Trapping of diffusing particles by small absorbers localized in a spherical region

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ABSTRACT

We study trapping of particles diffusing in a spherical cavity with an absorbing wall containing small static spherical absorbers localized in a spherical region in the center of the cavity. The focus is on the competition between the absorbers and the cavity wall for diffusing particles. Assuming that the absorbers and, initially, the particles are uniformly distributed in the central region, we derive an expression for the particle trapping probability by the cavity wall. The expression gives this probability as a function of two dimensionless parameters: the transparency parameter, characterizing the efficiency of the particle trapping by the absorbers, and the ratio of the absorber-containing region radius to that of the cavity. This work is a generalization of a recent study by Krapivsky and Redner [J. Chem. Phys. **147**, 214903 (2017)] who considered the case where the absorber-containing region occupies the entire cavity. The expression for the particle trapping probability is derived in the framework of a steady-state approach which, in our opinion, is much simpler than the time-dependent approach used in the above-mentioned study.

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I. INTRODUCTION

There is substantial interest in using near-infrared light for diagnostic purposes, in particular, to probe human tissues for localized heterogeneities such as tumors. Despite that human tissues are turbid media, transport of near-infrared photons in them are reasonably well described by the diffusion equation. Being motivated by the studies on absorption of photons diffusing in turbid media,^{1–11} Krapivsky and Redner¹² recently considered trapping of diffusing particles by dilute static absorbers randomly and uniformly distributed in a spherical cavity. They assumed that a particle touching the cavity wall exits the cavity and never comes back, and they modelled this by considering the wall as a perfectly absorbing boundary for diffusing particles. The focus was on whether the particle is trapped by the absorbers inside the cavity or exits, i.e., trapped by the cavity wall.

Krapivsky and Redner describe trapping of particles diffusing inside the cavity containing N static spherical absorbers of radius a , which is small compared to the cavity radius R , $a \ll R$, using the effective-medium approximation.^{13–22} In this approximation, a micro-heterogeneous medium with absorbers is replaced by a uniformly absorbing medium characterized by an effective trapping rate, K , which is given by¹²

$$K = k_{sm}n = 3DaN/R^3, \quad (1.1)$$

where $k_{sm} = 4\pi Da$ and $n = N/V_R$ are the Smoluchowski rate constant and the absorber number density in the cavity, with D and $V_R = 4\pi R^3/3$ denoting the particle diffusivity and the cavity volume (see Fig. 1), respectively. Assuming that the particle starting point is uniformly distributed inside the cavity, Krapivsky and Redner showed that the particle exit probability is determined by a single dimensionless

FIG. 1. Schematic representation of a sphere of radius R with the absorbing wall containing N small spherical absorbers of radius a , $a \ll R$; all absorbing surfaces are shown by bold lines [panel (a)]. Its effective-medium equivalent is shown in panel (b).

parameter λ

$$\lambda = N \frac{a}{R}. \quad (1.2)$$

They derived an expression for the exit probability, denoted by $E(\lambda)$, which can be written as

$$E(\lambda) = \frac{1}{\lambda} [\sqrt{3\lambda} \coth(\sqrt{3\lambda}) - 1]. \quad (1.3)$$

Here, we generalize the Krapivsky-Redner model. To make the model more relevant for the characterization of the tissue absorptive anomalies, we relax the assumption that the particle reaching the boundary of the spherical region of radius R containing the absorbers never returns to this region. We consider a process occurring in a larger spherical cavity of radius R_w , $R_w \geq R$, and impose the absorbing boundary condition on the external cavity wall at radius R_w (see Fig. 2). When the particle exits the region containing absorbers, it continues its diffusion and may return to the region with absorbers many times. As a consequence, the probability that the particle is trapped by the absorbers increases. Thus, the Krapivsky-Redner result in Eq. (1.3) provides an upper boundary for the probability that the particle is trapped by the cavity wall located at R_w .

The main result of the present work is an expression giving the latter probability, denoted by $P_w(\lambda, R/R_w)$, as a function of two dimensionless parameters, λ , and the radius ratio,

FIG. 2. Schematic representation of a spherical cavity of radius R_w with the absorbing wall surrounding a smaller spherical region of radius R (shown by a dashed line) that contains N spherical absorbers of radius a ($a \ll R \leq R_w$), which is located in the center of the cavity; all absorbing surfaces are shown by bold lines [panel (a)]. In panel (b), the smaller spherical region with the traps is replaced by its effective-medium equivalent shown in grey.

R/R_w ,

$$P_w(\lambda, R/R_w) = \frac{E(\lambda)}{\lambda E(\lambda)(1 - R/R_w) + 1}. \quad (1.4)$$

One can see that, as the radius ratio decreases from unity to zero, the probability $P_w(\lambda, R/R_w)$ decreases from its maximum value at $R_w = R$ given by the Krapivsky-Redner result for the exit probability,

$$P_w(\lambda, 1) = E(\lambda), \quad (1.5)$$

to its minimum value when $R_w \rightarrow \infty$

$$P_w(\lambda, 0) = \frac{E(\lambda)}{\lambda E(\lambda) + 1} = \frac{1}{\lambda} \left(1 - \frac{1}{\sqrt{3\lambda}} \tanh(\sqrt{3\lambda}) \right). \quad (1.6)$$

The latter is the probability of the particle escape to infinity in the infinite medium.

It is worth noting that the parameter λ has a simple physical interpretation.²² Since the mean lifetime of a particle diffusing in a medium, characterized by the trapping rate K , is $1/K$, the characteristic distance travelled by the particle in such a medium, l , is given by

$$l = \sqrt{D/K} = \sqrt{R^3/(3a\lambda)}, \quad (1.7)$$

where we have used the expression for K in Eq. (1.1). The ratio of the radius R of the trap-containing region to this distance, R/l , characterizing the region transparency is equal to $\sqrt{3\lambda}$

$$\frac{R}{l} = \sqrt{3N \frac{a}{R}} = \sqrt{3\lambda}. \quad (1.8)$$

The region is transparent for diffusing particles when $l \gg R$, leading to $\lambda \ll 1$. In the opposite limiting case of the absorbing region, $l \ll R$, and hence $\lambda \gg 1$.

A condition separating the two limiting cases is $\lambda = 1$, i.e., $N = R/(3a)$. Thus, the trap-containing region is absorbing when the number of traps, N , significantly exceeds the ratio R/a , $N \gg R/a$. It is informative to compare this constraint on the number of traps with the number of traps corresponding to dense packing (dp), which is given by $N_{dp} \propto (R/a)^3$. A comparison shows that the region can be absorbing even in the case of diluted traps. This happens when the number of traps satisfies the inequalities $R/a \ll N \ll (R/a)^3$.²² In a sense, this is similar to the Berg-Purcell result for trapping of diffusing ligands by circular receptors on the cell surface:²³ the cell surface becomes perfectly absorbing well before the surface fraction occupied by the receptors approaches unity.

The outline of this paper is as follows: The expression for the probability $P_w(\lambda, R/R_w)$ that the particle is trapped by the cavity wall [Eq. (1.4)], which is the main result of this work, is derived in Sec. II. Its discussion and some conclusions are given in Sec. III.

II. DERIVATION

Consider a particle diffusing in a spherical cavity of radius R_w with a perfectly absorbing wall. In the center of the cavity, there is a spherical region of radius R , $R \leq R_w$, which is uniformly absorbing with the particle trapping rate equal to K [see

Fig. 2(b)]. The particle can be trapped either in this region or by the cavity wall located at R_w . Our focus is on the probability that the particle is trapped by the wall on the condition that the particle starting point is uniformly distributed over the absorbing region. We find this probability, denoted by $P_w(\lambda, R/R_w)$, in two steps. First, we find the trapping probability by the wall for a particle starting at a distance r_0 from the cavity center, $r_0 \leq R$, denoted by $P_w(r_0)$. Then, we find $P_w(\lambda, R/R_w)$ by averaging $P_w(r_0)$ over r_0

$$P_w(\lambda, R/R_w) = \frac{3}{R^3} \int_0^R P_w(r_0) r_0^2 dr_0. \quad (2.1)$$

To find the trapping probability $P_w(r_0)$, consider a steady state where particles are constantly injected at a distance r_0 from the cavity center, $r_0 \leq R$. The injected particles are uniformly distributed over the spherical surface of radius r_0 . The steady-state concentration of the particles, denoted by $c(r)$, satisfies

$$\frac{D}{r^2} \frac{d}{dr} \left(r^2 \frac{dc}{dr} \right) - KcH(R-r) + \frac{J}{4\pi r_0^2} \delta(r-r_0) = 0, \quad (2.2)$$

subject to the boundary conditions

$$\left. \frac{dc}{dr} \right|_{r=0} = c(R_w) = 0. \quad (2.3)$$

Here, J is the particle injection rate, $H(z)$ is the Heaviside step-function, and $\delta(z)$ is the delta-function. Dividing both sides of Eq. (2.2) by D and introducing the notation j for the injected flux J divided by $4\pi D$,

$$j = \frac{J}{4\pi D}, \quad (2.4)$$

we can write this equation as

$$\frac{1}{r^2} \frac{d}{dr} \left(r^2 \frac{dc}{dr} \right) - \frac{c}{l^2} H(R-r) + \frac{j}{r_0^2} \delta(r-r_0) = 0, \quad (2.5)$$

where the length l is defined in Eq. (1.7).

The steady-state flux of particles trapped by the cavity wall, denoted by J_w , is given by

$$J_w = -4\pi D R_w^2 \left. \frac{dc}{dr} \right|_{r=R_w}. \quad (2.6)$$

The probability $P_w(r_0)$ is the ratio of this flux to the injected flux J

$$P_w(r_0) = \frac{J_w}{J} = -\frac{R_w^2}{j} \left. \frac{dc}{dr} \right|_{r=R_w}. \quad (2.7)$$

Thus, to find $P_w(r_0)$, we have to calculate the flux J_w by solving Eq. (2.5).

When the radius r satisfies $R < r < R_w$, Eq. (2.5) simplifies and reduces to

$$\frac{d}{dr} \left(r^2 \frac{dc}{dr} \right) = 0. \quad (2.8)$$

Solving this equation with the absorbing boundary condition at $r = R_w$, $c(R_w) = 0$, we find

$$c(r) = j_w \left(\frac{1}{r} - \frac{1}{R_w} \right), \quad (2.9)$$

where the positive constant j_w is related to the flux J_w , entering Eq. (2.7), by the relationship

$$j_w = \frac{J_w}{4\pi D}. \quad (2.10)$$

When the radius r lies in between the injection radius r_0 and the absorbing region radius R , $r_0 < r < R$, Eq. (2.5) takes the form

$$\frac{1}{r^2} \frac{d}{dr} \left(r^2 \frac{dc}{dr} \right) - \frac{c}{l^2} = 0. \quad (2.11)$$

Its solution is given by

$$c(r) = \frac{1}{r} \left[A \sinh\left(\frac{r-r_0}{l}\right) + B \cosh\left(\frac{r-r_0}{l}\right) \right], \quad (2.12)$$

where A and B are constants which can be expressed in terms of the flux j_w using the continuity of the concentration $c(r)$ and its derivative at $r = R$. The result is

$$A = -j_w \left[\frac{l}{R_w} \cosh\left(\frac{R-r_0}{l}\right) + \left(1 - \frac{R}{R_w}\right) \sinh\left(\frac{R-r_0}{l}\right) \right], \quad (2.13)$$

$$B = j_w \left[\left(1 - \frac{R}{R_w}\right) \cosh\left(\frac{R-r_0}{l}\right) + \frac{l}{R_w} \sinh\left(\frac{R-r_0}{l}\right) \right]. \quad (2.14)$$

For the values of r located in between the injection radius, r_0 , and the origin, $r < r_0$, the concentration $c(r)$ also satisfies Eq. (2.11), but this time with the reflecting boundary condition at the origin, $dc/dr|_{r=0} = 0$ [see Eq. (2.3)]. Here, the solution is given by

$$c(r) = \frac{C}{r} \sinh\left(\frac{r}{l}\right), \quad (2.15)$$

where C is a constant which can be expressed in terms of the flux j_w using the requirement of the particle concentration continuity at $r = r_0$. The result is

$$\begin{aligned} C &= \frac{B}{\sinh(r_0/l)} \\ &= \frac{j_w}{\sinh(r_0/l)} \left[\left(1 - \frac{R}{R_w}\right) \cosh\left(\frac{R-r_0}{l}\right) + \frac{l}{R_w} \sinh\left(\frac{R-r_0}{l}\right) \right]. \end{aligned} \quad (2.16)$$

As follows from Eqs. (2.7) and (2.10), the probability $P_w(r_0)$ is given by

$$P_w(r_0) = \frac{J_w}{J} = \frac{j_w}{j}. \quad (2.17)$$

To find the ratio j_w/j and hence the probability $P_w(r_0)$, we use the flux conservation condition at $r = r_0$, according to which we have

$$j = r_0^2 \left(\left. \frac{dc}{dr} \right|_{r_0-\varepsilon} - \left. \frac{dc}{dr} \right|_{r_0+\varepsilon} \right), \quad \varepsilon \rightarrow 0. \quad (2.18)$$

This leads to

$$P_w(r_0) = \frac{j_w}{j} = \frac{(l/r_0) \sinh(r_0/l)}{\cosh(R/l) \{1 - (R/R_w) [1 - (l/R) \tanh(R/l)]\}}. \quad (2.19)$$

Averaging this probability over the particle starting distance from the origin [see Eq. (2.1)], we arrive at the main result of this work

$$P_w(\lambda, R/R_w) = \left(3/R^3\right) \int_0^R P_w(r_0) r_0^2 dr_0 \\ = \frac{3[1 - (l/R) \tanh(R/l)]}{(R/l)^2 \{1 - (R/R_w)[1 - (l/R) \tanh(R/l)]\}}. \quad (2.20)$$

One can check that this is identical to the expression for the trapping probability in Eq. (1.4) using the relation $R/l = \sqrt{3}\lambda$, Eqs. (1.8) and (1.3) for $E(\lambda)$.

III. DISCUSSION AND CONCLUSIONS

In this work, we study trapping of a diffusing particle by dilute absorbers randomly and uniformly distributed in a spherical region of radius R located in the center of a spherical cavity of larger radius R_w with absorbing wall, $R_w \geq R$ (see Fig. 2). It is assumed that the particle starting point is uniformly distributed in the absorber-containing region. The focus is on the particle trapping probability by the wall. The main result of this work is the expression in Eq. (1.4). It shows how this probability, denoted by $P_w(\lambda, R/R_w)$, depends on the radii ratio, R/R_w , and the dimensionless parameter, λ , defined in Eq. (1.2), which characterizes the transparency of the absorber containing region [see Eq. (1.8)]. As the cavity radius, R_w , increases at a fixed value of the absorbing region radius, R , the probability $P_w(\lambda, R/R_w)$ decreases from the exit probability, $E(\lambda)$ [Eq. (1.3)], obtained by Krapivsky and Redner¹² for the case of $R_w = R$, to the particle escape probability in the infinite medium given by Eq. (1.6). The dependence of the probability $P_w(\lambda, R/R_w)$ on the radii ratio is illustrated in Fig. 3 for 3 values of the transparency parameter λ : $\lambda = 0.1, 1, 10$.

Finally, it is worth noting that we derive the probability $P_w(\lambda, R/R_w)$ using the steady-state approach, which is quite different from the time-dependent approach used by Krapivsky and Redner to obtain the exit probability, $E(\lambda)$. Following the latter approach, they first found the time-

dependent solution for the particle concentration in the region, which describes the decay of the initial uniform concentration. This solution was then used to find the time-dependent particle flux trapped by the absorbing cavity wall. The fraction of exiting particles, $E(\lambda)$, was found by integrating this flux over time from zero to infinity. In our opinion, the steady-state approach is much simpler than its time-dependent counterpart.

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FIG. 3. Probability of the particle trapping by the cavity wall, $P_w(\lambda, R/R_w)$, as a function of the radii ratio, R/R_w , for three value of the transparency parameter λ : $\lambda = 0.1, 1, 10$, from the top to bottom.