


## Non-self-averaging in random trapping transport: The diffusion coefficient in the fluctuation regime

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The nonstationary diffusion of particles in a medium with static random traps or sinks is considered. The question of the self-averaging of the diffusion coefficient (or, equivalently, of the mean-square displacement) is addressed for the fluctuation regime in the long-time limit. The property of self-averaging is needed for the result of a single measurement to be representative and reproducible. It is demonstrated that the diffusion coefficient of the surviving particles is a strongly non-self-averaging quantity: In a  $d$ -dimensional system its reciprocal standard deviation grows with time exponentially  $\approx \exp[\text{const}_{d,1} t^{d/(d+2)}]$ . The same result is reproduced in the “normalized” formulation “per one survivor on average.” The case when all the particles, both the survivors and the trapped ones, are contributing to the diffusion coefficient and its variance is considered also. Non-self-averaging is demonstrated for this case as well, the fluctuations of the diffusion coefficient being of the same order as its average value. The critical dimension, above which the mean-field result becomes exact, is infinite—due to the drastic difference between the classes of trajectories, upon which the corresponding results are being built. In high dimensions the strong non-self-averaging of survivors is preserved. For the case of all the particles taken into account, the nonstrong non-self-averaging is retained for any finite dimension. However, for  $d \rightarrow \infty$  the limiting value of the reciprocal standard deviation, calculated for all the particles, decreases to zero. This signifies restoration of the self-averaging in some sense. In all the cases, the time evolution of the average characteristics and of their variances is governed by the decaying concentration of the survivors in fluctuational cavities.

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### I. INTRODUCTION

Diffusion or hopping transport of particles in a system with random traps is quite a common problem in the kinetics of condensed media and in chemical kinetics. Some examples are exciton migration in molecular crystals, in amorphous solids and in biological systems, sensitized luminescence and photochemistry, conductivity of extrinsic semiconductors, spin diffusion, kinetics of diffusion-controlled chemical reactions, etc. [1–6].

Such processes are studied in the framework of chemical kinetics (both classical and fluctuational) and of the stochastic transport theory of disordered systems [1–38].

Diffusion with a chemical reaction (equivalent to trapping by mobile sinks) has been considered by Smoluchowski [7]. This classical approach is in fact an approximation of mean-field type and is accurate at short and intermediate time. The corresponding long-time limit is of fluctuation nature. It has been considered first within a nonperturbative approach in Ref. [8]. The obtained decay of the concentration of the surviving particles in the fluctuation regime at long time is a stretched exponential function  $\approx \exp[-\text{const}_{d,2} t^{d/(d+2)}]$ . It has been proven to be exact in Refs. [8–11].

In a reversible chemical reaction [13] (trapping corresponds to one of the components being frozen) the approach to equilibrium at long time is of a slow power-law type  $\approx (Dt)^{-d/2}$ ; see also Refs. [20–22].

A diagram technique has been developed for exciton migration in a solution with traps or sinks [14,15] and two- and three-site self-consistent approximations of mean-field type have been built. Hopping on a disordered chain in one dimension has been studied rigorously [16,17]. A diverse evolution in a system with diffusion, annihilation, and reproduction of particles has been examined [18,19].

The problem of particle migration on lattices with traps of random positions and random depths has been studied in detail on the basis of a constructed self-consistent cluster effective-medium approximation [20–25]. The method proved to be accurate in most limiting cases, except for the long-time trapping by sinks [20]. The evolution of the averaged parameters like the diffusion coefficient, ac conductivity, and the averaged kinetics of relaxation of the spectral population have been studied in detail.

Apart from the average kinetic characteristics, of considerable interest is the study of the fluctuations of these characteristics in disordered systems. These fluctuations come from the sample-to-sample fluctuations and from the fluctuations within the sample due to shifted initial positions. This question is of importance, as the stability and reproducibility of a single measurement relies on the decay of the fluctuations, both in real-world experiments and in computer simulations. To characterize the reproducibility and convergence of the results the term “self-averaging” is used. It specifies the evolution of the relative fluctuations of the measured quantity—whether its dispersion in different realizations vanishes with time or not. Self-averaging signifies that the reciprocal fluctuations of the quantity tend to zero. Then in the long-time limit the measurement in a single experiment produces a representative result.

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The question of the self-averaging of the diffusion coefficient in random-trapping transport has been addressed in the nonstationary formulation within the master-equation approach in Refs. [16,22–25]. The change of the self-averaging properties has been traced with the increase of the disorder (of the trap depth). In the regular regime of reversible trapping, the diffusion coefficient has been found to be self-averaging with a power-law decrease of the sample-to-sample fluctuations. In the stronger disordered subdispersive and dispersive regimes, the self-averaging slows down. For lower-dimensional systems in the first place, self-averaging becomes weak with a slow logarithmic decay of fluctuations. With the subsequent increase of trap depths it occasionally turns into non-self-averaging with nonvanishing sample-to-sample fluctuations. The most disordered case of irreversible trapping by infinitely deep traps has been studied in the regime of intermediate-long-time asymptotics. The reciprocal sample-to-sample fluctuations in this case do not decay in time in all dimensions. This signifies non-self-averaging.

The only case that has not been studied within the approach [20–25] corresponds to the long-time asymptotics for irreversible trapping, as the method becomes inaccurate in this limit. To fill the gap that is left in the nearly complete picture, we address the problem of self-averaging of the diffusion coefficient in the long-time regime with traps or sinks in the present study. To do that, we use a simple nonperturbative approach, termed the “method of an optimal fluctuation (or method of cavities)” [8,11,12]. This method has been proven to provide exact exponential factors for the concentration of particles in the problem of sinks [8–11].

In general, the problem of self-averaging has attracted considerable attention lately [16,22–38]. Initially it has been addressed for quantum disordered systems in the steady-state regime (see [26] and references therein). In the quantum steady-state context the self-averaging of a measurable quantity is understood as the decay of its fluctuations in the thermodynamic limit, as the system volume tends to infinity. However, in nonstationary stochastic systems, considered in the present paper, self-averaging is understood somewhat differently—as vanishing of relative fluctuations with time, as stated above.

In the stochastic formulation, the problem of self-averaging has been addressed in a number of studies [16,22–25,27–38]. Within the master-equation approach to disordered systems, it has been considered in Refs. [16,22]. Processes of spin relaxation for random walks on disordered lattices have been demonstrated to be self-averaging [27]. The spatial distribution function of diffusing particles in the presence of traps has been analyzed [28] with the help of the Lifshits method [26]. Directed percolation in a random medium is non-self-averaging [29]. Occupation-time distribution for the diffusion on a disordered chain demonstrates loss of self-averaging and big sample-to-sample fluctuations [30]. Population kinetics in some cases lacks self-averaging [31]. Stochastic processes with power-law distributions may be non-self-averaging [32]. Certain classes of percolation models are non-self-averaging [33]. Variance of the concentration of diffusing particles, reversibly bound to reaction centers on long macromolecules or on cell surfaces, has been calculated [34]. Certain bounded quantities become non-self-averaging, when the correlation

length reaches the order of the size of the system [35]. First-passage-time distribution in a disordered medium is sample dependent and non-self-averaging [36]. Non-self-averaging and ergodicity breaking in subdiffusion in quenched random media have been investigated [37]. Conditions for the self-averaging of the impurity-limited resistance in quasi-one-dimensional nanowires have been found [38].

The goal of this paper is the theoretical study of the self-averaging of the diffusion coefficient in nonstationary processes in the presence of traps or sinks. The measurements in the ensemble of samples with random traps or sinks will produce close and reproducible results—or not? If the fluctuations of the kinetic coefficients are large, will they decay in time, and if so—which way? To answer these questions we calculate and analyze the variance of the mean-square displacement of the particles—as a function of time, of the concentration of sinks, and of the dimensionality of the system. Another goal of this study is to fill the gap in the classification of self-averaging properties for the long-time fluctuation regime with irreversible trapping, which could not be considered with the help of the previous method [22–25].

## II. MODEL

Let us consider particles that diffuse in a medium with static traps or sinks. These can be quasiparticles in a solid, chemical reactants in a solvent, etc. In this study, we consider the case when the traps or the sinks have infinite “depth”; i.e. they capture the particle irreversibly. We will use the term “trap” for the case, when the particle, captured by the trap, is still counted in the normalization of the probability density. By the term “sink” we denote the case when the particle captured vanishes from the norm. Thus, the difference between traps and sinks is in the normalization of the particle concentration. In the case of traps, we consider all the particles, both the survivors and the captured ones, and their total concentration is always preserved. In the case of sinks, we deal with the concentration of the surviving particles, which decays in time.

The traps or sinks are distributed at random with concentration  $c$ . The diffusion coefficient we denote by  $D$ . Upon contacting a trap or sink, the particle gets trapped instantly.

We consider the nonstationary problem. At short and intermediate time the evolution is governed by the classical chemical kinetics of Smoluchowski [7]. However, here we are specifically interested in the long-time limit of the evolution—the long-time fluctuation regime [8]. In this regime the reactants in the regions, where the components (the particles and the sinks) are well mixed, have already vanished. The particles survive in rare cavities, where accidentally there happen to be no sinks. Within such cavities, the particles diffuse relatively slowly and vanish only while reaching their borders with sinks [8]. These rare occasional cavities build up the average concentration and the mean-square displacement of the survivors at long time. The rest of the space hardly contains surviving particles. In the present study we neglect the very rare intercavity transfers (cf. Ref. [28]).

Based on this picture, we calculate the variance of the mean-square displacement. The reciprocal variance, as always, provides the measure of the fluctuations of the quan-

tity under consideration. It gives the answer to the question we are interested in—is the mean-square displacement of the particles in the long-time fluctuation regime in the presence of sinks a self-averaging quantity or not? In other words: Are the fluctuations of the mean-square displacement comparable to (or even greater than) its average value? In such a case, this quantity is non-self-averaging and it cannot be reliably measured in a single experiment. Then special care and excessive averaging in the ensemble have to be taken for the study of the corresponding quantity.

### III. METHOD AND SOLUTION

The picture, stated above, suggests the method of solution [8–12]. It is often called the method of an optimal fluctuation or the method of cavities.

We consider sink-free cavities, where the particles have chances of survival at long time. The probability of the formation of such a  $d$ -dimensional cavity of volume  $V$ , free of traps or sinks, is  $\exp(-Vc)$ . The lowest mode in volume  $V$ , that corresponds to the slowest long-time decay, is in a spherical cavity—therefore we limit our consideration to  $d$ -dimensional spheres. This approach provides a lower estimate for the exact result. It has been demonstrated also [8–11] that the value of the exponent of the concentration of survivors, obtained within this approach in the long-time limit, is exact. The correct form of the leading exponent is sufficient for the determination of self-averaging or non-self-averaging.

In such a spherical cavity the particles diffuse,

$$\partial\rho(\mathbf{r}, t)/\partial t = D\Delta\rho(\mathbf{r}, t), \quad (1)$$

with absorption on the boundaries  $\Sigma$ ,

$$\rho(\mathbf{r}, t)|_{\Sigma} = 0. \quad (2)$$

The initial conditions correspond to one particle in a cavity, positioned in its center. This is in line with the formulation of Refs. [20–25]; its extension we are considering here. Differently distributed initial conditions within the cavity would modify the numerical coefficient in the preexponent of the corresponding long-time asymptotics, which is not of great importance for the consideration of self-averaging.

The solutions for the long-time asymptotics of the probability density  $\rho(\mathbf{r}, t)$  of the particles in a  $d$ -dimensional cavity of radius  $R$  are

$$\rho(r, t \rightarrow \infty) = \frac{1}{R} \cos\left(\frac{\pi r}{2R}\right) \exp\left(-\frac{\pi^2 Dt}{4R^2}\right), \quad d = 1, \quad (3)$$

$$\rho(r, t \rightarrow \infty) = \frac{1}{\pi R^2 J_1^2(j_{0,1})} J_0\left(\frac{j_{0,1} r}{R}\right) \exp\left(-\frac{j_{0,1}^2 Dt}{R^2}\right), \quad d = 2, \quad (4)$$

$$\rho(r, t \rightarrow \infty) = \frac{1}{2R^2 r} \sin\left(\frac{\pi r}{R}\right) \exp\left(-\frac{\pi^2 Dt}{R^2}\right), \quad d = 3, \quad (5)$$

where  $J_0$  is the Bessel function and  $j_{0,1}$  is the first zero of the Bessel function of order zero.

In the next section, we consider first the case of surviving particles in the problem of sinks. The quantities of interest are the mean-square displacement of the survivors,

$$\langle R^2(t) \rangle = \int_{V_\infty} dV c \exp(-Vc) \int_V dv r^2 \rho(r, t), \quad (6)$$

and its variance,

$$\text{Var}[R^2(t)] = \int_{V_\infty} dV c \exp(-Vc) \left[ \int_V dv r^2 \rho(r, t) \right]^2 - \left[ \int_{V_\infty} dV c \exp(-Vc) \int_V dv r^2 \rho(r, t) \right]^2. \quad (7)$$

The second integration in Eqs. (6) and (7) over the possible sizes of the cavities extends to the entire space  $V_\infty$ .

As always, the mean-square displacement measures the spreading of the particle. Its reciprocal standard deviation (square root of the variance, divided by the average value) measures the relative fluctuations of this spreading. As the number of surviving particles in the present formulation is not conserved, both the mean-square displacement (6) and its variance (7) decay in time.

The use of a non-normalized probability density  $\rho(\mathbf{r}, t)$  in Eqs. (6) and (7) poses no problem. One can consider the total probability density in a cavity  $\rho_\Sigma(\mathbf{r}, t) = \rho(\mathbf{r}, t) + \rho_{tr}(\mathbf{r}, t)$ , normalized to 1, where  $\rho(\mathbf{r}, t)$  refers to survivors, as above, while  $\rho_{tr}(\mathbf{r}, t)$  is for trapped particles. The contribution of survivors to  $\langle R^2(t) \rangle$  is  $\sim r^2$ ; the contribution of trapped particles in this case is 0. The calculation of this partial mean-square displacement leads back to (6) and (7).

Another measure of the spreading and of the fluctuations of the surviving particles can be adopted as well. It addresses the problem from the point of view of the diffusion coefficient as the characteristic of a single particle. Then the mean-square displacement and its variance are calculated “per one survivor” ([28] and references therein). In this case the probability density of the contributing particles is renormalized at any given moment of time to provide the constant norm of survivors, equal to 1. Such a “normalization” of the density function per one survivor can be provided in two ways; cf. [28].

In the first way the norm is calculated over all the possible cavity sizes. It is equivalent to averaging over realizations or to the integration over the entire volume with different starting positions. In our case, it leads to summation over independent cavities with a Poisson distribution. Thus, the density function (3)–(5) is multiplied by the inversed average survival probability of a particle in the system. This normalization coefficient is the same for all the realizations:

$$\rho_{n1}(r, t) = \rho(r, t) \left[ \int_{V_\infty} dV c \exp(-Vc) \int_V dv \rho(r, t) \right]^{-1}. \quad (8)$$

The norm of Eq. (8) over all the realizations is 1 at any time.

In the second way the density is normalized prior to averaging, within one realization. As we neglect intercavity transfers, the cavities are decoupled and the normalization

proceeds independently within each cavity of radius  $R$ :

$$\rho_{n2}(r, R, t) = \rho(r, t) \left[ \int_V dv \rho(r, t) \right]^{-1}. \quad (9)$$

Thus, the norm of  $\rho_{n2}(r, R, t)$  in a cavity  $R$  is always 1 at any time.

$$\langle \tilde{R}^2(t) \rangle = \int_{V_\infty} dV c \exp(-Vc) \left[ R^2 - \int_V dv \rho(r, t) (R^2 - r^2) \right]. \quad (10)$$

The first term in the square brackets is the long-time limit, when all the particles get trapped on the surface of the cavity at radius  $R$ . The second is the transition term at time  $t$ . The total number of particles within the cavity, both survivors and the trapped ones, is preserved at all times, as we neglect intercavity transfer. Consequently, no normalization procedure is applicable in this case.

The variance of the mean-square displacement, calculated for all the particles, is

$$\text{Var}[\tilde{R}^2(t)] = \int_{V_\infty} dV c \exp(-Vc) \left[ R^2 - \int_V dv \rho(r, t) (R^2 - r^2) \right]^2 - \left\{ \int_{V_\infty} dV c \exp(-Vc) \left[ R^2 - \int_V dv \rho(r, t) (R^2 - r^2) \right] \right\}^2. \quad (11)$$

In Eq. (11), the first term is the mean square of the square displacement. The second term is the mean-square displacement squared.

We note that the consideration of all the particles, (10) and (11), is the direct extension of our study in Ref. [25] to the present case of infinite trap depth. The true long-time asymptotics in this particular case could not be calculated there because of the limitations of the effective-medium approximation. The present nonperturbative study fills the last gap in the picture of self-averaging, presented there. With this study, the classification of self-averaging for all the possible cases, as the disorder strength increases from regular to irreversible trapping, on a qualitative level becomes complete.

#### IV. RESULTS AND DISCUSSION

First, we consider the case when only the surviving particles contribute to the concentration and to the mean-square displacement. The trapped particles are disregarded and vanish from the normalization of the probability density.

##### A. Survivors: Results

The calculation of the reciprocal standard deviation of the mean-square displacement, (6) and (7), at long time  $c^{2/d}Dt \gg 1$  with the account of Eqs. (3)–(5) provides the following result:

$$\frac{\text{Var}^{1/2}[R^2]}{\langle R^2 \rangle} = \frac{A_d}{(c^{2/d}Dt)^{d/(4d+8)}} \exp[B_d(c^{2/d}Dt)^{d/(d+2)}], \quad (12)$$

where the numerical coefficients are

$$A_1 = 2^{-1/12} 3^{1/4} \pi^{-5/12}, \quad B_1 = 2^{-4/3} (2^{2/3} - 1) 3\pi^{2/3}, \quad d = 1, \quad (13)$$

$$A_2 = 2^{5/8} \pi^{-3/8} j_{0,1}^{-1/4}, \quad B_2 = (2 - \sqrt{2}) \pi^{1/2} j_{0,1}, \quad d = 2, \quad (14)$$

$$A_3 = 2^{-11/20} 5^{1/4} \pi^{-13/20}, \quad B_3 = 3^{-1} (2^{2/5} - 1) 5\pi^{8/5}, \quad d = 3. \quad (15)$$

The two corresponding normalized results are discussed and compared to the previous formulation in the next section.

Of interest also is the problem of irreversible trapping with the account of all the particles, both the survivors and the trapped ones. The corresponding mean-square displacement we mark by a tilde to distinguish it from (6) and (7):

At long time the factor  $c^{2/d}Dt$  is the big dimensionless parameter of the problem.

The average value of the mean-square displacement (6) at long time  $c^{2/d}Dt \gg 1$ , calculated in a similar way with the help of Eqs. (3)–(5), is

$$\langle R^2 \rangle = A'_d c^{-2/d} (c^{2/d}Dt)^{(d+4)/(2d+4)} \times \exp[-B'_d (c^{2/d}Dt)^{d/(d+2)}]. \quad (16)$$

The numerical coefficients in Eq. (16) are

$$A'_1 = \frac{2^{7/3}(\pi^2 - 8)}{3^{1/2}\pi^{5/6}}, \quad B'_1 = \frac{3\pi^{2/3}}{2^{2/3}}, \quad d = 1, \quad (17)$$

$$A'_2 = \frac{2\pi^{1/4}}{j_{0,1}^{1/2} J_1^2(j_{0,1})} [2J_2(j_{0,1}) - j_{0,1} J_3(j_{0,1})], \quad (18)$$

$$B'_2 = 2\pi^{1/2} j_{0,1}, \quad d = 2,$$

$$A'_3 = \frac{2^{14/5}(\pi^2 - 6)}{5^{1/2}\pi^{3/10}}, \quad B'_3 = \frac{2^{2/5} 5\pi^{8/5}}{3}, \quad d = 3. \quad (19)$$

In one dimension the calculation and the results (12) and (13), and (16) and (17), are exact. In higher dimensions the long-time asymptotics (12), (14), and (15), and (16), (18), and (19), are calculated exactly within the approximate method of cavities. The leading exponents  $B_d, B'_d$  in (12) and (16) for  $d > 1$  are believed to be exact irrespective of the approximation of cavities.

##### B. Survivors: Discussion

Let us analyze the obtained results (12) and (16). Clearly, the expression (12) for the reciprocal standard deviation of the mean-square displacement signifies non-self-averaging. More to the point, this is strong non-self-averaging—exponential. The fluctuations grow with respect to the average value of the mean-square displacement up to infinity—as  $\exp[B_1(c^2Dt)^{1/3}]$  in one dimension, as  $\exp[B_2(cDt)^{1/2}]$  in 2D, and as  $\exp[B_3(c^{2/3}Dt)^{3/5}]$  in 3D.



In lower dimensions the relative fluctuations (12)–(15) are smaller and grow slower than in higher dimensions. The absolute values of the fluctuations (12) with the account of Eq. (16),

$$\text{Var}^{1/2}[R^2] = A_d A'_d Dt (c^{2/d} Dt)^{-3d/(4d+8)} \times \exp[-(B'_d - B_d)(c^{2/d} Dt)^{d/(d+2)}], \quad (20)$$

in lower dimensions are bigger and decay slower.

As noted above, the asymptotics for the survival probability  $P_s(t)$ , obtained with the method of cavities,

$$\langle P_s(t) \rangle = A''_d (c^{2/d} Dt)^{d/(2d+4)} \exp[-B''_d (c^{2/d} Dt)^{d/(d+2)}], \quad (21)$$

reproduce the exact stretched exponentials [8–11]. This fact provides good grounds for the belief that the exponents (12), (16), and (20) should be exact as well.

The unbounded growth of the reciprocal fluctuations (12), for sure, does not signify any divergence of any actual characteristic. In fact, both the standard deviation (20) and the average value (16) of the mean-square displacement of the survivors are extremely small and decay further in the fast exponential way. It is only their ratio that grows exponentially.

Both the average value (16) and the standard deviation (20) of the mean-square displacement are governed by the exponential terms. These originate from the concentration of the surviving particles. The  $R^2$  factor contributes to the preexponential term only.

A simple qualitative interpretation of these results can be provided. The correlators  $\langle (R^2)^2 \rangle$  and  $\langle R^2 \rangle^2$  in the variance are integrals with the local density function. In the difference  $\langle (R^2)^2 \rangle - \langle R^2 \rangle^2$ , the small exponential factor of the concentration enters the first term linearly, while in the second term it is squared. Therefore  $\langle R^2 \rangle^2$  is negligible in the long-time leading term of the variance. Thus, in the reciprocal standard deviation the numerator  $\langle (R^2)^2 \rangle^{1/2}$  effectively comprises the square root of the “stretched exponential” factor of the concentration. At the same time, the denominator  $\langle R^2 \rangle$  is linear in it. The result is the positive growing exponent (12) with the coefficient  $B_d$  that comprises the difference of the corresponding factors.

### C. All particles: Results

Next, we consider the case when all the particles, both the survivors and the trapped ones, contribute to the mean-square displacement and to its reciprocal variance.

The calculation of the reciprocal standard deviation of the mean-square displacement (10), (11) at long time  $c^{2/d} Dt \gg 1$  with the account of all the particles produces the following result:

$$\text{Var}^{1/2}[\tilde{R}^2]/\langle \tilde{R}^2 \rangle = \tilde{C}_d \{1 - \tilde{A}_d (c^{2/d} Dt)^{(d+8)/(2d+4)} \times \exp[-\tilde{B}_d (c^{2/d} Dt)^{d/(d+2)}]\}. \quad (22)$$

In Eq. (22) we made use of (3)–(5). The numerical coefficients in (22) we mark with a tilde as well to distinguish them

from (12)–(21):

$$\tilde{A}_1 = 2^6 3^{-1/2} 5^{-1} \pi^{1/2}, \quad \tilde{B}_1 = 2^{-2/3} 3 \pi^{2/3}, \quad \tilde{C}_1 = \sqrt{5}, \quad d = 1, \quad (23)$$

$$\tilde{A}_2 = 4\pi^{7/4} j_{0,1}^{1/2} J_1^{-2}(j_{0,1}) J_2(j_{0,1}), \quad \tilde{B}_2 = 2\pi^{1/2} j_{0,1}, \quad \tilde{C}_2 = 1, \quad d = 2, \quad (24)$$

$$\tilde{A}_3 = \frac{2^{61/15} 3^{5/3} \pi^{43/30}}{5^{1/2} [\Gamma(1/3) - \Gamma^2(2/3)]}, \quad \tilde{B}_3 = \frac{2^{2/5} 5 \pi^{8/5}}{3}, \quad \tilde{C}_3 = \left[ \frac{\Gamma(1/3)}{\Gamma^2(2/3)} - 1 \right]^{1/2}, \quad d = 3. \quad (25)$$

The expression of the mean-square displacement (10) at long time  $c^{2/d} Dt \gg 1$  is calculated in a similar way with the help of Eqs. (3)–(5):

$$\langle \tilde{R}^2 \rangle = \tilde{C}'_d c^{-2/d} \{1 - \tilde{A}'_d (c^{2/d} Dt)^{(d+4)/(2d+4)} \times \exp[-\tilde{B}'_d (c^{2/d} Dt)^{d/(d+2)}]\}. \quad (26)$$

The numerical coefficients in Eq. (26) are

$$\tilde{A}'_1 = 2^{19/3} 3^{-1/2} \pi^{-5/6}, \quad \tilde{B}'_1 = 3\pi^{2/3} 2^{-2/3}, \quad \tilde{C}'_1 = 1/2, \quad d = 1, \quad (27)$$

$$\tilde{A}'_2 = 2^2 \pi^{5/4} j_{0,1}^{-1/2} J_1^{-2}(j_{0,1}) J_2(j_{0,1}), \quad \tilde{B}'_2 = 2\pi^{1/2} j_{0,1}, \quad \tilde{C}'_2 = 1/\pi, \quad d = 2. \quad (28)$$

$$\tilde{A}'_3 = \frac{2^{62/15} 3^{4/3} \pi^{11/30}}{5^{1/2} \Gamma(2/3)}, \quad \tilde{B}'_3 = \frac{2^{2/5} 5 \pi^{8/5}}{3}, \quad \tilde{C}'_3 = \frac{\Gamma(2/3)}{6^{1/3} \pi^{2/3}}, \quad d = 3. \quad (29)$$

The one-dimensional results (22) and (23), and (26) and (27), are exact. The long-time asymptotics in higher dimensions (22), (24), and (25), and (26), (28), and (29) are calculated exactly within the approximate method of cavities. The exponents  $\tilde{B}_d, \tilde{B}'_d$ ,  $d > 1$  are supposedly exact.

### D. All particles: Discussion

Let us analyze the reciprocal standard deviation (22). Its long-time limit is a numerical constant  $\tilde{C}_d$ . This fact signifies non-self-averaging. However, in contrast to the case of survivors (12), this non-self-averaging is not a strong one. The previously considered survivors are the most fluctuating part of the concentration—their reciprocal variance grows exponentially. In the present case of all particles accounted for, the major part of the concentration is the regular one—the particles trapped. However, this regular component of the concentration got trapped on the surface of cavities of varying sizes. The result is the constant value of the reciprocal standard deviation in the long-time limit due to geometrical reasons. The magnitude of the fluctuations of the mean-square displacement is of the same order as its average value.

We note that the long-time limiting values  $\tilde{C}_d$  of the reciprocal standard deviation decay with the increase of the dimension: 2.23... in one dimension, 1 in two dimensions,

and  $0.67 \dots$  in three dimensions. The fluctuations of the mean-square displacement with respect to its average value get smaller in higher dimensions, as expected; the absolute fluctuations do as well. The difference with the previously considered case of survivors comes from the different role and different contributions of the fluctuating and of the regular components of the concentration.

It should be noted that the limiting values  $\tilde{C}_d$  of  $\text{Var}^{1/2}[\tilde{R}^2]/\langle \tilde{R}^2 \rangle$  for dimensions higher than 1 are approximately dependent. A better account of the possible geometry of cavities will produce a somewhat different constant for  $\tilde{C}_d$ . As before, the one-dimensional case is solved exactly.

The second term in curly braces (22) is the transition term. The reciprocal fluctuations of the mean-square displacement of all the particles at long time reach the constant limiting value  $\tilde{C}_d$  according to a stretched exponential law. The stretched exponential transition is typical for fluctuation-produced asymptotics.

As noted above, the fact that the method of cavities produces the exact exponent for the concentration of surviving particles suggests that the stretched exponents in (22) and (26) should be exact as well.

The present results, (22) and (26), complete the picture of self-averaging of the diffusion coefficient, addressed in [16,22–25] in the formulation of all the particles taken into account. Different cases that correspond to the qualitative increase of trap depths have been studied. Those started with regular reversible trapping by traps of limited depth, through the stronger disordered subregular, subdispersive, and dispersive cases, and up to irreversible trapping. Self-averaging started correspondingly from regular power-law decay of fluctuations, through the weak self-averaging with a logarithmic decay, and up to non-self-averaging, when the fluctuations are of the same order as the average value. The peculiarity of the irreversible case, studied there, is that the variances were calculated as the intermediate-long-time asymptotics. The true long-time asymptotics could not be found there due to the limitations of the effective-medium approximation. For the true long-time asymptotics a suitable nonperturbative treatment was required. The present study fills that gap.

The intermediate-long-time asymptotics [25] and the true long-time asymptotics (22) do not overlap, but complement each other. In doing so they agree qualitatively—in both cases the reciprocal standard deviation of the mean-square displacement tends to a constant. Both asymptotics reveal fluctuations of the mean-square displacement of the same order, as its average value. We note also that these two values do not have to coincide—(a) because of the different time intervals and (b) because of the approximate nature of both methods. These solutions, in fact, are built largely on different classes of trajectories, as discussed below.

### E. Normalized survivors: Results and discussion

Next, we address the commonly considered normalized cases (8) and (9) (see, for example, [28] and references therein).

The first case is the normalization over all realizations—over the cavity sizes (8). The mean-square displacement, normalized “per one survivor in the system on average” is calculated with the use of (6) and (8). It is a power-law

function of time [11]:

$$\langle R^2 \rangle_{n1} = C_d'' (Dt/c)^{2/(d+2)}. \quad (30)$$

Due to the normalization (8) it contains no exponential factor that otherwise would come from the survival probability; cf. (16).

In the reciprocal standard deviation, the normalization factor in the square brackets of (8) enters linearly both the numerator and the denominator. Due to its cancellation, the resulting expression of the normalized reciprocal standard deviation per one survivor in the system on average coincides exactly with the previous formulas (12)–(15). Therefore, the discussion of the reciprocal standard deviation for survivors remains valid in this case as well.

In short, the normalized mean-square displacement per one survivor in the system on average (8) is a strongly non-self-averaging quantity (12).

It should be noted, however, that this normalization is not quite satisfactory physically, as the normalization coefficient (the inversed survival probability) in (8) is not a self-averaging quantity. Therefore, strictly speaking, the density function (8) should be normalized for each realization separately with the corresponding realization-specific normalization coefficient and not with its average value [28].

Next we go over to the normalization of the second type.

It is the normalization within a single sample—in each cavity (9). The reciprocal standard deviation of the mean-square displacement, (6) and (7), calculated with the normalized density function (9) “per one survivor in a single sample prior to averaging” in the long-time limit tends to a constant:

$$\text{Var}^{1/2}[R^2]_{n2}/\langle R^2 \rangle_{n2} = C_{n2,d}. \quad (31)$$

The long-time limit of the normalized standard deviation (31) coincides exactly with the long-time limit for the case of all the particles taken into account, (22)–(25),  $C_{n2,d} = \tilde{C}_d$ . The identical coefficients  $\tilde{C}_d$  and  $C_{n2,d}$  are determined by the geometry and the statistics of the cavities and not by the kinetics.

The normalized mean-square displacement, (6) and (9), at long time tends to a constant as well:

$$\langle \tilde{R}^2 \rangle = C'_{n2,d} c^{-2/d}. \quad (32)$$

Equations (31) and (32) would signify non-self-averaging of the survivors, normalized per one survivor in a single sample prior to averaging. However, no conclusions from (31) and (32) can be derived, as this kind of normalization becomes invalid in the present formulation (cf. Ref. [28]).

The reason for it is the following. In our approach the intercavity transfers are neglected; the cavities are decoupled. Therefore, the normalization procedure (9) takes place not in the entire space of one sample, but within one cavity of some radius. Thus, cavities of different sizes get different normalization coefficients and this deteriorates their relative contributions to the final result. Smaller cavities at long time are overestimated as compared to bigger ones. Therefore, the reciprocal fluctuations at long times are underestimated considerably; cf. (12) and (31). The fluctuation mechanism was built on the interplay of the bigger survival probability in bigger cavities versus their smaller statistical weight. The

normalization procedure (9) in the case of decoupled cavities eliminates the first effect.

If other independent cavities are added to the normalization within one sample, then the procedure effectively becomes that of the previous type (8).

We note that there is no problem of such kind in the general case [28] with intercavity transfer. With cavities connected, the integration in (9) goes over the entire space of one sample. Consequently, all the participating cavities get the same normalization coefficient  $\int_{V_\infty} \rho(\mathbf{r}, t) d\mathbf{r}$  within one configuration of random traps or sinks. Therefore, the relative contributions of cavities of different radii are accounted for correctly. In the general case, the density function  $\rho_{n2}$  is normalized to 1 in the entire space of connected cavities within one realization. The cavity sizes in an infinite system cover the entire spectrum and the normalization is provided per one survivor in one sample prior to averaging over the random positions of the traps.

**F. Higher dimensions: Average characteristics**

Let us consider the higher-dimensional cases, average characteristics first.

The long-time fluctuation asymptotics of the averaged quantities for survivors are of the form  $\approx \exp[-\text{const}_{d,3} t^{d/(d+2)}]$ . The corresponding mean-field-type results are  $\approx \exp[-\text{const}_{d,4} t]$ . The comparison of the two leads to the conclusion that the critical dimension, above which the mean-field result becomes exact, in this case is infinite.

Obviously, this is due to the difference of the classes of trajectories, upon which the corresponding results are being built. For the self-consistent effective-medium approximation [20–25], these are non-all-self-intersecting trajectories—those that contain at least one site, visited only once. The fluctuation asymptotics, on the contrary, are formed by the trajectories, which wind densely at long time within the cavities, and, therefore, late enough, become typically all-self-intersecting. Thus, there is no finite dimension, where the class of trajectories that support the mean-field result would essentially coincide with the class of trajectories for the fluctuation asymptotics. These classes are to a large extent complementary. However, the increase of the dimension helps the trajectories to avoid self-intersections at every site visited and this somehow improves the mean-field picture of the fluctuation regime.

In other words, the uniform medium is not a good zero-order approximation for the long-time fluctuation regime in the presence of sinks in any finite dimension—when the system effectively splits into rare isolated cavities—up to  $d = \infty$ . Therefore the critical dimension of the system is infinite.

We note also that the order in which the limits (long-time versus high dimensions) are calculated is of importance. Obviously, it is physically relevant to choose the dimension  $d$  and fix it, and to consider the long-time limit  $c^{2/d}Dt \gg 1$  after that. The high- $d$  case can be studied as well with the assumption that the parameter  $c^{2/d}Dt$  is the biggest.

**G. Higher dimensions: Self-averaging, survivors**

Next, let us consider the self-averaging in higher dimensions.

The spherically symmetric solution of (1) and (2) in  $d$  dimensions at long time  $c^{2/d}Dt \gg 1$  is

$$\rho(r, t \rightarrow \infty) = K_d R^{-\frac{d+2}{2}} r^{-\frac{d-2}{2}} J_{\frac{d-2}{2}} \left( \frac{j_{(d-2)/2,1} r}{R} \right) \times \exp \left( -\frac{j_{(d-2)/2,1}^2 Dt}{R^2} \right), \tag{33}$$

where  $j_{(d-2)/2,1}$  is the first zero of the Bessel function  $J_{(d-2)/2}$  and the coefficient  $K_d$  is solely a function of the dimension  $d$ .

The mean-square displacement (6) of survivors in  $d$  dimensions at long time is calculated as

$$\langle R^2 \rangle = \text{const}_{d,5} c^{-\frac{2}{d}} (c^{\frac{2}{d}} Dt)^{\frac{d+4}{2(d+2)}} \times \exp \left[ -\frac{d+2}{d} \left( \frac{1}{2} \Omega_d j_{\frac{d-2}{2},1}^d \right)^{\frac{2}{d+2}} (c^{\frac{2}{d}} Dt)^{\frac{d}{d+2}} \right], \tag{34}$$

where  $\Omega_d = 2\pi^{d/2} \Gamma^{-1}(d/2)$  is the  $d$ -dimensional spherical angle and the numerical constant  $\text{const}_{d,5}$  is function of dimension only.

The expression of the mean-square displacement (34) is valid for any  $d$ . The exponent and the prefactor are calculated exactly up to an unspecified numeric coefficient  $\text{const}_{d,5}$  within the approximate method of cavities. As noted before, the exponent in (34) is supposed to be exact without recourse to the cavities approximation. The entire expression (34), certainly, agrees with the previous formula (16) up to the numeric coefficient  $A'_d$ .

The mean-square displacement (34) decays in time as a stretched exponential in any dimension. This originates from the decay of the survival probability.

In the limit of high dimensions  $d \gg 1$  the modulus of the exponent in (34) grows linearly with  $d$ :

$$\langle R^2 \rangle = \text{const}_{d,5} c^{-\frac{2}{d}} (c^{\frac{2}{d}} Dt)^{1/2} \exp \left[ -\frac{\pi ed}{2} c^{\frac{2}{d}} Dt \right]. \tag{35}$$

In higher dimensions, the decay (35) in the parameter  $c^{2/d}Dt$  is faster. This is due to the lower probability of the formation of big cavities with the growing dimension.

The reciprocal standard deviation is calculated with the help of (6), (7), and (33):

$$\frac{\text{Var}^{1/2}[R^2]}{\langle R^2 \rangle} = \text{const}_{d,6} (c^{\frac{2}{d}} Dt)^{-\frac{d}{4(d+2)}} \times \exp \left[ \frac{(1 - 2^{-\frac{2}{d+2}})(d+2)}{d} \left( \frac{1}{2} \Omega_d j_{\frac{d-2}{2},1}^d \right)^{\frac{2}{d+2}} (c^{\frac{2}{d}} Dt)^{\frac{d}{d+2}} \right]. \tag{36}$$

The formula (36) for the reciprocal standard deviation is valid for any  $d$  in the long-time domain  $c^{2/d}Dt \gg 1$ . The entire expression (36), except for the unspecified numerical constant  $\text{const}_{d,6}$ , is exact within the approximate method of cavities. As before, the exponent in (36) is supposedly exact without recourse to the approximation of cavities. The entire expression (36), certainly, agrees with the previous formula (12) up to the numeric coefficient  $A_d$ .

The reciprocal standard deviation (36) signifies strong non-self-averaging in any dimension. The relative fluctuations grow as a stretched exponential. As before, both the standard deviation and the mean-square displacement decay to zero with different exponents, and it is only their ratio (36) that grows. The evolution of these quantities is governed by the decay of the survival probability density in the cavities of varying sizes.

In the high-dimension limit  $d \rightarrow \infty$ , the reciprocal standard deviation (36) takes a simpler form:

$$\text{Var}^{1/2}[R^2]/\langle R^2 \rangle \approx \text{const}_{d,6} (c^{2/d} Dt)^{-1/4} 2^{\pi e c^{2/d} Dt}. \quad (37)$$

For survivors in infinite dimensions the self-averaging is not restored. The reciprocal fluctuations (37) grow exponentially with time, and this signifies strong non-self-averaging.

As  $d \rightarrow \infty$  the stretched exponential in (36) grows into a somewhat faster usual exponent (37) in  $c^{2/d} Dt$ , as it usually is in the problem of sinks. As for the  $d$  dependence, the limiting expression (37) reveals no significant dependence on dimension  $d$ . In infinite dimensions, the strong non-self-averaging of survivors is preserved.

### H. Higher dimensions: Self-averaging, all particles

The mean-square displacement (10) of all the particles, both the survivors and the trapped ones, in  $d$  dimensions at long time  $c^{2/d} Dt \gg 1$  is calculated with the help of (33)

$$\langle R^2 \rangle = \frac{2^{(d-2)/d} \Gamma(2/d) \Gamma^{2/d}(d/2)}{\pi d^{(d-2)/d} c^{2/d}} \left\{ 1 - \text{const}_{d,7} (c^{2/d} Dt)^{\frac{d+4}{2(d+2)}} \right. \\ \left. \times \exp \left[ -\frac{d+2}{d} \left( \frac{1}{2} \Omega_d J_{\frac{d-2}{2},1}^d \right)^{\frac{2}{d+2}} (c^{2/d} Dt)^{\frac{d}{d+2}} \right] \right\}. \quad (38)$$

In the long-time limit, the mean-square displacement (38) tends to a constant, as all the particles stick to the boundary of the cavities. The transition term is a stretched exponential.

Both terms in (38) are calculated exactly within the approximate method of cavities, the second term—up to the unspecified numeric prefactor  $\text{const}_{d,7}$ . The exponent of the transition term is supposed to be exact irrespective of the approximation of cavities. The entire expression (38) agrees with the previous results (26)–(29) up to the numeric coefficient  $\tilde{A}'_d$ .

In the high dimension limit  $d \gg 1$ , the formula (38) takes a simpler form:

$$\langle R^2 \rangle \approx \frac{d}{2\pi e c^{2/d}} \left\{ 1 - \text{const}_{d,7} (c^{2/d} Dt)^{\frac{1}{2}} \exp \left[ -\frac{\pi e d}{2} c^{2/d} Dt \right] \right\}. \quad (39)$$

The limiting value of the mean-square displacement in high dimensions is linear in  $d$ . This proportionality is of the same kind as in the usual case  $\langle R^2 \rangle = 2dDt$ . The dimensional factor in (39), as before, scales with trap concentration as  $c^{-2/d}$ . The exponent of the transition term for high dimensions becomes linear in  $d$  as well. The decay of the transition term in  $t$  is faster for higher dimensions. This is explained by the lower probability of the formation of big cavities.

The reciprocal standard deviation is calculated with the help of (10), (11), and (33):

$$\frac{\text{Var}^{1/2}[R^2]}{\langle R^2 \rangle} = \left[ d \Gamma \left( \frac{4}{d} \right) \Gamma^{-2} \left( \frac{2}{d} \right) - 1 \right]^{1/2} - \text{const}_{d,8} (c^{2/d} Dt)^{\frac{d+8}{2(d+2)}} \\ \times \exp \left[ -\frac{(d+2)}{d} \left( \frac{1}{2} \Omega_d J_{\frac{d-2}{2},1}^d \right)^{\frac{2}{d+2}} (c^{2/d} Dt)^{\frac{d}{d+2}} \right]. \quad (40)$$

Equation (40) was calculated exactly within the approximate method of cavities. The second transition term has been found up to an unspecified coefficient  $\text{const}_{d,8}$ , which is solely a function of dimension  $d$ . The stretched exponent of the transition term should be exact irrespective of the approximation of cavities. The entire expression (40) agrees with the previously found particular cases (22)–(25) up to the factor  $\tilde{A}_d$ .

At long time in any finite dimension  $d$ , the reciprocal standard deviation (40) tends to a constant. This signifies non-self-averaging, not a strong one. In the general case, the particular value of this constant in the long-time limit depends upon the adopted approximation for the shape and size distribution of the cavities.

An interesting question is whether the conclusion on non-self-averaging changes in the limit of infinite dimensions or not. For high values of  $d$  the expression (40) takes a simple form:

$$\frac{\text{Var}^{1/2}[R^2]}{\langle R^2 \rangle} \approx \frac{\sqrt{2}\pi}{\sqrt{3}d} - \text{const}_{d,8} (c^{2/d} Dt)^{\frac{1}{2}} \exp \left[ -\frac{\pi e d}{2} c^{2/d} Dt \right]. \quad (41)$$

The limiting value of the reciprocal standard deviation decreases to zero as  $d^{-1}$ . It means that, although in any finite dimension the mean-square displacement of all the particles is a non-self-averaging quantity, in the limit of infinite dimensions self-averaging is restored in some sense.

With the increasing dimension, the reciprocal standard deviation becomes smaller. On a qualitative level, in the limit of infinite dimensions the fluctuating system becomes more “regular.” The zero limit of the reciprocal standard deviation signifies some kind of restored self-averaging.

The decrease of the high- $d$  limit (40) is in line with the corresponding decrease for the cases  $d = 1, 2, 3$ , considered in (22)–(25) and discussed there.

We note also that in the case of survivors (37), the self-averaging was not restored in the limit of infinite dimensions. There the exponentially growing reciprocal standard deviation was formed by the most fluctuating part of the particles—the survivors. The “regularizing” effect of infinite dimensions was not enough to overcome the exponentially increasing reciprocal fluctuations.

In the case of all the particles accounted for, the major part of those, the trapped ones, forms the regular component. These particles produce nonstrong non-self-averaging. This “weaker” non-self-averaging can indeed be overcome by the “regularizing influence” of the growing dimension,  $d \rightarrow \infty$  (41).



It should be noted also that the onset time for the fluctuation regime with the increasing dimension  $d$  slowly decreases as  $t_f \approx D^{-1}c^{-2/d}$ . Therefore, the time range for the cited fluctuation asymptotics exists in all the cases considered above.

## V. CONCLUSIONS

The main results of the paper are the following.

The mean-square displacement of surviving particles is strongly non-self-averaging. Its reciprocal fluctuations grow as a stretched exponential (12)–(15).

The mean-square displacement of all the particles, both the survivors and the trapped ones, is also non-self-averaging. This non-self-averaging, however, is not a strong one—the reciprocal fluctuations tend to a constant (22)–(25). The transition proceeds via a stretched exponential.

The formulation of self-averaging normalized “per one survivor in the system on average” is equivalent to the result for survivors and corresponds to strong non-self-averaging, (12)–(15).

The normalization “per one survivor in a sample prior to averaging” in the case of decoupled cavities is irrelevant [Eqs. (31) and (32) and the subsequent discussion].

The critical dimension in the problem of sinks is infinite due to the drastic difference between the classes of trajectories

upon which the effective-medium and the fluctuational results are being built.

In higher dimensions, the strong non-self-averaging of survivors is preserved, (36) and (37).

For the case of all the particles taken into account, the nonstrong non-self-averaging diminishes with the increasing dimension (40) up to restoration of the self-averaging in some sense for  $d = \infty$  (41).

In all the cases, the non-self-averaging signifies the poor measurability and reproducibility of results in single measurements due to large fluctuations, both in real-world experiments and in computer simulations.

These results complete the entire picture of the self-averaging in the random-trapping problem, a function of the increasing disorder (trap depth) that ranges from the regular regime to irreversible trapping, started in [16,22–25].

The consideration of the self-averaging of the survival probability of particles and of some other related quantities we will provide in a future publication.

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