Kinetics of first-order phase transitions: Monte Carlo simulations, renormalization-group methods, and scaling for critical quenches

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Using a combination of renormalization-group (RG) methods and Monte Carlo simulations, we study the growth kinetics of the spin-flip (SFKI) and the spin-exchange (SEKI) kinetic Ising models subjected to a critical quench (in zero external field) from infinite to low temperatures. The method developed here allows one to establish, in a nonperturbative fashion, the RG equations developed by us elsewhere. In the case of the SFKI model we find agreement, as expected, with the curvature-driven dynamics of Lifshitz, Cahn, and Allen which gives a growth law for a typical domain size of $L(t)\sim t^{1/2}$. Our results for the SEKI model (spinodal decomposition) are qualitatively different from the SFKI case. While both show scaling behavior for quenches to nonzero temperatures, the growth kinetics for the SE case show a long-time logarithmic growth $L(t)\sim \ln t$. For intermediate times one can fit an effective exponent $L(t)\sim t^{a(t)}$ where $a(t)$ agrees well with existing direct Monte Carlo studies. This logarithmic behavior is associated with the freezing of this system for quenches to zero temperature.

I. INTRODUCTION

Our understanding of the growth kinetics of systems subjected to strong external changes has increased[1-5] rapidly in recent years. In particular, considerable attention has been focused on the properties of binary alloys and mixtures subjected to a rapid temperature quench from an initial temperature T_I, well above an ordering temperature T_c, to a final temperature T_F below T_c. In many cases the behavior of these systems can be described in terms of Ising-like models which are convenient for Monte Carlo (MC) simulations. We show in this paper, using renormalization-group (RG) ideas implemented through Monte Carlo simulations, that the growth kinetics for critical quenches (through the critical point or in zero external field in the magnetic case) are qualitatively different depending on whether the order parameter is conserved or not. In both cases one does have scaling for the structure factor, but the time evolution of the associated scaling length $L(t)$ and the associated scaling behavior are qualitatively different.

When the order parameter is not conserved (NCOP), as in an order-disorder transition in a binary alloy, it is well known that the growth kinetics are governed by the curvature-driven dynamics developed by Lifshitz,[6] Cahn and Allen[7] (LCA) which gives $L(t)\sim t^{1/2}$ for dimensions greater than one. It is commonly asserted that in the case of a conserved order parameter (COP) (phase separation or spinodal decomposition in a binary alloy) the long-time growth kinetics is controlled by the evaporation-condensation mechanism of Lifshitz and Slyozov[8,9,4] which gives $L(t)\sim t^{1/3}$. While there is convincing evidence that this is the correct description for off-critical quenches near the coexistence curve, we demonstrate in this paper that the growth kinetics for critical quenches can be quite different from the Lifshitz-Slyozov theory. We find, in particular, that the long-time behavior is not a power law, but $L(t)\sim \ln t$. The origin of this effect is associated with a low-temperature freezing behavior found in the COP case but not in the NCOP case. For intermediate times, over a few decades, one can fit this logarithmic behavior to a power law in time with an effective index that is in agreement with available MC simulations. Before presenting our methods for determining this unexpected behavior we will discuss some of the background and previous work on this heavily studied problem.

It is becoming widely recognized that in the study of first-order transitions scaling concepts are likely to be as useful as they proved to be in the second-order case. The current understanding of the theoretical underpinnings of scaling in nonequilibrium problems has been the subject of several reviews,[1-3] while the experimental evidence for scaling has steadily increased, as reviewed in Ref. 10. There have been many investigations of temperature quenches in binary alloys which lead to sublattice ordering (NCOP) or phase separation (COP). Information about these problems can be obtained not only from experiment,[11-22] but also from MC simulations,[23-38] since they can be modeled by an Ising lattice gas with spin-exchange (SE) ferromagnetic Kawasaki[39] dynamics for spinodal decomposition and with either spin-flip (SF) Glauber[40] or spin-exchange antiferromagnetic dynamics[28,30] for the order-disorder transition. (These two alternatives in the NCOP case give similar, although not identical results.[41]) In this paper we study critical quenches (corresponding to 50% concentration) of an Ising-lattice-gas model on a square lattice for both SF and SE dynamics.

In the case of order-disorder transitions, the presence

and nature of scaling and that the associated growth laws for $L(t)$ (a typical domain size) are characterized by time-independent exponents has been clear for some time from experimental[11,20] and theoretical[6,7,42–47] points of view. We have expressed our point of view on this question in past work.[47,48,44,41] Indeed, we have included in this paper a discussion of the SF case in part because it is a very persuasive check of our method in a problem where the result is not, in our opinion, in dispute.

The situation is quite different for spinodal decomposition. In that case it has been the generally accepted assumption that the growth of order is characterized by time independent exponents. Thus, for example, the position, q_m, of the maximum in the quasistatic structure factor $C(\mathbf{q},t)$ is assumed to by given by $q_m \sim t^{-a}$, while the value $C_m = C(q_m,t)$ is $C_m \sim t^{a'}$. Scaling requires $a' = da$, where d is the dimensionality of the system. It is often asserted,[49] in the case where the concentration of one species is small, that $a = \frac{1}{3}$; the Lifshitz-Slyozov result.[8] The arguments leading to the Lifshitz-Slyozov theory depend on the presence of droplets and the assumption of local equilibrium at an interface. In the case of 50% concentration, extensive MC simulations for the spin-exchange kinetic Ising (SEKI) model have been carried out[38] to check the morphology of the system as time evolves.[50] The results of Ref. 38 clearly show that the domain structure is percolative, and characterized by fractal exponents, and not at all dropletlike. In Fig. 1 we show the results of simulating a quench to $T_F = 0$ from a purely random initial state at $t = 0$. Again, we see that the structures are percolative, not compact. This is due to the conservation law and the fact that the up-down symmetry of the system is not broken. Note also that the system appears to freeze for times greater than 50 Monte Carlo steps (MCS). A similar freezing behavior at zero temperature was found in Ref. 36 in a case with a NCOP and it was[36] found that this dramatically and unexpectedly affects the growth laws for quenchings to finite temperature. The freezing we find indicates that diffusion across an interface is an activated process and the assumption of local equilibrium in the Lifshitz-Slyozov model might never hold at low temperatures. The possibility exists that the SEKI model may not lead to the Lifshitz-Slyozov result even for small concentrations. We have, however, not yet investigated quenches away from 50% concentration, and we cannot, therefore, make a definite statement on this point.

The available evidence from experiment, theory or simulation on the growth law for spinodal decomposition in 50% binary alloys is inconclusive. In particular there is a lack of definitive evidence for any value of the exponent a (or even, we shall see, of its existence over an extended time range). This lack of evidence has been, in fact, one of the primary motivations for undertaking the present work.

In the context of analyzing MC simulations for the three-dimensional Ising model, it has been recently[31] pointed out that the value of a is "difficult to pin down." In fact, the authors of Ref. 31 go further: after quoting a value of $a = 0.24$ (apparently depending on the temperature) they explicitly caution the reader that "it is difficult

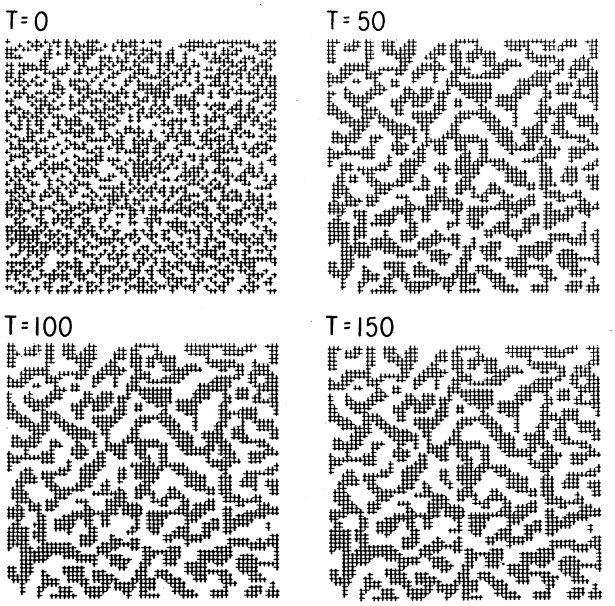

FIG. 1. Monte Carlo simulation for a quench from infinite to zero temperature for the SE model. The system consists of 64×64 spins and the times after the quench are given above the panel in MCS per spin. The crosses represent "up" spins and a blank represents "down" spins.

to extract from our simulations precise and reliable information about the analytic form of the behavior of $q_m(t)$ for late times" and go on to say that, as in real experiments, "almost any assumed form with some adjustable parameters can be made to fit the data." In other words over a time interval of, say, two or three decades, as for simulations and much less (as noted in Ref. 10) for experiment, one can always get a good fit to a fixed power law for $q_m(t)$ if that form is assumed. In MC simulations performed in two dimensions[23,25] the results stated[25] are $a = 0.2$ and $a' = 0.6$, which, since $a' \neq 2a$, would be in violation of scaling. However, in three dimensions the same group found[27] $a = 0.22$ and[24] $a = 0.21$—0.15; $a' = 0.65$—0.74 (it does not seem that the results show any definite trend with changing quenching temperature) which is consistent with scaling. Analysis of these results is hampered by very poor statistics, since the number of runs is in all cases extremely small. At longer times, where q_m is small, finite-size effects cannot be ruled out. We should add here that concentration dependence is quite obvious in all the MC results.

From the experimental point of view, an excellent compilation of results can be found in Ref. 10. The claim there is that none of the experiments really probed the asymptotic region. Widely differing exponents are quoted in the literature. In Ref. 51 the results $a = 0.2$, $a' = 0.7$ are given, for example, while a very extensive study of AlZn alloys performed in Ref. 17 gives an exponent close to $\frac{1}{3}$ at room temperature, but much smaller as the temperature is raised. These results of Ref. 51 are for small concentrations only. We think that the analysis of Ref. 10 and its conclusions are likely to apply.

We can only briefly discuss the theoretical status of the

problem here. A great deal of the work has been done from the point of view of time-dependent Ginzburg-Landau (TDGL) models.[52–57] In three dimensions the value $a=0.212$ was obtained in Ref. 57 using a truncation scheme for the set of coupled equations generated by the TDGL model. Other methods involving detailed studies of cluster dynamics[58–62] and the behavior of interfaces[63,64] have been proposed. In Refs. 59, 63, and 64, it is found that $a=\frac{1}{3}$ (in agreement with the Lifshitz-Slyozov theory), but $a=1/(d+2)$ at "earlier times," while in Refs. 58 and 62 one gets (from cluster dynamics) $a=0.16$, $a'=0.7$, at least at low concentrations and fairly early times (note that this last result violates scaling). Additional results can be obtained through the use of a Langevin-type equation: earlier[65,66] calculations yielded $a=(d+2)^{-1}$, but later results[67,68] seem to indicate a more complicated behavior, with an effective exponent depending on temperature, and on the time regime.

Our conclusion is that there is no hard evidence, from any of the above points of view, for any *single* power law. The fact that several exponents can be obtained in different "intermediate" and "late" time regimes points out the possibility of having a more complicated time dependence in this problem, which appears to be a power law when a relatively restricted time interval is considered.[10,31]

In order to clarify this issue we will use a combination of RG and MC methods. As discussed above, brute force MC simulations, for various reasons, have not been able to settle these questions. A primary benefit of RG methods is that they can be used to extract long-time and long-distance information from an analysis of quantities computed on modest time and distance scales. We have developed in this paper a procedure which uses a RG analysis of short-time MC simulations to extract this information, and to study the scaling behavior at long times. This is accomplished by using the MC simulations to verify the existence of a time-rescaling parameter Δ, which relates spatially rescaled correlation functions at different times. Scaling laws can be derived analytically from the existence of this parameter,[48,44] and long-time behavior follows, as we shall see, from its temperature dependence. Therefore, the question of the existence and properties of Δ is definitely worthy of study.

In our previous work[47,48,44,41,69] we developed a RG approach which made use of the time-rescaling factor Δ. We pointed out in Ref. 69 that although it is easy to determine Δ in the SF case further work remained to be done in the SE case. We show in this paper how this can be accomplished through MC simulations. In addition, the nonperturbative method we develop there to derive RG equations of the type used in Refs. 44, 41, 47, 48, and 69, clarifies the significance and validity of these equations, whose previous derivation has been characterized[70] as phenomenological.

This paper is organized as follows. In Sec. II we introduce the RG ideas, the MC procedure which enables us to implement them, and discuss the consequences of the existence of Δ and its implications for the long-time growth laws. In Sec. III the procedure is implemented for the case of SF dynamics (order-disorder transition). In this case we find scaling behavior as expected and the Δ we obtain leads to excellent agreement with the LCA growth law. In Sec. IV the same procedure is applied to spinodal decomposition (SE dynamics). We find that Δ still exists, but it has a nontrivial temperature dependence which leads to a weaker form of scaling compared to the SF case. We recapitulate our conclusions in Sec. V.

II. RENORMALIZATION-GROUP METHOD

In this section we develop the theoretical background which will then allow us to numerically verify the existence of the time-rescaling parameter Δ. This constitutes a nonperturbative approach to implementing the RG methods we developed elsewhere.[47,48,44,41]

Let us consider a system of spins $\sigma(\mathbf{n})$ defined on a d dimensional hypercubic lattice with N spins on a side. The positions of the spin are given by $\mathbf{n}=\sum_{i=1}^{d} n_i \hat{\mathbf{x}}_i$ where the n_i are integers ranging from 1 to N. All distances are measured in units of the lattice spacing. We assume that this system is subjected to a rapid temperature quench such that the time-dependent probability[71] governing the behavior of the spins $\{\sigma\}$ is given by $P(\sigma;t)$ and averages over the spin variable are given by

$$\langle A \rangle_t = \sum_{\sigma} P(\sigma;t) A(\sigma) . \tag{2.1}$$

We shall be interested in blocks of spins with $M(\leq N)$ spins on a side imbedded in the larger system. We can then define the "block" magnetization per spin

$$m_M = M^{-d} \sum_{\mathbf{n}\in \text{block}} \sigma(\mathbf{n}) . \tag{2.2}$$

The average of m_M is just the average magnetization per spin. We introduce m_M because we are interested in sampling the growth of spatial correlations by varying the block size M. Let us consider the quantity

$$R_M^N(t) = \langle m_M^2 \rangle_t - \langle m_M^2 \rangle_0 . \tag{2.3}$$

This quantity gives a good measure of the growth of order out to length scales comparable to M over the time interval 0 to t. We have defined[72] $R_M^N(t)$ such that it is zero initially. If we quench the system at that time from an initially disordered state, then $\langle m_M^2 \rangle_t$ will be a monotonically increasing function of t. This will be explicitly demonstrated in Secs. III and IV. We can easily see that if the initial state is completely disordered—as in an Ising system at infinite temperature and in zero external field—then $\langle m_M^2 \rangle_0 = M^{-d}$. If, however, the system eventually orders with some finite equilibrium magnetization per spin m_E, then

$$\langle m_M^2 \rangle_\infty = m_E^2 , \tag{2.4}$$

and for sufficiently large blocks we see that $R_M(t)$ is an increasing function of t ($m_M^2 \gg M^{-d}$). If one restricts the analysis to large blocks in an infinite system, then $\langle m_M^2 \rangle_0$ can be made arbitrarily small. We can gain some feeling for the quantity $R_M(t)$ by going over to the continuum limit and assuming M is sufficiently large so that we can ignore the subtraction term in (2.3). We have then

$$R_M(t)=M^{-2d}\int_0^M d^dr_1\int_0^M d^dr_2 C(\mathbf{r}_1-\mathbf{r}_2,t)\ . \quad (2.5)$$

Since the correlation function

$$C(\mathbf{r}_1-\mathbf{r}_2,t)=\langle\sigma(\mathbf{r}_1)\sigma(\mathbf{r}_2)\rangle_t$$

depends only on the distance between the spins at $\mathbf{r}_1$ and $\mathbf{r}_2$, we can do one set of the integrals in (2.5). Using $C(\mathbf{r},t)=C(-\mathbf{r},t)$, we obtain

$$R_M(t)=\left[\frac{2}{M^2}\right]^d\int_0^M d^dr\left[\prod_{i=1}^d(M-r_i)\right]C(\mathbf{r},t)\ . \quad (2.6)$$

There are two important limits we can check. For short times after the quench, $C(\mathbf{r},t)$ will be short ranged and the quantity $M^{-2d}\int d^dr r^d C(\mathbf{r},t)$ will be arbitrarily small for M sufficiently large. Therefore, for sufficiently short times and large M,

$$R_M(t)=\left[\frac{2}{M^2}\right]^d M^d\int_0^M d^dr C(\mathbf{r},t)=\frac{1}{M^d}\chi_M(t)\ , \quad (2.7)$$

where $\chi(t)=\chi_\infty(t)$ is just the time-dependent susceptibility or, equivalently, the $\mathbf{q}=0$ component of the quasistatic structure factor[48] $C(\mathbf{q},t)$. Thus for sufficiently short times (we shall pursue this more quantitatively in the next section) the M dependence of $R_M(t)\sim M^{-d}$. In the long-time limit, where $C(\mathbf{r},t)\sim m_E^2$, we find immediately that

$$R_M(t)=\left[\frac{2}{M^2}\right]^d m_E^2\left[\int_0^M dx(M-x)\right]^d=m_E^2 \quad (2.8)$$

as we suggested earlier.

We move now to the main point of this section. There are many problems of interest where there is an apparent self-similar behavior as order grows from length scale to length scale. As we indicated in earlier papers[48,69,44,41] this is reflected in a scaling equation of the form

$$\frac{C(\mathbf{r},\xi,t)}{m_E^2(\xi)}=\frac{C(\mathbf{r}',\xi',t')}{m_E^2(\xi')}\ , \quad (2.9)$$

where, on rescaling space by a factor $b(>1)$, $\mathbf{r}'=\mathbf{r}/b$, we must also rescale time, $t'=t'(t,b)$ and the correlation length[73] $\xi'=\xi/b$ characterizing the final equilibrium state. Note that we must divide $C(\mathbf{r},\xi,t)$ by m_E^2 if this relation is to hold for long times since $m_E^2(\xi)\neq m_E^2(\xi')$ except for quenches to $T=0$ or to above the transition temperature. Let us define

$$\nu^2=[m_E(\xi)/m_E(\xi')]^2\ . \quad (2.10)$$

Then, going back to Eq. (2.6) for $R_M(t)$ we obtain, in the scaling region,

$$R_M(t,\xi)=\nu^2 R_{M/b}(t',\xi')\ . \quad (2.11)$$

We see therefore that the quantity $R_M(t,\xi)$ serves as a good probe of scaling behavior *if such behavior exists.* We want, however, a method for investigating a system "growing order" which is capable of establishing the existence of scaling as well as quantifying its properties when it does exist.

Let us return to the discrete model which can be conveniently simulated using Monte Carlo methods. We can then *define* a quantity $t'(N,M,M',t,\xi)$ via

$$R_M^N(t,\xi)=\nu^2 R_{M'}^{N'}(t',\xi')\ , \quad (2.12)$$

where we assume $M>M'$ and $N'=NM'/M$. If $R_M^N(t,\xi)$ and $R_{M'}^{N'}(t',\xi')$ are monotonic functions of t, then we can solve (2.12) to obtain t'. We can then define

$$\Delta(N,M,M',t,\xi)=t'/t\ . \quad (2.13)$$

We obtain a scaling solution if, after sending $N\to\infty$, we find that Δ is only a function of $b=M/M'$, t, and ξ in the limit of large M and M' with b fixed. In that case we have the scaling relation (2.11). In many cases of interest, Δ will approach some constant value for long times

$$\lim_{t\to\infty}\Delta(b,t)=\Delta(b)\ . \quad (2.14)$$

Thus we have a well-defined unprejudiced numerical strategy for investigating possible scaling behavior in a wide variety of problems. The limitations are only those associated with making N, M, M', and t "large enough," and yet keeping the simulations within modest proportions. We discuss this in detail in the next two sections.

It is worth discussing here the scaling structure one obtains upon determining Δ. We will distinguish two important cases. In the first case, which we refer to a strong scaling, Δ is a nonzero constant essentially independent of temperature for low temperatures. In the second case of weak scaling, Δ is strongly temperature dependent for low temperatures, going to zero at zero temperature.

Let us take first the strong-scaling case, which is appropriate to the NCOP problem discussed in Sec. III. We have then, for low temperatures where $\xi=\xi'=0$ and $\nu=1$, the recursion relation

$$R_M(t)=R_{M/b}(\Delta t)\ , \quad (2.15)$$

which has a scaling solution,

$$R_M(t)=f(M/L(t))\ , \quad (2.16)$$

where $L(t)=bL(\Delta t)$. This in turn has a power-law solution:

$$L(t)=L_0 t^x;\ \ x=-\ln b/\ln\Delta\ . \quad (2.17)$$

We have the LCA growth-law behavior $x=\frac{1}{2}$, expected in the NCOP problem, if $\Delta=b^{-2}$.

We turn next to the slightly more involved weak-scaling case. When we look at spinodal decomposition in detail in Sec. IV we will find that Δ vanishes as $y=e^{-4K_F}$ (where $K_F=J/k_BT_F$ is the ratio of the exchange coupling J to the temperature) goes to zero. In this case we must include the temperature dependence in the analysis and use (2.11) which has a scaling solution,

$$R_M(t,\xi)=m_E^2 f(M/L(t,\xi),\xi)\ , \quad (2.18)$$

where

$$L(t,\xi)=bL(\Delta(\xi)t,\xi')\ . \quad (2.19)$$

If we find at low temperatures that

$$\Delta=\Delta_0 y^a=\Delta_0 e^{-2a/\xi}\ , \quad (2.20)$$

where a $[=a(b)]$ is a constant, then, using $\xi'=\xi/b$, we

find a solution to (2.19) of the form

$$L(t,\xi)=\xi l(t\xi^{\beta}/\Delta^{1/(b-1)}) , \tag{2.21}$$

where $\beta=\ln\Delta_0/\ln b$. We can determine the form of $l(x)$ by noting that $L(t,\xi)$ must be finite and nonzero in the zero-temperature limit. This means that $\lim_{\xi\to 0} l(t\xi^{\beta}/\Delta^{1/(b-1)})\sim 1/\xi$. Using (2.20) we see that we must choose $l(x)=L_0\ln x$ and the long-time growth goes as

$$L(t,\xi)=\xi L_0\ln(t\xi^{\beta}/\Delta^{1/(b-1)}) \tag{2.22}$$

for moderately low temperatures. Thus we obtain a logarithmic growth law at long times. Our numerical solution[74] of our recursion relations for the structure factor also show this logarithmic behavior. As indicated above and in Sec. IV this behavior is strongly tied to the fact that the SE system freezes at some finite time after a quench to zero temperature. In this case we find that Δ vanishes for long times [in Sec. IV we find that $\Delta=\Delta_0/(1+t/t_0)$] and there is no scaling behavior (growth develops out to some finite-length scale and stops).

It should be clear from our discussion above that our introduction of $R_M(t)$ is motivated by the convenience with which we can compute this quantity using Monte Carlo methods. The sampling of m_M is computationally fast, the ranges of M and M' needed are fairly narrow and the sizes N needed are manageable.

One point worth stressing is that our approach differs from finite-size scaling in that our finite system of size M^d is, in principle, embedded in an infinite ($N\gg M$) system. Thus we are justified, for example, in treating R_M as a function of the true correlation length ξ corresponding to an infinite system. In practice for systems where the order parameter is *not* conserved the distinctions between the two methods will not be important if various limits are taken appropriately. Thus, if we consider $R_N^N(t)$, we obtain, using (2.9),

$$\lim_{N\to\infty} N^d R_N^N(t)=\chi(t) , \tag{2.23}$$

and one has the scaling relation

$$\chi(t,\xi)=\lim_{N\to\infty} N^d v^2 R_{N/b}^{N/b}(t',\xi')=v^2 b^d \chi(\xi',t') , \tag{2.24}$$

in agreement with our previous renormalization-group work [Eq. (3.11) in Ref. 48 where we used the notation $\chi(t)=C_M(t)$]. This equation can be used to determine the time-rescaling factor Δ since the ansatz

$$\chi(t)\sim t^{y} \tag{2.25}$$

implies for $T_F=0$ that

$$\Delta=b^{-(d/y)} , \tag{2.26}$$

and y can be determined accurately using Monte Carlo methods. This determination of Δ is a weaker statement than obtaining it from (2.11) since it does not explicitly involve the rescaling of spatial coordinates. On the other hand, once one can establish (2.11), then (2.25) and (2.26) are useful ways of determining Δ explicitly.

Our method becomes particularly useful in the case where the order parameter is conserved. The point in that case is that R_N^N is time independent since m_N is conserved. Therefore the interesting range dynamically is not on length scales $M\sim N$ but for M considerably smaller than N. Thus our block-spin method is really necessary in the case of a conserved order parameter.

III. SINGLE SPIN-FLIP KINETIC ISING MODEL

As a first example of the use of the approach developed in the last section we consider the single spin-flip kinetic Ising (SFKI) model defined on a square lattice. This model has been treated previously by a number of authors and we think we understand the growth of order in some detail. In particular it is widely believed that there is scaling behavior, and the growth kinetics are characterized by the LCA law, $L(t)\sim t^{1/2}$. We wish to show how we can establish the more general scaling law given by (2.11). As we have indicated elsewhere[44,41] the growth kinetics of this system do not fundamentally depend on T_F for quenches below T_c. We shall, therefore, focus on the case where we quench from infinite to zero temperature. We will investigate the temperature dependence in the COP case (next section) where its effects are very important.

Let us specify our dynamics in more detail. In previous work we studied the "minimal" coupling[47,48,44] SFKI model and the antiferromagnetic spin-exchange model[41] as examples of growth of a NCOP. Both appeared to cross into the scaling region rather quickly as a function of time after the quench. In this paper we investigate the "Glauber" SFKI model which differs from that studied in Ref. 44 in the choice of the flipping probability. In the Glauber model the probability of flipping the spin at site $\mathbf{n}$ is given by

$$W_{\mathbf{n}}[\sigma]=\tfrac{1}{2}(1-\sigma_{\mathbf{n}}\tanh E_{\mathbf{n}}[\sigma]) \tag{3.1}$$

with $E_{\mathbf{n}}[\sigma]=K\sum_{\mathbf{m}}^{\mathbf{n}}\sigma(\mathbf{m})$ where the sum is over the four nearest neighbors of the spin at site $\mathbf{n}$, and K is the coupling defined in Sec. II.

We have computed the $R_M(t)$ over the ranges $M=4$ to 20 and $t=0$ to 5 MCS per spin[75] in steps of 0.25. We have chosen N such that $N\geq 2M$ in all of the runs and we have averaged over 700 runs in the analysis. In Table I we present the data for R_M. One sees that the data is smooth and monotonically increasing with time. Therefore $R_M(t)$ does intersect $R_{M'}(t')$, for $M>M'$, at a time $t>t'$ as discussed in the last section. We can then extract $t'(M,M',t)$ as defined in Sec. II.

The most direct method of determining $\Delta(M,M',t)=t'/t$ is to simply pick a pair, M,M' and numerically solve

$$R_M(t)=R_{M'}(t') \tag{3.2}$$

to obtain t' for $M>M'$. In Fig. 2 we have plotted the Δ one obtains from the pair $M=10$ and $M'=9$. One can see that Δ is well approximated by a constant, independent of time. This is true for all the nearest pairs $M=M'+1$. For the case $b=2$, $M=2M'$, one finds that there are some weak initial time-dependent transients for times less than 1, but for $t>1$, Δ is again very nearly constant in time. This direct approach is, however, a bit

TABLE I. Values of $R_M(t)\times 10^5$ obtained from Monte Carlo simulations for quenches from infinite to zero temperatures. The times after the quench are measured in units of MCS per spin. These data represent averages of 700 independent runs where $N > 2M$.

t \ M	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20
0.25	3394	2357	1654	1287	1010	812	640	554	501	413	331	334	294	234	221	206	181
0.50	6632	4534	3251	2507	1985	1628	1310	1171	1037	876	714	674	599	493	451	406	360
0.75	9637	6768	4946	3797	3021	2507	1987	1759	1565	1311	1080	911	882	749	689	633	562
1.00	12571	9019	6612	5080	4035	3374	2707	2407	2133	1779	1478	1359	1204	1022	934	863	770
1.25	15218	11100	8193	6342	5043	4247	3438	3036	2703	2248	1848	1726	1532	1301	1180	1086	964
1.50	17850	13129	9747	7593	6047	5088	4184	3657	3240	2725	2225	2076	1843	1583	1436	1309	1156
1.75	20321	15054	11233	8811	7082	5928	4879	4254	3783	3170	2618	2469	2197	1873	1692	1553	1376
2.00	22679	16997	12740	10001	8060	6724	5564	4807	4275	3616	3046	2880	2570	2159	1946	1797	1606
2.25	24755	18649	14121	11138	8980	7540	6230	5401	4799	4056	3467	3268	2913	2434	2196	2023	1818
2.50	26976	20309	15464	12174	9838	8318	6884	5983	5312	4526	3873	3640	3248	2726	2461	2263	2032
2.75	28744	21761	16716	13202	10673	9026	7517	6574	5848	5017	4277	3993	3571	2992	2698	2462	2200
3.00	30381	23149	18001	14243	11529	9753	8114	7102	6353	5465	4674	4305	3854	3267	2946	2676	2393
3.25	32133	24600	19346	15345	12446	10581	8847	7682	6862	5907	5030	4614	4133	3544	3185	2891	2577
3.50	33512	25997	20545	16283	13238	11337	9483	8194	7326	6331	5412	4959	4434	3806	3415	3106	2775
3.75	34904	27113	21638	17315	14092	12040	10082	8732	7812	6752	5757	5294	4736	4059	3640	3303	2944
4.00	36230	28276	22682	18137	14768	12647	10630	9198	8225	7119	6083	5630	5043	4334	3884	3511	3136
4.25	37413	29351	23676	18933	15437	13270	11210	9690	8642	7496	6455	5964	5337	4594	4118	3710	3306
4.50	38498	30376	24656	19706	16083	13785	11698	10244	9119	7901	6799	6278	5620	4835	4320	3888	3465
4.75	39542	31396	25676	20550	16799	14404	12280	10704	9526	8283	7144	6552	5859	5068	4525	4080	3638
5.00	40624	32491	26715	21339	17472	14912	12730	11122	9896	8626	7473	6857	6129	5320	4753	4276	3827

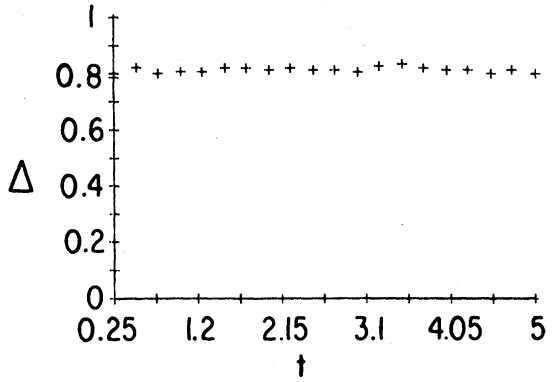

FIG. 2. Δ extracted from a numerical solution of (3.2), using the data in Table I for the case $M=10$ and $M'=9$, is plotted versus time measured in MCS per spin.

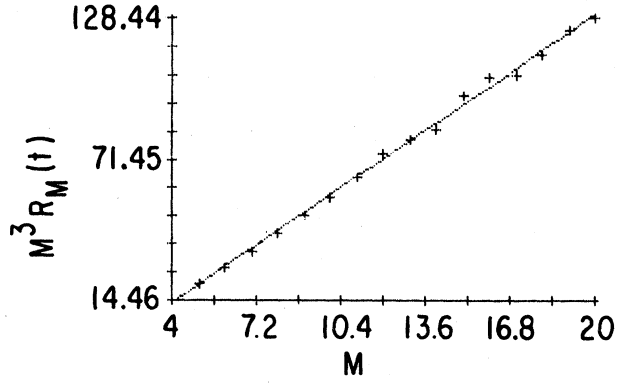

FIG. 3. Linear least-squares fit of the data (from Table I) for $M^3R_M(t)$ versus M (crosses) for $t=2$ MCS to a straight line.

cumbersome for determining the M,M' dependence of Δ since it does not make the best use of all the data. A more efficient approach for this purpose is to take advantage of our knowledge of the M dependence of $R_M(t)$ for relatively early times. If we allow for the first correction to (2.7), we can write for sufficiently short times that

$$R_M(t)=\frac{\chi(t)}{M^2}[1+c(t)/M+0(M^{-2})] . \tag{3.3}$$

The quantity $\chi(t)$ can be identified with the characteristic domain size $L(t)$ via

$$\chi(t)=L^2(t) . \tag{3.4}$$

This expansion should be valid over the region where $L(t)/M$ is small. We can determine $L(t)$ and $c(t)$ via a linear least-squares fit of $M^3R_M(t)$ to $\chi(t)(M+c(t))$. We show in Fig. 3 the fit[76] to the data for $t=2$. The quality of the fits is approximately the same for $t\geq 1$ (the coefficient of determination ≥ 0.997) but is slightly worse for shorter times. In Table II we give the values of $\chi(t)$ and $c(t)$. A plot of $\chi(t)$ shows that for times less than about 1.75 it grows with an exponent slightly greater than 1 [a fit to the data over the region 0 to 2 gives $\chi(t)\sim t^y$ with $y=1.07$]. A fit over the entire time region 0 to 5 gives $y=1.065$. As seen in Fig. 4, this does not fit the longest-time data as well as the shorter-time data. In order to properly fit the data from $t=4$ to 5, one must drop the data from 0 to 1.75. One then obtains the excellent fit given in Fig. 5 and the results

$$\chi(t)=3.70t^y , \tag{3.5}$$

$$y=0.999 , \tag{3.6}$$

with a coefficient of determination 0.9995. The quantity c is negative as expected and less than M for all M's treated. We see that $L(t=5)=4.28$ and $R_4(5)=0.406$ is showing strong saturation effects.

The behavior found here at short times differs from what we found for the minimal coupling operator in Ref. 44 where we found a very rapid crossover to the LCA growth law. The reason for this difference can be traced back to the very early time behavior for the two problems. One can show, exactly, that in the Glauber model studied here

TABLE II. Results of a linear least-squares fit of $M^3R_M(t)$ versus M to the form $\chi(M+c)$ for $M=4$ to 20 for each time listed in Table I.

t	χ	c
0.25	0.781 85	−1.3805
0.50	1.598 65	−1.4225
0.75	2.440 95	−1.5629
1.00	3.386 89	−1.7076
1.25	4.295 22	−1.7721
1.50	5.205 38	−1.8319
1.75	6.225 59	−1.9977
2.00	7.285 07	−2.1643
2.25	8.283 38	−2.2594
2.50	9.325 46	−2.3748
2.75	10.217 42	−2.3915
3.00	11.155 35	−2.4533
3.25	12.041 59	−2.4598
3.50	12.987 91	−2.5216
3.75	13.857 75	−2.5465
4.00	14.831 63	−2.6422
4.25	15.745 28	−2.7000
4.50	16.582 71	−2.7315
4.75	17.410 79	−2.7619
5.00	18.332 57	−2.8324

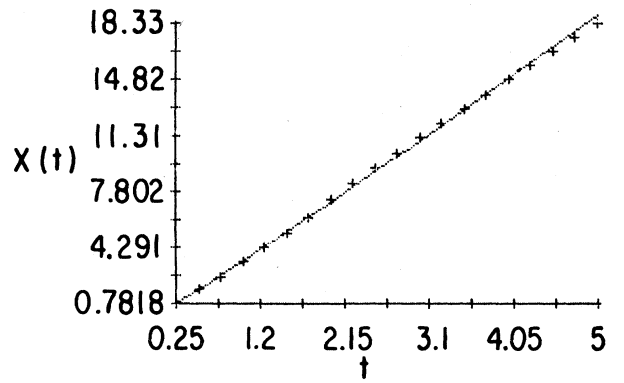

FIG. 4. Linear least-squares fit of the data (from Table II) for $\chi(t)$ versus t (crosses) to a power law over the time region $t=0.25$ to 5 MCS.

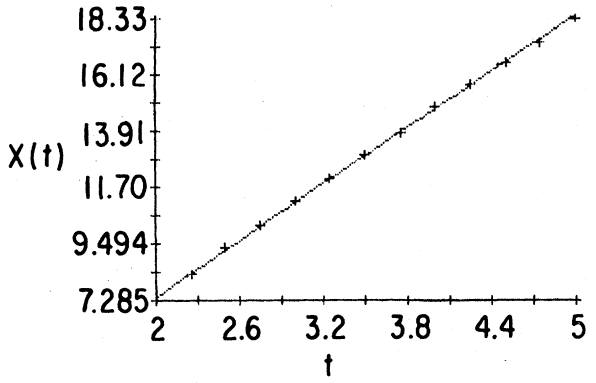

FIG. 5. Same as Fig. 4 except the fit is over the region $t=2$ to 5 MCS.

$$\chi_G(t)=\tfrac{3}{2}t+\tfrac{3}{32}t^2+O(t^3)\ , \tag{3.7}$$

while for the minimal coupling operator,

$$\chi_{\rm mc}(t)=4t+O(t)^3\ , \tag{3.8}$$

and the quadratic term is missing for quenches to zero temperature. In the Glauber case the quadratic term is present *and* positive (it is negative in the minimal coupling case for $T_F>0$). Thus one must wait for this initial "transient" to die down in the Glauber case.

Once we have determined $\chi(t)$ and $c(t)$, it is straightforward to determine Δ. We insert (3.3) for $R_M(t)$ into (3.2) and obtain

$$\frac{\chi(t)}{M^2}\left[1+\frac{c(t)}{M}+\cdots\right]=\frac{\chi(t')}{(M')^2}\left[1+\frac{c(t')}{M'}+\cdots\right]. \tag{3.9}$$

Using our excellent fit (3.5) for $\chi(t)$, we obtain

$$\Delta^y=\left[\frac{M'}{M}\right]^2\left[\frac{1+c(t)/M}{1+c(t')/M'}\right], \tag{3.10}$$

and in the appropriate limit (M,M' large $b=M/M'$ fixed),

$$\Delta=b^{-2/y}\ , \tag{3.11}$$

and with $y=0.999$ we recover the results of Refs. 44 and a good approximation to the LCA growth law.

IV. SPIN-EXCHANGE DYNAMICS

In this section we present the results obtained for the case of spin-exchange or Kawasaki dynamics. We quench the system as in Sec. III from a completely disordered state (infinite temperature) to a final temperature $T_F<T_c$. We assume only nearest-neighbor interactions are present and we choose[39] the spin-exchange probability between two neighboring sites **m** and **n** to be the form

$$W_{\rm m,n}(\sigma)=\frac{1}{2}\left[1-\tanh\frac{\Delta E}{2}\right], \tag{4.1}$$

where ΔE is the change in the energy of the system due to exchange of the two spins.

The MC simulation is carried out in a manner quite similar, in principle, to that performed in the SF case (Sec. III), but, as we shall see, there are important differences in practice. The main reason for this is that the growth of order is now much slower than in the NCOP case, which forces us to consider a much larger time scale. We define one MCS in this case as being one attempt to exchange any pair of spins.[77]

To extract the time-rescaling factor Δ we have to follow the evolution of the system for times sufficiently long so that the behavior of $R_M(t)$ contains information about the growth of spatial correlations in the scaling regime, that is, over distances appreciably longer than the lattice spacing. At short times, only the nearest-neighbor correlation function differs significantly from zero. Of course, one does not want to proceed to extremely long times, as this would vitiate the convenience of the method. Fortunately this turns out to be unnecessary, and even counterproductive in some cases where the long time growth of $R_M(t)$ becomes contaminated by finite size effects. We have found that it is convenient and sufficient to consider the time interval $10\le t\le 10^3$ (in MCS as defined above). While this is a much longer time interval than that considered in Sec. III, it is still quite short when compared to that of typical MC simulations.

We must choose the system size N and block size M so as to balance convergence and practicality. We have found in this case that (for the block sizes discussed below) it is sufficient to use $N=20$ or 25; both yield the same results for Δ within statistical error. Obviously, the values of M considered must be sufficiently small when compared to N, since if $M=N$, $R_N^N(t)=0$ and for values of M close to N, $R_M^N(t)$ is very sensitive to changes in N. On the other hand, they should be sufficiently large so that $R_M(t)$ contains information (in the time range considered) about the scaling region. To determine which values of M to use is not a completely trivial matter. We have, at zero temperature, computed $R_M(t)$ for $4\le M\le 16$ averaging over a sufficient number of runs (forty) to obtain convergence. The parameter Δ can be obtained from this data. Throughout this section we will use for this purpose the direct approach of solving (2.12) for $\Delta=t'/t$. As we shall see below, it turns out that Δ depends on time (and on temperature as well) for spin-exchange dynamics. We have obtained Δ at zero temperature by analyzing all pairs of $R_M(t)$ and $R_{M'}(t)$ with $5\le M\le 12$ and $4\le M'<M$ and verified that the resulting function $\Delta(t,T=0,b)$ depends on b but not on the value of M (and correspondingly M') chosen.

As the final quenching temperature is increased from zero, statistical fluctuations become larger, and the number of runs required correspondingly increases. Finite-size effects also seem to become more bothersome. We shall, therefore, concentrate on the lower temperature range. We have obtained data for $y=0$, 0.01, 0.03, 0.04, 0.05, and 0.08 ($y=e^{-4K_F}$). Even at $y=0.05$, 200 runs were required to obtain satisfactory results. Since $y_c=0.172$, one should be wary of extrapolating our results to quenches within the critical region. We present here results for

$y \leq 0.05$. Most of the simulations performed at finite temperature, covering the indicated range of y and t with a sufficient number of runs were carried out with $M=11$ and 9 and with a spatial-rescaling factor[78] $b=2$, that is, $M'=6$ and 5, respectively. In all cases both M,M' pairs yield the same result for $\Delta(t,y)$, indicating that it is a function of the ratio b only. This is despite the fact that $R_M(t)$ is, at constant time, a strongly varying function of M: $R_{11}(t)$ typically differs from $R_9(t)$ by nearly a factor of 2. These are the results that we discuss in detail below. In addition, over more restricted time and temperature ranges we have checked that the same $b=2$ results are obtained from $M=13$ and 7 (and corresponding M' values). We have also verified that for M,M' pairs corresponding to $b \approx 1.2$, Δ does not depend on M either. We have not carried out for this case the detailed analysis of the b dependence performed in Sec. III, instead we have concentrated on its temperature dependence, which is of crucial importance in this case.

Some of our data for $R_M(t)$ is displayed in Figs. 6 and 7. In Fig. 6 we plot $R_M(t)$ versus t at $y=0$ for several values of M. We can see that for $t>200$ MCS all the $R_M(t)$ are constant: the system is "frozen" within the distances considered. We have also verified in a separate MC simulation that the nearest-neighbor correlation function also freezes at a value considerably below its equilibrium value of unity.

This freezing behavior at zero temperature has important consequences for the scaling behavior of the system: it is obvious that the freezing of $R_M(t)$ implies that $\Delta \sim t^{-1}$ at long times. We have, therefore,

$$\lim_{t\to\infty} \Delta(t,y=0)=0 \ . \tag{4.2}$$

The discussion in Sec. II implies, then, that for quenches to zero temperatures the position $q_m(t)$ of the peak in the quasistatic structure factor $[\sim L^{-1}(t)]$ will tend toward a finite value:

$$\lim_{t\to\infty} q_m(t,y=0)=q_0 \neq 0 \ . \tag{4.3}$$

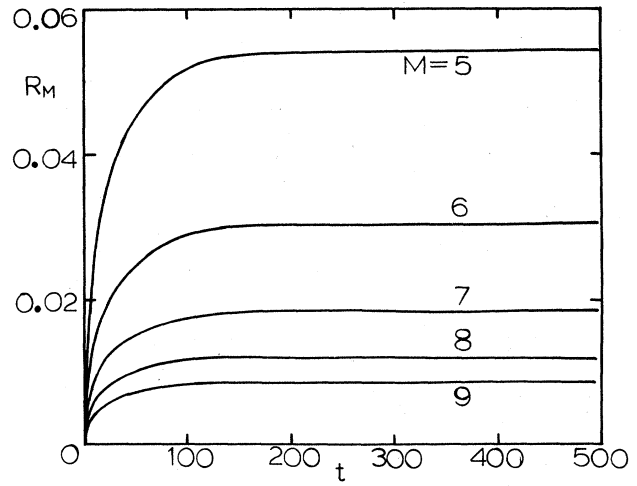

FIG. 6. $R_M(t)$ versus time for quenches to zero final temperature. Note that the system freezes. Curves represent the average over 40 runs, for a 20×20 system.

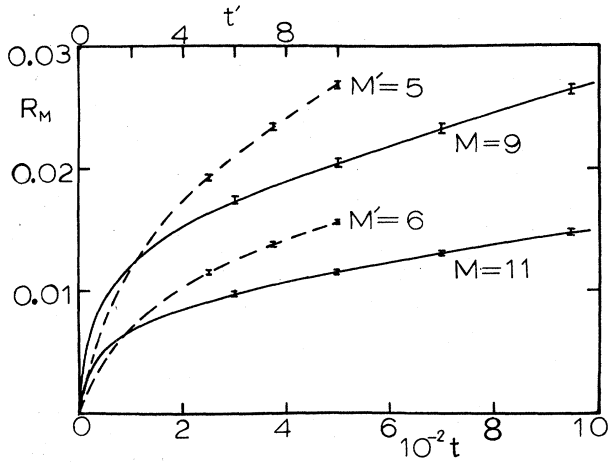

FIG. 7. $R_M(t)$ versus time (solid lines and lower scale) for quenchings to a final temperature corresponding to $y=e^{-4K_F}=0.02$, and $R_{M'}(t')$ versus t' (dashed lines and upper scale) for quenchings to the corresponding renormalized temperature. The error bars denote the statistical errors over 180 runs.

We have taken particular care to verify that this zero-temperature behavior is not a finite-size effect by performing additional zero-temperature simulations in a larger ($N=40$) system, which confirm the previous results. We conclude that the system indeed freezes—does not reach equilibrium—at zero temperature, quite in contrast with the NCOP case.

The situation is quite different for quenches to a finite temperature ($y\neq0$). In Fig. 7 we plot $R_M(t)$ versus time for $M=9$ and 11 (solid lines) at $y=0.02$. The error bars indicate the statistical fluctuation. We also plot $R_M(t)$ versus t' for $M'=5$ and 6 (dashed lines) at the corresponding renormalized temperature[73] (as determined from the condition $\xi'=\xi/2$). The parameter Δ is, of course, extracted from this data (and similar data obtained at other temperatures).

Our first, and most important result, is that, within statistical error, there indeed exists a time-rescaling parameter Δ which depends on the ratio b rather than on M and M' separately. Thus, we find that this model does exhibit scaling. It is, however, immediately seen from the data that Δ depends on y: since $R_M(t)$ does not saturate ("freeze") at any final quenching temperature T_F, where $T_c > T_F > 0$, it will not (at $y>0$) exhibit the t^{-1} long-time behavior which we found at zero temperature. We find, in fact, that

$$\lim_{t\to\infty} \Delta(t,y)=\Delta_\infty(y) \ , \tag{4.4}$$

where, within the range of y considered here, $\Delta_\infty(t)$ is proportional to y and vanishes as $y\to0$.

As explained in Sec. II, this kind of temperature dependence of Δ at long times implies that the system exhibits only a weaker form of scaling where the growth laws are *not* power laws in time but are (at finite temperature), logarithmic. This is a very fundamental result, which, when combined with our analytical RG methods developed previously,[48,69,44,41] leads[74] to excellent agreement with direct

MC simulations for the structure factor $C(\mathbf{q},t)$. We point out, nevertheless, that nearly any monotonic growth can be fitted to a power law over a restricted time range, as it has been customarily done for the present model. We are not, therefore, directly contradicting the many published power-law growth results: As we have explained in Sec. I, the power-law values reported in the various theoretical, experimental, and simulation papers referred to there, are actually consistent with a different growth law (logarithmic, in fact, according to our calculations) which appears to be a power law over a decade or two of the time variable.

In Fig. 8 we present our results (dots) for Δ at several values of the final quenching temperature (as specified by the variable y). These values are obtained from data for R and R' averaged over a number of runs ranging from 120 to 200 (depending on y) and over two M,M' pairs. For $y \leq 0.03$, we include times up to 10^3 MCS, and a smaller range for higher temperatures. We clearly see that Δ is indeed dependent on t and y. $\Delta(t,y)$ at constant y decreases rapidly in the region $t < 10^2$ MCS, and approaches $\Delta_\infty(y)$ as the time increases further. We have found that our results for $\Delta(t,y)$ can be represented, within their statistical accuracy, by the expression

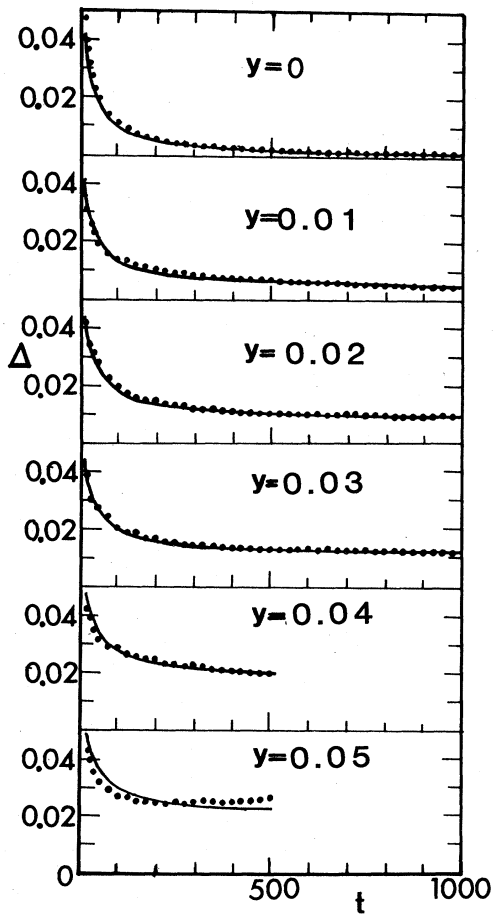

FIG. 8. The time rescaling parameter Δ versus for several values of the final quenching temperature. The dots represent our results, and the solid lines the best fits of the form (4.5).

$$\Delta(t,y)=\Delta_\infty(y)+\frac{\Delta_0(y)}{1+t/t_0(y)} . \tag{4.5}$$

The quantity $\Delta_\infty(y)$ can be determined from a linear least-squares fit of Δ versus $1/t$ for times $t > 150$ MCS. We find then that $\Delta_\infty(y)$ is given approximately by

$$\Delta_\infty(y)=0.41y . \tag{4.6}$$

We then use $\Delta_\infty(y)$ to find, from the data for $10 \leq t \leq 150$, the values of Δ_0 and t_0 in (4.5). We find that $\Delta_0(y)$ and $t_0(y)$ vary only very slightly, and unsystematically, as we vary y. We attribute this small variation to statistical uncertainties in the data and conclude that, independently of y, we have $\Delta_0=0.057$ and $t_0=20$. These values (together with those for Δ_∞) yield good fits to the data at all values of y considered, as can be seen from Fig. 8, where the fits are indicated by the solid lines.

We have, therefore, shown in this section that SE dynamics exhibits a very different kind of scaling behavior from that found in the Glauber case. There is still a time-rescaling parameter Δ, but it depends not only on t, but also on y, and it vanishes as long times, at $y=0$. As indicated in Sec. II this leads to logarithmic growth laws at finite temperatures T_F and freezes at zero temperature. Of course, it now becomes very appealing to develop a RG theory of spinodal decomposition along the lines of Refs. 69 and 41, incorporating this determination of Δ, and we do so in Ref. 74.

V. CONCLUSIONS

We have attempted in this paper to elucidate the long-time growth behavior of kinetic Ising systems subject to a critical quench with the order parameter being either conserved or nonconserved. We have used RG methods to verify scaling behavior and growth laws at long times from an analysis of Monte Carlo data for fairly small size systems and very modest time scales.

In the NCOP case our procedure reproduces previously known results: the usual scaling forms for the structure factor and associated quantities and the LCA power-law growth. In part, therefore, the results in this case (Sec. III) must be considered a check on the consistency of our method, as well as a demonstration of their power.

For spinodal decomposition, however, we have found a very different situation. There is still scaling for quenches to a temperature T_F, where $T_c > T_F > 0$, which is reflected in the existence of a time-rescaling parameter Δ associated with the self-similarity of the system under rescalings of both space and time. However, the long-time value of Δ turns out to depend strongly on the quenching temperature. The temperature dependence of Δ is connected to two interrelated effects: First, a freezing behavior for quenches to zero temperature such that the system never reaches equilibrium. Second, for quenches to finite temperature, we have found logarithmic, rather than power-law, long-time growth. As we have pointed out in the Introduction, this result does not contradict the many (and different) power laws reported by many authors for this problem: A logarithmic law does look like a power law over restricted time ranges. We will show in a future pub-

lication how, over the time ranges considered in traditional MC simulations, a RG analysis of spinodal decomposition reproduces the apparent power-law behavior (and the exponent) found in Ref. 25 very well, when one looks only at the time range in which the simulations are performed.

An important implication of four work here, and that in Ref. 36, is that a key ingredient in identifying the "universality" class associated with growth kinetics of a particular set of systems will be to establish whether the systems freezes after quenches to zero temperature or not.

Our work here also throws additional light on the RG methods which we have in the past used to treat several nonequilibrium problems. The nonperturbative procedures of Sec. II complement and generalize the perturbative analysis we have given elsewhere (see Refs. 44 and 48 for example). We will discuss additional applications of these procedures in future work.

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[75]This time unit corresponds to setting the fundamental flipping rate α, discussed in Refs. 48 and 44, equal to 1.

[76]These data were not all taken over the same 700 runs. The small fluctuations seen in the data could be reduced further by computing all the $R_M(t)$ over the same runs. Our procedure was to sample all blocks of side M which could be fit into the system of size N^2.

[77]For the case of SE dynamics, two of our MCS equal one time unit in Refs. 20–23.

[78]While we have used the definition $b=M'/M$ in Sec. III, we have used $b=(M'-1)/(M-1)$ in Sec. IV. This is irrelevant for sufficiently large M,M' pairs and does not lead to any changes in the physics of the problem.