

Low- and High-Dimension Limits of a Phase Separation Model

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We study a simple zero-temperature model for phase separation of a binary alloy, in which nearest-neighbor interchange can occur if the fraction of AB pairs is not thereby increased. We present analytic results for the one-dimensional case and numerical results for the infinite dimensionality limit on a Cayley tree. In neither limit does the final fraction of AB pairs agree with the dimension-independent result found previously in $d = 3, 4, 5$.

KEY WORDS: Phase separation; spinodal decomposition; simulation; non-ergodic processes.

1. INTRODUCTION

Levy, Reich, and Meakin⁽¹⁾ and subsequently Meakin and Reich⁽²⁾ have performed a zero temperature Monte Carlo simulation of spinodal decomposition in a 50–50 model alloy. They commence by generating a lattice of equal numbers of A and B atoms, either inserted at random or inserted in an alternating structure such that each atom has nearest neighbors of the opposite type. They consider several lattices in dimension $d = 1$ through $d = 5$. The stochastic reorganization of the 50–50 array of atoms is carried out with the following rules:

- (1) A nearest-neighbor pair of atoms is selected at random.
- (2a) If the exchange of the pair of atoms would result in enrichment of “good” (AA or BB) nearest-neighbor pairs it is performed.

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(2b) If the exchange would dilute good pairs it is never performed.

(2c) If the exchange would cause neither enrichment nor dilution it is carried out with probability p . This third case corresponds to diffusion. References 1 and 2 consider only $p = 1/2$, but other values are under study (P. Meakin, private communication).

For $d \geq 2$ with $p > 0$ the process usually never terminates, but eventually becomes purely diffusive. Either $d = 1$ or $p = 0$ results in eventual termination. In all cases the fraction B of "bad" AB bonds [$B = N_{AB}/(N_{AA} + N_{BB} + N_{AB})$] appears numerically to approach a limit considerably greater than zero; complete phase separation ($B \rightarrow 0$ as $N_A + N_B \rightarrow \infty$) is never achieved. For $3 \leq d \leq 5$ on a hypercubic lattice with $p = 0.5$ the limiting B is⁽²⁾ 0.2804 ± 0.001 for an alternating start and 0.274 ± 0.003 for a random start. Note that S/Z in Refs. 1 and 2 corresponds to $(1 - B)/2$. The values for $d = 2$ appear slightly different,⁽¹⁾ 0.2820 ± 0.0004 for an alternating start and 0.264 ± 0.002 for a random start. The dimensionality independence is surprising and warrants further investigation. We have examined the one-dimensional case (Section 2) and the "infinite" dimensionality limit on a Cayley tree (Section 3). We consider only the case of an alternating start.

Besides the intrinsic interest of this model, which mimics spinodal decomposition, it is an example of a physically relevant class of nonergodic Monte Carlo processes whose simplest properties (e.g., B) cannot be found by equilibrium consideration. Here the effective Hamiltonian is that of an Ising ferromagnet (with Kawasaki dynamics), which would show complete phase separation ($B \rightarrow 0$) at zero temperature in equilibrium. This is not seen and instead the evolution and final states depend crucially on the dynamics and on the starting state. Similar processes are found in spin glass models.

2. ONE DIMENSION

We consider a one-dimensional chain of N sites connected into a ring. We first treat the $p = 0$ case of no diffusion. Any state of the chain may be specified in terms of the runs of like spins, called singles, doubles (two As surrounded by Bs, or vice versa), triples, etc. Starting in the alternating ABAB... state (N even), after the first transition there are two adjacent doubles and $N - 4$ singles.

The crucial observation is that the sequence

$$S_n = \text{double}, n \text{ singles}, \text{double}$$

evolves independently of its surroundings, provided its immediate neighbors are not singles. The proviso is true initially, where we have S_{N-4} , and is preserved by the rules now discussed. For $n \geq 4$, S_n evolves in one of the following $n - 1$ equally likely ways:

$$S_n \rightarrow \begin{cases} (\text{triple}, S_{n-3}) \\ (S_0, S_{n-4}) \\ \vdots \\ (S_{n-4}, S_0) \text{ (for } n > 4) \\ (S_{n-3}, \text{triple}) \end{cases}$$

The omitted sequences are of the form (S_p, S_q) with $p + q = n - 4$. S_3 evolves into (triple, S_0) or the reverse, and S_2 evolves into $(\text{triple}, \text{triple})$. S_0, S_1 , and triples cannot evolve further. Let the bonds to the right of each atom in S_n be associated with S_n , so that S_n is associated with 2 good and $n + 2$ bad bonds. Now let $r_n (\leq n + 2)$ be the average number of bad bonds remaining upon completion of S_n 's evolution. The rules lead immediately to

$$r_0 = 2, \quad r_1 = 3, \quad r_2 = 2, \quad r_3 = 3$$

and, for $n \geq 4$,

$$r_n = \frac{1}{n-1} \sum_{p=0}^{n-4} (r_p + r_{n-4-p}) + \frac{2}{n-1} (r_{n-3} + 1)$$

This may be reduced to the recurrence relation

$$nr_{n+1} - (n-1)r_n = 2r_{n-2} \quad (n \geq 3)$$

which generates all r_n 's. In the large system limit B is given by

$$B = \lim_{N \rightarrow \infty} r_{n-4}/N = 0.45090 \quad (p = 0)$$

The result is easily obtained numerically, or by solving the differential equation for the generating function $\phi(s) = \sum r_n S^n$.

The addition of diffusion ($p > 0$) spoils the independent evolution of each S_n , and can generate sequences longer than triples. We have found no general solution, and comment that the dynamics may be expressed in terms of an unsolved four-femion Hamiltonian (using femion operators to create or annihilate bad bonds). However, the addition of an infinitesimal amount of diffusion ($0 < p \ll 1$) is easily treated because the evolution may then be divided into two stages. First the $p = 0$ evolution occurs, leaving triples, S_0 's, and S_1 's. Then the S_1 's evolve further via diffusion, each

eliminating two bad bonds. The number of S_1 s remaining after the first stage is given on average by q_{N-4} , where q_n can be shown to satisfy the same recurrence as r_n for $n \geq 3$, and $q_0 = 0$, $q_1 = 1$, $q_2 = q_3 = 0$. Thus

$$B = \lim_{N \rightarrow \infty} (r_{N-4} - 2q_{N-4})/N = 0.35132 \quad (0 < p \leq 1).$$

This is already most of the way from the $p = 0$ result to the $p = 1/2$ result

$$B = 0.3355 \pm 0.0005 \quad (p = 1/2)$$

which we obtain by simulation. Reference 1 quotes $B = 0.338$ at $p = 1/2$. It seems that B is rather insensitive to p , as long as $p > 0$, although of course the completion time increases as $1/p$. We also simulated the $p = 0.01$ and $p = 1$ cases, with results

$$B = 0.3506 \pm 0.0005 \quad (p = 0.001)$$

$$B = 0.3292 \pm 0.0005 \quad (p = 1)$$

We found no significant size dependence above $N = 1000$. It is clear that the $d = 1$ version of this process is very different from $d = 2$ through 5.

3. CAYLEY TREES

A Cayley tree often plays the role of an infinite-dimensional lattice. We have therefore tried some simulations of the present model on a Cayley tree of coordination number Z , with n generations starting from a central generation 0 site. We consider only $p = 1/2$ and use an alternating start. We allow boundary pairs, between generations $n - 1$ and n , to interchange according to the same rule (Section 1) as for other pairs, even though the outermost atoms have no generation $n + 1$ neighbors. The simulation is run for $100N$ steps past the last B -changing interchange observed, where N is the number of nearest-neighbor pairs,

$$N = [Z(Z - 1)^n - 2]/(Z - 2) - 1$$

This gives good convergence for $Z \geq 4$; $Z = 3$ appears to need even longer runs for reliable results, and is excluded here. We repeat each (Z, n) case 100 times and average the values obtained for B .

Figure 1 shows our results for $Z \geq 4$, $n \geq 4$. We plot $B - 1/Z$ versus $1/N$ (labeled with N) because we find a leading $1/Z$ behavior for B . Error bars are derived from our 100 runs at each (Z, n) . The limit of interest for representing an infinite-dimensional hypercubic lattice is

$$B_\infty = \lim_{Z \rightarrow \infty} \lim_{N \rightarrow \infty} B(Z, n)$$

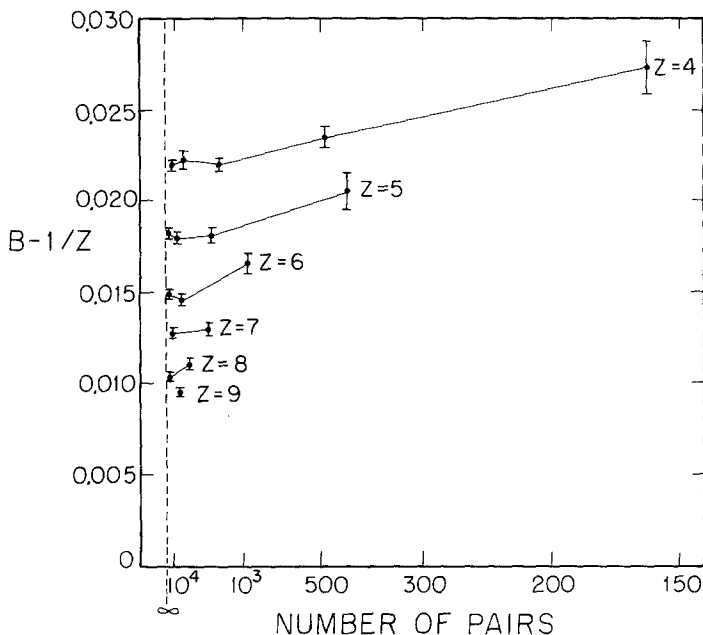


Fig. 1. The fraction B of "bad" AB pairs, minus $1/Z$, after running the dynamical process to completion on Cayley trees of coordinates number Z . The lines join points of the same Z , with $n = 4, 5, \dots$ generations from right to left. The abscissa is linear in $1/N$.

The inner limit appears reasonably well converged and suggests $B_\infty = 0$ for the outer limit. In any case we can be confident in stating $B_\infty < 0.01$, implying almost complete phase separation. If indeed the Cayley tree represents the $d \rightarrow \infty$ limit of the finite d lattices the conjecture⁽²⁾ that B is independent of d for $d \geq 3$ is thereby excluded.

The result $B \sim 1/Z$ is not hard to understand. For large Z almost all the pairs are between generations $n-1$ and n , and interchanging one of these for each generation $n-1$ site leaves no bad pairs among the $n-2:n-1$ pairs, and a fraction $1/Z-1$ bad among the $n-1:n$ pairs. This gives $B = 1/Z$ overall if the few pairs inside generation $n-2$ are ignored. Our simulation results agree with this picture, particularly in that very few bad pairs are found among the $n-2:n-1$ ones. Those pairs inside generation $n-2$ have a much larger bad fraction, which appears to decrease toward a limit as N increases, the limit increasing with Z . We estimate $B_\infty \gtrsim 0.5$ for these inner pairs alone. Again, this excludes the d -independence conjecture.

In summary, we have studied the $d=1$ process analytically and by simulation, and the $d \rightarrow \infty$ Cayley tree by simulation. These limiting cases differ from the intermediate dimensions reported in Refs. 1 and 2.

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