

Kinetics of Domain Growth in Two Dimensions

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The growth of ordered domains following a quench of a system from a temperature T in the disordered phase to a state below the critical temperature T_c is simulated by Monte Carlo computations. For a square-lattice gas with (fourfold-degenerate) 2×1 order the domain size $L(t)$ grows with time t as $L(t) \propto t^x$, $x \cong 0.35$ (density conserved) and $x = \frac{1}{2}$ (density not conserved), respectively. Quenches to T_c and to $T=0$ are also discussed.

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A system which is quenched from a disordered state to a state below a transition temperature where an ordered phase appears first forms small domains separated by walls, since the order parameter always has some degeneracy p corresponding to the symmetry which is broken at the transition. As the time t after the quench increases, these domains coarsen and thus the (unfavorable) excess free energy due to the walls is reduced. In the late stages the domain sizes are much larger than all microscopic lengths, and by analogy with critical phenomena—where the order-parameter correlation length ξ is much larger than all microscopic lengths and power laws, scaling, and universality result¹—one also expects² the growth law for the domain size $L(t)$ to have a power-law form, $L(t) \propto t^x$, where x is some “universal” exponent, and the structure factor $S(k, t)$ studied in scattering experiments can be cast in scaled form,²⁻¹²

$$S(k, t) = [L(t)]^d \tilde{S}\{kL(t)\}, \quad (1)$$

d being the dimensionality, where the wave vector \vec{k} is measured relative to the Bragg positions of the ordered structure, and $\tilde{S}(z)$ is some scaling function. But in contrast to critical phenomena,¹ our understanding of the “universality classes,” i.e., classes of systems for which x and $\tilde{S}(z)$ are the same, is quite incomplete: Only for a nonconserved one-component order parameter $\{p=2\}$, all theories agree that $x = \frac{1}{2}$,^{4, 5, 7, 8, 12, 13} and this is also true if the density is conserved.¹³⁻¹⁶ While for conserved order parameter and $p=2$ there is increasing evidence^{3, 6, 10} for the Lifshitz-Slyozov¹⁷ result $x = \frac{1}{3}$, much less is known for larger p . Larger p 's do occur in ordered monolayers at surfaces, where $p=3$ for the $\sqrt{3} \times \sqrt{3}$ structure, $p=4$ for the 2×1 structure, etc.,¹⁷ and for ordered alloys on the fcc lattice (Cu-Au and Cu₃Au orderings, etc.),¹⁸ for instance. While it

was suggested that $L(t) \propto \ln t$ for $p > d+1$ (and thus $x=0$),¹⁹ a recent simulation of the p -state Potts model yielded²⁰ $x \cong \frac{1}{2}$ for $p \leq 6$ and $x \cong 0.38$ for large p .

In the present Letter we wish to contribute to this interesting problem of sorting out the universality classes of domain growth by showing that for order-disorder transitions with $p=4$ one obtains $x = \frac{1}{2}$ if the density is not conserved, which result agrees with Ref. 20, while for conserved density we obtain $x \cong 0.35$. This result contrasts with the case $p=2$, where the conservation law for the density did not make a difference. Our study is of potential interest to experiments studying O on W(110) surfaces, for instance, where a (2×1) structure with $p=4$ occurs²¹ and the coverage of the monolayer is held fixed.

We study a square lattice with repulsive interactions of equal strength between nearest- and next-nearest neighbors. Although this model is not directly applicable to any real system, in contrast to some more complicated lattice-gas models (e.g., in Kinzel, Selke, and Binder²²), the choice of this model is reasonable as its static properties²³ and its collective diffusion constant D ²⁴ have been studied in detail, and it is simple enough so that we are able to study $N \times N$ lattices with N as large as $N=800$ (with periodic boundary conditions). Thus we are able to ascertain that any finite-size effects, entering via the fact that decay factors $\exp[-Dk^2t]$ can involve a minimum wave number $k_{\min} = (2\pi)/N$ only, are completely irrelevant, and also percolation of one kind of domain [which we find to become important when $L(t) \approx N/3$] plays no role. As an initial state for the quench, we always choose a random configuration (i.e., infinite temperature). We monitor the decay of the excess energy $\Delta E(t) = E(t) - E_T$, where E_T is the internal energy of a monodomain

sample in thermal equilibrium at the temperature T chosen for the quench, and define an effective mean domain size $L(t)$ as

$$L(t) \equiv N[\psi_1^2(t) + \psi_2^2(t)]^{1/2} / \psi_T. \quad (2)$$

Here $\psi_1(t)$, $\psi_2(t)$ are the (instantaneous) order-parameter components of the (2×1) structure,²⁵ ψ_T being the equilibrium value of the order parameter. Of course, the square of the numerator of Eq. (2) is identical to the sum of the scattering intensities $S(0, t)$ at the superstructure Bragg positions. In order to average out fluctuations which are quite pronounced in individual runs, the quenching experiment typically was repeated 32 times for $N=200$ (and even more often for smaller N , such as $N=80, 120$). For comparison, quenches to $T > T_c$ were also performed, and it was established that $\Delta E(t)$ decreased exponentially fast as $t \rightarrow \infty$ for the nonconserved case, while $\Delta E(t) \propto t^{-1}$ for conserved density.²⁶ The density was chosen stoichiometric [$\rho = \frac{1}{2}$ for the (2×1) structure] in most cases, but also a few cases with $\rho < \frac{1}{2}$ were studied.²⁶

Figure 1 shows typical results for the nonconserved case at two temperatures. At the lower temperature (data for $T=0.75$ look similar), after a short transient period of about $t \approx 10$ MCS we find $L(t) \propto t^x$ with $x = \frac{1}{2}$, and also $\Delta E(t) \propto t^{-y}$ with $y = \frac{1}{2}$ (from scaling² one expects $x=y$). At $T=2.0$ and 2.1 , on the other hand, both exponents x, y are distinctly smaller (and no longer equal to each other). At these temperatures, $L(t)$ is not large in comparison to ξ , and hence the exponents x_c, y_c then reflect the dynamic critical behavior: One expects²⁷ $2x_c = \gamma/\nu z$, $y_c = (1-\alpha)/\nu z$, where α , γ , and ν are the static critical exponents of specific heat, susceptibility, and correlation length, and z is the dynamic exponent.¹¹ Using the estimates for the (nonuniversal) exponents²³ $\alpha \approx 0.3$, $\gamma \approx 1.5$ we would obtain $z \approx 2.1$ (from $x_c \approx 0.42$) and $z \approx 2.6$ (from $y_c \approx 0.32$). The discrepancy between these two numbers indicates that our data are still affected by correction terms to the leading asymptotic behavior seen at T_c , and an accurate estimate of z cannot yet be obtained. While the above exponent $x=y = \frac{1}{2}$ is the same for both the present model and the four-state Potts model, the critical behavior belongs to different universality classes. Estimates of z for the four-state Potts-model range from $z \approx 2.5$ to $z \approx 2.7$.²⁸ It would be interesting to study the critical behavior in two dimensions experimentally by performing quenching experiments of adsorbed layers at T_c .

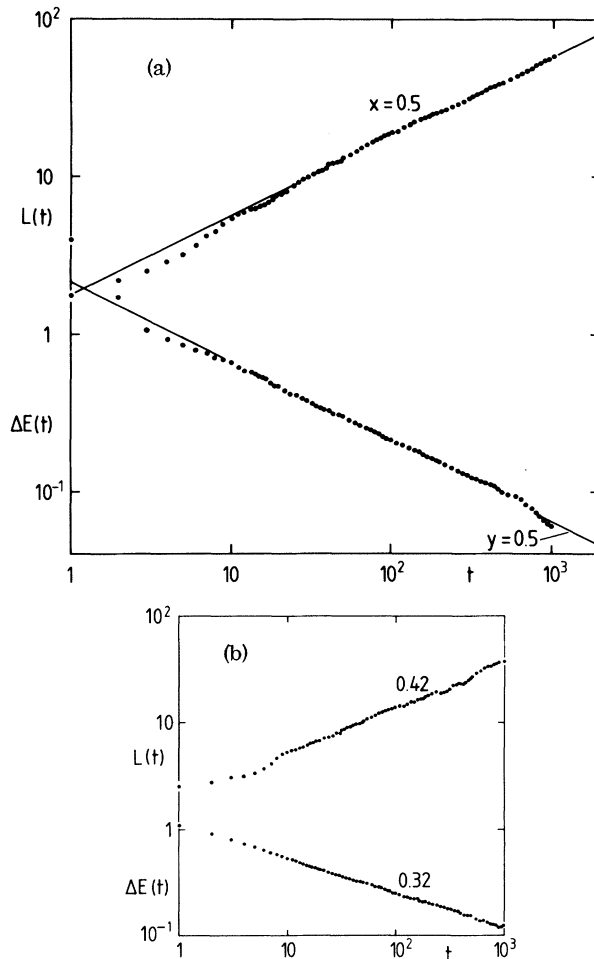


FIG. 1. Log-log plot of $E(t)$ and $L(t)$ vs time, for the temperatures (measured in units of nearest-neighbor exchange) (a) $T=1.33$ and (b) $T=2.0$, respectively ($T_c \approx 2.1$ at the chosen density $\rho = \frac{1}{2}$). Numbers at the curves are estimates of the exponent x . Time in the nonconserved case is measured in units of Monte Carlo steps (MCS) per site. Energies are measured in units of the nearest-neighbor exchange constant.

The case of conserved density was simulated by allowing the atoms to jump from a given site to a neighboring empty lattice site.^{24,26} There is clear evidence that for T distinctly less than T_c the exponents are $x=y = 0.35 \pm 0.05$, and hence are different from the nonconserved case (Fig. 2). Unfortunately, the slight curvature visible on the log-log plot does not allow us to give a more precise estimate of this exponent. At temperatures closer to T_c similar data are obtained; within our precision we cannot distinguish x_c, y_c from x, y in the conserved case. But still another crossover occurs at lower temperatures: Already at T

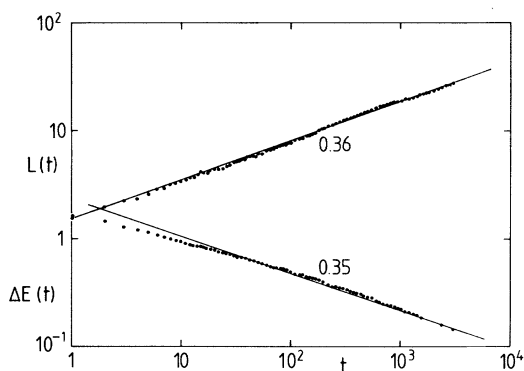


FIG. 2. Log-log plot of $E(t)$ and $L(t)$ vs time, for the temperature $T = 1.33$ and $\rho = \frac{1}{2}$. Time in this case with conservation law is measured in units of Monte Carlo steps (MCS) per particle. Points are due to 32 samples of a 120×120 system (sizes from 80×80 to 800×800 give identical results within the statistical error).

$= 0.75$ over several decades of time a rather slow domain growth (described by effective exponents $\chi_{\text{eff}} \approx \nu_{\text{eff}} \approx 0.2$) is observed [Fig. 3(a)], and at $T = 0$ the time evolution even seems to stop at a fi-

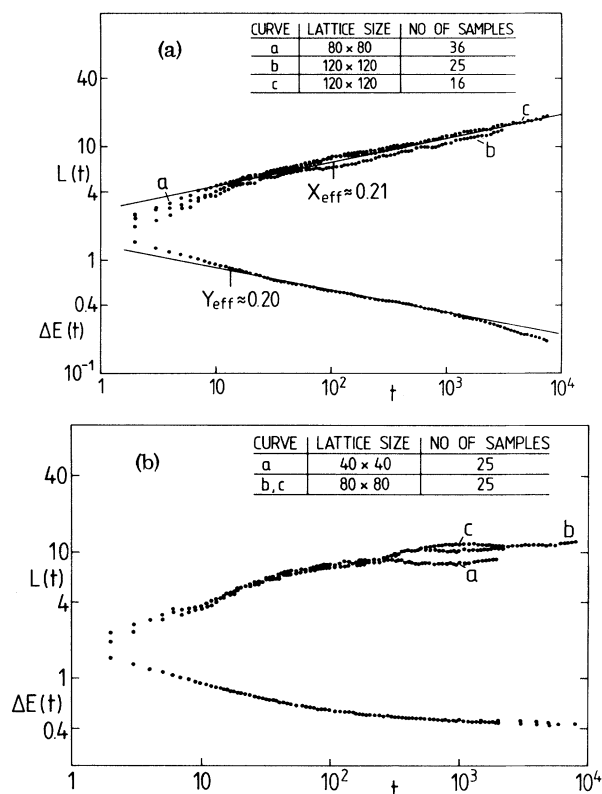


FIG. 3. Log-log plot of $\Delta E(t)$ and $L(t)$ vs time, for temperatures (a) $T = 0.75$ and (b) $T = 0.00$. Size and number of samples are indicated in the figure.

nite domain size, a metastable domain pattern being frozen in.

In conclusion, we have studied the kinetics of growth in a model exhibiting four-fold-degenerate (2×1) ordering. If there is no conservation law, domain sizes simply grow as $L(t) \propto t^{1/2}$ at all temperatures $T < T_c$ [but near T_c this is seen only for $L(t) \gg \xi$, while for $L(t) \leq \xi$ the domain growth can be related to the dynamic critical exponent z]. In the conserved case the exponent χ is distinctly less than $\frac{1}{2}$, and at low temperatures there occurs another crossover due to frozen-in domain patterns occurring in quenches to $T = 0$. Neither of these results agrees with the suggestion¹⁹ $L(t) \propto \ln t$; see also Ref. 16. Hence a theoretical understanding of our findings as yet is completely lacking. We hope that the present work will stimulate both further theoretical work and experimental studies of domain growth in adsorbed layers at surfaces.

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²⁵ $\psi_1 = \pm 1$ if completely filled rows alternate with empty rows oriented along the x direction, while $\psi_2 = \pm 1$ if the rows are oriented along the y direction. Change of sign corresponds to interchange of filled and empty sublattices. Thus $p = 4$ choices of the order result.

²⁶More details, particularly about quenches at $T > T_c$, will be given elsewhere. Of course, at $T > T_c$ the factor ψ_T^{-1} in Eq. (2) is omitted.

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