Trapping of diffusing particles by clusters of absorbing disks on a reflecting wall with disk centers on sites of a square lattice

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A simple approximate formula is derived for the rate constant that describes steady-state flux of diffusing particles through a cluster of perfectly absorbing disks on the otherwise reflecting flat wall, assuming that the disk centers occupy neighboring sites of a square lattice. A distinctive feature of trapping by a disk cluster is that disks located at the cluster periphery shield the disks in the center of the cluster. This competition of the disks for diffusing particles makes it impossible to find an exact analytical solution for the rate constant in the general case. To derive the approximate formula, we use a recently suggested approach [A. M. Berezhkovskii, L. Dagdug, V. A. Lizunov, J. Zimmerberg, and S. M. Bezrukov, *J. Chem. Phys.* **136**, 211102 (2012)], which is based on the replacement of the disk cluster by an effective uniform partially absorbing spot. The formula shows how the rate constant depends on the size and shape of the cluster. To check the accuracy of the formula, we compare its predictions with the values of the rate constant obtained from Brownian dynamics simulations. The comparison made for 18 clusters of various shapes and sizes shows good agreement between the theoretical predictions and numerical results. © 2013 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4790370>]

I. INTRODUCTION

This paper focuses on trapping of diffusing point particles by clusters of perfectly absorbing circular disks located on the otherwise reflecting flat wall. This problem arises, for example, in theoretical treatments of various biological processes such as ligand binding to cell-surface receptors, transport of ions and molecules through membrane channels and transporters, etc. All these processes involve a step of finding by diffusing particles right trapping sites on the membrane surface. The efficiency of finding increases with the number of these sites, however, the presence of multiple traps in the vicinity of each other also affects the effective trapping due to their competition for diffusing particles. This is particularly important in the case when receptors or transporters cluster together or form multimeric complexes (dimers, trimers, etc.).

The goal of this work is to find the rate constant, which characterizes the steady-state flux of the particles trapped by a cluster, as a function of the cluster shape and size. Trapping by clusters is an obviously many-body problem, since the disks located at the cluster periphery shield the disks in the center of the cluster from the particles. Many-body effects make it impossible to develop an exact analytical theory of the steady-state trapping rate in the general case. Development of approximate approaches to the problem was pioneered by Goldstein and Wiegel.¹ One can learn about sophisticated formalisms suggested to deal with the many-body effects in this problem in papers cited in Ref. 2.

Recently, we proposed a simple approach to the problem, which is based on the replacement of the cluster of perfectly absorbing disks by an effective uniform spot, which is partially absorbing.³ Using this approach we derived an approximate formula for the rate constant k_N of a N -disk cluster assuming that disk centers occupy neighboring sites of a triangular lattice.³ We compared the values of the rate constant predicted by the formula with those obtained from Brownian dynamics simulations for 14 clusters of various shapes and found good agreement between the two.

The question naturally arises whether this good agreement between the theory and simulations is an artifact of our choice of the triangular lattice. To shed some light on this question, in the present paper we use this approach to derive an approximate formula for k_N in the case where the disk centers occupy neighboring sites of a square lattice. We compare the values of k_N predicted by the formula and obtained from Brownian dynamics simulations for 18 clusters of various shapes over wide ranges of the cluster sizes. The results presented in Fig. 1 and Table I demonstrate good agreement between the theoretical predictions and numerical results.

Though our motivation came from numerous observations of clustering of receptors, ion channels, and other transporters on the cell surface,^{4–6} one can also use the obtained expression for the rate constant to find the capacitance of a cluster of metallic disks on a flat dielectric surface. The point is that there is a general relation between the rate constant k

FIG. 1. Comparison of the rate constants predicted by Eqs. (2.11) and (3.2) (solid curves) with the rate constant values obtained from Brownian dynamics simulations (symbols). Panels (a)–(d) show the results for linear, rectangular, cross-shaped, and gulf-shaped clusters, respectively.

of a diffusion-limited reaction and the capacitance C of the trapping body, $k = 4\pi DC$,⁷ where D is the diffusion constant of the particle. Therefore, the capacitance C_N of a cluster of N metallic disks is $C_N = k_N/(4\pi D)$.

The outline of this paper is as follows. We derive the formula for the rate constant in Sec. II. In Sec. III, we compare the values of the rate constant given by the formula with those obtained from Brownian dynamics simulations. Some concluding remarks are made in the final Sec. IV.

II. THEORY

Consider trapping of diffusing particles by a cluster formed by N perfectly absorbing, non-overlapping disks of radius a . Disk centers are located in the neighboring sites of a square lattice of period l , $l \geq 2a$. To find the trapping rate constant k_N of the cluster, we use the approach proposed in Ref. 3. The key steps of this approach are (i) the replacement of the cluster by a uniform partially absorbing spot and (ii) the use of the Collins-Kimball-like formula for the rate constant. The Collins-Kimball-like formula is an analog of the Collins-Kimball formula,⁸ which provides an exact solution for the steady-state rate constant of a partially absorbing spherical trap, k_{CK} . The formula gives k_{CK} in terms of the Smoluchowski rate constant of a perfectly absorbing sphere, k_{Sm} , and the chemical rate constant k , which is the rate con-

stant of the partially absorbing sphere in the limiting case of infinitely fast diffusion,⁸

$$k_{CK} = \frac{k_{Sm}k}{k_{Sm} + k}. \quad (2.1)$$

For a spherical trap of radius R ,

$$k_{Sm} = 4\pi DR, \quad (2.2)$$

and

$$k = \kappa A_{sph} = 4\pi R^2 \kappa, \quad (2.3)$$

where κ is the surface trapping rate and $A_{sph} = 4\pi R^2$ is the area of the sphere.

The Collins-Kimball-like formula used in our further analysis is

$$k_N = \frac{k_D \kappa A}{k_D + \kappa A}, \quad (2.4)$$

where k_D is the trapping rate constant of the perfectly absorbing spot, A is the spot area, and κ is the effective trapping rate of the spot surface. This formula has been used in the case of trapping by a partially absorbing circular disk on the otherwise reflecting flat wall,⁹ where k_D is given by the Hill formula for the rate constant of a perfectly absorbing circular disk of radius a ,¹⁰

$$k_D = k_H = 4Da. \quad (2.5)$$

TABLE I. Maximum relative errors in predictions of the rate constants k_N , Eq. (2.11), and k_D , Eq. (2.7).

Cluster	Error in k_N in %	Error in k_D in %
12.9	4.3
7.6	2.7
6.6	1.1
2.2	1.2
1.5	0.64
0.82	0.78
3.5	2.9
2.7	2.7
1.1	0.8
2.2	1.3
2.5	1.9
1.6	3.1
2.9	3.4
2.4	2.3

TABLE I. (Continued.)

Cluster	Error in k_N in %	Error in k_D in %
4.3	4.4
4.5	4.0
3.7	3.0
3.3	2.2

The rate constant k_N , Eq. (2.4), considered as a function of κ monotonically increases with the trapping rate from its minimum value $k_N = \kappa A$, as $\kappa \rightarrow 0$, to its maximum value $k_N = k_D$, as $\kappa \rightarrow \infty$ (perfectly absorbing spot).

To find k_N one needs to know the shape of the spot, k_D , and κ . We take that the spot is a unification of N squares of the square lattice of period l , centers of which are occupied by the centers of the disks forming the cluster. In Fig. 1 and Table I, these squares are shown by dashed lines. The area A of the spot is

$$A = Nl^2. \quad (2.6)$$

We find the rate constant k_D of the perfectly absorbing spot using the approximate formula,¹¹

$$k_D = \left(\frac{32}{\pi^2} AP \right)^{1/3} D, \quad (2.7)$$

where P is the spot perimeter. The expression in Eq. (2.7) can be considered as a generalization of the Hill formula, Eq. (2.5).

Finally, we assume that the trapping rate κ of the spot surface is identical to the effective trapping rate of an infinite flat surface covered by perfectly absorbing circular disks of radius a arranged in a square lattice of period l , $l \geq a$. An approximate expression for κ is found in Ref. 12. It gives the effective trapping rate as a function of the surface fraction σ occupied by the disks, $\sigma = \pi a^2/l^2 = \pi a^2 n$, where $n = l^{-2}$ is the disk concentration on the surface. As l increases, the disk surface fraction monotonically decreases from its maximum value $\sigma_{\max} = \pi/4$ when disks are in contact to zero as $l \rightarrow \infty$. At sufficiently large l ($\sigma \rightarrow 0$), the disks do not interact with

each other, and

$$\kappa = k_1 n = \frac{4D}{\pi a} \sigma, \quad l \rightarrow \infty, \quad \sigma \rightarrow 0, \quad (2.8)$$

where $k_1 = k_H$ is the Hill rate constant for an isolated disk given in Eq. (2.5). A general formula for κ , which is applicable over the entire range of σ , $0 < \sigma \leq \pi/4$, is given by¹²

$$\kappa = \frac{4D}{\pi a} \sigma f(\sigma), \quad f(\sigma) = \frac{1 + 1.75\sqrt{\sigma} - 2.02\sigma^2}{(1 - \sigma)^2}. \quad (2.9)$$

Function $f(\sigma)$ monotonically increases with σ from unity at $\sigma = 0$ to its maximum value at $\sigma = \sigma_{\max} = \pi/4$, which is much larger than unity.

As the lattice period l tend to infinity, both σ and κ tend to zero. As a result, the rate constant in Eq. (2.4) takes the form

$$k_N = \kappa A = \frac{4D}{\pi a} \sigma A = 4DaN = k_1 N, \quad (2.10)$$

which shows that as $l \rightarrow \infty$, k_N reduces to the sum of the rate constants of N isolated disks, as it must be since well separated disks do not affect each other. A general expression for the rate constant, which is applicable over the entire range of the lattice period l and, hence, σ , can be obtained by substituting into Eq. (2.4) the expressions for A , k_D , and κ given in Eqs. (2.6), (2.7), and (2.9) respectively. This leads to

$$k_N = \frac{4DaN}{f(\sigma)^{-1} + [2\pi^2 N^2 / (P/l)]^{1/3} a / l}. \quad (2.11)$$

This formula is the main result of the present paper. It shows how k_N depends on the number of disks in the cluster, the disk radius, distance l between the centers of neighboring disks, which is the lattice period, disk surface fraction $\sigma = \pi a^2 / l^2$, and the cluster perimeter P . The formula allows one to analyze the dependence of k_N not only on the size of the cluster, but also on its shape, since clusters of the same size, but different shapes, may have different perimeters.

III. NUMERICAL RESULTS

To check the accuracy of the approximate formulas for the rate constants k_N and k_D , given by Eqs. (2.11) and (2.7), respectively, we compare their predictions with the values of the rate constants obtained from Brownian dynamics simulations. In simulations, we find the mean lifetime τ of a particle diffusing in a cavity of volume V containing a cluster of perfectly absorbing disks or a perfectly absorbing spot on its otherwise reflecting wall. If the particle starting position is uniformly distributed over the cavity volume, the rate constant k is given by the ratio of the cavity volume to the mean lifetime,

$$k = \frac{V}{\tau}. \quad (3.1)$$

We ran simulations in a cubic cavity of size $8 \times 8 \times 8$. The disk radius was $a = 5 \times 10^{-2}$. Particle diffusion constant was unity, $D = 1$. The time step was $\Delta t = 10^{-6}$, so that the mean length step was $\sqrt{6\Delta t} = \sqrt{6} \times 10^{-3}$. For each cluster and perfectly absorbing spot, the mean lifetime was found by

averaging over 5×10^4 trajectories. We found that the relative errors of our simulation results did not exceed 2% for perfectly absorbing spots and 5% for clusters of perfectly absorbing disks.

Simulations were run for 18 clusters of various shapes, which are shown in Fig. 1 and Table I. For each cluster, the distance between the centers of neighboring disks varied from $l = 2a$ to $l = 20a$. In Fig. 1, the numerical results are shown by symbols, whereas the solid curves represent the l -dependences, which follow from Eq. (2.11). As l increases from $2a$ to infinity, the ratio $k_N/(4Da)$ monotonically increases from

$$\left. \frac{k_N}{4Da} \right|_{l=2a} = \frac{N}{f(\pi/4)^{-1} + 2[(\pi/4)^2 N^2 a / P]^{1/3}} \quad (3.2)$$

at $l = 2a$ to N , as $l \rightarrow \infty$.

In Fig. 1(a), we compare analytical and numerical results for linear clusters. The perimeter of a linear (l) N -disk cluster, $P_N^{(l)}$, is given by

$$P_N^{(l)} = 2(N + 1)l. \quad (3.3)$$

Substituting this into Eq. (2.11), we arrive at a general formula for the rate constant of a linear N -disk cluster,

$$k_N^{(l)} = \frac{4DaN}{f(\sigma)^{-1} + [\pi^2 N^2 / (N + 1)]^{1/3} a / l}. \quad (3.4)$$

One can see very good agreement between the analytical and numerical results over the entire range of l for clusters of 5 and more disks. For smaller clusters, Eq. (3.4) predicts larger values of the rate constant than the values obtained from the simulations. As N increases, the maximum relative error decreases (see Table I). It is worth mentioning that for a two-disk cluster an exact solution for the rate constant k_2 has been obtained by Strieder (two identical disks) and Saddawi and Strieder (two disks of arbitrary radii) in the papers cited in Ref. 2. In contrast to our approximate solution, Eq. (3.4), which for $N = 2$ takes the form $k_2 = 8Da/[f(\sigma)^{-1} + (4\pi^2/3)^{1/3} a/l]$, the exact solution is extremely complicated. For clusters with $N = 2$ and 3, Eq. (3.4) predicts non-monotonic l -dependences of the rate constant with the maximum values that are slightly larger than $4DaN$, which is obviously wrong. This happens at large values of l where the rate constants are close to $4DaN$. Therefore, the relative errors of the theoretical predictions at large l do not exceed the maximum relative errors at $l = 4a$, which are 12.9% for $N = 2$ and 7.6% for $N = 3$ (see Fig. 1(a) and Table I).

In Ref. 3, we studied trapping by linear clusters of absorbing disks, assuming that the disk centers occupy neighboring sites of a triangular (Δ) lattice, and obtained the following formula for the rate constant $k_{N,\Delta}^{(l)}$:

$$k_{N,\Delta}^{(l)} = \frac{4DaN}{f_{\Delta}(\sigma_{\Delta})^{-1} + [\pi^2 N^2 / (N + 1/2)]^{1/3} a / l}, \quad (3.5)$$

where $\sigma_{\Delta} = (2\pi/\sqrt{3})(a/l)^2$ is the surface fraction occupied by disks of radius a arranged in a triangular lattice of period

l , $l \geq 2a$, and $f_{\Delta}(\sigma_{\Delta})$ is the analog of $f(\sigma)$ in Eq. (2.9) for a triangular lattice of absorbing disks,¹²

$$f_{\Delta}(\sigma_{\Delta}) = \frac{1 + 1.62\sqrt{\sigma_{\Delta}} - 1.36\sigma_{\Delta}^2}{(1 - \sigma_{\Delta})^2}. \quad (3.6)$$

Although the formulas in Eqs. (3.4) and (3.5) are different, they give practically the same values of the rate constant. Maximum deviation of the ratio of the rate constants $k_N^{(l)}/k_{N,\Delta}^{(l)}$ from unity does not exceed 3.7% for $N = 2$, 2.5% for $N = 3$, 1.7% for $N = 4$, 1.2% for $N = 5$, and is less than 1% for longer clusters, $N \geq 6$.

The results for rectangular N -disk clusters are shown in Fig. 1(b). The perimeter of the $m \times n = N$ rectangular (r) cluster $P_{m \times n}^{(r)}$ is

$$P_{m \times n}^{(r)} = 2(m + n)l. \quad (3.7)$$

Substituting this into Eq. (2.11), we obtain a general formula for the rate constant of a rectangular ($m \times n$)-disk cluster,

$$k_{m \times n}^{(r)} = \frac{4Damn}{f(\sigma)^{-1} + [\pi^2 m^2 n^2 / (m + n)]^{1/3} a / l}. \quad (3.8)$$

One can see that this formula accurately predicts the rate constant over the entire range of l even for the smallest rectangular cluster of size 2×2 .

Comparison of the analytical and numerical results for clusters of exotic shapes is given in Figs. 1(c) (cross-shaped clusters) and 1(d) (gulf-shaped clusters). One can see that the rate constant values predicted by Eq. (2.11) are in good agreement with the numerical results.

We summarized the maximum relative errors of the values of the rate constant k_N predicted by Eq. (2.11) in Table I. In this table, we also give the relative errors of the theoretically predicted rate constants of perfectly absorbing spots of different shapes. One can see that the formula for k_D , Eq. (2.7), works reasonably well even for the spots of exotic shapes.

IV. CONCLUDING REMARKS

In our recent paper,³ we proposed a general approach, which allows one to analyze trapping of diffusing particles by a cluster of non-overlapping perfectly absorbing disks located on the otherwise reflecting flat wall. In Ref. 3, the approach is used to find the rate constant that describes the steady-state flux of diffusing particles through a disk cluster assuming that the disk centers occupy neighboring sites of a triangular lattice. In the present paper, we use the approach to find the rate constant in the situation when the disk centers occupy neighboring sites of a square lattice. This rate constant, Eq. (2.11), is the main result of the present paper. The formula in Eq. (2.11) shows how the rate constant depends on the size and shape of the cluster. To be more specific, the formula shows how the rate constant depends on the number of disks in the cluster, the disk radius, the distance between centers of neighboring disks, and the perimeter of the equivalent spot.

The dependence on the spot perimeter leads to the dependence of the rate constant on the cluster shape. For example, one can compare trapping by linear and rectangular

clusters containing the same number of identical disks with the same distance between the disk centers. Using Eqs. (3.8) and (3.4), one can find the ratio of the rate constants $k_N^{(l)}$ and $k_{m \times n}^{(r)}|_{mn=N}$,

$$\frac{k_N^{(l)}}{k_{m \times n}^{(r)}|_{mn=N}} = \frac{f(\sigma)^{-1} + [\pi^2 N^2 / (m + n)]^{1/3} a / l}{f(\sigma)^{-1} + [\pi^2 N^2 / (N + 1)]^{1/3} a / l}. \quad (4.1)$$

The ratio shows that $k_N^{(l)} > k_{m \times n}^{(r)}|_{mn=N}$ when $m, n \geq 2$, i.e., linear clusters absorb diffusing particles more efficiently than rectangular ones. This is a consequence of the fact that shielding effects are more pronounced in rectangular clusters. Since the sum $m + n|_{mn=N}$ has a minimum at $m^* = n^* = \sqrt{N}$, the ratio in Eq. (4.1) reaches a maximum for square clusters, in which shielding is the most effective.

To summarize, this paper shows that the approach proposed in Ref. 3 to account for shielding effects in trapping by clusters of perfectly absorbing disks works equally well when the disk centers occupy neighboring sites of either a triangular or square lattice.

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