

Kinetic Models of a Binary Alloy at Zero Temperature

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We study theoretically two types of kinetic models of a binary alloy at zero temperature. In the phase separation model, a nearest-neighbor interchange can occur if the fraction of AB bonds (where A and B denote distinct species of atoms in a binary alloy) is thereby decreased. The crystallization model is defined by the opposite evolution rule. We examine these models in one dimension and obtain exact analytical results for the densities of domain walls, defects, and for a number of other correlators. Nonergodic zero-temperature dynamics leads to final states strongly dependent on initial conditions. For generalized models, in which nearest-neighbor interchange is also performed if the portion of AB bonds is not changed, a very rich kinetic behavior is observed.

KEY WORDS: Phase separation; crystallization; binary alloy; nonergodic processes.

1. INTRODUCTION

In this paper we examine a collection of interacting particle systems, motivated by the study of binary alloys. We consider a zero-temperature dynamics on a uniform one-dimensional lattice whose atoms exchange in a nearest-neighbor pairs. Models are formulated as computer games whose evolution to stationary states is strongly dependent on their initial states. The latter are prepared by generating sequences of atoms of species A and B. The dynamics of these models manifestly conserves the order parameter. The asymptotic long-time behavior of the phase separation model in which a nearest-neighbor interchange can occur if the fraction of AB bonds is thereby decreased was studied numerically by Meakin and Reich^(1,2) for cubic lattices of various dimensions ($d=1-5$) and then by Frisch and co-workers numerically for the Bethe lattice⁽³⁾ and analytically in $d=1$.^(3,4)

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Some analytical results concerning the approach to equilibrium were found by Elskens and Frisch.⁽⁴⁾

In this study we give a description of the kinetic behavior of the phase separation model and the dual crystallization model, based on analytical solutions of one-dimensional models. Besides the intrinsic interest of these models, they also exemplify a physically relevant class of nonergodic processes whose simplest properties (e.g., the degree of phase separation) cannot be found by equilibrium considerations. Similar properties are also typical for models of glass. Therefore, although we investigate simple one-dimensional models, time-dependent analytical solutions are interesting since they provide some understanding of nonergodic dynamic processes.

The outline of this paper is as follows. In Section 2, we describe the evolution of the simplest one-dimensional phase separation model to its stationary state. We focus on sequences of atoms of alternating species, write down a master equation for the densities of these alternating clusters, solve this equation exactly for factorizable initial conditions, and then discuss in detail the evolution for the most natural initial conditions, that is, for the alternating and the random one. Exact results clearly reveal the crucial role played by initial conditions. Some generalized phase separation models are discussed in Section 3. The mathematical analysis of these interacting particle systems is remarkably simple—by mapping the generalized phase separation model onto a special version of the random sequential adsorption (RSA) process one can use well-established procedure and solve the model exactly in one dimension. In Section 4, we describe parallel results for the crystallization model. In Section 5, we investigate the crystallization model in which the nearest-neighbor interchange is also performed if the portion of AB bonds is kept fixed. In addition to the deposition events (on the language of the RSA), this model allows also the diffusion events. The interplay between deposition and diffusion processes leads to a rich kinetic behavior strongly dependent on initial conditions. Finally, conclusions appear in Section 6.

2. PHASE SEPARATION MODEL

Our model is an irreversible one-dimensional phase separation model with nearest-neighbor particle exchange dynamics. Each site of the infinite linear lattice is occupied by one A or one B particle. Two neighbor particles of different species can be exchanged if the number of interfaces (i.e., AB bonds) is thereby decreased. All possible exchange events are assumed to be independent with an exponent waiting time. Thus we have the Kawasaki-like particle exchange dynamics which conserves the total number of particles of both species. Since two neighbors exchange if and only if the total number of interfaces is decreased by a maximum amount

(equal to 2) by this exchange event, we see that the present model is just the zero-temperature Kawasaki dynamics. Indeed, at zero temperature the exchange event which leads to the maximum decreasing of the number of interfaces has the infinite Boltzmann factor compared to the exchange events.

Denote by X_N a sequence of N atoms of the same species, N A's or N B's, and by Y_M a sequence of M atoms of alternating species. Any sequence of A's and B's is then uniquely (up to substituting of all A's and B's) described by a sequence of the form $\dots X_{N_1} Y_{M_1} X_{N_2} Y_{M_2} \dots$, which $N_i \geq 2$ and $M_i \geq 0$ to avoid ambiguities. For instance,

$$\dots \text{AABABBBAAA} \dots = \dots X_2 Y_2 X_3 Y_0 X_3 \dots$$

From the definition of the model one finds that the exchange events take place only inside sequences Y_M . Furthermore, the most interesting quantities, such as the densities of domain walls, defects, and interfaces, may be immediately extracted from the densities of Y_M 's. Therefore in the following we focus on the behavior of the densities of alternating clusters, although other correlators, e.g., the densities of clusters of the same species, X_N , are also of interest.

Consider now the evolution of alternating clusters. The definition of the model implies that:

(i) $X_{N_1} Y_0 X_{N_2}$ cannot evolve further, i.e., a domain wall ($\dots \text{AABB} \dots$) is steady.

(ii) $X_{N_1} Y_1 X_{N_2}$ cannot evolve further, i.e., a defect ($\dots \text{AABAA} \dots$) is steady, too.

(iii) $X_N Y_P X_M$ can evolve in $P - 1$ equally likely ways:

$$X_N Y_P X_M \Rightarrow \begin{cases} X_{N+1} Y_0 X_2 Y_{P-3} X_M \\ X_N Y_Q X_2 Y_0 X_2 Y_{P-Q-4} X_M \\ X_N Y_{P-3} X_2 Y_0 X_{M+1} \end{cases} \quad (0 < Q < P - 4)$$

The stationary state is thus a sequence of X_N 's, Y_0 's, and Y_1 's only. Both the density of domain walls and that of defects grow with time and a full phase separation cannot be achieved.

Let us denote by $Y_M(t)$ a portion of Y_M 's at time t . Then a master equation for this problem has the following form:

$$\frac{d}{dt} Y_0 = Y_2 + \sum_{K \geq 3} (K + 1) Y_K \tag{2.1a}$$

$$\frac{d}{dt} Y_N = -(N - 1) Y_N + 2 \sum_{K \geq 3} Y_{N+K}, \quad N > 0 \tag{2.1b}$$

All terms in Eqs. (2.1) are simply deduced from the evolution law (iii).

To solve Eq. (2.1b), we make the ansatz

$$Y_N = AB^{N-1}, \quad N \geq 1 \tag{2.2}$$

where $A(t)$ and $B(t)$ are functions of time to be found. By inserting (2.2) into (2.1), we reduce the infinite system of Eq.(2.1b) to the finite one

$$\frac{d}{dt} A = 2AB^3(1 - B)^{-1} \tag{2.3a}$$

$$\frac{d}{dt} B = -B \tag{2.3b}$$

while Eq. (2.1a) becomes

$$\frac{d}{dt} Y_0 = AB \frac{1 + 2B - 2B^2}{(1 - B)^2} \tag{2.4}$$

Thus, if initial data are in agreement with the ansatz (2.2), we must solve Eqs. (2.3) and (2.4). Such is the case for the alternating initial sequence Y_∞ . We have the following initial data:

$$Y_N(0) = 0 \quad \text{for all } N \geq 0, \quad \lim_{t \rightarrow 0} \sum_{N \geq 1} NY_N(t) = 1 \tag{2.5}$$

Using the relation $\sum_{N \geq 1} NY_N = A(1 - B)^{-2}$, we deduce from Eq. (2.5)

$$A(0) = 0, \quad B(0) = 1, \quad \lim_{t \rightarrow 0} A(1 - B)^{-2} = 1, \quad Y_0(0) = 0 \tag{2.6}$$

and find the exact solution

$$\begin{aligned} A(t) &= [1 - \exp(-t)]^2 \exp[\exp(-2t) + 2 \exp(-t) - 3] \\ B(t) &= \exp(-t) \end{aligned} \tag{2.7}$$

$$Y_0(t) = \int_{\exp(-t)}^1 du (1 + 2u - 2u^2) \exp(u^2 + 2u - 3)$$

The degree of phase separation in the model may be measured by the fraction $\rho(t)$ of “bad” bonds (A–B):

$$\rho = \sum_{N \geq 0} (N + 1) Y_N = Y_0 + A(2 - B)(1 - B)^{-2} \tag{2.8}$$

At the stationary state, $t = \infty$, we find the following values for the density of domain walls Y_0 , defects Y_1 , and bad bonds ρ :

$$\begin{aligned}
 Y_0(\infty) &= 0.351323887\dots \\
 Y_1(\infty) &= 0.049787068\dots \\
 \rho(\infty) &= 0.450898024\dots
 \end{aligned}
 \tag{2.9}$$

As a second example, we consider a random chain where the probability of meeting an A atom and a B atom is equal. Here initial data are again compatible with the ansatz (2.2). Simple combinatorics gives

$$Y_N(0) = 2^{-N-3}$$

or

$$A(0) = 1/16, \quad B(0) = 1/2, \quad Y_0(0) = 1/8 \tag{2.10}$$

The exact solution for these initial data is

$$\begin{aligned}
 A(t) &= \frac{1}{4} [1 - \frac{1}{2} \exp(-t)]^2 \exp[\exp(-2t) + \frac{1}{4} \exp(-t) - \frac{5}{4}] \\
 B(t) &= \frac{1}{2} \exp(-t) \\
 Y_0(t) &= \frac{1}{8} + \frac{1}{4} \int_{\exp(-t)/2}^1 du (1 + 2u - 2u^2) \exp(u^2 + 2u - \frac{5}{4})
 \end{aligned}
 \tag{2.11}$$

At the stationary state we have

$$\begin{aligned}
 Y_0(\infty) &= 0.219704245\dots \\
 Y_1(\infty) &= 0.071631199\dots \\
 \rho(\infty) &= 0.36296644\dots
 \end{aligned}
 \tag{2.12}$$

Comparison of (2.9) and (2.12) shows that the evolution to the final stationary state is strongly dependent on the initial state, i.e., an asymptotic distribution cannot be evaluated from the single requirement of stationarity. The system therefore has an infinite number of stationary states. So, the kinetic model is not ergodic.^(3,4)

It is worth pointing out that analytical solutions to the master equation (2.1) can be also found for some cases when the ansatz (2.2) becomes invalid. One natural such example is provided by the binary alloy with *different* initial concentrations and random initial conditions. Denoting initial concentrations of A and B species by p and q , respectively ($p + q = 1$), one finds distinct expressions for even and odd $Y_N(0)$, namely

$$Y_{2N}(0) = 2(pq)^{N+2}, \quad Y_{2N+1}(0) = (p^3 + q^3)(pq)^{N+1} \tag{2.13}$$

However, the initial data are in agreement with the generalized version of the ansatz (2.2)

$$Y_{2N+1} = A_1 B^{2N}, \quad Y_{2N+2} = A_0 B^{2N+1}, \quad N \geq 1 \quad (2.14)$$

By inserting (2.14) into the master equation one can find a closed system of equations for functions $A_0(t)$, $A_1(t)$, $B(t)$, and $Y_0(t)$ and then solve these equations. We omit these explicit but rather cumbersome results.

3. GENERALIZED PHASE SEPARATION MODELS

In this section we discuss some generalized one-dimensional phase separation models. First, we describe a class of models amenable to exact solvability like the simple model of Section 2. For this purpose, it is convenient to use a new representation of a lattice occupied by A and B atoms. Let us denote any “bad” bond (i.e., A–B or B–A) by 0 and any “good” bond (i.e., A–A or B–B) by 1. An arbitrary sequence of A’s and B’s is then uniquely (up to substituting of all A’s and B’s) described by a sequence of 0’s and 1’s. For instance,

$$\dots \text{AABABBAABBB} \dots = \dots 1000101011 \dots$$

The evolution law for the phase separation model of Section 2 can be depicted as follows:

- (1.PS) Any 000-triplet can evolve, $000 \Rightarrow 101$.
- (2.PS) Other triplets cannot evolve further.

One can interpret 0’s and 1’s as vacant and filled sites, respectively, and a process of spinodal decomposition as a version of a random sequential adsorption (RSA) process. In such processes one attempts to place immobile objects at random on a lattice in such a way that they do not overlap. In our model two vacant next-neighbor sites, divided by a vacant site, are filled simultaneously. Because of the condition of immobility, RSA configurations differ from their equilibrium counterparts.

The RSA process is of interest in a variety of fields, including statistical,^(5,6) chemical,^(7,8) and biological physics.^(9,10) Exact results are sparse and largely confined to one-dimensional systems and lattices with tree properties. For one-dimensional RSA, several authors have derived the analytical expressions for the coverage versus time⁽¹¹⁻¹³⁾ and for more complex correlation functions.⁽¹⁴⁻¹⁶⁾ The special solvable case in which single sites fill randomly with nearest-neighbor cooperative effects has been studied extensively.

These studies suggest that a generalization of our RSA model taking into account some cooperative effects must be tractable, too. The simplest such generalization permits us to take into account boundary sites of a triplet. We modify the rule (1.PS) as follows:

(1.PS)' Any triplet 000 evolves to 101 with the rates λ_0 , λ_1 , and λ^2 depending on a number of vacant sites on its boundary, e.g., 000 \Rightarrow 01010 with the rate λ_0 , etc.

The rule (2.PS) remains unchanged.

To display the solvability of a model defined by the rules (1.PS)' and (2.PS), we return to the XY representation of a lattice. Then the master equation for this problem has the following form:

$$\frac{d}{dt} Y_0 = \lambda_2 Y_2 + 2\lambda \sum_{K \geq 4} Y_K + 4\lambda_1 Y_3 + \lambda_0 \sum_{K \geq 4} (K-1) Y_K \quad (3.1a)$$

$$\frac{d}{dt} Y_1 = 2\lambda_1 Y_4 + 2\lambda_0 \sum_{K \geq 5} Y_K \quad (3.1b)$$

$$\frac{d}{dt} Y_2 = -\lambda_2 Y_2 + 2\lambda_1 Y_5 + 2\lambda_0 \sum_{K \geq 6} Y_K \quad (3.1c)$$

$$\frac{d}{dt} Y_N = -\lambda_0(N-3) Y_N - 2\lambda_1 Y_N + 2\lambda_1 Y_{N+3} + 2\lambda_0 \sum_{K \geq 4} Y_{K+N} \quad (3.1d)$$

All terms in Eq. (3.1) are deduced the evolution law (1.PS)' after straightforward manipulations.

To solve Eq. (3.1d) we use the ansatz

$$Y_N = AB^{N-3}, \quad N > 2 \quad (3.2)$$

and reduce the infinite system (3.1d) to the following equations for the functions $A(t)$ and $B(t)$:

$$\begin{aligned} \frac{d}{dt} A &= 2A[\lambda_0 B^4(1-B) - \lambda_1(1-B^3)] \\ \frac{d}{dt} B &= -\lambda_0 B \end{aligned} \quad (3.3)$$

The general solution to these equations has the form

$$\begin{aligned} A &= C_1(1-B)^2 B^{2\mu} \exp[2B + B^2 + \frac{2}{3}(1-\mu)B^3], \quad \mu = \lambda_1/\lambda_0 \\ B(t) &= C_2 \exp(-\lambda_0 t) \end{aligned} \quad (3.4)$$

where the constants C_1 and C_2 can be defined from initial data. One can further reduce Eqs. (3.1a)–(3.1c) to a closed system relating $Y_0(t)$, $Y_1(t)$, and $Y_2(t)$ and then solve these equations analytically. We omit the resulting explicit but rather cumbersome formulas.

Let us now consider the approach to equilibrium for the alternating initial sequence Y_∞ . Substituting the corresponding initial data $A(0)=0$ and $B(0)=1$ into the exact solution (3.4), we obtain all constants

$$C_1 = \exp\left[-3 - \frac{2}{3}(1 - \mu)\right], \quad C_2 = 1 \quad (3.5)$$

For the random start, we have $A(0)=1/64$ and $B(0)=1/2$. By inserting these initial data into (3.4), we obtain

$$C_1 = 2^{2\mu-4} \exp\left[-\frac{5}{4} - \frac{1}{12}(1 - \mu)\right], \quad C_2 = \frac{1}{2} \quad (3.6)$$

The approach to equilibrium appears to be exponential.

4. CRYSTALLIZATION MODEL

Turn now to the crystallization model which is dual to the phase separation model of Section 2. We consider a sequence of atoms of species A and B on the infinite line, in which nearest neighbor interchange can occur only if the portion of AB bonds is thereby increased. Using the 0–1 representation introduced in Section 2, we see from the definition of the model that the only evolution occurs inside triplets. Moreover:

(1.C) Any 010-triplet can evolve, $010 \Rightarrow 111$.

(2.C) Other triplets cannot evolve further.

Our crystallization model with RSA-type kinetics proves to be solvable. To demonstrate this, it is convenient to denote by Z_N , $N \geq 1$, a sequence of N recovering evolutionary triplets of the type 010 (e.g., 0101010 means Z_3). Then the master equation for the fraction of Z_N 's in a chain at time t , $Z_N(t)$, has the following form:

$$\frac{d}{dt} Z_N = -N Z_N + 2 \sum_{K \geq 2} Z_{N+K} \quad (4.1)$$

All terms in Eq. (4.1) are simply deduced from the evolutionary rules (1.C) and (2.C).

To solve the master equation, we again use the ansatz

$$Z_N = AB^N \quad (4.2)$$

where $A(t)$ and $B(t)$ are functions of time to be found. Substituting (4.2) into (4.1), we come to the reduced system

$$\frac{d}{dt} A = 2AB^2(1-B)^{-1}, \quad \frac{d}{dt} B = -B \quad (4.3)$$

Solving Eq. (4.3) leads to

$$A = \text{const} \cdot (1-B)^2 \exp(2B), \quad B = \text{const} \cdot \exp(-t) \quad (4.4)$$

First, we investigate the approach to equilibrium for a random chain. Then the probability to meet Z_N at time $t=0$ is

$$Z_N(0) = (3/4)^2 2^{-N} 2^{-N-1} \quad (4.5)$$

where the first factor $3/4$ is a consequence of the boundary conditions on every side of Z_N [boundary bond(s) are given by 0 or 11]; the second factor accounts for N units in a sequence Z_N and the last factor accounts for $N+1$ zeros in a sequence Z_N . Thus the initial data (4.5) are in agreement with the ansatz (4.2) and they may be rewritten as follows:

$$A(0) = 9/32, \quad B(0) = 1/4 \quad (4.6)$$

Combining (4.4) and (4.6), we obtain

$$A(t) = \frac{1}{2} \left[1 - \frac{1}{4} \exp(-t) \right]^2 \exp \left[\frac{\exp(-t) - 1}{2} \right], \quad B(t) = \frac{1}{4} \exp(-t) \quad (4.7)$$

The degree of crystallization in our model may be measured by the fraction $\rho(t)$ of bad bond, i.e., the fraction of zeros. All zeros outside of Z_N 's will survive, while other zeros may disappear. Calculating these loss terms yields a simple equation for $\rho(t)$:

$$\frac{d}{dt} \rho = -2 \sum_{K \geq 1} K Z_K = -2AB^2(1-B)^{-2} \quad (4.8)$$

Combining (4.7) and (4.8), we obtain

$$\rho(t) = \frac{1}{2} \exp \left[\frac{\exp(-t) - 1}{2} \right] \quad (4.9)$$

i.e., when t increases from zero to infinity, $\rho(t)$ decreases from $\rho(0) = \frac{1}{2}$ to $\rho(\infty) = \frac{1}{2} \exp(-\frac{1}{2}) = 0.303265\dots$

Second, we investigate the approach to equilibrium for an alternating, in 0-1 representation, chain

$$\dots 0101010101010\dots = \dots AABBAABBAABBAABB\dots$$

To define $A(0)$ and $B(0)$, we use obvious initial data

$$A_N(0) = 0 \quad \text{for all } N \geq 1, \quad \sum_{N \geq 1} NZ_N(0) = 1/2 \quad (4.10)$$

and find

$$A(0) = 0, \quad B(0) = 1, \quad \lim_{t \rightarrow 0} A(1 - B)^{-2} = 1/2 \quad (4.11)$$

Combining (4.4) and (4.11), we find the exact solution

$$A = \frac{1}{2}(1 - B)^2 \exp(2B - 2), \quad B(t) = \exp(-t) \quad (4.12)$$

Then we arrive at the following equation for the fraction $\rho(t)$ of bad bonds (A-A or B-B):

$$\rho(t) = \frac{1}{2} \exp[2 \exp(-t) - 2] \quad (4.13)$$

Thus we conclude that $\rho(t)$ decreases from $\rho(0) = \frac{1}{2}$ to $\rho(\infty) = \frac{1}{2} \exp(-2) = 0.067668\dots$ when t increases from zero to infinity and that asymptotic decay appears to be exponential.

Comparing (4.8) and (4.13) shows that the evolution to the final stationary state is strongly dependent on its initial state, i.e., an asymptotic distribution cannot be computed directly from the single requirement of stationarity in Eq. (4.1). The system, therefore, has an infinite number of stationary states, i.e., the kinetic model of crystallization is not ergodic.

In closing, we note that an approach to equilibrium appears to be exponential independent of initial conditions. Indeed, by inspection of (4.1), one finds the universal asymptotic decay

$$Z_N(t) = C_N \exp(-Nt) + \dots \quad \text{at } t \gg 1 \quad (4.14)$$

with some nonuniversal constants C_N .

5. GENERALIZED CRYSTALLIZATION MODEL

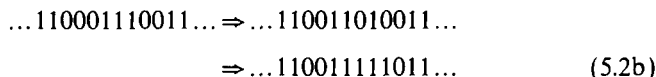
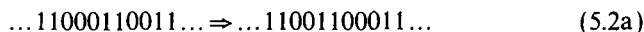
In this section we discuss a natural generalization of the model for crystallization at zero temperature. We will assume that the exchange of a pair of neighboring atoms that would cause neither enrichment nor dilution of good nearest-neighbor pairs is also possible and is performed with the probability ϵ . Returning to the evolution laws (1.C) and (2.C), we conclude that any 011- or 110-triplet can now evolve, $011 \Rightarrow 110$ or $110 \Rightarrow 011$, with the rate ϵ . Thus, the RSA interpretation shows that the generalized model takes into account "diffusion" processes.

Contrary to the simple RSA case, studies of generalized RSA processes with diffusion have begun very recently.⁽⁷⁾ Analytical results for generalized RSA processes are absent, while an approximate treatment⁽⁷⁾ predicts interesting unexpected results, which must, however, be confirmed rigorously or numerically.

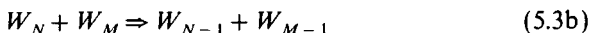
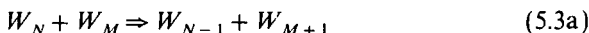
In the present generalized crystallization model the addition of diffusion spoils the independent evolution of each Z_N , and we have found no general solution. However, the case of very weak diffusion ($\varepsilon \ll 1$) can be partly treated because the evolution may be divided into two stages. The first stage was investigated in the preceding section. To describe the evolution in the second stage, we introduce the scaled time

$$T = \varepsilon t \quad (5.1)$$

and use the stationary states of the first stage as the initial data for the second one. Our goal here is a determination of the concentration of clusters of zeros (in the 0-1 representation) of length N at time t , $W_N(t)$. Two basic elementary kinetic processes governing the evolution of W_N 's can be depicted as follows:



The first step in the reaction (5.2b) occurs due to the diffusion process (5.2a), while the second happens instantly in terms of the scaled time T due to the reaction (1.C). The reaction scheme (5.2) may be symbolized as follows:



Notice that the former reaction, (5.3a), explicitly shows that the generalized crystallization process is no longer irreversible, while the latter reaction, (5.3b), describes the irreversible part of the crystallization process.

The basic theoretical approach to describing the reaction scheme (5.3) is based on rate equations. This is an approximation of a mean-field character, as fluctuations are ignored. Clusters are assumed to be distributed homogeneously at all times throughout the system, i.e., there is perfect mixing. While the rate equation approach provides a useful and comprehensive account of a wide range of kinetic processes, recent results by a number of authors show that local spatial fluctuations which are

ignored in a mean-field approximation will dominate the large-time reaction kinetics in low-dimensional systems, namely, at $d < d_c$, where d_c is an upper critical dimension (for a review, see, e.g., ref. 17). However, as will be seen later, d_c is probably equal to 2 in our model because for some initial conditions the present model can be reduced to the annihilation model with known upper critical dimension $d_c = 2$. Therefore, we must treat the model outside the scope of mean-field approximation.

Unfortunately, we have not succeeded in finding an exact treatment of our system of interacting clusters for arbitrary initial data. However, this may be done for particular initial conditions, e.g., for the alternating chain. The first stage of this process, which is described in Section 4, gives the initial conditions for the second stage

$$W_1(T=0) = \rho(t = \infty) = 0.067668\dots$$

$$W_N(T=0) = 0 \quad \text{for all } N \geq 2$$

Thus, in the second stage we start with defects but without domain walls. Furthermore, a simple analysis shows that the fusion reaction, $W_1 + W_1 \rightarrow W_2$, is now impossible because distances between any defects appears to be even. Accordingly, we have a system of defects which execute a random walk along a one-dimensional lattice and annihilate if they land on next-nearest-neighbor sites simultaneously. This reaction may be symbolized as $D + D \rightarrow 0$, and the system would evolve into a trivial, completely empty state in the long-time limit, $T \gg 1$. Thus we see that for the alternating initial data the generalized crystallization process remains *irreversible* and the system reaches the close-parked configuration. So we rigorously confirm, although for a particular situation, the conclusion of Tarjus *et al.*⁽⁷⁾ that in generalized RSA processes with diffusion a system should reach close-parked configurations.

The upper critical dimension for the annihilation process is equal to 2.⁽¹⁷⁻¹⁹⁾ In one dimension, the problem of diffusive annihilation has been investigated extensively and the power-law decay, $W_1(T) \simeq (4\pi T)^{-1/2}$ at $T \gg 1$, is known to be an exact result⁽²⁰⁻²⁴⁾ for the usual variant of a model where particles hop on nearest-neighbor sites and annihilate if they land on the same site. For the crystallization model with the alternating initial data, in the second stage, particles execute a random walk along the even-site sublattice and a small difference between the reaction rules also exists. However, it is clear that these modifications do not change the asymptotic results. From the above analysis, the asymptotic behavior of the defect density can be deduced:

$$W_1(T) \simeq \frac{1}{2}(4\pi T)^{-1/2} = (16\pi\epsilon t)^{-1/2} \quad (5.4)$$

Similar behavior can be found for some other ordered initial conditions. For instance, the chain

$$\dots AAAA BBBB AAAA BBBB \dots = \dots 0001 0001 0001 0001 \dots$$

simply evolves in the first stage

$$Z_1(t) = \frac{1}{4}e^{-t}, \quad Z_N(t) = 0 \quad \text{for all } N \geq 2$$

and defects with the density $W_1 = 1/4$ are produced at the end of this stage. The distance between any two neighboring defects is equal to 4 and they execute a random walk annihilate as for the alternating chain. For another ordered chain

$$\dots AAAA BBBB AAAA BBBB \dots = \dots 0000 1000 0100 0010 0000 \dots$$

the evolution in the first stage produces 2-clusters only, with the density $W_2 = 1/5$ at the end of this stage. There are three elementary reactions in the system: elementary reactions in the system: a breakup of a 2-cluster onto two defects, left and right; an annihilation of some left defect and a right defect of the preceding 2-cluster; and a fusion of defects of a single cluster. The system consists of 1- and 2-clusters during a whole process of evolution. A simple analysis shows that the density of 2-clusters is negligible in comparison with the density of the defects at $T \gg 1$ and we again obtain the asymptotic behavior (5.4) for the density of the defects. On the basis of these examples one can describe the evolution of all ordered lattices with an elementary cell of length $(m+3)$. The first stage is obvious

$$Z_1(t) = (m+3)^{-1}e^{-t}, \quad Z_N(t) = 0 \quad \text{for all } N \geq 2$$

During the second stage the system consists of 1-, ..., m -clusters. Thus asymptotically we again obtain preceding results, Eq. (5.4).

To illustrate other possibilities, we consider the evolution of a lattice of defects with odd period. One can find that the annihilation process is impossible for these initial conditions. Consequently the "mass" of all clusters, i.e., the degree of crystallization, is conserved and a full crystallization cannot be achieved. Thus in these situations the conclusion of Tarjus *et al.*⁽⁷⁾ that in generalized RSA processes with diffusion a system should reach close-packed configurations becomes invalid.

6. SUMMARY

We studied the kinetics of two simple zero-temperature models of a binary alloy on a one-dimensional lattice. For the first model, which

mimics a phase separation process, the nearest-neighbor exchange can occur if the portion of AB bonds is thereby decreased. The crystallization model is defined by the opposite evolution law. Both models prove to be soluble and many features are computed analytically. When a diffusion is taken into account, i.e., the nearest-neighbor exchange is performed if the portion of AB bonds remains invariable, the behavior is drastically changed even for infinitely weak diffusion.

In the absence of diffusion, the evolution to the final stationary states appears to be exponential and strongly dependent on the initial data. There are infinitely many stationary states, i.e., these kinetic models are not ergodic. Assuming the diffusion to be small, we divided the evolution into a first stage in which diffusion is negligible and a second one in which the diffusion plays a crucial role. We observed a very rich kinetic behavior strongly dependent on the initial data. For an alternating chain we have succeeded in describing the kinetics of crystallization in the last stage by mapping our crystallization process onto the one-dimensional annihilation process. For these ordered chains we found that the average crystalline size L scales with time as $t^{-1/2}$ and confirmed the conclusion of Tarjus *et al.*⁽⁷⁾ that in generalized RSA processes with diffusion a system should reach close-packed configurations. We also found initial conditions for which the above conclusion is violated.

Finally we note that very recently exact results for the phase separation model on a one-dimensional lattice have been obtained by Privman⁽²⁵⁾; exact solution of the phase separation model on the Bethe lattice has been found in ref. 26.

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