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Cluster structures and cluster–cluster aggregations in a two-dimensional ferromagnetic colloidal system

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Abstract. We studied cluster structures and cluster aggregations in a two-dimensional ferromagnetic colloidal system numerically. We developed a Brownian dynamics calculation method in which both the translational and rotational motions of ferromagnetic particles were taken into account. The dependence of the cluster structures on the nondimensional parameters λ and ξ was investigated, where λ is the ratio of the magnetic dipole energy of a particle to thermal energy and ξ is the ratio of the interactive energy between the dipole moment and the external magnetic field to thermal energy. The fractal dimension was 1.3 in the absence of a magnetic field irrespective of λ. On the other hand, the fractal dimension was very close to 1.0 when the system was subjected to a strong magnetic field. Cluster–cluster aggregations were also investigated and the validity of the dynamic scaling law was examined. The exponent in the dynamic scaling law was obtained as a function of λ. It has been found that the exponent increases with λ and becomes constant when $\lambda > 12$ but that the fractal dimension does not change with λ. The values of the fractal dimensions and the exponents obtained by the present simulation are compared with those obtained by other experiments and model simulations.

1. Introduction

It is known that ferromagnetic particles dispersed in a solvent fluid form clusters under certain conditions. The main parameters which govern the cluster formations and structures are the concentration of the particles, the temperature of the system, the magnetic dipole moment of a particle, the diameter of a particle and the strength of an external magnetic field. It is very important from a theoretical point of view to make clear the mechanism of the cluster formations and the mode of the cluster–cluster aggregations (CCAs) because it is believed that there may be universal laws in the mechanism and the mode [1]. It is also necessary to make clear the mechanism and the mode from a practical point of view as the structures of clusters determine the rheological or optical characteristics of ferromagnetic colloids [2–5]. Cluster formations in magnetic colloidal systems have been investigated experimentally, theoretically and numerically based on statistical mechanics, the Brownian dynamics method and the Stokesian dynamics method by several researchers. Helgesen *et al* [6], Promislow *et al* [7], Fermiger and Gast [8], Jansen *et al* [9], Kim and Brock [10], Liu *et al* [11] and Hagenbüchle and Liu [12] carried out experimental investigations of cluster structures of ferromagnetic or paramagnetic colloidal systems. Gennes and Pincus [13], Sano and Doi [14], Jordan [15] and

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Hayter and Pynn [16] analysed cluster structures by statistical mechanics. Satoh *et al* [17] and Ikuhara and Cawley [18] analysed cluster structures of ferromagnetic colloids by the Monte Carlo method. Mohebi *et al* [19] studied the cluster structure formation in magnetorheological fluids by the Stokesian dynamics method. Models of irreversible growth of linear chains by CCAs have been developed by Miyazima *et al* [20], Eriksson and Jonson [21] and Mors *et al* [22]. However, the cluster growth process and the dependence of the cluster structures on parameter λ are still open questions. The following points are still unknown and therefore must be investigated: (1) Does the fractal dimension of ferromagnetic clusters change with λ? (2) Does the dynamic scaling law, that is $n_s(t) \sim t^{-w}s^{-\tau}f(s/t^z)$ where n_s, t, s and f are, respectively, the number of clusters which are composed of s particles, time, the number of particles in a cluster and a scaling function, apply to the cluster growth of ferromagnetic particles? If the dynamic scaling law applies, the exponent z determines the cluster growth rate as shown in section 3.2 and therefore the dependence of z on λ must be made clear.

The purpose of this paper is to analyse the cluster formations and structures in a ferromagnetic colloidal system by the Brownian dynamics method and to make clear the effect of parameters λ and ξ on the fractal dimensions of the clusters and the exponent z in the dynamic scaling law. In the second section, we develop a Brownian dynamics method in which both translational and rotational motions of ferromagnetic particles are taken into account. The potential energy between two particles is described and the numerical technique and procedure are explained. In the third section, the cluster formations are analysed. The effect of parameters λ and ξ on the fractal dimensions of the clusters are investigated. CCAs are also analysed. The validity of the dynamic scaling law is investigated and discussed. In the final section, the result of the analysis is summarized.

2. Analysis

In this section, a model of a ferromagnetic colloidal system is introduced and the numerical procedure is described.

2.1. Modelling

As we focus on the motions of colloidal particles, we make a model of a ferromagnetic colloidal system based on the Langevin equation. In this case, the effect of the collisions of solvent molecules with ferromagnetic particles are represented by a white noise. The Langevin equations for both translational and rotational motions are expressed as follows:

$$\frac{\partial \boldsymbol{u}}{\partial t} = -\frac{\zeta_t}{m}\boldsymbol{u} + \frac{\boldsymbol{F}}{m} + \boldsymbol{P} \tag{1}$$

$$\frac{\partial \boldsymbol{\omega}}{\partial t} = -\frac{\zeta_r}{I}\boldsymbol{\omega} + \frac{\boldsymbol{T}}{I} + \boldsymbol{\tau} \tag{2}$$

where $\boldsymbol{u}$ and $\boldsymbol{\omega}$ are, respectively, the velocity and angular velocity of a particle; m is the mass and I is the moment of inertia. $\boldsymbol{F}$ and $\boldsymbol{T}$ are, respectively, the force and torque acting on a particle through the interaction with the other particles and an external magnetic field; ζ_t and ζ_r are the friction coefficients. We assume that Stokes law applies to the coefficients. $\boldsymbol{P}$ and $\boldsymbol{\tau}$ are the random force and torque caused by collisions with solvent molecules. The mean values and autocorrelations of the random force and torque satisfy the following relations:

$$\langle \boldsymbol{P}(t) \rangle = \langle \boldsymbol{\tau}(t) \rangle = 0 \tag{3}$$

$$\langle \boldsymbol{P}(t) \cdot \boldsymbol{P}(t + \Delta t) \rangle = \frac{6\zeta_t kT}{m^2}\delta(\Delta t) \tag{4}$$

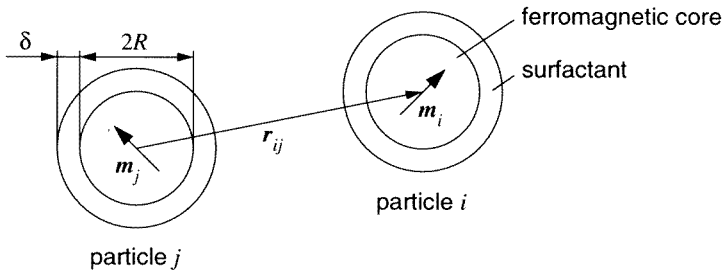

Figure 1. Ferromagnetic particles. Each particle is composed of a single magnetic domain and the surface is coated with a surfactant material.

$$\langle \boldsymbol{\tau}(t) \cdot \boldsymbol{\tau}(t + \Delta t) \rangle = \frac{6\zeta_r kT}{I^2} \delta(\Delta t) \tag{5}$$

where $\langle \cdots \rangle$ represents the time average. k and T are the Boltzmann constant and temperature and δ is the Dirac delta function.

2.2. Potential energy

As is shown in figure 1, we assume that ferromagnetic particles are spherical and the surface is coated with a surfactant material. Each particle is composed of a single magnetic domain and therefore the magnetic interaction between particles is expressed by a dipole potential u_m [13]. Furthermore, we take into account potentials based on the van der Waals attraction u_v and the repulsion caused by surfactant–surfactant contact u_s [23].

$$u_m = -\boldsymbol{m}_i \cdot \boldsymbol{H}_{ij} = \frac{1}{4\pi\mu_0} \left\{ \frac{\boldsymbol{m}_i \cdot \boldsymbol{m}_j}{r_{ij}^3} - \frac{3}{r_{ij}^5} (\boldsymbol{m}_i \cdot \boldsymbol{r}_{ij})(\boldsymbol{m}_j \cdot \boldsymbol{r}_{ij}) \right\} \tag{6}$$

$$u_v = -\frac{A}{6} \left\{ \frac{2R^2}{r_{ij}^2 - 4R^2} + \frac{2R^2}{r_{ij}^2} + \ln\left(\frac{r_{ij}^2 - 4R^2}{r_{ij}^2} \right) \right\} \tag{7}$$

$$u_s = 2\pi R^2 N k T \left\{ 2 - \frac{\boldsymbol{r}_{ij} - 2R}{\delta} - \frac{\boldsymbol{r}_{ij}}{\delta} \ln\left(\frac{2R + 2\delta}{\boldsymbol{r}_{ij}} \right) \right\} \tag{8}$$

where μ_0, $\boldsymbol{m}_i$, R, δ, A and N are, respectively, the permeability, the magnetic moment of particle i, the radius of the particle, the thickness of the surfactant layer, the Hamaker constant and the surface density of the surfactant molecules. $\boldsymbol{H}_{ij}$ is the magnetic field at the position of particle i produced by particle j. The interactive energy between a particle and an external magnetic field is expressed as

$$u_H = -\boldsymbol{m}_i \cdot \boldsymbol{H} \tag{9}$$

where $\boldsymbol{H}$ is the external magnetic field.

$\boldsymbol{F}$ and $\boldsymbol{T}$ in equations (1) and (2) are obtained as follows:

$$\boldsymbol{F} = -\sum_{j=1(j\neq i)}^{n} \frac{\partial}{\partial \boldsymbol{r}_{ij}} (u_m + u_v + u_s) \tag{10}$$

$$\boldsymbol{T} = \boldsymbol{m}_i \times \left(\sum_{j=1(j\neq i)}^{n} \boldsymbol{H}_{ij} + \boldsymbol{H} \right) \tag{11}$$

where n is the total number of particles. Although we included the van der Waals attraction, the system is a so-called soft-sphere dipolar fluid: that is, the dipole interactions and short-range repulsion are dominant compared with the van der Waals attraction.

Table 1. Calculation conditions.

Diameter of ferromagnetic core	10 nm
Thickness of surfactant	2 nm
Mass of particle	2.9×10^{-21} kg
Surface density of surfactant molecules	10^{18} molecules m^{-2}
Dynamic viscosity of solvent	10^{-3} Pa s
Hamaker constant	10^{-19} J
Temperature of solvent	300 K
Number density of particles	121 particles (0.352 μm)^2

2.3. Numerical technique and procedure

The calculation conditions and physical properties are summarized in table 1. We carried out calculations assuming that the diameter of the particles was 10 nm and the thickness of the surfactant layer was 2 nm. Nondimensional parameters λ and ξ are defined as

$$\lambda \equiv \frac{|\boldsymbol{m}|^2}{4\pi\mu_0 d^3 kT} \tag{12}$$

$$\xi \equiv \frac{|\boldsymbol{m}|H}{kT} \tag{13}$$

where d is the diameter of a particle, λ is the ratio of the magnetic dipole moment energy to thermal energy and ξ is the ratio of the interactive energy between the dipole moment and the external magnetic field to thermal energy.

121 particles were placed in a 0.352 μm $\times$ 0.352 μm cell. Periodic boundary conditions were employed to overcome the surface problem. The cut-off distance of the interactive potentials was 80 nm, that is eight times longer than the particle diameter. A uniform magnetic field was applied to the square cell and the effect of the magnetic field on the cluster structures was analysed. Because of the random force and torque (see equations (1)–(5)), the system becomes canonical. The Langevin equations (1) and (2) were integrated based on Ermak's procedure [24]. The integration procedure is summarized in the appendix. The time step of the calculation was 10 ps.

3. Result and discussion

The calculation was successfully carried out without any numerical instabilities. In this section, the result of the numerical calculation is analysed and the structures of clusters and the CCA modes are investigated.

3.1. Steady-state cluster structures

We calculated the pair-correlation functions of particles for different λ and defined the cluster formations as follows: if the distance between two particles is shorter than $1.2d$, where d is the diameter of a particle, those two particles form a cluster. This definition is correct for the following reason: there was little effect of λ and ξ on the first and second peak positions in the pair-correlation functions; the first peak position $= 0.98d$ and the second peak position $= 1.96d$. The standard deviation was less than $0.04d$ in both cases. The position corresponding to the minimum point between the first and second peak positions was $1.55d \pm 0.12d$. Therefore, the distance, r_c, which defines the cluster formation, has to be $0.98d < r_c < 1.55d$. We carried out simulations changing the distance $r_c = 1.1d$, $1.2d$,

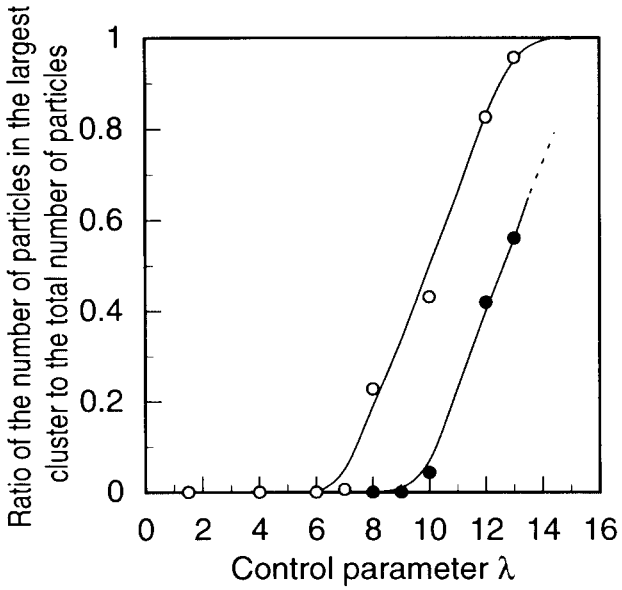

Figure 2. Dependence of the ratio of the number of particles in the largest cluster to the total number of particles in the system on control parameter λ. Data are shown for $\xi = 0$ (●) and $\xi = 30$ (○).

$1.5d$. The cluster growth process did not change even if the definition was changed as long as $0.98d < r_c < 1.55d$.

After the system had reached a steady state, the cluster structures were investigated. The ratio of the number of particles in the largest cluster to the total number of particles in the system was calculated. The dependence of the ratio on λ is shown in figure 2. The ratio starts increasing at $\lambda \sim 9$ in the absence of a magnetic field ($\xi = 0$), whereas the corresponding value is $\lambda \sim 7$ in the magnetic field ($\xi = 30$).

The fractal dimension of the clusters in the system can be calculated from the relation between the number of particles in a cluster and the cluster radius [1]. The fractal dimensions in the absence of a magnetic field are shown in figures 3(*a*) and (*b*) for $\lambda = 10$ and 13. The fractal dimensions in the presence of a magnetic field are shown in figures 3(*c*) and (*d*) for $\lambda = 8$ and 13. The fractal dimensions are almost constant despite any changes in parameter λ in both cases. The fractal dimensions are, respectively, 1.3 and 1.0 in the absence of a magnetic field and in the magnetic field.

3.2. Cluster–cluster aggregations

The CCA mode was analysed from the time histories of the cluster size and the number of clusters. The time variations of cluster aggregations are shown in figure 4. After the particle–particle diffusion growth in the early stage, CCAs are observed. The clusters are curved in the absence of a magnetic field, while straight clusters are formed in the magnetic field. When λ is larger than the critical value, the size of the largest cluster formed in the system reaches the system size (see also figure 2).

As was mentioned above, after the particle–particle diffusion growth, the clusters grow through the CCAs. According to the dynamic scaling law [25], the number of clusters which

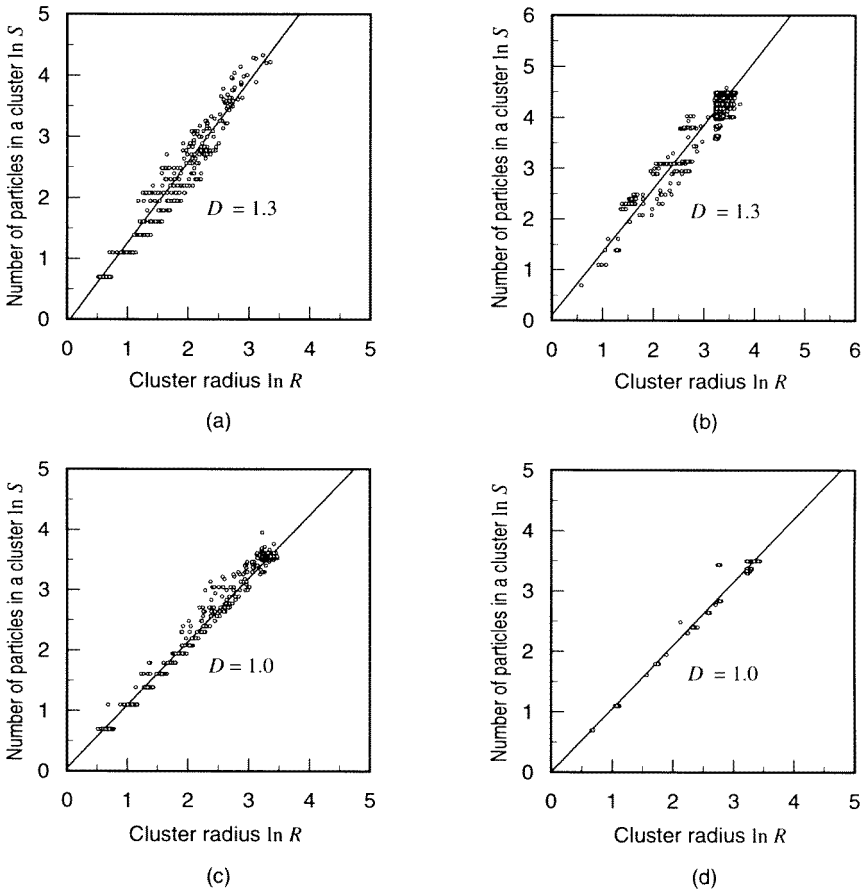

Figure 3. Relation between the number of particles in a cluster and the cluster radius: (*a*) $\xi = 0$, $\lambda = 10$; (*b*) $\xi = 0$, $\lambda = 13$; (*c*) $\xi = 30$, $\lambda = 8$; (*d*) $\xi = 30$, $\lambda = 13$. The slope gives the fractal dimension. The fractal dimension is 1.3 in the absence of a magnetic field ($\xi = 0$) and 1.0 in the magnetic field ($\xi = 30$).

are composed of s particles, $n_s(t)$, is expressed by the following relation:

$$n_s(t) \sim t^{-w} s^{-\tau} f(s/t^z) \tag{14}$$

where t and s are, respectively, time and the number of particles in a cluster; f is a scaling function and w, τ and z are the exponents which characterize the cluster growth process. If the dynamic scaling relation (14) is correct, the second moment $S(t)$ (the average number of particles in a cluster), the mean radius of inertia of a cluster, $R(t)$, and the zeroth moment, $N(t)$ (that is, the total number of clusters) change with time as follows [25, 26]:

$$S(t) = \frac{\sum_s n_s(t) s^2}{\sum_s n_s(t) s} \sim t^z \tag{15}$$

$$R(t) \sim S(t)^{\frac{1}{D}} \sim t^{\frac{z}{D}} \tag{16}$$

$$N(t) = \sum_s n_s(t) \sim \begin{cases} t^{-z} (\tau < 1) \\ t^{-w} (\tau > 1) \end{cases} \tag{17}$$

where D is the fractal dimension.

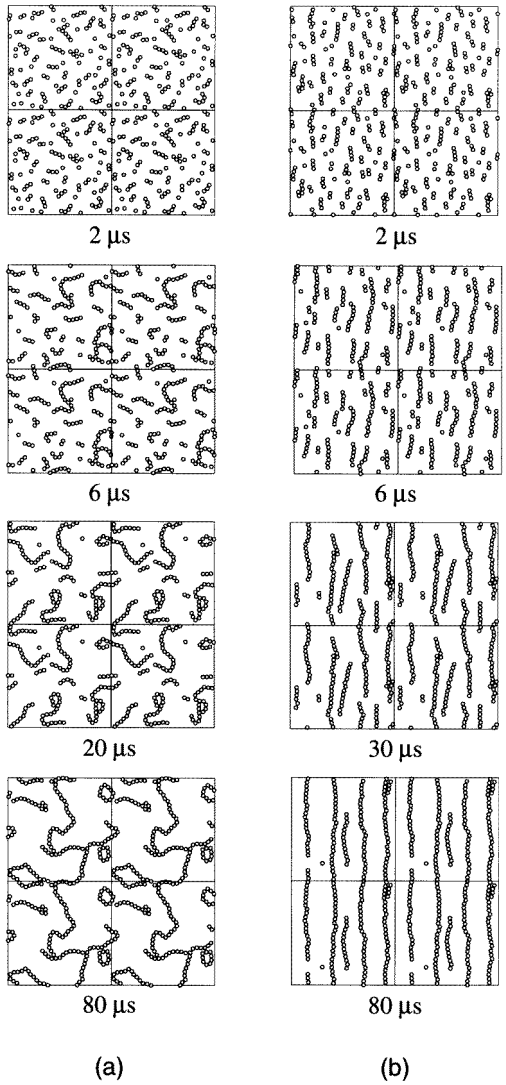

Figure 4. Time variations of cluster aggregations: (*a*) $\xi = 0$, $\lambda = 13$; (*b*) $\xi = 30$, $\lambda = 13$. After the particle diffusion growth in the early stage, clusters grow through CCAs. Clusters are curved in the absence of a magnetic field ($\xi = 0$) and straight in the magnetic field ($\xi = 30$).

The time variations of $S(t)$, $R(t)$ and $N(t)$ are shown in figure 5. The exponent z calculated by equation (15) coincides with that calculated by equation (17), which means that $\tau < 1$ (see equation (17)). The particle–particle aggregations and CCAs are determined by the diffusion-limited process; $S(t)$ increases with time as $S(t) \sim t^z$ and $N(t)$ decreases with time as $N(t) \sim t^{-z}$. These relations are also confirmed by a phenomenological model [20] and an experiment of paramagnetic colloidal system in a magnetic field [8]. We also carried out simulations with a larger system of 900 particles per 0.960 μm $\times$ 0.960 μm and confirmed that the values of z were the same as those obtained in the original smaller system. Note that the standard deviations of the exponent z was within 10% of the average values.

The fractal dimensions calculated by equation (16) using the exponent z obtained by equations (15) and (17) are very close to those of clusters at the steady state. The fractal structures during the CCAs are almost the same as those at the steady state. In our simulations, the fractal dimension was 1.3 in the absence of a magnetic field for $8 \leqslant \lambda \leqslant 13$. According

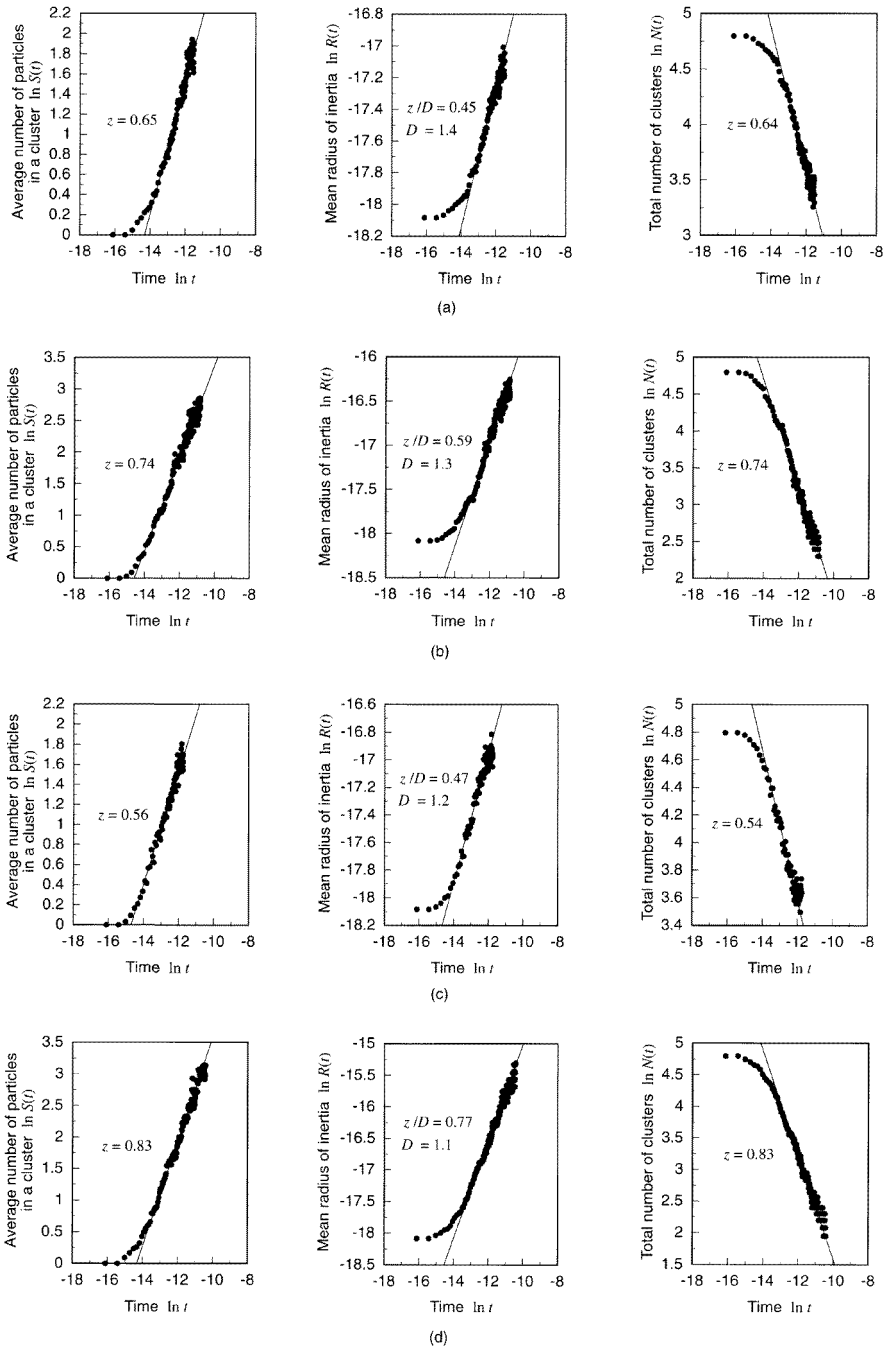

Figure 5. Time variations of the average number of particles in a cluster $S(t)$, the mean radius of inertia of a cluster $R(t)$ and the total number of clusters in the system $N(t)$: (*a*) $\xi = 0$, $\lambda = 10$; (*b*) $\xi = 0$, $\lambda = 13$; (*c*) $\xi = 30$, $\lambda = 8$; (*d*) $\xi = 30$, $\lambda = 13$. $S(t)$ and $N(t)$ change with time as $S(t) \sim t^z$ and $N(t) \sim t^{-z}$. The fractal dimensions of clusters during CCA are almost the same as those at the steady state.

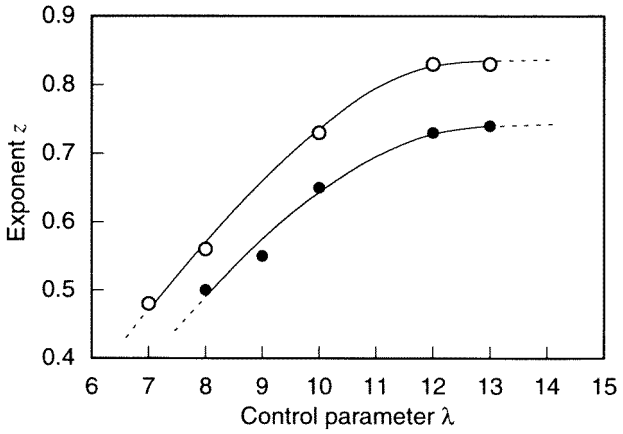

Figure 6. Dependence of exponent z on control parameter λ. Data are shown for $\xi = 0$ (●) and $\xi = 30$ (○). z increases with λ and becomes constant when $\lambda > 12$.

to statistical analysis carried out by Morimoto and Maekawa [27], the fractal dimension of clusters in the absence of a magnetic field in a two-dimensional system is $\frac{4}{3}$ irrespective of λ, which agrees with our present result. Helgesen *et al* carried out two-dimensional experiments and reported D in the range from 1.49 ± 0.06 to 1.16 ± 0.15 for $\lambda = 0$–1360 [6]. If we focus on the fractal dimension for $8 \leqslant \lambda \leqslant 13$, $D = 1.47$–1.46. The fractal dimension obtained by Helgesen *et al* is slightly higher than ours and is almost constant for $8 \leqslant \lambda \leqslant 13$. The SAW model and the tip-to-tip model [28] showed $D = \frac{4}{3}$ and 1.28 ± 0.03, respectively, in a two-dimensional case. These values are very close to our value, 1.3. Because of the highly directional dipole–dipole interactions, clusters tend to be formed through tip-to-tip or tail-to-end coagulations as in the case of the tip-to-tip model. According to the chain–chain aggregation model [29], $D = 1.32 \pm 0.04$ in a two-dimensional case. The value of the fractal dimension is almost the same as ours. A cluster aggregation model in which dipole–dipole interactions are taken into account was also developed by Helgesen *et al* for $\lambda \to \infty$ [6]. The fractal dimension was 1.23 ± 0.12.

The dependence of the exponent z on parameter λ is shown in figure 6. The exponent increases with λ and becomes constant when $\lambda \geqslant 12$ although the fractal dimension is constant despite any changes to λ. The exponent in the magnetic field is larger than that in the absence of a magnetic field. A similar dependence of the exponent z on the nondimensional parameter λ is obtained by experiment [6]. CCA processes in the absence of a magnetic field were observed by Helgesen *et al* [6] in a two-dimensional system. Although z increased with λ, which agrees with our result, z was 1.7 ± 0.2 for $\lambda = 1360$. The value of z is much larger than ours. We assume that in their experiment, the three-dimensional motion of magnetic moment encouraged the aggregations. Note that the diameter of the magnetic particle was 3.6 μm and the depth of the liquid layer was 5 μm in their experiment. According to Morimoto and Maekawa [30], the exponent in a three-dimensional system is larger than that in a two-dimensional system. Promislow *et al* [7] reported that z decreases with λ, mentioning that chain clusters are elastically bent when λ is small. However, such flexible chain clusters were not observed in our simulations. The chain–chain aggregation model gives $z = 0.72 \pm 0.02$ in a two-dimensional case [29], which agrees with our z for $\lambda > 12$. Hagenbüchle and Liu [12] measured the time history of cluster length in a magnetic field by the dynamic scattering method and obtained $z/D = 0.78$ for $22.5 < \lambda < 263$, which agrees with our simulation

result; $z/D = 0.77$ for $\lambda = 12$ and $\xi = 30$. Although Hagenbüchle and Liu's result was obtained in a three-dimensional system, the value of z/D in a three-dimensional system is the same as that in a two-dimensional system when the intensity of magnetic field is high [30]. Miyazima *et al* [20] investigated CCAs by a phenomenological model and mean field theory in which particles were allowed to aggregate only in one direction, which is very similar to cluster aggregations in a magnetic field. They reported that mean field theory applied to the coagulation process when the system dimension d was 2 and 3 and therefore the exponent z for $d = 3$ was the same as that for $d = 2$.

According to our simulations, the rate of the dissociations of clusters is almost the same as that of the aggregations of clusters when λ is small. In other words, the cluster growth process is reversible, which explains why z is small when λ is small. When λ is large, however, the interparticle potential becomes strong and therefore clusters cannot dissociate. The cluster growth process changes from reversible to irreversible mode as λ increases. This process change corresponds to the saturation point of z.

4. Conclusions

We studied cluster structures and CCAs in a two-dimensional ferromagnetic colloidal system numerically. We developed a Brownian dynamics method in which both the translational and rotational motions of particles were taken into account. The calculations were successfully carried out. The fractal dimensions were, respectively, 1.3 and 1.0 in the absence of a magnetic field and in the magnetic field, irrespective of λ. CCAs were also investigated. We found that the dynamic scaling relation expressed by equation (14) applies to the CCA of ferromagnetic particles and, as a result, the power laws expressed by equations (15)–(17) apply to the time variations of the average number of particles, the mean radius of inertia of a cluster and the total number of clusters. The fractal dimensions of clusters during the CCA process coincided with those at the steady state. Although the fractal dimension was constant despite any changes to λ, the exponent z increased with λ and became constant when $\lambda \geqslant 12$.

Acknowledgments

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Appendix A. Procedure of numerical integration

We integrated the Langevin equations numerically. The integration procedures are as follows:

(1) Integration of the Langevin equation of the translational motion;

$$\boldsymbol{u}(t+\Delta t) = \mathrm{e}^{-\beta_t \Delta t}\boldsymbol{u}(t) + \frac{\boldsymbol{F}(t)}{m\beta_t}(1-\mathrm{e}^{-\beta_t \Delta t}) + \boldsymbol{\Psi}_u \tag{A.1}$$

$$\boldsymbol{r}(t+\Delta t) = \boldsymbol{r}(t) + \frac{1}{\beta_t}\frac{1-\mathrm{e}^{-\beta_t \Delta t}}{1+\mathrm{e}^{-\beta_t \Delta t}}\left\{\boldsymbol{u}(t+\Delta t) + \boldsymbol{u}(t) - 2\frac{\boldsymbol{F}(t)}{m\beta_t}\right\} + \frac{\boldsymbol{F}(t)}{m\beta_t}\Delta t + \boldsymbol{\Psi}_r \tag{A.2}$$

where $\beta_t = \zeta_t/m$ and $\boldsymbol{\Psi}_u$ and $\boldsymbol{\Psi}_r$ are the normalized random vectors which satisfy the

following relations:

$$\begin{aligned}
&\langle \boldsymbol{\Psi}_u \rangle = \langle \boldsymbol{\Psi}_r \rangle = 0 \\
&\langle \boldsymbol{\Psi}_u \cdot \boldsymbol{\Psi}_r \rangle = 0 \\
&\langle |\boldsymbol{\Psi}_u|^2 \rangle = \frac{3kT}{m}\{1 - \mathrm{e}^{-2\beta_t \Delta t}\} \\
&\langle |\boldsymbol{\Psi}_r|^2 \rangle = \frac{6kT}{m\beta_t^2}\left\{\beta_t \Delta t - 2\frac{1 - \mathrm{e}^{-\beta_t \Delta t}}{1 + \mathrm{e}^{-\beta_t \Delta t}}\right\}.
\end{aligned} \tag{A.3}$$

(2) Integration of the Langevin equation of the rotational motion;

$$\boldsymbol{\omega}(t + \Delta t) = \mathrm{e}^{-\beta_r \Delta t}\boldsymbol{\omega}(t) + \frac{\boldsymbol{T}(t)}{I\beta_r}(1 - \mathrm{e}^{-\beta_r \Delta t}) + \boldsymbol{\Psi}_\omega \tag{A.4}$$

$$\Delta\boldsymbol{\Omega} = \frac{1}{\beta_r}\frac{1 - \mathrm{e}^{-\beta_r \Delta t}}{1 + \mathrm{e}^{-\beta_r \Delta t}}\left\{\boldsymbol{\omega}(t + \Delta t) + \boldsymbol{\omega}(t) - 2\frac{\boldsymbol{T}(t)}{I\beta_r}\right\} + \frac{\boldsymbol{T}(t)}{I\beta_r}\Delta t + \boldsymbol{\Psi}_{\Delta\Omega} \tag{A.5}$$

where $\beta_r = \zeta_r / I$ and $\boldsymbol{\Psi}_\omega$ and $\boldsymbol{\Psi}_{\Delta\Omega}$ are the normalized random vectors which satisfy the following relations:

$$\begin{aligned}
&\langle \boldsymbol{\Psi}_\omega \rangle = \langle \boldsymbol{\Psi}_{\Delta\Omega} \rangle = 0 \\
&\langle \boldsymbol{\Psi}_\omega \cdot \boldsymbol{\Psi}_{\Delta\Omega} \rangle = 0 \\
&\langle |\boldsymbol{\Psi}_\omega|^2 \rangle = \frac{3kT}{I}\{1 - \mathrm{e}^{-2\beta_r \Delta t}\} \\
&\langle |\boldsymbol{\Psi}_{\Delta\Omega}|^2 \rangle = \frac{6kT}{I\beta_r^2}\left\{\beta_r \Delta t - 2\frac{1 - \mathrm{e}^{-\beta_r \Delta t}}{1 + \mathrm{e}^{-\beta_r \Delta t}}\right\}.
\end{aligned} \tag{A.6}$$

The magnetic dipole moment vector $\boldsymbol{m}$ is calculated by the following equation:

$$\boldsymbol{m}(t + \Delta t) = \boldsymbol{m}(t)\cos\Delta\phi + \boldsymbol{n}(\boldsymbol{n} \cdot \boldsymbol{m}(t))(1 - \cos\Delta\phi) + (\boldsymbol{n} \times \boldsymbol{m}(t))\sin\Delta\phi \tag{A.7}$$

where $\Delta\phi = |\Delta\boldsymbol{\Omega}|$ and $\boldsymbol{n} = \Delta\boldsymbol{\Omega}/\Delta\phi$.

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