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Diffusion-limited-aggregation processes with three-particle elementary reactions

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A diffusion-limited-aggregation process, in which clusters coalesce by means of a three-particle reaction, $A+A+A\rightarrow A$, is investigated. In one dimension we give a heuristic argument that predicts logarithmic corrections to the mean-field asymptotic behavior for the concentration of clusters of mass m at time t, $C_m(t)\sim m^{-1/2}[\ln(t)/t]^{3/4}$, for $1\ll m\ll\sqrt{t/\ln(t)}$. The total concentration of clusters, $C(t)$, decays as $C(t)\sim\sqrt{\ln(t)/t}$ at $t\rightarrow\infty$. We also investigate the problem with a localized steady source of monomers and find that the steady-state concentration $C(r)$ scales as $r^{-1}[\ln(r)]^{1/2}$, r^{-1}, and $r^{-1}[\ln(r)]^{-1/2}$, respectively, for the spatial dimension $d=1$, 2, and 3. The total number of clusters, $N(t)$, grows with time as $[\ln(t)]^{3/2}$, $t^{1/2}$, and $t[\ln(t)]^{-1/2}$ for $d=1$, 2, and 3. Furthermore, in three dimensions we obtain an asymptotic solution for the steady-state cluster-mass distribution, $C_m(r)\sim r^{-1}[\ln(r)]^{-1}\Phi(z)$, with the scaling function $\Phi(z)=z^{-1/2}\exp(-z)$ and the scaling variable $z\sim m/\sqrt{\ln(r)}$.

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I. INTRODUCTION

Diffusion-limited-aggregation processes have attracted considerable recent interest in many fields of science and technology [1]. Typically, aggregation processes can be described by the binary reaction scheme

$$A_i+A_j\xrightarrow{K(i,j)}A_{i+j}\ .$$

Here, A_i denotes a cluster consisting of i monomers, an i-mer, and $K(i,j)$ is the rate at which the reaction between an i-mer and a j-mer proceeds. Much of the understanding of the kinetics of binary aggregation processes is based on the analysis of rate equations and their exact and scaling solutions [1]. For sufficiently low dimensions, the diffusion mechanism is not efficient enough and fluctuations in the densities of diffusing reactants result in dimension-dependent kinetic behavior at long times [2].

In a view of the richness of the kinetic behavior observed in the bimolecular model, it is of interest to investigate more complicated many-particle diffusion-limited-aggregation processes. In the present study, we focus on the three-particle reaction scheme

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

both for homogeneous and inhomogeneous situations, and outline a generalization for the n-particle case.

We will study the simplest n-particle aggregation process for which both reaction rates and diffusion coefficients do not depend on masses of clusters, $K(i_1,\ldots,i_n)=\text{const}$ and $D_k=\text{const}$. A notable feature of this process is that it reduces to a simple chemical reaction scheme, $nA\rightarrow A$, if one considers only the concentration of clusters. Hence, in the rate equation description, the concentration $C(t)$ obeys

$$\frac{dC}{dt}=-\gamma C^n\ , \tag{2}$$

with γ being the rate constant. In the long-time limit, the concentration behaves as

$$C(t)\simeq[\gamma(n-1)t]^{-1/(n-1)}\ . \tag{3}$$

However, the mean-field rate equation approach provides an accurate description of the kinetics only above the upper critical dimension d_c; when $d\leq d_c$, the mean-field theory does not predict correct long-time behavior. For the aggregation model with constant reactivities and diffusivities, the upper critical dimension is known: $d_c=2/(n-1)$ [3–5]. Thus, for the binary reaction in one

dimension, the kinetics is anomalous and the concentration decays as $t^{-1/2}$ (see, e.g., [2] and references therein), while the mean-field result is $C(t)\sim t^{-1}$. On the other hand, for $n\geq 4$ the mean-field answer (3) gives a correct asymptotic description of the kinetics in one dimension. The three-particle case is marginal in one dimension and hence a logarithmic correction has been expected [3–5]. After a number of attempts [3–6], the logarithmic correction of the form $C(t)\sim\sqrt{\ln(t)/t}$ has indeed been observed in a very recent study [7].

In Sec. II we explore the diffusion-limited three-particle aggregation process (1) in one dimension. We justify heuristically the appearance of the logarithmic correction for the concentration of clusters $C(t)$. Moreover, we obtain a complete scaling description of the cluster-mass distribution function:

$$C_m(t)\sim[\ln(t)/t]\Phi(z)\ ,$$

with the scaling function $\Phi(z)=z^{-1/2}\exp(-z)$ and the scaling variable $z\sim m\sqrt{\ln(t)/t}$.

In Sec. III we examine the three-particle aggregation process with a spatially localized source of monomers. We show that the system reaches the steady state for arbitrary spatial dimension d. We present evidence of two critical dimensions, $d_c=1$, as in the homogeneous system, and $d^c=3$, which demarcates the pure diffusive regime ($d>3$) from the diffusion-reaction regime ($d\leq 3$). In three dimensions, we derive a complete asymptotic solution for the steady-state cluster-mass distribution.

II. AGGREGATION PROCESS ON A LINE

Consider a linear lattice on which point clusters undergo a random walk. If n clusters happen to occupy the same lattice point, they aggregate irreversibly into a single cluster whose mass is equal to the sum of the masses of n parent clusters. This is the n-body particle coalescence model (PCM) [8]. The binary PCM in one dimension has been solved by Spouge [9]; see also [10–14] for other exact solutions of several generalization of the PCM. Note that we must keep the lattice spacing Δx finite, even in one dimension, since otherwise the reaction will be absent in the n-particle model for all $n\geq 3$. The possibility of passing to the continuum limit in the binary PCM significantly simplifies the analysis (see, e.g., [10,11,14]), while for $n\geq 3$ the spacing Δx, which may be considered as the size of particles, appears in the final results.

A simple heuristic argument explains the logarithmic corrections for the three-body PCM. Let T be a typical time between successive three-particle collisions in which cluster takes place. So the reaction rate is proportional to C/T,

$$\frac{dC}{dt}\simeq-\frac{C}{T}\ . \tag{4}$$

We will estimate T as follows: Consider a reference frame at rest with respect to an arbitrary "target" particle. When two other particles will be at the origin simultaneously, the target particle will die. Now consider all possible pairs of original particles. In the following, we will refer to these pairs as *imaginary* particles. For any pair, let us choose the location of one partner as the x coordinate and the other location as the y coordinate of the corresponding imaginary particle. Thus, we map the original diffusion process on the one-dimensional (1D) lattice onto a diffusionlike process on the two-dimensional (2D) square lattice with the same lattice spacing Δx. Although imaginary particles do not undergo a simple random walk, we shall assume that the asymptotic behavior of this diffusion process is similar to the one encountered in the simplest 2D random walk.

Now let us estimate the collision time T by considering an idealized 2D simple random walk of imaginary particles. The target particle will die when some imaginary particle will arrive at the origin. The density $P(r,t)$ of imaginary particles is governed by

$$\frac{\partial P(r,t)}{\partial t}=D\Delta P(r,t)\ , \tag{5}$$

with the initial condition

$$P(r,t=0)=C^2\ , \tag{6}$$

indicating the obvious fact that the density of imaginary particles is just the square of the density of original ones, and with the adsorbing boundary condition

$$P(r=\Delta x,t)=0\ . \tag{7}$$

A simple way to find an approximate solution of Eqs. (5)–(7) is to use a quasistatic approximation (see, e.g., [15]). In this approximation one solves the steady diffusion equation and accounts for the time dependence by a moving boundary condition. This very simple approach often gives asymptotically exact results (see, e.g., [16]). In the present problem, the quasistatic approximation yields

$$P(r,t)=C^2\frac{\ln(r/\Delta x)}{\ln(\sqrt{Dt}/\Delta x)}\ . \tag{8}$$

Now the collision time T may be evaluated by computing the flux to the origin and then by equating the flux to the unity:

$$\int_0^T 2\pi D\frac{\partial P(r=\Delta x,t)}{\partial r}dt=1\ . \tag{9}$$

This gives the final estimate,

$$T\simeq\ln(1/C\Delta x\sqrt{2\pi})/2\pi DC^2\ .$$

Substituting this result into (4), we arrive at the closed-form approximate differential equation for the concentration of clusters:

$$\frac{dC}{dt}\simeq\frac{2\pi DC^3}{\ln(C\Delta x\sqrt{2\pi})}\ . \tag{10}$$

Solving this equation at the long-time limit, one gets

$$C(t)\simeq\left[\frac{\ln(2Dt/\Delta x^2)}{8\pi Dt}\right]^{1/2}\ . \tag{11}$$

This result confirms previous suggestions [3–6] of possible logarithmic corrections to the mean-field power-law decay. Moreover, it is in a complete agreement with re-

cent simulational results [7]. A similar logarithmic correction has been found in the binary PCM in two dimensions, $C(t)\sim\ln(Dt/\Delta x^2)/Dt$ [17,8,18]. From these findings for two- and three-body PCM's in their critical dimensions, $d_c=2$ and $d_c=1$, respectively, one can expect the appearance of similar logarithmic corrections for the n-body PCM at the corresponding critical dimension $d_c=2/(n-1)$:

$$C(t)\sim[\ln(Dt/\Delta x^2)/Dt]^{1/(n-1)} .$$

Very recently, a similar result has been established by a renormalization group calculation for the n-particle annihilation process $nA\rightarrow 0$ (see [19]). Notice also that for $d<d_c$ dimensional analysis suggests a power-law decay with the exponent dependent on d and independent on n, $C(t)\sim(Dt)^{-d/2}$.

Let us now consider the three-body aggregation process (1). On the mean-field level, the process is described by the (generalized) Smoluchowski equation. For constant reactivities, the Smoluchowski equation has the form

$$\frac{dC_m}{dt}=\sum_{i+j+k=m}C_iC_jC_k-3C_mC^2 , \tag{12}$$

with $C=\sum_{i=1}^{\infty}C_i$. Notice that in Eq. (12) we set the reaction rate equal to 1 by an appropriate choice of units.

For the monodisperse initial conditions, $C_i(0)=N_0\delta_{i1}$, one can find the solution to Eq. (12) (see [4]). The densities of even-mass clusters vanish due to our choice of initial data, while the odd-mass densities are

$$C_{1+2m}=C_1\frac{\Gamma(m+\frac{1}{2})}{\Gamma(\frac{1}{2})\Gamma(m+1)}\left[1-\frac{C}{N_0}\right]^m , \tag{13}$$

where Γ is the Γ function. For example, the concentrations of monomers and clusters are given by

$$C_1=N_0(1+4N_0^2t)^{-3/4} , \quad C=N_0(1+4N_0^2t)^{-1/2} . \tag{14}$$

Notice that in the long-time limit the cluster-mass distribution (13) exhibits a scaling behavior of the form

$$C_{1+2m}\simeq\frac{C^2}{N_0}\frac{\exp(-z)}{\sqrt{\pi z}} , \tag{15}$$

with the scaling variable z, $z=mC/N_0$. Equations (13) and (15) show that the concentration decays as $C_{1+2m}\sim m^{-1/2}t^{-3/4}$ for $1\ll m\ll\sqrt{t}$. The exponents describing the scaling behavior might be obtained without appealing to the complete solution. For example, the exponent describing the time dependence, $C_{1+2m}\sim t^{-3/4}$, can be derived by solving Eq. (12) directly for small m.

In analogy with the analysis of the chemical reaction scheme, $3A\rightarrow A$, at the upper critical dimension one can suppose that the aggregation reaction process (1) at $d=d_c=1$ should be described by improved rate equations:

$$L\frac{dC_m}{dt}=\sum_{i+j+k=m}C_iC_jC_k-3C_mC^2 , \tag{16}$$

with L being the logarithmic factor

$$L=\ln(1/C\Delta x\sqrt{2\pi})/(\pi D) .$$

Upon summing equations (16) over all m, one can reproduce Eq. (10) for the cluster concentration, thus providing a useful check of self-consistency.

By applying a generating function technique, one can find an asymptotic solution to the modified Smoluchowski equation (16). The concentrations are given formally by the same expression (13), but with the modified monomer density

$$C_1\simeq N_0^{-1/2}\left[\frac{\ln(2Dt/\Delta x^2)}{8\pi Dt}\right]^{3/4} . \tag{17}$$

Both the scaling form and the scaling variable are identical to the corresponding mean-field result (15), although the normalization factors and widths of distributions differ by logarithmic factors.

A straightforward generalization to the n-body PCM shows that at the critical dimension $d=d_c=2/(n-1)$, the cluster-mass distribution exhibits scaling behavior of the mean-field form [4]:

$$C_{1+(n-1)m}(t)\simeq\frac{C^2}{N_0\Gamma\left[\frac{1}{n-1}\right]}z^{-(n-2)/(n-1)}e^{-z} , \tag{18}$$

with the scaling variable z,

$$z=mC/N_0\sim m[\ln(Dt/\Delta x^2)/Dt]^{1/(n-1)} .$$

In particular, the concentration of monomers decays as

$$C_1(t)\sim\left[\frac{\ln(Dt/\Delta x^2)}{Dt}\right]^{n/(n-1)^2} . \tag{19}$$

Thus, for n-body PCM's we have obtained the scaling description of the cluster-mass distribution function. Our approach is heavily based on the assumption of equal diffusivities of the reactants. (The second assumption of equal reactivities follows directly from the assumption of equal diffusivities since "sizes" of clusters in the PCM are the same.) However, from our results one can forecast some features of the general case. Consider, for example, the three-body PCM with the cluster diffusion coefficients D_m related to their mass m by $D_m\sim m^{-\delta}$. Since the typical mass of the cluster grows inversely proportional to the total concentration of clusters, $m_{\rm typ}\sim C^{-1}$, one can insert an estimate for the typical diffusion coefficient, $D_{\rm typ}\sim m_{\rm typ}^{-\delta}\sim C^{\delta}$, into expression (11) for the total concentration of clusters. This yields the following heuristic estimate for $C(t)$:

$$C(t)\sim\left[\frac{\ln(t)}{t}\right]^{1/(2+\delta)} , \tag{20}$$

but such an approach cannot predict the scaling form of the cluster-mass distribution function.

III. AGGREGATION PROCESS WITH A LOCALIZED STEADY SOURCE

In this section we consider n-body PCM with a steady spatially localized monomer input. We again focus on

the three-particle case and also write some final results for arbitrary n. Let the source of monomers be placed at the origin of d-dimensional space and J be the strength of the source. Within the continuum approximation, the concentration of clusters at time t and at distance r from the source, $C(r,t)$, satisfies the reaction-diffusion equation

$$\frac{\partial C(r,t)}{\partial t}=D\Delta C(r,t)-DR+J\delta(\mathbf{r}) . \tag{21}$$

Here, DR is the reaction term: $R=C^3$, $-C^3/\ln(C)$, and $C^{1+2/d}$ for $d>1$, $d=1$, and $d<1$, respectively. For $d>1$, the reaction term has the mean-field form; for the marginal case $d=1$, the form of the reaction term has been derived in Sec. II (we ignore numerical factors and set $\Delta x=1$ by an appropriate choice of the length scale); and for $d<1$, one can choose the reaction term of the form $C^{1+2/d}$ since it gives the correct long-time decay for the homogeneous system, $C(t)\sim(Dt)^{-d/2}$.

For the binary PCM, a qualitative investigation of similar reaction-diffusion equations has been performed in [12]. Following the same line of reasoning, let us assume that the system reaches the steady state and try a solution of the power-law form. By inserting such a form into the governing equation, one finds that dimension $d=3$ also plays a role of critical dimension in the present problem. For $d>3$, clusters do not interact far away from the source and the concentration decays as $r^{-(d-2)}$, i.e., as in the limit of no reaction. The reaction leads to the renormalization of the strength of the source but does not change the behavior qualitatively. In other dimensions, the reaction is relevant at all scales, and far away from the source $C(r)$ behaves as

$$C(r)\sim\begin{cases} r^{-1}[\ln(r)]^{-1/2} & \text{if } d=3 \\ r^{-1} & \text{if } 1<d<3 \\ r^{-1}[\ln(r)]^{1/2} & \text{if } d=1 \\ r^{-d} & \text{if } d<1 . \end{cases} \tag{22}$$

The critical cases $d=3$ and 1 have been treated separately. Since we expected logarithmic corrections to the power-law behavior $C(r)\sim r^{-1}$ for $1<d<3$, we tried a solution of the form $C(r)\sim r^{-1}[\ln(r)]^{\nu}$, inserted this form into Eq. (21), and finally found the exponent ν by asymptotically equating the most significant terms.

Notice that the system with a localized source but without reaction reaches the steady state only for sufficiently large spatial dimension, $d>2$. On the other hand, the system with reaction reaches the steady state for any d. Notice also the appearance of logarithmic corrections to the power-law behavior in two critical dimensions, $d_c=1$ and $d^c=3$: The former results from the logarithmic factor in the reaction term, while the latter reflects the fact that at $d=d^c$, the reaction just becomes relevant.

Since clusters perform a random walk and since the source was turned on at $t=0$, clusters will propagate diffusively up to the distance of the order $\sqrt{t}$. Therefore, at $r<\sqrt{t}$ the concentration of clusters approaches the steady-state limit given by Eq. (22); on the other hand, $C(r)\to 0$ rapidly for $r>\sqrt{t}$. Hence, the total number of clusters, $N(t)$, may be estimated from the relation $N(t)\sim\int_0^{\sqrt{t}} r^{d-1}C(r)dr$. This yields

$$N(t)\sim\begin{cases} t & \text{if } d>3 \\ t[\ln(t)]^{-1/2} & \text{if } d=3 \\ t^{(d-1)/2} & \text{if } 1<d<3 \\ [\ln(t)]^{3/2} & \text{if } d=1 \\ \ln(t) & \text{if } d<1 . \end{cases} \tag{23}$$

A straightforward generalization to the n-body PCM shows that, again, two critical dimensions d_c and d^c demarcate different behaviors: $d_c=2/(n-1)$ and $d^c=2n/(n-1)$. For the steady-state cluster concentration, one finds

$$C(r)\sim\begin{cases} r^{-(d-2)} & \text{if } d>d^c \\ r^{-d_c}[\ln(r)]^{-d_c/2} & \text{if } d=d^c \\ r^{-d_c} & \text{if } d_c<d<d^c \\ r^{-d_c}[\ln(r)]^{d_c/2} & \text{if } d=d_c \\ r^{-d} & \text{if } d<d_c , \end{cases} \tag{24}$$

while the total number of clusters scales as

$$N(t)\sim\begin{cases} t & \text{if } d>d^c \\ t[\ln(t)]^{-d_c/2} & \text{if } d=d^c \\ t^{(d-d_c)/2} & \text{if } d_c<d<d^c \\ [\ln(t)]^{d^c/2} & \text{if } d=d_c \\ \ln(t) & \text{if } d<d_c . \end{cases} \tag{25}$$

Consider now the behavior of the steady-state concentrations $C_m(r)$, for the three-body PCM in the most interesting three-dimensional (3D) case. The concentrations $C_m(r)$ obey the reaction-diffusion equations

$$\left[\frac{d^2}{dr^2}+\frac{2}{r}\frac{d}{dr}\right]C_m+\sum_{i+j+k=m}C_iC_jC_k-3C_mC^2 =-(J/D)\delta_{m1}\delta(\mathbf{r}) . \tag{26}$$

From these equations one can subsequently find asymptotic solutions for the total cluster concentration

$$C(r)\simeq r^{-1}[4\ln(r)]^{-1/2} , \tag{27}$$

for the concentration of monomers

$$C_1(r)\sim r^{-1}[\ln(r)]^{-3/4} , \tag{28}$$

etc. These results suggest the ansatz

$$C_m(r)=F_m(\rho)/r , \tag{29}$$

with $\rho=\ln(r)$ for the behavior of $C_m(r)$ for general m.

Substituting this ansatz into the governing equation, we obtain

$$\frac{dF_m}{d\rho}-\frac{d^2F_m}{d\rho^2}=\sum_{i+j+k=m}F_iF_jF_k-3F_mF^2 , \tag{30}$$

with

$$F=\sum_{m=1}^{\infty} F_m(\rho)=rC(r)\simeq(4\rho)^{-1/2} .$$

Far away from the source, one can omit the second term in the left-hand side of this equation. After replacement of all $F_m(\rho)$ by $C_m(t)$, the resulting approximate equation for source-induced aggregation at the steady state becomes *identical* to Eq. (12) for irreversible aggregation, with ρ, $\rho=\ln(r)$, playing the role of time t. For the latter problem, the cluster-mass distribution $C_m(t)$ approaches the scaling form not only in the monodisperse case but also for arbitrary (rapidly vanishing) initial conditions [4]. In the general case, the scaling solution still has the form (15), where the parameter N_0 is equal to the total mass of the system, $N_0=\sum_{m=1}^{\infty} mC_m(t)$ [4]. Since for the former problem the density of mass $M(r)$, $M(r)=\sum_{m=1}^{\infty} mC_m(r)$, satisfies the Laplace equation

$$D\left[\frac{d^2}{dr^2}+\frac{2}{r}\frac{d}{dr}\right]M(r)=-J\delta(\mathbf{r}) , \tag{31}$$

one gets $M(r)=J/4\pi Dr$. Therefore,

$$\sum_{m=1}^{\infty} mF_m(\rho)=rM(r)=J/4\pi D$$

will play the role of N_0 in the scaling solution. Combining all these findings, we finally obtain that in the scaling limit

$$m\to\infty , \quad r\to\infty , \quad z=2\pi Dm/J\sqrt{\ln(r)}=\text{finite} , \tag{32}$$

the steady-state cluster-mass distribution reaches the scaling form

$$C_{1+2m}(r)\simeq\frac{\pi D}{Jr\ln(r)}\frac{\exp(-z)}{\sqrt{\pi z}} . \tag{33}$$

Thus, in the steady state in three dimensions, the typical mass of clusters at distance r from the source grows unexpectedly slowly, $m_{\text{typ}}\sim\sqrt{\ln(r)}$.

From the steady state (33), one can find the following limiting behavior:

$$C_{1+2m}(r)\simeq\left[\frac{D}{J}\right]^{1/2} m^{-1/2}r^{-1}[\ln(r)]^{-3/4} \tag{34}$$

for $m\ll J\sqrt{\ln(r)}/D$. Notice that in the steady state the total cluster concentration $C(r)$ does not depend on the strength of the source, J, while all other concentrations $C_m(r)$ do depend on J.

For the general n-particle PCM, a complete asymptotic solution for the steady-state cluster-mass distribution may be found at the critical dimension $d=d^c$ following the procedure used for the three-body case. By applying the ansatz $C_m(r)=F_m(\rho)/r^{d_c}$, one can recast the steady-state reaction-diffusion equation to the n-particle Smoluchowski equation. Making use of the scaling solution of the latter equation (18), one obtains

$$C_{1+(n-1)m}(r)\sim[r\ln(r)]^{-d_c}z^{-(n-2)/(n-1)}e^{-z} , \tag{35}$$

with $z\sim m\ln(r)^{-1/(n-1)}$ being the scaling variable.

In conclusion, we have obtained the complete asymptotic solution for the steady-state cluster-mass distribution for the n-particle aggregation model with a spatially localized source of monomers in the critical dimension $d=d^c=2n/(n-1)$; the binary case corresponds to $d=4$, while the ternary case corresponds to $d=3$. The solution exhibits an unexpectedly slow logarithmic growth of the typical mass of clusters versus the distance r from the source: $m_{\text{typ}}\sim\ln(r)^{1/(n-1)}$. For sufficiency large dimensions $d>d^c$, we have found that clusters do not interact far away from the source and hence concentrations decay diffusively as $r^{-(d-2)}$. Therefore, for $d>d^c$ the reaction leads to the renormalization of the strength of the source but does not change the behavior qualitatively. For sufficiently small dimensions, $d^c>d>d_c$, the steady-state cluster-mass distribution is expected to be of the scaling form $C_m(r)\sim r^{-\beta}\Phi(mr^{-\alpha})$. From two known moments of the cluster-mass distribution

$$C(r)=\sum_{m=1}^{\infty} C_m(r)\sim r^{-d_c}$$

[see Eq. (21)] and

$$M(r)=\sum_{m=1}^{\infty} mC_m(r)\sim r^{-(d-2)} ,$$

one can find the exponents α and β, $\alpha=d^c-d$ and $\beta=d_c+d^c-d$. However, the scaling function $\Phi(mr^{-\alpha})$ satisfies a rather difficult integro-differential equation which I could not solve. Further progress is also possible in one dimension where exact solutions for the total density of clusters and for the densities of clusters of small mass may be readily found and behaviors even in the vicinity of the source may be investigated.

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