Impurity effects on domain-growth kinetics. I. Ising model

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The development of order for the Ising model in the presence of static, random impurities is studied following a quench from high temperature ($T \gg T_c$) to $T < T_c$. We find that for quenches to $T=0$, the system becomes pinned for long times for any value of $c > 0$ and never reaches its final equilibrium ferromagnetic ground state. The average linear pinned domain size scales as the inverse square root of the concentration c. For quenches to a final $T > 0$, the long-time behavior of the correlation length R and the energy E are slower than a power law, suggesting a logarithmic growth law for long times. The time that is required to reach this asymptotic logarithmic behavior increases as the impurity concentration decreases and/or the temperature increases.

I. INTRODUCTION

The kinetics of a nonconserved Ising model without impurities which has been quenched from high temperature, $T \gg T_c$, to a final temperature less than T_c has been well studied.[1] We know that the correlation length R grows algebraically as $t^{1/2}$ in both two and three dimensions, where t is the time. This result has been well documented by both analytical[2–7] and computer studies[5,8–10] as well as experimental studies on ordering alloys[3] (e.g., FeAl and Cu-Au). However, the effect of quenched impurities, or nonmagnetic ions, on the kinetics of domain growth have not received similar attention. In this paper, we present the first Monte Carlo simulations of domain growth for the nonconserved Ising model in the presence of impurities.

It is well known that diluting a magnetic system with nonmagnetic atoms reduces the critical temperature T_c. Monte Carlo estimate of the phase diagram have been made for both two-[11] and three-[12]dimensional lattices. In addition, we know by Harris's criterion[13] that if the specific-heat exponent $\alpha < 0$, the critical properties are unaffected by the impurities. However, if $\alpha > 0$, a new random fixed point becomes stable, leading to new critical exponents.

The effect of dilution on the kinetics of growth are less well understood. It is expected that kinetics should be slower in the presense of nonmagnetic atoms, since the dilution of the magnetic spins would reduce the driving force responsible for domain growth. Near the percolation threshold,[14] this effect should be quite large. Since the percolating clusters, just above the percolation threshold, are ramified,[14] then opposite spin orientation would meet two adjacent domains at only a few points as in Fig. 1, instead of along a line as in the nondilute case. This greatly reduces or eliminates the driving force for growth (see Fig. 1). We found that quenches from $T \gg T_c$ directly to $T=0$ always become pinned very quickly for all values of c studied. As the concentration of nonmagnetic impurities is reduced, the final domain area increases, but pinning still occurs at long times. For quenches to a final temperature T_F ($T_c > T_F > 0$), impurities continue to play an important role. We find that the asymptotic growth for long times is slower than a power law. This supports the recent results of Huse and Henley,[15] who suggested that in two dimensions the long-time growth would be $(\ln t)^4$. Though we are unable to distinguish a $\ln t$ from a power of $\ln t$, we do find that the growth becomes very slow and is not a power law in t.

The outline of the paper is as follows. In Sec. II we discuss the Monte Carlo procedure and the methods of analysis. In Sec. III we present our results for quenches to a final temperature $T_F=0$, while the $T_F > 0$ results are presented in Sec. IV. Finally, in Sec. V both our conclusions and comments on possible experimental verification of our results are presented. In the following paper we present our Monte Carlo results for the Q-state Potts model[16] for $Q > 2$ in the presence of impurities.[17]

FIG. 1. Example of a pinned domain configuration for $c=0.40$ in which most spins cannot flip for $T=0$. Solid and open circles correspond to up and down spins, and impurity sites are vacant.

II. PROCEDURE

Since we will address the evolution of the Q-state Potts model for $Q>2$ in the following paper,[17] we write the Hamiltonian for the Ising model in terms of the $Q=2$ Potts model,

$$H=-J\sum_{\mathrm{NN}}\delta_{S_iS_j}\ , \tag{1}$$

where $S_i=\pm 1$. The sum is over all nearest-neighbor (NN) pairs, the exchange constant $J>0$, and δ_{AB} is the Kronecker delta. To convert this Hamiltonian to the usual Ising convention we write

$$H_I=-(J/2)\sum_{\mathrm{NN}}S_iS_j-J/2\ . \tag{2}$$

Since the effective coupling is reduced by a factor of 2 in Eq. (2), the transition temperature is $T_c=1.82J/k_B$ for the nondilute triangular lattice instead of $3.64J$ for the usual Ising model. The conversion of the energy E presented in later figures to that for the usual Ising model convention is straightforward. In all simulations we start by randomly placing Nc immobile, nonmagnetic impurities ($S_i=0$) on the lattice, where N is the number of lattice sites and c is the impurity concentration. We then randomly assign a spin orientation, $S_i=\pm 1$, to the remaining sites and rapidly quench to $T<T_c$. All of our simulations are performed on a triangular lattice of size 200×200 or 500×500 with periodic boundary conditions. Most of our results are for a 500×500 lattice, in which we have averaged over at least ten runs for quenches to $T_F=0$ and at least two runs for $T_F>0$. We have chosen to make fewer runs on very large systems, instead of many runs on smaller systems, because the domains in the Ising model[18] quickly become very large for small values of c. We found that the energy E varied insignificantly (less than 0.1%) from run to run, suggesting that these runs provide more than adequate statistics. We found this also to be true for quenches to $T_F>0$, except for T_F near T_c where fluctuations were stronger. This will be discussed in more detail in Sec. IV.

Glauber spin dynamics were employed in these simulations. A trial spin, chosen at random, is flipped with probability

$$W=\begin{cases}\exp(-\Delta E/k_BT), & \Delta E\geq 0\\ 1, & \Delta E\leq 0\end{cases} \tag{3}$$

where ΔE is the change in energy resulting from the spin flip and k_B is the Boltzmann factor. We define a unit of time as 1 Monte Carlo step (MCS) per spin, corresponding to N microtrials or spin-flip attempts. While this standard MC technique was used for the early stages of the simulation, after a few hundred MCS's the probability of flipping a spin becomes very small. The computer time necessary to study the system for long times would then become prohibitive. In order to overcome this problem, we applied the continuous-time method (the "n-fold" way developed by Bortz, Kalos, and Lebowitz[19]). Though this method has not been widely used,[19,20] it turns out to be very important in the present simulations. Rather than choosing a spin at random and then carrying out a spin-flip attempt, in this method a spin is flipped at each trial and the time which elapsed since the previous flip is determined. The procedure we adopt is a slight generalization of the original procedure, since the presence of impurities gives rise to $n=13$ possible values of ΔE or classes of spin environments on the triangular lattice. With no impurities, this number is reduced to seven. The probability of flipping any spin in a class is identical and equal to

$$P_i=\begin{cases}N_i\exp(-\Delta E/k_BT), & \Delta E>0\\ N_i, & \Delta E\leq 0\end{cases} \tag{4}$$

where N_i is the number of spins in class i and the total probability is

$$P=\sum_{i=1}^{13}P_i\ . \tag{5}$$

A random number between zero and P is used to determine which class of spin shall be chosen and then a second random number is used to determine which spin in that class is flipped. The clock is advanced after each flip by

$$\Delta t=-(1/P)\ln R\ , \tag{6}$$

where R is a random number ($0<R<1$). Because of the extra record keeping necessary to apply this method, it is not efficient for the first few hundred steps after a quench from an initially random state. Standard MC techniques are employed in this early time regime. However, after this initial time, the n-fold way becomes more efficient since P [Eq. (5)] decreases with time.

In simulations for the Ising model, the question of an accurate, computationally efficient measure of the order always arises. A convenient method which we employ in our Q-state Potts model study is simply determination of the average cluster area A using a cluster-enumeration routine. This works very well for $Q>2$, since the domains are relatively compact and do not percolate for $d=2$. However, for the nonconserved Ising model, there are always two very large domains[18] (one of up spins and the other down) which span the sample. Thus measuring the average area (except possibly for very large values of c) is not a good measure of ordering for $Q=2$. A second measure of the average domain size is the inverse perimeter density,[10] $\overline{R}(t)$. $\overline{R}(t)=-E_0/[E(t)-E_0]$, where $E(t)$ is the average energy of the system at time t and E_0 is the energy of the equilibrium, ferromagnetic system for $T=T_F$. This function simply counts the number of broken bonds. In Secs. III and IV the time dependence of $E(t)$ will be employed as a measure of the growth of order. A third efficient measure of the average domain size is the mean chord length $\overline{L}(t)$. $\overline{L}(t)$ measures the linear one-dimensional distance between domain boundaries. $\overline{R}(t)$ and $\overline{L}(t)$ were found to have the same time dependence.

In addition to these three order parameters, two other measures of the domain size are possible, but both are computationally more time consuming. One is the second moment, $k_2(t)$, of the structure factor[1]

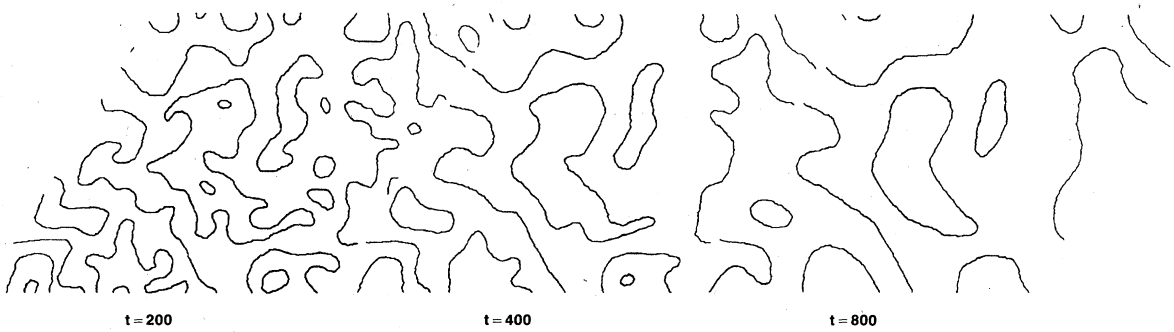

FIG. 2. Evolution of the domain boundary for the nonconserved Ising model quenched from $T \ll T_c$ to $T=0$ on a triangular lattice of size 500×500 with no impurities.

$$S(k,t)=\left\langle \left| \sum_{i,j} S_i S_j e^{i\mathbf{k}\cdot\mathbf{r}_{ij}} \right| \right\rangle , \tag{7}$$

where $\mathbf{r}_{ij}$ is the distance between spins and $\langle\ \rangle$ is the thermal average. Thus $k_2(t)$ is given by

$$k_2(t)=\frac{\sum_{0\le k\le k_{\max}} k^2 S(k,t)}{\sum_{0\le k\le k_{\max}} S(k,t)} . \tag{8}$$

The sum is over all allowed k vectors with magnitude between 0 and $k_{\max}$. The second[21] is

$$L(t)=[S(0,t)/N]^{1/2}/\psi(T) , \tag{9}$$

where $\psi(T)$ denotes the equilibrium value of the order parameter. Since $S(0,t)$ is simply the average of $|\sum_i S_i|^2$, $L(t)$ for one sample measures the amount by which the symmetry between the two orientations is broken for that particular sample. Mazenko and Valls[7] have pointed out that one must average over many samples to determine $L(t)$ correctly, as there are large fluctuations from sample to sample. Since they also found that the number of samples needed to obtain proper averages of $L(t)$ did not decrease significantly with increases in sample size, we did not measure $L(t)$. The quantity $k_2(t)$ turns out to be more suitable than $L(t)$ since it does not fluctuate significantly between different runs. This should not be surprising since $k_2(t)$ weights all k's and not just the $k=0$ component of the structure factor.

Thus, of the five possible measures of the order, we found that $\bar{R}(t)$, $\bar{L}(t)$, and $k_2^{-1}(t)$ were the most practical and meaningful, as they do not flucurate greatly between different runs for the large system sizes studied here. While keeping track of $\bar{R}(t)$ and $\bar{L}(t)$ took little computer time, the evaluation of $k_2(t)$ was quite slow, thus limiting how often it could be calculated.

III. $T_F=0$ KINETICS

In Fig. 2 we show the evolution of the spin configuration for the nonconserved Ising model quenched from $T=\infty$ to $T=0$, with no impurities present. We see that the growth is very rapid. The two largest spanning clusters quickly consume the smaller clusters. This should be constrasted with the evolution in the presence of static impurities, shown in Fig. 3 for $c=0.01$. Though the two largest domains still consume many of the smaller clusters, the growth becomes much slower, as seen by comparing the $t=400$ and 2000 MCS configurations in Fig. 3. In fact, for quenches directly to $T=0$ the growth essentially stops for long times. In Fig. 4 we show a 200×200 section of the pinned configurations for three values of the impurity concentration with the up-spin clusters shaded. As seen from Fig. 4, even for $c=0.10$, most of the spins are in one of the two spanning clusters and therefore the average area of a cluster, as discussed in Sec. II, is not a good measure of the order.

In Figs. 5(a) and 5(b) we show the change in energy $E(t)$ or, equivalently, $\bar{R}(t)=-E_0/[E(t)-E_0]$ as a function of time. Since $E(t)$ decays as $t^{-1/2}$ for $c=0$, we

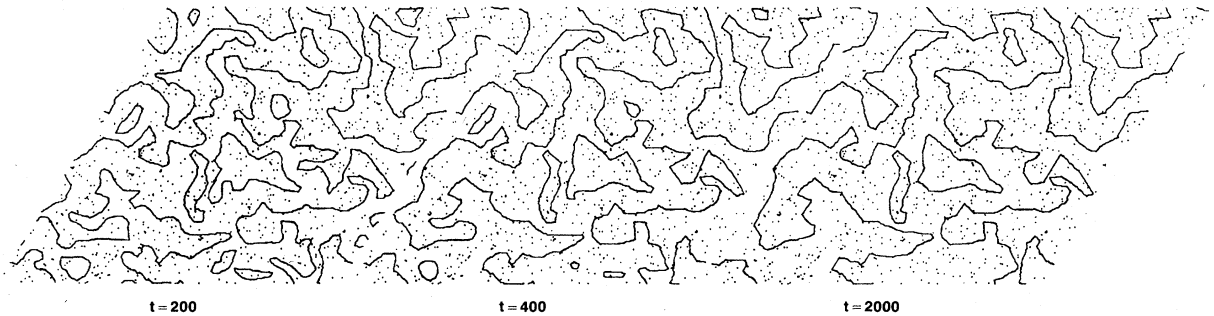

FIG. 3. Evolution of the domain boundary for the nonconserved Ising model quenched from $T \gg T_c$ to $T=0$ on a triangular lattice of size 500×500 with $c=0.005$.

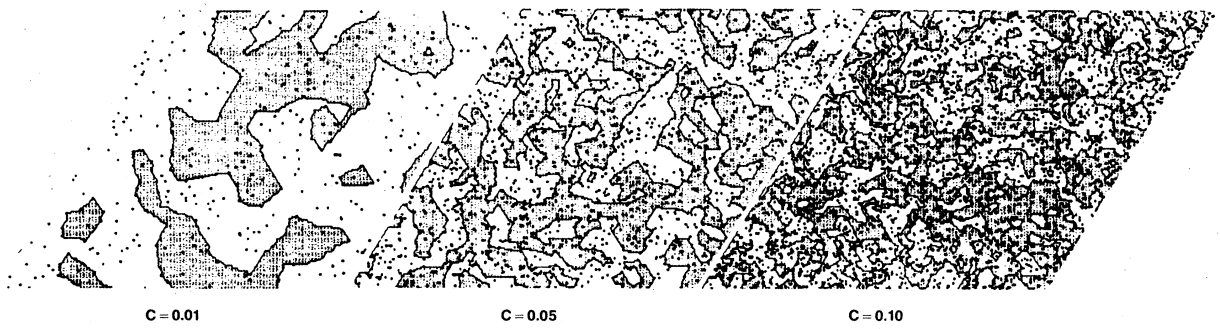

FIG. 4. Pinned microstructure for the Ising model quenched from $T \gg T_c$ to $T=0$ for three values of the impurity concentration c. A 200×200 subsection of each 500×500 lattice is shown. The up-spin domains have been shaded as a guide to the eye.

show $E(t)$ versus $t^{-1/2}$ in Fig. 5(a) for five values of c. For $c>0$ the systems pin quickly. In Fig. 5(b) we show the same data plotted in double-logarithmic form: $\ln(E-E_0)$ versus $\ln t$. Note that an attempt to fit t^{-n} for the early-time data would give values of n which are less than the known result $n=\frac{1}{2}$ for $c=0$. However, as c decreases and the crossover time for the system to become pinned increases, the early-time region approaches a $t^{-1/2}$ behavior. Thus it is likely that for $c \ll 1$, when the domain size is small compared to the spacing between impurities, the early-time regime can be well described by the power law $t^{-1/2}$. This is important experimentally, since all real samples will have some concentration of impurities. As we will see in the next section, this crossover time also increases with increasing T_F.

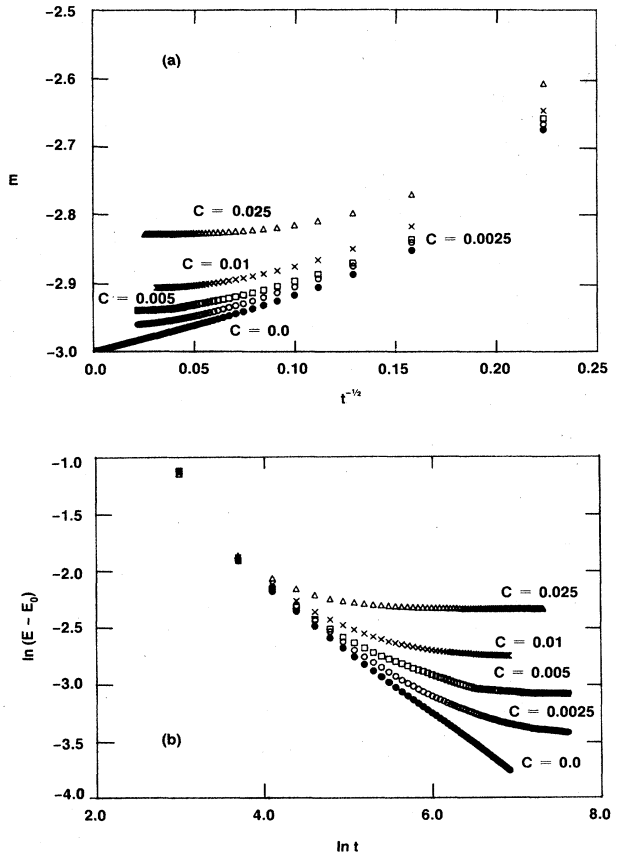

FIG. 5. (a) Energy vs $t^{-1/2}$ for the Ising model quenched from $T \gg T_c$ to $T=0$ for five values of the impurity concentration c. (b) Same data plotted on a log-log scale, where E_0 is the equilibrium, ferromagnetic ground-state energy. The data are averaged over 10 runs for each value of c on a 500×500 lattice.

In Fig. 6 the logarithm of R_f, the size of the pinned configurations after quenching directly to $T_F=0$, versus the inverse concentration c^{-1}, is plotted. The results are obtained for long times, after growth has become very slow or ceased. We used three measures of R_f. The first is the mean perimeter $\overline{R}(t)$, obtained from $E(t)$, and the second is the mean chord length $\overline{L}(t)$. These two quantities were measured using data from both the 200^2 and 500^2 lattices and averaged over at least 25 configurations for $c>0.01$ and 10 runs on the 500^2 lattice for $c \leq 0.01$, where R_f is too large to use the 200^2 lattices. The third measure that we used for R_f was $2\pi k_2^{-1/2}$, obtained from

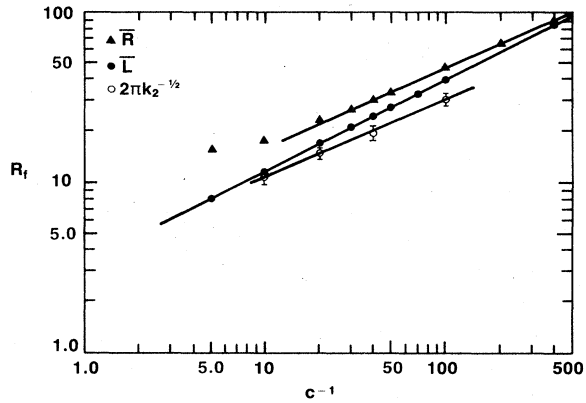

FIG. 6. Log-log plot of R_f vs the inverse concentration c^{-1}. Here R_f measures the size of the pinned configurations after quenches directly to $T_F=0$. The three measures of R_f used are the mean perimeter $\overline{R}(t)=-E_0/[E(t)-E_0]$, which counts the number of broken bonds, the mean chord length $\overline{L}(t)$, and the inverse square root of the second moment of the structure factor, $2\pi k_2^{-1/2}$.

averaging at least 10 runs on the 200^2 lattice.[22] Since the number of runs and lattice size were smaller, the uncertainty in $k_2^{-1/2}(t)$ is larger than for $\overline{R}$ and $\overline{L}$. From these results, we see that the three measures give slightly different results for the size of the domain, but all three show a power-law dependence on c. Over the range of concentration available, we find that $\overline{R} \sim c^{-a_1}$, $\overline{L} \sim c^{-a_2}$, and $k_2^{-1/2} \sim c^{-a_3}$, where $a_1 = 0.48 \pm 0.03$, $a_2 = 0.54 \pm 0.02$, and $a_3 = 0.45 \pm 0.05$. This difference in the value of these exponents indicates that c is not sufficiently small to obtain the true asymptotic behavior. However, from these results and our results for the $Q \geq 3$ Potts model,[17] we expect that this exponent should equal 0.5 as $c \to 0$.

Since the average domain size $\overline{R}(t)$ depends on $t^{1/2}$ for $c=0$ and scales approximately as $c^{-1/2}$ for $t \to \infty$ for $c > 0$, $R(t)$ should have the scaling form

$$R(t) = t^{1/2} f(ct) \ . \tag{10}$$

The scaling functions $f(x)$ must equal a constant for $x \to 0$ and $x^{-1/2}$ as $x \to \infty$. In Fig. 7 we replot the data for the four values of $c > 0$ from Fig. 5 in the scaling form $\ln[R(t)/t^{1/2}]$ versus $\ln(ct)$. Except for very small t, where one expects initial transients in the data, the results fall on one smooth curve. For small x $(=ct)$, $f(x)$ is only approximately constant, since even for $c=0.0025$, the smallest impurity concentration studied, there is no observable early-time $t^{1/2}$ region as seen from Fig. 5. Only for much smaller values of c would $f(x)$ become constant as $x \to 0$.

IV. $T_F > 0$ KINETICS

Since the quenches directly to $T_F = 0$ were pinned by the presence of impurities, it is important to determine if this behavior also occurs for $T_F > 0$. For $T_F > 0$ we expect that the system can never be truly pinned, as thermal fluctuations will eventually drive the system over the barriers that caused the pinning and, asymptotically, the true ferromagnetic ground state must be reached. The question to ask is, however, whether the growth law will be a power law or logarithmic in time. From our simulation results we will show that the long-time growth is probably logarithmic for all $c > 0$ and $T_F < T_c$.

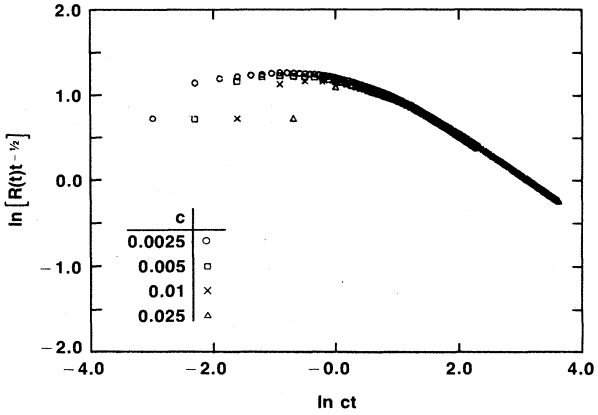

FIG. 7. Logarithm of $R(t)/t^{1/2}$ vs logarithm of ct for a quench from $T \gg T_c$ to $T=0$ for the four nonzero values of the impurity concentration c shown in Fig. 5.

In Fig. 8 we show the evolution of domain configuration for the nonconserved Ising model quenched from $T=\infty$ to $T=1.3J$ for $c=0.01$. This corresponds to a temperature of approximately $0.7T_c$. From studies of the time dependence of the energy for this value of T and c, the power law $E(t)-E_0 \sim t^{-1/2}$ is established. However, this is expected since c is small and T is large and the crossover to logarithmic growth occurs at very long times. We did observe this crossover at $c=0.01$ for very low T, but the crossover time quickly moved beyond the timescales for our simulations as T increased.

To study the dependence of the crossover from the power-law regime to the logarithmic regime over a wider temperature range, we looked in detail at two concentrations: $c=0.10$ and 0.40. For these cases, we followed the kinetics for up to 25 000 MCS's on a 500×500 lattice. In Fig. 9(a) we plot the average energy versus $t^{-1/2}$, for three values of T_F for $c=0.10$. In Fig. 9(b) we show the same data as well as data for two additional values of T_F plotted on a log-log plot. In Fig. 10 we show similar results

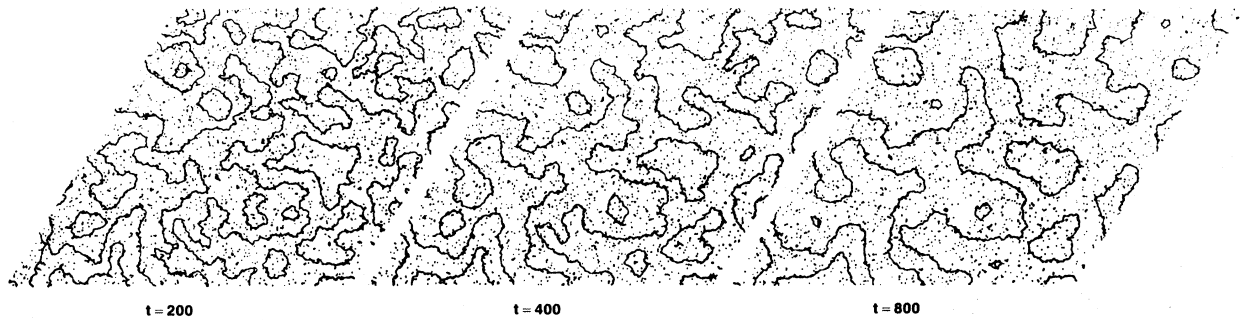

FIG. 8. Evolution of the domain boundary for the nonconserved Ising model quenched from $T \gg T_c$ to $T=1.3J/k_B$ on a triangular lattice of size 500×500 with $c=0.01$. Note that static impurities and reversed spins are not distinguished in plotting.

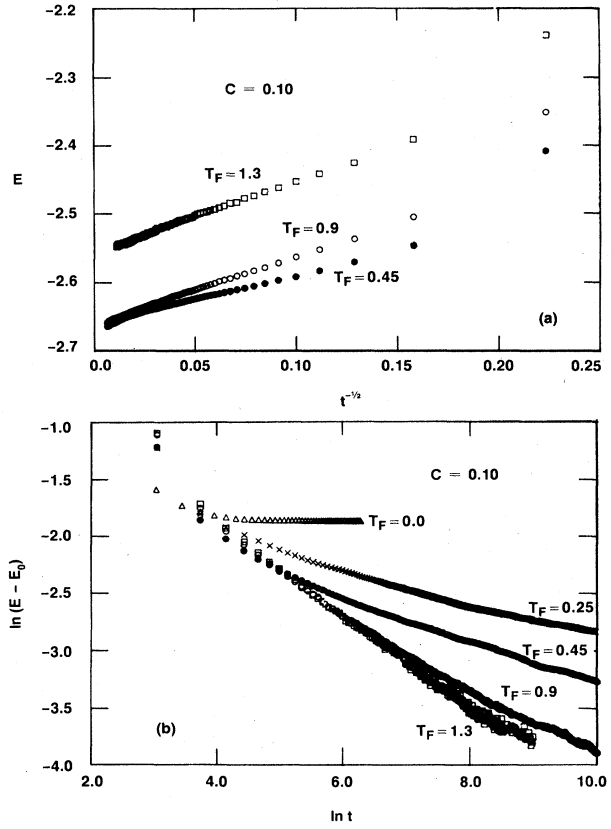

FIG. 9. (a) Energy vs $t^{-1/2}$ for the Ising model with $c=0.10$ quenched from $T=\infty$ to $T_F<T_c$ for three values of k_BT_F/J. (b) Same data as well as data for two lower values of T_F plotted on a log-log scale, where E_0 is the equilibrium energy for the long-range-ordered ferromagnet at $T=T_F$.

for $c=0.40$ on a log-log plot. T_c for these concentrations are roughly $1.55J$ and $0.9J$, respectively.

From Fig. 9(b) we see that for $c=0.10$ the crossover time (to a regime which is slower than power law) is clearly observable, only for $T_F=0$, while for the other temperatures it is not. However, if one attempts to fit a power law to the $T_F>0$ data, it does not fit the $T_F=0.25J$ data over the entire measured regime and gives only an adequate fit to the $T_F=0.45J$ data with a slope of approximately 0.3. Larger slopes are obtained for $T_F=0.9J$ and $1.3J$. Only the data for the highest T measured, $1.3J$, is consistent with a power law $\sim t^{-1/2}$ over the entire range measured. The results for $c=0.40$ are very different, in that for all temperatures studied, there is a clear crossover to a logarithmic growth regime. Collectively, we interpret these latter as suggesting, quite strongly, that the asymptotic growth law for any concentration c is logarithmic, but that the crossover time is a sensitive function of the impurity concentration and final quench temperature T_F. Depending on how large this crossover time is, a fit to the early-time data may or may not fit a power law of the form $t^{-1/2}$. Lower values of n indicate the strong influence on the growth law of the eventual crossover at late times.

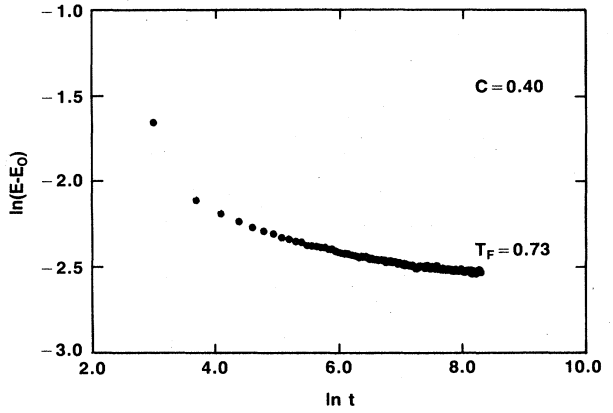

FIG. 10. $\ln(E-E_0)$ vs $\ln t$ for an Ising model with $c=0.40$ quenched from $T\gg T_c$ to $T_F=0.73J/k_B$, where $T_c\approx 0.9J/k_B$. Since $c=0.40$ is near the percolation threshold, only the largest spanning cluster was included in the simulations.

V. CONCLUSION

We have studied domain growth in the two-dimensional nonconserved Ising model with quenched impurities. For quenches from $T>T_c$ to $T_F=0$ the mean linear domain size $\bar{R}$ increases as $t^{1/2}$ at early times. However, for all impurity concentrations $c>0$ the spin configurations eventually become pinned and $\bar{R}$ tends to a constant. This constant is a function of impurity concentration and scales as $c^{-1/2}$. The time and concentration dependence of the mean linear domain size of Ising models with quenched impurities may collectively be described as $\bar{R}(t)=t^{1/2}f(ct)$, where $f(ct)$ is a scaling function plotted in Fig. 7. Results for quenches from $T>T_c$ to T_F ($T_c>T_F>0$) are similar to the results for quenches to $T_F=0$. The major difference is that instead of becoming pinned at long times, like the quenches to $T_F=0$, the spin configurations in the $T_F>0$ simulations continue to evolve, albeit at a slowing rate.[15] Analysis of these kinetics show that while the mean linear domain size continues to increase with time, the growth is slower than power law and is possibly logarithmic. The transition from the initial power-law kinetics ($\bar{R}\sim t^{1/2}$) to the "slower-than-power-law" kinetics occurs at longer times with increasing temperatures and decreasing impurity concentration.

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