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The Role of Dimensionality in the Kinetic Ising Model of Spinodal Decomposition: Evidence from Zero-Temperature Quenches

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We study a three-dimensional Ising lattice gas model with spin-exchange dynamics quenched from infinite to zero temperature. We consider a wide range of values of the binary composition (i.e., magnetization) and annealed vacancy concentration. We find that, as in two dimensions, the system freezes in a configuration very far from equilibrium, and that the interface energy per bond in the frozen state, which is very large, in all cases takes very nearly the same values as in two dimensions. We discuss the implications of these results regarding the irrelevance of dimensionality in this problem.

KEY WORDS: Kinetic Ising model; nonequilibrium phenomena; dimensionality; relevant variables; spinodal decomposition; interface energy.

1. INTRODUCTION

There is a widespread expectation that the laws that describe the late stages of domain growth in an unstable system may be universal (see Refs. 1 for recent reviews). The main reason for this is an analogy with second-order phase transitions. Even though there is evidence^(2,3) to support this expectation to some extent, the domain-growth universality classes seem to be more restrictively defined than in critical phenomena, in that the number of relevant variables is larger. Among the factors known to be relevant are, for example, the number of coexisting low-temperature phases,^(2,4) the conservation laws for the order parameter (see Refs. 1 for numerous references to this point), the number of components of the order parameter,^(5) the

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presence of a lattice structure,(6) and perhaps the quench rate, the range of interactions, and the concentration.

On the other hand, the dimensionality, which plays a most crucial role in critical phenomena, is not always a relevant variable from the viewpoint of domain growth. It is known, for example, that the Cahn–Allen(7) 1/2 exponent power law for growth after a deep temperature quench in a system with a nonconserved order parameter holds in any dimensionality d, provided only that $d > 1$. This can be attributed to the fact that the Cahn–Allen growth process is driven by the curvature of the growing domains. However, the important question of the relevance of dimensionality has not been the subject of any general discussion.

This question is of more than academic interest. Numerical simulations are very extensively used to study nonequilibrium phenomena. Experience has shown that very large numbers of runs and large-size systems are often required to obtain reliable results. Simulations in three dimensions are necessarily less extensive than in two because of the greatly increased costs in computer time. A degree of confidence in the irrelevance of dimensionality is then needed to make the leap from simulation results in two dimensions to the experimental, usually three-dimensional world.

In this paper we attempt to throw some light on some aspects of this subject. We consider a three-dimensional Ising lattice-gas model with a conserved order parameter (i.e., Kawasaki spin-exchange dynamics(8)) quenched from infinite to zero temperature. In two dimensions this system freezes after such a quench (see, for example, Ref. 9) in a relatively short time and in a fairly disordered structure, that is, a very large amount of interface energy is locked in the frozen state. In the present work we study by Monte Carlo (MC) simulation a very wide range of concentrations in the binary mixture represented by the Ising lattice gas model, with annealed vacancies present in various amounts. We find not only that the system in all cases freezes when quenched to zero temperature, but, more important, that the freezing structure is the same in three and in two dimensions in that the amount of interface energy locked in the frozen state is, for all binary compositions and vacancy concentrations studied here, numerically nearly the same in three and in two dimensions. This similarity extends to the energy of the system as a function of Monte Carlo time. Thus, dimensionality is *quantitatively* irrelevant here even though order extends only to rather short distances.

We also believe that our results are a strong indication, but not a conclusive proof, that the growth law for this system is independent of the dimensionality for quenches to all temperatures. However, the growth law for quenches to finite temperature in two dimensions for this system is still a matter of dispute. An old argument by Lifshitz and Slyozov(10) gives

$L(t) \sim t^a$ with $a = 1/3$ [$L(t)$ is the domain size and t the time]. Experimentally, power law fits give values of a between 0.09 and 0.3 for alloys.[2] Many experimental results can be fit to a logarithmic law,(9,13) which might follow from the fact that the growth mechanism involves an activated energy related to the curvature of the domain walls. We do not wish to address this controversy(14) here. In our opinion, however, the quantitative agreement between zero-temperature quenches in two and three dimensions is a strong (although not conclusive) argument for the dimensionality being an irrelevant parameter for this problem. Regardless of the exact form of the growth law, at least for quenches to low temperatures the growth will be dominated by the activated processes unfreezing the locked-in surface energy, which, as shown here, is present in the same amount in two and three dimensions. The activation energies involved differ in two and three dimensions by factors involving the coordination number that are taken into account [see Eq. (4) below] in the standard normalization of the energy. This conclusion seems to be borne out by the rather limited finite-temperature simulation results available in three dimensions.(15)

2. MODEL AND RESULTS

As stated above, we study the standard spin-exchange kinetic Ising model (SEKI).(8) In the absence of vacancies, we introduce Ising variables σ_i, which are defined on the sites i of a cubic lattice and take the values $\sigma_i = \pm 1$. These correspond to two different chemical species A and B. We will include (as was done in Ref. 9 in two dimensions) only nearest neighbor interactions, with coupling $K = -\beta J = -J/KT$, where J is the negative exchange constant and β the inverse temperature. The nearest neighbor exchange probability is given by

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

where ΔE is the energy difference between configurations before and after the exchange. In the case $\beta \to \infty$ considered here W_{ij} is a step function. Whenever $\Delta E = 0$ we take $W_{ij} = 1/2$. Vacancies are included by allowing σ_i to take a third value $\sigma_i = 0$. We consider here only annealed vacancies, which are allowed to exchange positions with neighboring A or B atoms according to the probability given by Eq. (1). We use standard MC procedures. The pair of spins to be exchanged is chosen at random, to avoid spurious correlations.

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[2] See, e.g., Ref. 11. An extensive review can be found in Ref. 12.

We have obtained results for many different values of the composition c and vacancy concentration v. We define these quantities as

$$c=\frac{N_A}{N_A+N_B} \tag{2}$$

and

$$v=\frac{N_v}{N_A+N_B+N_v} \tag{3}$$

where N_A, N_B, and N_v are the number of A particles, B particles, and vacancies, respectively. Note that $N_A+N_B+N_v=N$, where N is the number of sites.

Since preliminary runs indicated that in three as in two dimensions the system exhibits only short-range order in the frozen state, we have focused here on the nearest neighbor correlation function:

$$\varepsilon(t)=\frac{1}{zN}\sum_{i,\delta}\langle\sigma_i\sigma_{i+\delta}\rangle \tag{4}$$

where z is the number of nearest neighbors (six) and δ represents a lattice vector connecting nearest neighbors. Following Ref. 9, we introduce a quantity $r(t)$, which measures how far the system has progressed toward equilibrium at time t. A convenient definition is

$$r(t)=\frac{\varepsilon(t)-\varepsilon(0)}{\varepsilon_E-\varepsilon(0)} \tag{5}$$

where $\varepsilon_E=1-v$ is the equilibrium value of ε at zero temperature. At infinite temperature one has $\varepsilon(0)=[(1-v)(1-2c)]^2$.

Since $\varepsilon(t)$ is the energy per bond in units of J, the quantity $\varepsilon_E-\varepsilon(t)$ is, in the same units, the amount of interface energy present in the system at time t, while $\varepsilon_E-\varepsilon(\infty)$ represents the amount of interface energy locked in the frozen state of the system. This quantity would be zero in the limit of very large N if the system reached equilibrium, in which case $r(t)$ would tend to unity at long times. This is what would happen if the model discussed here had been quenched to a nonzero temperature, although the time and size scales required to establish this conclusively would be extremely large.

In all cases we have started the quench from infinite temperature ($K=0$). We have obtained data for $c=1/2$, $1/3$, $1/6$, and $1/10$ and $v=0$, $1/10$, and $1/4$. For all values the system freezes after a few hundred MC steps (MCS). We carried our simulations out to several thousand MCS.

Because of the short range of the ordering, we found a value of $N=25^3$ amply sufficient to eliminate finite-size effects, which we checked by obtaining data also at $N=20^3$ and in several cases at $N=32^3$. The results are averaged over several runs. Typically about 10–15 runs are required to obtain at least 5% precision for $\varepsilon(t)-\varepsilon(0)$.

The most striking feature of our results is that they are virtually indistinguishable from those obtained in two dimensions.(9) This can first be seen in Table I, where we show $\varepsilon(\infty)-\varepsilon(0)$ and $r(\infty)$ for the values of c and v indicated above. One can see that in all cases the small differences (which appear unsystematic) are nearly compatible with the *combined* uncertainties of the numerical results. It is also noteworthy that there is no qualitative difference between critical and noncritical quenches, and that annealed vacancies do not contribute in any way to unlocking the freeze. There is no evidence of any change in the behavior of $r(\infty)$ in the small-c ($c=1/10$) case, where initially only monomers are present, nor of any any effects related to percolation. In three as in two(9) dimensions these factors appear to have only secondary importance. Since uncertainties have been estimated rather conservatively, we believe that actual differences of order of 5% may exist between two- and three-dimensional results. For the particular case where no vacancies are present and the concentration is 50%, our results fully confirm those of Ref. 16, where it was found that for $v=0$, $c=1/2$ the average number of satisfied bonds in the frozen configuration is very approximately independent of d for $2\leqslant d\leqslant 5$.

Table I. The Quantities $\epsilon(\infty)-\epsilon(0)$ and $r(\infty)$, defined in Eqs. (4) and (5), for Several Values of Composition c and Vacancy Concentration v [Eqs. (2) and (3)][a]

c	v	$\varepsilon(\infty)-\varepsilon(0)$	$r(\infty)$
1/2	0	0.46 (0.46)	0.46 (0.46)
1/2	1/10	0.41 (0.42)	0.46 (0.47)
1/2	1/4	0.38 (0.37)	0.51 (0.50)
1/3	1/10	0.35 (0.38)	0.44 (0.45)
1/3	1/4	0.34 (0.33)	0.50 (0.48)
1/6	1/10	0.23 (0.20)	0.41 (0.36)
1/6	1/4	0.24 (0.22)	0.48 (0.44)
1/10	1/10	0.15 (0.14)	0.39 (0.36)
1/10	1/4	0.19 (0.17)	0.47 (0.44)

[a] The numbers in parentheses are the corresponding two-dimensional results, taken from Ref. 9.

The morphology of the frozen state of the system in three dimensions is very much the same as in two, and is characterized by a highly convoluted percolative structure near $c = 1/2$ and, at small c, very many small and fairly compact but not droplet like domains of the minority component embedded in a matrix of the majority phase. Vacancies have some tendency to concentrate near the boundaries between A and B domains. We do not include here any figures of the frozen structure, as cross sections are extremely similar to the two-dimensional structures depicted in Figs. 1 and 2 of Ref. 9. In three dimensions, however, domains that appear disconnected in cross-section view may actually be connected in the third dimension, often forming intricate three-dimensional structures.

The similarity between the two- and three-dimensional results is not limited to the $t \to \infty$ limit. We have obtained data for $\varepsilon(t)$ at time bins separated by $\Delta t = 10$ MCS. For all values of c and v studied here, we find that the resulting values of $\varepsilon(t)$ are very close to the corresponding two-dimensional results. Thus, any characteristic time one may define to characterize the approach of the system to its $t \to \infty$ value takes similar values in two and in three dimensions. If one chooses, as in Ref. 9, to define τ by the condition $\varepsilon(\tau) = 0.97\varepsilon(\infty)$, one finds that τ is in the range of 10–60 MCS, depending on c and v values.

3. CONCLUSIONS

We have presented in this paper detailed evidence that dimensionality is an irrelevant parameter for quenches to zero temperature in the spin-exchange kinetic Ising model. If, as seems reasonable, the growth law for quenches to nonzero temperature is predominately determined by the freeing up of the interface energy by means of activated processes that cannot operate at zero temperature, the implication is that the growth law in three and two dimensions will be the same. Our arguments cannot be considered a rigorous proof. However, given the high degree of difficulty encountered in numerically determining this law in two dimensions, the practical importance of universality is considerable.

More generally, we believe that the reason for the irrelevance of dimensionality in this problem is due to the fact that, as in the Cahn–Allen process, the growth is curvature-driven , as shown in Ref. 9. This argument does not explain, however, the detailed quantitative agreement in $\varepsilon(t)$.

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