Proof of Dynamical Scaling in Smoluchowski's Coagulation Equation with Constant Kernel

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Abstract

Smoluchowski's coagulation equation for irreversible aggregation with constant kernel is considered in its discrete version

$$\dot{c}_\ell \;=\; \sum_{k=1}^{\ell-1} c_{\ell-k} c_k \;-\; 2c_\ell \sum_{k=1}^{\infty} c_k$$

where $c_\ell = c_\ell(t)$ is the concentration of ℓ-particle clusters at time t. We prove that for initial data satisfying $c_1(0) > 0$ and the condition $0 \leq c_\ell(0) < A(1+\Delta)^{-\ell}$ $(A, \Delta > 0)$, the solutions behave asymptotically like $c_\ell(t) \sim t^{-2}\tilde{c}(\ell t^{-1})$ as $t \to \infty$ with ℓt^{-1} kept fixed. The scaling function $\tilde{c}(\xi)$ is $\frac{1}{\rho}\exp\left(-\frac{1}{\rho}\xi\right)$ where $\rho = \sum_\ell \ell c_\ell(0)$, a conserved quantity, is the initial number of particles per unit volume . An analogous result is obtained for the continuous version of Smoluchowski's coagulation equation

$$\frac{\partial}{\partial t} c(v,t) \;=\; \int_0^v du\; c(v-u,t)c(u,t) \;-\; 2c(v,t)\int_0^\infty du\; c(u,t)$$

where $c(v,t)$ is the concentration of clusters of size v.

Keywords: *Smoluchowski's coagulation equations, dynamical scaling, cluster growth, kinetics of first-order phase transitions*

1 Introduction

In the theory of phase separation processes, cluster dynamics is based on kinetic equations which model the detailed mechanisms leading to the formation and evolution of clusters of molecules ([1], [2]). These are equations for the time evolution of variables $c_\ell(t)$ representing the average concentrations of minority-phase clusters of size ℓ at time t, which neglect some of the complications of real systems, such as grains, boundaries and vacancies.

The kinetic equation of Smoluchowski ([3], [4])

$$\dot{c}_\ell \;=\; \sum_{k=1}^{\ell-1} a_{\ell-k,k} c_{\ell-k} c_k \;-\; 2\sum_{k=1}^{\infty} a_{\ell,k} c_\ell c_k \tag{1}$$

models a process where two clusters of sizes k and ℓ can coagulate to form a cluster of size $k+\ell$, with a probability proportional to $a_{k,\ell} = a_{\ell,k} > 0$. In this model there is no fragmentation of clusters. This infinite system of nonlinear ordinary differential equations is of interest in polymer science as a simple model for polymerization ([5]), and also in aerosol physics ([6]). As the equations (3) conserve the mass density $\rho = \sum_\ell \ell c_\ell(t)$ for all finite times if the coagulation rates $a_{k,\ell}$ do not grow too rapidly as $k, \ell \to \infty$ (see, for example [7]), the model considered here can be classified as model B in the Hohenberg-Halperin scheme of dynamical phase transitions ([8]).

The hypothesis of *dynamical scaling* ([1], [2], [9], [10]), asserts that after a sufficiently long time t the cluster distribution $c_\ell(t)$ for the large clusters becomes independent of the initial distribution and approaches a distribution which on suitable time and length scales has a self-similar profile , i.e.

$$c_\ell(t) \;\simeq\; t^y \tilde{c}(\ell t^{-x}) \quad , \quad \ell \to \infty \;,\; t \to \infty \tag{2}$$

where x and y are constants and $\tilde{c}(\cdot)$ is the so-called dynamical scaling function. This hypothesis is widely used for cluster models with a conserved order parameter. For aerosols, such as tobacco smoke, the hypothesis (2) is also known as Friedlander's self-similarity ansatz (see for example [6], [11]).

Although there exists experimental confirmation of the scaling hypothesis (2) (e.g. [12]) and evidence from computational and formal studies ([1], [5], [6], [9] - [11], [13] - [16]) not very much is known from the rigorous point of view ([6]). Even though the conjecture of a self-similar cluster distribution (2) described only a few macroscopic parameters (such as the mass density for example) was formulated over 50 years ago by Schuhmann ([13]), it has not been derived rigorously from Smoluchowski's kinetic equation (1).

In this paper we give such a derivation for the special case where the coefficients $a_{k,\ell}$ are independent of k and ℓ. On a time scale chosen to make $a_{k,\ell} = 1$, equation (1) becomes

$$\dot{c}_\ell \;=\; \sum_{k=1}^{\ell-1} c_{\ell-k} c_k \;-\; 2c_\ell \sum_{k=1}^{\infty} c_k \tag{3}$$

In this investigation we prove both for (3) and its continuous analogue

$$\frac{\partial}{\partial t} c(v,t) \;=\; \int_0^v du\, c(v-u,t)c(u,t) \;-\; 2c(v,t) \int_0^\infty du\, c(u,t) \tag{4}$$

that equation (2) holds if the initial cluster distribution satisfies some mild conditions. Our results can be stated as follows:

$$\begin{aligned} \lim_{\ell,t\to\infty\ ,\ \ell/t=\xi} t^2 c_\ell(t) &= \tilde{c}(\xi) \quad \text{for (3)} \\ \lim_{v,t\to\infty\ ,\ v/t=\xi} t^2 c(v,t) &= \tilde{c}(\xi) \quad \text{for (4)} \end{aligned}$$

with the dynamical scaling function $\tilde{c}$ given by

$$\tilde{c}(\xi) \;=\; \frac{1}{\rho} \exp\left(-\frac{1}{\rho}\xi\right). \tag{5}$$

2 The Discrete Case

We first collect some properties of the solution of (3) which are known from the work of [6], [7] and [17].

Proposition I *Let the initial data for eqn (3)have finite second moment, i.e.* $M_2(0) = \sum_{\ell=1}^\infty \ell^2 c_\ell(0) < \infty$. *Then this equation has a unique solution, which is positive, and the second moment* $M_2(t) = \sum_{\ell=1}^\infty \ell^2 c_\ell(t)$ *is bounded on bounded intervals of the positive* t*-axis. Moreover the series*

$$n(t) \;=\; \sum_{\ell=1}^{\infty} c_\ell(t) \tag{6}$$

$$\rho(t) = \sum_{\ell=1}^{\infty} \ell c_\ell(t) \tag{7}$$

$$\Phi(\zeta,t) = \sum_{\ell=1}^{\infty} \zeta^\ell c_\ell(t) \tag{8}$$

where ζ is any complex number satisfying $|\zeta| \leq 1$, converge for all t and satisfy the equations

$$\frac{d}{dt}\, n(t) = -[n(t)]^2 \tag{9}$$

$$\frac{d}{dt}\, \rho(t) = 0 \tag{10}$$

$$\frac{\partial}{\partial t}\Phi(\zeta,t) = [\Phi(\zeta,t)]^2 - 2\Phi(\zeta,t)n(t) \tag{11}$$

Proof:

The existence, uniqueness, positivity and absolute continuity of the solutions of (3) are proven e.g. in [7]. As shown in [17] these solutions have the following finite moment property for any $p > 1$:

$$0 \leq \sum_{\ell=1}^{\infty} \ell^p c_\ell(0) < \infty \quad \Rightarrow \quad 0 \leq \sum_{\ell=1}^{\infty} \ell^p c_\ell(t) \leq K_p(t) < \infty \tag{12}$$

where the function $K_p(t)$ is bounded on any bounded interval of the t-axis. With p set equal to 2, this property implies the desired result about $M_2(t)$, and also gives the following bound on the $c_\ell(t)$:

$$0 \leq c_\ell(t) \leq \frac{K_2(t)}{\ell^2} \tag{13}$$

To prove (9) we consider the integrated version of (3)

$$c_\ell(t) = c_\ell(0) + \int_0^t d\tau \sum_{k=1}^{\ell-1} c_{\ell-k}(\tau)c_k(\tau) - 2\int_0^t d\tau c_\ell(\tau) \sum_{k=1}^{\infty} c_k(\tau) \tag{14}$$

Summing (14) over ℓ from 1 to N and taking the limit $N \to \infty$ we obtain, with the help of (13), the continuity of the $c_\ell(\tau)$ and Weierstrass' uniform convergence theorem, the result

$$\begin{aligned} \sum_{\ell=1}^{\infty} c_\ell(t) &= \sum_{\ell=1}^{\infty} c_\ell(0) + \int_0^t d\tau \sum_{\ell=1}^{\infty}\sum_{k=1}^{\ell-1} c_{\ell-k}(\tau)c_k(\tau) - 2\int_0^t d\tau \sum_{\ell=1}^{\infty} c_\ell(\tau) \sum_{k=1}^{\infty} c_k(\tau) \\ &= \sum_{\ell=1}^{\infty} c_\ell(0) - \int_0^t d\tau \left(\sum_{\ell=1}^{\infty} c_\ell(\tau) \right)^2 \end{aligned}$$

After differentiation the first macroscopic equation (9) is obtained, since the integrands are continuous. Eqn (10) can be proved by a similar method; we omit the proof since the same result has already been proved in [7] for more general initial conditions. The other equation (11) can be derived by analogous arguments. Q.E.D.

The following lemma gives information about the location of the roots of the equation $\phi(\zeta) = n_0 + \tau$, which will be needed for the main theorem.

Lemma II *Let A, Δ be any positive numbers and let $c_1(0), c_2(0), ...$ be a sequence satisfying $0 < c_1(0),\ 0 \leq c_k(0) \leq A(1+\Delta)^{-k}$ $(k = 1, 2, ...)$. Furthermore set $n_0 = \sum_{k=1}^{\infty} c_k(0)$ and $\rho = \sum_{k=1}^{\infty} kc_k(0)$ and define*

$$\phi(\zeta) = \Phi(\zeta, 0) = \sum_{k=1}^{\infty} \zeta^k c_k(0) \quad , \quad |\zeta| < 1 + \Delta$$

Define $\tau_1 = \phi(1+\Delta) - n_0$, which is either a positive number or $+\infty$. Then the following statement holds true for the roots of the equation $\phi(\zeta) = n_0 + \tau$ for given τ :

1. *For all $\tau \in [0, \tau_1)$ there is a simple root, call it $\zeta_1(\tau)$, satisfying*

$$1 \leq \zeta_1(\tau) = 1 + \frac{1}{\rho}\tau + R^{(2)}(\tau) < 1 + \Delta \tag{15}$$

   where $R^{(2)}(\tau) = O(\tau^2)$ as $\tau \to 0$.

2. *All other roots $\zeta_m(\tau)$ $(m = 2, ...)$ lie outside the closed unit disk and are uniformly bounded away from it as $\tau \to 0$; that is to say, there exist $\tau_2 \in (0, \tau_1)$ and $\delta \in (0, \Delta)$ such that $|\zeta_m(\tau)| > 1 + \delta$ $(m = 2, 3, ...)$ for $\tau \in [0, \tau_2]$.*

Proof:

Define the function $\phi(\zeta) = \sum_{k=1}^{\infty} c_k(0)\zeta^k$, analytic in $|\zeta| < 1 + \Delta$. As ζ increases from 1 to $1 + \Delta$ the value of $\phi(\zeta)$ increases from n_0 to $n_0 + \tau_1$; so for each $\tau \in [0, \tau_1)$ the equation $\phi(\zeta) = n_0 + \tau$ has a unique positive solution, call it $\zeta_1(\tau)$, which is monotone increasing in τ.

Since the function $\phi(\zeta)$ is analytic in a neighbourhood of $\zeta = 1$ we can apply the implicit function theorem, obtaining, since $0 = d[n_0 + \tau - \phi(\zeta)]/d\tau|_{\tau=0} = 1 - \phi'(1)d\zeta/d\tau|_{\tau=0} = 1 - \rho d\zeta/d\tau|_{\tau=0}$,

$$\zeta_1(\tau) = 1 + \frac{1}{\rho}\tau + R^{(2)}(\tau)$$

where $R^{(2)}(\tau) = O(\tau^2)$ for $\tau \to 0$.

For the other roots $\zeta_m(\tau)$, $\quad (m = 2, ...)$ the argument is as follows: since the function $\phi(\zeta) - n_0$ is analytic in the open disk $|\zeta| < 1 + \Delta$, it has only a finite number of zeros inside the smaller disk $|\zeta| < 1 + \Delta/2$. Moreover, the only zero inside the closed unit disk is the one at $\zeta = 1$, since $\zeta \leq 1$ implies $|\phi(\zeta)| = |\sum_l c_l \zeta^l| \leq \sum_l c_l |\zeta|^l \leq \sum_l c_l = n_0$, with equality if and only if $\zeta = 1$. The rest of these zeros, which in the notation of the theorem are $\zeta_m(0)$ $(m = 2, 3, ...)$, lie in the annulus $1 < |\zeta| < 1 + \Delta/2$. Let the smallest of their moduli be $1 + 2\delta$. As τ increases from 0 these zeros move continuously (e.g. [18]) and so for sufficiently small τ none of their moduli can be less than $1 + \delta$. Q.E.D.

Now we can state our main result for the discrete case in the folowing theorem:

Theorem III *For all initial configurations satisfying the two conditions*

(i) $0 < c_1(0)$

(ii) $\exists A, \Delta > 0 \ : \ 0 \leq c_\ell(0) \leq \frac{A}{(1+\Delta)^\ell}$ $(\ell = 1, 2, ...)$

the solution to (3) has the following asymptotic behaviour as $t \to \infty$ *and* $\ell \to \infty$ *with* $\ell/t = \xi > 0$ *fixed*

$$\lim_{\ell, t \to \infty, \ell/t = \xi} t^2 c_\ell(t) \quad = \quad \tilde{c}(\xi) \tag{16}$$

where the dynamical scaling function $\tilde{c}(\cdot) \ : \ I\!R^+ \to I\!R^+$ *is given by*

$$\tilde{c}(\xi) \ = \ \frac{1}{\rho} e^{-\frac{1}{\rho}\xi} \tag{17}$$

The convergence in (16) is uniform on compact intervals of the positive ξ*-axis.*

Proof:

The initial conditions guarantee that Proposition I can be applied. Solving the macroscopic rate equations (9) and (10) with initial conditions $n(0) = n_0$ and $\rho(0) = \rho$ we obtain

$$n(t) = \frac{1}{n_0^{-1} + t}, \qquad \rho(t) = \rho \tag{18}$$

The equation (11) for the generating function for $|\zeta| < 1$ can also be solved explicitly subject to its initial condition $\Phi(\zeta,0) = \phi(\zeta) = \sum_{\ell=1}^{\infty} \zeta^\ell c_\ell(0)$ ([5], [6]), giving

$$\Phi(\zeta,t) = t^{-2}\frac{1}{(n_0+t^{-1})}\,\frac{\phi(\zeta)}{n_0+t^{-1}-\phi(\zeta)} \quad , \quad t>0 \;,\; |\zeta|<1 \tag{19}$$

By Proposition I, $\Phi(\zeta,t)$ is analytic in the unit disk of the ζ plane, and so by Cauchy's integral formula and (8) we can express the solution of (3) as

$$t^2 c_\ell(t) = \frac{1}{2\pi i}\,\frac{1}{(n_0+\tau)}\oint_{\Gamma_0}\frac{d\zeta}{\zeta^{\ell+1}}\quad\frac{\phi(\zeta)}{n_0+\tau-\phi(\zeta)} \tag{20}$$

where τ means t^{-1} and Γ_0 denotes the circle $\{\zeta : |\zeta| = r_0\}$ for some r_0 satisfying $0 < r_0 < 1$.

By condition (ii), $\phi(\zeta)$ is analytic in the disk $|\zeta| < 1+\Delta$, and hence, by (19) $\Phi(\zeta,t)$ is analytic in this disk except for poles at the zeros of the denominator. Defining $\zeta_1(\tau)$, τ_2 and δ as in Lemma II and setting $\tau_3 = \phi(1+\delta/4) - n_0 > 0$, we can chose a positive number $\tau_4 = \min\{\tau_2,\tau_3\}$ such that $1 \le \zeta_1(\tau) < 1+\delta/2$ for $\tau \in [0,\tau_4]$. Hence we see that for $\tau \in [0,\tau_4]$ the only such pole inside the circle $\Gamma_1 = \{\zeta : |\zeta| = 1+\delta/2\}$ is at $\zeta_1(\tau)$. Deforming the contour of integration in (20) we obtain

$$\oint_{\Gamma_0}\frac{d\zeta}{\zeta^{\ell+1}}\,\frac{\phi(\zeta)}{n_0+\tau-\phi(\zeta)} \;=\; \oint_{\Gamma_1}\frac{d\zeta}{\zeta^{\ell+1}}\,\frac{\phi(\zeta)}{n_0+\tau-\phi(\zeta)} \;-\; \oint_{\Gamma_2}\frac{d\zeta}{\zeta^{\ell+1}}\,\frac{\phi(\zeta)}{n_0+\tau-\phi(\zeta)} \tag{21}$$

where $\Gamma_2 = \{\zeta : |\zeta - \zeta_1(\tau)| = \delta/4\}$ lies inside the circle Γ_1 for $\tau \in [0,\tau_4]$.

To estimate the first integral on the right we define the function $f(\zeta,\tau) = |n_0+\tau-\phi(\zeta)|$. By Lemma II this function is positive throughout the closed set $\{\zeta,\tau : |\zeta| = 1+\delta/2, 0 \le \tau \le \tau_4\}$ because for $\tau \in [0,\tau_4]$ the only root of f inside the circle Γ_1 is stricly less than $1+\delta/2$. Since the function f is continuous its infimum in this domain must be attained; and this infimum, which we denote by f_{min}, is therefore positive:

$$\min_{\tau\in[0,\tau_4]}\;\min_{|\zeta|=1+\delta/2}|n_0+\tau-\phi(\zeta)| = f_{min} > 0 \tag{22}$$

Using this result and condition (ii) in (21) we obtain, since $\delta < \Delta$,

$$\begin{aligned}\left|\oint_{\Gamma_1}\frac{d\zeta}{\zeta^{\ell+1}}\,\frac{\phi(\zeta)}{n_0+\tau-\phi(\zeta)}\right| &\le \frac{2\pi}{(1+\delta)^\ell}\frac{1}{f_{min}}\sum_{k=1}^{\infty}A\left(\frac{1+\delta}{1+\Delta}\right)^k \\ &= \frac{2\pi A}{(1+\delta)^\ell}\frac{1}{f_{min}}\frac{1+\delta}{\Delta-\delta}\end{aligned} \tag{23}$$

provided that $0 \leq t^{-1} = \tau = \xi/\ell \leq \tau_4$. If ξ is restricted to some interval $[\xi_1, \xi_2]$, then (23) holds throughout that interval as soon as $\ell \geq \xi_2/\tau_4$, and so the integral approaches 0 as $\ell, t \to \infty$, uniformly on this set of values for ξ.

The second contour integral can be evaluated by the residue theorem using l'Hôpital's rule and equation (15) of Lemma II:

$$
\begin{aligned}
\frac{1}{2\pi i} \oint_{\Gamma_2} \frac{d\zeta}{\zeta^{\ell+1}} \frac{\phi(\zeta)}{n_0 + \tau - \phi(\zeta)} &= \mathrm{Res}_{\zeta=\zeta_1(\tau)} \left[\frac{1}{\zeta^{\ell+1}} \frac{\phi(\zeta)}{n_0 + \tau - \phi(\zeta)} \right] \\
&= \lim_{\zeta \to \zeta_1(\tau)} \left[\frac{(\zeta - \zeta_1(\tau))}{\zeta^{\ell+1}} \frac{\phi(\zeta)}{n_0 + \tau - \phi(\zeta)} \right] \\
&= \frac{1}{[\zeta_1(\tau)]^{\ell+1}} \frac{\phi(\zeta_1(\tau))}{-\phi'(\zeta_1(\tau))}
\end{aligned}
\tag{24}
$$

Now the result (15) in Lemma II implies that

$$
\zeta_1(\tau)|_{\tau=\frac{\xi}{\ell}} = 1 + \frac{1}{\rho}\frac{\xi}{\ell} + R^{(2)}(\xi/\ell) \qquad \text{as } \ell \to \infty \text{ with } \xi = \ell\tau \text{ fixed} \tag{25}
$$

where, if $\xi \in [\xi_1, \xi_2]$ and $\ell > \ell_0$ for some positive ℓ_0 then $|R^{(2)}(\xi/\ell)| \leq K(\xi^2/\ell^2)$, where K is a number depending on ℓ_0 and ξ_2 but not on ξ or ℓ. It follows that as $\ell \to \infty$ the right-hand side of (24) approaches the limit

$$
\frac{\phi(1)}{\phi'(1)} \lim_{\ell \to \infty} \left[1 + \frac{1}{\rho}\frac{\xi}{\ell} + R^{(2)}\left(\frac{\xi}{\ell}\right) \right]^{-\ell-1} = \frac{n_0}{\rho} e^{-\frac{1}{\rho}\xi} \tag{26}
$$

and that the converegence is uniform in ξ on the set $[\xi_1, \xi_2]$.

Putting (23) and (26) into (21) and using (20) completes the proof. Q.E.D.

The condition on the exponential decay of the initial cluster distribution and its derivatives is not restrictive from the physical point of view: usually the system does not contain any large clusters at all at time $t = 0$.

That the dynamical scaling as stated in Theorem III does not hold for arbitrary initial data can be seen by the following example. Consider the pure monodisperse initial condition $c_\ell(0) = \delta_{\ell,m}$ for some integer $m > 1$. Then it can be shown by direct computation that the solution for this initial condition is

$$
c_\ell(t) = \begin{cases} t^{-2}(1 + t^{-1})^{-\frac{\ell}{m}-1} & \text{if } \frac{\ell}{m} \in I\!N \\ 0 & \text{otherwise} \end{cases}
$$

For these functions the limiting behaviour stated in Theorem III does not hold. To see this, let ℓ tend to ∞ through a sequence $\{\ell_i\}_{i\in I\!N}$ of integers that are not multiples of m. We have then $c_{\ell_i}(t) = 0$ for all i and therefore $\lim_{i,t\to\infty\ ,\ \ell_i/t=\xi}\ t^2 c_{\ell_i}(t) = 0$. But if ℓ tends to ∞ through a sequence $\{\ell'_i\}_{i\in I\!N}$ that are multiples of m then we obtain a different limit, namely $\exp\left(-\frac{1}{m}\xi\right)$. Thus the limit discussed in Theorem II does not exist in this case. Notice also, that in this case two poles move towards the unit circle as $t \to \infty$.

3 The Continuous Case

As noted in [19], following a suggestion of van Kampen, equations like (4) can be solved by the Laplace transform method. In the next proposition we show that the Laplace transform method can be applied rigorously. Proposition IV for the continuous version (4), which makes extensive use of previous work of [20] and [21], is the analogue to Proposition I for the discrete case.

Proposition IV *Let $c(v,0)$ $(v \geq 0)$ be a continuous, non-negative, bounded and integrable function such that*

$$n(0) = \int_0^\infty dv\ c(v,0) \tag{27}$$

$$\rho = \int_0^\infty dv\ vc(v,0) \tag{28}$$

Then equation (4) has a unique solution $c(v,t)$, which is continuous, non-negative, bounded, integrable in v for each t, and analytic in t for each v. Furthermore the following integrals

$$n(t) = \int_0^\infty dv\ c(v,t) \tag{29}$$

$$\rho(t) = \int_0^\infty dv\ vc(v,t) \tag{30}$$

$$\Phi(s,t) = \int_0^\infty dv\ e^{-sv} c(v,t) \tag{31}$$

where s is any complex number satisfying $Re(s) \geq 0$, all converge and satisfy the equations

$$\frac{d}{dt}\, n(t) = -[n(t)]^2 \tag{32}$$

$$\frac{d}{dt}\,\rho(t) \quad = \quad 0 \tag{33}$$

$$\frac{\partial}{\partial t}\Phi(s,t) \quad = \quad [\Phi(s,t)]^2 - 2\Phi(s,t)n(t) \tag{34}$$

Proof:

The existence and uniqueness of a solution $c(v,t)$, which is continuous, non-negative, bounded, integrable in v for each t, and analytic in t for each v was first proven in [20]. That the solution conserves density, i.e. that equation (33) holds, is proven e.g. in [21]. By boundedness we conclude that there exists a positive constant K such that

$$0 \le c(v,t) < K \qquad v \ge 0\ ,\ t \ge 0 \tag{35}$$

The particle number $n(t)$ definied in (29) can be bounded with help of (35) and the density conservation as follows, keeping the non-negativity of the solution in mind,

$$0 \ \le\ n(t) = \Phi(0,t) \ =\ \int_0^\infty dv\; c(v,t) \ \le\ \int_0^1 dv\; c(v,t) + \int_1^\infty dv\; vc(v,t) \ \le\ K + \rho \tag{36}$$

For any $t > 0$ let us consider the integrated version of (4)

$$c(v,t) \ =\ c(v,0) + \int_0^t d\tau \int_0^v du\; c(v-u,\tau)c(u,\tau) - 2\int_0^t d\tau\; c(v,\tau) \int_0^\infty du\; c(u,\tau) \tag{37}$$

Multiplying equation (37) by e^{-sv}, where $\mathrm{Re}(s) \ge 0$, integrating over v from 0 to ∞ and using (29) we obtain

$$\begin{aligned}\int_0^\infty dv\; e^{-sv}c(v,t) \quad = \quad \int_0^\infty dv\; e^{-sv}c(v,0) \ &+\ \int_0^\infty dv\; e^{-sv}\int_0^t d\tau \int_0^v du\; c(v-u,\tau)c(u,\tau) \\ &-\ 2\int_0^\infty dv\; e^{-sv}\int_0^t d\tau\; c(v,\tau)n(\tau)\end{aligned} \tag{38}$$

where the integrals exists by the bound (35) and the fact that $n(\tau)$ is bounded by (36). We notice that for $\mathrm{Re}(s) \ge 0$ (36) implies

$$\int_0^t d\tau \int_0^\infty dv\; \left|e^{-sv}c(v,\tau)n(\tau)\right| \ \le\ \int_0^t d\tau\; [n(\tau)]^2 \ <\ \infty \tag{39}$$

$$\int_0^t d\tau \int_0^\infty du\; \left|e^{-su}c(u,\tau)\right| \int_u^\infty du\; \left|e^{-s(v-u)}c(v-u,\tau)\right| \ \le\ \int_0^t d\tau\; [n(\tau)]^2 \ <\ \infty \tag{40}$$

These guarantee that Tonelli's theorem can be applied to equation (38) so that the orders of integration can be interchanged to obtain with the help of definition (31)

$$\Phi(s,t) \;=\; \Phi(s,0) + \int_0^t d\tau\, [\Phi(s,\tau)]^2 - 2\int_0^t d\tau\, [\Phi(s,\tau)]n(\tau) \tag{41}$$

In particular when $s=0$ this gives, by (29)

$$n(t) \;=\; n(0) - \int_0^t d\tau [n(\tau)]^2 \tag{42}$$

Differentiation with respect to t proves equations (32) and (34) since the integrands are continuous. Q.E.D.

The following lemma is the analogue of Lemma II. It provides information about the location of the roots of the equation $\psi(s) = n_0 + \tau$, and about the behaviour of $\psi(s)$ at infinity, which will be needed for the main theorem about the continuous case.

Lemma V *Let the initial data in (4) be twice differentiable with respect to v, the second derivative being of bounded variation, and let them satisfy*

$$|c(v,0)| < Ae^{-\Delta\, v},\left|\frac{\partial}{\partial v}c(v,0)\right| < Ae^{-\Delta\, v} \text{ and } \left|\frac{\partial^2}{\partial^2 v}c(v,0)\right| < Ae^{-\Delta\, v} \qquad (v \in I\!R^+) \tag{43}$$

where A, Δ are positive numbers. Furthermore set $n_0 = \int_0^\infty dv\, c(v,0)$ and $\rho = \int_0^\infty dv\, vc(v,0)$ and define

$$\psi(s) \;=\; \Phi(s,0) \;=\; \int_0^\infty dv\; e^{-sv}c(v,0) \qquad (\mathrm{Re}(s) > -\Delta) \tag{44}$$

Define $\tau_1 = \psi(-\Delta) - n_0$, which is either a positive number or $+\infty$. Then:

1. *The integral $\int_{-\infty}^{+\infty}[\psi(s_1+is_2)-c(0,0)/(s_1+is_2)]ds$, where s_1 and s_2 denote the real and imaginary parts of s, converges for any fixed non-zero $s_1 > -\Delta$. The integrand is bounded on the path of integration and is $O(1/s_2)^2$ for large $|s_2|$.*

2. *For any $s_1 > -\Delta$ we have $\lim_{|s_2|\to\infty}\psi(s_1+is_2) = 0$. The convergence is uniform with respect to s_1 on any interval $-\delta \le s_1 < \infty$ with $\delta < \Delta$.*

3. *For all $\tau \in [0, \tau_1)$ the equation $\psi(s) = n_0 + \tau$ has a simple root, call it $\sigma_1(\tau)$, which increases monotonically to 0 as τ decreases to 0, and satisfies*

$$-\Delta < \sigma_1(\tau) = -\frac{1}{\rho}\tau + R^{(2)}(\tau) \leq 0 \tag{45}$$

where $R^{(2)}(\tau) = O(\tau^2)$ as $\tau \to 0$.

4. *All other roots $\sigma_m(\tau)$ $(m = 2, ...)$ of the equation $\psi(s) = n_0 + \tau$ are complex, and there is a positive δ such that they lie to the left of the line $Re(s) = -\delta$ for all sufficiently small τ. That is to say, there exist $\tau_2 \in (0, \tau_1)$ and $\delta \in (0, \Delta)$ such that $Re(\sigma_m(\tau)) < -\delta$ $(m = 2, 3, ...)$ for $\tau \in [0, \tau_2]$.*

Proof:

The conditions on the initial cluster distribution $c(v, 0)$ ensure that the integral (44) defining $\psi(s)$ converges whenever $\mathrm{Re}(s) > -\Delta$. Moreover, it is an analytic function of s in this domain. By partial integration applied to this integral we obtain

$$\psi(s) = s^{-1}c(0,0) + s^{-2}c'(0,0) + s^{-3}c''(0,0) + s^{-3}\int_0^\infty e^{-sv}dc''(v,0) \quad (\mathrm{Re}(s) > -\Delta) \tag{46}$$

where the primes indicate partial differentiations of $c(v, 0)$ with respect to v. Writing $s_1 = \mathrm{Re}(s)$ and $s_2 = \mathrm{Im}(s)$ we have, by a theorem of Widder [22] closely related to the Riemann-Lebesgue lemma,

$$\int_0^\infty e^{-(s_1+is_2)v}dc''(v,0) = o(|s_2|) \quad \text{as } |s_2| \to \infty. \tag{47}$$

uniformly on any interval $-\nu \leq s_1 < \infty$ of the s_1 axis with $\nu < \Delta$. Hence, by (46), we find that

$$\psi(s_1 + is_2) - c(0,0)/(s_1 + is_2) = c'(0,0)/(s_1 + is_2)^2 + o(1/s_2)^2 = O(1/s^2) \quad \text{as } |s_2| \to \infty \tag{48}$$

uniformly on $-\nu \leq s_1 < \infty$. The first part of the Lemma now follows, since the left-hand side of (48), being analytic in $-\Delta < \mathrm{Re}(s) < \infty$ apart from a pole at the origin, is bounded on the path of integration.

The second part of the Lemma also follows directly from (48).

As s increases through real negative values from $-\Delta$ to 0, the value of $\psi(s)$ decreases from $\tau_1 + n_0$ to n_0; therefore for each $\tau \in [0, \tau_1)$ the equation $n_0 + \tau - \phi(s) = 0$ has a unique root in $(-\Delta, 0]$, increasing monotonically from $-\Delta$ to 0 as τ decreases from τ_1 to 0. We denote this root by $\sigma_1(\tau)$. Since $\psi(s)$ is analytic in a neighbourhood of $s = 0$ and its derivative $\psi'(s)$ satisfies $\psi'(0) = -\rho$, it follows by the implicit function theorem that

$$\sigma_1(\tau) = -\frac{1}{\rho}\tau + O(\tau^2) \tag{49}$$

This completes the proof of the third part of the Lemma.

To locate the other roots, we note first that the definitions of $\psi(s)$ (eqn (44)) and of n_0 imply that $|\psi(s)| \leq n_0$ for $\mathrm{Re}(s) \geq 0$, with equality only if $s = 0$. Therefore, since n_0 is positive and τ non-negative, the equation $\psi(s) = n_0 + \tau$ can have no roots in the closed right-hand half of the s-plane, other than the root at $s = 0$ when $\tau = 0$.

In the strip $-\Delta < \mathrm{Re}(s) \leq 0$ there is one real root, as we know from part 3 of the Lemma, and there may be complex ones as well. Consider first the case when $\tau = 0$. There is one root at $s = 0$ and the others, as shown above, must lie to the left of the real axis. It is shown below that the number of such roots in the strip $-\Delta/2 < \mathrm{Re}(s) < 0$ is finite. The largest of the real parts of the complex roots must therefore be negative: denote it by -2δ (if there are no roots in the strip set $\delta = \Delta/2$). Now consider the the case $\tau > 0$. Since the positions of the roots depend continuously on τ (e.g. [18]) and there are only a finite number of them in the relevant part of the complex s-plane, we see that that for sufficiently small τ all the roots except possibly $\sigma_1(\tau)$ lie to the left of the line $\mathrm{Re}(s) = -\delta$.

It remains to show that the number of roots in the strip $-\Delta/2 < \mathrm{Re}(s) < 0$ is finite. Part 2 of the Lemma shows that we can find a constant M such that $|\psi(s)| < n_0$ for all s in the strip for which $|\mathrm{Im}(s)| > M$; since $n_0 > 0$ and $\tau > 0$ the equation $\psi(s) = n_0 + \tau$ can therefore have no roots at all outside the part of the strip where $|\mathrm{Im}(s)| \leq M$. The number of roots inside this part of the strip is finite, because the analytic function $\psi(s) - n_0 - \tau$ can have only a finite number of zeros in a finite region of the s-plane.

This completes the proof of the Lemma. Q.E.D.

Now we can state our theorem concerning asymptotic behaviour in the continuous case.

Theorem VI *Let the initial cluster distribution $c(v,0)$ in (4) satisfy the same conditions as in Lemma V. Then equation (4) has a unique solution and this solution $c(v,t)$ has the following asymptotic behaviour with $\xi = vt^{-1}$ fixed*

$$\lim_{t\to\infty} t^2 c(\xi t, t) \;=\; \tilde{c}(\xi) \tag{50}$$

where the dynamical scaling function $\tilde{c}(\cdot) \;:\; I\!R^+ \to I\!R^+$ is given by

$$\tilde{c}(\xi) \;=\; \frac{1}{\rho} e^{-\frac{1}{\rho}\xi} \tag{51}$$

The convergence is uniform on compact intervals of the positive ξ axis.

Proof:

Solving the differential equations (32) and (34) with initial conditions $n(0) = n_0$, $\Phi(s,0) = \psi(s)$ we obtain

$$\Phi(s,t) \;=\; \frac{\tau^2}{(n_0+\tau)} \frac{\psi(s)}{n_0+\tau-\psi(s)} \quad , \quad t > 0 \;,\; \mathrm{Re}(s) > -\Delta \tag{52}$$

where, as before, τ means t^{-1}. The inversion formula for the Laplace transform (31) gives (see, for example [22])

$$\left(\frac{n_0+\tau}{\tau^2}\right) c(v,t) \;=\; \frac{1}{2\pi i}\int_{\gamma-i\infty}^{\gamma+i\infty} ds\; e^{sv} \frac{\psi(s)}{n_0+\tau-\psi(s)} \tag{53}$$

where γ is any positive number.

We shall estimate the integral in (53) for values of τ in the interval $[0,\tau_4]$ where $\tau_2 > 0$ is defined in Lemma V and we define $\tau_3 = \psi(-\delta/4) - n_0 > 0$ and set $\tau_4 = \min\{\tau_2, \tau_3\}$. These definitions ensure that when τ lies in this interval the denominator in the integrand has no zeros in the strip $-\delta < \mathrm{Re}(s) < \delta/4$: in the case of $\sigma_1(\tau)$, this is a consequence of the definition of τ_3 and the monotonic behaviour noted in part 3 of Lemma V; in the case of the other roots $\sigma_2, ...$ it is a consequence of part 4 of Lemma V.

The conditions of the theorem ensure that $\psi(s)$ is analytic in the region $\mathrm{Re}(s) > -\Delta$; consequently the integrand in (53) is meromorphic in this region, with poles at the roots of the equation $\psi(s) = n_0 + \tau$. By parts 3 and 4 of Lemma V one of these poles is at the point $s = \sigma_1(\tau)$ and the others, if any, all lie to the left of the abscissa $\mathrm{Re}(s) = -\delta <$. Moreover, we know from part 2 of Lemma V that $\psi(s) \to 0$ as $\mathrm{Im}(s) \to \pm\infty$, uniformly in $\mathrm{Re}(s)$ for $-\delta/2 <\mathrm{Re}(s) < \gamma$. Hence we may move the contour in (53) to the left, obtaining

$$\left(\frac{n_0+\tau}{\tau^2}\right) c(v,t) \;=\; \frac{1}{2\pi i}\int_{-\frac{\delta}{2}-i\infty}^{-\frac{\delta}{2}+i\infty} ds\, e^{sv}\frac{\psi(s)}{n_0+\tau-\psi(s)} \;+\; \frac{1}{2\pi i}\oint_\Gamma ds\, e^{sv}\frac{\psi(s)}{n_0+\tau-\psi(s)} \tag{54}$$

where Γ is a small circle surounding the pole $s = \sigma_1(\tau)$.

By the residue theorem, the second term on the right is equal to the residue of the integrand at $\sigma_1(\tau)$, which is

$$e^{\sigma_1(\tau)v}\frac{\psi(\sigma_1(\tau))}{-\psi'(\sigma_1(\tau))}$$

The first term, as we show below, has an upper bound $Ke^{-v\delta/2}$, where $K > 0$ is independent of τ for $\tau \in [0,\tau_4]$. This term therefore goes to 0 in the limit $v \to \infty$, and it follows that

$$\begin{aligned}\lim_{t\to\infty} t^2 c(\xi t,t) &= \lim_{\tau\to 0}\frac{c(\xi/\tau,1/\tau)}{\tau^2}\\ &= \frac{1}{n_0}\lim_{\tau\to 0} e^{\xi\sigma_1(\tau)/\tau}\frac{\psi(\sigma_1(\tau)}{-\psi'(\sigma_1(\tau)} + \frac{1}{n_0}\lim_{\tau\to 0} Ke^{-(\xi/\tau)\delta/2}\\ &= \frac{1}{\rho}e^{-\xi/\rho} \quad \text{by (49)}\end{aligned} \tag{55}$$

In this way the main result (50) is reached. If ξ is restricted to some positive interval $[\xi_1,\xi_2]$ then we see that the limit in (55) holds uniformly on this set of values for ξ as $v,t \to \infty$.

To complete the proof we estimate the first term on the right of (54). We first show that the denominator $n_0 + \tau - \psi(s)$, call it $g_\tau(s)$, is bounded away from zero on the path of integration. For large values of $\mathrm{Im}(s)$ this follows from part 2 of Lemma V, which shows that there is a number M_1 such that $\psi(-\delta/2+i\lambda) < n_0/2$ whenever $|\lambda| > M_1$, so that $|g_\tau(-\delta/2+i\lambda)| > n_0/2$ for these values of λ. For smaller values of $\mathrm{Im}(s)$, we use the fact noted above that there are no zeros of $g_\tau(-\delta/2+i\lambda)$ on the line with $-M_1 \le \lambda \le M_1$ for all τ in $[0,\tau_4]$. Since $|g_\tau(s)|$ is a continous function of the two variables λ,τ its infimum on the closed set $[-M_1,M_1]\times[0,\tau_4]$ must be attained and must therefore be different from zero (since the value 0 is not attained). We denote this infimum by g_{min}. Defining $\mu = \min\{n_0/2, g_{min}\}$, we conclude that

$$|n_0 + \tau - \psi(-\delta/2 + i\lambda)| \geq \mu > 0 \qquad (\tau \in [0, \tau_4]). \tag{56}$$

A partial integration of the type used in the proof of Lemma V gives

$$\psi(s) = c(0,0)/s + R_1(s) \tag{57}$$

where

$$\begin{aligned} R_1(s) &= s^{-1} \int_0^\infty e^{-sv} c'(v,0)\, dv & (58)\\ &= c'(0,0)/s^2 + s^{-2} \int_0^\infty e^{-sv} c''(v,0)\, dv & (59)\end{aligned}$$

so that

$$\begin{aligned} |R_1(-\delta/2 + i\lambda)| &\leq K_0/s \\ \text{and} \qquad &\leq K_1/s^2, \qquad \lambda \in I\!R \end{aligned} \tag{60}$$

with $K_0 = \int_0^\infty e^{v\delta/2} A e^{-\Delta v} dv$ and $K_1 = |c'(0,0)| + \int_0^\infty e^{v\delta/2} A e^{-\Delta v} dv$ independent of τ. It follows, using (57), that for $\mathrm{Re}(s) = -\delta/2$

$$\begin{aligned} \frac{\psi(s)}{n_0 + \tau - \psi(s)} &= \frac{\psi(s)}{n_0 + \tau} + \frac{\psi(s)^2}{(n_0 + \tau)(n_0 + \tau - \psi(s))} \\ &= \frac{c(0,0)}{(n_0 + \tau)s} + R_2(s) \end{aligned} \tag{61}$$

where

$$R_2(s) = \frac{R_1(s)}{(n_0 + \tau)} + \frac{\psi(s)^2}{(n_0 + \tau)(n_0 + \tau - \psi(s))} \tag{62}$$

so that, by (56) and (60),

$$|R_2(s)| < K_2/s^2 \tag{63}$$

the constant $K_2 = K_1/n_0 + [|c(0,0)| + K_0]^2/n_0\mu$ not depending on τ.

Substituting (61) into the integral to be estimated and using the standard result ([22])

$$\frac{1}{2\pi i} \int_{-\delta/2 - i\infty}^{-\delta/2 + i\infty} ds\, \frac{e^{sv}}{s} = 0 \qquad (\delta > 0, v > 0) \tag{64}$$

we obtain

$$\left|\frac{1}{2\pi i}\int_{-\delta/2-i\infty}^{-\delta/2+i\infty} ds \frac{e^{sv}\psi(s)}{n_0+\tau-\psi(s)}\right| = \left|\frac{1}{2\pi i}\int_{-\delta/2-i\infty}^{-\delta/2+i\infty} e^{sv} R_2(s)\, ds\right| \leq e^{-v\delta/2}K \tag{65}$$

where

$$K = \frac{1}{2\pi}\int_{-\infty}^{+\infty} \max_{\tau\in[0,\tau_4]} |R_2(-\delta/2+i\lambda)|\, d\lambda, \tag{66}$$

an integral whose convergence is guaranteed by (63). The bound (66) justifies the treatment of the second term in (55) and so the proof of Theorem VI is complete. Q.E.D.

4 Discussion

Theorems III and VI provide a rigorous proof of the dynamical scaling hypothesis for Smoluchowski's coagulation equation with constant coefficients $a_{k,\ell}$, subject to mild conditions on the inital data; it thus confirms previous studies ([1], [2], [5], [6], [9] - [11], [13] - [16]) in which this hypothesis was used successfully in a less rigorous way. Unfortunately, owing to our use of the explicit solutions of the Smoluchowski equation, our proof cannot be extended to the general coagulation equations with non-constant coefficients (1). There is one other case where an explicit solution is known; this is the case $K(x,y) = \text{const.}xy$ [23, 24]. However, our method appears not to work for this case [25] because of the gelation transition at a finite time.

Our analysis leads to the same asymptotic form in both discrete and continuous cases: this is not surprising in view of the fact that the continuous equation can be thought of as a continuous approximation to the discrete one, whose validity is likely to be better the larger the cluster size ℓ. This asymptotic form is also the same as the one given by Friedlander's theory of self-similar spectra (i.e. self-similar cluster-size distributions).

For both discrete and continuous versions our result confirms the standard values for the dynamical scaling exponents in eqn (2), namely $x = 1$, $y = -2$. These numbers can, of course, be calculated by simpler methods. The fact that $x = 1$ can be seen as a consequence of the fact that the mean cluster size $\ell^* = \sum \ell c_\ell / \sum c_\ell = \rho/n(t)$ is, by (18) and the constancy of ρ, asymptotically proportional to the first power of

t; the value of y can then be deduced from the scaling relation $y = -2x$ which is a consequence of mass conservation (see for example [2]).

Using the above formula for mean cluster size, the asymptotic formula for the cluster distribution can be expressed in the form introduced by Lifshitz and Slyozov ([26])

$$c_\ell(t) \sim \frac{\rho}{[\ell^*]^2} e^{-\frac{\ell}{\ell^*}} \tag{67}$$

The scaling function for the Smoluchowski equation is, however, different from the one given by Lifshitz and Slyozov. The Smoluchowski coagulation mechanism is not to be confused with the Lifshitz-Slyozov coarsening mechanism ([1], [26], [27]), despite the agreement of the dynamical exponents x, y for $d = 3$. The Smoluchowski model has no fragmentation, whereas Lifshitz and Slyozov consider both coagulation (condensation) and fragmentation (evaporation). The essential difference between these two mechanisms and their physical relevance for solid mixtures and binary fluids was pointed out by Binder ([1]).

Finally we point out that the obvious way of generalizing Smoluchowski's equation, by including fragmentation as well as coagulation terms ([5], [19]) is not as innocent as it looks: it can lead to a dramatic change in the physics of the model. In a certain sense, the model without fragmentation investigated here has a phase transition at $t = \infty$: although the total mass $\sum \ell c_\ell(t)$ at any finite time t is equal to its initial value, the individual terms in the sum tend to 0 as $t \to \infty$, so that at $t = \infty$ there is no longer any mass in clusters of finite size. We may say that at $t = \infty$, when the system has reached equilibrium, all the mass has gone into a cluster or clusters of infinite size. This behaviour may be interpreted as a phase transition of first order, analogous to the one found in the mass-conserving Becker-Döring equations ([28]). On the other hand if constant fragmentation is included the physical (mass-conserving) solution to the discrete coagulation-fragmentation equation will in many cases ([29]) approach an equilibrium having the conserved value for the mass: even at $t = \infty$ the total mass in finite clusters is the same as at $t = 0$ and thus there is no first-order phase transiton.

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