Nearest-neighbor distance at a single mobile trap

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We consider a one-dimensional system with an infinite number of identical particles A diffusing in the presence of a single diffusing perfect trap B. We study numerically the average distance $\langle l(t)\rangle$ from the trap to the nearest unreacted A, and confirm the claim that in the long-time limit $\langle l(t)\rangle \propto t^{\alpha}$, where α is an exponent depending on the ratio of diffusivities D_A and D_B of the particles A and the trap B, respectively. We also confirm the validity of a conjecture for the value of α, but show that it should be limited to a specific choice of the initial distribution of particles A. [S1063-651X(98)03711-8]

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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 [1,2]. Because of the many-body nature of the problem, rigorous analysis of the depletion zone formed around the reactants is a formidable task, and only phenomenological theories and results of numerical simulations are available [1,2]. Therefore, analogous to the Smoluchowski approximation, which is commonly used in the reaction rate theory [3–5], one often investigates a simplified problem of segregation between a single trap B and particles A surrounding it [6–14].

In the original Smoluchowski model there is only a *single*, *immobile* spherical particle B surrounded by a swarm of freely diffusing particles A [3–5]. B is assumed to be a trap (sink) that swallows up any A that hits its surface ($A+B\rightarrow B$), and the reaction rate is computed from the flux of particles A into the sphere. The case where the diffusion constants D_A and D_B of the particles A and the trap B, respectively, are nonzero is then treated using the concept of the effective diffusion constant $D'=D_A+D_B$ applied to the above-mentioned system with an immobile trap B. Widespread use of this grossly simplified model in the literature of chemical physics [3] suggests the importance of studying its more realistic extensions [3–5].

A simple extension of the Smoluchowski model consists in allowing the trap B to diffuse. One is tempted to reduce it to the original Smoluchowski model by working in a coordinate system in which the trap is at rest. However, as was already pointed out by Noyes [4], in this reference system displacements of particles A are correlated, since the motion of the trap changes all of its relative distances to molecules A by the same amount. This effect is most pronounced when particles A are *static*; in the coordinate system attached to the mobile trap, all of their displacements will be the same, and by no means is it obvious that these correlations can be neglected in an analysis of the depletion zone, especially in low-dimensional systems.

The distribution of particles about the trap in low-dimensions can be studied through $\langle l(t)\rangle$, the average distance between the trap B and the particle A that is nearest to it. This measure of segregation was calculated exactly for the original Smoluchowski model (static B and mobile A's) in dimensions $d=1,2,3$ [6,8–11], and for a similar model with a single, *mobile* trap B, and *immobile* particles A for $d=1$ [7]. In the particular case of one-dimensional systems it was found that $\langle l(t)\rangle \propto t^{1/4}$ if the trap is static and particles A are mobile, and $\langle l(t)\rangle \propto t^{1/2}$ in the opposite case of a mobile trap and immobile particles A. The asymptotic distribution function for $\langle l(t)\rangle$ was obtained in Refs. [6,8].

The intermediate, general case of a one-dimensional system in which both the trap B and particles A are mobile has been studied numerically by Schoonover *et al.* [12]. This conceptually simple extension of the Smoluchowski model makes its rigorous mathematical treatment very difficult, since now the many-body problem cannot be reduced to a two-body one, and no analytical method of treating it has been suggested. Schoonover *et al.* [12] conjectured that asymptotically

$$\lim_{t\rightarrow\infty}\frac{\ln\langle l(t)\rangle}{\ln t}=\alpha, \tag{1}$$

with

$$\alpha(D)=\frac{1}{\pi}\arctan(\sqrt{1+2D}), \tag{2}$$

where

$$D\equiv D_B/D_A. \tag{3}$$

The form of Eq. (2) has been suggested based on a heuristic analogy with an exponent obtained in a related problem of the survival probability of a single particle A surrounded by two traps B [15]. It is exact for $D=0$ and $D\rightarrow\infty$, yielding $\alpha=1/4$ and $\alpha=1/2$, respectively. For other values of D, an agreement with the results of computer simulations has been claimed. However, the agreement was only qualitative. In fact, the numerical results in Ref. [12] were consistently smaller by about 5% from those predicted by Eq. (2). Moreover, those simulations were performed for relatively small systems and only for a particular initial condition in which particles A were placed on only one side of the trap B.

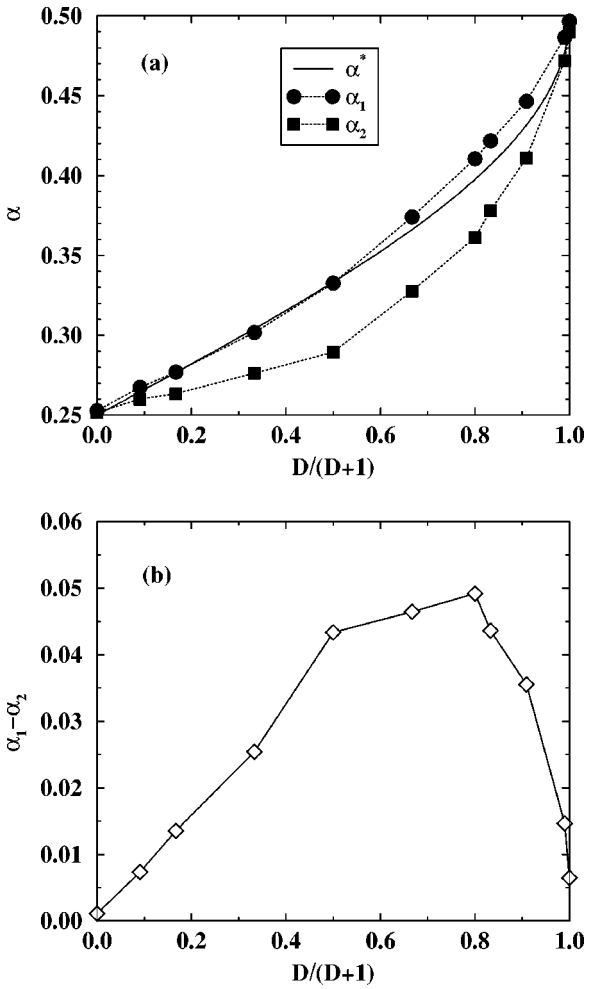

FIG. 1. (a) Comparison of the values of α, estimated from simulations, α_1 for the ''one-side'' and α_2 for the ''two-side'' initial conditions, respectively, with the values predicted in Eq. (2), denoted by α^*; (b) the difference between α_1 and α_2 as a function of $D/(D+1)$. The parameters are $t=10^5$, $L>7000$, and $N=31\,000$.

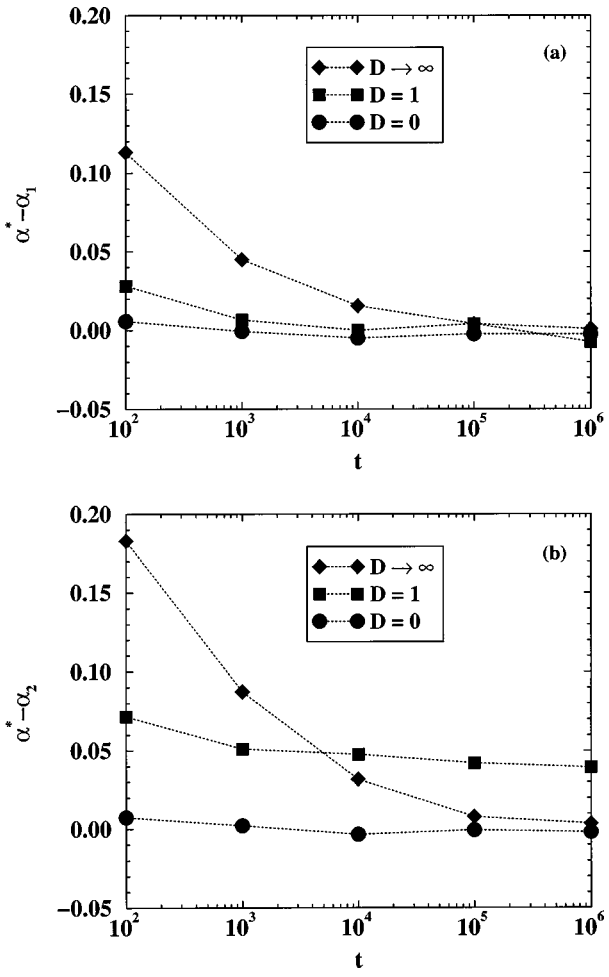

FIG. 2. The convergence of the exponent α to the asymptotic value α^*, computed from Eq. (2) for $D\to\infty$, $D=1$, and $D=0$, and two different initial conditions: (a) particles A distributed on one side of the trap (α_1), (b) particles A distributed on both sides of the trap (α_2). Note that α_2 does not converge to α^* for $D=1$. The parameters are: $t_{\max}=10^6$, $L=15\,001$, and $N=10\,000$.

In this Brief Report we present results of refined simulations of the generalized Smoluchowski model in one dimension, carried out for longer times and larger systems, for two different initial conditions: the ''one-side'' initial condition where particles A are placed on one side of the trap, and the ''two-side'' initial state in which particles A are distributed on both sides of the trap B. We find that when both the trap and the particles A are mobile, the value of α is not only nonuniversal, but also depends on the initial conditions. For the initial condition studied in Ref. [12], i.e., when particles A are initially deposited on one side of the trap, we show that within statistical error, α assumes values close to or even a little larger than those predicted in Eq. (2). However, in the case where they are distributed on both sides of the trap B, the values of α become significantly smaller than those predicted in Eq. (2). A similar dependence on the initial conditions was also found in a recent study of a system with a single particle diffusing in the presence of many diffusing traps [16].

Our computer simulations are based on the cellular-automata model of diffusion in one dimension (see Ref. [14] for a detailed description). At time $t=0$ a single trap B was placed at $x=0$, and particles A were randomly distributed either on both sides of it (the two-side initial condition), or only for $x>0$ (the one-side initial condition). The initial concentration a_0 of particles A was equal to 0.8, which is the value used in Ref. [12]. At subsequent time steps each particle A, as well as the trap B, performed a random jump. The jump length of particles A was fixed and equal to the lattice constant λ_A; the jump length of the trap could assume any value $\lambda_B\geqslant 0$ so that we could perform our simulations for arbitrary values of $D=D_B/D_A=\lambda_B^2/\lambda_A^2$. Upon contact with the trap, particles A immediately reacted and were removed from the system. In particular, if the trap was jumping from some x_0 to $x_0+\lambda_B$, then it reacted with any particle A located at any $x_0\leqslant x\leqslant x_0+\lambda_B$. To investigate the case $D\to\infty$ we performed a separate set of simulations with a special algorithm assuming that particles A are immobile ($D_A=0$) and that the trap diffuses ($D_B>0$). The simulations were carried out for $t_{\max}=10^5$ time steps. For each D the results were averaged over $N=31\,000$ independent runs. The number L of the lattice sites was chosen to be large enough to eliminate the boundary effects and varied from 7001 for $D\leqslant 1$ to 38 001 for $D=100$. These values are significantly bigger than those employed in Ref. [12], where

$N=500$, $L=2000$, and $t_{\max}=5000$ were used.

The results of our simulations are presented in Fig. 1(a). The estimated asymptotic values of α, for the case where particles A are initially deposited on one side of the trap, are shown as circles and will be denoted as α_1. The corresponding values for the two-side initial condition are depicted as squares and will be denoted as α_2. Both α_1 and α_2 were calculated for succesive times; for a given value of t they were determined from the slope of $\ln\langle l(t)\rangle$, as a function of $\ln t$, using the data from the interval $t/10<\tau\leqslant t$. The asymptotic values conjectured in [12] [see Eq. (2)] are plotted as a solid line and will be denoted as α^*. The results are plotted as a function of $D/(D+1)$ to allow for showing data points at $D=0,\infty$. For $D=0$, both α_1 and α_2 go to 1/4, and we obtained $\alpha_1=0.252$ and $\alpha_2=0.253$; for $D\to\infty$, both α_1 and α_2 converge to 1/2 and we obtained $\alpha_1=0.496$ and $\alpha_2=0.492$. In Fig. 1(b) we plot the difference $\alpha_1-\alpha_2$ as a function of $D/(D+1)$ estimated at time $t=10^5$. It shows that this difference assumes values much larger than statistical error (which will be discussed below), and goes to 0 only as $D\to0$ or $D\to\infty$.

Our estimations of α_1 and α_2 can be affected by two kinds of errors. The statistical errors, resulting from averaging over a finite number of samples N, can be estimated using the χ-square fitting or by performing several independent simulations with fixed parameters of the system; both methods lead to the conclusion that the accuracy of our simulations for a given time t amounts to at least two significant digits. For example, in the case $D=1$, the 99% confidence intervals for α_1 and α_2, computed using the χ-square fitting, gave (0.327,0.333) and (0.291,0.297), respectively, whereas Eq. (2) gives $\alpha^*=1/3$. The other difficulty is related to the fact that we extrapolate the asymptotic values of α_1 and α_2 using the results obtained for finite values of time t. To determine the relevance of this factor, in Figs. 2(a) and 2(b) we show the time evolution of $\alpha^*-\alpha_1(t)$ and $\alpha^*-\alpha_2(t)$, respectively, for three characteristic values of $D=0,1,\infty$, and using higher values of time steps and chain length, i.e., $t_{\max}=10^6$, $L=15\,001$, and $N=10\,000$. We can see that these quantities assume almost constant values (within statistical error) quite quickly, especially for small values of D, which suggests that extrapolation errors in our simulations are sufficiently small. These figures also show that the values of $\alpha_1(t)$ and $\alpha_2(t)$ for small values of t tend to underestimate their asymptotic values, which explains why the values of α_1, obtained in Ref. [12] for time $t=5000$, were smaller than expected.

We suggest that the sensitivity of α to the initial conditions is related to the fact that in each case the major contribution to the ensemble average of the nearest-neighbor A-B distance $\langle l(t)\rangle$ comes from entirely different realizations of the system. In the case where particles A are distributed on one side of the trap, the most important contribution to the average comes from the systems in which the trap moves in the direction opposite to the location of particles A; however, if particles A were distributed on both sides of the trap, this kind of motion would make the trap diffuse deep into a region densely occupied by particles A, and so the corresponding nearest-neighbor distance $l(t)$ would be very small. The above heuristic argument can easily be extended and used to prove that, in general, $\alpha_1\geqslant\alpha_2$. It remains a challenge to find a rigorous relation between α_1 and α_2 for any $0<D<\infty$.

In summary, we performed extensive simulations of a one-dimensional system with a single, mobile, perfect trap B, interacting with many diffusing particles A. We found that the value of the exponent α, which characterizes the asymptotic properties of the distance between the trap and the nearest particle A, depends on the initial conditions. Its conjectured form (2) can be used only when particles A are distributed on one side of the trap; in the case where they can be found on both sides of B, Eq. (2) overestimates the value of α by up to 15%, with the difference diminishing as $D\to0$ or $D\to\infty$.

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