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## LETTER TO THE EDITOR

### Ordered phase *AB* diffusive growth

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**Abstract.** Ordered phase *AB* formation and diffusive growth and its boundary evolution on a square lattice are investigated by computer simulation. It is found that the boundary is a self-similar fractal ( $D=7/4$ ) and its width and length grow as a power law with exponents  $1/3$  and  $1/4$ , correspondingly. The phase is found to exist over the 0.31-0.69 concentration range. It is shown that a phase formed via a second order-phase transition may have a boundary if the physical properties of the system have a gradient.

In the present work we simulated ordered phase formation and diffusive growth in a two-dimensional system consisting of atoms *A* and *B* which have negative mixing energy (this is taken to correspond to attraction of different-type atoms) so that it is thermodynamically justified for the system to be ordered into phase *AB*. In contrast to the traditional treatment of problems on phase diffusive growth which considers the boundary to remain smooth in the course of the diffusion, such an approach allows investigation of the geometry and the laws governing the evolution of the boundary upon interdiffusion.

Previously we investigated boundary evolution upon interdiffusion. In [1, 2] we investigated the evolution of a diffusion couple consisting of components which have zero mixing energy and which therefore form an ideal solution. We found that the diffusion front thus formed has a fractal geometry and that its evolution is characterized by some critical exponents. We also investigated another case [3] where the mixing energy is great enough so that it is thermodynamically justified for the system to be separated into two phases by a boundary. We showed that the boundary is a self-affine fractal and that its evolution is consistent with the predictions the linear version of the Langevin equation, quite different from the diffusion front in an ideal solution.

In recent years surfaces of aggregates formed by other stochastic processes, such as deposition and growth, were investigated both by computer simulation and by theoretical methods (see, for example, references in [4]). As was first shown by Family and Vicsek [5], for such aggregates the dependence of the surface width on time and on substrate size can be written in a double scaling form. Edwards *et al* [6] derived a linear equation of Langevin type for such surface growth. Later, Kardar *et al* generalized this equation by inserting a nonlinear term.

However, the results of this work are quite different both from the predictions of the Langevin equation and from the results of our work [3] where the positive mixing energy case was studied.

The atoms were placed on a square lattice having  $N_x \times N_y$  sites. Originally the atoms formed two pure phases *A* and *B* separated by a straight line joining the middle points of the lattice boundaries. These two boundaries were 'joined' (periodic boundary conditions) while the other two were made reflecting.

A vacancy moved over the lattice and jumped each time to one of the four neighbour sites thus carrying out the interdiffusion. The activation energy of the vacancy jump was taken to be  $E = E_0 + n\epsilon/2$ , where  $n$  is the change in the number of heterogeneous bounds in the sample as a result of the jump and  $\epsilon = \epsilon_{AB} - 0.5(\epsilon_{AA} + \epsilon_{BB})$  is the mixing energy of the atoms  $A$  and  $B$  ( $\epsilon_{AB}$ ,  $\epsilon_{AA}$  and  $\epsilon_{BB}$  are the binding energies for the corresponding atom pairs). Then the difference between the activation energies of the direct jump and the reverse one is just  $nu$ . It means that relative probability of a diffusion jump to a given site is proportional to  $\exp(-nu/2)$  where  $u = \epsilon/kT$  is the reduced mixing energy. The simulation was performed at  $u = -1$  and  $u = -2$ .

In the course of the interdiffusion, the ordered phase  $AB$  formed having a square unit cell with two  $A$  atoms on one diagonal and two  $B$  atoms on the other. At first stage ordered phase clusters were formed along the original boundary between atoms  $A$  and  $B$  (figure 1(a)) which then coalesced into one cluster. We shall call it the main cluster in contrast to the class of clusters formed in an outlying area. The main cluster consisted of antiphase domains of two types depending on which of the two sublattices was occupied by  $A$  or  $B$  atoms (figure 1(b)). Interdomain wall migration will be studied in detail in a further work; here we only note that the domains coalesced into one after the phase spread over the whole sample.

The ordered phase, ignoring domain type difference and the main cluster boundary, are shown in figure 2. We studied the growth kinetics for the total phase width (the total number of phase atoms normalized on  $N_v$ ) and for the boundary length and width (doubled mean square deviation from the central position). The results for  $256 \times 128$  sample at  $u = 1$  are shown in figure 3 from the moment when the main cluster is formed. The values under study exhibit scaling; the exponents are: for the total

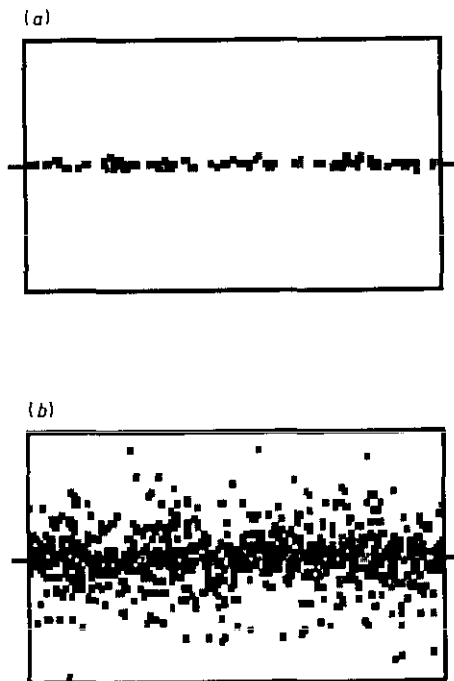
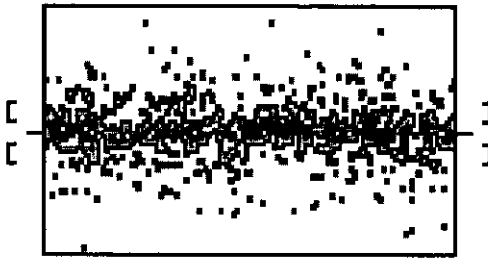
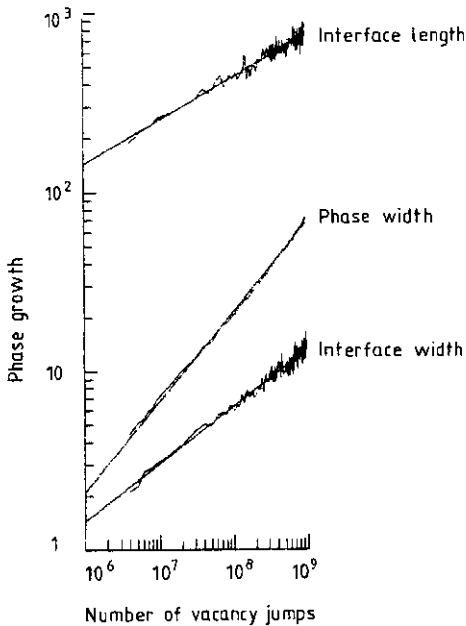


Figure 1. Ordered phase after diffusion on a  $64 \times 128$  lattice at  $u = 1$  after (a)  $10^6$  and (b)  $10^8$  vacancy jumps. The domains are indicated grey and black.



**Figure 2.** Ordered phase (grey) and the boundaries of its main cluster (black) on  $64 \times 128$  lattice after  $10^8$  vacancy jumps at  $u = -1$ . The domains are not indicated. The marks show the boundary mean widths.



**Figure 3.** Boundary evolution on a  $256 \times 128$  lattice at  $u = -1$ .

phase width  $\beta_w = 0.50 \pm 0.01$ , for the boundary width  $\beta = 0.33 \pm 0.01$  and for the boundary length  $\beta_N = 0.24 \pm 0.01$ . This allows the assumption that exact values for the exponents are:  $\beta_w = 1/2$ ,  $\beta = 1/3$  and  $\beta_N = 1/4$ . The same results were obtained for  $u = -2$ .

We also studied the geometry of the ordered phase main cluster boundary. It appeared to be a self-similar fractal. We determined the value of its fractal dimension from the dependence of the number of atoms in the box against the linear size of the box. Due to finite size effects the fractal dimension rose in the course of diffusion; it was equal to  $D = 1.60$  after  $10^9$  vacancy jumps. However, we can remove finite size effects. From the relationship  $\beta_N = (D - 1)\beta$  [1, 2] we get  $D = 7/4$  equal to the fractal dimension of the percolation cluster external boundary [8].

A self-similar fractal is essentially a multivalued function of any Cartesian coordinate. Hence although the value of the exponent  $\beta = 1/3$  corresponds to the prediction of the nonlinear version of the Langevin equation [7], the latter is hardly applicable

to the description of the  $AB$  phase boundary evolution because it supposes the boundary to be a single-valued function of the coordinates.

From the equation  $c_b = (1/N) \sum_{i=1}^N c_i(A) n_i$ , a mean concentration of  $A$  atoms corresponding to the boundary position was found. Here  $c_i(A)$  and  $n_i$  are concentration of  $A$  atoms and number of boundary atoms in the  $i$ th layer, and  $N$  is total number of the boundary atoms. In course of phase growth the concentration appeared to remain constant and equal to  $c_b = 0.31 \pm 0.01$  for one boundary and, correspondingly,  $0.69 \pm 0.01$  for the other, both for  $u = -1$  and  $u = -2$ . So, the phase exists at the 0.31–0.69 concentration range.

In contrast to the case of positive mixing energy, when phases  $A$  and  $B$  available are stable and therefore the boundary thickness saturates to a constant value [3], in the case under study the boundaries grew until they reached the lattice edge. To establish whether the boundaries are stable or not we simulated diffusion in a system consisting originally of the phase  $AB$  and the pure component  $A$  for mixing energies from  $u = -1$  to  $u = -6$ . At any (negative) energy the phase  $AB$  appeared to dissolve an unlimited quantity of the pure component (however, the phase becomes disordered if its concentration falls out of the range mentioned above). Hence (i) there is no concentration step on the phase  $AB$  boundary and (ii) the boundary is unstable. The absence of the concentration step on the boundary means that a second-order phase transition takes place in the regions adding to the phase  $AB$  (ordering). The energy of such a boundary is zero. Therefore, in contrast to the boundary of phases  $A$  and  $B$  [3], the boundary under study is a self-similar fractal rather than a self-affine fractal.

It seems to be appropriate to discuss here the problem of applicability of boundary concept to phase growing via a second-order phase transition. Indeed, while at first-order phase transitions, regions having physical property steps on their boundaries are formed, at second-order phase transitions an infinitesimal new property normally arises simultaneously in the whole volume, so that one cannot speak about a boundary in this case. However, the latter argument is valid only for systems having uniform physical characteristics. For instance, if the system temperature has a gradient, a paramagnetic–ferromagnetic transition cannot take place in the whole volume simultaneously. Hence, a boundary must occur between the paramagnetic and the ferromagnetic regions. The phase  $AB$  boundary formed under a concentration gradient is of a similar nature.

The foregoing simulation and analysis give rise to the following conclusions: a phase growing via a second-order phase transition may have a boundary if the physical properties of the system have a gradient; as a result of interdiffusion of atoms  $A$  and  $B$  having negative mixing energy, an ordered phase  $AB$  forms in the diffusion zone which exists over the 0.31–0.69 concentration range; in contrast to the positive mixing energy case [3] the phase boundary is a self-similar fractal having the same fractal dimension ( $D = 7/4$ ) as the diffusion front formed at zero mixing energy [1, 2]; in contrast to both the zero [1, 2] and positive [3] mixing energy cases the boundary width and length grow as a power law with exponents  $1/3$  and  $1/4$ , respectively; there is no concentration step on the  $AB$  phase boundary and the boundary is unstable.

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