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Cluster survival and polydispersity in aggregation

E. K. O. Hellén, P. E. Salmi and M. J. Alava

Laboratory of Physics, Helsinki University of Technology
P.O. Box 1100, FIN-02150 HUT, Finland

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Abstract. – We study the probability, $P_{\mathrm{S}}(t)$, of a cluster to remain intact in one-dimensional cluster-cluster aggregation when the cluster diffusion coefficient scales with size as $D(s) \sim s^{\gamma}$. $P_{\mathrm{S}}(t)$ exhibits a stretched exponential decay for $\gamma < 0$ and the power laws $t^{-3/2}$ for $\gamma = 0$, and $t^{-2/(2-\gamma)}$ for $0 < \gamma < 2$. A random walk picture explains the discontinuous and non-monotonic behavior of the exponent. The decay of $P_{\mathrm{S}}(t)$ determines the polydispersity exponent, τ, which describes the size distribution for small clusters. Surprisingly, $\tau(\gamma)$ is a constant $\tau = 0$ for $0 < \gamma < 2$.

Many models of aggregation phenomena lead to scale-invariance: the average cluster size increases as a power law, $S(t) \sim t^z$, which defines a dynamical exponent z. This kind of behavior is met in various contexts ranging from chemical engineering to materials science to atmosphere research to, ultimately, even astrophysics [1]. It is of interest to explore the statistics of aggregation as a dynamical process, beyond the length- and timescales defined through z.

In this letter we introduce a new quantity in aggregation systems, the cluster survival, defined as the probability $P_{\mathrm{S}}(t)$ that a cluster present at $t = 0$ remains unaggregated until time t. This is a first-passage problem [2] in a many-body system and analogous to persistence which is often studied by measuring the fraction of a system that preserves its initial condition for all times $[0, t]$ [3]. The cluster survival turns out to decay in a nontrivial and counterintuitive manner. The behavior can be understood by a mean-field–like random walk analysis. Even on the mean-field level the question reduces to a novel, unsolved random walk problem, which we analyse in the long-time limit. More importantly, by solving the decay of the cluster survival we are able to determine the polydispersity exponent characterizing the cluster size distribution.

We concentrate on a common and important example: diffusion-limited cluster-cluster aggregation (DLCA) [4]. In the lattice version of DLCA any set of nearest-neighbor occupied lattice sites is identified as a cluster. Each of these performs a random walk with a size-dependent diffusion constant, $D(s) \sim s^{\gamma}$, where γ is the diffusion exponent. Colliding clusters are irreversibly merged together and the aggregate diffuses either faster ($\gamma > 0$) or slower ($\gamma < 0$) than a monomer. In the following, cluster survival is investigated in one dimension for numerical and analytical simplicity. One can discern three separate cases: i) $0 < \gamma < 2$, which results in a power law decay for the survival, $P_{\mathrm{S}}(t) \sim t^{-\theta_{\mathrm{S}}}$, ii) $\gamma = 0$, which is exactly solvable [5], and iii) $\gamma < 0$, when $P_{\mathrm{S}}(t) \sim \exp[-Ct^{\beta_{\mathrm{S}}}]$. Here $\theta_{\mathrm{S}}(\gamma)$ is the survival exponent, $C > 0$ a constant, and $\beta_{\mathrm{S}}(\gamma)$ the stretching exponent. For $\gamma > 2$ the system has a gelation transition and is not of interest here.

The numerics is made transparent by mapping the behavior of surviving clusters to a three-particle random walk (RW) picture: two particles with a time-dependent diffusion coefficient confine a surviving one which diffuses at a constant rate. This is an analogy of the famous independent interval approximation often used in persistence studies [6]. Although the kinetics in one-dimension is fluctuation-dominated [7], the mean-field RW approach turns out to capture the essential ingredients for $\gamma \geq 0$. This is demonstrated by comparing the random walk survival to that of the full DLCA one. The first main result is that $\theta_{\rm S}(\gamma) = 2/(2-\gamma)$ when $0 < \gamma < 2$. Thus the survival exponent is *discontinuous* and *non-monotonic* since $\theta_{\rm S}(0) = 3/2$ but $\theta_{\rm S}(0^+) = 1$. This non-intuitive result follows since for $\gamma > 0$ the problem becomes asymptotically separable: the ratio of the diffusivity of a surviving cluster to that of a typical one goes to zero as $t \to \infty$. For $\gamma < 0$ the spatial fluctuations remain relevant in the sense that the mean-field RW survival and the DLCA one decay with different stretching exponents. For the DLCA simulations suggest an expression $\beta_{\rm S}(\gamma) = -2\gamma/[3(2-\gamma)]$.

One of the main interests in aggregation is the cluster size distribution, $n_s(t)$ (the number of cluster of size s per lattice site at time t). For DLCA simulations and experiments have validated the scaling [4] $n_s(t) = S(t)^{-2} f\left(s/S(t)\right)$, where the scaling limit, $s \to \infty$, $S(t) \to \infty$ with $x \equiv s/S(t)$ fixed, is taken. In one dimension $z = 1/(2-\gamma)$ [8].

There is a fundamental difference between $\gamma < 0$ and $\gamma \geq 0$. For $\gamma < 0$ the cluster size distribution is bell-shaped and $f(x)$ decays faster than any power at both tails. For $\gamma \geq 0$ it is broad so that $f(x) \sim x^{-\tau}$ ($x \to 0$) defines the polydispersity exponent, τ, which characterizes the density of small clusters. In this region we show evidence for the scaling relation $\theta_{\rm S} = (2-\tau)z$. Therefore τ is determined by the cluster survival strategy, shedding light on the non-trivial problem on how to compute it [9,10]. The final main result follows thus $\tau(\gamma) = 0$ for $0 < \gamma < 2$, indicating a flat cluster size distribution. This is confirmed by simulations.

Next we present a mean-field random walk analysis to calculate the survival exponent $\theta_{\rm S}(\gamma)$. In the limit $t \gg 0$ the average distance between clusters grows as t^z and between the unaggregated ones as $t^{\theta_{\rm S}}$. The latter become separated by aggregated clusters at late times, since evidently $\theta_{\rm S} > z$. Thus it is sufficient to consider only one trial cluster and its two neighbors. These two grow still by collisions with neighbors on the opposite side from the trial cluster. In the mean-field approximation the discrete growth events can be substituted by a continuous process and the neighboring clusters grow as the average cluster does. The finite extent of clusters is irrelevant and they can be considered as point particles. Let $x_i(t)$ ($i = 1, 2, 3$) denote their positions at time t with $x_1(0) < x_2(0) < x_3(0)$.

The motion of these particles is described by the Langevin equations

$$\dot{x}_i(t) = \xi_i(t) \tag{1}$$

with Gaussian white noises $\langle \xi_i(t) \rangle = 0$ and $\langle \xi_i(t)\xi_j(t') \rangle = 2\mathcal{D}_i(t)\delta_{ij}\delta(t-t')$, in the standard notation. The diffusion coefficients read as $\mathcal{D}_1(t) = \mathcal{D}_3(t) = D_1 t^{\gamma z}$ and $\mathcal{D}_2(t) = D_2$. The time-dependent diffusion constant, say $\mathcal{D}_1(t)$, implies that the particle 1 will follow a simple diffusive motion with a constant diffusion coefficient D_1 in a time scale $T(t) = \int_0^t {\rm d}t' \mathcal{D}_1(t')/D_1 = t^{\gamma z+1}/(\gamma z + 1)$.

The survival of the center particle is determined by the termination of the process, given by either $x_1(t) = x_2(t)$ or $x_2(t) = x_3(t)$. It is natural to consider the distances between the particles: $x_{12}(t) = x_2(t) - x_1(t) \geq 0$ and $x_{23}(t) = x_3(t) - x_2(t) \geq 0$. Starting from eq. (1) the following Fokker-Planck equation is reached:

$$\frac{\partial \rho}{\partial t} = (D_2 + D_1 t^{\gamma z}) \left(\frac{\partial^2 \rho}{\partial x_{12}^2} + \frac{\partial^2 \rho}{\partial x_{23}^2} \right) - 2D_2 \frac{\partial^2 \rho}{\partial x_{12} \partial x_{23}}, \tag{2}$$

where $\rho(x_{12}, x_{23}; t)$ is the probability density for the two distances at time t. The initial condition is $\rho(x_{12}, x_{23}; 0) = \delta(x_{12}-x_{12}(0))\delta(x_{23}-x_{23}(0))$. The termination of the process when two particles collide gives absorbing boundary conditions along the axis, *i.e.*, $\rho(x_{12}, 0; t) = 0$ and $\rho(0, x_{23}; t) = 0$ for all times t. The survival probability

$$P_{\mathrm{RW}}(t) = \int_0^\infty \mathrm{d}x_{12} \int_0^\infty \mathrm{d}x_{23}\ \rho(x_{12}, x_{23}; t) \sim t^{-\theta_{\mathrm{RW}}(\gamma)}, \tag{3}$$

where the exponent θ_{RW} is the survival exponent for the RW problem.

Equation (2) cannot be solved for t arbitrary since the absorbing boundary conditions together with the two different time scales make the standard methods inappropriate. However, the survival exponent is given by the leading-order asymptotic behavior when $t \to \infty$. We consider the large time limit and the three different cases separately: $\gamma = 0$, $\gamma > 0$, and $\gamma < 0$.

In the size-independent case, $\gamma = 0$, the collisions of the clusters surrounding a surviving one with other clusters do not matter. This is an old problem of three similar annihilating random walkers, for which the survival exponent $\theta_{\mathrm{RW}}(0) = 3/2$ [11]. This agrees with the exact solution of the DLCA problem, $\theta_{\mathrm{S}}(0) = 3/2$ [5]. When the initial distances are $x_{12}(0) = x_{23}(0) = l_0$, the solutions including the first correction to scaling read [5,12] $P_{\mathrm{S}}(t) = P_{\mathrm{RW}}(t) = \frac{1}{4\sqrt{2\pi}}\left(\frac{l_0^2}{Dt}\right)^{3/2} \times \left[1 - \frac{3}{16}\left(\frac{l_0^2}{Dt}\right) + \mathcal{O}\left(\left(\frac{l_0^2}{Dt}\right)^{-2}\right)\right]$ so that the correction becomes negligible for times much larger than the crossover time $t_{\mathrm{cr}} = 3l_0^2/(16D)$. For $\gamma \neq 0$ the ratio of the diffusion coefficients, $D_1 t^{\gamma z}/D_2$, controls the corrections. According to the simulations, this ratio is of order 30 when the asymptotic scaling becomes valid. Hence, the crossover time depends on $|\gamma|$ as $t_{\mathrm{cr}} \sim r^{(2-\gamma)/|\gamma|}$, where $r \approx 30$. As t_{cr} diverges for $|\gamma| \to 0$ the asymptotic scaling regime can be reached in simulations only for relatively large values of $|\gamma|$.

Consider now eq. (2) for $\gamma > 0$. The mixed spatial derivative can be eliminated by making the transformation $y_1 = x_{12} + x_{23}$, $y_2 = g(t)(x_{12} - x_{23})$, where the choice $g(t) = \sqrt{D_1 t^{\gamma z}/(2D_2 + D_1 t^{\gamma z})}$ ensures isotropic diffusion. The absorbing boundaries transform into a wedge with a time-dependent wedge angle. This makes the exact solution hard. However, by further making a change to the timescale $T(t)$ the leading term of the survival probability at late times can be obtained by solving a simple diffusion equation, $\partial_T \rho = 2D_1(\partial^2_{y_1} + \partial^2_{y_2})\rho$, in the final wedge angle $\Theta_\infty = \pi/2$. Thus [2] $P_{\mathrm{RW}}(t) \sim T^{-\pi/2\Theta_\infty} \sim T^{-1} \sim t^{-(1+\gamma z)}$. Since $z = 1/(2-\gamma)$ the survival exponent reads $\theta_{\mathrm{RW}}(\gamma) = 2z = 2/(2-\gamma)$. The approximation obtained by replacing the time-dependent angle by the final opening angle corresponds to putting $D_2 = 0$ in eq. (2), *i.e.*, to taking the center particle to be at rest. This guess can be validated by directly solving equation (2) and analyzing the limiting behavior of the solution [13]. Note that in this limit $\theta_{\mathrm{RW}}(\gamma)$ can be simply determined from two independent random walkers with a *fixed* absorbing boundary in between.

Figure 1 shows the survival probabilities obtained from simulations. $P_{\mathrm{RW}}(t)$ clearly decays as a power law for large times. For $\gamma = 0$ the survival exponent saturates to the asymptotic value $\theta_{\mathrm{RW}} = 3/2$ around $t \approx 10^4$ as shown in the inset, where the local exponent is presented. The exponent saturates also for $\gamma = 0.75$ and $\gamma = 1$. For $0 < \gamma < 0.75$ it is slowly approaching the asymptotic value given by $\theta_{\mathrm{RW}} = 2/(2-\gamma)$. The variations of the local exponents for the last time points are due to statistical fluctuations. For $\gamma = 0.75$ the ratio $D_2 t^{\gamma z}/D_1 \approx 30$ at the time when the exponent saturates. This corresponds to $t_{\mathrm{cr}} \approx 3 \times 10^4$ and 2×10^{10} for $\gamma = 0.50$ and 0.25, respectively. Thus we cannot reach the asymptotic regime for $\gamma \lesssim 0.5$.

The RW results are tested by comparing them to the DLCA simulations. These are done in the usual fashion [14], with a monodisperse initial condition and equal distances between neighboring clusters. $P_{\mathrm{S}}(t)$ is independent of the initial distribution except for transient effects [15]. As fig. 2 shows, the DLCA and RW survival probabilities agree except for a small difference between the amplitudes. The initial inter-particle distances are taken to be the

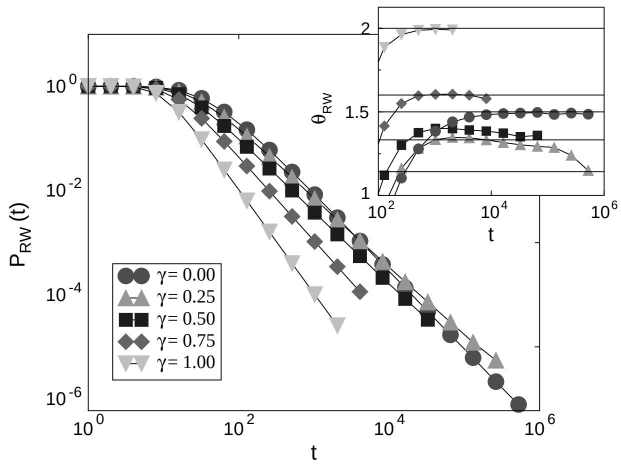

Fig. 1 – The survival probability of three random walkers. The inset shows the corresponding local exponents. The horizontal lines correspond to the analytic values given by $\theta_{\mathrm{RW}} = 2/(2-\gamma)$. The data are averaged over a variable number of realizations ranging from 10^9 for $\gamma = 0$ to 2×10^7 for $\gamma = 0.5$. The initial distance between particles is 10.

same, in order that the RW-picture be as close to DLCA as possible. Notice that the figure contains a case ($\gamma = 0.33$) in which the asymptotic regime is not reached.

For $\gamma < 0$ the situation is more tricky and will be considered in more detail elsewhere [13]. Briefly, analysing eq. (2) similarly as above leads to a closing wedge with the final angle $\Theta_\infty = 0$. This corresponds to a situation where now the particles 1 and 3 are fixed and therefore to a simple exponential decay for the survival [2]. However, simulations show that the survival probabilities are stretched exponentials for both RW and DLCA systems. The stretching

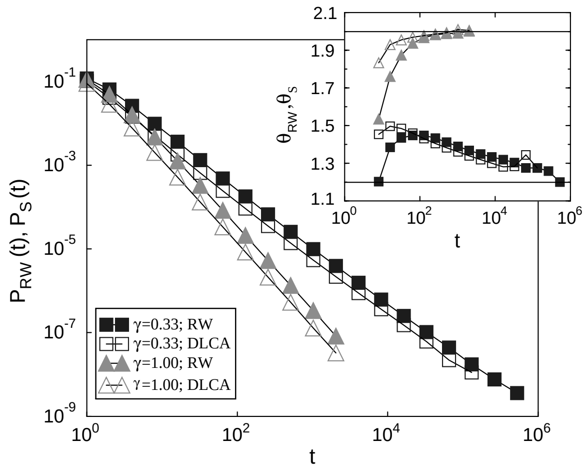

Fig. 2 – Comparison between the random walk and DLCA survival probabilities. The inset shows the corresponding local exponents. The horizontal lines correspond to the analytic values given by $2/(2-\gamma)$. The RW simulations are averaged over 2.5×10^{10} ($\gamma = 0.33$) and 2×10^{10} ($\gamma = 1.00$) realizations with the initial distance between particles being 2. The DLCA simulations are averaged over 50000 realizations on a system of size 55555.

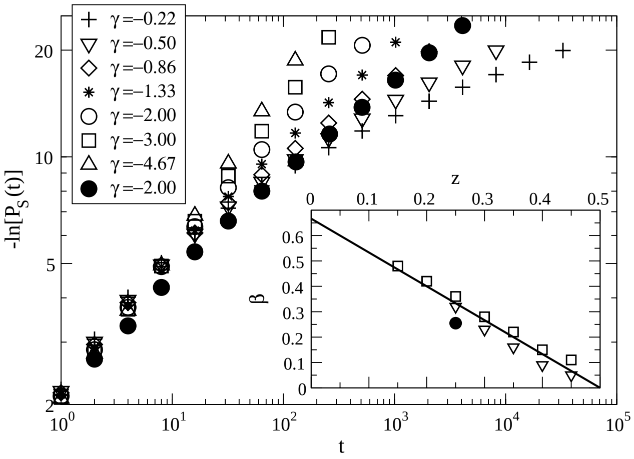

Fig. 3 – The DLCA survival probability for $\gamma < 0$ (open symbols). The inset shows the upper (□) and lower (▽) limits for the stretching exponent. The solid line is given by $2(1-2z)/3$. The data are averaged over 50000 realizations on a system of size 55555. The filled circles show an example of the RW survival.

exponents differ from each other implying that the mean-field RW picture is insufficient to describe the DLCA survival for $\gamma < 0$ (see fig. 3). We concentrate here only on the DLCA case.

In fig. 3 the DLCA survival probabilities for several $\gamma < 0$ are plotted in such a way that a straight line corresponds to a stretched exponential decay and the slope of the line gives the stretching exponent β_S. The smaller the γ the longer it takes to reach the asymptotic scaling regime. This is consistent with $t_\mathrm{cr} \sim r^{(2-\gamma)/|\gamma|}$. The inset shows the stretching exponent as a function of the dynamic exponent z. The squares correspond to the values obtained by fitting a line to the three or four largest time points. For γ close to zero the asymptotic regime is not yet reached and the method gives an upper bound for the exponent. We also considered the change in the local exponent and extrapolated to $1/t = 0$. This approach neglects the saturation of the local exponent after t_cr and gives a lower bound for β_S.

As a result, the numerics suggest an expression $\beta_\mathrm{S} = 2(1-2z)/3$, which lies between the bounds. We also checked that the average distance between the neighbors of unaggregated clusters grows as t^z. If the distance grew *deterministically* as t^z, this would lead to a stretched exponential decay with $\beta_\mathrm{det} = 1-2z$ [16]. Here the fluctuations violate this relation. Naturally $\beta_\mathrm{S} < \beta_\mathrm{det}$ as the fluctuations making the distance longer have a larger weight than those making it shorter when calculating the survival (for an interval of fixed length l the survival probability $\sim \exp[-\pi^2 Dt/l^2]$). Note that $\lim_{\gamma\to-\infty} \beta_\mathrm{S} = 2/3$ and not a simple exponential ($\beta_\mathrm{S} = 1$) as one might expect based on the case of two immobile neighbors. It is also quite surprising, that the mean-field approximation works better for a broad cluster size distribution ($\gamma \geq 0 : f(x) \sim x^{-\tau} (x \to 0)$) than in the case, where the distribution is narrow around the mean ($\gamma < 0 : f(x)$ decays faster than any power law at both ends).

To summarize the results the survival probability decays as

$$P_\mathrm{S}(t) \sim \begin{cases} \exp[-Ct^{\beta_\mathrm{S}}], & \gamma < 0, \\ t^{-3/2}, & \gamma = 0, \\ t^{-2/(2-\gamma)}, & 0 < \gamma < 2. \end{cases} \tag{4}$$

The survival exponent is discontinuous at $\gamma = 0$, *i.e.*, $3/2 = \theta_\mathrm{S}(0) > \theta_\mathrm{S}(0^+) = 1$. This

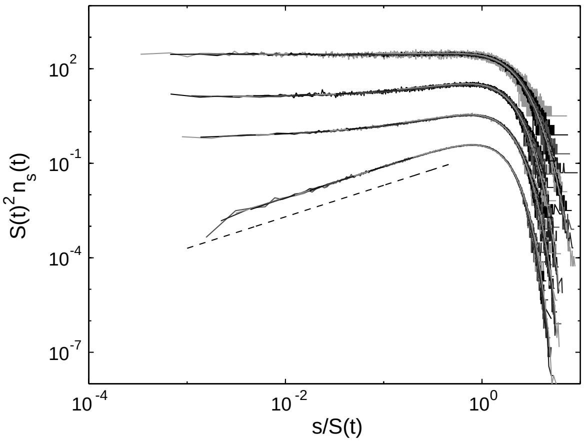

Fig. 4 – The scaling of the cluster size distributions for $\gamma = 0.00$, 0.40, 0.57, and 1.00 (from bottom to top). Distributions are shown at times from $t = 2^3$ to $2^{k_{\max}}$, where $k_{\max} = 17$, 15, 14, and 11 for $\gamma = 0.00$, 0.40, 0,57, and 1,00, respectively. The dashed line has a slope 1. The data are averaged over 50000 realizations on a system of size 55555. The distributions (except the $\gamma = 0$ distribution) have been shifted in the vertical direction for clarity.

seems first counterintuitive since making some of the particles diffuse faster helps the other to survive longer! This is the best since the surviving particle eventually "discovers" the optimal strategy [17] of remaining stationary. This is further confirmed by the fact that the probability of finding a site never occupied by any of the clusters decays as power law with the exponent $2/(2-\gamma)$ for all $\gamma < 2$ [15].

Next we show what these results imply for the cluster size distribution. The third exponent of interest in DLCA scaling is the decay exponent, w, which describes the decrease of the number of clusters of a fixed size s as a function of time $n_s(t) \sim t^{-w}$. As the other exponents z and τ, it is expected to depend on γ. The three exponents are related by the scaling relation $w = (2-\tau)z$ [18]. Therefore, dynamic scaling is fully characterized by any two of the exponents. Even on mean-field level (Smoluchowski's equation) the only easy exponent is z since it does not involve the full scaling function as in the case of τ and w [9].

For $\gamma = 0$ an exact solution of the cluster size distribution $n_s(t)$ is possible, with $w = 3/2$ for any short-range correlated initial distribution $n_s(0)$ [5]. For a monodisperse initial condition, $n_s(0) = \delta_{1,s}$, the survival probability is simply $n_1(t)$, yielding the exponent $\theta_{\rm S}(0) = 3/2$. For $\gamma > 0$ the dynamics of the clusters in the $s \ll S(t)$ part of the size distribution is dictated by collisions with larger, faster ones, that remove such small clusters from the tail. The mechanism by which clusters stay in the tail should be the same as for the survival problem. Thus for $0 < \gamma < 2$ one should have for the decay exponent $w = \theta_{\rm S} = 2/(2-\gamma) = 2z$ together with the scaling relation

$$\theta_{\rm S} = (2-\tau)z. \tag{5}$$

The numerically estimated values for the exponents fulfill eq. (5) within the error bars for all values of the diffusion exponent γ [13]. Hence, the polydispersity exponent is discontinuous at $\gamma = 0$ since $\tau(0) = -1 \neq 0 = \tau(0^+)$, similarly to some examples on the mean-field level [19]. It is also surprisingly enough independent of the value of γ.

Simulations confirm this although crossover effects make the analysis intractable near $\gamma = 0$. Figure 4 shows the scaling plots for the cluster size distribution for various values of the diffusion exponent. The bigger the γ is the faster the scaling function approaches a constant near $x = 0$. For $\gamma \lesssim 0.5$ the times reached in simulations are too small to reveal the asymptotic scaling behavior. In this region also the measurements of w from $n_s(t) \sim t^{-w}$ give information only on the crossover effects [14].

In summary, we have studied the survival of clusters in DLCA in one dimension. The decay of the initial state equals the density of clusters that stay intact by not aggregating with others. This can be analyzed on the mean-field level as the survival of a random walker bounded by two others with time-dependent diffusion coefficients. For $\gamma > 0$ the surviving particles are such that they effectively become immobile. The resulting survival exponent is non-monotonic and discontinuous at $\gamma = 0$. Cluster survival determines the polydispersity exponent τ that characterizes the small cluster size tail. It is discontinuous and constant, $\tau = 0$, for $0 < \gamma < 2$. For $\gamma < 0$ the survival probability decays stretched exponentially with the exponent $\beta_S = 2(1-2z)/3 = -2\gamma/[3(2-\gamma)]$ and the spatial fluctuations are relevant in the sense that the mean-field RW picture gives only a qualitative understanding of the survival.

Above one dimension the study of cluster survival will be both interesting and much less straightforward. A similar mean-field picture in terms of first-passage times of random walks is not directly applicable. For example, one lacks much of the theory needed to analyze the survival behavior of many interacting particles. We conclude with the conjecture that the solution of the survival problem is related to the cluster size distribution also in higher dimensions. There it should be possible to find experimental realizations, to study the survival phenomenon [20].

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