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Field theoretic approach to a source-enhanced aggregation process

Hisao Hayakawa†

Department of Physics, Kyushu University, Fukuoka 812, Japan

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Abstract. A diffusion-controlled cluster-cluster aggregation system with a source is analysed using the Fock space formalism and diagrammatic perturbation technique. The effective coagulation kernel and equation for low-dimensional cases ($d \leqslant 2$) are derived. The asymptotic size distribution obeying a power law is obtained for any dimension. The application to the aggregation process of particles undergoing Lévy flight is discussed. Through the analysis in this paper, we clarify a relation between critical phenomena and a source-enhanced aggregation.

1. Introduction

Aggregation is a typical irreversible process in which basic units (monomers) stick together to form clusters. Recently, interest in this problem has increased, because this process appears in many areas of science (e.g. Family and Landau 1984).

A source-enhanced cluster-cluster aggregation can be observed in various natural phenomena such as atmospheric aerosols (Klett 1975, Friedlander 1977, White 1982), star formation (Field and Saslaw 1965), the formation of interstellar dust grains (Salpeter 1977, Hayakawa and Hayakawa 1988), vapour-deposited thin films (Family and Meakin 1988) and diffusion-controlled deposition (Rácz and Vicsek 1983, Matsushita and Meakin 1988). The source-enhanced aggregation process also has a relation to a mathematical model, the so-called unbiased voter model (Griffeath 1979). Recently, Takayasu and Nishikawa (1986) have found a power-law size distribution for a source-enhanced aggregation model from their simulation (see also Takayasu *et al* 1988, Hayakawa *et al* 1987). In the mean-field approximation, i.e. using the Smoluchowski coagulation equation, Hayakawa (1987) obtained a solution for size distribution obeying a power law. We need a more basic theory going beyond the mean-field approximation to describe source-enhanced aggregation.

On the other hand, Vicsek *et al* (1985) investigated a similar system in which particles are injected into the system at a constant rate and larger clusters are removed by sedimentation. They found that the number of clusters n takes the scaling form involving the injection rate h as $n = h^a F(h^b t)$, where $F(x)$ is a scaling function. Rácz (1985a, b) pointed out the similarity between source-enhanced aggregation and dynamical critical phenomena. If we can construct a basic theory for an aggregation system, we will be able to understand the similarity on a level going beyond phenomenological arguments.

† Present address: Department of Physics, Emory University, Atlanta, Georgia 30322, USA.

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In this paper, we study a source-enhanced aggregation process. We derive the effective coagulation kernel and discuss the size distribution. In the course of our analysis, we find the scaling law between a source strength and number density of clusters.

The plan of this paper is as follows. In the next section we formulate the master equation in Fock space following the method of Mikhailov and Yashin (1985). Section 3 is devoted to an explanation of diagrammatic perturbation technique. Section 4 is the central part of this paper, where we derive a 'renormalised' coagulation equation and obtain a scaling solution of the size distribution. This section is derived into three parts. In § 4.1 we give the analysis for the zero-density limit which describes time evolution of the system. In § 4.2 we analyse a finite-density system which is applicable to a steady state. In § 4.3 we mention the logarithmic correction when the spatial dimension is two. In § 5 we apply our theory to aggregation particles undergoing Lévy flight. In the final section we discuss the similarity between source-enhanced aggregation and traditional critical phenomena, and summarise our results.

2. The Fock space formalism

The Fock space formalism (formulated by Doi (1976), Grassberger and Scheunert (1980) and Peliti (1985)) is a powerful tool for studying various reaction-diffusion systems (Ohtsuki and Keyes 1987a, b and references therein). This formalism is also effective in studying aggregation systems, because we believe that diffusive annihilation is equivalent to irreversible aggregation, in which the reaction rate is independent of coalescing clusters. Thus we can construct a theory based on the work by Mikhailov and Yashin (1985) who gave a detailed analysis for source-enhanced diffusive annihilation (see also Peliti (1986) and Elderfield (1987); the validity of the Smoluchowski equation was discussed in the latter paper). In this paper, by using the Fock space formalism, we analyse a source-enhanced aggregation system. We exclude the reaction-limited aggregation (RLA) because there often occurs a sol-gel transition at a finite time (Ziff 1980) and we cannot assume the Markov process in RLA.

A source-enhanced aggregation process is often expressed by the following equations:

$$X_i + X_k \xrightarrow{K_{ij}} X_{i+j} \qquad 0 \xrightarrow{h} X_1 \tag{2.1}$$

where X_k $(k = i, j, i+j)$ denotes a k-mer which is formed from k monomers and K_{ij} is the rate coefficient or reaction kernel. The second equation in (2.1) represents the creation or injection of monomers with the rate h.

The system of classical reacting clusters can be described by the Fock space formalism. In this formalism the time evolution of system is represented by the Liouville equation as

$$\frac{\partial}{\partial t}|\varphi\rangle = L|\varphi\rangle \tag{2.2}$$

where L is the 'Liouvillian' expressed by creation and annihilation operators. The state vector can be expressed as $|\varphi\rangle = |\varphi_1\rangle \otimes |\varphi_2\rangle \otimes \ldots \otimes |\varphi_N\rangle$ where $|\varphi_s\rangle$ denotes the state vector for clusters with size s. Let us assume the medium is divided into cells which contain a macroscopic number of clusters. We characterise every cell by its centre position $\boldsymbol{r}_m$. The probability distribution function $P_s(\{n_s(\boldsymbol{r}_m)\}, t)$ gives the

probability of finding n_s clusters with size s contained in the cell located at $\boldsymbol{r}_m$. The vector $|\varphi_s\rangle$ is defined as

$$|\varphi_s\rangle = \sum_{\{n_s(\boldsymbol{r}_m)\}} P_s(\{n_s(\boldsymbol{r}_m)\}, t) \prod_m (\Psi_s^\dagger(\boldsymbol{r}_m))^{n_s(\boldsymbol{r}_m)}|0\rangle \tag{2.3}$$

where the summation is performed over all possible cluster numbers $\{n_s(\boldsymbol{r}_m)\}$. $\Psi_s^\dagger(\boldsymbol{r}_m)$ is the creation operator of a cluster with size s which satisfies the commutation relation

$$[\Psi_i(\boldsymbol{r}_n), \Psi_j^\dagger(\boldsymbol{r}_m)] = \Delta_{n,m}\Delta_{i,j} \tag{2.4}$$

where $\Psi_i(\boldsymbol{r}_n)$ is the annihilation operator with size i and Δ_{ij} is the Kronecker delta. The Fourier transformations of creation and annihilation operators are expressed as $\pi_s(\boldsymbol{k})$ and $a_s(\boldsymbol{k})$. For source-enhanced aggregation, the Liouvillian L has the form

$$L = L_0 + L_{\text{int}} \tag{2.5a}$$

where

$$L_0 = -\sum_s \sum_k D_s k^2 \pi_s(\boldsymbol{k}) a_s(\boldsymbol{k}) \tag{2.5b}$$

and

$$L_{\text{int}} = \frac{1}{2V}\sum_{i,j}\sum_{k,p} K_{ij}[\tfrac{1}{2}(\boldsymbol{p}-\boldsymbol{k})]\pi_{i+j}(\boldsymbol{p}+\boldsymbol{k})a_i(\boldsymbol{p})a_j(\boldsymbol{k})$$

$$-\frac{1}{2V}\sum_{i,j}\sum_{k,p,q} K_{ij}(\boldsymbol{k})\pi_i(\boldsymbol{p}+\boldsymbol{k})\pi_j(\boldsymbol{q}-\boldsymbol{k})a_i(\boldsymbol{p})a_j(\boldsymbol{q}) + Vh(\pi_1(0)-1). \tag{2.5c}$$

Here L_0 and L_{int} are the free and interactive Liouvillians, respectively, D_s is the diffusion coefficient of cluster with size s, $K_{ij}(\boldsymbol{k})$ is the Fourier transformation of the reaction kernel $K_{ij}(\boldsymbol{r})$ and V is the volume of the medium. The injection rate of monomers h is assumed to be uniform per unit time per unit volume.

As the Liouvillian is non-Hermitian, we introduce the covector satisfying the normalisation condition following Mikhailov and Yashin (1985), namely, $|\bar{\varphi}\rangle = |\bar{\varphi}_1\rangle \otimes \ldots \otimes |\bar{\varphi}_N\rangle$ is defined by

$$a_s(k)|\bar{\varphi}(k)\rangle \equiv \Delta(k)|\bar{\varphi}(k)\rangle. \tag{2.6}$$

where $\Delta(\boldsymbol{k}) = 1$ for $\boldsymbol{k} = 0$ and $\Delta(\boldsymbol{k}) = 0$ for $\boldsymbol{k} \neq 0$. In terms of the covector, the normalisation relation for distribution function becomes $\langle\bar{\varphi}|\varphi\rangle = 1$. The observable number of clusters is the average of the number operator as

$$N_s = \left\langle \bar{\varphi} \middle| \sum_k \pi_s(k)a_s(k) \middle| \varphi \right\rangle = \langle\bar{\varphi}|a_s(0)|\varphi\rangle \equiv \langle a_s(0)\rangle. \tag{2.7}$$

From (2.6) and (2.7) we find $[a_s(0), \pi_s(0)] = 1$ and $\langle\pi_s(0)\rangle = 1$. Hence $\pi_s(0)$ and $a_s(0)$ can be treated as c numbers for $N \gg 1$.

3. Diagrammatic perturbation

According to (2.2), (2.5) and (2.7) time evolution of the number density of clusters $n_s = N_s/V$ is described by the equation

$$\frac{\partial}{\partial t} n_s = \frac{1}{V}\langle a_s(0)L\rangle$$

$$= \frac{1}{2V^2}\sum_k \left(\sum_{i+j=s} K_{ij}(\boldsymbol{k})\langle a_i(\boldsymbol{k})a_j(-\boldsymbol{k})\rangle - 2\sum_{j=1}^{N} K_{sj}(\boldsymbol{k})\langle a_s(\boldsymbol{k})a_j(-\boldsymbol{k})\rangle \right) + h\Delta_{s,1} \tag{3.1}$$

where we use (2.6) and the commutation relation $[a_i(\boldsymbol{k}),\ \pi_j(\boldsymbol{p})] = \Delta_{ij}\Delta(\boldsymbol{p}-\boldsymbol{k})$. If we adopt an approximation as $\langle a_i(\boldsymbol{k})a_j(-\boldsymbol{k})\rangle = N_iN_j$, (3.1) reduces to the Smoluchowski coagulation equation.

Introducing the interaction representation we can develop a perturbation expansion. In this representation we have $\tilde{a}_s(\boldsymbol{k}, t) = \exp(L_0t)a_s(\boldsymbol{k})\exp(-L_0t)$ and

$$\frac{\partial}{\partial t}|\tilde{\varphi}(t)\rangle = \tilde{L}_{\text{int}}(t)|\tilde{\varphi}(t)\rangle \tag{3.2}$$

where $\tilde{L}_{\text{int}}(t) = \exp(L_0t)L_{\text{int}}\exp(-L_0t)$ and $|\tilde{\varphi}(t)\rangle = \exp(L_0t)|\varphi(t)\rangle$.

As (3.2) can be formally solved, we can represent the correlations in (3.1) as

$$\begin{aligned}\langle\bar{\varphi}|a_i(\boldsymbol{k})a_j(-\boldsymbol{k})|\varphi\rangle &= \langle\bar{\varphi}|\tilde{a}_i(\boldsymbol{k}, t)\tilde{a}_j(-\boldsymbol{k}, t)U(t, -\infty)|\tilde{\varphi}(-\infty)\rangle \\ &\equiv \langle\tilde{a}_i(\boldsymbol{k}, t)\tilde{a}_j(-\boldsymbol{k}, t)U(t, -\infty)\rangle_0\end{aligned} \tag{3.3}$$

with the evolution operator

$$U(t, -\infty) = T\exp\left(\int_{-\infty}^{t}\tilde{L}_{\text{int}}(t')\,\mathrm{d}t'\right) \tag{3.4}$$

where T denotes the time-ordering operator. Here we assume that the correlations among particles are absent in the initial state $|\tilde{\varphi}(-\infty)\rangle$. By expanding $U(t, -\infty)$ into the series of products of $\langle T\tilde{a}_l\tilde{\pi}_m\rangle_0$ and $\langle\tilde{a}_s(0, t)\rangle_0 = \langle a_s(0)\rangle$ with the help of Wick's theorem, we can perform a perturbative calculation. Here the bare propagator is defined by

$$\begin{aligned}&\langle T\tilde{a}_i(\boldsymbol{k}, t)\tilde{\pi}_j(\boldsymbol{k}', t')\rangle_0 \\ &\quad = \Delta_{ij}\Delta(\boldsymbol{k}-\boldsymbol{k}')G_i^0(\boldsymbol{k}-\boldsymbol{k}', t-t') \\ &\quad = \Delta_{ij}\Delta(\boldsymbol{k}-\boldsymbol{k}')\exp(-D_ik^2(t-t'))\theta(t-t')\end{aligned} \tag{3.5}$$

where $\theta(t)$ is the step function. Let us note the following points in the perturbation procedure. (As we assume $N_s \gg 1$, the contribution from $\langle\pi_s(0)\rangle = \langle\tilde{\pi}_s(0, t)\rangle_0 = 1$ is negligible.) From $h(\langle\pi_1(0)\rangle - 1) = 0$, there is no explicit contribution from the injection terms. The injection term affects the result through $N_s = \langle a_s(0)\rangle$. We now introduce the real propagator of clusters as

$$G_{ij}(\boldsymbol{k}, \tau) = \langle T\tilde{a}_i(\boldsymbol{k}, t+\tau)\tilde{\pi}_j(\boldsymbol{k}, t)U(t, -\infty)\rangle_0. \tag{3.6}$$

We note that the size of the cluster is not conserved, in general, during the propagation of clusters. Therefore the non-diagonal elements of the real popagator exist. From now on, we use the propagator of Laplace transformation, i.e. the free propagator is $G_s^0(\boldsymbol{k}, z) = (z + D_sk^2)^{-1}$. For simplicity, the notation for the Green function shall not change under the Laplace transformation. From (2.5), (3.4) and (3.6) we find that the propagator has the following form for $N_s \gg 1$:

$$G_{ij}(\boldsymbol{k}, z) = G_i^0(\boldsymbol{k}, z)\Delta_{ij} - \sum_{l,m} 2G_i^0(\boldsymbol{k}, z)\Gamma_{il,m}G_{mj}(\boldsymbol{k}, z). \tag{3.7}$$

Here the first term on the right-hand side expresses the diagonal part and the second term expresses the size increase due to coalescence during the propagation of the cluster. $\Gamma_{il,m} = \lim_{p\to 0}\Gamma_{il,m}(\frac{1}{2}(\boldsymbol{k}-\boldsymbol{p}), \boldsymbol{k}, \boldsymbol{p}; z)$ represents the process in which two clusters (size i and momentum $\boldsymbol{k}$, and size l and momentum $\boldsymbol{p}$) stick together to form a cluster with size m. However, we must note that the renormalised kernel $\Gamma_{il,m}$ can contain multiple coalescence processes. Thus the renormalised reaction rate is defined by

$$\Gamma_{il}(z)n_in_l \equiv K_{il}(\boldsymbol{k})\langle a_i(\boldsymbol{k})a_j(-\boldsymbol{k})\rangle/V^2. \tag{3.8}$$

The discussion to obtain (3.7) is parallel to the discussion by Mikhailov and Yashin (1985) (see figure 1). On the left-hand side of (3.8) $\Gamma_{il}(z)$ is equal to $\lim_{k,p\to 0} \Sigma_m \Gamma_{ij,m}(k, p, p-2k; z)$.

In the low-density case, i.e. the small injection rate case, the ladder approximation is effective, because the correction for such an approximation appears to higher order of the number density of clusters. Therefore Γ_{ij} is determined by

$$\Gamma_{ij,n+s}(\boldsymbol{k}, \boldsymbol{p}, \boldsymbol{p}-2\boldsymbol{k}; z)$$

$$= K_{ij}(\boldsymbol{k}) - 2\sum_{l,m}\int_c \frac{d\Omega}{2\pi i}\int \frac{d^d q}{(2\pi)^d} K_{ns}(\boldsymbol{k}-\boldsymbol{q})\Gamma_{ij,l+m}(\boldsymbol{q}, \boldsymbol{p}, \boldsymbol{p}-2\boldsymbol{k}; \Omega)$$

$$\times G_{ln}(\boldsymbol{p}-\boldsymbol{q}; z-\Omega)G_{ms}(\boldsymbol{p}-2\boldsymbol{k}+\boldsymbol{q}, \Omega) \tag{3.9}$$

with $\boldsymbol{k}, \boldsymbol{p} \to 0$ (see figure 2). Equation (3.9) is not simple, because the Green functions contain effective reaction rates.

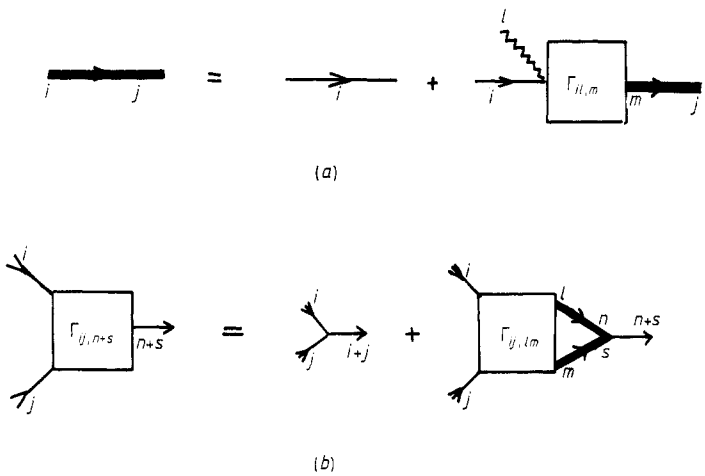

Figure 1. (*a*) Diagrammatic representation of (3.7), where thin, thick and zigzag lines represent the bare propagator, the real propagator and the number density n_1, respectively. Each subscript denotes the size of the cluster. The first diagram on the right-hand side represents the diagonal element and the second diagram corresponds to the size increase because of coalescence where the summations are taken over l and m. (*b*) Diagrammatic representation of (3.9) where $\Gamma_{ij,lm} = -\Gamma_{ij,l+m}$ from (2.5*c*).

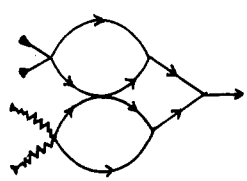

Figure 2. A simple example of a diagram which is not contained in (3.9). The contribution from this diagram can be neglected in the low-density case, because the order of this diagram is n_s^2.

4. Effective rate and size distribution

In this section, calculating the correction to the vertex function, we obtain an effective reaction rate and the size distribution functions obeying power laws. We also confirm the scaling law pointed out by Rácz (1985a, b).

4.1. Zero-density limit (time evolution)

First, we consider the time evolution of a source-enhanced aggregation system. Since we are interested in the behaviour at low density, we adopt the zero-density approximation in (3.9) where the real propagator is replaced by the bare propagator. Thus, we can express (3.9) as

$$\Gamma_{ij}(z) \simeq K_{ij}(0)[1+2K_{ij}(0)I(0,0;z)]^{-1} \tag{4.1a}$$

with

$$I(\boldsymbol{p},\boldsymbol{p}';z) = \int_c \frac{d\Omega}{2\pi i} \int \frac{d^d q}{(2\pi)^d} G_i^0(\boldsymbol{p}-\boldsymbol{q}, z-\Omega) G_j^0(\boldsymbol{p}'-\boldsymbol{q},\Omega). \tag{4.1b}$$

Since we are interested in the behaviour at long wavelengths of diffusion-limited aggregation, we use an approximation as $\boldsymbol{K}_{ij}(k) \simeq K_{ij}(0)$ for $|\boldsymbol{k}| < R_{ij}^{-1}$ where R_{ij} is the reaction radius whose order is the sum of radii of the i-mer and j-mer. In such a case the integral $I(0,0;z)$ can easily be performed:

$$\begin{aligned} I(0,0;z) &= \int \frac{d^d q}{(2\pi)^d} \frac{1}{z + D_{ij} q^2} \\ &= \frac{\pi K_d (z/D_{ij})^{(d-2)/2}}{D_{ij} \sin(\pi d/2)} \end{aligned} \tag{4.2}$$

where $K_d = 2^{1-d}/\pi^{d-2}\Gamma(d/2)$ and $D_{ij} = D_i + D_j$. From the first equation to the second equation we use the property of the beta function, $B(d/2, 1-d/2) = \pi/\sin(\pi d/2)$. This integral diverges at $z \to 0$ when the spatial dimension, d, is less than two. On the other hand, the integral can be neglected in the same limit for $d \geqslant 2$, namely the upper critical dimension d_c which ensures the validity of the mean-field approximation is two.

From (4.1) and (4.2) we can obtain the effective reaction rate:

$$\Gamma_{ij}(z) \simeq \frac{(D_{ij})^{d/2} \sin(\pi d/2) z^{(2-d)/2}}{\pi K_d} \tag{4.3}$$

for $d < 2$. Although the inverse Laplace transformation of (4.3) does not exist for any time, we can analyse the asymptotic behaviour of a source-enhanced aggregation system. Since we discard diagrams except for the ladder type in our approximation, the dimension analysis is exact (Amit 1978). Thus we estimate the effective rate as

$$\frac{\partial n_s}{\partial t} \simeq \frac{1}{2} \sum_{i+j=s} A_{ij} t^{-\varepsilon/2} n_i n_j - n_s \sum_{j=1} A_{sj} t^{-\varepsilon/2} n_j + h\Delta_{s,1} \tag{4.4}$$

where $\varepsilon = 2-d$ and A_{ij} is the contribution from the inverse Laplace transformation and the prefactor of $z^{(2-d)/2}$ in (4.3).

Next, we consider the size distribution of clusters. We assume that the asymptotic solution of (4.4) has the scaling form

$$n_s \simeq \sigma \langle s \rangle^{-\tau} f(s/\langle s \rangle) \tag{4.5}$$

where $f(x)$ is a scaling function. We consider the time region which is long enough to ensure (4.4) and short enough for the maximum size of clusters not to reach the cutoff size. In this time region the mean cluster size $\langle s\rangle$ grows as time increases. We adopt a scaling analysis developed by van Dongen and Ernst (1988), which is used for a sourceless aggregation in the mean-field approximation. Let us introduce the mass flux $\dot{M}^{(s)}$ from clusters of size $j \leqslant s$ to the clusters of size $j > s$:

$$\dot{M}^{(s)}(t) \equiv \sum_{j=1}^{s} j\dot{n}_j(t) = h - \sum_{j=s+1}^{N} j\dot{n}_j(t) \tag{4.6}$$

where N is the cutoff size. In § 4.1 we replace the cutoff N by ∞. From (4.5) and (4.6) we obtain

$$\dot{M}^{(s)}(t) \simeq h + \sigma\langle s\rangle^{1-\tau}\langle\dot{s}\rangle \int_x^\infty \mathrm{d}y[y^2 f'(y) + \tau y f(y)] \tag{4.7}$$

where $f'(x) = \mathrm{d}f/\mathrm{d}x$ and $x = s/\langle s\rangle$. From (3.1), (3.9) and (4.6) we find another expression for $\dot{M}^{(s)}$:

$$\begin{aligned}\dot{M}^{(s)} &= h - \sum_{i=1}^{s}\sum_{j=s-i+1}^{\infty} \mathrm{i}\Gamma_{ij} n_i n_j \\ &\simeq h - \sigma^2\langle s\rangle^{3+\lambda'-2\tau} t^{-\varepsilon/2} \int_0^x \mathrm{d}u \int_{x-u}^\infty \mathrm{d}v\, uA(u, v) f(u) f(v)\end{aligned} \tag{4.8}$$

where we assume the homogeneity of A_{ij} as $A(ai, aj) = a^{\lambda'} A(i, j)$ where the exponent λ' is determined by the diffusion coefficient (see (4.3)). For example, $\lambda' = d/6$ (Friedlander 1977) when the cluster is spherical. From (4.7) and (4.8) we can separate the x and t dependence of the coagulation equation as

$$\langle\dot{s}\rangle\langle s\rangle^{\tau-\lambda'-2} = wt^{-\varepsilon/2} \tag{4.9a}$$

and

$$-w\int_x^\infty [\tau y f(y) + y^2 f'(y)]\,\mathrm{d}y = \sigma \int_0^x \mathrm{d}u \int_{x-u}^\infty \mathrm{d}v\, uA(u, v) f(u) f(v) \tag{4.9b}$$

where w is a separation constant. This separation supports the validity of the scaling ansatz (4.5). Equation (4.9a) can be easily solved. The solution is given by

$$\langle s(t)\rangle \simeq \left(\frac{\tau - 1 - \lambda'}{1 - \varepsilon/2}\, w\right)^{1/(\tau-1-\lambda')} t^{(1-\varepsilon/2)/(\tau-1-\lambda')} \tag{4.10}$$

for $t \gg [d\langle s(0)\rangle^{\tau-\lambda'-1}/2(\tau-\lambda'-1)]^{2/d}$. From (4.9$b$) we find that $f(x)$ has a solution obeying a power law for small x as $x^{-\tau}$. From (4.8) and (4.10) we obtain

$$h - \dot{M}^{(s)} \simeq \sigma^2\langle s(t)\rangle^{3+\lambda'-2\tau-\varepsilon/2z}\left(\frac{\tau-\lambda'-1}{1-\varepsilon/2}\, w\right)^{\varepsilon/d} J_1(\tau, x) \tag{4.11}$$

where we use (4.10) with $z = (1-\varepsilon/2)/(\tau-1-\lambda')$ and $J_1(\tau, x) = \int_0^x \mathrm{d}u \int_{x-u}^\infty \mathrm{d}v\, uA(u, v)(uv)^{-\tau}$. The left-hand side of (4.11) tends to zero after a finite time, owing to a balance between injection h and dissipative mass flux $\dot{M}^{(s)}$. Therefore the power index of $\langle s\rangle$ must be zero for large $\langle s\rangle$. As a result we obtain the exponent

$$\tau = 2(3 - \varepsilon + \lambda')/(4 - \varepsilon) = 2(d + 1 + \lambda')/(d + 2). \tag{4.12}$$

Since the prefactor σ in (4.5) is proportional to the separation constant w (see (4.9b)), σ is determined by (4.10) and (4.11) as

$$\sigma = [hC'/J_1(\tau)]^{d/(2+d)} \tag{4.13}$$

with the numerical factor C' (see (4.9b) and (4.11)). The expression (4.13) gives a proof for the scaling relation predicted by Rácz (1985a) for $1 < d < 2$. We add a comment here: since (4.9b) is the familiar equation in the scaling theory of Smoluchowski's equation, we know that the scaling function has an exponential form as $f(x) \sim Ax^{-\lambda} \exp(\alpha x)$ for large x where α and A are constants whose ratio can be represented by known quantities (van Dongen and Ernst 1988).

From the above analysis we find that the size distribution is characterised by the exponent τ, which is the region of size distribution a power law increases as time increases. These statements supports the numerical results by Hayakawa *et al* (1987) and Takayasu *et al* (1988). When we set $\lambda' = 0$ and $d = 1$, we obtain $\tau = \frac{4}{3}$, which agrees with the result of a simulation by Takayasu *et al* (1988).

4.2. Finite density (steady state)

In § 4.1 we use an approximation in which the density of clusters is negligible. However, this approximation leads to some contradictions: the true steady state is not attainable, because the largest cluster grows due to coalescence. The total mass M involved in the medium diverges as $M = ht$ in the long-time limit if we do not consider the effect of a sink. In order to solve these contradictions, we must consider the density of clusters and introduce the cutoff of cluster sizes which represents the effect of a sink.

When spatial dimension is larger than two, we can use the Smoluchowski equation. In this case we can show a power-law size distribution in a steady state as follows. We consider a system with cutoff N, where clusters larger than N are removed due to sedimentation. As time increases, the system reaches a steady state because of a balance between a source and a sink. First, we assume that the cutoff size is infinite. From (4.6) we obtain

$$\dot{M} = \lim_{s\to\infty} \dot{M}^{(s)} = h - \lim_{s\to\infty} \sum_{i=1}^{s} \sum_{j=N-i+1}^{\infty} iK_{ij} n_i n_j. \tag{4.14}$$

In order to balance between the source and the sink, the second term on the right-hand side in (4.14) must be finite. This is possible if n_s has the algebraic form

$$n_s \simeq B(h) s^{-\tau} \qquad (s \gg 1). \tag{4.18}$$

Substituting the ansatz (4.15) into (4.14) with $\dot{M} = 0$ we obtain

$$h \simeq B(h)^2 s^{3+\lambda-2\tau} J_2(\tau). \tag{4.16}$$

Here $J_2(\tau)$ is defined as

$$J_2(\tau) = \int_0^1 dx \int_{1-x}^{\infty} dy\, xK(x, y)(xy)^{-\tau}. \tag{4.17}$$

When we assume $s \gg 1$, the right-hand side of (4.16) must be independent of s. Therefore we obtain

$$\tau = (\lambda + 3)/2 \tag{4.18}$$

for $\lambda < 1$ (non-gelling model) and

$$B(h) = [h/J_2(\tau)]^{1/2}. \tag{4.19}$$

Equations (4.18) and (4.19) give a generalisation of the result obtained by Hayakawa (1987).

When $d \leqslant 2$, we must consider contributions from the diagram in figure 1. In this case the expression of the renormalised kernel is complicated, because we must use the real propagator which is not scalar. When the reaction kernel is independent of cluster size, we can use a simplified description. In such a case, aggregation can be regarded as diffusive annihilation $X+X \to X$ where X represents a cluster. Diffusive annihilation has already been analysed by Mikhailov and Yashin (1985) and Peliti (1986). In this case the Green function is given by

$$G(\boldsymbol{k}, z) = (z + Dk^2 + 4n\Gamma(\boldsymbol{k}, z))^{-1} \tag{4.20}$$

where Γ and n are the effective reaction rate and the number density of clusters, respectively. $\Gamma(\boldsymbol{k}, z)$ is determined by an equation similar to (4.1), where we replace G^0 by G. Thus

$$\Gamma(0,0) \simeq D\left(\frac{2\sin(\pi d/2)}{\pi K_d}\right)^{2/d} (4n)^{(2-d)/d} \tag{4.21}$$

is obtained for $d<2$. In the case of $d=1$ we obtain $\Gamma = 16Dn$, which recovers the result obtained by Mikhailov and Yashin (1985) except for a numerical factor.

The size distribution can be obtained when the reaction kernel is Γ. In the following discussion we consider the system with a finite cutoff size N. Here we assume the scaling ansatz

$$n_s \simeq \sigma' s^{-\tau} g(s/N). \tag{4.22}$$

This ansatz is natural when we consider the previous discussions. Let us consider the size distribution for $s \sim N$. Substituting (4.21) with $\Gamma = A_d n^{(2-d)/d}$ and the ansatz (4.22) into the first equation of (4.8), we also find that the size distribution obeys a power law when we observe the size distribution near the cutoff size with

$$\tau = \frac{2(d+1)}{d+2} \qquad \sigma' = \left(\frac{h}{J_3(\tau)}\right)^{d/(2+d)} \tag{4.23}$$

where

$$J_3(\tau) = A_d\left(\int_0^1 \mathrm{d}z z^{-\tau} g(z)\right)^{(2-d)/d} \int_0^1 \mathrm{d}x \int_{1-x}^1 \mathrm{d}y\, x(xy)^{-\tau} g(x) g(y).$$

4.3. Logarithmic correction

When the spatial dimension is equal to two, the size distribution function has a logarithmic correction. In §4.3 we analyse the simplest case, that the coagulation rate is independent of the size of the clusters. Following the discussion to obtain (4.21), we find that the reaction kernel has a logarithmic correction in a steady state as

$$\Gamma \simeq \frac{\pi D}{\ln(D/4n\Gamma R^2)} \tag{4.24}$$

where R is the reaction radius. The expression (4.24) is a self-consistent equation.

Since the higher-order corrections are small, (4.24) can be approximated as

$$\Gamma \simeq \pi D/\ln(L/R) \tag{4.25}$$

where $L \simeq n^{-2}$ is the mean distance between the clusters. From the steady equation of motion in (3.1) the solution has a form (see Hayakawa 1987)

$$\begin{aligned} n_s &\simeq (h \ln(L/R)/2\pi^2 D)^{1/2} s^{-3/2} \\ &\simeq (h \ln N/2\pi^2 D)^{1/2} s^{-3/2} \qquad \text{for } N \gg 1. \end{aligned} \tag{4.26}$$

From these arguments the steady-state distribution has a logarithmic correction whose form is given by (4.26) with the cutoff size N. Before the system reaches a steady state, we have to replace N by $\langle s \rangle$. In such a case the size distribution has a logarithmic correction as $n_s \simeq [h \ln(ct)/2\pi^2 D]^{1/2} s^{-3/2}$ where c is some constant.

When we assume the power-law size distribution without the logarithmic correction, we find that the power-law exponent is smaller than the true value. Takayasu *et al* (1988) reported that the exponent of size distribution is smaller than the mean-field value for the two-dimensional case from their numerical calculation. Then they discussed the possibility that the upper critical dimension is four. However, there is little possibility of their assertion being true, because their analysis did not consider the logarithmic correction such as (4.26).

5. Application

In this section, we discuss an application of the above formalism to the aggregation of particles undergoing Lévy flight (Takayasu *et al* 1988). Lévy flight (Hughes and Prager 1983) is characterised by the following transition probability of a particle:

$$W(r) \propto r^{-\beta-1} \tag{5.1}$$

where r is the hopping distance. From the definition of Lévy flight, we obtain the free propagator (Hughes and Prager 1983)

$$G^0(\boldsymbol{k}, z) = (z + Dk^\beta)^{-1} \qquad (0 < \beta \leqslant 2). \tag{5.2}$$

When β is equal to two, (5.2) reduces to the ordinary diffusion propagator.

Let us consider an aggregation in which diffusing particles, described in (5.2), stick together to form clusters. In this section we assume that the coalescence probability is independent of the sizes of the clusters. Following the procedure in the previous section, we obtain the effective kernel in a steady state as

$$\Gamma \simeq D\left(\frac{\beta \sin(\pi d/\beta)}{\pi K_d}\right)^{\beta/d} (4n)^{(\beta-d)/d}. \tag{5.3}$$

From (5.3) and the discussion in the previous section a power-law size distribution is derived. In such a case, the power exponent of time $\varepsilon/2$ in (4.9a) is replaced by $(\beta - d)/\beta$, and ε and z in (4.11) are replaced by $\beta - d$ and $z' = d/\beta(\tau - 1)$, respectively. Thus we obtain the power exponent of size $\tau = (\beta + 2d)/(\beta + d)$ for $d < \beta$ and $\tau = \frac{3}{2}$ for $d > \beta$.

Takayasu *et al* (1988) simulated the one-dimensional aggregation of particles undergoing Lévy flight, where they discussed a relation between the exponent of cluster size

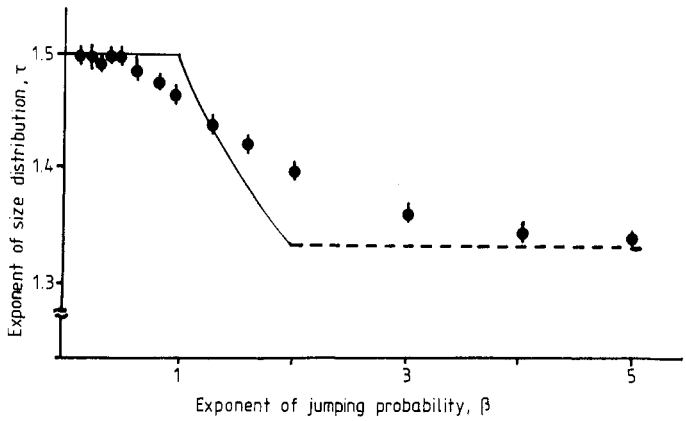

Figure 3. The relation between the exponent characterising the jumping probability β and the exponent of the power of the size distribution τ. The full circles and error bars are obtained by the simulation (Takayasu *et al* 1988). The full curve is predicted by (5.4). The broken line is given by extrapolation using the effective dimension $d_{\text{eff}} = 1$.

distribution and the value of the parameter β (see figure 3). In our theory we predict the exponent of size distribution

$$\begin{aligned} \tau &= \frac{2+\beta}{1+\beta} \qquad (1 < \beta \leq 2) \\ \tau &= \tfrac{3}{2} \qquad (\beta < 1) \end{aligned} \tag{5.4}$$

where $\tau = \frac{3}{2}$ represents a mean-field description because one dimension is larger than the upper critical dimension β. In the case of $\beta = 1$ there is a logarithmic correction for the size distribution function. When β is larger than 2, the system cannot be described by the propagator (5.2). We expect that the results for the case $\beta \geq 2$ reduce to those of $\beta = 2$, because Lévy flight is characterised by an effective dimension $d_{\text{eff}} = 1$ for $\beta \geq 2$ (Takayasu *et al* 1988). The theoretical values do not agree with the numerical results of Takayasu *et al* (1988). Such disagreements are probably based on the numerical error as mentioned in their paper.

6. Discussion and conclusion

As discussed by Rácz (1985a, b) there is a similarity between source-enhanced aggregation and traditional critical phenomena. Our analysis shows a connection between source-enhanced aggregation and critical phenomena for the following reasons. When we assume $h = 0$, then the number density of clusters disappears in a long-time limit. When we impose an external source, the number density appears, namely the number density can be regarded as the order parameter under the source h.

The method used in §4.2 suggests another relation to critical phenomena. Source-enhanced aggregation has a mass flux into a sink. On the other hand, there is a mass flux into an infinite cluster in the course of gelation. It is an interesting point that a source-enhanced aggregation system maintains a 'critical state', due to the balance between a source and a sink. Such a steady state seems to be one of the reasons there are so many fractal objects in nature.

It seems natural that source-enhanced aggregation has scaling properties, because this process is a kind of cascade process. Let us compare source-enhanced aggregation with three-dimensional homogeneous turbulence (Levich 1987). The turbulence occurs at extremely large Reynolds number (Re) and has fractal structures in the inertial range. The system attains a steady state by a balance of the creation of vortices and the energy dissipation, with characteristic lengths L(~system size) and l_d ($\sim Re^{-3/4}L$) respectively. As the Reynolds number goes to infinity, i.e. $l_d \to 0$, the system exhibits a universal behaviour. When we see the cascade process of turbulence in k space, there is a source with the small k value and a sink with the large k. These situations correspond to the case of source-enhanced aggregation in size space. Therefore, we can extract universal properties when the cutoff size tends to infinity.

Let us note that the spatial dimension expresses the effective dimension of the diffusion field, i.e. the aggregation under strong anisotropic diffusion can be regarded as the low-dimensional case. Thus it is important to analyse the coagulation of systems of lower (in general, fractional) dimensions.

We now summarise our results. In this paper, we analyse source-enhanced aggregation based on the Fock space formalism and obtain the size distribution which obeys a power law. We consider a logarithmic correction when the spatial dimension is 2. In addition, we give the answer to some of the questions posed by Takayasu *et al* (1988).

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