Nontrivial polydispersity exponents in aggregation models

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We consider the scaling solutions of Smoluchowski's equation of irreversible aggregation, for a nongelling collision kernel. The scaling mass distribution $f(s)$ diverges as $s^{-\tau}$ when $s\to 0$. τ is nontrivial and could, until now, only be computed by numerical simulations. We develop here *general methods* to obtain exact bounds and good approximations of τ. For the specific kernel $K_D^d(x,y)=(x^{1/D}+y^{1/D})^d$, describing a mean-field model of particles moving in d dimensions and aggregating with conservation of "mass" $s=R^D$ (R is the particle radius), perturbative and nonperturbative expansions are derived. For a general kernel, we find exact inequalities for τ and develop a *variational approximation* which is used to carry out a systematic study of $\tau(d,D)$ for K_D^d. The agreement is excellent both with the expansions we derived and with existing numerical values. Finally, we discuss a possible application to $2d$ decaying turbulence. [S1063-651X(97)02304-0]

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I. INTRODUCTION

Aggregation phenomena are widespread in nature. They have such an impact on materials sciences, chemistry, and astrophysics that a large amount of literature has been devoted to them [1–4]. In such dynamical processes, particles or objects as different in geometry and size as colloidal particles, galaxies, small molecules, vortices in fluids, droplets, and polymers can merge to form a new entity when they come into close contact or interpenetrate, through diffusion (Brownian coagulation [5,6]), ballistic motion (ballistic agglomeration [7–9]), exogenous growth (droplet growth and coalescence [10]), or droplet deposition [11].

One is usually interested in the evolution of the statistical distribution of the "mass" s, a quantity characteristic of each particle, that is conserved in the coalescence process: it can be either the actual mass, the volume, the area, the electric charge, or any other physical quantity, depending on the underlying physics.

Great progress was achieved when it was proposed [12] and observed both in real experiments and in numerical simulations that the mass distribution $N(s,t)$ exhibits scale invariance at large time:

$$N(s,t)\sim S(t)^{-\beta}f\left(\frac{s}{S(t)}\right), \quad S(t)\sim t^{z}. \tag{1.1}$$

The divergence of the mass scale $S(t)$ bears on the oblivion of initial conditions and physical cutoff or discreteness, as does the diverging correlation length of critical phenomena: universality arises in dynamics as well, with new universality classes.

The exponents z and β are often easily derived from conservation laws and physical arguments, but in many cases a polydispersity exponent τ defined by $f(x)\sim x^{-\tau}$ when $x\to 0$ is observed, whose value is nontrivial though universal. The prediction of τ is still a challenge.

Except for a few [usually one-dimensional (1D)] exactly solvable models [13,14], analytical results are still lacking. The most popular approach to these aggregation problems is Smoluchowski's equation [5], a master equation [15] for the one-body distribution $N(s,t)$:

$$\frac{\partial N(s,t)}{\partial t}=\frac{1}{2}\int_0^s N(s_1,t)N(s-s_1,t)K(s_1,s-s_1)ds_1 - N(s,t)\int_0^{+\infty}N(s_1,t)K(s,s_1)ds_1, \tag{1.2}$$

where the aggregation kernel $K(x,y)$ is symmetric and is characteristic of the physics of the aggregation process on a more or less coarse-grained level. Such kinetic equations are usually derived within a mean-field approximation, where density fluctuations are ignored. Mean-field approximation is expected to be valid above an upper critical spatial dimension. This dimension is usually 2 for reaction-diffusion models, but van Dongen showed that it can depend on the kernel [16]. Including some proper approximation of the density-density correlations in the kernel may improve Smoluchowski's approach [17].

Mean field as it may be, Smoluchowski's equation is still highly nontrivial. No exact solution is available, except in a very few specific cases (see below), and extracting the nontrivial exponent τ for a specific system from the proper kinetic equation is not an easy task. The problem was clarified by van Dongen and Ernst [18], who classified the kernels according to their homogeneity and asymptotic behavior:

$$K(bx,by)=b^{\lambda}K(x,y), \tag{1.3}$$

$$K(x,y)\sim x^{\mu}y^{\nu}(y\gg x). \tag{1.4}$$

For a given physical system, the homogeneity λ is easily determined using scaling arguments. We consider only nongelling systems with $\lambda\leq 1$ [18]. For $\mu>0$, the exponent τ is trivial and found to be $\tau=1+\lambda$, whereas for $\mu=0$, τ depends on the whole solution f of the scaling equation derived from Eq. (1.2) [see Eq. (2.5) below]. $\mu<0$ does not lead to any power law behavior but rather to a bell-shaped scaling function f [18].

In the following, we shall focus on the $\mu=0$ case for which the exponent τ has so far only been determined numerically by direct simulation of Smoluchowski's equation [19,20], not an easy task [2,19], by time series [21], and of course by direct simulation of the physical system supposed to be described by the considered Smoluchowski equation [2,6,7,9–12,19]. In the latter case, direct comparison with mean-field results is in principle rather delicate. These methods are quite heavy, which explains that very few values of τ are known [20,21], most of them concerning a specific kernel, $K_D^d(x,y)=(x^{1/D}+y^{1/D})^d$ $(0\leqslant d\leqslant D)$, which appears in various physical applications [2–4,17,22–24].

Considering the ubiquity and the importance of the $\mu=0$ case leading to nontrivial polydispersity exponents, analytical results as well as more effective numerical methods, making it possible to carry out extensive studies, are certainly needed to use Smoluchowski's approach in a predictive way. The purpose of this article is to provide both and use them to perform a complete study of $\tau(d,D)$ for the kernel $K_D^d=(x^{1/D}+y^{1/D})^d$. These analytical methods consist of exact bounds, perturbative and nonperturbative expansions around exactly solvable limits, while we introduce a *variational* scheme, leading to excellent approximations of τ at extremely low computational cost, without directly solving Smoluchowski's equation. We end the paper with a practical application of our results in the field of two-dimensional turbulence.

In Sec. II we present a mean-field model of aggregation of D-dimensional spheres diffusing in a d-dimensional space and coalescing with conservation of their volume, for which we derive a Smoluchowski equation with the kernel $K_D^d=(x^{1/D}+y^{1/D})^d$. Under the scaling hypothesis, we write down the equation for the scaling function, determine the exponents z and β, and derive an integral equation for τ as well as a series of integral equations for the moments of the scaling function f. This section is intended merely to clarify notations, to present the state of the art, and to make a few useful remarks.

Sections III and IV present analytical results for the previously introduced kernel K_D^d. Section III describes a method to obtain exact bounds for any kernel, based on integral equalities established in Sec. II.

Section IV deals with expansions of τ around its value for exactly solvable kernels. Starting from the remark that K_D^d reduces to the constant kernel in both $d\to 0$ and $D\to\infty$ limits, for which an explicit exponential solution is known, we find some perturbative expansions in both limits. In the large D limit with $d/D=\lambda$ fixed, the kernel reduces to $2^d(xy)^\lambda$ and we show that $\tau\to 1+\lambda$, the first correction being exponentially small at large d, and thus nonperturbative.

In Sec. V we present a variational approximation based on integral equations for the moments of f, and valid for *any homogeneous kernel*. This method reproduces some known exact results, and is used to compute τ for a wide range of d and D, the results being summarized in Fig. 2. The approximation is compared to the few existing numerical results [20,21] as well as with analytical expansions derived in Sec. IV, with excellent agreement and very low computational cost.

Section VI presents a possible application in the field of two-dimensional turbulence. We consider a model of diffusing and merging coherent vortices, and Smoluchowski's equation leads to non-Batchelor energy spectra with exponents in qualitative agreement with direct simulations found in the literature [25,26].

II. MODEL AND SCALING

Consider hyperspherical particles in a d-dimensional box, of polydisperse radii R with distribution $F(R,t)$, evolving the following way: at time t we choose the positions of their centers with uniform probability in d space. Then each pair of overlapping spheres of radii R_1 and R_2 merges to form a new sphere of radius,

$$R=(R_1^D+R_2^D)^{1/D}, \tag{2.1}$$

where D is a parameter with $D\geqslant d$. D can be the actual dimension of the spheres, as for instance in the case of $D=3$ spheres deposited on a $d=2$ plane [11]. Once each coalescence has been resolved, we have reached time $t+\delta t$.

A. Derivation of Smoluchowski's equation

The conserved variable is $s=R^D$, and is continuous. We shall call s_0 the physical lower cutoff, that is, the charge of the smallest sphere in the initial condition. Since the radius of a surviving sphere can only increase through coalescence, $N(s,t)=0$ for $s<s_0$ and for any time $t>0$. Smoluchowski's equation consists just in a balance of collisions. The number of collisions between two spheres of radius $s_1^{1/D}$ and $s_2^{1/D}$ randomly and independently deposited in the d-dimensional medium is $N(s_1,t)N(s_2,t)\Omega_d(s_1^{1/D}+s_2^{1/D})^d$ where Ω_d is the d-dimensional total solid angle. We obtain the equation

$$\begin{aligned} N(s,t+\delta t)-N(s,t)& \\ =\Omega_d\bigg\{\frac{1}{2}\int_0^s & N(s_1,t)N(s-s_1,t)K_D^d(s_1,s-s_1)ds_1 \\ & -N(s,t)\int_0^{+\infty}N(s_1,t)K_D^d(s,s_1)ds_1\bigg\}, \end{aligned} \tag{2.2}$$

with $K_D^d(x,y)=(x^{1/D}+y^{1/D})^d$. We can get rid of the multiplicative constant, by properly choosing the time unit δt and by replacing the finite difference in time by a partial derivative to exactly obtain Eq. (1.2). We notice that the only approximation used to derive the equation is to neglect multiple collisions, for the system is intrinsically mean-field.

The kernel $K_D^d(x,y)=(x^{1/D}+y^{1/D})^d$ has been introduced in many contexts from molecular coagulation [17] to cosmology [20,22] for specific values of d and D, and is one of the most studied in the literature [17,18,20–22,27–29] although very few analytical results are known. This kernel has $\lambda=d/D$ and $\mu=0$. Exact solutions are available in the case $d=0$ or $D=\infty$ (constant kernel) [5], and $d=D=1$ [27].

B. Scaling

Now, we introduce the scaling form of $N(s,t)$. We first write the conservation law. The total mass in the system is $\int_{s_0}^{+\infty} sN(s,t)ds \sim S(t)^{2-\beta}\int_0^{+\infty} xf(x)dx$ and is conserved, which implies $\beta=2$, implicitly assuming that the integral of $xf(x)$ converges, i.e., in terms of the small x divergence of f, that $\tau<2$, which will be shown below. We consider the total number of particles in the system $n(t)=\int_0^{+\infty} N(s,t)ds$. It behaves at large time as $S(t)^{1-\beta}\int_{s_0/S(t)}^{+\infty} f(x)dx$. If $\tau<1$, $n(t)\sim S(t)^{1-\beta}\int_0^{+\infty} f(x)dx$ whereas if $\tau>1$, $n(t)\propto S(t)^{\beta-\tau}$. If $\tau=1$, the integral diverges as $\ln[S(t)]$, hence $n(t)\propto S(t)^{1-\beta}\ln[S(t)]$.

As promised, we are now able to show that $\tau<2$. If $\tau>2$, the total charge in the system is proportional to $S(t)^{\tau-\beta}$, enforcing $\beta=\tau$. As a consequence, $n(t)$ would have a nonzero limit, which is impossible. To summarize these results, we have, with $n(t)\propto t^{-z'}$,

$$\beta=2, \tag{2.3}$$

$$z'=\begin{cases} z & \text{if } \tau<1 \\ z(2-\tau) & \text{if } \tau>1. \end{cases} \tag{2.4}$$

The derivation of the scaling equation is rigorously described in [30], where it is shown that $S(t)\sim wt^z$, w being some positive constant characteristic of the time dependent equation. Plugging the scaling form of the distribution into Smoluchowski's equation, and matching the large t behavior of both sides of the equation, yields $z=D/(d-D)$ and the equation for the scaling function,

$$w[sf'(s)+2f(s)] = f(s)\int_0^{+\infty} f(s_1)K_D^d(s_1,s)ds_1 - \frac{1}{2}\int_0^s f(s_1)f(s-s_1)K_D^d(s_1,s-s_1)ds_1. \tag{2.5}$$

If $\tau\geqslant 1$ each term of the right hand side of Eq. (2.5) is separately divergent and they should be properly grouped, for instance,

$$w[sf'(s)+2f(s)] = f(s)\int_{s/2}^{+\infty} f(s_1)K_D^d(s_1,s)ds_1 - \int_0^{s/2} f(s_1)\{f(s-s_1)K_D^d(s_1,s-s_1) - f(s)K_D^d(s_1,s)ds_1. \tag{2.6}$$

Another way of taking care of these divergences is to be found in [18,30].

As we are only interested in the exponent affecting the small s behavior of f, we shall set w to unity by changing f to $w^{1/2}f$. If $f(s)$ is a solution of Eq. (2.5), then $b^{1+d/D}f(bs)$ is also a solution. The value of b is often fixed by imposing $\int xf(x)dx=1$, but we will make a different choice for reasons that will become clear later.

A careful study of the large s behavior of f shows that if $\lambda<1$ $(d<D)$, $f(s)\sim c_\infty \delta s^{-\lambda}e^{-\delta s}$, with $c_\infty^{-1}=\int_0^{1/2}K_D^d(x,1-x)x^{-\lambda}(1-x)^{-\lambda}dx$ [30]. We choose the solution corresponding to $\delta=1$, which fixes b, and leads to a nontrivial value for $\int xf(x)dx$. This asymptotic behavior is not valid for $\lambda=1$ $(d=D)$.

For $d=0$ or $D=\infty$, Eq. (2.5) reduces to the constant kernel equation with exact solution $f_0(x)=2e^{-s}$ and $f_\infty(s)=2^{1-d}e^{-s}$ (note that the large s asymptotics become the exact solution for all s in these cases). For $d=1$ and $D=1$, an exact analytic solution is also known for the time dependent equation, the scaling function being $f(s)\propto s^{-3/2}e^{-s}$ [27], with $z=\infty$ and $S(t)\propto e^t$.

Now, for given d and D, and plugging the expected small s behavior $f(s)\sim s^{-\tau}$ into Eq. (2.5), one first gets that $\tau<1+\lambda=1+d/D$. Then, matching the behavior of both sides of Eq. (2.5) [18,30], one finds

$$\tau=2-\int_0^\infty f(x)x^\lambda dx. \tag{2.7}$$

If $\alpha>\tau-1$ we obtain by multiplying Eq. (2.5) by x^α and integrating [18,28]

$$2(1-\alpha)\int_0^\infty x^\alpha f(x)dx=\int\int_0^\infty f(x)f(y)K_D^d(x,y) \times[x^\alpha+y^\alpha-(x+y)^\alpha]dxdy. \tag{2.8}$$

All these results are valid for any homogeneous kernel with $\lambda<1$ and $\mu=0$ [18,30].

C. Existing analytical and numerical results

Most existing analytical results for $\mu=0$ kernels are to be found in the beautiful series of papers by van Dongen and Ernst [16,18,28,30]. Apart from results mentioned earlier, they determined the small x subleading behavior of the scaling function, and they found some inequalities for τ in the cases $d=1$ and $D=1$. In 1984, Leyvraz [29] proposed the analytical result $\tau=1+1/2D$ for the kernel K_D^d with $d=1$, but in 1985, using exact inequalities, van Dongen and Ernst showed that this result was erroneous and explained why it was so [28]. The argument of Leyvraz leading to this result is perfectly valid for class I kernels with $\mu>0$ for which it predicts the correct exponent, but it breaks down for $\mu=0$ kernels. We mention this fact for some references to the wrong result $\tau=1+1/2D$ can still be found in some recent articles.

We now review various kinds of numerical studies concerning the polydispersity exponent τ. These studies deal with the kernel K_D^d.

Kang *et al.* [19] simulated a model of particle diffusion and coalescence (PCM) that can be shown to be exactly equivalent to Smoluchowski's equation. They also numerically directly computed the solution of the equation itself. Their results concern the $d=1$ case. They surprisingly found values of τ in contradiction with the exact bound $\tau\geqslant 1$ (see Sec. III) (for $D=4$, they found $\tau=0.63$). By comparison between their two methods of computation, they concluded

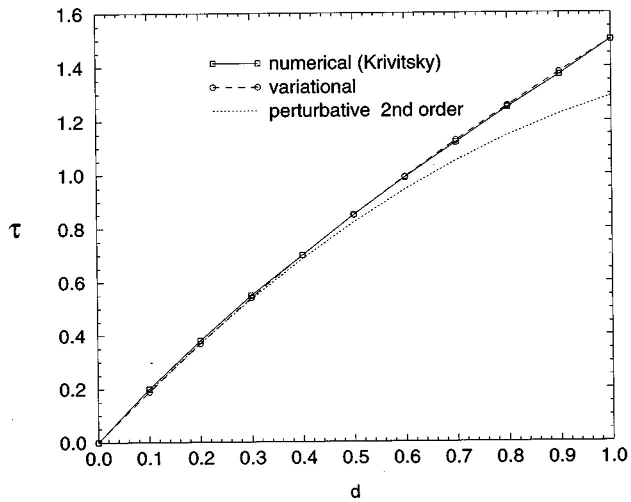

FIG. 1. In $D=1$, the comparison between the results obtained in [20] by Krivitsky, the variational approximation with three parameters and eight moments, and the $O(d^2)$ perturbative expansion of τ, illustrates the efficiency of the variational approximation. Indeed, the agreement between the numerical solution of Smoluchowski's equation [20] and the variational approximation is excellent. The variational approximation is even in closer agreement with the small d perturbative expansion than Krivitsky's result, and although both methods recover the exact result $\tau=3/2$ for $D=1$, Krivitsky's curve seems to have an accident in the vicinity of $D=1$, whereas the variational result is smooth.

that in both cases they observed a pseudoasymptotic state, with wrong exponents but apparent scaling, and that the actual asymptotic scaling regime appeared at times too large to be seen by their simulations. This illustrates the drawback of considering the direct time evolution of the system: the actual asymptotic regime may not be reached within the accessible numerical simulation time scale.

Krivitsky [20] numerically solved Smoluchowski's equation for the time dependent distribution for the kernel K_D^d, for $D=1,d\leqslant 1$, for which he determined ten values for τ (see Fig. 1). Comparison with analytical results obtained by analysis of the scaling equation (infinite time limit) in the present article will assess the fact that in this case the asymptotic regime was actually reached by Krivitsky's solution. These numerical results will be found to be in excellent agreement with our variational method of Sec. V.

Song and Poland [21], computed the large time evolution of the number of clusters $n(t)\propto t^{-z'}$, and as $z'=z(2-\tau)$ when $\tau>1$, and $z'=z$, when $\tau<1$, we can extract τ from their data (for which $\tau>1$). Their method consists in solving the equation for $n(t)$ as a power series in time t, and to extract the exponent z' by manipulations of this series. They treated only the cases $d=1,D=2$ and $d=2,D=3$. In the case $d=1,D=2$, they present two different results in the text. They first consider K_2^1 and find $1/z'=0.57\pm 0.01$, then they extend their method to K_d^{d-1} and in the case $d=2$, which is exactly the same as previously, they find $1/z'=0.588$ (they do not give any error estimate in this case). In the following, we shall see that we believe the first result to be closer to the exact one. In the next section, we shall see that their result in $d=2,D=3$ strongly violates exact inequalities, and thus is wrong.

The conclusion of this section is that no complete study of the value of τ had been performed until now because of a lack of appropriate numerical tools. More precise analytical results would also certainly be welcome to guide numerical works. We see that simulating or solving for the time evolution of the distribution function may not enable us to reach the asymptotic scaling regime, and a guideline of the present work will be to directly rely on the scaling equation corresponding to the infinite time asymptotic state itself.

III. EXACT BOUNDS

In the next three sections, our workhorses will be both Eqs. (2.7) and (2.8).

We first show that $\tau\geqslant 1$, for $d\geqslant 1$. Suppose $\tau<1$ and consider Eq. (2.8) with $\alpha=0$,

$$2\int_0^{+\infty} f(x)dx=\int\int_0^{+\infty} f(x)f(y)(x^{1/D}+y^{1/D})^d dxdy. \tag{3.1}$$

For $d\geqslant 1$, we have $(x^{1/D}+y^{1/D})^d\geqslant x^{d/D}+y^{d/D}$, which leads to $\int f(x)dx\geqslant\int f(x)dx\int f(x)x^{d/D}dx$ (in the bulk of the text, all integrals should be understood from 0 to ∞). Comparing with Eq. (2.7), this leads to $1\geqslant 2-\tau$ or $\tau\geqslant 1$, which is contradictory. Notice that Eq. (2.8) with $\alpha=2$ for $d=1$ and $D=1$ leads to $\int x^2 f(x)dx=2[\int x^2 f(x)dx][\int xf(x)dx]$, and we recover the exact result $\tau=2-\int xf(x)dx=3/2$ [27] in a very simple way. These results were already obtained by van Dongen and Ernst [28,30], who were able to find in the case $D=1$ the exact inequality, $2d<\tau<2-2^{1-d}(1-d)/(2-2^d)$, which shows that $\tau=2d+O(d^2)$ when $d\to 0$. This interesting result will be generalized to any D in the next section and the $O(d^2)$ term will be computed in $D=1$. They also found weaker inequalities in $d=1$, but no result was obtained for general d and D.

In order to deal with the general case, we introduce an extremely simple method to get lower and upper bounds for τ. We rely on Eq. (2.8) valid for $\alpha>\tau-1$. Combining Eqs. (2.7) and (2.8), we get

$$\tau=2-(1-\alpha)\frac{\int\int_0^{\infty} g(x,y)dxdy}{\int\int_0^{\infty} g(x,y)A(x/y)dxdy}, \tag{3.2}$$

where $A(u)=[1+u^\alpha-(1+u)^\alpha](1+u^{1/D})^d/(u^\alpha+u^{d/D})$ satisfies $A(u)=A(1/u)$ and $g(x,y)=(x^\alpha y^{d/D}+x^{d/D}y^\alpha)f(x)f(y)$. The ratio in Eq. (3.2) can then be interpreted as the inverse of a kind of *average* of $A(x/y)$ with the weight $g(x,y)$. For a given $\alpha\leqslant d/D$, we numerically determine the maximum M_α and minimum m_α of the function $A(u)$. Using Eq. (3.2), this gives

$$2-(1-\alpha)/m_\alpha\leqslant\tau\leqslant 2-(1-\alpha)/M_\alpha. \tag{3.3}$$

We then choose the best values of $\alpha\leqslant d/D$ compatible with $\alpha>\tau-1$ leading to the tightest bounds. More precisely, we proceed the following way: we start with $\alpha=d/D$ (as $\tau<1+d/D$), from which we obtain some upper and lower bound τ_m and τ_M. If $\tau_M<1+d/D$, we can repeat the operation with $\tau_M-1<\alpha<d/D$ which leads to new τ_m and τ_M, otherwise we cannot improve the trivial upper bound $1+d/D$. If the new τ_M is bigger than $\alpha+1$, we must reject this attempt, and keep the old values of both the upper and lower bound, but if it is smaller, then we can repeat the process and keep on this way until we obtain the tightest bounds.

For a general $\mu=0$ kernel $K(x,y)$, this method can be straightforwardly extended, with $A(u)=[1+u^{\alpha}-(1+u)^{\alpha}]$ $K(1,u)/(u^{\alpha}+u^{\lambda})$ and $g(x,y)=(x^{\alpha}y^{\lambda}+x^{\lambda}y^{\alpha})f(x)f(y)$.

A superficial plot of the function $A(u)$ for K_D^d may lead to the incorrect conclusion that its minimum is always obtained at $u=0$ with $A(0)=1$. In fact a more careful study of A shows that for certain values of α, the actual minimum is at $u>0$ but very close to 0. For $u\to 0$, $A(u)\sim 1+du^{1/D}-u^{d/D-\alpha}$, and we see that if $\alpha>(d-1)/D$, there is a local minimum for $u_m>0$ with $A(u_m)<1$. For $d>1$, and $\alpha=(d-1)/D+\varepsilon$, we get $u_m\sim\exp[-\ln(d)/\varepsilon]$, which vanishes exponentially when $\varepsilon\to 0$ $(d>1)$. Indeed, even when α is not so close to $(d-1)/D$, u_m may be very small. For instance, for $d=2,D=3$, and $\alpha=0.585\,98>(d-1)/D=0.333\ldots$, we find that $u_m=1.365\times 10^{-4}$, and $A(u_m)=0.7322$, which leads to a nontrivial lower bound of 1.4349 for τ.

Actually, it is easily seen that the inequalities obtained by van Dongen and Ernst (in the case $d=1$ or $D=1$) correspond to $\alpha=d/D$. In fact, even in this case, M_α and m_α are nontrivial, and they used some *explicit* bounds of M_α and m_α, which do not lead to the tightest bounds for τ.

Thus our method consists in computing the *actual* value of m_α and M_α, and varying α to optimize these bounds, which allows us to *greatly improve* van Dongen and Ernst's explicit inequalities for $D=1$ or $d=1$, and to obtain new exact bounds for $d>1$. For instance, for the physically interesting cases (see below) $(d=1,D=2)$, $(d=1,D=4)$, and $(d=2,D=4)$ we, respectively, found $1.084\leqslant\tau\leqslant 1.147$, $1\leqslant\tau\leqslant 1.075$ (compared to $1\leqslant\tau\leqslant 1.28$ and $1\leqslant\tau\leqslant 1.109$ in [28]) and $1.25\leqslant\tau\leqslant 1.5$.

For $d=2,D=3$, we find $1.4349\leqslant\tau\leqslant 1.585$, which just discards the value $\tau=1.244$ found by Song and Poland [21], and strongly questions the validity of their approach. The exact bounds we obtained in $d=1,D=2$ are violated by their alternative value 1.150 for τ but not by their first result 1.123 (see Sec. II C).

It is useful to note that for any D, with $\alpha=d/D$, $A(u)\to 1/2$ when $d\to 0$, which entails that $\tau\to 0$ [from Eq. (3.3)] in this limit.

To conclude with this topic of inequalities, let us consider Eq. (3.3) with $\alpha=d/D$. In this case, when $D\to\infty$,

$$A(u)=\frac{1}{2}(1+u^{-1/D})^{d}[1+u^{d/D}-(1+u)^{d/D}]$$
$$\to\begin{cases}2^{d-1}, & 0<u\leqslant 1\\ \frac{1}{2}, & u=0\end{cases}\tag{3.4}$$

hence $m_\alpha\to 1/2$ and $M_\alpha\to 2^{d-1}$. Therefore the upper bound for τ in Eq. (3.3) tends to $2-2^{1-d}$. This is strictly less than 1 for $d<1$, which means that for any $d<1$, there exists a finite critical $D_c(d)$, such that $\tau<1$ for any $D>D_c$. This result will be used in Sec. IV.

IV. PERTURBATIVE AND NONPERTURBATIVE EXPANSIONS

In this section we use the exactly solvable limits $d=0$ and $D=\infty$ as a basis for a perturbative expansion. We also consider the case $d\to\infty$, keeping $d/D=\lambda$ constant, for which we find a nonperturbative expansion.

We saw that $\lim_{d\to 0}\tau=0$. What about the $D\to\infty$ limit of τ? In fact, although strictly at $D=\infty$, τ is equal to 0, as $f(x)=2^{1-d}e^{-x}$, we will see that $\tau_\infty=\lim_{D\to\infty}\tau>0$. This result was already noticed by van Dongen and Ernst in $d=1$ [28]. Since $\tau<1+d/D$ we get that

$$\tau_\infty\leqslant 1.\tag{4.1}$$

What can we learn from Eq. (2.7) in the large D limit? We see that the limit for τ is

$$\tau_\infty=2-\int_0^{+\infty}f_\infty(x)dx=2-2^{1-d}\tag{4.2}$$

provided that

$$\lim_{D\to\infty}\int_0^{+\infty}[f_D(x)-f_\infty(x)]x^{d/D}dx=0.\tag{4.3}$$

For $d<1$, this result is consistent, since, from the last remark of Sec. III, we get $\tau_\infty\leqslant 2-2^{1-d}<1$.

However, for $d\geqslant 1$ we know that $\tau\geqslant 1$, hence $\tau_\infty=1$, which means that for $d>1$,

$$\lim_{D\to\infty}\int_0^{+\infty}[f_D(x)-f_\infty(x)]x^{d/D}dx=1-2^{1-d}>0\tag{4.4}$$

while in $d=1$, Eq. (4.3) is true.

Now that we know the large D limit of τ ($\tau_\infty=1$ for $d>1$ and $\tau_\infty=2-2^{1-d}$ for $d\leqslant 1$), as well as its small d limit ($\tau\to 0$), let us compute the corresponding asymptotic corrections.

A. Small *d* expansion

First, consider the limit $d\to 0$. We expand f in series in d: $f(x)=f_0(x)+df_1(x)+O(d^2)$, $f_0(x)=e^{-x}$. A systematic way of expanding τ would be to write down a linear (self-consistent) differential equation for f_1 to solve it and plug the result into Eq. (2.7).

However, as far as the first order is concerned we can get it without solving for f_1. By expanding the integral expression of τ, Eq. (2.7), we get

$$\tau=2-\int_0^{+\infty}f(x)x^{d/D}dx=-\frac{d}{D}\int_0^{+\infty}f_0(x)\ln x\,dx$$
$$-d\int_0^{+\infty}f_1(x)dx+O(d^2).\tag{4.5}$$

Then we expand both sides of Eq. (3.1) to get an equation for $\int f_1(x)dx$:

$$\int_0^{+\infty} f_1(x)dx=\frac{1}{2}\int\int_0^{+\infty} f_0(x)f_0(y)\ln(x^{1/D}+y^{1/D})dxdy-\int_0^{+\infty} f_0(x)dx\int_0^{+\infty} f_1(x)dx \tag{4.6}$$

hence $\int f_1(x)dx=-\int\int e^{-x-y}\ln(x^{1/D}+y^{1/D})dxdy$. After eliminating $\int f_1(x)dx$, we get

$$\tau=2dJ_D+O(d^2),$$

$$J_D=\int_0^1 \ln\left[1+\left(\frac{1-u}{u}\right)^{1/D}\right]du. \tag{4.7}$$

Let us mention that this result can be systematically generalized to the case of any homogeneous kernel of the form $[g(x,y)]^d$, leading to $\tau=2d\int_0^1 \ln g(1,(1-u)/u)du+O(d^2)$.

Although it may seem a bit tedious, it is interesting to recover this result in another way, as it shows that the small x behavior of f_1 is consistent with the $d\to 0$ expansion of the power law $x^{-\tau}=1-2dJ_D\ln x+O(d^2)$. Let us write down the linear equation for f_1,

$$xf_1'(x)+2e^{-x}\int_0^x f_1(y)e^y dy=2e^{-x}\int_0^{+\infty} f_1(y)dy+4e^{-x}\int_0^{+\infty} e^{-y}\ln(y^{1/D}+x^{1/D})dy-2e^{-x}\int_0^x \ln[y^{1/D}+(x-y)^{1/D}]dy. \tag{4.8}$$

With $u=e^x f_1$ we get the following equation:

$$x(u'-u)+2\int_0^x u(y)dy=2\int_0^{+\infty} u(y)e^{-y}dy+4\int_0^{+\infty} e^{-y}\ln(y^{1/D}+x^{1/D})dy-2xJ_D-\frac{2}{D}(x\ln x-x), \tag{4.9}$$

which implies, after taking the derivative of Eq. (4.9),

$$xu''+(1-x)u'+u=-\frac{2}{D}\ln x-2J_D+\frac{4}{D}\int_0^{+\infty} e^{-y}\frac{x^{1/D-1}}{y^{1/D}+x^{1/D}}dy. \tag{4.10}$$

The solution u of Eq. (4.10) involves two integration constants, one being fixed by the fact that f_1 should go to zero at large x, the other, c_0, by writing the compatibility with Eq. (4.9), which can be done by taking the $x\to 0$ limit the latter equation. From the expression of the solution (Appendix C), or directly from Eq. (4.10), it is easily seen that u has the asymptotic expansion for $x\to 0$:

$$u(x)=b_0\ln x+O(1), \tag{4.11}$$

with $b_0=c_0-2/D$.

We know that $f(x)\sim cx^{-\tau}$ when $x\to 0$. When $d\to 0$, $c\to 2$, and $\tau=d\tau_1+O(d^2)$, hence up to order d we expect

$$f(x)\sim 2\tau_1\ln x \tag{4.12}$$

so that we interpret b_0 as $-2\tau_1$,

$$\tau=-d\frac{b_0}{2}+O(d^2). \tag{4.13}$$

The $x\to 0$ limit of Eq. (4.9) is

$$b_0=2\int_0^{+\infty} f_1(x)dx-\frac{4}{D}\int_0^{+\infty} e^{-x}\ln x dx. \tag{4.14}$$

The integration of Eq. (4.10) between 0 and $+\infty$ yields

$$-b_0+\int_0^{+\infty} f_1(x)dx=2J_D-\frac{2}{D}\int_0^{+\infty} e^{-x}\ln x dx. \tag{4.15}$$

The combination of Eqs. (4.14) and (4.15) yields b_0, which, substituted into Eq. (4.13), eventually leads to the same result for τ as previously obtained through the expansion of Eq. (3.1) and Eq. (2.7).

For $D=1$, we get $\tau=2d+O(d^2)$, in good agreement with direct numerical integration of Smoluchowski's equation performed by Krivitsky [20] and shown in Fig. 1 (see below). This result for $D=1$ also coincides up to order $O(d)$ with the inequalities for τ that we obtained above, as noticed in Sec. III. This is not the case for other values of D.

The order $O(d^2)$ requires the computation of f_1. However, in the special case $D=1$ it is possible to obtain explicitly the $O(d^2)$ term by expanding Eq. (2.8) for $\alpha=d/D$ (see Appendix B). We obtain

$$\tau=2d+\left(\frac{\pi^2}{3}-4\right)d^2+O(d^3). \tag{4.16}$$

In Sec. V (see Fig. 1) we shall see that this result is in excellent agreement with both Krivitsky's results and a method of approximation that we shall introduce in Sec. V.

B. Large D expansion

Now, we perform an expansion in powers of $1/D$ for $d\leqslant 1$, expanding $f(x)=f_\infty(x)+(1/D)f_1(x)+(1/D^2)f_2(x)+O(1/D^3)$.

Perturbative expansion in $d<1$. In $d<1$, as mentioned in Sec. III, $\tau<1$ for any D above a finite critical $D_c(d)$. As a consequence, Eq. (2.8) can be written for any $D>D_c(d)$. Therefore we can expand this equation for large D in powers of $1/D$, and we find at first order,

$$\int_0^{+\infty} f_1(x)dx = d\,2^{d-2}\int\int_0^{+\infty} f_\infty(x)f_\infty(y)(\ln x+\ln y)dxdy + 2^d\int_0^{+\infty} f_\infty(x)dx\int_0^{+\infty} f_1(x)dx \quad (4.17)$$

hence

$$\int_0^{+\infty} f_1(x)dx = -d\int_0^{+\infty} f_\infty(x)\ln(x)dx = 2^{1-d}d\gamma, \quad (4.18)$$

where γ is Euler's constant, while from Eq. (2.7),

$$\tau=\tau_\infty-\frac{d}{D}\int_0^{+\infty} f_\infty(x)\ln(x)dx-\frac{1}{D}\int_0^{+\infty} f_1(x)dx+O\left(\frac{1}{D^2}\right). \quad (4.19)$$

We conclude, using Eq. (4.18), that the first order correction to τ_∞ is zero.

The same method also gives access to the next term:

$$\tau=\tau_\infty-\frac{d^2}{2D^2}\int_0^{+\infty} f_\infty(x)(\ln x)^2dx-\frac{d}{D^2}\int_0^{+\infty} f_1(x)\ln(x)dx - \frac{1}{D^2}\int_0^{+\infty} f_2(x)dx+O\left(\frac{1}{D^3}\right) \quad (4.20)$$

while

$$\int_0^{+\infty} f_2(x)dx=\frac{1}{2}\int\int_0^{+\infty} f_\infty(x)f_\infty(y)\frac{2^d}{8}\{(d+1)[(\ln x)^2+(\ln y)^2]+2(d-1)\ln(x)\ln(y)\}dxdy + 2^{d-1}d\int\int_0^{+\infty} f_1(x)f_1(y)(\ln x+\ln y)dxdy+2^{d-1}\left(\int_0^{+\infty} f_1(x)dx\right)^2+2\int_0^{+\infty} f_2(x)dx. \quad (4.21)$$

Using the known value of $\int f_1$ we get

$$-\int_0^{+\infty} f_2(x)dx=\frac{d^2}{4}\int_0^{+\infty} f_\infty(x)(\ln x)^2dx + d\int_0^{+\infty} f_1(x)\ln(x)dx + \frac{2^{1-d}d}{4}\left(\frac{\pi^2}{6}+d\gamma^2\right) \quad (4.22)$$

(γ being Euler's constant), which leads to

$$\tau=2-2^{1-d}+\frac{\pi^2 2^{-d}d(1-d)}{12D^2}+O\left(\frac{1}{D^3}\right). \quad (4.23)$$

Once again we were able to obtain a highly nontrivial expansion for τ without solving for f_1 and f_2 themselves, although this can also be achieved this way. Note that in the limit of large D *and* small d, Eqs. (4.7) and (4.23) coincide up to order $O(d/D^2)$.

Perturbative estimate for $d>1$. In the case $d\geq1$, we have shown that $\tau\geq1$ and since $\tau<1+d/D$, we see that $\tau\to1$ for $D\to\infty$ and finite $d\geq1$. As f_1 is nonintegrable, Eq. (2.8) cannot be used with $\alpha=0$, and the previous perturbation breaks down.

Nevertheless we can try to obtain an estimate of τ in the following way: we make the ansatz $f\sim f_\infty+c/s^{1+\varepsilon}e^{-s}$. We plug it into Eq. (2.7) and Eq. (2.8) for $\alpha=d/D$, and after some algebra (see Appendix D) we see that for consistency ε must be of order $1/D$ and that $c=(1-2^{1-d})(d/D-\varepsilon)$, and eventually that $\varepsilon=\kappa/D+O(1/D^2)$ where κ is the solution of the nonlinear equation:

$$\frac{2}{1+2^{1-d}}=\int_0^1(1+v^{1/(d-\kappa)})^d dv. \quad (4.24)$$

This equation always has a solution consistent with the exact bound $1<\tau<1+d/D$. For instance, in the case $d=2$, $D=4$ we obtain $\tau\approx1.462$. Though it is still of order $1/D$, the obtained perturbative estimate depends on the choice of α. $\alpha=d/D$ seems, however, to be the most natural choice.

In $d=1$, c vanishes and we do not learn much. All terms of the $d<1$ series for τ in powers of $1/D$ vanish for $d\to1$, as can be seen in Eq. (4.23) for the two leading ones. The reason is the following: the perturbation is derived from Eq. (3.1) under the assumption that $\tau<1$. In $d=1$, such an assumption yields $2\int f(x)dx=2[\int x^{1/D}f(x)dx][\int f(x)dx]$ hence $\tau=1$. Consequently the perturbative value of τ tends to 1 when $d\to1^-$. As will be illustrated below by numerical results, for a given $d>1$ the critical $D=D_c(d)$ above which $\tau<1$ tends to infinity when $d\to1^-$, entailing the vanishing of the perturbation validity domain in D. Thus the correction to $\tau=1$ for large D may be *nonperturbative* in $d=1$.

If we now take the $d\to\infty$ limit in Eq. (4.24), we obtain $\tau\simeq1+\lambda-2^{-d}\lambda$ ($\lambda=d/D$), a nonperturbative behavior in d which is to be related to the results below, obtained for $d\to\infty$, $D\to\infty$, keeping λ constant.

C. Large *d* and *D*

We now present a nonperturbative calculation in the limit of large d *and* D, keeping the ratio $\lambda=d/D$ fixed. In this limit, the kernel can be written

$$(x^{1/D}+y^{1/D})^d=2^d(xy)^{\lambda/2}[1+O(d/D^2)] \quad (4.25)$$

and surprisingly transforms into the well-studied ''product'' kernel [2,18,30,19–21,29]. Assuming scaling (a still controversial subject [20]), one can easily show that $\tau=1+\lambda=1+d/D$ [18] [see also Eqs. (1.3) and (1.4) and the discussion below them, as it corresponds to $\mu=\lambda/2>0$].

We can show that including higher order corrections in power of $1/D$ does not change the value of τ, so that the correction to $\tau=1+\lambda$ is certainly nonperturbative. Consider the expansion of the kernel:

$$K(x,y)=2^d(xy)^{\lambda/2}[1+2^{-d}O(1/d^2)]. \quad (4.26)$$

The rescaled function $\tilde{f}=2^d f$ is the solution of the scaling form of Smoluchowski's equation with the kernel $\tilde{K}=2^{-d}K(x,y)$, which is equal to $(xy)^{\lambda/2}$ *at every order in* $1/d=1/(\lambda D)$. In fact, we can estimate this correction by assuming that for finite d and D,

$$\tilde{f}(s)\sim c_\lambda/s^{1+\lambda-\varepsilon_d} \quad (4.27)$$

for $s\to 0$. Plugging this estimate into Eq. (2.7) with the limit kernel of Eq. (4.25), we first get

$$\varepsilon_d\approx 2^{-d}\frac{c_\lambda}{(1-\lambda)}. \quad (4.28)$$

c_λ can be determined by matching the coefficients of the leading terms in Eq. (2.5) using the kernel of Eq. (4.25). After a straightforward calculation, one gets in the $d\to\infty$ limit,

$$c_\lambda=2(1-\lambda)I_\lambda^{-1}, \quad (4.29)$$

$$I_\lambda=\int_0^1[u(1-u)]^{-1-\lambda/2}[u^\lambda+(1-u)^\lambda-1]du, \quad (4.30)$$

which leads to

$$\tau=1+\lambda-2^{1-d}I_\lambda^{-1}. \quad (4.31)$$

We thus find a nonperturbative (exponentially small) correction to τ in the large d and large D limit, consistent with the result obtained above for $d>1$ and large D. Note that Eq. (4.29) is also consistent with the exact result that $\tau\to 1$ as $D\to\infty$ for finite $d>1$, a result that we obtain by setting $\lambda=0$ (as I_λ diverges).

D. Summary of the results

We have shown that when $D\to\infty$, $\tau\to 1$ for $d\geqslant 1$, whereas $\tau\to 2-2^{1-d}<1$ for $d<1$. We were able to derive an $O(1/D^2)$ perturbative expansion in $d<1$, and we convinced ourselves that the leading corrective term in $d>1$ was of order $1/D$, by giving an estimate of this correction. In $d=1$ both approaches break down and the large D corrections to $\tau_\infty=1$ are probably nonperturbative.

When $d\to 0$, τ goes to zero, and we gave a first order perturbative expression in d, for any D. For $D=1$, we also found the explicit coefficient in d^2.

Eventually, we showed that for a fixed homogeneity $\lambda=d/D$, τ tends exponentially to $1+\lambda$ at large d. In the following section we present a general numerical method to compute τ and we confirm our analytical result by performing an extensive study of the function $\tau(d,D)$.

V. VARIATIONAL APPROACH

In this section we present a practical way of obtaining good approximate values for τ, without explicitly solving Smoluchowski's equation. Once again, we rely on Eq. (2.8), which holds for the exact scaling function [solution of Eq. (2.5)], for any $\alpha>\tau-1$. This equation is *general*, and does not depend on the specific kernel we study in this article. As a consequence, the methods we develop are general and do apply to *any homogeneous kernel*. We emphasize the fact that this method does not intend to approach the whole scaling function, but sets the focus on the computation of τ [in fact, numerically solving the scaling equation (2.5) for the scaling function seems to be very difficult, and at least as difficult as directly solving the time-dependent equation [31]].

A. Principles of the method

The simplest way of approximating τ is to evaluate the ''average'' in Eq. (3.2) using a reasonable trial weight function $g(x,y)$ instead of the unknown exact one. As a simple start, we will expose a crude, but straightforward algorithm, that illustrates the basic idea. Then we will develop the variational method itself, which is not much more intricate, but much more effective.

A one-parameter choice for a trial weight function is obtained by replacing in the above expression of $g(x,y)$ the exact $f(x)$ by $f_\tau(x)=x^{-\tau}\exp(-x)$, which has the correct leading asymptotics for small x (by definition of τ) and decays exponentially at large x, although not with the exact asymptotics $x^{-d/D}e^{-x}$ $(d/D<1)$ [16]. Still, this functional form is known to be a good approximation of the actual $f(x)$ obtained in simulations [20], and is even the exact solution, but for a multiplicative constant, in the $d=D=1$ case, which belongs to the special class $\lambda=1$ [27]. The first idea that comes to mind is just to determine τ self-consistently such that Eq. (3.2) holds for f_τ, with a specific choice of α, for instance, $\alpha=d/D$. This is readily done, by an iterative method: starting from an initial τ_0, verifying previously obtained exact bounds, we construct the sequence

$$\tau_{n+1}=(1-\varepsilon)+\varepsilon[2-(1-\alpha)R_\alpha(f_{\tau_n})], \quad (5.1)$$

with

$$R_\alpha(\phi)=2\frac{\int\int_0^{+\infty}x^\alpha\phi(x)y^{d/D}\phi(y)dxdy}{\int\int_0^{+\infty}\phi(x)\phi(y)K(x,y)[(x+y)^\alpha-x^\alpha-y^\alpha]}, \quad (5.2)$$

which converges, with a proper choice of $1>\varepsilon>0$, to a fixed point corresponding to an f_τ verifying Eq. (3.2). The numerical evaluation of $R(\tau)$ can be achieved with utter celerity and arbitrary precision, since it reduces to the calculation of one-dimensional integrals, and of a few values of the Γ function, thanks to a very convenient transformation (see Appendix A). We notice that it is unnecessary to include any multiplicative constant into f_τ, since it would just cancel out in Eq. (3.2).

Of course, this algorithm should yield different values of τ for different choices of α, except in the special case when the exact solution is of the form f_τ. This corresponds to $d=0$, $D=\infty$ and $d=1,D=1$, and this method converges by construction, to the exact value of τ, but for the round-off errors. In the generic case, the variation can be non-negligible (in $d=2,D=4$, $\tau\approx 1.371$ for $\alpha=d/D$, while $\tau\approx 1.398$ for $\alpha=0.403$) and the fixed point τ may even violate exact bounds. For instance, in the case $d=1,D=3$ with $\alpha=d/D$ we get $\tau=0.9894$ whereas we know that $\tau>1$. The variation with α makes the method unreliable. In $d=2,D=4$, it gives $\tau\approx 1.385\pm 0.015$, compared to $\tau\approx 1.434\pm 0.004$ with the variational approximation, that we now introduce, which, starting from the same basic idea, proves to be much more effective.

Variational approximation. A much better and hardly more intricate method is to choose a reasonable sample of values of α, and minimize an error function measuring the violation of the corresponding Eqs. (3.2). This method can be systematically improved by allowing for n free ''fitting'' parameters (including τ itself) in the trial weight $g(x,y)$. In the following we will proceed by replacing the exact f by a variational function of the form

$$f_v(x,\tau_0,\tau_1,\ldots,\tau_n,c_1,\ldots,c_i)=x^{-\tau_0}e^{-x}+\sum_{j=1}^{n} c_j x^{-\tau_j}e^{-x} \tag{5.3}$$

and we will minimize the error function,

$$\chi^2(f_v)=\sum_i\,[\tau_0-2+(1-\alpha_i)R_{\alpha_i}(f_v)]^2, \tag{5.4}$$

to get a variational approximation $\tau_v=\tau_0$ of τ. Brute force should not be used in the evaluation of χ^2: once again, Eq. (A1) makes it possible to drastically reduce the computation time, and to perform the evaluation of χ^2 with an excellent precision.

Of course, the values of the exponents in f_v should not be blindly chosen. van Dongen and Ernst [30] showed that the subleading term in the small x asymptotic expansion of f is

$$\propto\begin{cases} x^{1+\lambda-2\tau} & \text{if} \quad \tau>1+\lambda-\mu_1 \\ x^{\mu_1-\tau} & \text{if} \quad \tau<1+\lambda-\mu_1 \\ x^{-\tau}\ln x & \text{if} \quad \tau=1+\lambda-\mu_1, \end{cases} \tag{5.5}$$

with $K(x,y)-x^\lambda\propto y^{\mu_1}x^{\lambda-\mu_1}$ when $x\to\infty$, whereas the exact asymptotic at large x is $\propto x^{-\lambda}e^{-x}$. Therefore a good three-parameter class of trial functions should be

$$f_v(x,\tau_0,c_1,c_2)=\left(\frac{1}{x^{\tau_0}}+\frac{c_1}{x^{\tau_1(\tau_0)}}+\frac{c_2}{x^{\lambda}}\right)e^{-x}, \tag{5.6}$$

τ_1 being either $2\tau_0-1-\lambda$ (if $\tau_0>1+\lambda-\mu_1$), or $\tau_0-\mu_1$ (if $\tau_0<1+\lambda-\mu_1$). The small x leading term in f_v is τ_0 provided that $\tau_0>\lambda$. The approximate value τ_v is the value of τ_0 at the minimum.

By construction, this method reproduces the exact results for the constant kernel and $d=1,D=1$, since the exact scaling function is contained in those cases in the class of variational function we chose. In general, this method is inadequate to approach f itself, and is just designed to compute τ, in the same way as the variational approach in quantum mechanics is designed to obtain eigenvalues but, in principle, not eigenfunctions.

B. Implementation

With a small number n of variational parameters, we choose to perform the minimization with the downhill simplex method described in [32] (steepest descent, conjugate gradient, or other methods could also be used, with the drawback that these methods require extra evaluations of χ^2 to compute its gradient). This method starts from an n-dimensional simplex, i.e., $n+1$ points in the n-dimensional parameter space, and performs a sequence of geometric deformations until it contracts to a local minimum of the function. It is not the fastest algorithm, but it easily converges, and in our case where the computational burden is low we do not need more sophisticated devices.

As in any optimization problem, the initial condition is a crucial parameter, but here there is the additional complication that the smallest moment $\alpha_{\min}$ used in the computation of χ^2 should be bigger than $\tau-1$, and bigger than τ_0-1 at any step of the algorithm. What information on the value of τ we may *a priori* gather (exact bounds, perturbation expansion) should guide our choice. Anyway, we do know that $\tau<1+\lambda$: starting with an initial τ_0 smaller than $1+\lambda$ and $\alpha_{\min}>\lambda$ should avoid any trouble. As we get a first approximation of τ we will be able to decrease the value of $\alpha_{\min}$ and make it closer to τ_v-1, while refining the initial conditions. A few Monte Carlo minimization steps can also be used to find a proper initial condition (but we scarcely needed this functionality in this work).

Why should we choose as small an $\alpha_{\min}$ as possible? The answer is that small moments probe the small x divergence of $f(x)$, which is precisely what we are interested in. However, we also need some intermediate and higher moments to probe the intermediate x and the large x decay to stabilize consistent values of c_1 and c_2. There should be at least as many moments as variational parameter, otherwise there would be an infinite number of minima. Too many moments would cause excessive numerical round-off errors in the computation of χ^2.

We tested round-off errors by computing τ_v for the exactly solvable model K_1^1 for which $f(x)\propto x^{-3/2}e^{-x}$, since, were we endowed with infinite numerical precision, our algorithm would yield the exact result in this case, as said before, whatever the α_i may be, provided that they all are bigger than $1/2=\tau-1$.

With the three-parameter function introduced above, and moments 0.55, 0.667, 0.783, 0.9, and 2, we find $\tau=1.499\,97\pm 4\times 10^{-6}$ ($\chi^2=1.94\times 10^{-8}$), the uncertainty being due to variations with different choices for the initial

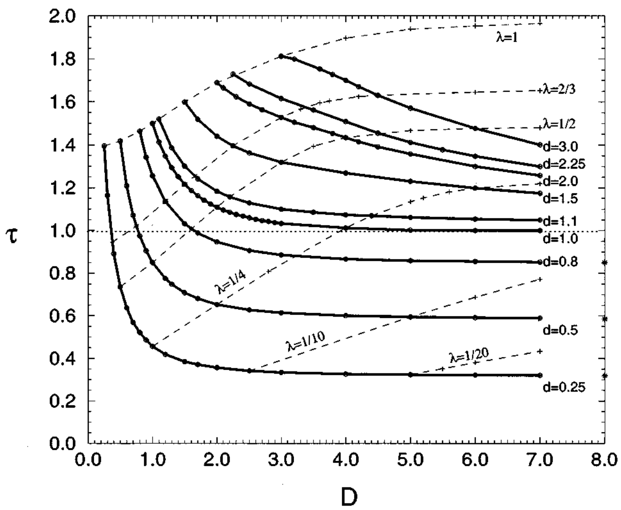

FIG. 2. The exponent τ was computed by the variational method for various values of d and D. We show here some iso-d (solid lines) and iso-λ (dashed) ($\lambda = d/D$) lines. The iso-d lines tend to $\tau = 2 - 2^{1-d}$ (stars on the right axis) if $d < 1$, and to 1 if $d \geqslant 1$. The critical D above which τ becomes smaller than 1 tends to infinity when $d \to 1^-$, entailing the breakdown of the large D perturbative expansion in $D \geqslant 1$. The $d = 1$ iso-d line seems to tend exponentially to 1, while for $d > 1$ the relaxation to 1 is slower. An inflection point appears above $d \approx 2$. The iso-λ lines exponentially saturate to $1 + \lambda$ at large D.

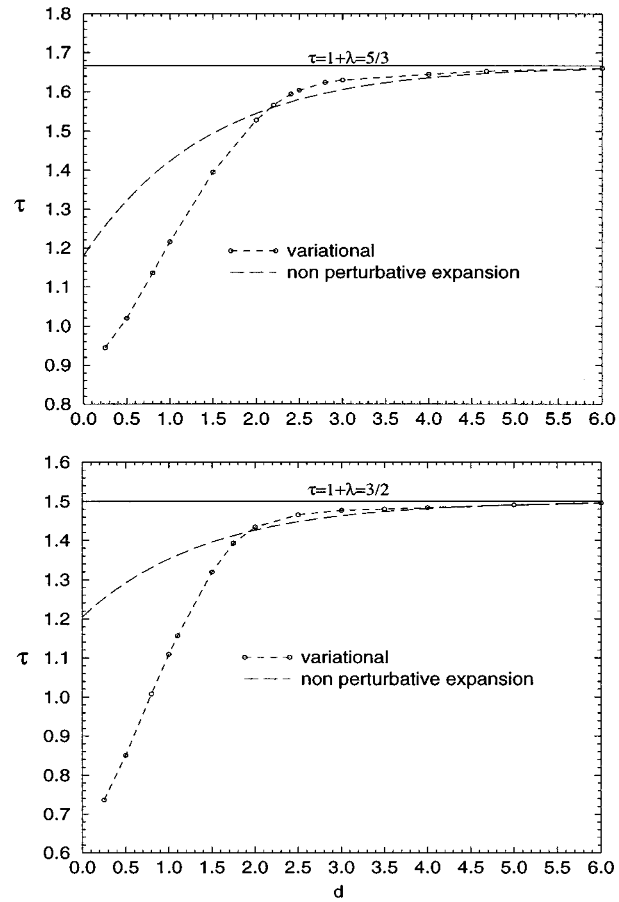

FIG. 3. Iso-λ curves computed by the variational method (solid lines), as a function of d, for $\lambda = 1/2$ and $\lambda = 2/3$. As analytically established, τ tends to $1 + \lambda$ at large D. The agreement is good at large d with the nonperturbative expansion (dashed lines).

values of the parameters and the tolerance on the size of the simplex (the minimization algorithm stop criterion). The round-off errors increase with the number of moments and the number of variational parameters. The error is much bigger on c_1 and c_2, we find $c_1 = 0.11 \pm 0.1$ and $c_2 = -0.12 \pm 0.1$, instead of strictly 0. This means that the sensitivity on c_1 and c_2 is small in the vicinity of the minimum, and this method is not the right one to determine the scaling function (a negative c_2 is unphysical here), but it just was not devised for this purpose: we just meant to compute τ, and for this quantity the accuracy is excellent.

C. Numerical results

We used this method to determine approximations of τ for the kernel $(x^{1/D} + y^{1/D})^d$. We compared our results to numerical values obtained for $d \leqslant 1$, $D = 1$ by Krivitsky [20], and to our perturbative and nonperturbative expansions.

All values were obtained from the three-parameter variational functions introduced earlier in this text. We used eight moments, six in the interval $[\alpha_{\min}, 0.9]$, plus $\alpha = 2$ and $\alpha = 3$. $\alpha_{\min}$ was adjusted to be as close to $\tau_v - 1$ as possible. The computation time was from 1 to 10 seconds per run on a HP workstation. Two to five runs per point were necessary to adjust the parameters.

We also computed a few points with a different repartition of moments: five in the range $[\tau - 1, d/D]$, $\alpha = 0.9$, 2, 3, as well as with only two variational parameters ($c_1 = 0$), and with four variational parameters [the additional exponent being $\mu_1 - \tau$ in the case when $\tau > 1 + (d-1)/D$]. The observed *relative* variations of τ_v were at most of a few 10^{-3}. In all cases, τ was found to be consistent with exact bounds.

First, we consider the case $D = 1$. Figure 1 shows the comparison between variational approximations of τ obtained with the modus operandi we just exposed, values extracted by Krivitsky [20] from a numerical solution of Smoluchowski's equation, and the $O(d^2)$ perturbative expansion. The agreement between the variational approximation and Krivitsky's results is excellent, which confirms the effectiveness and efficiency of the method: the ratio computation time (a few seconds)/accuracy is impressive. Actually, the variational approximation looks smoother than Krivitsky's curve, which has two visible accidents (small cusps) near $d = 1$ and $d = 0.4$, and the variational approximation is fully consistent with the exact $O(d^2)$ expansion at small d to which it clearly tends asymptotically, whereas Krivitsky's result tends to remain parallel to the perturbative curve, though close to it. Its good agreement with our infinite time results assesses the fact that Krivitsky's solution actually reached the scaling regime, which, as said in Sec. II, was not obvious *a priori*. We conclude that in this regime, the variational approximation recovers and confirms the results obtained by numerical integration of Smoluchowski's equation.

Once the effectiveness of the method was established, we were able to carry out a systematic study of $\tau(d,D)$, and to control its validity thanks to the analytical results obtained in Secs. III and IV.

We show in Fig. 2 the function $\tau(d,D)$ $(0.25 \leqslant d \leqslant 3,$

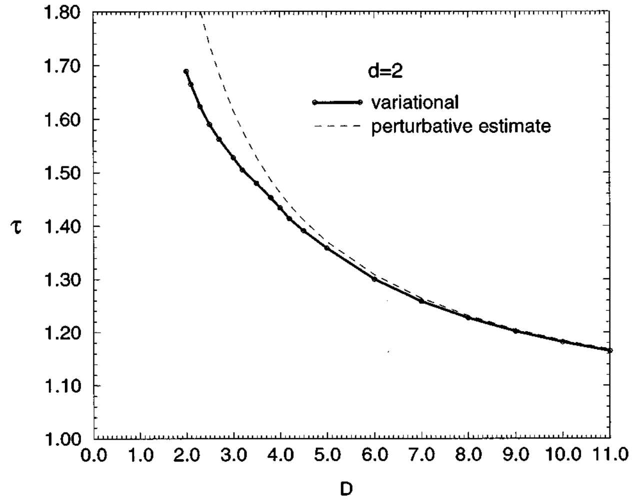

FIG. 4. In $d=2$, the exponents computed by the variational approximation are in good agreement with the perturbative large D estimate $\tau=1+1.849/D$. From data, the actual asymptotic correction seems to be closer to $1.82/D$. The cusp on the variational curve corresponds to the change of behavior with the occurrence of an inflection point for above $d=2$.

$d\leqslant D<8$) plotted in a (τ,D) diagram. Two kinds of curves are shown. Solid lines represent some iso-d lines, i.e., the function $\tau(D)$ for a fixed value of d, whereas dashed lines are iso-λ ($\lambda=d/D$) lines. The reliability of the approximation is assessed by the comparison with analytical results. As established in Sec. IV iso-d lines tend to $\tau=2-2^{1-d}$ (stars on the right axis of Fig. 2) if $d<1$, and to 1 if $d\geqslant1$. As expected, the critical D above which τ becomes smaller than 1 tends to infinity when $d\to1^-$, entailing the breakdown of the large D perturbative expansion in $D\geqslant1$. The $d=1$ iso-d line seems to tend exponentially to 1, which is consistent

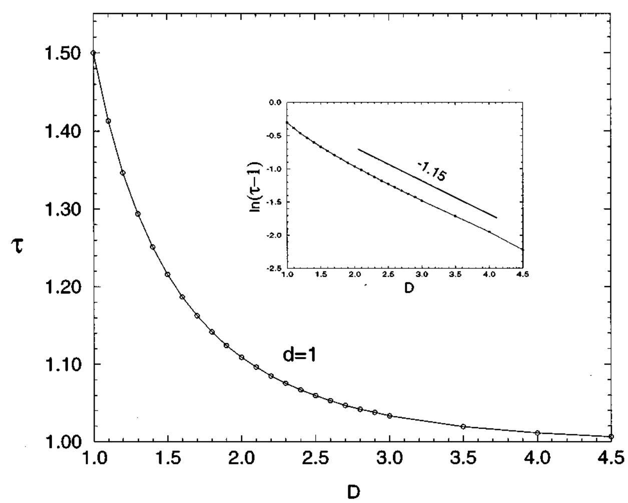

FIG. 5. For $d=1$, the exponents computed by the variational approximation display a much faster decay to their $D=\infty$ limit ($\tau_\infty=1$), than for $d>1$. Indeed, as shown in this figure, the decay seems to be exponential in D, with roughly $\tau-1\propto e^{-1.15D}$, a nonperturbative behavior to be related to the breakdown of the large D perturbative approaches for $d=1$.

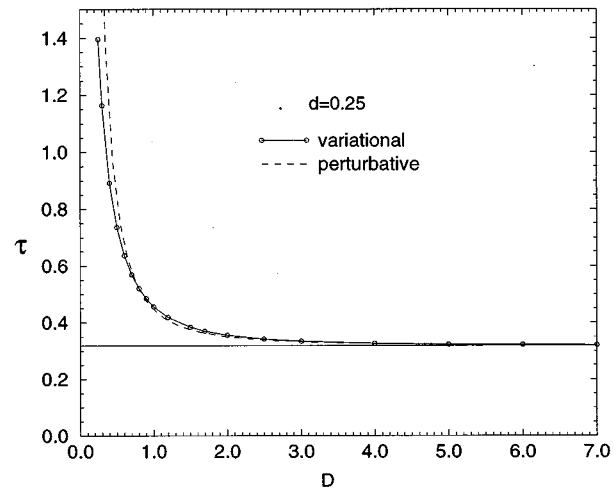

FIG. 6. In $d=0.25$, the exponents computed by the variational approximation are in good agreement with the perturbative large D estimate $\tau=2-2^{1-d}+\pi^2 2^{-d}d(1-d)/12D^2+O(1/D^3)$.

with a nonperturbative decay in $1/D$ (see below). For $d>1$ the large D decay is slower, as analytically predicted (we found a $1/D$ perturbative correction, see below). For $d\geqslant2$ the curves qualitatively shape changes and an inflection point appears.

Iso-λ lines exponentially saturate to $1+\lambda$ at large D, as analytically established before. Figure 3 shows the comparison between the variational approximation and the nonperturbative large d expansion of Eq. (4.31) in two cases, $\lambda=1/2$ and $\lambda=2/3$. The agreement is once again excellent at large d.

In $d=1,D=2$, Song and Poland [21] found $\tau=1.123\pm0.016$ (using their first result), which compares well with our $\tau=1.109$. In $d=2,D=3$, we find $\tau=1.528$ which, unlike their result (1.243), is perfectly consistent with the exact bounds $1.4349<\tau<1.585$. In $d=2,D=4$, we find $\tau=1.434$, which is in fair agreement with the perturbative large D estimate $\tau=1.462$ of Sec. IV. In fact, as shown in Fig. 4, the perturbative estimate is indeed a good approximation of τ in $d=2$ for $D\geqslant6$, and the $\propto1/D$ decay is confirmed by the variational results. The cusp on the variational curve is confirmed by the existence of an inflection point on $d>2$ curves, as mentioned above. In $d=1$, a nonperturbative exponential large D decay to $\tau_\infty=1$ is confirmed by Fig. 5. We roughly find $\tau-1\propto e^{-1.15D}$.

Eventually, we show in Fig. 6 (for $d=0.25$) that the variational result is also in good agreement with the large D second order perturbative expansion in $d<1$ ($\propto1/D^2$).

As this section draws to a close, we shall say that this variational method, although very simple, seems to be very well adapted to the determination of the exponent τ, as it is fast and, at least in the case we studied in this article, very accurate. It made it possible to acquire quantitative knowledge of τ in the whole parameter space of the K_D^d kernel, the most studied and the prototype of the notorious class II kernels. The method is general and could help shed some light on the whole class of kernels, thus increasing the practical use of Smoluchowski's approach to understand aggregation phenomena. This point is worth an example. This is precisely what is dealt with in Sec. VI.

VI. APPLICATION IN TWO-DIMENSIONAL DECAYING TURBULENCE

In this section we would like to illustrate the results obtained in this article by presenting an original application outside the field of massive particle aggregation, namely, the dynamics of vortices in two-dimensional decaying turbulence.

Recently, a statistical numerical model was introduced [25,26] which describes the dynamics and the merger of vortices with the assumption that the typical core vorticity ω and the total energy $E \sim \int v^2 d^2x \sim \Sigma_i \omega^2 R_i^4$ are conserved (R_i is the radius of the ith vortex) throughout the merging processes. This model reproduces the main features observed in direct numerical simulations (see [25,26] for details). For instance, after noting that a distribution of vortex radii satisfying $P(R) \sim R^{-\beta}$ is equivalent to a Gaussian energy spectrum $E(k) \sim k^{\beta-6}$ [26], the simulation of this model was able to reproduce the fact that starting from a Batchelor spectrum $E(k) \sim k^{-3}$ $(\beta=3)$, the system evolves systematically to a steeper spectrum $E(k) \sim k^{-\gamma}$ with $\gamma=6-\beta$ in the range $\gamma \approx 3 \sim 5$ [26].

Now, one expects that the collision kernel between two vortices is somewhat intermediate between the ballistic hard-disk form $\sigma \sim (R_1+R_2)$ [21], and the totally uncorrelated form $\sigma \sim (R_1+R_2)^2$ [where the probability of colliding is proportional to the probability that two randomly placed vortices overlap, see also below Eq. (2.1)]. Thus one can describe approximately the decay of vortices due to mergers by means of Eq. (2.5) with $1 \leqslant d \leqslant 2$ and $D=4$, as two colliding vortices merge into a new one with $R=(R_1^4+R_2^4)^{1/4}$ in order to conserve energy and core vorticity. One thus expects a power law radius distribution $P(R) \sim R^{-\beta}$, with $\beta=D(\tau-1)+1$ and τ given by our model. We find values of γ ranging from $\gamma \approx 3.26$ for $d=2$ (taking $\tau=1.434$) to $\gamma \approx 4.95$ (taking $\tau=1.012$) for $d=1$, in good qualitative agreement with observed exponents. As also found in direct simulations, the actual exponent (and here the value of the effective correct d) could depend on the actual initial conditions (ω, area occupied by the vortices $\sim$ enstrophy). Note that the Batchelor limit case $\gamma=3$ is obtained when taking the naive strict upper bound $\tau=1+d/D$ with $d=2$ and $D=4$.

VII. CONCLUSION

In this article, we tackled the notoriously difficult problem of nontrivial polydispersity exponents in Smoluchowski's approach to aggregation from an original angle. We chose to directly start from the scaling (infinite time limit) equation, and we did not focus on the determination of the whole scaling function, which is the object of solving Smoluchowski's equation, to concentrate on τ itself, which actually mainly depends on global (integral) equations. We think, and illustrated this point on the example of a simplified model of two-dimensional turbulence, that in some cases, the only knowledge of τ would still be a good step towards the understanding of the phenomenon. The choices we made were fruitful and gave birth to new analytical and numerical results.

From an analytical viewpoint, we were able to use integral equations to find some exact bounds for τ, and, in the specific case of $K_D^d=(x^{1/D}+y^{1/D})^d$, we obtained some perturbative and nonperturbative expansions of τ, without explicitly computing the corresponding expansions for the whole scaling function.

From a numerical viewpoint, we devised a variational approximation scheme, that recovers by construction known exact results, and can be used as a tool for extensive determination of τ, since it is both very economical and accurate. In addition, it is likely that the scaling function obtained in the variational approach is in many cases qualitatively, if not quantitatively, right. To illustrate its effectiveness, we performed a comprehensive study of τ for a wide range of the parameters (d,D) of the kernel K_D^d. This is a noticeable advance, since very little quantitative knowledge was available for this kernel, although it was the prototype kernel with a nontrivial τ, and the object of much attention in the past [17–24,27–29].

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APPENDIX A: A USEFUL FORMULA

$$\int\int_0^{+\infty} x^{-\tau_1} y^{-\tau_2} e^{-x-y} K(x,y)[(x+y)^\alpha - x^\alpha - y^\alpha] dx dy = \Gamma(2+\lambda+\alpha-\tau_1-\tau_2)[\mathcal{X}(\tau_1,\alpha,\tau_1+\tau_2)+\mathcal{X}(\tau_2,\alpha,\tau_1+\tau_2)], \tag{A1}$$

where Γ is the gamma function, and

$$\mathcal{X}(t,\alpha,q)=\int_0^1 \frac{K(1,u)[(1+u)^\alpha-1-u^\alpha]}{u^t(1+u)^{2+\lambda+\alpha-q}} du. \tag{A2}$$

To demonstrate this formula is straightforward: just make the change of variable $x=uv, y=v$, and use the definition of the Γ function:

$$\Gamma(x)=\int_0^{+\infty} t^{x-1} e^{-t} dt. \tag{A3}$$

From a numerical viewpoint this formula makes it possible to implement very rapid and accurate code for the variational approximations we developed before. It would be very awkward and inefficient to use two-dimensional numerical integration (especially here, as the integrand is singular at the origin). A startlingly economical way of computing the Γ function is due to Lanczos and is described in [32] (it is not much slower than the built-in exponential function).

APPENDIX B: THE $O(d^2)$ TERM IN $D=1$

We derive the $O(d^2)$ correction to $\tau=2d$ for $D=1$, by computing the d^2 order of, respectively, Eqs. (2.7) and (2.8) with $\alpha=d$, to get

$$4-2a_2=c+4\int_0^{+\infty} f_1(x)\ln x\,dx+4\int_0^{+\infty} f_2(x)\,dx, \tag{B1}$$

$$-a_2=\frac{1}{2}\int_0^{+\infty} e^{-x}(\ln x)^2 dx+\int_0^{+\infty} f_1(x)\ln x\,dx+\int_0^{+\infty} f_2(x)\,dx, \tag{B2}$$

where $\tau=2d+a_2d^2+O(d^3)$, and

$$c=4\int_0^{+\infty} e^{-x}[\ln(x)]^2dx+4\int\int_0^{+\infty} e^{-x-y}\ln(x+y)\times\ln\frac{xy}{x+y}dx\,dy+4\left(\int_0^{+\infty} f_1(x)\,dx\right)\left(\int_0^{+\infty} e^{-y}\ln y\,dy\right)+\left(\int_0^{+\infty} f_1(x)\,dx\right)^2. \tag{B3}$$

c can be computed since $\int f_1$ is known from the first order calculation. After some elementary transformations, we find that $c-4\int e^{-x}(\ln x)^2dx=2\pi^2/3-4$. Combining Eqs. (B1) and (B2), we find $4+2a_2=c-4\int e^{-x}(\ln x)^2dx$, hence eventually

$$a_2=\frac{\pi^2}{3}-4. \tag{B4}$$

APPENDIX C: THE LINEARIZED SCALING FUNCTION

We find the solution of the second order differential equation (4.10) for the linear coefficient $f_1(x)$ in the small d expansion of the scaling function. With $u(x)=e^x f_1(x)$, the latter equation is

$$xu''+(1-x)u'+u=-\frac{2}{D}\ln x-2J_D+\frac{4}{D}\int_0^{+\infty} e^{-y}\frac{x^{1/D-1}}{y^{1/D}+x^{1/D}}dy. \tag{C1}$$

With $v(x)=u(x)/(x-1)$, this equation reduces to a first order differential equation for v', and we find

$$f_1(x)=c_0u_0(x)e^{-x}+c_1(x-1)-2J_D-\frac{2}{D}(1+\ln x)+\frac{4}{D}e^{-x}\int_0^x dy_1\frac{e^{y_1}}{y_1(y_1-1)^2}\int_0^{y_1}dy_2y_2^{1/D-1}e^{-y_2}\times(y_2-1)\int_0^{+\infty}dy_3\frac{e^{-y_3}}{y_3^{1/D}+y_2^{1/D}} \tag{C2}$$

and

$$u_0(x)=e^x-(x-1)\mathrm{P}\left(\int_{-\infty}^x\frac{e^y}{y}dy\right) \tag{C3}$$

(‘‘P’’ means ‘‘principal value’’).

In fact, the triple integral can be transformed into a simple integral involving special functions. For our purpose, we only need to know that this integral goes to zero when $x\to 0$, which is easily seen.

APPENDIX D: PERTURBATIVE ESTIMATE

For $d>1$, $\tau=1+\varepsilon(D)$ where $\varepsilon\to 0$ when $D\to\infty$. We make the ansatz

$$f(x)\approx f_\infty(x)+\frac{c}{x^{1+\varepsilon}}e^{-x} \tag{D1}$$

and plug it into Eq. (2.7) to obtain $1-\varepsilon=2^{1-d}+c\Gamma(d/D-\varepsilon)$, which means that, when $D\to\infty$, $c\approx(1-2^{1-d})(d/D-\varepsilon)$. Then we make use of Eqs. (A1) and (2.8) to obtain

$$2\left(1-\frac{d}{D}\right)(1-\varepsilon)=2^{2-2d}\int\int e^{-x-y}(x^{1/D}+y^{1/D})^d[x^{d/D}+y^{d/D}-(x+y)^{d/D}]dx\,dy+2^{2-d}c\Gamma(1+2d/D-\varepsilon)[\mathcal{X}(0,d/D,1+\varepsilon)+\mathcal{X}(1+\varepsilon,d/D,1+\varepsilon)]+2c^2\Gamma(2d/D-2\varepsilon)\mathcal{X}(1+\varepsilon,d/D,2+2\varepsilon). \tag{D2}$$

The next step is to write down the limit of this equation when $D\to\infty$. We know that $\Gamma(x)\sim_{x\to 0}1/x$, and a change of variable $v=u^{d/D-\varepsilon}$ in the integral factors $\mathcal{X}$ shows that

$$\mathcal{X}(1+\varepsilon,d/D,1+\epsilon)\sim\mathcal{X}(1+\varepsilon,d/D,2+\varepsilon)\sim(d/D-\varepsilon)^{-1}\textstyle\int_0^1(1+v^{1/(d-D\varepsilon)})^d dv.$$

We obtain

$$2=2^{2-d}+2^{2-d}\frac{c}{d/D-\varepsilon}\int_0^1(1+v^{1/(d-\kappa)})^d dv+\frac{c^2}{(d/D-\varepsilon)^2}\int_0^1(1+v^{1/(d-\kappa)})^d dv. \tag{D3}$$

κ is the limit of $D\varepsilon$. Taking into account the value of c, we finally get

$$\frac{2}{1+2^{1-d}}=\int_0^1(1+v^{1/(d-\kappa)})^d dv=J(\kappa,d), \qquad \text{(D4)}$$

$$\tau=1+\frac{\kappa}{D}+O\left(\frac{1}{D^2}\right). \qquad \text{(D5)}$$

Equation (D4) has a unique solution $0<\kappa<d$ since the integral $J(\kappa,d)$ is a decreasing function of κ, and $J(0,d)=2^d>2/(2^{1-d}+1)>1=J(d,d)$ (for $d>1$).

[1] S. K. Friedlander, *Smoke, Dust and Haze* (Wiley Interscience, New York, 1977).
[2] P. Meakin, Phys. Scr. **46**, 295 (1992).
[3] *On Growth and Form*, edited by H. E. Stanley and N. Ostrowsky (Martinus Nijhoff Publishers, Dordrecht, 1986).
[4] T. Vicsek, *Fractal Growth Phenomena* (World Scientific, Singapore, 1992).
[5] M. von Smoluchowski, Z. Phys. Chem. **92**, 129 (1918).
[6] P. Meakin, Phys. Rev. Lett. **51**, 1119 (1983); M. Kolb, R. Botet, and R. Jullien, *ibid.* **51**, 1123 (1983).
[7] G. F. Carnevale, Y. Pomeau, and W. R. Young, Phys. Rev. Lett. **64**, 2913 (1990).
[8] E. Ben-Naim, S. Redner, and F. Leyvraz, Phys. Rev. Lett. **70**, 1890 (1993).
[9] E. Trizac and J.-P. Hansen, Phys. Rev. Lett. **74**, 4114 (1995).
[10] P. Meakin, Rep. Prog. Phys. **55**, 157 (1992).
[11] F. Family and P. Meakin, Phys. Rev. A **40**, 3836 (1989).
[12] T. Vicsek and F. Family, Phys. Rev. Lett. **52**, 1669 (1984).
[13] H. Takayasu, Phys. Rev. Lett. **63**, 2563 (1989); S. Majumdar and C. Sire, *ibid.* **71**, 3729 (1993).
[14] *Nonequilibrium Statistical Mechanics In One Dimension*, edited by V. Privman (Cambridge University Press, Cambridge, England, 1996).
[15] N. G. van Kampen, *Stochastic Processes in Physics and Chemistry* (North-Holland, Amsterdam, 1981).
[16] P. G. J. van Dongen, Phys. Rev. Lett. **63**, 1281 (1989).
[17] D. Poland, J. Chem. Phys. **97**, 470 (1992).
[18] P. G. J. van Dongen and M. H. Ernst, Phys. Rev. Lett. **54**, 1396 (1985).
[19] K. Kang, S. Redner, P. Meakin, and F. Leyvraz, Phys. Rev. A **33**, 1171 (1986).
[20] D. S. Krivitsky, J. Phys. A **28**, 2025 (1995).
[21] S. Song and D. Poland, Phys. Rev. A **46**, 5063 (1992).
[22] J. Silk and S. D. White, Astrophys. J. **223**, L59 (1978).
[23] E. Ruckenstein and B. Pulvermacher, AIChE J. **19**, 356 (1973).
[24] P. G. Saffman and J. S. Turner, J. Fluid Mech. **1**, 16 (1956).
[25] G. F. Carnevale, J. C. McWilliams, Y. Pomeau, J. B. Weiss, and W. R. Young, Phys. Rev. Lett. **66**, 2735 (1991); J. B. Weiss and J. C. McWilliams, Phys. Fluids A **5**, 608 (1993).
[26] R. Benzi, M. Colella, M. Briscoloni, and P. Santangelo, Phys. Fluids A **4**, 1036 (1992).
[27] R. M. Ziff, M. H. Ernst, and E. M. Hendriks, J. Colloid Interface Sci. **100**, 220 (1984).
[28] P. G. J. van Dongen and M. H. Ernst, Phys. Rev. A **32**, 670 (1985); J. Phys. A **18**, 2779 (1985).
[29] F. Leyvraz, Phys. Rev. A **29**, 854 (1984).
[30] P. G. J. van Dongen and M. H. Ernst, J. Stat. Phys. **50**, 295 (1988).
[31] S. Cueille (unpublished).
[32] W. H. Press, B. P. Flannery, S. A. Teukolsky, and W. T. Vetterling, *Numerical Recipes* (Cambridge, Cambridge, England, 1986).