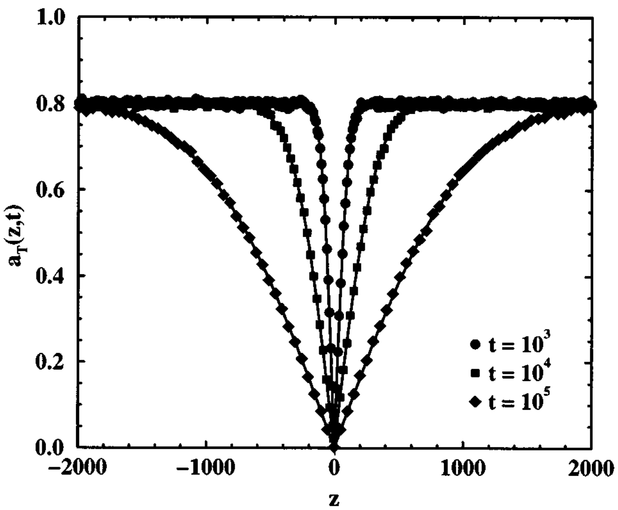

FIG. 4. The average concentration of particles A seen from the reference system attached to the trap at time $t=10^3$ (circles), 10^4 (squares), and 10^5 (diamonds). The parameters are $D_T/D_A=5$, $\rho_0=0.8$ and $L=10\,001$. The solid lines were computed from Eq. (8).

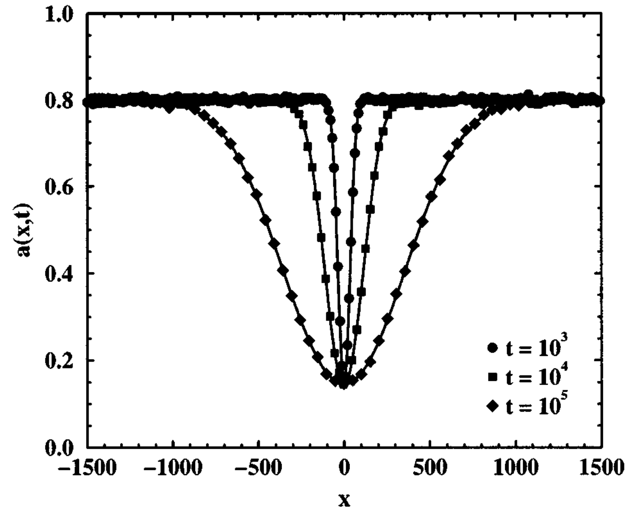

FIG. 5. The average concentration of particles A in the laboratory coordinate system. The parameters are $D_T/D_A=1$, $\rho_0=0.8$ and $L=5001$. The solid lines were computed from Eqs. (38) and (31). Note the constant value of $a(x,t)$ at $x=0$. As $t\to\infty$, it becomes the value of $a(x,t)$ for any x.

V. COMPUTER SIMULATIONS

To examine the properties of discrete systems we performed numerical simulations based on the cellular-automata model of diffusion [20]. In our approach λ_A is fixed, but the trap can perform off-lattice jumps of any length λ_T, hence any value of D_T/D_A can be studied in simulations. This goal is achieved by storing information about positions of particles using integer arithmetic for particles A, and floating-point arithmetic for the mobile trap T. We used the full initial condition (4) with the initial concentration $\rho_0=0.8$ and the probability of reaction $\kappa=1$. The value of D_T/D_A varied from 0.1 to 10; we also made additional runs for $D_T=0$ (immobile trap), and $D_A=0$ (immobile particles A). The lattice size L varied from 5001 for $D_T/D_A\leq 1$ to 12 001 sites for $D_T/D_A=10$. The results were averaged over 31 000 independent runs.

The concentration of particles A as seen from the reference system attached to the mobile trap, $a_T(z,t)$, is depicted in Fig. 4 for times $t=10^3$, 10^4, and 10^5, and $D_T/D_A=5$. This corresponds to $\lambda_T=\sqrt{5}$, i.e., to the trap performing off-lattice jumps. The solid lines were computed using Eq. (8). A similar plot of the concentration of particles A in the laboratory coordinate system, for $D_T/D_A=1$, is shown in Fig. 5; here the solid lines were computed from Eqs. (38) and (31). In both cases the agreement between the continuous theory and the discrete simulations is excellent for all times.

The difference between $a_T(z,t)$ and $a(x,t)$ is most pronounced at the origin. While $a_T(0,t)$ asymptotically goes to 0, $a(0,t)$ converges to a positive value. Figure 6 shows $a(0,t)$ as a function of time. We can see that initially it exhibits sharp fluctuations, caused by the discreteness of the system, but quickly approaches the asymptotic value of $2a_1(0,t)\approx 0.1404$, as predicted theoretically in Eq. (36) and represented by the dashed line. Simulations performed for other values of D_T/D_A gave similar results.

VI. CHARACTERISTIC LENGTH SCALES

A number of recent papers have been devoted to the problem of finding a quantitative description of the self-segregation observed in diffusion-limited bimolecular reactions [21,22]. In analogy with the Smoluchowski approximation one can try to investigate a simplified problem of segregation between a single trap and surrounding particles A. Three characteristic measures of this separation were suggested, but explicit calculations were performed only for the case where the trap is immobile.

One of these lengths, the so called θ distance, is defined [8] as the distance from the trap to the point r_θ at which the

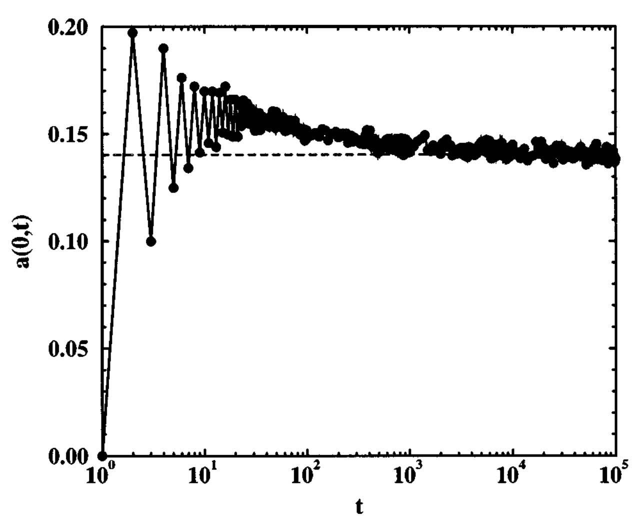

FIG. 6. The average concentration of particles A at the origin of the system as a function of time t. The circles represent the results of numerical simulations, and the dashed line corresponds to the theoretical value $2a_1(0,t)\approx 0.1404$ computed from Eq. (36). The parameters are $D_T/D_A=2$, $\rho_0=0.8$, and $L=7001$.

concentration $a_T(z,t)$ of A's is equal to a given fraction θ $(0<\theta<1)$ of its bulk value. Another one, the characteristic minimum distance $x_{\min}$, is defined [23] by requirement that on average there should be exactly one particle A at a distance $z\leqslant x_{\min}$ from the trap,

$$\int_0^{x_{\min}} a_T(z,t) J_d(z)\, dz = 1, \tag{40}$$

where $J_d(z)$ is an appropriate Jacobian in d dimensions. Both of these lengths are related to $a_T(z,t)$, the concentration of particles A in the reference system in which the trap is at rest. The results of Refs. [7–10] where $a_T(z,t)$ and $r_\theta(t)$ were computed for the static trap in $d=1,2,3$ can be therefore easily applied to the case of mobile trap, yielding $r_\theta(t)\propto t^{1/2}$ for $d=1$, $r_\theta(t)\propto t^{\theta/2}$ for $d=2$, and $r_\theta(t)\sim$const for $d=3$. Note the nonuniversal behavior of the θ distance in two-dimensional systems. Using the form of $a_T(z,t)$ derived in Refs. [7–10] we can also immediately conclude that for arbitrary values of D_A and D_T there is $x_{\min}\propto t^{1/4}$ for $d=1$, $x_{\min}\propto(\ln t)^{1/2}$ for $d=2$, and $x_{\min}\sim$const for $d=3$.

The third characteristic length is the average distance from the trap to the nearest particle A [6–10,24]. However, the problem of finding its properties when both the trap and particles A are mobile remains unsolved. Since $f(x,t|y)$ turned out to be the essential quantity used to determine this distance if either D_T or D_A is zero [7,25], we hope that our exact, general formula for $f(x,t|y)$ will prove useful in solving this problem for arbitrary values of D_T and D_A.

VII. DISCUSSION AND CONCLUSIONS

We have studied the behavior of a one-dimensional system initially uniformly filled with diffusing particles A, and a single diffusing trap T placed at $x=0$. We found that the total rate of trapping can be most easily calculated in the reference system attached to the mobile trap. This reduces the problem to that of the trap being immobile and particles A possessing relative diffusion constant $D'=D_T+D_A$, i.e., in the way suggested by Smoluchowski. As this trick can be applied to homogeneous systems of any space dimensionality, the results of Ref. [7–10], where the immobile trap was studied, can be immediately used to calculate the total reaction rate $R(t)$ and characteristic lengths $r_\theta(t)$ and $x_{\min}(t)$ for any d.

Using dimensional analysis and explicit calculations for $D_A=D_T$, we found that as $t\to\infty$, the mean concentration of particles A at any point x asymptotically goes to a constant value. We showed that this quantity is greater than zero except when the trap or the particles are immobile. In this sense we can call the trapping of mobile particles by a mobile trap inefficient. It is reasonable to expect this property to hold also in higher dimensions.

Note finally that in our model the local reaction rate $R(x,t)$ *is not* proportional to the product of the local concentration $a(x,t)$ and the probability to find the trap T at (x,t). This implies that despite its simplicity, our system cannot be described in terms of classical, mean-field theories.

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