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Nonsingular self-preserving regimes of coagulation-condensation process

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Growth of disperse particles is considered assuming that the preexisting disperse particles coagulate and grow simultaneously by condensing a low volatile substance (vapor, in what follows) whose concentration is permanently refreshed by a spacially uniform and constant in time source. The kinetics of the condensation-coagulation process is studied under the assumption that the condensation rate and coagulation kernels are homogeneous functions of the particle masses. The power exponents characterizing these functions define the asymptotic self-preserving regimes of the particle growth. Four such regimes are detected: (i) the mass of the disperse phase consumes all vapor and grows linearly with time, while the vapor concentration grows (or even drops) with time as its power $s<1$; (ii) the mass of the disperse phase grows slower than a linear function of time, while the vapor concentration grows asymptotically as time; (iii) the mass of disperse phase remains finite; and (iv) both, the mass of disperse phase and the vapor concentration grow linearly with time. For all above regimes the equations are derived defining the shape of the asymptotic mass distribution. The latter is shown to depend on a combination of the particle mass and time. The theory is illustrated by two exactly soluble models, and numerical results for the condensation-coagulation growth of aerosol particles in free molecular regime.

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I. INTRODUCTION

The self-preserving dynamics is not a rarity for nonlinear systems. Still every new self-preserving solution of a nonlinear dynamic equation is always an event. Self-preserving regimes are usually attributed to the asymptotic behavior of complex nonlinear systems when the system "forgets" the initial conditions at large times. The self-preserving regimes in coagulating systems are of special importance and have been studied already for more than half a century [1–5].

The investigations of coagulation processes received much attention in the last several decades from the specialists of very diverse areas (and not only physics). There are deep reasons for this. First, the list of the phenomena related to coagulation is impressive: aging of aerodisperse systems and hydrosols [1,2,4–6], formation of astrophysical objects [7], formation of traffic jams [8,9], sol-gel transitions (see Ref. [10] and references therein), evolution of random graphs [11]—the problem, which is, in turn, related to a number of social phenomena [12], and evolution of fractal systems [10]. Particularly coagulation and condensation processes are essential in the formation of nanomaterials (see, e.g., [13]), and in the formation of the atmospheric aerosols [2,5,14]. The latter process is of great importance in considering the climate changes via aerosol-cloud-climate interactions. Second, being simple from the first sight, this process reveals a huge diversity of regimes and even unexpected new phenomena like sol-gel transitions [15–18].

The physics of the coagulation process is surprisingly simple: two clusters containing respectively, g and l elements (monomers) coalesce and produce irreversibly one cluster of the total mass $g+l$,

$$(g)+(l)\rightarrow(g+l).$$

Once the rate of the elementary coalescence act is known as a function of the masses of colliding particles, the kinetics of coagulation processes is described by the Smoluchowskii kinetic equation, the right-hand side of which (the collision term) balances the gain and the loss in the cluster population of given mass. The collision term is usually quadratic in the cluster concentrations, which permits one to construct the self-preserving solution to this equation once the collision rates are homogeneous functions of the masses of colliding particles. These self-preserving solutions have played an extremely essential role in understanding the regimes of disperse particle coagulation.

However, the reality destroys the sweet self-preserving picture of the asymptotic behavior of coagulating systems. The homogeneity of the collision term is often broken by the intervenience of some important accompanying processes like particle sources, the processes of particle losses, condensational growth of particles, etc. In these cases the invariance with respect to the scaling transformations, providing the existence of the self preserving regimes is broken, and *exact* self-preserving regimes simply do not exist.

Still the asymptotical self-preservation can exist. Recently [19], while studying the nucleation-condensation-coagulation formation and growth of aerosol particles we encountered such a case. The problem of the existence of self-preserving solutions, however, remained unsolved. Only now we resolved this problem completely, found convincing arguments in favor of the existence of self-preserving regimes of coagulation condensation, and derived the equations describing the shape of asymptotic particle mass spectra.

This paper focuses on the consideration of the time evolution of coagulating systems, in which a constant in time source produces a condensable substance (condensable vapor, in what follows) whose molecules deposit on the surfaces of coagulating disperse particles. The mass of the disperse phase grows with time, and the coagulation process accelerates.

After formulating the model and the basic assumptions in Sec. II, we give a classification of the asymptotic self-preserving regimes and derive the equations defining the shapes of the particle-mass spectra (Sec. III). Section IV considers a couple of exactly soluble models that help one to understand how the system drives at the self-preserving regimes. In Sec. V we summarize the results and illustrate the theory by a realistic example: coagulation-condensation growth of aerosols in free molecular regime. This example had been considered by us in Ref. [19] within the log-normal approximation.

In what follows, we deal with *nonsingular* mass distributions, i.e., all integrals appearing in our consideration are assumed to be convergent. Most authors investigating the asymptotic behavior of coagulating mass spectra operate with these distributions. There exists another type of distributions: *singular* mass distributions [20–22], which require a much less straightforward approach.

II. BASIC EQUATIONS

Consider a spacially uniform disperse system and assume that:

(i) There is a time independent source of condensable vapor of productivity *I*.

(ii) Initially existing disperse particles can coagulate and grow simultaneously by condensing the vapor molecules (monomers).

(iii) The rates of coagulation and condensation are homogeneous functions of the particle masses.

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

$$\frac{dC}{dt}=I-\alpha C\varphi_\gamma, \tag{1}$$

where I is the productivity of the external source of monomers, and

$$\alpha(g)=\alpha g^\gamma \tag{2}$$

is the condensation rate [α is a constant, g is the particle mass (in the units of the monomer mass)]. The moments of particle-mass distribution $\varphi_\gamma(t)$ are defined as follows:

$$\varphi_\gamma(t)=\int_0^\infty g^\gamma \mathcal{N}(g,t)dg, \tag{3}$$

with $\mathcal{N}(g,t)$ being the particle-mass spectrum ($\mathcal{N}(g,t)dg$ is the number concentration of g-mers within the mass interval $[g,g+dg]$).

The first term on the right-hand side (rhs) of Eq. (1) increases the monomer concentration because of the action of the source. The last one is responsible for depleting the concentration of monomers due to their condensation onto the surfaces of disperse particles.

The continuity equation

$$\frac{\partial \mathcal{N}}{\partial t}+\alpha C\frac{\partial}{\partial g}g^\gamma \mathcal{N}=(K\mathcal{N}\mathcal{N})_g \tag{4}$$

governs the time evolution of the particle-mass spectrum due to condensation [the second term on the left-hand side of Eq. (4)] and coagulation (the rhs of this equation). The coagulation term $(K\mathcal{N}\mathcal{N})_g$ has well known form

$$(K\mathcal{N}\mathcal{N})_g=\frac{1}{2}\int_0^g K(g-l,l)\mathcal{N}(g-l,t)\mathcal{N}(l,t)dl \\ -\mathcal{N}(g\cdot t)\int_0^\infty K(g,l)\mathcal{N}(l,t)dl. \tag{5}$$

Here $K(g,l)$ is the coagulation kernel [the rate of the process $(g)+(l)\rightarrow(g+l)$].

Equations (1) and (4) are subject to initial conditions whose details are of no importance in the asymptotic analysis.

Two integral equalities will be of use further on. Integrating Eq. (4) over all g gives

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

where $N(t)=\int_0^\infty \mathcal{N}(g,t)dg$ is the total number concentration of particles. The second equality reflects the mass conservation. Let us multiply both sides of Eq. (4) by g and again integrate over all g. Then, noticing that $\int_0^\infty g(K\mathcal{N}\mathcal{N})_g)dg=0$, one finds

$$\frac{dM}{dt}=-\alpha C\int_0^\infty g\frac{\partial g^\gamma \mathcal{N}}{\partial g}dg=\alpha C\varphi_\gamma(t), \tag{7}$$

where $M(t)=\int_0^\infty g\mathcal{N}(g,t)dg$ is the total mass concentration of the disperse phase. On combining this result with Eq. (1) gives $d_tM=d_tC+I$ or

$$M(t)-M_0=It-C(t). \tag{8}$$

Here $M_0=M(0)$ is the initial mass concentration. The zero initial condition is imposed on $C(t)$: $C(0)=0$.

In what follows $K(g,l)$ is assumed to be a homogeneous function of its variables,

$$K(ag,al)=a^\lambda K(g,l), \tag{9}$$

with λ being the homogeneity exponent.

We restrict our consideration with $0\leqslant\lambda<1$, $0\leqslant\gamma\leqslant1$. This choice is stipulated by some physical reasons (see, e.g., [5]), although negative λ and $\lambda>1$ are not forbidden. In the latter case the coagulation process gives rise to gelatin, i.e., the formation of objects whose concentration is zero in the thermodynamic limit (see, e.g., [15]), and the Smoluchowskii equation does not work after the gel has formed or does not work at all. The case of negative λ is not of wide physical interest.

In what follows the system of units $\alpha = I = 1$ is used, i.e., all concentrations are measured in the units of $\sqrt{I/\alpha}$, and time in $1/\sqrt{I\alpha}$.

III. ASYMPTOTIC STAGE

Now let us return to Eq. (1). At the late stage of evolution of the system the values of $C(t)$ and $\varphi_\gamma(t)$ are expected to be monotonous functions of time. Then it is possible to imagine four situations.

(i) The monomer concentration grows with time slower than t, then $d_t C$ on the lhs of Eq. (1) can be neglected and

$$C(t) \approx \frac{1}{\varphi_\gamma(t)}. \tag{10}$$

The mass of disperse fraction grows as $M \approx t$, i.e., all vapor mass converts to disperse phase.

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

$$M(t) \propto t^s \quad \text{with} \quad 0 < s < 1. \tag{11}$$

(iii) The monomer concentration grows as t, while the mass of the disperse fraction goes to a constant.

(iv) The monomer concentration grows with time linearly,

$$C(t) \approx at, \tag{12}$$

with $a < 1$. The mass of the disperse phase then also grows linearly,

$$M(t) - M_0 = (1-a)t. \tag{13}$$

Below we derive the conditions for the realization of each case and the equations for the asymptotic mass spectra.

(a) Case (i). Let us rewrite continuity equation (4) taking into account Eq. (10). The result is

$$\frac{\partial \mathcal{N}}{\partial t} + \frac{1}{\varphi_\gamma(t)} \frac{\partial g^\gamma \mathcal{N}}{\partial g} = (K\mathcal{N}\mathcal{N})_g . \tag{14}$$

We state that Eq. (14) admits a self-preserving solution of the form

$$\mathcal{N}(g,t) = A(t)\psi(gB(t)), \tag{15}$$

with $A(t)$, $B(t)$, and $\psi(x)$ being yet unknown functions. Indeed, substituting Eq. (15) into Eq. (14) gives

$$\dot{A}\psi + A\frac{\dot{B}}{B}x\psi' + \frac{B^2}{\phi_\gamma}(x^\gamma\psi)' = \frac{A^2}{B^{1+\lambda}}(K\psi\psi)_x , \tag{16}$$

where

$$\phi_\gamma = \int_0^\infty x^\gamma \psi(x)\,dx. \tag{17}$$

The final equation for ψ should not contain any t dependencies that imposes the conditions

$$\dot{A} \propto A\frac{\dot{B}}{B} \propto B^2 \propto \frac{A^2}{B^{1+\lambda}}. \tag{18}$$

Set (18) is overdetermined, i.e., it comprises three equations for two unknown functions $A(t)$ and $B(t)$. Since exact equalities are not implied in Eq. (18), there exists a solution to this set: power functions of t meet conditions (18),

$$A(t) = at^{-\xi}, \quad B(t) = bt^{-\eta}. \tag{19}$$

The power exponents ξ and η are found balancing the powers of time in Eq. (18),

$$\xi = \frac{3+\lambda}{1-\lambda}, \quad \eta = \frac{2}{1-\lambda}. \tag{20}$$

The constants a and b can be included to the definition of the function ψ and thus put unity $a = b = 1$ in the final equation for ψ,

$$-\frac{3+\lambda}{1-\lambda}\psi - \frac{2}{1-\lambda}x\psi' + \frac{1}{\phi_\gamma}(x^\gamma\psi)' = (K\psi\psi)_x . \tag{21}$$

Multiplying both sides of Eq. (21) by x and integrating over all x give

$$\phi_1 = \int_0^\infty x\psi(x)\,dx = 1. \tag{22}$$

Equation (15) together with Eqs (19) and (20) provides the asymptotically linear growth of the total mass concentration with time,

$$M(t) - M_0 = \int_0^\infty gN(gt)\,dg \approx t. \tag{23}$$

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 (3), and the self-similar distribution Eq. (15) give $C(t) = 1/\varphi_\gamma(t) \propto B^{(1+\gamma)}/A \propto t^r$ with

$$r = \xi - \eta(1+\gamma) = \frac{1+\lambda-2\gamma}{1-\lambda}. \tag{24}$$

The condition allowing for ignoring $d_t C$ in Eq (1) is $r < 1$ or

$$\gamma > \lambda. \tag{25}$$

(b) Case (ii). If $\varphi_\gamma(t)$ drops with time sufficiently fast, then at large t the vapor concentration $C \propto t$. The mass concentration of the disperse phase grows slower than t. Continuity equation (4) in this case takes the form

$$\partial_t \mathcal{N} + t\partial_g g^\gamma \mathcal{N} = (K\mathcal{N}\mathcal{N})_x . \tag{26}$$

The substitution of $\mathcal{N}(g,t)$ in the form of Eq. (15) and the requirement the function ψ to be independent of t yield

$$\dot{A} \propto A\frac{\dot{B}}{B} \propto tAB^{1-\gamma} \propto \frac{A^2}{B^{1+\lambda}}. \tag{27}$$

Again, the power functions $A \propto t^{-\xi}$, $B \propto t^{-\eta}$ meet conditions (27) if

$$\xi = \frac{3+2\lambda-\gamma}{1-\gamma}, \quad \eta = \frac{2}{1-\gamma}. \tag{28}$$

Using Eqs. (15) and (28) allows for deriving the condition for the mass concentration not to grow very fast,

$$M(t) = \frac{A}{B^2}\int_0^\infty x\psi(x)dx \propto t^s, \tag{29}$$

where

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

Condition (11) $0 < s < 1$ gives two simultaneous restrictions,

$$\gamma < \lambda$$

and

$$\gamma > 2\lambda - 1. \tag{30}$$

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

$$-\frac{3+2\lambda-\gamma}{1-\gamma}\psi - \frac{2}{1-\gamma}x\psi' + \frac{d}{dx}x^\gamma\psi = (K\psi\psi)_x. \tag{31}$$

On multiplying this equation by x and integrating over all x give

$$\phi_1 = \frac{1-\gamma}{1-2\lambda+\gamma}\phi_\gamma. \tag{32}$$

(c) Case (iii). At $\gamma \leqslant 2\lambda - 1$ the condensation process is slow, and only a finite part of the condensable substance converts to the disperse phase. This means that the condensation process becomes ineffective at large time and can thus be ignored.

If the total mass concentration of the coagulating system does not change with time, the functions $A(t)$ and $B(t)$ are linked by the relation

$$A(t) = M_\infty B^2(t), \tag{33}$$

where [see Eq. (7)]

$$M_\infty = M_0 + \int_0^\infty C(t)\varphi_\gamma(t)dt. \tag{34}$$

Let us return to Eq. (27), which gives now $\dot{B} = -\kappa B^{2-\lambda}$ or

$$B(t) = [(1-\lambda)\kappa M_\infty t]^{-1/(1-\lambda)}, \tag{35}$$

where κ is a separation constant.

We can now find the condition at which it is possible to ignore the condensation term [the third term in Eq. (27)]. To this end we have to compare the term $\dot{A} \propto B\dot{B} \propto t^{-(3-\lambda)/(1-\lambda)}$ with $tAB^{1-\gamma} \propto t^{-(2-\gamma+\lambda)/(1-\lambda)}$. The latter expression should drop down faster than the former, or $3-\lambda < 2-\gamma+\lambda$. This condition is equivalent to

$$\gamma < 2\lambda - 1. \tag{36}$$

The equation for the universal function $\psi(x)$ describing the asymptotic mass spectra in this case has the form

$$-2\psi - x\psi' = \kappa M_\infty (K\psi\psi)_x. \tag{37}$$

The factor κM_∞ can be removed by a redefinition of the function ψ and then found by using the normalization condition $\int_0^\infty x\psi(x)dx = 1$ that fixes the value of the separation constant κ.

It is seen that if $\psi(x)$ is a solution to Eq. (37), then the function

$$\psi_1(x) = \frac{1}{x_0^{1+\lambda}}\psi\left(\frac{x}{x_0}\right) \tag{38}$$

also meets this equation, with x_0 being an arbitrary scale whose value is finally fixed by the condition (22).

At $\gamma = 2\lambda - 1$ the mass concentration is expected to grow with time as a power of logarithm, $M \propto \ln^\sigma t$. The mass distribution is then sought in the form

$$\mathcal{N}(g,t) = D\ln^\sigma t B^2(t)\psi(gB(t)). \tag{39}$$

The term responsible for condensation can still be neglected (it will be shown a bit later). Then the power balance in the equation

$$\ln^\sigma t B\dot{B} \propto \ln^{2\sigma} t B^{3-\lambda} \tag{40}$$

gives within the logarithmic accuracy,

$$B(t) \propto (t\ln^\sigma t)^{1/(1-\lambda)}. \tag{41}$$

The exponent σ is readily found from Eq. (7),

$$C\phi_\gamma(t) \propto t\ln^\sigma t B^{2(1-\lambda)} \propto \frac{1}{t\ln^\sigma t}. \tag{42}$$

Then, integrating Eq. (7) gives $M \propto \ln^{1-\sigma} t$. On the other hand, we assumed $M(t) \propto \ln^\sigma t$. Hence, $\sigma = 1/2$ and

$$B(t) \propto (t\sqrt{\ln t})^{-1/(1-\lambda)}. \tag{43}$$

The mass concentration grows as

$$M(t) \propto \sqrt{\ln(t)}. \tag{44}$$

We are now ready to show that the condensation term does not contribute asymptotically and can be thus ignored. To this end we find the ratio $B\dot{B}/AB^{(1-\gamma)}$. Taking into account that $A = B^2$ and $\gamma = 2\lambda - 1$, we find that this ratio grows with time as $\ln t$.

(d) Case (iv). At $\gamma = \lambda$ both, the vapor and the mass concentrations grow linearly with t. On substituting $C = at$ into

Eq. (4) now gives the same balance of powers as in the case (ii) [which coincides now with the result Eq. (20)], and the equation for the universal function ψ,

$$-\frac{3+\lambda}{1-\lambda}\psi-\frac{2}{1-\lambda}x\psi'+a(x^{\gamma}\psi)'=(K\psi\psi)_x. \tag{45}$$

Instead of Eq. (22), however, we find

$$\phi_1=a\phi_{\gamma}. \tag{46}$$

The mass also grows linearly as prescribed by Eq. (13).

IV. EXACTLY SOLUBLE MODELS

Here we analyze two exactly soluble models of the condensation-coagulation process: (i) $K(x,y)=1$, $\lambda=0$, $\gamma=0$, (ii) $K(x,y)=1$, $\lambda=0$, $\gamma=1$.

A. Model $K=1$, $\gamma=0$

We analyze first the model $K=1,\gamma=0$ and restrict ourselves by considering the exact self-preserving spectrum whose form is easily conjectured,

$$\mathcal{N}(g,t)=A(t)e^{-gB(t)}. \tag{47}$$

Substituting such $\mathcal{N}(g,t)$ into Eqs. (1) and (4) yields the set of equations for C, A, and B,

$$\dot{C}=1-\frac{CA}{B},\quad \dot{A}=CAB-\frac{A^2}{B},\quad \dot{B}=-\frac{1}{2}A. \tag{48}$$

The initial conditions to these equations are: $A(0)=A_0$, $B(0)=B_0$, $C(0)=0$. Set (48) has the integral

$$C(t)+\frac{A(t)}{B^2(t)}=t+M_0, \tag{49}$$

where $M_0=A_0/B_0^2$. This is just the mass conservation: $C+\int_0^\infty g\mathcal{N}(g,t)dg=t+t_0$. This integral allows one to find the asymptotic powerlike solution to set (48),

$$A(t)\approx\frac{20}{t^3},\quad B(t)\approx\frac{5}{t^2}. \tag{50}$$

Numerical analysis of the solution of these equations confirms the asymptotics Eq. (50). The time dependencies of the vapor concentration $C(t)$, particle number concentration $N(t)$, and particle-mass concentration $M(t)$ are shown in Figs. 1(a)–1(c) (curves A) as the functions of time.

The universal function

$$\psi(x)=20e^{-5x} \tag{51}$$

is seen to be normalized to 4/5 rather than to unity, which corresponds to the linear growth of $C(t)$ with time: $C(t)\approx t/5$, i.e., 1/5 of the total mass does not convert to disperse phase. Of course, thus found function $\psi(x)$ meets Eq. (45). The universal function is shown in Fig. 2 (curve A).

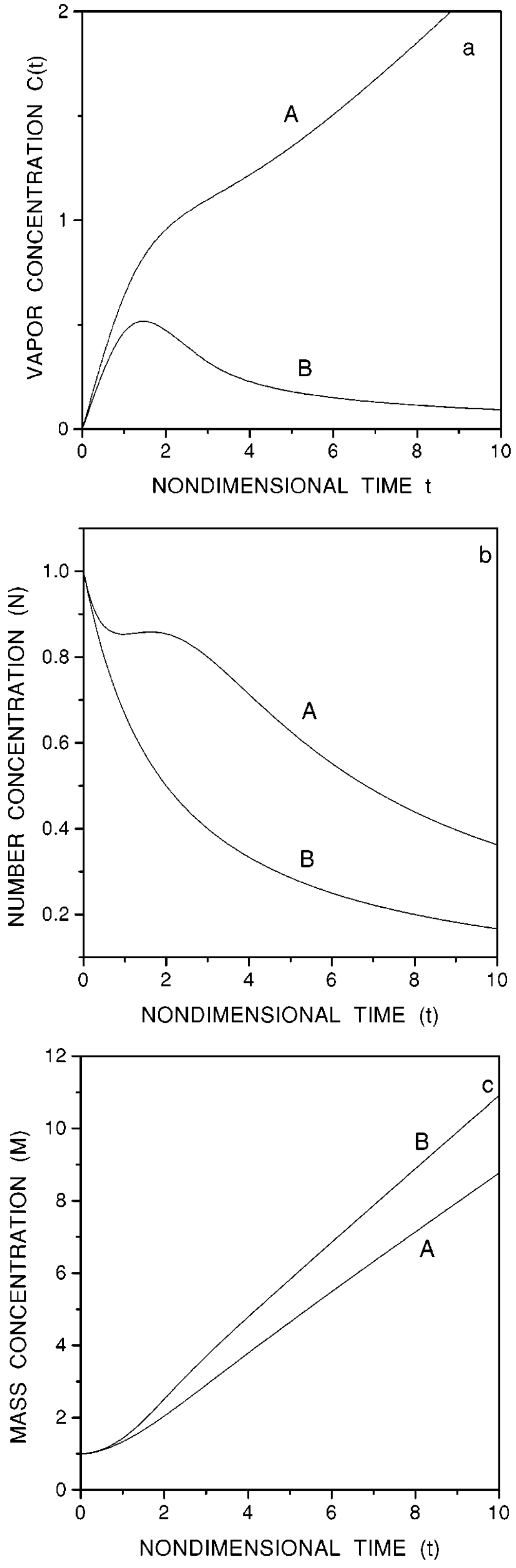

FIG. 1. Time dependencies of vapor concentration (a), particle number concentration (b), and mass concentration (c) for exactly soluble models A and B. The concentrations are given in the units $\sqrt{I/\alpha}$ and time in $\sqrt{1/I\alpha}$.

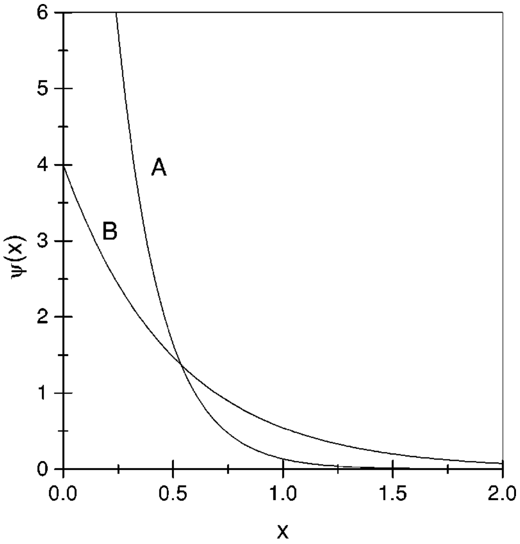

FIG. 2. Universal functions $\psi(x)$ describing the self-preserving mass distributions for two exactly soluble models *A* and *B*. Both axes are nondimensional.

B. Model $K=1,\ \gamma=1$

We again will try to find the particle mass distribution in the exponential form of Eq. (47). The equations for $A(t)$, $B(t)$, and $C(t)$ take the form at $\lambda=0,\ \gamma=1$:

$$\dot{A}+CA=-\frac{A^2}{B},\quad \dot{B}+CB=-\frac{A}{2}, \tag{52}$$

$$\dot{C}=1-C(t+M_0-C). \tag{53}$$

Equation (53) has the asymptotics $C(t)\approx 1/t$. On substituting $A\approx a/t^{\xi}$ and $B\approx b/t^{\eta}$ into Eq. (52) we find $\xi=3$ and $\eta=2$, in accordance with Eq. (20). Two equations for *a* and *b* give only one relation between these values $a=2b$. The constant *a* is defined from the condition $M(t)\approx t$ at large *t* or $A/B^2\approx t$. This condition gives $a=4$ and $b=2$, or

$$\psi(x)=4e^{-2x}. \tag{54}$$

Now let us try to reproduce this result using the asymptotic theory. Equation (21) takes the form

$$-2\psi-x\psi'=\frac{1}{2}\int_0^x\psi(x-y)\psi(y)dy-\psi(x)\int_0^{\infty}\psi(y)dy. \tag{55}$$

The attempt to find the solution to this equation in the form $\psi(x)=ae^{-bx}$ gives the result Eq. (54).

Figures 1(a)–1(c) show the time dependencies of the functions $C(t)$, $N(t)$, and $M(t)$ for this case (curves *B*). In contrast to the just considered model the vapor condenses much faster, and its concentration drops as reciprocal time at large *t* [Fig. 1(a)]. The mass is seen to grow linearly with time. The function $\psi(x)$ for this case is shown in Fig. 2 (curve *B*).

V. RESULTS AND DISCUSSION

The main results of this paper may be summarized as follows.

(1) We have shown that the self-preserving regimes of coagulation condensation can exist even in source-enhanced systems. The mass spectrum of the disperse phase at the asymptotic stage has the form

$$\mathcal{N}(g,t)=A(t)\psi(gB(t)). \tag{56}$$

The function $\psi(x)$ defining the shape of the self-preserving mass spectrum is the solution of Eqs. (21), (31), (37), and (45). Since the function $\psi(x)$ was assumed to be integrable at $x=0$ (nonsingular distribution), it is possible to express the functions $A(t)$ and $B(t)$ in terms of the number $N(t)$ and the mass $M(t)$ concentrations of disperse particles. Indeed, since $A(t)=M(t)B^2(t)$ and $B(t)=N(t)/[M(t)\phi_0]$, the asymptotic mass spectrum can be rewritten as

$$\mathcal{N}(g,t)=\frac{N^2(t)}{M(t)\phi_0^2}\psi\left(\frac{gN(t)}{\phi_0 M(t)}\right). \tag{57}$$

Equation (57) is the Friedlander form of the self-preserving solutions of the coagulation kinetic equation widely applied for analyzing the asymptotic behavior of free coagulating systems (see [2–4]). Our result Eq. (57), in addition, includes the time dependence of the particle-mass concentration coming from the accompanying condensation process. Of course, the equations defining the shape of the self-preserving mass spectra are different from the case of free coagulation, except for Eq. (37) which describes coagulation in the system with finite mass concentration.

(2) We have considered four possible regimes of coagulation-condensation.

(i) The disperse particles consume (asymptotically) all condensable vapor and their mass grows linearly with time. The coagulation process is so slow that the vapor condensation is efficient enough despite the coagulation diminishing the particle number concentration, average surface, or other moments responsible for the rate of condensation process. The asymptotic time dependencies of mass and number concentrations of disperse phase and the vapor concentration are

$$M(t)\approx t,\quad N(t)\propto t^{-(1+\lambda)/(1-\lambda)},$$
$$C(t)\propto t^{(1+\lambda-2\gamma)/(1-\lambda)}. \tag{58}$$

The condition for the realization of this case is $\lambda<\gamma$, $\gamma>2\lambda-1$.

(ii) In the opposite case, when condensation goes slower than coagulation, the mass of disperse fraction grows slower than *t*, i.e., the coagulating disperse phase becomes a noneffective sink for the condensable matter. In this case

$$M(t)\propto t^{(1-2\lambda+\gamma)/(1-\gamma)},$$
$$N(t)\propto t^{-(1+2\lambda-\gamma)/(1-\gamma)},\quad C(t)\approx t. \tag{59}$$

The condition for the realization of this situation is $\lambda>\gamma$.

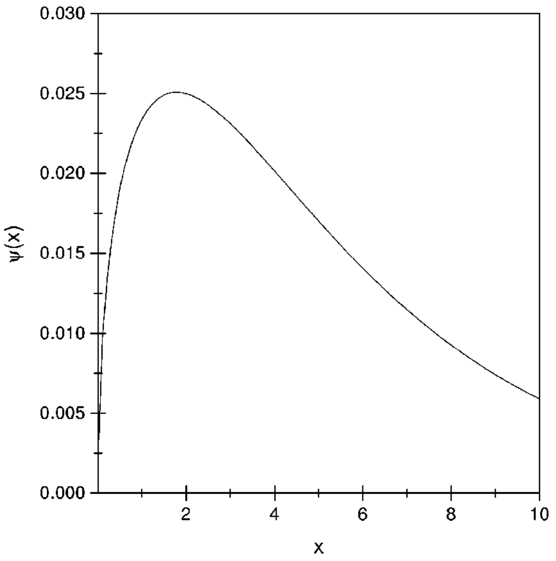

FIG. 3. Universal function $\psi(x)$ describing the self-preserving mass distribution of disperse particles growing by simultaneous coagulation and condensation in free molecular regime. Both axes are nondimensional.

(iii) Coagulation goes so swiftly that only a *finite* part of the vapor mass is able to deposit onto the particle surfaces. In this case the coagulation process goes like in free systems, i.e.,

$$M(t) = \text{const}, \quad N(t) \propto t^{1/(1-\lambda)}, \quad C(t) \approx t. \tag{60}$$

This happens at $\gamma < 2\lambda - 1$. At $\gamma = 2\lambda - 1$ the mass concentration grows logarithmically with time. Although the equation for ψ still retains the form (37), the dependencies of M, N, and C on t are different and look as follows:

$$M(t) \propto \sqrt{\ln t}, \quad N(t) \propto (t \ln^{\lambda/2} t)^{-1/(1-\lambda)},$$

$$C(t) \propto \frac{1}{t\sqrt{\ln t}}. \tag{61}$$

(iv) At $\gamma = \lambda$ the disperse and vapor phases share the mass of vapor from the source and grow linearly with time. In this case

$$M(t) \propto t, \quad N(t) \propto t^{\{(1+\lambda)/1-\lambda\}}, \quad C(t) \propto t. \tag{62}$$

The models of preceding section correspond to the cases (iv) and (i), respectively.

We conclude this section by demonstrating the numerically found self-preserving mass spectrum for the free molecular regime of aerosol particle growth (when the mean free path of the carrier gas molecules exceeds the particle size). In this case

$$K_1 = (g^{1/3} + l^{1/3})^2 \sqrt{\frac{1}{g} + \frac{1}{l}}, \quad \lambda = 1/6, \quad \gamma = 2/3. \tag{63}$$

Although this coagulation kernel is an idealization (its homogeneity is broken due to the van der Waals attraction of colliding particles) the simple expression Eq. (64) is widely used for practical needs (see, e.g., Ref. [23] and references therein).

The result of calculations is shown in Fig. 3. The self-similarity distribution has a humplike form with no singularity at $x = 0$ that allows one to find the asymptotic time dependencies of the moments,

$$N(t) \propto t^{-7/5}, \quad \phi_{1/3}(t) \propto t^{-11/5}, \quad \phi_{2/3}(t) = t^{1/5}. \tag{64}$$

VI. CONCLUSION

Coagulation processes in condensing systems display a number of self-preserving regimes. These regimes are attributed to the asymptotic stage of evolution of the disperse systems when they ‘‘forget’’ the initial conditions and is characterized by some self-established scales.

The simplifications resulting from the application of the self-preservation hypothesis are enormous. In many cases the self-preserving asymptotic solutions even replace the straightforward numerical analysis, when the latter becomes powerless to give conclusive results on the evolution of coagulating systems at large times. The fact that the self-preserving regimes are realizable in describing the deep stages of evolution of coagulating-condensing systems is far from being self-evident.

From the practical point of view the free molecular regime of particle growth is of most interest, because all aerosol technologies for nanoparticle production deal with the aerosol particles in free molecular regime. In this case the asymptotic mass spectrum is a nonsingular humplike function, which, in principle, can be approximated by a log-normal distribution.

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