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Phase transition of binary-component DLA (diffusion-limited aggregation)–SQL (square lattice) system contacted with thermal bath in the perimeter

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Abstract. To make the DLA model associate with more realistic processes of crystal growth, we construct and study a new model including the experimental variables required. Diffusion particles of multicomponents diffuse on the square lattice as in the DLA model; the aggregation perimeter contacts with a thermal bath of temperature T and the sticking probability $\mathscr{P}$ consists of a constant probability $\mathscr{P}_c$ and the thermal one $\mathscr{P}_t$ at a neighbouring site of the perimeter, as $\mathscr{P}=(1-\alpha)\mathscr{P}_c+\alpha\mathscr{P}_t$ (α is a parameter which includes the non-equilbrium-equilibrium tendency of the system).. $\mathscr{P}_t$ is evaluated by the thermodynamic distribution of the Ising system including up to next-nearest-neighbour interactions and chemical potentials. Our system has the possibility of phase transitions. We show the phase transitions, aggregation patterns, correlation functions, fractal dimensions, and so on.

1. Introduction

Many substances in nature form individual, peculiar patterns. Some of the most beautiful and familiar examples can be found in the growth of crystals [1, 2]. The basic interests in the pattern formation problem are, for example, (i) how the characteristic patterns appear in a structureless environment; (ii) how the original patterns depend on certain circumstances and change into other patterns from the static and dynamic points of view, including their special sensitivities; and (iii) what are the order parameters (OP) which distinguish these characteristic patterns. Within the past few years, remarkable progress has been made in understanding structure (pattern) formations in various random process systems. Recent developments are summarised in monographs by Stanley and Ostrowsky [3] and Pietronero and Tosatti [4]. Their works are connected with pattern formation processes over many fields, e.g. the Eden model, diffusion-limited aggregation (DLA) and epidemics, including the mole's labyrinth and clustering of clusters for colloids, aerosols, etc. Their model systems are greatly simplified and are constructed with the smallest number of parameters still able to distinguish their model differences. Their main interest is in studying the growth patterns and comparing them with certain natural forms. Furthermore, it is interesting to look for a complete set of order parameters to describe the pattern formation

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processes and to determine the corresponding fractal (anomalous) dimensions. These works have succeeded in some sense as simplified systems and have approached their goals for the initial stage. However, from the microscopic point of view, the systems do not undergo phase transitions, and from the macroscopic point of view they are too simplified to be compared with natural patterns (e.g. crystal growth forms). Generally, crystal growth processes depend strongly on symmetries of atomic arrangements and surface anisotropies, and therefore on those near-equilibrium properties which are dominated by atomic and crystallographic effects. Their formation processes are intrinsically concerned with non-equilibrium phenomena and have no internally controlling mechanism. The growth at any instant must be controlled by a simple set of spatially uniform external conditions, e.g. the temperature, the concentrations of various atoms and the pressure. In the relatively well known example of the snowflake, the crystal growth processes are separated into several steps, i.e. (i) strong, (ii) weak and (iii) intermediate steps of molecular binding at the crystallographic planes, and show different behaviour. We propose to go beyond the initial stage and try to make the above-mentioned random process approaches more realistic. That is, we construct new more physical models which include the experimental variables required, and we discuss their physical properties.

In the present paper we consider the DLA model. This was studied by, among others, Witten and Sander [5] and Meakin [6] and has been pointed out as a central problem in many fields of applied science, e.g. dielectric breakdown and viscous fingering (see the review articles by Nittmann *et al* [7] and Bensimon *et al* [2]). Actual crystal growth processes are strongly associated with the concentrations, the interparticle interactions and the chemical potentials of the composite particles, together with the temperature and the pressure. It is especially important to take into account the frustration mechanism among sticking particles, in addition to the introduction of the experimentally changeable variables [8]. The diffusion particles of the system consist of multicomponents $\{i\}$ with concentrations $\{c_i\}$ $(i = 1, 2, \ldots, M)$. In this paper we consider a binary component system $(M = 2)$ on the square (SQ) lattice for simplicity. After a component and some starting position of a diffusion particle are randomly selected, it randomly diffuses on the lattice in the same way as the usual DLA model. Generally, the sticking probability is supposed to correlate strongly with the temperature, the competition of the short-ranged interactions and the chemical potentials at the surface of the aggregation cluster (seed) (which is called the aggregation perimeter below), in addition to the constant probability. The irreversible (in the sense that stuck particles never dissociate) sticking probability $\mathscr{P}$ is assumed to consist of a constant probability $\mathscr{P}_c$ and a thermal probability $\mathscr{P}_t$ at a neighbouring site of the perimeter, as $\mathscr{P} = (1-\alpha)\mathscr{P}_c + \alpha\mathscr{P}_t$ with a parameter α. The thermal probability is assumed to be described with the Ising system energy including up to next-nearest-neighbour interactions and chemical potentials. If a diffusion particle visits a site adjacent or next adjacent to the aggregation cluster and its sticking probability is larger than the random number, then the particle constructs the aggregation cluster. Otherwise it is removed and another introduced. Our system has the following feature. It corresponds to the usual DLA model (not contacted with a thermal bath) for $\alpha = 0$, and to the thermal DLA system (completely contacted with a thermal bath) for $\alpha = 1$. Here we conventionally call the former the non-equilibrium DLA, the latter the equilibrium DLA, and the remaining the non-equilibrium–equilibrium DLA. Our system suggests the possibility of phase transitions which belong to a new type of phase transition associated with non-equilibrium–equilibrium states. It is also a very interesting system which we may

approach both from the DLA-like (non-equilibrium, finite size) point of view and from the statistical-mechanics-like (equilibrium, infinite size) point of view. As a first step we focus our attention on the phase transitions and the DLA-like properties (the aggregation patterns, the correlation functions and the fractal dimensions) of the system for a few typical physical parameters. In § 2 we define our new system and the computing method, in § 3 the physical properties obtained are summarised and we make some concluding remarks in § 4.

2. System and method

Our system is a variant of the DLA model and consists of binary atoms A and B. We form a set of physical parameters: concentrations $\{c_A, c_B(=1-c_A)\}$, temperature T, nearest- and next-nearest-neighbour interactions $\{J_1, J_2\}$ and chemical potentials $\{h_A, h_B\}$, together with constant sticking probability $\mathscr{P}_c$ and the non-equilibrium-equilibrium parameter α. As in the initial state an A-atom seed is put at the origin on the SQ lattice. At first diffusion, the particle (A or B atom), randomly chosen under the fixed concentrations, is added at some random site at a large distance from the origin. This particle walks randomly until it visits a site which is the nearest- or next-nearest-neighbour to the seed (aggregation cluster). There we evaluate the sticking probability, e.g. for the A atom

$$\mathscr{P}_A = (1-\alpha)\mathscr{P}_{cA} + \alpha\mathscr{P}_{tA}$$

using

$$\mathscr{P}_{tA} = \exp(-\beta\Delta E_A)/[1+\exp(-\beta\Delta E_A)] \qquad (\Delta E_A \equiv E_A - E_B;\ \beta^{-1} \equiv k_B T)$$

and

$$E_i \equiv -S_i\left(\sum_{\delta:\mathrm{NN}} J_1 S_{i+\delta} + \sum_{\rho:\mathrm{NNN}} J_2 S_{i+\rho} + h_i\right) \qquad (i = \mathrm{A}, \mathrm{B})$$

where $\delta(\rho)$ stands for the nearest- (next-nearest)-neighbour vector, and S takes a value of one (zero) for the A(B) atom. Here we assume that a same-atom pair (A-A, B-B) yields the same perimeter energy increment, while a different-atom pair (A-B, B-A) is different from it. We consider the case of $J_1 < 0$ and $J_2 > 0$, i.e. a non-frustrated case from the interaction point of view. This case is very interesting, because of taking into consideration a fine interaction mechanism in the neighbourhood of the perimeter, and for the sake of comparison with the frustrated case of the triangular lattice in the near future. Notice that the following identities hold:

$$\sum_{i=\mathrm{A},\mathrm{B}} \mathscr{P}_{ci} = 1 \qquad \sum_{i=\mathrm{A},\mathrm{B}} \mathscr{P}_{ti} = 1 \qquad \sum_{i=\mathrm{A},\mathrm{B}} \mathscr{P}_i = 1.$$

The visiting particle sticks and forms a part of the aggregation cluster if the sticking probability is larger than the random number. Otherwise it is removed and another particle is again introduced at a random large distant point. The same procedures are repeated until the cluster forms an N-particle aggregation cluster (N sufficiently large). If a particle touches the boundaries of the lattice in its random walk it is removed and another introduced also.

The successive computing procedure for an N-particle cluster described above supplies one random process datum for one set of physical parameters. We have to

compute N_R random process data repeatedly (N_R sufficiently large) and to take an average over N_R data. We call this average the statistical average. This average value corresponds to the physical value for the fixed physical parameters.

For our study below, such physical values must be computed for a large number of the required physical parameter sets. Actual computations were performed for finite-size(N) systems and statistically averaged over finite (N_R) random process data. The method of extrapolating the properties of the infinite system will be described in § 3.

3. Physical properties

In the usual DLA model we could not think of phase transitions. We consider whether or not the *possibility* of phase transitions exists in our infinite-size system. In the limit of the uniform crystal we can divide the lattice into two sublattices (A and B sublattices), and regard the sublattice magnetisations $\{M_A, M_B\}$ as an order parameter of the system. These concepts would be available for our system when the aggregation cluster size is large enough. We divide our lattice into the A and B sublattices (where the origin is the A sublattice). On our N-particle aggregation cluster we define the following quantities: the number of A(B) sites $N_A(N_B)$; the number of A(B) atoms occupying the A sites $N_{AA}(N_{BA})$; and that of the B sites $N_{AB}(N_{BB})$. Furthermore we define the sublattice commensurabilities (C_A, C_B) and incommensurabilities (IC_A, IC_B) as $C_i \equiv N_{ii}/N_i$, $IC_i \equiv N_{ji}/N_i$ ($i, j = A, B; j \neq i$). They relate to the sublattice magnetisations as $M_i \equiv C_i - IC_i$ ($i = A, B$). In the uniform crystal limit, i.e. the SQL–AF magnetic Ising system, phase transitions occur as the ordered phase to the paraphase. In our aggregation cluster system it is also expected that phase transitions would appear in the infinite-size limit if they exist at all, and that the transition temperature should become considerably lower than those of the uniform crystal. There may exist some new types of phase transitions being characteristic for non-linear, non-equilibrium and finite-size systems, but we will investigate the phase transition and its behaviour in the infinite-size limit as a first step in our study.

Before we enter the discussion of our computations, we emphasise some important points in the process of the derivation of our results. Firstly, our system is strongly concerned with a finite-size, non-equilibrium and random process system. This fact makes the determination of the transition point and the behaviour of the system difficult. The finite size of the system modifies the singular behaviour at the transition point of the corresponding infinite system to regular behaviour. In a non-equilibrium system there is the possibility of the appearance of different behaviour from the ordinary critical behaviour in the equilibrium state. A random process system requires us to take the statistical average for each set of physical parameters. Therefore a computing error comes from the finite size of the aggregation clusters and the lack of data for the random processes, and it makes the singular (critical) point and its characteristic behaviour vague. Our method of attacking these difficulties (though it is not complete) is as follows: (i) we choose a cluster size N as large as possible; (ii) we adopt as many good (i.e. the deviation from the average value is small) random process data as possible; and (iii) we use an extrapolation procedure similar to the finite-size scaling method used in the usual critical phenomena, for data sets for 3–4 different size clusters (including the maximum cluster size). Because of various actual limitations we have studied a relatively small system with few good data values.

Now let us proceed to our computations and results. A list of our computational objectives is as follows: (i) differences from the usual DLA, (ii) differences between the binary-component and the single-component systems, (iii) the concentration effects, (iv) the α and $\mathcal{P}_c$ effects, (v) the temperature effect, and (vi) the chemical potentials and the pressure effects, in the process of evaluation of the possibility of phase transitions and the DLA-like properties. These basic issues are argued as follows.

(i) As the usual DLA ($M=1$, $c=1$) corresponds to the (A+B) cluster system with $\alpha=0$, this item can be argued by comparing the behaviour with that of the (A or B or A+B) cluster system with $\alpha \neq 0$.

(ii) As the single-component ($M=1$) system coincides with the (A+B) cluster system, we may discuss this item from differences among the {A, B, (A+B)} cluster systems.

(iii) Over various concentration values c_A, we compare physical quantities and the cluster patterns. To derive especially interesting properties in our system, we choose small values for c_A.

(iv) This item can be argued by comparing the results for various values of α and $\mathcal{P}_c$, but the qualitative features of our system may be derived from the α dependence for $\mathcal{P}_c=0.5$.

(v) The main interest consists of (1) whether there exists a possibility of phase transitions, and (2) the comparison of differences among the ordered state, the parastate and the temperature-independent ($\alpha=0$) case.

(vi) We can discuss this by computing the changes of physical quantities and cluster patterns for various values of chemical potentials and pressure. As this issue belongs to the second step of our study, we do not discuss it below.

The actual computations have been performed for a *representative* case ($J_1=-1$, $J_2=0.5$, $\mathcal{P}_c=0.5$) with $c_A=0.1$, which matched with our objectives. In this particularly interesting case we took $N=10\,000$ and $\alpha=\{0, 0.3, 0.5, 0.8, 1\}$, and averaged over several values. The numerical results are shown in figure 1(a) for the sublattice magnetisations (M_A, $-M_B$) and in figure 1(b) for the sublattice commensurability C_A (incommensurability IC_B). Using data both for the maximum-size cluster and for the intermediate-size ($N \sim 5000$, 7000, 9000) clusters, we extrapolated the transition point of the infinite-size system in terms of the finite-size scaling approach. Though we could neither rigorously prove the existence of the transition point nor show the behaviour near the point from our actual limitations, phase transition points in the infinite-system point of view were determined and we obtained the qualitative phase diagram illustrated in figure 2. In the following we summarise the characteristic features on the phase transitions obtained from these figures.

(i) The physical quantities $\{(M_A, -M_B), C_A, IC_B\}$ for $\alpha=0$ are independent of the temperature, the interactions $\{J_1, J_2\}$ and the chemical potentials (h_A, h_B), and they give the lower limit.

(ii) The above quantities for any α depend on the concentration c_A and α at zero temperature. This fact requires to redefine the OP as $(M-M_{min})/(M_{max}-M_{min}) \to M$.

(iii) It is very interesting that the sublattice magnetisations are negative for small concentrations of c_A.

(iv) For the cases $c_A \geqslant 0.5$, the sublattice magnetisations $(M_A, -M_B)$ are non-negative.

(v) At least there exists a possibility of the phase transitions, from the ordered phase $\{(M_A, -M_B)$ for $M_A \neq 0\}$ to the paraphase [9].

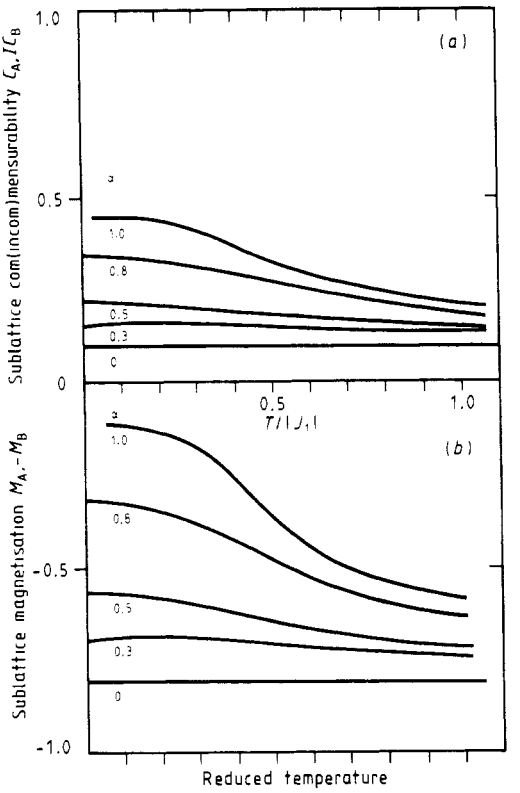

Figure 1. Phase transition behaviour in the case $c_A = 0.1$ ($J_1 = -1$, $J_2 = 0.5$, $\mathscr{P}_c = 0.5$) for the α parameters. (*a*) Sublattice com(incom)mensurability C_A, IC_B against reduced temperature; (*b*) sublattice magnetisation M_A, $-M_B$ against reduced temperature $T/|J_1|$.

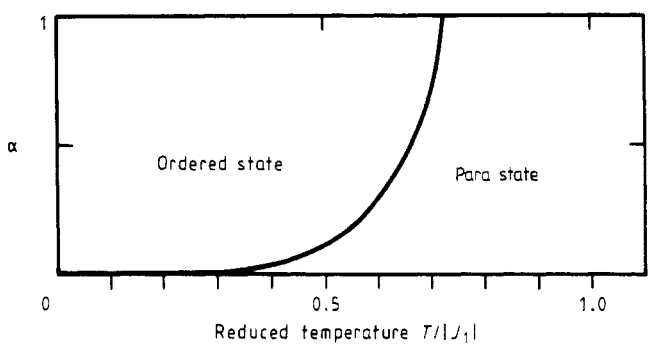

Figure 2. Phase diagram for the same case as figure 1.

(vi) The qualitative α-dependent phase diagram shows also a simple phase transition from the ordered phase to the paraphase. However, there should exist a lower critical value α_c for the existence of this ordered phase, which has not been determined yet. The critical temperature T_c in our system is considerably lower than that of the corresponding uniform crystal system. (S Miyajima (private communication) found that there is a critical point in a ferromagnetic Ising system on the DLA cluster and that its T_c is very small.) The reason is that the aggregation clusters behave as a

network having a small effective coordination number, i.e. they are of a low-dimensional network which is strongly influenced by finite-size effects.

Next we consider the DLA-like properties: the aggregation patterns, the correlation functions and the fractal dimensions. The patterns of the first two cases supply us with physically important information for the system. Especially for researchers in this field, they give important detailed data concerning the growth process. The weight of one pattern in the random process is not heavy but a selected typical pattern is very instructive. With the aid of an electronic computer we drew three types of aggregation pattern constructed with (A+B) atoms, with A atoms, and with B atoms. Typical examples ($c_A = 0.3$, $J_1 = -1$, $J_2 = 0.5$, $\mathscr{P}_c = 0.5$, $T = 0.4|J_1|$) of $\alpha = 0.8$ for $N = 6000$ are shown in figure 3. To understand the physical tendencies in the case of $\alpha = 1$ ($c_A = 0.1$), we show a few patterns of (A+B) atoms for $N = 10\,000$ in figure 4. Information about the particle distribution can be obtained from the density-density correlation function. We define the i-atom density $\rho_i(\boldsymbol{r})$ as 1 for the sites occupied by the i atom and otherwise 0. The correlation functions in a N_{ij}-particle aggregation cluster are defined by

$$C_{i,j}(r) \equiv N_{i,j}^{-1} \sum_{\boldsymbol{r}'} \rho_i(\boldsymbol{r}')\rho_j(\boldsymbol{r}'+\boldsymbol{r}) \qquad (i, j = \mathrm{A}, \mathrm{B})$$

and are an approximation to the ensemble averaged correlation function

$$\langle \rho_i(\boldsymbol{r}')\rho_j(\boldsymbol{r}'+\boldsymbol{r})\rangle / (\langle \rho_i(\boldsymbol{r}')\rangle \langle \rho_j(\boldsymbol{r}+\boldsymbol{r}')\rangle)^{-1/2}.$$

These functions are assumed to depend only on the distance r separating the two sites, for r much less than the cluster size. We calculated $C_{ii}(r)$ for the i-atom clusters

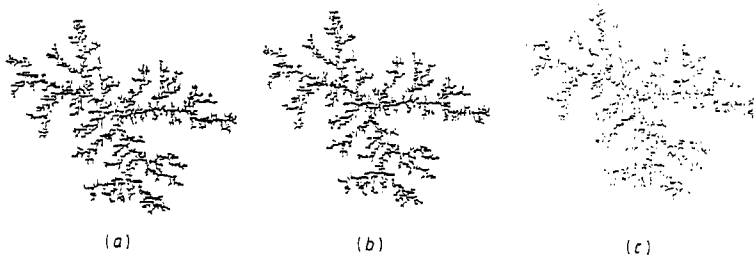

Figure 3. Aggregation clusters of $N \simeq 6000$ particles in the case $c_A = 0.3$ ($J_1 = -1$, $J_2 = 0.5$, $\mathscr{P}_c = 0.5$, $T = 0.4|J_1|$) for $\alpha = 0.8$. (*a*) (A+B)-atom cluster; (*b*) A-atom cluster; (*c*) B-atom cluster.

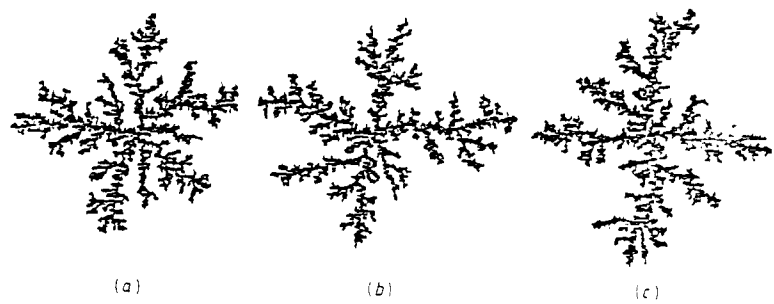

Figure 4. (A+B)-atom aggregation clusters of $N = 10\,000$ particles in the case $c_A = 0.1$ ($J_1 = -1$, $J_2 = 0.5$, $\mathscr{P}_c = 0.5$) for $\alpha = 1$: (*a*) $T/|J_1| = 0.005$; (*b*) $T/|J_1| = 0.1$; (*c*) $T/|J_1| = 0.4$.

($i = A, B$) and $C(r)$ for (A+B)-atom clusters, but the results for these three types of correlation function were the same within the range of numerical accuracy. These supply an answer to objective (i). Figure 5 shows an example of the $C(r)$ correlation function averaged over directions and over several clusters consisting of $N = 10\,000$ particles for the case $c_A = 0.1$ ($J_1 = -1$, $J_2 = 0.5$, $\mathcal{P}_c = 0.5$) with $\alpha = 0.3$, $T/|J_1| = 0.05$. The characteristic features of the aggregation properties are summarised as follows.

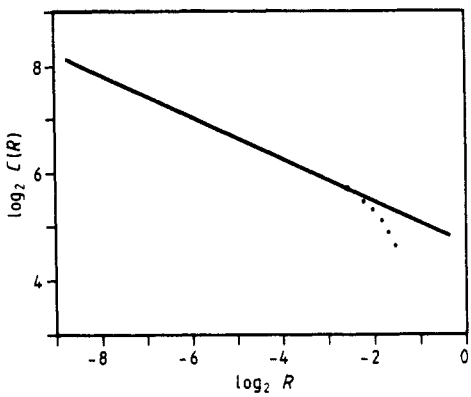

Figure 5. Density-density correlation function averaged over several aggregation clusters in the case $c_A = 0.1$ ($J_1 = -1$, $J_2 = 0.5$, $\mathcal{P}_c = 0.5$) with $\alpha = 0.3$, $T/|J_1| = 0.05$. R stands for the distance measured in the lattice constant. The line is $C(R) \sim R^{-0.401}$.

(i) Firstly we consider the α, c_A dependence of three types of cluster. When $c_A = 0(1)$, the A(B)-atom cluster does not appear. When c_A is a small value ($0 \leq c_A < c_{Ab}$), the A-atom cluster is constructed with fewer number of atoms than the B-atom cluster for small values of α ($0 \leq \alpha < \alpha_b$), and vice versa for α near unity ($\alpha_b < \alpha \leq 1$). Here c_{Ab}, α_b stand for certain balancing values which depend on the quantities $[c_A, \alpha, T, \{J_1, J_2\}, \{h_A, h_B\}, \mathcal{P}_c]$. In contrast, when c_A is large ($c_{Ab} < c_A \leq 1$), the composite-atom number on the A-atom cluster is larger than that on the B-atom cluster for any α. The physical reason is that the symmetry of the binary-component system was broken by choosing the background constructed with the B atoms.

(ii) Next let us consider the α, T dependence of the (A+B)-atom clusters. The $\alpha = 0$ case depends on c_A but not on T. This case belongs to the paraphase. The behaviour of the $\alpha \neq 0$ case is divided into two types for the ordered phase and for the paraphase. Notice that the paraphase behaviour for $\alpha \neq 0$ is influenced by the physical factors near the crystal surface (e.g. the competition of interactions, the chemical potential differences and the temperature) and the $\alpha = 0$ behaviour depends only on the constant sticking probabilities ($\mathcal{P}_{cA}$, $\mathcal{P}_{cB}$), in addition to the common factors $\{c_A, c_B\}$. The differences in the behaviour of two types are very difficult to distinguish by only looking at figures, since they accompany the statistical randomness. We will study them through the fractal dimensions below. Compared with the usual DLA, the characteristic properties of our SQ lattice clusters, including up to the next-nearest-neighbour interactions, are that the number of closed loops increases and dense clusters are formed.

(ii) Finally we discuss the characteristic properties of the fractal dimensions. The correlation functions $C(\equiv\{C_{ij}\})$ show a power law over the region of a few lattice

spacings to the aggregation cluster size. They are expressed as

$$C(r) \sim r^{-A}$$

where their exponents and the corresponding fractal dimensions are denoted by $A(\equiv\{A_{ij}\})$ and $2-A$ respectively. When we compute the exponent in a finite-size system, diffusion particles stick at a far distance (r large) perimeter with high probability, and screen near-distance (r small) areas (where they can no longer stick). In other words, the behaviour of the correlation function is settling down from near-distance regions with increasing cluster size. But such behaviour in certain domains near the origin is different from that desired, because the number of lattice sites allowed on a circle with radius r is too few and the increment by one atom occupation (which is associated with the statistical error) influences a large change. On the other hand, the aggregation processes are still going on outside a certain far-distance domain. Therefore we note that the power laws in aggregation problems are derived from the rather short-distance (not too short and not too long) behaviour, contrary to the long-distance behaviour in critical phenomena. Accordingly, in the computation of critical exponents we may well aim their values in the infinite correlation length limit. Therefore any ambiguity does not enter their evaluations. On the other hand, fractal dimensions must be evaluated over some distance interval between the lower and the upper limits. These two limits are evaluated with the statistical random average over aggregation clusters. Actual individual data give considerably good agreements for the exponents if we suppose two arbitrary values for the two limits. Concepts of the renormalisation group (RG) theories have been applied, on the one hand, to the critical phenomena in condensed matter with infinite particles and, on the other hand, in confinement problems of high-energy physics with finite particles. Their essential critical behaviour is described by the short- or long-distance behaviour respectively. Our system must be evaluated in the same way as the latter application. From such a point of view, according to our method described above, we readjusted our data and obtained the following result. Contrary to the expectation that our system may belong to a new type of phase transition with temperature-dependent exponents A and with different exponents A_{ij} $(i,j=\mathrm{A},\mathrm{B})$, their temperature dependencies and their magnitude differences seem to be smaller than the range of our numerical accuracies. The exponent values are evaluated for the case $c_{\mathrm{A}}=0.1$ ($J_1=-1$, $J_2=0.5$, $\mathscr{P}_{\mathrm{c}}=0.5$) as in table 1.

From this it follows that the exponent values are almost the same (slightly different) in the same (different) phase regions, and increase slightly with increasing α. The other parameter dependence of these exponents is currently under study.

We summarise the characteristic tendencies between the usual DLA and our system on the SQ lattice.

Table 1.

	α				
$T/\lvert J_1\rvert$	0	0.3	0.5	0.8	1
0.05	0.326	0.353	0.357	0.359	0.360
0.1	0.326	0.355	0.359	0.360	0.361
0.2	0.326	0.357	0.560	0.361	0.362
0.4	0.326	0.358	0.361	0.362	0.363
1	0.326	0.359	0.361	0.363	0.363

(i) The constant sticking probability effects can be understood in the case of $\alpha = 0$. The exponent value and the number of sticking particles per unit time increase with increasing sticking probability.

(ii) The far-neighbour interaction effects can be drawn in the $\alpha \neq 0$ case. The interactions slightly enhance the exponent values.

Finally we summarise the range of validity of our approximation and the remarkable features for our objectives (i)-(v). With respect to the accuracy of the exponent, we suppose the error to be within 0.02.

(i) In contrast to the usual DLA system having no phase transitions, our system features the possibility of phase transitions characterised by the physical variables and parameters, in the infinite-size cluster limit, in the parameter regions above α_c, c_{Ac}, and nowhere else. The exponent values in our system are slightly larger than those in the DLA system.

(ii) By taking into account two kinds of atoms we could investigate the interesting (ordered, para) phase and their behaviour, while the single-component system (cf (A+B) system) displays rather simple but interesting behaviour, in comparison with the usual DLA system. Our system has almost the same values depending upon the physical parameters, within our numerical accuracy, but the regimes characterised by different exponent values may exist in the parameter regions below α_c, c_{Ac}.

(iii) By introducing the concentrations for the binary components we investigated the regions below and above the critical (percolation) concentration for each component. When c_A is small but larger than c_{Ac}, negative sublattice magnetisation appears over wide ranges of temperature.

(iv) The irreversibility of the system comes from the constant part $(1-\alpha)\mathscr{P}_c$ and the thermal part $\alpha\mathscr{P}_t$ of $\mathscr{P}$. The phase transition of the ordered phase to the paraphase takes place for $\alpha > \alpha_c$.

(v) By introducing the concept of temperature we could discuss the phase transitions and their behaviour. A possibility of the existence of phases was suggested in our system. The exponent values are independent of temperature within our numerical accuracy.

4. Concluding remarks

Improving the DLA model, we constructed a new physical multicomponent model, reflecting a slightly more complex interaction mechanism near the crystal surface, chemical potentials of composite particles and temperature, in addition to the diffusion particle concentrations and the constant sticking probabilities. Further we introduced the non-equilibrium-equilibrium parameter α.

We found that our system has a possibility of a new type of phase transition from the ordered state to the parastate, but these states are different from the usual equilibrium states. The critical temperature are considerably lower than those of the corresponding Ising system. The sublattice magnetisations are associted with one of the OP of the system, but it is required to redefine them to change in the interval $(0, 1)$. A very interesting feature is that negative sublattice magnetisations appear for $0 < c_A < 0.5$. Within the range of our numerical accuracies, we could not distinguish the differences among the density-density correlation functions $\{C_{AA}, C_{BB}, C_{(A+B)(A+B)}\}$, and the temperature dependence of the critical exponent A. But these are still open questions. That is, we think that there may be new phase regions in our system, where the OP are

different. We need to look for a complete set of OP in our system. In order to get accurate results in a simple systematic treatment, we tried to attack in terms of a finite-size RG approach, a real-space RG approach and further analytic RG approach, but we have not yet arrived at satisfactory results.

Now let us discuss the relation of our results to experiments. Historically, in order to discuss interfacial stability and pattern formation in many, common, physical problems as non-equilibrium dynamics problems, the Saffman-Taylor problem [9] (the interfacial motion of two incompressible fluids with different viscosities in a two-dimensional channel devised by Hele-Shaw [10], models for dendritic growth [1] and for the DLA [5, 6] and so on, have been well investigated. Notably, the Hele-Shaw system was translated into a quite effective computing model with the DLA-like random walk picture, and provides the foundation for the Monte Carlo study of the hydrodynamic process. It was pointed out by Paterson [11] that a similarity exists between the interfacial motion of two fluids in porous media and a set of the mean-field DLA equations, and by Pietronero and Wiesmann [12] that the same analogy exists between the dendritic breakdown and the DLA system. They compared their DLA simulations with the same experiments. Tang [13] showed that the mean-field limit of DLA is exactly the Saffman-Taylor problem. Bensimon *et al* [2] found a more general connection between the Saffman-Taylor problem and the DLA. Nittmann *et al* [7] performed the same experiments and DLA simulations of fluids in a Hele-Shaw cell in the zero-surface-tension case. From the theoretical [14-17] and experimental [18-20] studies the DLA looks like the zero-surface-tension case of the Saffman-Taylor problem, i.e. zero surface tension makes an interface highly unstable and, as a result, the aggregation cluster of DLA forms tiny fingers or wisps which have a fractal structure. This is different from any structure formed in a real fluid flow with non-zero surface tension. In order to improve the usual DLA model and to derive reasonable fluid behaviour, Tang required that no motion occurs until some number M of walkers arrived. Comparing his result to the exact steady state solution of Saffman and Taylor [9] or to the exact time-dependent solution of Shraiman and Bensimon [14], he found an excellent agreement between them. Another improvement of the DLA model is to take into account the surface tension correctly. In our system we did not take into account any kind of averaging M but some kind of temperature-dependent surface tension at the perimeter (interface). Later we shall devise both some kind of averaging method, like the M averaging, and some sort of growth mechanism, such as the snowflake, as we intend to compare our results with experiments.

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