Singular self-preserving regimes of coagulation processes

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(Received 30 October 2001; published 2 April 2002)

The late stages of the time evolution of disperse systems when either coagulation alone governs the temporal changes of particle mass spectra or simultaneous condensation complicates the evolution process are studied under the assumption that the condensation efficiencies and coagulation kernels are homogeneous functions of the particle masses, with γ and λ being their homogeneity exponents, respectively. In considering the asymptotic behavior of the particle mass distributions the renormalization-group approach is applied to three types of coagulating systems: free coagulating systems in which coagulation alone is responsible for disperse particle growth; source-enhanced coagulating systems, where an external spacially uniform source permanently adds fresh small particles, with the particle production being a power function of time; and coagulating-condensing systems in which a condensation process accompanies the coagulation growth of disperse particles. The particle mass distributions of the form $\mathcal{N}_A(g,t)=A(t)\psi(gB(t))$ are shown to describe the asymptotic regimes of particle growth in all the three types of coagulating systems (g is the particle mass). The functions $A(t)$ and $B(t)$ are normally power functions of time whose power exponents are found for all possible regimes of coagulation and condensation as the functions of λ and γ. The equations for the universality function $\psi(x)$ are formulated. It is shown that in many cases $\psi(x)\propto x^{-\sigma}$ ($\sigma>1$) at small x, i.e., the particle mass distributions are singular. The power exponent σ is expressed in terms of λ and γ. Two exactly soluble models illustrate the general theoretical consideration.

DOI: 10.1103/PhysRevE.65.041604 PACS number(s): 68.03.Fg, 05.70.−a, 64.60.Qb

I. INTRODUCTION

The term "coagulation" refers to a large number of very diverse phenomena whose manifestations are related to coalescence of clusters—the parts of an evolving system. The simplest example of the coagulation process is the evolution of a system of N monomeric units that are able to form g-mers resulting from the process

$$(g_1)+(g_2)+\cdots+(g_k)\rightarrow(g_1+g_2+\cdots+g_k). \qquad (1)$$

Aging of aerosols and hydrosols [1–5], formation of traffic jams [6,7], cloud and precipitation formation [1,3,4], formation of fractals [8], evolution of random graphs [9], formation of the spectra of atmospheric aerosols [1,4,10–12], and even formation of bubbles in cheeses are only some of the phenomena, where coagulation plays a key role.

A special attention has been given to the study of the asymptotic regimes of coagulation [1–4], when the coagulation process has formed sufficiently large objects compared to initially existing ones. The reason for this enormous interest to this very stage of the process is not only practical. The far stages of coagulation processes obey the laws similar to those met already in the theories of phase transitions [8,13–16]. Respectively, the methods for studying the deep stages of the evolution of coagulating systems are also similar to those used in the theory of phase transitions, theory of turbulence, and theories of quantum fields [15]. However, in contrast to the above examples, where no closed equations are formulated, the kinetics of coagulation is more simple because it can, mostly, but not in every case, be described by the Smoluchowski kinetic equation. The latter is an integrodifferential equation analogous to Boltzmann's equation, but describing a deeply nonequilibrium process: in contrast to the gas-kinetic situations the coagulating systems have generally no final equilibrium state, or, better to say, this state contains no clusters.

In what follows we consider only binary coalescence ($k=2$) for which the Smoluchowski equation has the structure

$$\partial_t\mathcal{N}=(K\mathcal{N}\mathcal{N}). \qquad (2)$$

Here $\mathcal{N}=\mathcal{N}(g,t)$ is the population of g-mers at time t, and $(K\mathcal{N}\mathcal{N})$ stands for a functional quadratic in $\mathcal{N}$. The kernel K is a homogeneous function of the masses of colliding clusters. Because of the uniformity of Eq. (2) one expects it to possess the self-similar solutions

$$\mathcal{N}_A(g,t)=A(t)\psi(gB(t)), \qquad (3)$$

which can be likely candidates for describing the asymptotic stages. This concept had come up already more than half a century ago in Ref. [17]. In a more perfect form the theory of self-preserving mass distributions appeared later in Ref. [18] and found successful applications. Attempts to apply the renormalization group (RG) methods were made in Ref. [14], where the RG equation fixing the arguments of self-preserving mass spectra was formulated. The next very important step had been done in Ref. [19], where *singular* self-preserving regimes were discovered, i.e., the functions describing the asymptotics occured singular at small values of the self-similarity argument. It was shown that these singular mass spectra are not exceptional and are expected to be encountered in many practical situations.

This paper focusses on the study of the singular self-preserving regimes in coagulating and coagulating-condensing systems. The nonsingular spectra were considered in our recent paper [20]

In the following section we formulate necessary starting equations for the following three types of coagulation processes.

(i) Free coagulation, where an initial particle mass distribution evolves due to coagulation alone.

(ii) Source-enhanced coagulation, where a source permanently supplies the system with small fresh particles.

(iii) Coagulation in condensing systems in which a source of condensable substance (vapor, in what follows) provides the system with vapor condensing onto the particle surfaces. Section III applies the RG arguments to the three types of coagulating systems. The asymptotic particle mass spectra are found in the form of Eq. (2). The arguments in favor of the existence of the singular asymptotic regimes in coagulating systems are given in Sec. IV, where a classification of singular particle mass distributions is proposed. In Sec. V two examples of singular self-preserving mass spectra are given. The results are summarized in Sec. VI.

Nondimensional systems of units are used throughout the paper. They are introduced differently for each case listed above (see Sec. II).

II. BASIC EQUATIONS

Coagulation is a surprizingly simple process: two clusters containing, respectively, g and l monomeric units coalesce and produce irreversibly one cluster of the total mass $g+l$,

$$(g)+(l)\rightarrow(g+l). \tag{4}$$

The rate of this process $K(g,l)$ is assumed to be a known function of the masses g and l of colliding particles.

In this section we formulate the kinetic equations for three types of coagulation processes.

(i) Free coagulation, where an initial mass distribution of particles evolves because of coagulation alone.

(ii) Source-enhanced coagulation, where in addition to the process given by Eq. (4) a source providing the system with fresh particles is added.

(iii) Coagulation-condensation process, where the coagulating particles grow by simultaneous condensation of vapor molecules.

A. Free coagulation

Once the rate of elementary coalescence process $K(g,l)$ is known as a function of the masses of colliding particles, the kinetics of coagulation processes is described by the Smoluchowski kinetic equations the right-hand side of which (the collision term) balances the gain and the loss in the cluster population of given mass. This famous equation has the form

$$\partial_t \mathcal{N}(g,t)=(K\mathcal{N}\mathcal{N})_g. \tag{5}$$

Here $\mathcal{N}(g,t)$ is the particle mass spectrum (the number concentration of the particles of mass g within the mass interval $[g,g+dg]$ at time t), and

$$(K\mathcal{N}\mathcal{N})_g=\frac{1}{2}\int_0^g K(g-l,l)\mathcal{N}(g-l,t)\mathcal{N}(l,t)dl - \mathcal{N}(g,t)\int_0^\infty K(g,l)\mathcal{N}(l,t)dl \tag{6}$$

is the collision term. In most studies on coagulation the collision kernel $K(g,l)$ was assumed to be a homogeneous function of its variables

$$K(ag,al)=a^\lambda K(g,l), \tag{7}$$

with λ being the homogeneity exponent. We will also follow this tradition, but for simplicity restrict the consideration to separable coagulation kernels [21]

$$K(g,l)=\kappa(g^\alpha l^\beta+g^\beta l^\alpha), \tag{8}$$

where κ is a dimensionality carrier. It is apparent that

$$\lambda=\alpha+\beta. \tag{9}$$

In what follows we assume that $0\leqslant\lambda\leqslant 1$, $\alpha\geqslant\beta$.

The system of units $\kappa=M=1$ is used, with M being the particle mass concentration,

$$M=\int_0^\infty \mathcal{N}(g,t)g\,dg=\text{const.}$$

B. Source-enhanced coagulation

A spacially uniform source of fresh particles added to the coagulating system modifies the Smoluchowski equation as follows:

$$\partial_t \mathcal{N}(g,t)=J(g,t)+(K\mathcal{N}\mathcal{N})_g, \tag{10}$$

where $J(g,t)$ is the production of the particle source. In what follows we assume the source to produce the particles with masses much smaller than those formed in the course of the coagulation process at large time. The particle mass concentration is considered to grow with time as its power,

$$M(t)=Jt^s, \tag{11}$$

with $J=\int J(g)dg$ being the total production of fresh particles.

The system of units used in this case is $J=\kappa=1$.

C. Coagulation condensation

Consider a spacially uniform disperse system and assume that: (i) there is a time independent source of vapor of production I; (ii) initially existing particles whose mass spectrum is a known function of their mass g can coagulate and grow by simultaneous vapor condensation.

According to above assumptions the set of evolution equations looks as follows.

The rate of change with time in the monomeric concentration $C(t)$ is

$$\frac{dC}{dt}=I-\alpha C\varphi_{\gamma}, \tag{12}$$

where I is the production of the external source of vapor and

$$\alpha(g)=\alpha g^{\gamma} \tag{13}$$

is the condensational efficiency (α is a constant). The moments of the particle mass distribution φ_{γ} are defined as follows:

$$\varphi_{\gamma}(t)=\int_0^{\infty} g^{\gamma}\mathcal{N}(g,t)dg. \tag{14}$$

The first term on the right-hand side (rhs) of Eq. (12) increases the vapor concentration because of the action of the source. The last one is responsible for depleting the concentration of vapor due to its condensation onto the surfaces of disperse particles.

The continuity equation

$$\frac{\partial \mathcal{N}}{\partial t}+\alpha C\frac{\partial}{\partial g}g^{\gamma}\mathcal{N}=(K\mathcal{N}\mathcal{N})_g \tag{15}$$

describes the time evolution of the particle mass spectrum due to condensation [the second term on the left-hand side (lhs) of Eq. (15)] and coagulation (the rhs of this equation).

Two integral equalities will be of use further on. On integrating Eq. (15) over all g yields

$$\frac{dN(t)}{dt}=-\frac{1}{2}\int_0^{\infty}K(g,l)\mathcal{N}(g,t)\mathcal{N}(l,t)dg\,dl, \tag{16}$$

where $N(t)=\int_0^{\infty}\mathcal{N}(g,t)dg$ is the total particle number concentration.

The second equality reflects the mass conservation. Let us multiply both sides of Eq. (15) by g and again, integrating over all g. Then, noticing that $\int_0^{\infty}g(K\mathcal{N}\mathcal{N})_g dg=0$ one finds

$$\frac{dM}{dt}=-\alpha C\int_0^{\infty}g\frac{\partial g^{\gamma}\mathcal{N}}{\partial g}dg=\alpha C\varphi_{\gamma}(t), \tag{17}$$

where $M(t)=\int_0^{\infty}g\mathcal{N}(g,t)dg$ is the total mass concentration of disperse phase. Combining this result with Eq. (12) gives $d_tM=d_tC+I$ or

$$M(t)-M(0)=It-C(t). \tag{18}$$

In what follows the system of units $\alpha=I=1$ is used in this case.

III. RG APPROACH

Here the RG approach is applied for deriving the asymptotic mass spectra in the coagulating systems. Some results in this direction had been obtained earlier in Ref. [14].

The idea of application of RG is very simple. First, we investigate the invariance properties of the particle mass spectra with respect to possible scaling transformations. Then, a dynamical restriction and the mass conservation put additional constrains on the rescalings retaining free only one scale. The requirement of the independence of the asymptotic mass distribution of any scale whose value is defined by the initial mass distribution leads to the RG equation [15].

A. Free coagulation

It is easy to check that if $\mathcal{N}(g,t)$ is a solution to Eq. (5) then a rescaled function

$$\mathcal{N}_1(g,t)=\frac{1}{\mathcal{N}_0}\mathcal{N}\left(\frac{g}{g_0},\frac{t}{t_0}\right) \tag{19}$$

is also a solution once yet arbitrary scales meet the condition

$$\frac{g_0^{1+\lambda}t_0}{\mathcal{N}_0}=1. \tag{20}$$

The mass conservation

$$\int_0^{\infty}g\mathcal{N}(g,t)dg=\int_0^{\infty}g\mathcal{N}_1(g,t)dg \tag{21}$$

imposes another condition

$$g_0^2=\mathcal{N}_0. \tag{22}$$

The independence of the asymptotic mass distribution of the initial conditions implies its independence of the scales $\mathcal{N}_0$, g_0, and t_0. If we differentiate Eq. (19) over g_0, take into account the links Eqs. (20) and (22), and then put $g_0=1$, we derive the RG equation for the asymptotic mass distribution $\mathcal{N}_A$,

$$g\frac{\partial \mathcal{N}_A}{\partial g}+(1-\lambda)t\frac{\partial \mathcal{N}_A}{\partial t}+2\mathcal{N}_A=0. \tag{23}$$

The solution to this equation has the form

$$\mathcal{N}_A(g,t)=t^{-2/(1-\lambda)}\psi(gt^{-1/(1-\lambda)}), \tag{24}$$

where $\psi(x)$ is yet unknown function. The mass distribution Eq. (24) conserves the total particle mass concentration, i.e., the value

$$M=\int_0^{\infty}g\mathcal{N}_A(g,t)dg=\int_0^{\infty}x\psi(x)dx \tag{25}$$

does not change with time.

Substituting Eq. (24) into Eq. (5) results in the equation for the universality function $\psi(x)$,

$$-2\psi-x\psi'=(1-\lambda)(K\psi\psi)_x. \tag{26}$$

In studying the singular mass spectra it is much more convenient to rewrite Eq. (26) in terms of the functions

$$\phi_\sigma(x)=\int_x^\infty y^\sigma \psi(y)dy \quad (\sigma=\alpha,\beta). \tag{27}$$

Noticing that $-2\psi - x\psi' = (x\phi_0)''$ and using Eq. (A3) in the Appendix we get

$$x\phi_0(x)=(1-\lambda)(\phi_\alpha^* \phi_\beta), \tag{28}$$

where $(f*g)$ stands for the Laplace convolution $(f*g) = \int_0^x f(x-y)g(y)dy$. The equation linking the functions ϕ_σ and ϕ_0,

$$x^\sigma \frac{d\phi_0}{dx}=\frac{d\phi_\sigma}{dx} \tag{29}$$

follows from the definition of ϕ_σ [Eq. (27)].

Equations (26) and (28) are invariant with respect to the scaling transformation

$$\psi_1(x)=\frac{1}{x_0^{1+\lambda}}\psi\left(\frac{x}{x_0}\right), \tag{30}$$

that is, $\psi_1(x)$ is also a solution to Eqs. (26) or (28). This transformation, however, changes the total mass concentration

$$M_1 = x_0^{1-\lambda}M. \tag{31}$$

Another transformation

$$\psi_1(x)=\frac{1}{x_0^2}\psi\left(\frac{x}{x_0}\right) \tag{32}$$

leaves the mass unchanged, but changes Eq. (28). The function ψ_1 meets the equation

$$-x_0^{1-\lambda}(2\psi_1 + x\psi_1')=(1-\lambda)(K\psi_1\psi_1)_x. \tag{33}$$

At $\lambda=1$ the RG argumentation should be modified. It is expected that exponential time dependencies replace the power ones in Eq. (24). These dependencies contain a time scale fixed with the initial conditions to Eq. (5). We, therefore, replace $\partial_t = \xi\partial_\xi$ in Eq. (5) and redefine the rescaled function in Eq. (19),

$$\mathcal{N}_1(g,t)=\frac{1}{\mathcal{N}_0}\mathcal{N}\left(\frac{g}{g_0},\frac{\xi}{\xi_0}\right). \tag{34}$$

The function $\mathcal{N}_1$ is again, a solution to Eq. (5) if the condition Eq. (22) is fulfilled. The dynamical condition Eq. (20) adds nothing new, and the link between the scales ξ_0 and g_0 should be introduced differently. We use the link

$$\xi_0(g_0)=a g_0^\kappa \tag{35}$$

introducing no new scales. Here a and κ are constants. On differentiating $\mathcal{N}_1$ over g_0 and applying Eqs. (22) and (35) we get

$$g\frac{\partial \mathcal{N}_A}{\partial g}+t_a\frac{\partial \mathcal{N}_A}{\partial t}+2\mathcal{N}_A=0, \tag{36}$$

where $t_a = a\kappa$ is a time scale whose value is defined by the initial conditions.

The solution to Eq. (36) has the form

$$\mathcal{N}_A(g,t)=e^{-2t/t_a}\psi(ge^{-t/t_a}). \tag{37}$$

The equation for ψ is readily obtained on substituting Eq. (37) into Eq. (5),

$$x\phi_0 = t_a(\phi_\alpha^* \phi_{1-\alpha}). \tag{38}$$

The transformation property Eq. (30) remains valid at $\lambda=1$, but in contrast to other cases it changes neither the scale t_a nor the total mass [the value of $\phi_1(0)$].

It is important to notice that the scale t_a is proportional to the reciprocal mass concentration,

$$t_a \propto M^{-1}. \tag{39}$$

In order to prove it we notice that the asymptotic mass distribution has the structure

$$\mathcal{N}_A = M g_0^{-2}\psi(g/g_0). \tag{40}$$

The value of $\varphi_2 \propto g_0 M$ [see Eq. (14)]. On the other hand, $\dot{\varphi}_2 = \int g^2 (K\mathcal{N}_a\mathcal{N}_A)_g dg \propto M^2 g_0$. Hence, $\dot{g}_0 \propto g_0/M$, which proves Eq. (39).

B. Source-enhanced coagulation

The existence of self-preserving regimes in source-enhanced systems is less evident, for the presence of the source term in Eq. (10) makes it impossible for a straightforward application of RG. This difficulty can be avoided by assuming that the source can be ignored in Eq. (10) and replaced by the condition that the total mass concentration grows as $M(t)\propto t^s$. This step restores the possibility to apply the RG approach (see also Ref. [22]).

Once again, we compare two asymptotic mass spectra $\mathcal{N}_A(g,t)$ and $\mathcal{N}_0^{-1}N_A(g/g_0, t/t_0)$ and find the condition for the rescaled spectrum to describe the same regime as nonrescaled one. The first condition does not differ from Eq. (20),

$$\mathcal{N}_0 = g_0^{1+\lambda}t_0. \tag{41}$$

Next, the mass concentration should grow as the power s of time, i.e.,

$$\int_0^\infty g\mathcal{N}_A(g,t)dg=\frac{1}{\mathcal{N}_0}\int_0^\infty g\mathcal{N}_A\left(\frac{g}{g_0},\frac{t}{t_0}\right)dg = Jt^s. \tag{42}$$

This condition gives

$$\mathcal{N}_0=\frac{g_0^2}{t_0^s}. \tag{43}$$

Combining Eqs. (41) and (43) yields

$$\mathcal{N}_0 = g_0^{[2+s(1+\lambda)]/(1+s)}, \quad t_0 = g_0^{(1-\lambda)(1+s)}. \tag{44}$$

Our final requirement of independence of the asymptotic distribution of the scale g_0 leads to the RG equation

$$\frac{2+s(1+\lambda)}{1+s}\mathcal{N}_A + \frac{1-\lambda}{1+s}t\frac{\partial \mathcal{N}_A}{\partial t} + g\frac{\partial \mathcal{N}_A}{\partial g} = 0. \tag{45}$$

The solution to this equation has the form

$$\mathcal{N}_A(g,t) = t^{-\xi}\psi(gt^{-\eta}), \tag{46}$$

where

$$\xi = \frac{2+s(1+\lambda)}{1-\lambda}, \quad \eta = \frac{1+s}{1-\lambda}. \tag{47}$$

The equation for the universality function $\psi(x)$ is readily derived by substituting Eq. (46) into Eq. (5) for *free* coagulation. The action of the source is accounted for by condition (42) providing the power growth of the particle mass concentration with time. The equation for ψ looks as follows:

$$-\frac{2+s(1+\lambda)}{1-\lambda}\psi - \frac{1+s}{1-\lambda}x\psi' = (K\psi\psi)_x. \tag{48}$$

Integrating twice both sides of this equation from x to ∞ gives

$$\frac{1+s\lambda}{1-\lambda}x\phi_0 + s\phi_1 = (\phi_\alpha^*\phi_\beta). \tag{49}$$

At $\lambda=1$ the RG equation Eq. (39) should be modified by replacing the scale t_a with $t_a(t) \propto 1/M(t)$. This step can be done, for the power dependence of the mass concentration on time is slow compared to the exponential dependence of the characteristic particle mass [see Eq. (51) below]. The RG equation is then

$$g\frac{\partial \mathcal{N}_A}{\partial g} + \frac{t_a^{s+1}}{t^s}\frac{\partial \mathcal{N}_A}{\partial t} + 2\mathcal{N}_A = 0, \tag{50}$$

The solution to this equation again, has the form of Eq. (40) with

$$g_0(t) = \exp[-(t/t_1)^{s+1}]. \tag{51}$$

The function $\psi(x)$ meets the same equation [Eq. (38)] as in the case of free coagulation. The strightforward substitution of Eq. (40) with g_0 given by Eq. (51) readily proves this statement.

C. Coagulation condensation

Now let us return to Eq. (12). At the late stage of evolution of the system the vapor concentration $C(t)$ and the moments of particle mass distribution $\varphi_\gamma(t)$ are expected to be monotonous functions of time. Then it is possible to imagine two situations.

(i) The vapor concentration grows with time slower than t, and d_tC on the lhs of Eq. (12) can be neglected, i.e.,

$$C(t) \approx \frac{1}{\varphi_\gamma(t)}. \tag{52}$$

The mass of disperse phase then grows as $M \approx t$, i.e., all vapor mass converts to the particles. The case when monomer concentration grows linearly with time

$$C(t) \approx at, \tag{53}$$

but $a<1$, with the mass of the disperse phase growing linearly,

$$M(t) - M_0 = (1-a)t \tag{54}$$

can also be attributed to this item.

(ii) The vapor concentration grows with time as $C(t) \approx t$, while the mass of the disperse phase also grows, but slower than t,

$$M(t) \propto t^s \quad \text{with} \quad 0 \leqslant s < 1. \tag{55}$$

Below we derive the conditions for the realization of these cases and the equations for the asymptotic mass spectra.

Case (i). Let us rewrite continuity equation (15) taking into account Eq. (52),

$$\frac{\partial \mathcal{N}}{\partial t} + \frac{1}{\varphi_\gamma(t)}\frac{\partial g^\gamma \mathcal{N}}{\partial g} = (K\mathcal{N}N)_g. \tag{56}$$

Since the mass concentration grows with time as t, we can put $s=1$ in Eqs. (46) and (47) and find that the asymptotic regime is described by the distribution

$$\mathcal{N}(g,t) = A(t)\psi(gB(t)) \tag{57}$$

with

$$A(t) = t^{-\xi}, \quad B(t) = t^{-\eta}. \tag{58}$$

The power exponents ξ and η found from Eq. (47) are

$$\xi = \frac{3+\lambda}{1-\lambda}, \quad \eta = \frac{2}{1-\lambda}. \tag{59}$$

The equation for $\psi(x)$ is derived by substituting Eq. (56) into Eq. (15). The result is

$$-\frac{3+\lambda}{1-\lambda}\psi - \frac{2}{1-\lambda}x\psi' + \frac{1}{\phi_\gamma(0)}(x^\gamma\psi)' = \frac{d^2}{dx^2}(\phi_\alpha^*\phi_\beta). \tag{60}$$

The parentheses on the rhs of Eq. (60) stand for the Laplace convolution [see Eq. (A2) of the Appendix]. The lhs of Eq. (60) can be rewriten as

$$-\frac{3+\lambda}{1-\lambda}\psi - \frac{2}{1-\lambda}x\psi' + \frac{(x^\gamma\psi)'}{\phi_\gamma(0)} = \frac{d^2}{dx^2}\left(\frac{1+\lambda}{1-\lambda}x\phi_0(x) + \phi_1(x) - \frac{\phi_\gamma(x)}{\phi_\gamma(0)}\right). \tag{61}$$

Integrating Eqs. (60) and (61) twice from x to ∞ gives

$$\frac{1+\lambda}{1-\lambda}x\phi_0(x)+\phi_1(x)-\frac{\phi_\gamma(x)}{\phi_\gamma(0)}=(\phi_\alpha^*\phi_\beta). \tag{62}$$

The identity

$$\phi_1(x)=x\phi_0(x)+\int_x^\infty \phi_0(y)dy \tag{63}$$

allows one to cast Eq. (62) into another useful form,

$$-\int_0^x \phi_0(y)dy+\frac{2}{1-\lambda}x\phi_0(x)+\frac{1}{\phi_\gamma(0)}\int_0^x y^\gamma\psi(y)dy =(\phi_\alpha^*\phi_\beta). \tag{64}$$

Putting $x=0$ in Eq. (62) yields

$$\phi_1(0)=\int_0^\infty x\psi(x)dx=1. \tag{65}$$

Equation (57) together with Eqs. (58) and (59) provides the asymptotically linear growth of the total mass concentration with time,

$$M(t)-M_0=\int_0^\infty g\mathcal{N}_A(g,t)dg\approx t. \tag{66}$$

Case (ii). If $\varphi_\gamma(t)$ drops with time sufficiently fast, then the concentration $C\propto t$ at large t, the mass concentration of the disperse phase growing slower than t. The continuity equation (15) takes the form

$$\partial_t\mathcal{N}+t\partial_g g^\gamma\mathcal{N}=\frac{\partial^2}{\partial g^2}(\Phi_\alpha^*\Phi_\beta). \tag{67}$$

The substitution of $\mathcal{N}(g,t)$ in the form of Eq. (46) and balancing the powers of time in Eq. (67) give

$$\xi=\frac{3+2\lambda-\gamma}{1-\gamma},\quad \eta=\frac{2}{1-\gamma}. \tag{68}$$

Using Eq. (47) allows us to find the growth exponent s in Eq. (42),

$$s=\frac{1-2\lambda+\gamma}{1-\gamma}. \tag{69}$$

The condition $0<s<1$ puts two restrictions on γ and λ,

$$\gamma<\lambda \quad \text{and} \quad \gamma>2\lambda-1. \tag{70}$$

The equation for the universality function $\psi(x)$ looks as follows:

$$-\frac{3+2\lambda-\gamma}{1-\gamma}\psi-\frac{2}{1-\gamma}x\psi'+\frac{d}{dx}x^\gamma\psi=\frac{d^2}{dx^2}(\phi_\alpha^*\phi_\beta). \tag{71}$$

Integrating twice from x to ∞ both sides of Eq. (71) yields

$$\frac{1+\gamma-2\lambda}{1-\gamma}\int_x^\infty \phi_0(y)dy+\frac{2}{1-\gamma}x\phi_0(x)-\phi_\gamma(x)=(\phi_\alpha^*\phi_\beta). \tag{72}$$

It is seen, that the coefficient in the second term on the lhs of Eq. (72) is positive because of the second condition (70).

At $\gamma=2\lambda-1$ Eq. (69) gives $s=0$. The mass of disperse phase ceases to grow. At this and smaller γ the condensation process is slow and only a finite part of vapor converts to the disperse phase ($s=0$). This means that the condensation process becomes ineffective at large time and can be thus ignored. The coagulation process goes like in free systems.

IV. TYPES OF SINGULAR DISTRIBUTIONS

The term "singular distribution" appeared for the first time in Ref. [19]. It refers to the asymptotic distributions having a singularity at small particles masses. Such distributions had been known before this work, e.g., Junge's distributions of atmospheric aerosols [1,4], mass distributions in source-enhanced system [22–24], the asymptotic distribution in the Golovin-Scott model [25,26] (the model $\alpha=1$, $\beta=0$). In Refs. [19,22] it was shown that the singular distributions should appear in many realistic situations such as diffusion controlled formation of supported metal crystallites, coagulation of aerosols in shear viscous flows and in turbulent atmosphere, coagulation of fractals, source-enhanced coagulation of aerosols in the continuum regime, etc.

A. Singular distributions

Let us return to Eq. (24) and assume the function $\psi(x)$ to be integrable at $x=0$. Then the asymptotic time dependence of the total particle number concentration can be found from Eq. (24),

$$N(t)=\int_0^\infty \mathcal{N}(g,t)dg\approx\int_0^\infty \mathcal{N}_A(g,t)dg\propto t^{-1/(1-\lambda)}. \tag{73}$$

And now we are showing that for $\alpha=\beta>0$ the asymptotics Eq. (73) cannot hold. To this end, we consider the discrete version of the Smoluchowski equation, which, in particular, follows from Eq. (5) if the latter is subject to the initial condition $\mathcal{N}(g,0)=\delta(g-1)$ [$\delta(x)$ is Dirac's delta function]. In this case the mass spectrum has a discrete form

$$\mathcal{N}(g,t)=\sum_{k=1}^\infty c_k(t)\delta(g-k). \tag{74}$$

The concentrations $c_k(t)$ obey the set of equations

$$\frac{dc_s}{dt}=\frac{1}{2}\sum_{k=1}^{s-1}K(s-l,l)c_{s-l}c_s-c_s\sum_{k=1}^\infty K(s,l)c_l. \tag{75}$$

Let us consider the kernel $K(g,l)=g^\alpha l^\alpha$. From Eq. (75) one finds

$$c_1(t)=\exp\left(-\int_0^t \varphi_\alpha(t')dt'\right) \tag{76}$$

and

$$N(t)=1-\frac{1}{2}\int_0^t \varphi_\alpha^2(t')dt', \tag{77}$$

where the moment $\varphi_\alpha(t)$ is now defined as

$$\varphi_\alpha(t)=\sum_{k=0}^{\infty} k^\alpha c_k(t). \tag{78}$$

Since at $t\to\infty$ the monomer concentration goes to zero, the integral on the rhs of Eq. (76) must diverge, i.e., $\sigma\leqslant 1$. On the other hand, the integral $\int_0^\infty \varphi_\alpha^2(t)dt=1$, which corresponds to $N(\infty)=0$. Two inequalitiy $2\sigma>1$ and the law $N(t)\propto t^{-(2\sigma-1)}$ then follow immediately from the assumption that $\varphi_\alpha(t)\propto t^{-\sigma}$. And finally, the condition $\alpha>0$ leads to the obvious inequality $N(t)\leqslant\varphi_\alpha(t)$, i.e., $2\sigma-1\geqslant\sigma$. Combining these three inequalities, $\sigma\leqslant 1$, $2\sigma>1$, and $2\sigma-1\geqslant\sigma$ gives $\sigma=1$ instead of $\sigma=1/(1-2\alpha)$, as follows from Eq. (73).

The question then comes up: how to reconcile this result with the self-preservation in the form of Eq. (24)?

The answer was found in Ref. [19]. It is: one must sacrifice the assumption that the function $\psi(x)$ is regular at $x=0$. It was shown [19] that the function $\psi(x)$ having the singularity

$$\psi(x)\propto\frac{1}{x^{1+\lambda}} \quad \text{at} \quad x\ll 1 \tag{79}$$

removes the contradiction, once the diverging integrals be regularized by introducing a cutoff parameter $\zeta\propto 1$. For example,

$$N(t)=\int_0^\infty \mathcal{N}(g,t)dg\approx\int_\zeta^\infty \mathcal{N}_A(g,t)dg$$
$$\propto t^{-1/(1-\lambda)}\int_{\zeta b(t)}^\infty x^{-(1+\lambda)}dx\propto\frac{1}{t}, \tag{80}$$

where $b(t)\propto t^{-1/(1-\lambda)}$. All other moments $\phi_\alpha(t)$ with $\alpha<\lambda$ containig the divergency at small x also behave like t^{-1}.

Not all, however, is yet in order. The point is that the singular distribution cannot be so strightforwardly substituted into Eq. (5), for its rhs [Eq. (6)] contains divergent integrals. Once again, the situation is saved by introducing the cutoff parameter that *exactly cancels* in the limit $\epsilon=\zeta t^{-1/(1-\lambda)}\to 0$ (the proof shown in the Appendix). This extremely important property of the Smoluchowski equation was first noticed in Ref. [19].

B. Free systems

Let us analyze first the types of singularities arising in the asymptotic mass spectra in free systems. The substitution of the power function $\psi(x)\propto x^\sigma$ into Eq. (28) and balancing the powers lead to the result ($\alpha>\beta>0$):

$$\psi(x)=\mathcal{A}x^{-(1+\lambda)} \tag{81}$$

with

$$\mathcal{A}=\frac{\alpha\beta}{\lambda(1-\lambda)B(1-\alpha,1-\beta)}. \tag{82}$$

Here $B(x,y)$ is Euler's beta function.

Equations (81) and (82) give *an exact* solution of Eq. (28). Unfortunately, this exact solution is unnormalizable, i.e., the integral $\int_0^\infty x\psi(x)dx$ diverges. There exist other solutions having the singularity $\psi(x)\propto x^{-(1+\lambda)}$. This is a consequence of the scaling invariance of the function $\psi(x)$ [see Eq. (30)].

At $\alpha=\beta=0$ it is possible to find the normalizable solution

$$\psi(x)=e^{-ax}. \tag{83}$$

The scale a is defined by the normalization condition.

At $\beta=0$ the singularity is weaker than $x^{-(1+\lambda)}$, and the integral $\phi_\lambda(0)=\int_0^\infty x^\lambda\psi(x)dx$ converges. Hence, at small x Eq. (28) can be rewritten as

$$x\phi_0=(1-\lambda)\phi_\lambda(0)\int_0^x \phi_0(y)dy. \tag{84}$$

The solution to this equation is

$$\phi_0(x)=bx^{-\kappa}, \tag{85}$$

where $\kappa=1-(1-\lambda)\phi_\lambda(0)$ and b is a constant. If $q(x)=\phi_0(x)-bx^{-\kappa}\neq 0,\infty$ at $x\to 0$ then Eq. (84) reproduces the result of Ref. [27] $\kappa=\lambda/2$. We, however, did not find convincing arguments for rejecting the possibilities $q(0)=0$ or $q(0)=\infty$.

C. Source-enhanced coagulation

At small x the second term on the lhs of Eq. (49) goes to 1, and the asymptotics of $\psi(x)$ is defined by the equation

$$\int_0^x \phi_\alpha(x-y)\phi_\beta(y)dy=s. \tag{86}$$

Again, we seek the solution in the powerlike form. On substituting $\psi(x)=Ax^{-\omega}$ into Eq. (86) yields

$$\omega=\frac{3+\lambda}{2} \tag{87}$$

and

$$\mathcal{A}^2=s\frac{(\omega-\alpha-1)(\omega-\beta-1)}{B(\alpha-\omega+2,\beta-\omega+2)}, \tag{88}$$

Now let us notice that at $\omega=(3+\lambda)/2$ the sum of the arguments of beta function in Eq. (88) is unity, which allows one to use the well known theorem for Euler's beta function,

$$B(\alpha-\omega+2\beta-\omega+2)=B\left(\frac{1-\mu}{2},\frac{1+\mu}{2}\right)=\frac{\pi}{\cos(\pi\mu/2)}. \tag{89}$$

Finally,

$$\mathcal{A}=\sqrt{s\frac{(1-\mu^2)\cos(\pi\mu/2)}{4\pi}}, \tag{90}$$

where $\mu=\alpha-\beta$, $\mu\geqslant 0$. No singular solutions exist at $\mu<0$.

D. Coagulation condensation

Let us first analyze the behavior of the solution to Eq. (64) at small x. At $x\ll 1$,

$$\psi(x)\approx Ax^{-\omega}. \tag{91}$$

Since the first two terms on the lhs of Eq. (64) can be neglected at $x\ll 1$, the power balance gives

$$\omega=2-(\gamma-\lambda). \tag{92}$$

The coefficient A in Eq. (91) is then readily found from Eq. (64),

$$\mathcal{A}=\frac{(1+\alpha-\gamma)(1+\beta-\gamma)}{(2\gamma-1-\lambda)B(\gamma-\alpha,\gamma-\beta)\phi_\gamma(0)}. \tag{93}$$

Now let us formulate the conditions for realizing the case (i) in terms of λ and γ. Using the definition of $\varphi_\gamma(t)$ Eq. (14) and the distribution in the form Eq. (57) gives

$$C(t)=\varphi_\gamma(t)\propto AB^{-(1+\gamma)}\propto t^r \tag{94}$$

with

$$r=\xi-\eta(1+\gamma)=\frac{1+\lambda-2\gamma}{1-\lambda}. \tag{95}$$

The condition allowing for ignoring d_tC in Eq. (12) is $r<1$ or

$$\gamma>\lambda. \tag{96}$$

Another restriction on γ follows from the convergence of $\int_0^x y^\gamma\psi(y)dy$ at the lower limit,

$$2\gamma-\lambda>1. \tag{97}$$

At $\lambda<\gamma<(1+\lambda)/2$ the condensation term in Eq. (56) can be neglected, and the asymptotic regimes do not differ of those in source-enhanced coagulating systems.

The singular asymptotics never realizes at $\gamma<\lambda$. Neither attempt to balance the powers of x in Eq. (72) gives consistent results.

V. EXAMPLES

This section considers two models in which the singular universality functions can be found analytically.

A. Model $\alpha=1$, $\beta=0$

Let us consider first the model $\alpha=1$, $\beta=0$. As was shown above, the asymptotic regimes of free and source-enhanced coagulation and coagulation-condensation processes at $\gamma=1$ are described by the same universality function $\psi(x)$, which is the solution of the equation

$$t_a x\phi_0=(\phi_0^*\phi_1), \tag{98}$$

where t_a is the time scale introduced in deriving Eq. (38). The link

$$\phi_1'=x\phi_0' \tag{99}$$

closes Eq. (98).

In terms of Laplace's images Eqs. (98) and (99) have the form

$$-t_a\frac{d\Phi_0}{dp}=\Phi_0(p)\Phi_1(p) \tag{100}$$

and

$$p\Phi_1(p)-1=-\frac{d}{dp}p\Phi_0(p), \tag{101}$$

where $\Phi_\sigma(p)=\int_0^\infty e^{-px}\phi_\sigma(x)dx$.

Equation (100) allows one to find the constant t_a. To this end, we put $p=0$ in Eq. (100) and notice that $\Phi_0'(0)=\int_0^\infty x\Phi_0 dx$. On integrating by parts gives $\Phi_0'(0)=1/2\int_0^\infty\psi(x)x^2dx=0.5\phi_2(0)$. Similarly one finds $\Phi_1(0)=\int_0^\infty x\phi_1(x)dx=\phi_2(0)$. Then Eq. (100) gives

$$t_a=2. \tag{102}$$

There is no problem now to derive a first-order differential equation for Φ_0 alone,

$$2p\frac{\Phi_0'}{\Phi_0}=\Phi_0+p\Phi_0'-1. \tag{103}$$

The result Eq. (102) is taken into account in Eq. (103). Equation (103) is readily solved to give

$$\Phi_0(p)=\frac{\sqrt{1+4p}-1}{2p}. \tag{104}$$

The function $\psi(x)$ corresponding to Eq. (104) is

$$\psi(x)=\frac{1}{\sqrt{2\pi x^3}}e^{-x/4}. \tag{105}$$

The function $\psi(x)$ can always be rescaled according to Eq. (32).

The asymptotic solution of this problem was found in Ref. [19] for free coagulation.

B. Model $\alpha=\beta=0$

Let us consider the source-enhanced coagulation process for the model with constant coagulation kernel. According to the consideration of Sec. IV the asymptotic mass distribution has the singularity $\psi(x)\propto x^{-3/2}$. Here we find the exact asymptotic mass distribution. To this end, we must solve the equation [see Eq. (49)],

$$x\phi_0(x)+s\phi_1(x)=(\phi_0^*\phi_0). \tag{106}$$

Let us again introduce the Laplace image $\Phi_0(p)$ and use Eq. (101). These steps result in the first-order differential equation

$$\left(s\frac{\Phi_0-1}{p}+(s+1)\frac{d\Phi_0}{dp}\right)=-\Phi_0^2. \tag{107}$$

Noticing that

$$s\frac{\Phi_0}{p}+(s+1)\frac{d\Phi_0}{dp}=(s+1)p^{-s/(s+1)}\frac{d}{dp}p^{s/(s+1)}\Phi_0 \tag{108}$$

reduces this equation to

$$\frac{dy}{d\zeta}+y^2-s\zeta^{s-1}=0. \tag{109}$$

Here we introduced the new unknown function $y=p^{s/(s+1)}\Phi_0$ and the variable $\zeta=p^{1/(s+1)}$. At $s=1$ this equation has a simple solution $y=\tanh\zeta$ or

$$\Phi_0(p)=\frac{1}{\sqrt{p}}\tanh\sqrt{p}. \tag{110}$$

The original is now readily restored,

$$\phi_0(x)=\Theta_2(0|i\pi x), \tag{111}$$

where

$$\Theta_2(z|y)=2\sum_{n=0}^{\infty}\exp\left[i\pi y\left(n+\frac{1}{2}\right)^2\right]\cos(2n+1)z \tag{112}$$

is Jacobi's theta function [28]. At $x\ll 1$ it has the asymptotics

$$\phi_0(x)=\frac{1}{\sqrt{\pi x}}\left[1-2\exp\left(-\frac{1}{x}\right)\right]. \tag{113}$$

Equation (109) can be reduced to a linear second-order differential equation

$$\frac{d^2Z}{d\zeta^2}-s\zeta^{s-1}Z=0. \tag{114}$$

Although the solution to this equation can be found in terms of known special functions the inversion of the Laplace transform in the analytical form is hardly possible.

VI. RESULTS AND DISCUSSION

In considering the asymptotic behavior of the particle mass distributions the RG approach has been applied to three types of coagulating systems: free coagulating systems in which coagulation alone is responsible for the disperse particle growth, source-enhanced coagulating systems, where an external spacially uniform source permanently adds fresh small particles, with the particle production being a power function of time, and coagulating-condensing systems in which a condensation process accompanies the coagulation growth of disperse particles. The particle mass distributions of the form [see Eq. (3)]

$$\mathcal{N}_A(g,t)=A(t)\psi(gB(t))$$

have been found to describe the asymptotic regimes of the particle growth in all the three types of coagulating systems.

In free coagulating systems Eq. (3) absolutely naturally follows from the Smoluchowski equation. Of course, this self-similarity solution corresponds to some fixed initial conditions, and the step done by earlier authors reduced to the assumption that the asymptotic regime of the coagulation process was independent of the form of the initial conditions. It was natural, therefore, to reject the singular asymptotics as irrelevant, for the integrals entering the collision term in the Smoluchowski equation were divergent.

The RG approach introduces the assumption of the independence of the initial conditions explicitly via the RG equation [Eqs. (23) and (45)], and the derivation of Eq. (3) then relies only on the transformation properties of the Smoluchowskii equation allowing for the formulation of the dynamical constraint Eq. (20). Another constraint Eq. (22) related to the mass conservation is entirely independent of the form of the kinetic equation. Moreover, the dynamical condition can also be derived relying upon a consideration in the spirit of earlier works on the RG [13], rather than the kinetic equation. For free coagulation it was done in Ref. [19]. The RG approach is in no way related to the concrete properties of the function ψ and thus readily admits its singular behavior. The fact that the singularity does not prevent the derivation of a closed equation for ψ resolves the problem of finding and classifying the asymptotic regimes.

In the source-enhanced coagulating systems and in coagulating-condensing ones no exact self-similarity solutions exist. Still the RG approach has been shown to work and to give rather impressive results.

We have given the classification of the singular self-preserving regimes in coagulating systems and have defined the conditions for their realization. They are listed below.

In the free coagulating systems $\psi(x)\propto 1/x^{1+\lambda}$ at $x\ll 1$, which corresponds to the mass distribution of the form

$$\mathcal{N}_A(g,t)\propto\frac{1}{g^{1+\lambda}t}. \tag{115}$$

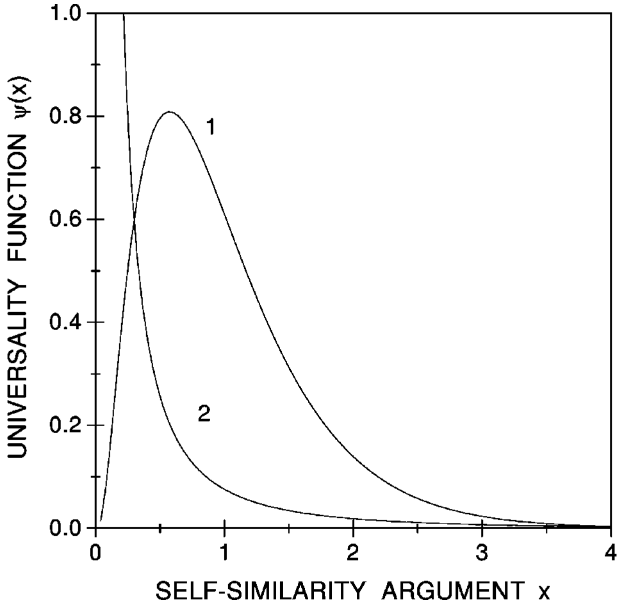

FIG. 1. Nonsingular (curve 1) and singular (curve 2) particle mass distributions. Shown are the universality functions $\psi(x)$ found for diffusion controlled coagulation of particles [the kernel $K(g,l) = (g^{-1/3}+l^{-1/3})(g^{1/3}+l^{1/3})$]. Curve 1 (nonsingular)—free coagulation process. Curve 2—source-enhanced coagulation process.

The condition for the realization of this asymptotics is $\alpha,\beta > 0$. At $\beta=0$ the singularity is weaker, $\psi(x)\propto 1/x^{1+\gamma}$, where $\gamma<\lambda$ [see Eq. (85)]. It is not so difficult to understand the physical meaning of this condition: the rate of interaction of small particles ($g\propto 1$) with large ones ($g\gg 1$) is of the order of $K(1,g)\propto g^{\alpha}$ and $K(g,g)\propto g^{\lambda}$, respectively, i.e., the smaller particles interact with the larger ones much slower than the latters between themselves ($\alpha\leqslant\lambda$). Strongly polydisperse mass spectra thus form in which the part of larger particles is less than that of smaller ones. The situation changes drastically once $\beta<0$. In this case $K(1,g)\gg K(g,g)$, i.e., the larger particles "eat" the smaller ones much faster than each others. A hump in the distribution at large masses develops, while the concentrations of small particles drops down with time. A singular and a nonsingular distributions are shown in Fig. 1.

Two inequalities $\alpha+\beta\leqslant 1$ and $\alpha-\beta<1$ define the conditions for the singular distributions to exist in source–enhanced coagulating systems. These are simply the conditions for the convergency of the integral on the rhs of Eq. (86). The singularity of the mass spectra in the source–enhanced coagulating systems is $\psi(x)\propto x^{-(3+\lambda)/2}$ [22,23] or, in terms of the particle masses

$$\mathcal{N}_A(g,t)\propto g^{-(3+\lambda)/2}t^{-(1-s)(1+\lambda)/2(1-\lambda)}. \tag{116}$$

At $s=1$ (a constant in time source) the time dependent multiplier turns to unity. The mass spectrum has a steady-state left wing, i.e., the spectrum of highly disperse fraction is independent of time, although the source permanently supplies the system with the fresh portions of small particles. These particles deposite mainly on the larger ones providing the right wing of the spectrum to move to the right along the mass axis. The steady-state regimes of coagulation in source-enhanced systems had been considered in [22–24].

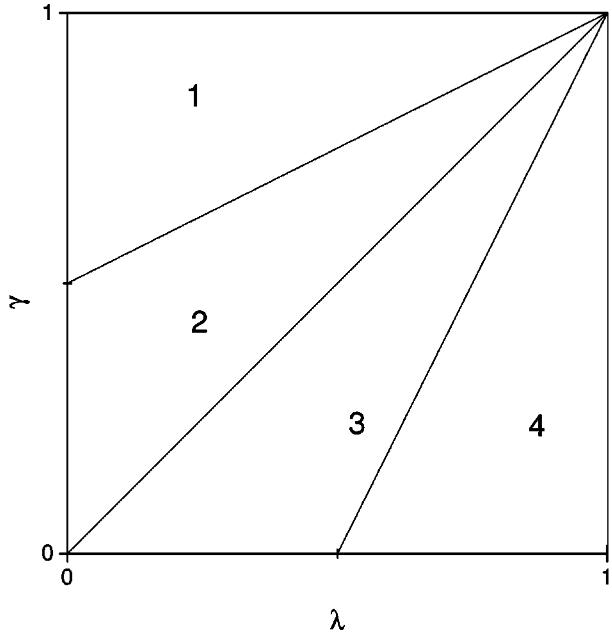

FIG. 2. The regimes of particle growth in coagulating-condensing systems. (1) $\gamma>(1+\lambda)/2$. The mass of disperse phase grows as t, i.e., the coagulating particles deplete the vapor. (2) $(1+\lambda)/2>\gamma>\lambda$. The mass of disperse phase still grows linearly with time. The universality function is the same as in source-enhanced coagulating systems with constant in time source. (3) $\lambda<\gamma<2\lambda-1$. The mass of disperse phase grows with time slower than linearly. The mass of condensable vapor grows linearly with time. No singular regimes are detected. (4) $\gamma<2\lambda-1$. Only a finite part of vapor condenses on the disperse particles. The universality function has the same shape as in free coagulating systems. No singular regimes can exist.

We have considered the systems of coagulating particles in which a constant in time source produces a low volatile vapor condensing onto the particle surfaces. The particle growth in such systems is similar in many respect to that in source–enhanced and (sometimes) free systems. Several regimes have been detected.

(i) The disperse phase consumes all mass of the vapor. In this case $\psi(x)\propto 1/x^{2-\gamma+\lambda}$, or

$$\mathcal{N}_A(g,t)\propto 1/t^{2\gamma-1-\lambda}g^{2-\gamma+\lambda}. \tag{117}$$

The conditions for realizing these distributions are: $\gamma<1$, $2\gamma>1+\lambda$. At $\lambda<\gamma<(\lambda+1)/2$ the coagulating-condensing system behaves like a source-enhanced coagulating system with linearly growing mass concentration.

(ii) When the mass of the disperse phase grows slower than t the asymptotic mass distribution in coagulating-condensing systems is the same as in source-enhanced systems. The singular asymptotics, however, never realizes. At $\gamma\leqslant 2\lambda-1$ condensation is so slow that the coagulating disperse system consumes only a finite part of vapor and the coagulation process goes like in free coagulating systems.

The whole situation is clearly seen from Fig. 2.

VII. CONCLUDING REMARKS

Coagulation processes display a number of very diverse self-preserving asymptotic regimes. The RG approach occurs to be a very fruitful tool for their investigation. Here we have focussed on the singular asymptotic regimes that realize in a number of practically important situations: source-enhanced coagulation of aerosols in continuum regime, coagulation-condensation growth of aerosol particles in turbulent flows, etc. Once the conditions for realizing the singular asymptotics are known, it becomes easy to diagnose them. It is much harder to solve respective equations and to find the function ψ either analytically or numerically. Even the simplest analytically soluble examples are far from trivial.

ACKNOWLEDGMENT

One of us (A.L.) thanks ISTC for financial support through Grant No. 1908.

APPENDIX: THE IDENTITY

Let $f(x)$ and $g(x)$ be arbitrary functions and

$$F(x)=\int_x^\infty f(y)dy \quad \text{and} \quad G(x)=\int_x^\infty g(y)dy. \tag{A1}$$

We also introduce the notation

$$(q*p)=\int_0^x q(x-y)p(y)dy \tag{A2}$$

for the Laplace convolution of a couple of functions $p(x)$ and $q(x)$.

Now we are ready to prove a very important identity

$$\frac{d^2}{dx^2}(F*G)=(f*g)-f(x)G(0)-g(x)F(0). \tag{A3}$$

To this end let us find the first derivative of $(F*G)$,

$$d_x(F*G)=F(0)G(y)+(F_x'*G). \tag{A4}$$

Next, we integrate by parts the second term on the rhs of Eq. (A4). The result is

$$(F_x'*G)=-(F_y'*G)=-G(y)F(0)+F(y)G(0)+(F*G_y'). \tag{A5}$$

On combining this result with Eq. (A4) yields

$$d_x(F*G)=G(0)F(y)+(F*G_y'). \tag{A6}$$

Differentiating once again Eq (A6) over x and taking into account that $F'=f$ and $G'=-g$ prove Eq. (A3).

Above proof assumed that the functions f, g, F, and G have no singularities at $x=0$. The extension to singular f and g is almost straightforward. We apply the identity Eq. (A3) to the functions

$$f_\epsilon(x)=f(x)\Theta(x-\epsilon) \quad \text{and} \quad g_\epsilon(x)=g(x)\Theta(x-\epsilon), \tag{A7}$$

where $\Theta(x)$ stands for the Heaviside step function. The result is

$$\frac{d^2}{dx^2}(F_\epsilon^*G_\epsilon)=\int_\epsilon^{x-\epsilon} f(x-y)g(y)dy-f(x-\epsilon)G(\epsilon) -g(x-\epsilon)F(\epsilon). \tag{A8}$$

Now it is seen that in the limit $\epsilon\rightarrow 0$ the rhs of Eq. (A8) is finite and independent of ϵ. Indeed, at small ϵ we have

$$\frac{\partial}{\partial\epsilon}\int_\epsilon^{x-\epsilon} f(x-y)g(y)dy-f(x-\epsilon)G(\epsilon)-g(x-\epsilon)F(\epsilon) =-f(x-\epsilon)g(\epsilon)-f(\epsilon)g(x-\epsilon)+f(x)g(\epsilon)+g(x)f(\epsilon). \tag{A9}$$

At small ϵ the rhs of Eq. (A9) goes to zero. In proving Eq. (A9) the convergence of the integral on its lhs was assumed, i.e., not very strong singularities of f and g are admissible.

The identity (A3) allows the Smoluchowski equation $\partial_t N(g,t)=(KNN)_g$ to be rewritten as

$$\frac{\partial N}{\partial t}=\frac{\partial^2}{\partial g^2}\int_0^g \Phi_\alpha(g-l,t)\Phi_\beta(l,t)dl. \tag{A10}$$

This form of the Smoluchowski equation appeared for the first time in Ref. [19].

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