Exact Calculation of the Probabilities of Rare Events in Cluster-Cluster Aggregation

R. Rajesh,[1,2,*] V. Subashri,[1,2,†] and Oleg Zaboronski[3,‡]
[1]The Institute of Mathematical Sciences, C.I.T. Campus, Taramani, Chennai 600113, India
[2]Homi Bhabha National Institute, Training School Complex, Anushakti Nagar, Mumbai 400094, India
[3]Mathematics Institute, University of Warwick, Gibbet Hill Road, Coventry CV4 7AL, United Kingdom

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We develop an action formalism to calculate probabilities of rare events in cluster-cluster aggregation for arbitrary collision kernels and establish a pathwise large deviation principle with total mass being the rate. As an application, the rate function for the number of surviving particles as well as the optimal evolution trajectory are calculated exactly for the constant, sum, and product kernels. For the product kernel, we argue that the second derivative of the rate function has a discontinuity. The theoretical results agree with simulations tailored to the calculation of rare events.

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The study of cluster-cluster aggregation (CCA), a nonequilibrium, irreversible phenomenon where particles, or clusters coalesce on contact to form larger clusters, has a long history dating back to Smoluchowski in 1917 [1]. It has been studied extensively because of its occurrence in diverse physical phenomena such as blood coagulation [2], cloud formation [3,4], aerosol dynamics [5], coagulation of dust and gas particles forming Saturn's rings [6], aggregation of particulate matter in oceans [7], protein aggregation [8,9], charged biopolymers [10,11], ductile fracture [12], etc. CCA also finds applications in applied fields such as river networks [13], mobile networks [14], population genetics [15], and explosive percolation [16,17], etc.

CCA has been analyzed using different approaches. The most common approach is to solve the deterministic mean-field Smoluchowski equation that describes the change in the number of clusters of a given mass due to coagulation events (see Refs. [18–21] for reviews). The Smoluchowski equation for the mean mass distribution is exactly solvable when the rate of collision is independent of the masses (constant kernel), is the sum of the masses (sum kernel), and product of the masses (product kernel). For the product kernel, a sol-gel transition is observed wherein the total mass is not conserved beyond a gelling time. For the sum kernel, the gelling occurs at infinite time [20]. In lower dimensions, spatial density fluctuations become important and have been studied using both analytical and numerical techniques [22–25]. These approaches are, however, restricted to studying the mean or typical mass distribution and the low order moments of the mean mass distributions and do not give information about either the probabilities of rare or atypical events or the trajectories that lead to atypical events. In this Letter, we present an exact calculation of these probabilities.

The tails of a probability distribution describe events, which while rare, are important to study because their impact may be significant. Examples of impactful rare events include heat waves [26,27], earthquakes [28], extreme events in climate and ecosystems, such as the loss of sea ice in the Arctic region [29], etc. In particular, examples of rare events in CCA include neurological disorders such as Alzheimers disease ([30]), mad cow disease [31], the clustering of raindrops leading to rapid onset of rainfall [32], etc. The probabilities of rare events is captured by the large deviation function (LDF) or the rate function, and falls into the general framework of large deviation theory. The LDF can be interpreted as a nonequilibrium generalization of entropy or free energy.

Consider a collection of massive particles which evolves in time through binary mass-conserving aggregation (also known as the Marcus-Lushnikov model [33–36]):

$$A_i + A_j \xrightarrow{\lambda K(i,j)} A_{i+j}, \tag{1}$$

where A_k denotes a particle of mass k and $\lambda K(i,j)$ is the rate at which two particles of masses i and j aggregate. We note that all the spatial information has been encoded into the collision kernel, $K(i,j)$. Let $N(t)$ denote the number of particles at time t. Initially, there are $N(0) = M$ particles of equal mass (set equal to 1). A quantity of interest is the probability density function $P(M,N,t_f)$, of having exactly N particles remain at time t_f. Additionally, we ask what the most probable trajectory is for a given M,N,t_f.

In this Letter, we study the LDF for CCA using the Doi-Peliti-Zeldovich (DPZ) method [37–42], a path integral method. The LDF is calculated exactly for the constant,

*Contact author: rrajesh@imsc.res.in
†Contact author: subashriv@imsc.res.in
‡Contact author: olegz@maths.warwick.ac.uk

sum, and product kernels. For the product kernel, we argue that the LDF is singular with a discontinuity in the second derivative, indicating the sol-gel transition. Gelation transition has been studied using large deviation theory in the probability literature (see [43] and references within). These results are based on special factorization properties of the law of the mass distribution. In contrast, we derive the pathwise large deviation principle which, at least formally, is valid for arbitrary kernels. This more general point of view allows us to determine the optimal evolution trajectories as solutions to the Euler-Lagrange equations for effective action functional.

We first express $P(M,N,t_f)$ in terms of an effective action [44,45]. Let $\tilde{P}(\vec{N},t)$ denote the probability of a system being in a configuration $\vec{N}$ at time t, where $\vec{N}(t)=\{N_1(t),N_2(t),\ldots N_M(t)\}^{\mathsf{T}}$, and $N_i(t)$ is the number of particles of mass i at time t. Then,

$$P(M,N,t_f)=\sum_{\vec{N}}\tilde{P}(\vec{N},t_f)\delta\left(\sum_{i=1}^{M}N_i(t_f)-N\right). \quad (2)$$

The time evolution of $\tilde{P}(\vec{N},t)$ is described by the master equation:

$$\frac{d\tilde{P}(\vec{N})}{dt}=\sum_{i,j}\frac{\lambda K(i,j)}{2}[(N_i+1+\delta_{i,j})(N_j+1)\tilde{P}(\vec{N}+\mathcal{I}_i+\mathcal{I}_j-\mathcal{I}_{i+j})-N_i(N_j-\delta_{i,j})\tilde{P}(\vec{N})], \quad (3)$$

where $\mathcal{I}_k$ is the M-dimensional column vector whose jth component equals δ_{jk}. The first term in the right-hand side of Eq. (3) enumerates all possible collisions that lead to $\vec{N}$ while the second term enumerates all possible collisions that lead to the system exiting $\vec{N}$.

The DPZ formalism allows one to rewrite the master equation in the form of a Schrödinger equation in imaginary time. The corresponding effective Hamiltonian is a polynomial in annihilation and creation operators $a_m, a_m^\dagger$ of particles of mass $m\geq 1$. These satisfy the canonical commutation relations $[a_m,a_n^\dagger]=\delta_{mn}$, $[a_m,a_n]=[a_m^\dagger,a_n^\dagger]=0$. Using the Trotter formula and the complete set of coherent states $\{\langle\overline{z_i},i=1,2,\ldots|,|z_j,j=1,2,\ldots\rangle\}_{z,\bar{z}\in\mathbb{C}}$, a solution to the master equation can be constructed in the form of a path integral. In particular, the probability $P(M,N,t_f)$, after substituting $\phi=N/M$ and $\tau=M\lambda t_f$, can be written as (see [46] for the derivation)

$$P(M,N,t_f)=\sum_{k_i=1}^{k^*}{}'\int\frac{\mathcal{D}\tilde{z}_i\mathcal{D}z_i}{N!}\prod_{n=1}^{N}z_{k_n}(\tau_f)e^{-MS(\phi,\tau_f;\{z_i,\tilde{z}_i\})}, \quad (4)$$

where $k^*=M-N+1$, and $'$ denotes the constraint $\sum_i k_i=M$. The variables $z,\tilde{z}$ parametrize the symbol $H(\{z_i\},\{\tilde{z}_i\})$ of the evolution operator. The action S is given in terms of the effective Hamiltonian H as

$$S=\int_0^{\tau_f}d\tau\left[\sum_{m=1}^{M}\tilde{z}_m\dot{z}_m+H(\{z_i,\tilde{z}_i\})\right]-\ln\tilde{z}_1(0)+1, \quad (5)$$

$$H(\{z_i\},\{\tilde{z}_i\})=-\frac{1}{2}\sum_{i,j}K(i,j)(\tilde{z}_{i+j}-\tilde{z}_i\tilde{z}_j)z_iz_j. \quad (6)$$

The action is invariant under the transformation $z_m\to c^m z_m$ and $\tilde{z}_m\to c^{-m}\tilde{z}_m$. Hence, we can set $\tilde{z}_1(0)=1$.

In the limit $M\to\infty$, keeping ϕ and τ_f fixed, the functional integral in Eq. (4) is dominated by the minimum of S, and hence can be calculated using the Laplace method. The corresponding Euler-Lagrange equations for $z_m,\tilde{z}_m$, $m=1\ldots M$, are

$$\frac{dz_m}{d\tau}=\frac{1}{2}\sum_j K(m-j,j)z_jz_{m-j}-\sum_j K(m,j)\tilde{z}_jz_mz_j, \quad (7)$$

$$\frac{d\tilde{z}_m}{d\tau}=-\sum_j K(m,j)(\tilde{z}_{m+j}-\tilde{z}_m\tilde{z}_j)z_j, \quad (8)$$

with the boundary conditions $z_m(0)\tilde{z}_1(0)=\delta_{m,1}$ and $\tilde{z}_m(\tau_f)z_m(\tau_f)=M^{-1}\sum_{k_n=1}^{k^*}\delta_{k_n,m}$. These give M boundary conditions for z_i at $\tau=0$, and M boundary conditions for $\tilde{z}_i$ at $\tau=\tau_f$. The time evolution of $n=\sum_i z_i\tilde{z}_i$, the fraction of particles, is then given by

$$\frac{dn}{d\tau}=\frac{-\sum_{i,j}K(i,j)n_in_j}{2}+E,\quad n(0)=1,\quad n(\tau_f)=\phi. \quad (9)$$

For $\tilde{z}_i$, z_i satisfying the Euler Lagrange equations, it can be shown that H reduces to $2H=\sum_i z_i(d\tilde{z}_i/d\tau)$. Also, H is a constant of motion (see [46] for the proof), and we denote its value by E. We note that Eq. (8) is satisfied by $\tilde{z}_i(\tau)=1$, in which case, $E=0$ [see Eq. (6)]. For this special case, Eq. (7) for z_m is identical to the Smoluchowski equation for the mean mass distribution, and thus will correspond to the typical solution for a given time. We now discuss the general case, $E\neq 0$, corresponding to atypical solutions. Evaluating the integral Eq. (5), we then obtain

$$P(M,N,\tau_f)\sim\max_{\{k_i\}}{}'\prod_{n=1}^{N}z_{k_n}(\tau_f)e^{-M[\phi\ln\phi-E\tau_f]}. \quad (10)$$

Equations (7), (8), and (10) describe the calculation of the LDF for an arbitrary kernel.

Since $N=\phi M$, it is clear that in the limit $M\to\infty$, keeping ϕ and τ_f fixed, we can define a LDF

$$f(\phi,\tau)=\lim_{M\to\infty}\frac{-1}{M}\ln P(M,M\phi,\tau/M), \quad (11)$$

thus establishing a large deviation principle for any collision kernel. We will now present an exact calculation of $f(\phi,\tau)$ for the constant, sum, and product kernels.

Constant kernel [$K(i,j)=1$]—The instanton equation, Eq. (9), reduces to $dn/d\tau=-n^2/2+E$. Since $n(\tau)$ decreases with time, $E<n^2/2$. The solution for $n(\tau)$ is (see [46] for more details)

$$n(\tau)=\begin{cases}\frac{1}{1+\tau/2}, & E=0,\\ \sqrt{2E}\coth\frac{\sqrt{2E}(\tau-\tau_0)}{2}, & E\neq 0,\end{cases} \tag{12}$$

where the constants E,τ_0 are determined in terms of ϕ and τ_f through the boundary conditions in Eq. (9). For determining the LDF, we also need to determine $z_m(\tau_f)$ and $\tilde{z}_1(0)$. Writing $z_m(\tau)$ in terms of its generating function, $Y(x,\tau)=\sum_m z_m(\tau)x^m-n(\tau)$, we obtain

$$\frac{\partial Y}{\partial\tau}=\frac{Y^2}{2}-E,\qquad Y(x,0)=y_1(0)x. \tag{13}$$

Solving for Y and hence $z_m(\tau)$, we obtain

$$z_m(\tau)=\begin{cases}\frac{4\tau^{m-1}(z_1(0))^m}{(2+\tau)^{m+1}}, & E=0,\\ \frac{2Ez_1(0)^m\sinh^{m-1}\sqrt{E/2}\tau}{[\sinh\sqrt{E/2}\tau+\sqrt{2E}\cosh\sqrt{E/2}\tau]^{m+1}}, & E\neq 0.\end{cases} \tag{14}$$

The maximization over $\{k_i\}$ in Eq. (10) is easily done and leads to a factor $\binom{M-1}{N-1}$. Substituting for $z_m(\tau)$ in Eq. (10), the LDF is

$$f(\phi,\tau)=\begin{cases}\phi\ln\frac{\phi^2}{-2E+\phi^2}+\ln(1-2E)-E\tau, & E<0,\\ 0, & E=0,\\ -E\tau-\phi\ln\frac{2E}{\phi^2}-(1-\phi)\ln\frac{\sinh\tau\sqrt{E/2}}{1-\phi}+ & \\ (1+\phi)\ln(\sqrt{2E}\cosh\tau\sqrt{E/2}+\sinh\tau\sqrt{E/2}), & E>0,\end{cases} \tag{15}$$

where $E<0$, $E=0$, $E>0$ correspond to final times $\tau<\tau_{\rm typ}$, $\tau=\tau_{\rm typ}$ and $\tau>\tau_{\rm typ}$, respectively, and $\tau_{\rm typ}$ is the typical time for the fraction of particles to reach ϕ.

We demonstrate the correctness of the solution as well as the procedure by comparing $f(\phi,\tau)$ with results from both Monte Carlo simulations and the exact expression for $P(M,N,t)$. The simulations are based on a biased Monte Carlo scheme for fixed number of particles [47] that accurately determines the probabilities of rare events and the instanton trajectory. We generalize the algorithm to allow for number of particles to fluctuate (see [46] for more details). For the constant kernel, the reaction rate does not explicitly depend on the mass distribution and hence it is possible to write $P(M,N,t)$ as a sum over exponentials [47]. We note that it is difficult to extract the LDF from this expression, however, it can be evaluated numerically. We find an excellent agreement of $f(\phi,\tau)$ with the simulations and exact answer both for fixed τ and varying ϕ [see Fig. 1(a)], and fixed ϕ and varying τ [see Fig. 1(b)]. The analytical results for the instanton solution [see Eq. (12)] are also in excellent agreement with the numerical results for short, typical, and long times [see Figs. 1(c) and 1(d)].

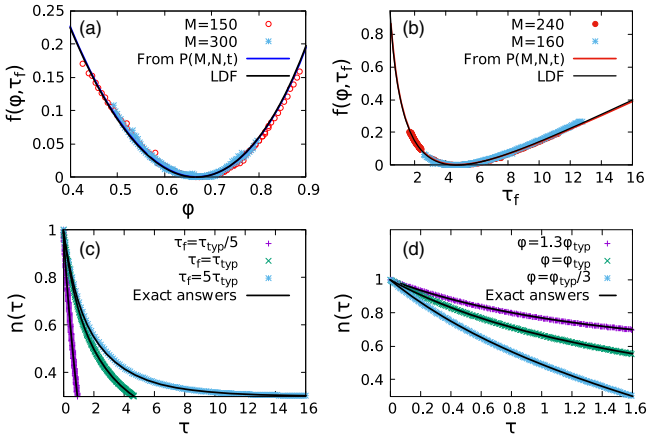

FIG. 1. Constant kernel: Comparison of $f(\phi,\tau)$ with simulation data and exact expression for $P(M,N,t)$ for (a) varying ϕ for fixed $\tau_f=1$, (b) varying τ_f for $\phi=0.3$. The instanton trajectory in Eq. (12) is compared with simulation data for (c) $\phi=0.3$ and different τ_f and (d) $\tau_f=1.6$ and different ϕ.

Sum kernel [$K(i,j)=(i+j)/2$]—The instanton equation, Eq. (9), reduces to $dn/d\tau=E-n/2$, with solution

$$n(\tau)=\frac{\phi-e^{-\tau_f/2}}{1-e^{-\tau_f/2}}-\left(\frac{\phi-1}{1-e^{-\tau_f/2}}\right)e^{-\frac{\tau}{2}}. \tag{16}$$

The Euler-Lagrange equations for z_i [see Eq. (7)] can now be solved (see [46]) to give

$$z_i(\tau)=\frac{i^{i-1}a_1^i}{i!}(1-e^{-\tau/2})^{i-1}e^{-\int d\tau' \frac{in+1}{2}}, \tag{17}$$

where a_1 is a constant. The maximization over $\{k_i\}$ in Eq. (10) is then

$$\sum_{k_i=1}^{k^*}{}'\prod_{n=1}^{N}\frac{k_n^{k_n-1}}{k_n!}=e^{M(1-\phi)[1-\ln(1-\phi)]}. \tag{18}$$

Substituting $z_m(\tau)$ and the prefactor in Eq. (10), we obtain LDF for sum kernel to be

$$f(\phi,\tau)=-(1-\phi)\ln\frac{1-e^{-\frac{\tau}{2}}}{1-\phi}+\frac{\tau\phi}{2}+\phi\ln\phi. \tag{19}$$

We find an excellent agreement of $f(\phi,\tau)$ with the simulations both for fixed τ and varying ϕ [see Fig. 2(a)],

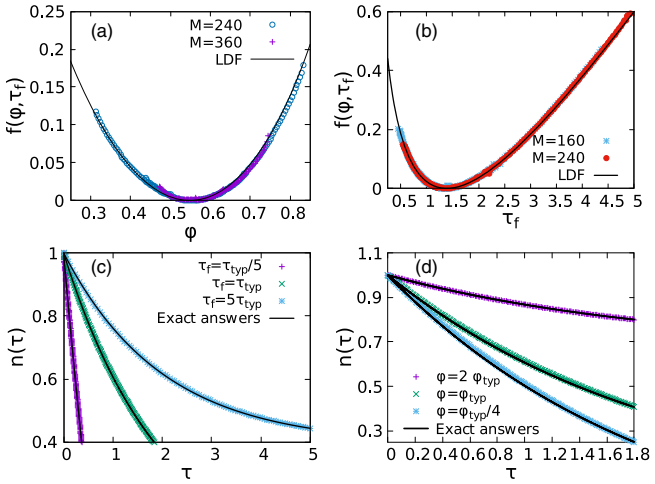

FIG. 2. Sum kernel: Comparison of $f(\phi,\tau)$ with simulation data for (a) varying ϕ for fixed $\tau_f = 1.2$, (b) varying τ_f for $\phi = 0.5$. The instanton trajectory in Eq. (16) is compared with simulation data for (c) $\phi = 0.4$ and different τ_f and (d) $\tau_f = 1.8$ and different ϕ.

and fixed ϕ and varying τ [see Fig. 2(b)]. The analytical results for the instanton solution [see Eq. (16)] are also in excellent agreement with the numerical results for short, typical and long times [see Figs. 2(c) and 2(d)].

Product kernel—For the product kernel the Smoluchowski equation predicts that a gel that contains a finite fraction of the mass forms at gelling time $\tau_g = 1$ and gelling density $\phi_g = 0.5$. In the discussion following Eq. (9), we showed that $E = 0$ corresponds to the solution to the Smoluchowski equation. However, this solution cannot be correct for $\tau \geq 1$ as mass is not conserved, violating the strict conservation of mass in the Marcus-Lushnikov model. We, therefore, modify the solution for product kernel as follows.

We rewrite the unscaled Hamiltonian using number operator $\hat{n}_i$ and total mass operator $\hat{M}$, breaking normal ordering. To restore normal ordering, we use the relation $\sum_i \hat{M}|\psi(0)\rangle = M|\psi(0)\rangle$, where $|\psi(0)\rangle = a_1^{\dagger M}|\vec{0}\rangle$, to rewrite $P(M,N,t_f)$ as

$$P(M,N,t_f) = \langle \vec{N}| \frac{(\sum_i a_i)^N}{N!} e^{-H'(\{a_i^\dagger\},\{a_i\})t_f}|\psi(0)\rangle, \tag{20}$$

$$H' = -\frac{1}{2}\sum_i \sum_j ija_{i+j}^\dagger a_i a_j + \sum_j \frac{(Mj - j^2)a_j^\dagger a_j}{2}. \tag{21}$$

On introducing coherent states, we obtain the Euler-Lagrange equations to be

$$\dot{z}_k = \frac{1}{2}\sum_{l=1}^{k-1} l(k-l)z_l z_{k-l} - Mkz_k + \frac{k^2 z_k}{2}, \tag{22}$$

$$\dot{\tilde{z}}_k = -\sum_{l=1}^{k^*} kl\tilde{z}_{l+k}z_l + kM\tilde{z}_k - \frac{k^2\tilde{z}_k}{2}. \tag{23}$$

We note that we could have followed the same procedure of introducing the operators $\hat{M}$ and $\hat{n}_i$ for the constant and sum kernels. For these kernels, the extra terms are always subleading in M and thus, we obtain the same LDF. However, for the product kernel, the extra terms become important when a gel is present, and hence cannot be neglected.

Equation (22) can be solved exactly. Let $G(x,t) = \sum_{m=1}^{M} z_m(t)x^m$. Then, making the Cole-Hopf transformation $G(x,t) = \ln D[p(x,t),t]$, where $p(x,t) = xe^{-Mt}$, and solving the resulting partial differential equation using Knuth identity [48] (see [46] for details), we obtain z_m to be

$$z_m(\tau) = \frac{(e^{\tau/M}-1)^{m-1}F_{m-1}(e^{\tau/M})M^{m-1}e^{-m\tau}}{m!}, \tag{24}$$

where $F_m(x)$ are the Mallows-Riordan polynomials [34,48]. From Eqs. (22) and (23), we find that $\sum_i \dot{z}_i\tilde{z}_i = -E' - M^2/2$, where E' is the value of H'. Substituting for z_i in Eq. (10), and computing the combinatorial prefactor, we obtain the LDF for the product kernel:

$$f(\phi,\tau) = \ln\frac{\phi^\phi e^{\tau/2+1-\phi}}{\tau^{1-\phi}} + \min_x\{\ln x - \phi h(x)\}, \tag{25}$$

$$h(x) = \sum_{k=1}^{k^*}\frac{x^k F_{k-1}(e^{\tau/M})}{k!}. \tag{26}$$

We find an excellent agreement of $f(\phi,\tau)$ with the simulations for both pregelling and postgelling regimes

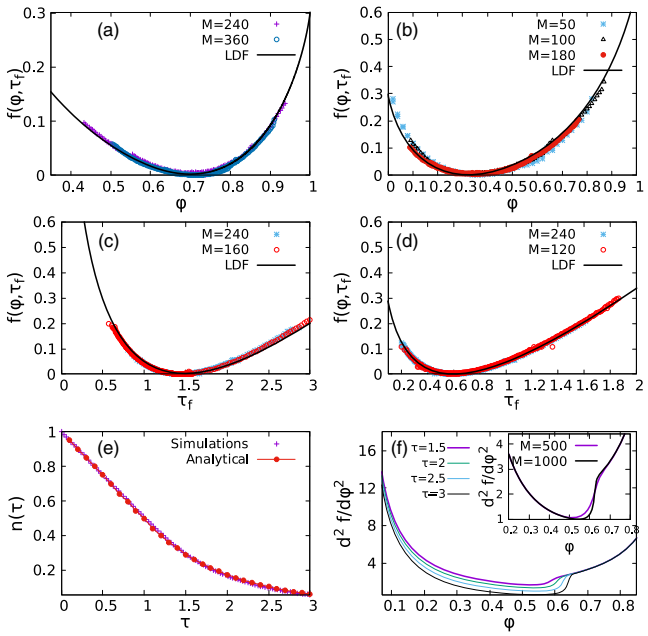

FIG. 3. Product kernel: Comparison of $f(\phi,\tau)$ with simulation data for (a) fixed $\tau_f = 0.6 < \tau_g$, (b) fixed $\tau_f = 1.4 > \tau_g$, (c) fixed $\phi = 0.3 < \phi_g$, and (d) fixed $\phi = 0.7 > \phi_g$. (e) The value of ϕ at minimum of $f(\phi,\tau)$ is compared with Monte Carlo simulations of the typical trajectory. (f) $d^2f/d\phi^2$ is discontinuous with ϕ. Inset: the discontinuity becomes sharper with increasing M.

[see Figs. 3(a)–3(d)]. We also confirm that the minimum of the action corresponds to the typical solution [see Fig. 3(e)]. Finally, we find that the $\partial^2 f/\partial^2 \phi$ has a discontinuity at a critical ϕ [see Fig. 3(f)]. The discontinuity becomes sharper with M [see inset of Fig. 3(f)], suggesting the presence of a second order phase transition.

In summary, we developed a formalism to calculate the probabilities of rare events in cluster-cluster aggregation and demonstrated the existence of a large deviation principle for any collision kernel. The LDF is calculated exactly for the constant, sum, and product kernels. The known sol-gel transition for the product kernel is reflected as a singular behavior in the LDF. Our general method allows us to obtain the optimal evolution trajectory corresponding to any rare event. These exact solutions will serve as a guideline for the numerical investigation of rare events in aggregation with collision kernels applicable to particular physical systems.

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