

## Slow equilibration in systems undergoing diffusion-controlled phase separation on a lattice

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We consider binary alloys undergoing phase separation after a deep temperature quench. We present Monte Carlo simulation results showing that such systems freeze when quenched to zero temperature. This holds for a wide range of atomic and vacancy concentrations. We interpret these results in terms of an equilibration mechanism dominated by activated diffusive motion of atoms on a lattice across an interface with an activation energy dependent on the local curvature. We show that in such a case the domain size increases logarithmically for quenches to low temperatures. This logarithmic law is shown to be in excellent agreement with neutron scattering experiments.

### I. INTRODUCTION

A common mechanism for equilibration in solids cooled to below some critical temperature is phase separation associated with the motion of atoms moving diffusively. The form of the growth law as a function of time  $t$  after a quench for the typical domain size  $L(t)$  has usually been assumed<sup>1</sup> to be a power law. In this paper we argue that for binary alloys subjected to a deep temperature quench the phase separation process proceeds, quite generally, more slowly than a power law. The physical origin of this slow equilibration is that diffusion across an interface at low temperatures is an activated process with an activation energy inversely proportional to the local curvature. For quenches to zero temperature we find, using Monte Carlo (MC) simulations, that such systems freeze in a nontrivial fashion<sup>2</sup> after growing domains up to some finite size. The structure of the frozen system is highly disordered and the domains form either percolative structures or, at small minority concentrations, small nonspherical compact structures. These glassy configurations are formed dynamically in the absence of any quenched disorder in the system. As we discuss in detail below, the freezing behavior appears for critical or off-critical quenches, in the presence of both thermal and quenched vacancies, for several two-dimensional lattices, and for quench rates which are not infinitely rapid.

We have recently pointed out<sup>1,3</sup> that this freezing behavior occurs in the particular case of an infinitely rapid quench in a binary system with a critical composition ( $c=0.5$ ) and no vacancies on a square lattice. Following that work, however, the possibility existed that this freezing was restricted to that special case because of the high symmetry and the resulting percolative structure which is in contrast to the dropletlike structure assumed in most of the theoretical work, as, for example, in the Lifshitz-Slyozov (LS) theory.<sup>4</sup> A second concern was that the

freezes might unlock if thermal vacancies are introduced into the system. The vacancies would naturally "wet" the interfaces and therefore increase diffusion of particles across an interface and possibly permit the system to avoid the freeze found in the absence of vacancies. The concentration of vacancies needed might be thought to be related to the percolation concentration. The assumption of infinitely fast quench rates and a specific (square) lattice were additional concerns. A key result of the present paper is that, in fact, neither the relative concentration of atoms nor the number of annealed vacancies have, alone or combined, any qualitative effect and only a small quantitative effect, on the freezing behavior of the spin exchange kinetic Ising model used in Refs. 1 and 3 (and defined in Sec. II below).

In Refs. 1 and 3, by using a combination of renormalization-group (RG) and MC methods we showed that, at least in the particular case considered in that work, the freezing of the system for quenches to a final temperature  $T_F=0$  was tied to a logarithmic growth law for  $L(t)$ , for quenches to  $T_F>0$ , in disagreement with previous expectations of a power-law form. Given the generality of the freezing behavior, it is natural to investigate the possibility that the logarithmic law is also generally valid. Our arguments for this are presented in Sec. III. Furthermore, we show also in Sec. III that the logarithmic growth law gives a superior fit to recent neutron scattering experiments<sup>5,6</sup> on binary alloys when compared to power-law fits.

We believe that these findings may have important consequences with regard to the question of the universality of growth kinetics associated with first-order phase transitions. According to the analysis of Refs. 1, 7, and 8, the growth of order in kinetic Ising models can be classified in terms of the renormalization-group fixed-point structure underlying the scaling behavior found in these systems: if spatial coordinates are rescaled by a factor  $b > 1$ , then the system is self-similar upon rescaling of the

time variable by a factor  $\Delta(b) < 1$ . For quenches from high temperatures to temperatures  $T_F$  below the critical temperature  $T_c$ , the renormalization-group flows for  $T_F$  iterate toward the zero-temperature fixed point. The long-scale behavior of the system is governed by the behavior at  $\Delta(b, T_F)$  as  $T_F \rightarrow 0$ . When the order parameter is not conserved it has been well established<sup>1,7-9</sup> that  $\Delta = b^{-2}$  in that limit. It is straightforward to show that this leads to the Lifshitz-Cahn-Allen<sup>10</sup> growth law [ $L(t) \sim t^{1/2}$ ]. However, the situation for phase separation on a lattice, with a conserved order parameter, is qualitatively different. The time-rescaling factor  $\Delta$  does not tend to a finite constant at low temperatures, but it approaches zero instead.<sup>11</sup> The vanishing of  $\Delta$  at zero temperature follows from the fact that the system freezes.<sup>12</sup> At finite temperatures the activated behavior of  $\Delta$  leads to a logarithmic growth law, as shown in Ref. 1. Turning the argument around, if the growth law is logarithmic, as discussed in Sec. III, and given by  $L(t, \xi) = L_0 + \xi \ln(V_0 t / \xi)$ , then the assumption of self-similarity,  $L(t, \xi) = bL(\Delta t, \xi/b)$ , determines  $\Delta$  to be of the form  $\Delta = b^{-1} e^{-L_0(b-1)/\xi}$ .

This paper is organized as follows: We define, in Sec. II, the model studied in the MC simulations and give the MC results for quenches to zero temperature under a variety of conditions. Then, in Sec. III, we present our interpretation of the MC results and their implications for quenches to finite temperatures in terms of simple considerations of interface dynamics. We also present a detailed comparison with the results of Ref. 5. In Sec. IV we recapitulate and point to directions for future work.

## II. MODEL AND MONTE CARLO RESULTS

The model we study is the standard Kawasaki<sup>13</sup> spin-exchange kinetic Ising (SEKI) model of a binary alloy. We consider a set of variables  $\sigma_i$  defined on lattice sites  $i$ . In the absence of vacancies, the  $\sigma_i$  are Ising variables,  $\sigma_i = \pm 1$ , the two values representing two chemical species  $A$  and  $B$ . We limit ourselves to the case where there are only nearest-neighbor interactions with coupling constant  $K = -\beta J = -J/k_B T$ , where  $J$  is the exchange constant which, in the present case of phase separation, must be taken to be positive. For fast quenches to a final temperature  $T_F$ , the mode assumes nearest-neighbor exchange probability (due to interactions with a thermal bath) of the form

$$W_{ij} = \frac{1}{2} [1 - \tanh(\beta \Delta E / 2)], \quad (2.1)$$

where  $\Delta E$  is the energy difference between the configurations before and after the exchange. Equation (2.1) implicitly defines the unit of time. We will measure our time in Monte Carlo steps (MCS) defined as 1 MCS =  $N$  exchange attempts, where  $N$  is the number of sites. Most of the work reported here was performed on a square lattice, but some of the results were also obtained on a triangular lattice as we shall see.

To introduce vacancies, we allow the variables  $\sigma_i$  to take on a third value,  $\sigma_i = 0$ . Thus, the energy associated with a nearest-neighbor bond between an atom and a vacancy vanishes. We will be concerned here mostly with

the case of annealed vacancies, which are free to move by exchanging positions with neighboring atoms according to the probability given by (2.1). Note that  $W_{ij}$  satisfies the detailed balance condition whether vacancies are present or not. We will also briefly consider the case of quenched, fixed vacancies where all attempts to exchange an atom and a vacancy fail.

As explained in the Introduction, we are primarily concerned here with quenches from a totally disordered state ( $T_I = \infty$  or  $K_I = 0$ ) to zero temperature ( $T_F = 0$  or  $K_F = \infty$ ), for various values of the relative atomic concentration and number of annealed vacancies. We have performed our MC simulations, for the above model, for different values of the concentration  $c = N_A / (N_A + N_B)$  of  $A$  particles and of the vacancy density  $v = N_v / N$  ( $N$  is the total number of sites). We have investigated the values of  $c$ ,  $c = \frac{1}{2}, \frac{1}{3}, \frac{1}{6}$ , and  $\frac{1}{10}$ , and 15 values of  $v$  ranging from 0 to 0.85. In all cases the system freezes: further exchanges lowering the energy are impossible.

To see how this happens, and to understand why, it is useful to first consider, qualitatively, the variables  $v$  and  $c$  separately. When  $v = 0$  but  $c < \frac{1}{2}$ , the morphology of the system changes from a percolative structure similar to that found at  $c = \frac{1}{2}$  (as given, e.g., in Fig. 1 of Ref. 1) to a collection, when  $c$  is very small, of many compact structures imbedded in a matrix of the majority phase. The domains of the minority phase are, however, not droplet-like but similar to those found in Refs. 1 and 14. This can be clearly seen in Fig. 1.

Let us now turn to what happens for quenches with different values of  $v > 0$ , and  $c = \frac{1}{2}$ . When  $v$  is small, the system freezes with vacancies occupying positions in the walls between  $A$  and  $B$  domains, but which are otherwise random. This "wetting" behavior would raise expectations that, when  $v$  reaches a value related to the percolation threshold,<sup>15</sup> a change in behavior may occur. What actually happens as the vacancy density increases is that part of the vacancies phase separate: they start to form their own domains (although of a smaller characteristic size) and therefore have no effect on the rest of the system. The overall appearance of the system is again as found in Refs. 1 and 14, but with three kinds of domains. An example of the morphological appearance of the system in its frozen configuration is given in Fig. 2(a). Further increases in  $v$  have no qualitative effect. Nothing whatever happens as  $v$  is increased beyond the percolation concentration. For noncritical quenches, the morphology of the system is still the same: Fig. 2(b) corresponds to the same vacancy concentration as Fig. 2(a) ( $v = 0.25$ ) but for a noncritical quench ( $c = \frac{1}{3}$ ). It is hard to distinguish between the two figures without actually counting the plus and minus signs.

This behavior can more easily be understood by recalling the equilibrium phase diagram of an Ising model with a fixed number of annealed vacancies.<sup>16</sup> At low temperatures two phases coexist, one phase has a small vacancy concentration, and the other is vacancy-rich: Once it is established that a small density of vacancies has little effect on the system, the separation tendency (one must emphasize that separation is only very partial) negates the ef-

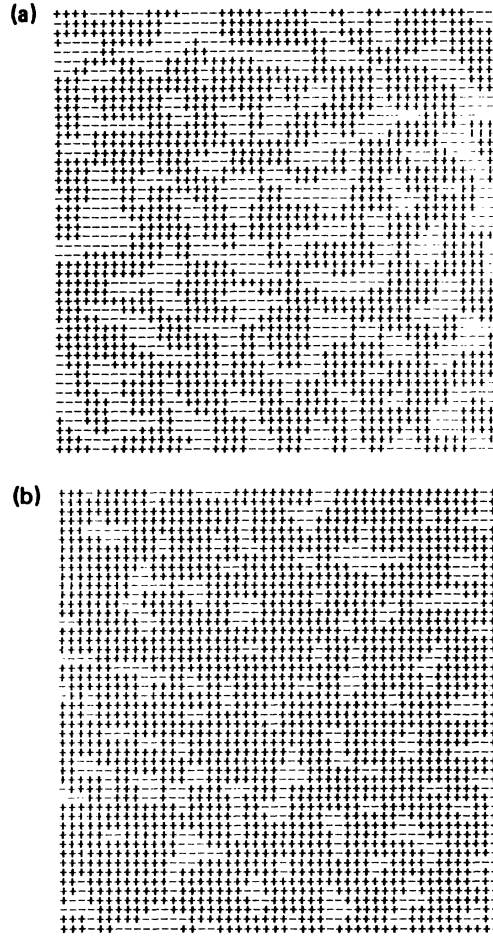


FIG. 1. Morphology of the quenched structure for noncritical quenches. The  $-$  signs represent  $A$  atoms and the  $+$  signs  $B$  atoms. The concentration  $c$  of  $A$  atoms is  $c = \frac{1}{3}$  in panel (a) and  $c = \frac{1}{6}$  in panel (b). Both panels correspond to snapshots taken at  $t = 400$  MCS, at which time no further exchanges lowering the energy are possible. The size of the system is  $48 \times 48$  sites.

fect of increasing vacancy density. In fact, the notion that increasing either  $c$  or  $v$  will increase diffusion is based on the idea that differences in chemical potential will drive the interface velocity. But, since the competing phases have the same chemical potential, this does not happen.

Clearly, it is desirable to make the above considerations more quantitative. A good measure of how far the system has progressed toward equilibrium is given by the quantity

$$r(t) = \frac{\varepsilon(t) - \varepsilon(0)}{\varepsilon_E - \varepsilon(0)}, \quad (2.2)$$

where  $\varepsilon(t)$  is just the nearest-neighbor correlation function (or energy in units of  $-4J$ ), and  $\varepsilon_E = 1 - v$  is the equilibrium value of  $\varepsilon$  associated with the final state  $T_F = 0$ , while  $\varepsilon(0)$  corresponds to the initial state  $T_I = \infty$ . Note also that  $\varepsilon_E - \varepsilon(t)$  is a measure of the amount of interface

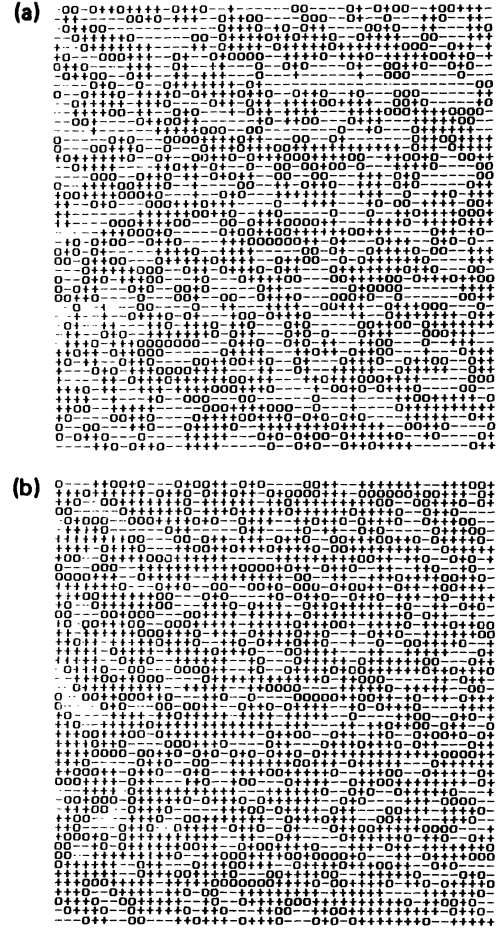


FIG. 2. Morphology of the system when annealed vacancies are present. The symbol 0 represents a vacancy. In panel (a)  $c = \frac{1}{2}$  (critical quench) and the vacancy concentration is  $v = 0.25$ , while in panel (b)  $c = \frac{1}{3}$  and  $v = 0.25$ . All other parameters are as in Fig. 1.

present in the system. The quantity  $r(\infty)$  measures how far the system has progressed toward equilibrium before freezing. If the system reaches equilibrium  $r(\infty) = 1$ .

We have obtained the quantity  $\varepsilon(t)$  (and  $r$ ) from our MC simulations, for the values of  $c$  and  $v$  indicated above. The simulations have been carried out for square-lattice systems ranging in size from  $16 \times 16$  to  $48 \times 48$  (we have verified that finite-size effects are eliminated<sup>17</sup>) and for times long enough (several hundred MC steps) so that the system locks up: additional exchanges increasing  $\varepsilon(t)$  are impossible. The results are averaged over enough runs (20–25) to allow for at least two-digit precision in  $\varepsilon(t)$ . The time it takes for the system to freeze can be associated with a characteristic time  $\tau$  defined by  $\varepsilon(\tau) = 0.97\varepsilon(\infty)$ . Although  $\varepsilon(t)$  is a smooth, monotonic function, the way in which  $\varepsilon(t) \rightarrow \varepsilon(\infty)$  cannot be characterized by an exponential or a single power law.

For critical quenches ( $c = \frac{1}{2}$ ) the results are reported in Table I, as a function of  $v$ . One can see that  $r(\infty)$  at first increases slightly with  $v$ , to a maximum of only 0.52 at  $v = 0.4$ : the annealed vacancies favor equilibration very

TABLE I. Results for critical quenches to  $T_F=0$ , as a function of the vacancy concentration  $v$ . The quantity  $\varepsilon(t)$  is the energy (in units as defined in the text), and  $r(t)$  is given by Eq. (2.2).

$v$	$\varepsilon(t=\infty)$	$r(\infty)$
0.0	0.46	0.46
0.05	0.44	0.46
0.1	0.42	0.47
0.22	0.39	0.49
0.25	0.37	0.50
0.28	0.35	0.50
0.33	0.34	0.51
0.35	0.33	0.51
0.40	0.31	0.52
0.45	0.27	0.51
0.47	0.27	0.50
0.5	0.25	0.49
0.6	0.19	0.48
0.75	0.11	0.44
0.85	0.05	0.33

slightly. When  $v$  is increased further, however,  $r(\infty)$  declines sharply, down to a value of 0.33 at  $v=0.85$ . The time  $\tau$  remains at  $\sim 60$  MCS, independent of  $v$ , for  $c=\frac{1}{2}$ . Thus, we see that the vacancies make only a small quantitative contribution to the equilibration of the system when  $0 < v < 0.4$ .

On the other hand, the main effect of increasing  $c$  at constant  $v$  is to sharply decrease  $\tau$ , e.g., to  $\sim 10$  MCS at  $v=0.1$  and  $c=0.1$ . For noncritical quenches the system already has, at  $t=0$ , a finite value of  $\varepsilon(t)$ ; therefore  $\varepsilon(t)$  saturates faster, even though  $\varepsilon(\infty)$  does increase somewhat with  $c$ . In all cases we have investigated,  $r(\infty)$  does not exceed 0.55.<sup>18</sup> These results are summarized in Table II. Note that the values of  $r(\infty)$  and  $\varepsilon(\infty)$  that we find represent a high degree of disorder. For the equilibrium system, with  $c=\frac{1}{2}$  and  $v=0$  a value of  $\varepsilon=0.55$  occurs only when the temperature is well above  $T_c$ , and the system is well into the disordered phase. Thus, the quenched system is very far from equilibrium when it freezes.

The annealed, mobile vacancies discussed above must be distinguished from quenched, fixed vacancies, which, as is well known, are often an important factor in preventing a system from reaching equilibrium. Indeed, if we assume quenched, instead of annealed vacancies, we find that  $r(\infty)$  always decreases as a function of vacancy density: at  $c=\frac{1}{2}$  and  $v=0.25$ , for example,  $r(\infty)$  is 0.28 for quenched vacancies and 0.50 for annealed. The two kinds of vacancies have different effects which should not be confused. Quenched vacancies obviously do not phase separate and equilibrium conditions are then determined<sup>19</sup> from percolation considerations.

We have performed two additional checks of the generality of the freezing effect. First, we have performed similar simulations for a triangular lattice and found the same behavior: the system freezes. Second, to make sure that the effect is not an artifact from the assumption of a sudden quench to  $T_F=0$ , that is, coupling  $K_F=\infty$  or  $u_F=\tanh K_F=1$ , we have performed quenches with a

TABLE II. Results for noncritical quenches as a function of atomic concentration  $c$  and vacancy concentration  $v$ . The time  $\tau$  is defined in the text, and it is given in MCS.

$v$	$c$	$\varepsilon(\infty)-\varepsilon(0)$	$r(\infty)$	$\tau$
0.1	$\frac{1}{2}$	0.42	0.47	50
0.1	$\frac{1}{3}$	0.38	0.45	50
0.1	$\frac{1}{6}$	0.20	0.36	20
0.1	$\frac{1}{10}$	0.14	0.36	10
0.25	$\frac{1}{2}$	0.37	0.50	60
0.25	$\frac{1}{3}$	0.33	0.48	40
0.25	$\frac{1}{6}$	0.22	0.44	30
0.25	$\frac{1}{10}$	0.17	0.44	20
0.5	$\frac{1}{2}$	0.25	0.50	60
0.5	$\frac{1}{3}$	0.25	0.53	60
0.5	$\frac{1}{6}$	0.22	0.55	50

time-dependent bath temperature:  $u_F(t)=1-e^{-\gamma t}$ . This corresponds to the equilibrium correlation length at the bath temperature vanishing as  $1/t$  at long times. We have used times  $\gamma^{-1}$  ranging from 1 to 100 MCS. (The case  $\gamma=\infty$  is the sudden-quench limit.) We again find that the system freezes. Although a finite  $\gamma$  has a very noticeable effect on  $\tau$  (since obviously  $\tau > \gamma^{-1}$ ), it has only a fairly small effect on  $\varepsilon(\infty)$ .

The inescapable conclusion is that the freezing phenomena are in fact quite robust, and not at all a result of the limitations and assumptions of the specific case discussed in Ref. 1. In Sec. III we turn to further interpretation of these results.

### III. ANALYSIS

Let us now turn to some general considerations which are helpful in understanding the results presented in the preceding section and in relating these results to the behavior of the system when quenched to finite, low temperatures. This analysis is qualitative but leads to a physical understanding of the more rigorous renormalization-group analysis given in Ref. 1. Let us assume that our system has evolved sufficiently that essentially any further motion occurs at an interface. Then for the SEKI model we can define a local interface such that as one crosses the interface the sites above and below the interface correspond to different chemical species (see Fig. 3). The "velocity" or rate  $V$ , at which a particle, for example, the circled particle in Fig. 3(a), will cross the interface is proportional to the probability that this particle will exchange positions with a neighboring particle across the interface. As seen in Fig. 3, each exchange requires a finite amount of energy  $\Delta E$ , given by

$$\Delta E = 2J(z-1-n), \quad (3.1)$$

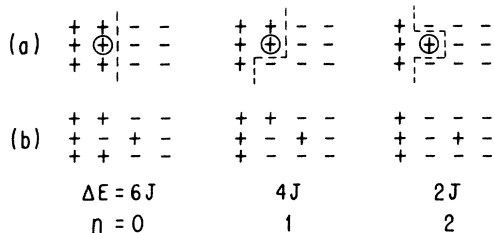


FIG. 3. The top of the figure, (a), shows the three possible configurations seen by the circled + particle at the interface. The position of the interface is shown by a dotted line. The configurations (b) show the system after exchange. Below each configuration is the change in energy in the exchange, ΔE, and the number n of chemically different neighbors which generate a kink in the interface.

where n is the number of chemically different neighbors of the exchanging particles which generate a kink in the interface (see Fig. 3). We can then, in a rough sense, define a local curvature κ via

$$\kappa = (z - 1 - n)^{-1} \tag{3.2}$$

As n takes on its various discrete values, κ increases as n increases. Since n = 0 corresponds to a locally flat interface, this identification makes sense. We have then, from (2.1),

$$V = W = \frac{e^{-2K/\kappa}}{1 + e^{-2K/\kappa}} \tag{3.3}$$

It is clear then, at zero temperature, after one has set up a collection of interfaces of the type defined above, that V = 0 and the system will freeze.

The analysis above concerns the introduction of the local “curvature” κ. We now consider a situation where there are a collection of droplets, and a given droplet is characterized by a “radius” R. It is, of course, impossible to go from a local, discrete variable which can take only a small number of values to a continuous variable ranging from zero to infinity. Note, however, that the important point is not the range of κ, but the range of κξ which is from zero to infinity. We believe it is reasonable, in developing a theory for the velocity of the interface of this droplet, to assume one can use (3.2), but with the local “radius” κ<sup>-1</sup> replaced by the average radius (R - L<sub>0</sub>), where L<sub>0</sub> reflects the fact that for small enough R one obtains monomers which are mobile (not activated) even at zero temperature (ΔE = 0 for monomers). For sufficiently low temperatures the equilibrium correlation length ξ is proportional to 1/K<sub>F</sub> and one can write that the interface velocity is given by

$$V = V_0 e^{-(R-L_0)/\xi} \tag{3.4}$$

While one can then, using this assumption, develop a theory of the droplet growth or decay for this system, for our purposes here it will be sufficient to use a scaling argument to extract the growth law associated with (3.4). Assuming

$$V = \frac{dR}{dt} \tag{3.5}$$

and R ~ L we have

$$\frac{dL}{dt} = V_0 e^{-(L-L_0)/\xi} \tag{3.6}$$

Clearly this has the solution

$$L(t) = L_0 + \xi \ln(V_0 t / \xi), \tag{3.7}$$

which is essentially the result obtained in Refs. 1 and 3.

This analysis should be contrasted with that due to Lifshitz and Slyozov<sup>4</sup> which has been supposed to be applicable to this problem. In this case the minority phase is assumed to form compact structures (droplets) which can be characterized by some effective R<sub>i</sub> for the i<sup>th</sup> droplet. It is then argued, invoking local equilibrium at the interface and the Gibbs-Thomson relation, that the velocity of the interface of this compact structure is given by

$$V_i = \frac{D}{R_i} (\Delta - \alpha / R_i), \tag{3.8}$$

where D is a diffusion coefficient, Δ is the degree of local supersaturation, and α depends on the surface tension and the concentration of saturated solution. Combining this result with the constraint of conservation of particles, one can develop a statistical theory for the growth of droplets in this system. The main result for our purposes here is that the characteristic droplet size L(t) grows as t<sup>1/3</sup>.

This result is quite different from what we have found and what is found experimentally (see below). It follows from our arguments that the assumption of local equili-

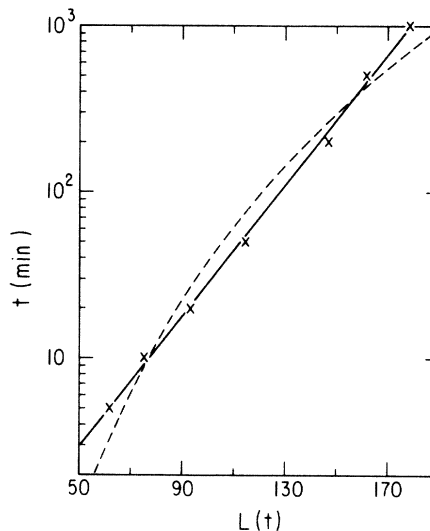


FIG. 4. Two different fits to the experimental results of Ref. 5. The crosses represent the results for the quantity L(t) (in angstroms) as given in Table VII of Ref. 5 for sample A. The dashed line is the best least-squares power-law fit, L(t) = 48.75t<sup>(0.198)</sup>. The fit is clearly not as good as that obtained from a least-squares fit to a logarithmic law, L(t) = 22.10 ln t + 25.85, given by the straight solid line.

TABLE III. Comparison of power-law and logarithmic fits for the data on Table VII of Ref 5 (samples *A-D*, *A'-D'*) and on Table I of Ref. 6 (which we label *H*). In the second column is the best-fit power-law exponent for each sample. In the last two columns we have the coefficients of determination for the power-law and logarithmic fits. The overall size of these coefficients reflects the quality of the corresponding data, which varies from sample to sample. We see that the logarithmic fit is always better, except for sample *C'* where the exponent is smallest.

Sample	Exponent	$r^2$ (power)	$r^2$ (log)
<i>A</i>	0.20	0.97	0.995
<i>B</i>	0.11	0.57	0.63
<i>C</i>	0.22	0.98	0.99
<i>D</i>	0.12	0.59	0.66
<i>A'</i>	0.13	0.98	0.99
<i>B'</i>	0.16	0.83	0.87
<i>C'</i>	0.088	0.95	0.93
<i>D'</i>	0.10	0.77	0.81
<i>H</i>	0.30	0.97	0.99

brium and the use of the related Gibbs-Thomson relation are not justified in the case of solid-on-solid diffusion as manifested by the SEKI model at low temperatures. There simply does not appear to be any compelling justification for assuming that the rate at which particles leave the surface of a droplet at low temperatures is proportional to the equilibrium averaged diffusion coefficient *D* governing the minority set of particles. Our statements are, at this point confined to the case of solids. Phase separation involving fluids may be quite different. Indeed the Lifshitz-Slyozov<sup>4</sup> model may well then apply. Thus, in contrast to critical phenomena, the existence of an underlying lattice and the associated processes at low temperature can play an important qualitative role in the growth kinetics of certain systems.

Our logarithmic growth law gives a superior fit to recent neutron scattering experiments<sup>5,6</sup> on binary alloys when compared to power-law fits. This can clearly be seen in Fig. 4 and Table III where experimental results of Ref. 5 are compared with a power-law fit and the logarithmic law. For all samples in Ref. 5 (except one for which the exponent is very small) we find that the logarithmic law gives a better fit to the experimental data than a power law. The logarithmic fit is even superior to the four-parameter, two-power-law form used in Ref. 5 to fit some of their data. Furthermore, the exponents fitted in Ref. 5 (none of which is near  $\frac{1}{3}$ ) vary wildly from sample

to sample, and it is hard to believe that they could represent a general law. The logarithmic law agrees also with the experimental results of Ref. 6 better than any power law. Thus, the experimental evidence clearly favors our conclusions.

#### IV. CONCLUSIONS AND FUTURE WORK

The main conclusion of this paper is that the freezing phenomena associated with zero-temperature quenches are in fact quite robust and not at all a result of the limitations and assumptions of the specific case discussed in Ref. 1. We have also presented physical arguments based on the hypothesis of the existence of an activated domain interface velocity. These arguments are consistent with the conclusion that the characteristic domain size increases only logarithmically for quenches to finite temperature. This conclusion appears to be substantiated by experiment, as we have shown by comparing with the results of Refs. 5 and 6.

The physical arguments given in Sec. III, while appealing to physical intuition, are certainly not conclusive. For the case of critical quenches, a detailed analysis of the problem (leading to the same logarithmic law) was performed by a combination of MC simulations and RG techniques in Ref. 1 (see also Ref. 3). It is obviously desirable to generalize the analysis of Ref. 1 to noncritical quenches. Note that it is virtually impossible to conclusively prove the existence of a logarithmic law by direct "brute force" MC simulations. As shown in Ref. 3 over the range of times accessible to MC simulations, the logarithmic law mimics quite well a power law with some effective exponent.

Our statements are restricted to solids. For spinodal decomposition in fluids the Lifshitz-Slyozov theory may apply. Thus, in contrast to critical phenomena, the existence of an underlying lattice and the associated activated processes at low temperatures can play an important qualitative role in the growth kinetics of certain systems. There are many systems which freeze after being quenched to zero temperature and it is likely that at least some of these fit into the same class of dynamic behavior for the growth kinetics as discussed here.

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<sup>1</sup>G. F. Mazenko, O. T. Valls, and F. C. Zhang, Phys. Rev. B 31, 4453 (1985).

<sup>2</sup>By nontrivial we mean that this behavior arises specifically from the dynamics of the system. The system does not freeze for an antiferromagnetic coupling.

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