Growth of unstable domains in the two-dimensional Ising model

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We investigate the ferromagnetic Ising model with spin-flip dynamics by Monte Carlo computer simulation. The system is prepared at time $t=0$ by deeply quenching from a high-temperature disordered state, to a low-temperature nonequilibrium state. We analyze the growth of domains of the ordered phase through two measures of the average size of these domains: the fluctuation in magnetization and the perimeter density. Systems of size 60^2, 75^2, 105^2, 150^2, and 240^2 are studied over large numbers of quenches (from 48 to 450 on a given lattice). We find that domains grow self-similarly following the Allen-Cahn law (domain area proportional to time). The effects of different updating procedures, finite size, and varying number of runs on the evolution and the statistics of the data are studied. We find that the time evolution given by random updating or a multispin coding algorithm are the same. We estimate the percentage error in the observed size of domains from a simple zero-time sum rule, which is independent of system size. This is found to be a reasonable estimate of error throughout the self-similar scaling regime.

I. INTRODUCTION

The development of order in a rapidly quenched system is a problem of great current interest.[1–4] In a binary alloy undergoing an order-disorder transition (a two-state degenerate, nonconserved system), convoluted domains form which grow self-similarly through their interfacial motion. A typical evolution following a deep quench is shown in Fig. 1. According to theory,[3–6] the average size of the domains $\bar{R}$ grows in time via the Allen-Cahn growth law

$$\bar{R}(t) \propto t^{1/2} . \tag{1}$$

Furthermore, lengths are predicted to scale with $\bar{R}$ during the growth process. In particular, the nonequilibrium (elastic) structure factor scales to a good approximation,

$$S(k,t)=\bar{R}^{d}(t)F[k\bar{R}(t)] , \tag{2}$$

in d dimensions, where k is the wave number.

These predictions have been investigated by experiment and computer simulation. A recent experiment by Wang and Lu studied oxygen chemisorbed on W(112).[7] Their data are consistent with the $t^{1/2}$ law. Two lattice gas models which have been extensively studied are the Ising antiferromagnet with spin-exchange[15] dynamics [which corresponds to the O/W(112) experiment] and the Ising ferromagnet with spin-flip[16] dynamics. Both models are expected to have the same growth law as the experimental order-disorder systems. The antiferromagnet has received the most attention: Phani *et al.*[8] studied it in three dimensions, while Sahni *et al.*[9] and Kaski *et al.*[10] considered two dimensions. The two-dimensional ferromagnet has been studied by Kawabata and Kawasaki,[11] and quite recently by Mazenko and Valls.[6] All these studies observed scaling and the $t^{1/2}$ law. Mazenko and Valls also find extremely good agreement between their simulation and a phenomenological renormalization-group theory of domain growth which they have proposed.

The application of the Monte Carlo method to dynamics is somewhat controversial, however.[17] The principal problem arises from the fact that the computer algorithm one chooses for updating the spins becomes an integral part of the model. That is, for every different updating procedure employed, a new dynamical equation is, in effect, being simulated. Furthermore, the criteria for

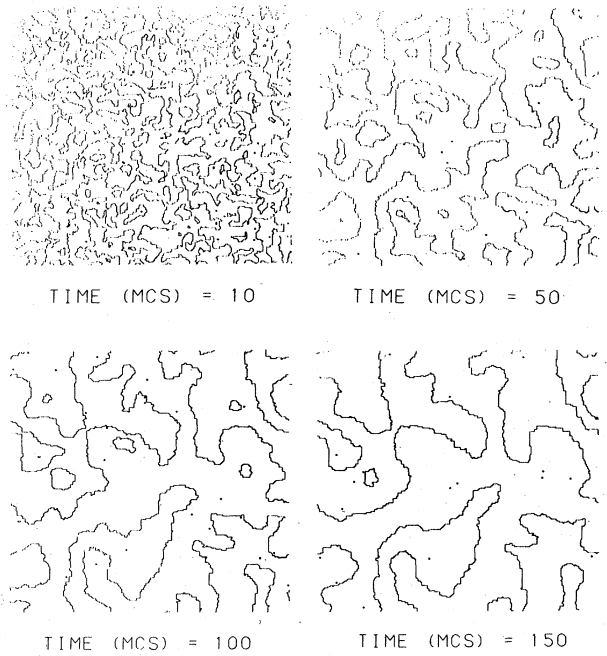

FIG. 1. Typical evolution of a 240^2 system after a deep quench to $T=1$. The interfaces separating the domains are drawn for $t=10$, 50, 100, and 150 MCS. Note the self-similarity of the domain structure in time.

minimizing statistical error in computer-simulation data involve balancing N (the number of spins) versus n (the number of quenches) in an appropriate way. The subtlety of this problem has been discussed in the literature.[8–12] Recently, in an excellent Monte Carlo study of a four-state degenerate model, Sadiq and Binder[12] showed that increasing N does not necessarily lead to better statistics. This has also been emphasized by Mazenko and Valls.[6] As more complex lattice models are studied, this problem becomes increasingly important.

In this study we return to the Ising ferromagnet with spin-flip dynamics which models the kinetics of an order-disorder transition in a binary alloy. We have done this for five reasons: (i) to test the effect of various computer algorithms for updating spins, (ii) to test scaling and the $t^{1/2}$ law on large lattices with large numbers of runs, (iii) to study finite-size effects, (iv) to investigate the consequences of varying the number of quenches, and (v) to provide a basis for comparison in a separate paper on the Ising model in a random magnetic field.[13]

We have prepared the two-dimensional Ising ferromagnet in an unstable state. At $t=0$, the system is quenched from infinite temperature, through the critical temperature, to a low temperature $T/J=1$, where J is the Ising-interaction constant and Boltzmann's constant is unity. The Ising Hamiltonian is

$$\mathscr{H}=-J\sum_{\langle ij\rangle}\sigma_i\sigma_j\ , \tag{3}$$

where the sum runs over nearest neighbors, and $\sigma=\pm 1$. The dynamics of this model are given by a stochastic interaction with a heat bath, via the spin-flip probability

$$W_i=\begin{cases} e^{-\Delta E_i/T}, & \Delta E_i\geq 0 \\ 1, & \Delta E_i\leq 0 \end{cases} \tag{4}$$

where ΔE_i is the change in energy due to flipping spin σ_i. A spin flips if W_i exceeds a random number between 0 and 1. We define a Monte Carlo step (MCS), our fundamental unit of time, as N attempts to flip spins. The updating procedure is discussed below. Our attention has focused on two quantities: the nonequilibrium mean square of the magnetization fluctuations per spin,

$$\langle M^2\rangle(t)=\left\langle\left[\frac{1}{N}\sum_{i=1}^{N}\sigma_i\right]^2\right\rangle\ , \tag{5}$$

and the average energy per spin

$$E(t)=\left\langle-\frac{J}{N}\sum_{\langle ij\rangle}\sigma_i\sigma_j\right\rangle\ . \tag{6}$$

Since the $k=0$ peak of the structure factor is $S(0,t)=N\langle M^2\rangle(t)$, scaling [Eq. (2)] implies the length measure

$$\bar{R}_M^2(t)=N\langle M^2\rangle(t)\ , \tag{7}$$

originally introduced by Sadiq and Binder.[12] It is straightforward to see that the average perimeter length per unit area is $\bar{R}_p^{-1}$, where[12]

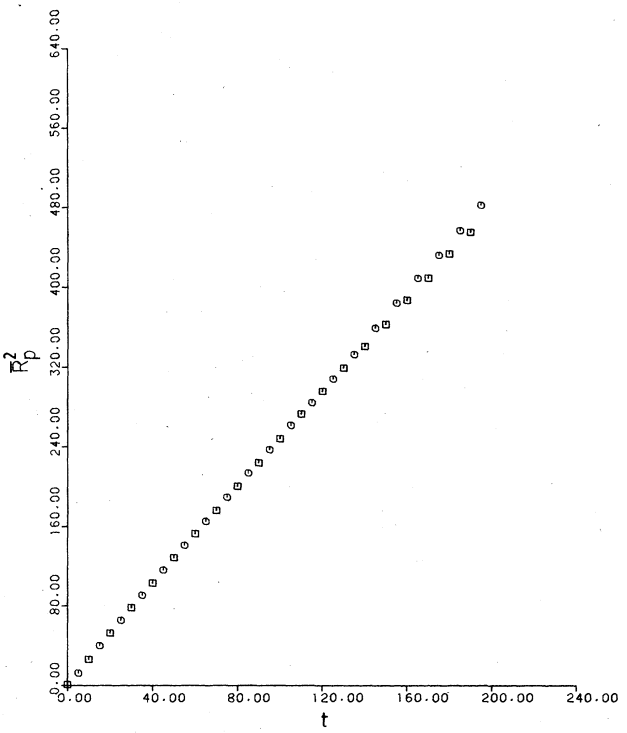

FIG. 2. $\bar{R}_p^2(t)$ vs. t for random updating (⊡) and multispin coding (⊙). $N=60^2, T=1$, 1000 quenches each. Note the excellent agreement with Allen-Cahn theory.

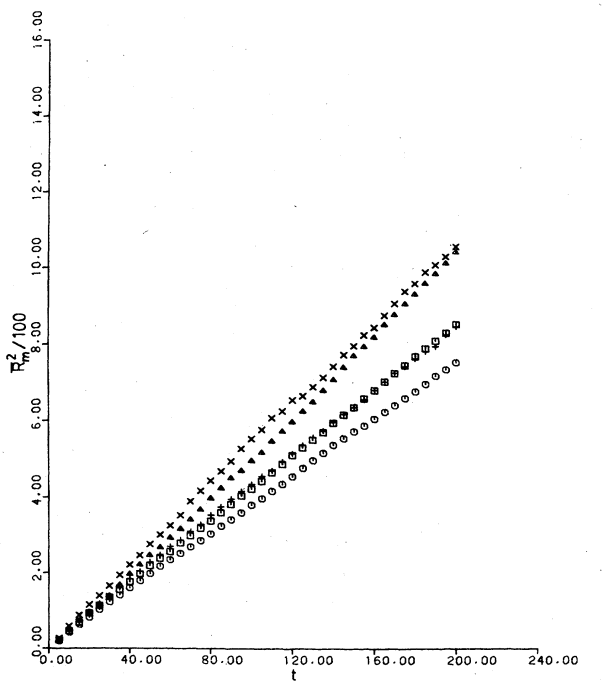

FIG. 3. $\bar{R}_M^2/100$ vs t(MCS) where $\bar{R}_M^2=N\langle M^2\rangle$. These data indicate good agreement with the Allen-Cahn growth law $\bar{R}^2\propto t$. No systematic finite-size effects are apparent. ⊡, ⊙, △, +, and × represent system sizes $N=60^2$, 75^2, 105^2, 150^2, and 240^2, respectively.

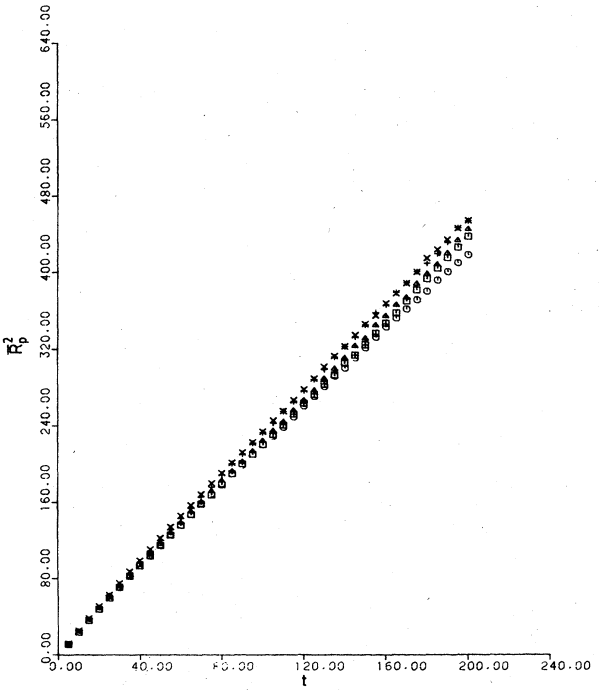

FIG. 4. $\bar{R}_p^2=[2/(2+E/J)]^2$ vs t(MCS). Again note the good Allen-Cahn agreement. Plotting symbols indicate the same system sizes as in Fig. 2.

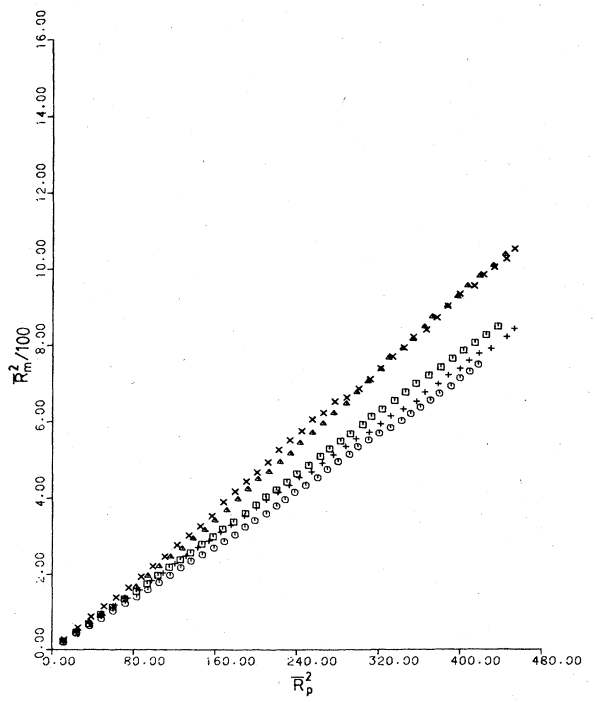

FIG. 5. $\bar{R}_M^2/100$ vs $\bar{R}_p^2$. Note that $\bar{R}_M^2 \propto \bar{R}_p^2$ indicating that these independently determined lengths have the same time dependence. This is a consequence of dynamic scaling. Time range is from 0 to 200 MCS.

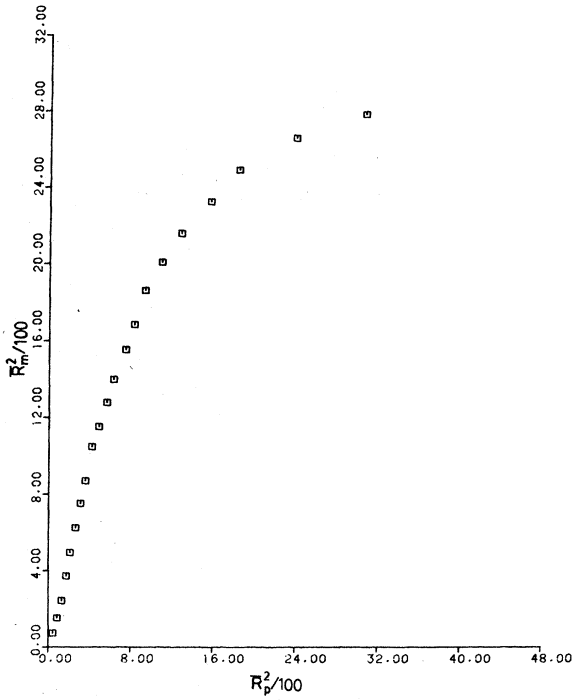

FIG. 6. $\bar{R}_M^2/100$ vs $\bar{R}_p^2/100$ for a 60^2 system. Time range is from 0 to 525 MCS. The breakdown of scaling is apparent for $\bar{R} \gtrsim 0.4\sqrt{N}$ due to finite-size effects.

$$\bar{R}_p(t)=\frac{2}{2+E(t)/J} \ . \tag{8}$$

Our results for these lengths in the different sized systems are given in Figs. 2–6.

II. MASTER EQUATION AND THE MONTE CARLO ALGORITHM

In this section we will examine the relationship between the standard master equation and the Monte Carlo algorithm. We will see that, in a very real sense, it is the algorithm which defines the dynamical model. Nevertheless, we will present empirical evidence demonstrating that there are many dynamical equations (that is, many algorithms) which give the salient dynamical properties in a universal way. Our discussion is partly based on a recent paper by Choi and Huberman.[17] In discrete time the standard master equation for the probability distribution function $\rho(\sigma,t)$ can be written in the form

$$\rho(\sigma,t+1)-\rho(\sigma,t) = -\sum_{i=1}^{N}[W_i(\sigma_i)\rho(\sigma_i,t)-W_i(-\sigma_i)\rho(-\sigma_i,t)] \ . \tag{9}$$

The Monte Carlo algorithm for the same spin-flip probability can be written in the form

$$\rho(\sigma,t+1)-\rho(\sigma,t) = -\sum_{i=1}^{N}{}' \left[W_i(\sigma_i)\rho\left[\sigma_i,t+\frac{i-1}{N}\right] - W_i(-\sigma_i)\rho\left[-\sigma_i,t+\frac{i-1}{N}\right]\right], \quad (10)$$

where the unit of time is now one Monte Carlo step (N attempts to update spins). The prime on the summation means that the spins σ_i are updated in some particular order.

Clearly these two equations are different because of the additional time dependence of ρ on the right-hand side of Eq. (10). (Both dynamical equations have the same condition for detailed balance.) In Eq. (9) all N spins flip in the same environment, and in no particular order. In Eq. (10), however, the spins flip in some specified order. To be explicit, while the spin σ_1 flips in the environment of the tth Monte Carlo step, σ_N flips in the environment of the $(t+1)$th Monte Carlo step [to order $1/N$ in Eq. (10)], which is not the case for the master equation. Thus, if the Monte Carlo algorithm is to simulate the standard master equation, there must be no significant evolution during a Monte Carlo step.

A concrete way to express this is as follows. One must require that a physical quantity $R(t)$ (e.g., a moment of ρ) satisfies

$$\left|\frac{R(t+1)-R(t)}{R(t)}\right| \ll 1 \quad (11)$$

for the Monte Carlo algorithm, where t is measured in Monte Carlo steps. While this is certainly a necessary condition, it is by no means sufficient. To our knowledge, there is no proof, rigorous or otherwise, of any limit in which Eqs. (9) and (10) are identical. Instead there are only plausible arguments (folklore) and empirical evidence. We note that rigorous proofs would be of great value and considerable interest. Nevertheless, for the particular problem we are studying (the growth of unstable domains following a temperature quench) there are some additional requirements which must be satisfied. In particular, for late times, we should observe the Allen-Cahn growth law, Eq. (1). (Although this result is usually derived by field-theoretic analysis, it is to be expected that it also follows from the master equation.) Furthermore, by integrating the *continuum* master equation, following Mazenko and Valls,[6] we find that

$$\bar{R}_M^2(t)=1+\alpha t+O(t^2), \quad (12)$$

as $t\to 0$, where $\alpha\simeq 3.0$ for $T/J=1$. [Equation (12) is completely unrelated to the Allen-Cahn growth law and simply describes initial transient behavior.] These three constraints, Eq. (11), the Allen-Cahn growth law, and Eq. (12), will enable us to provide empirical evidence relating the Monte Carlo algorithm to the master equation.

Any differences between the algorithm and the master equation are intimately connected to the way spins are updated in the primed summation of Eq. (10). Certainly the simplest way to update spins is to go through the lattice sequentially, row by row. It is evident, however, that this can lead to the sort of spurious evolution during a Monte Carlo step discussed above. Indeed, test runs we have carried out indicate that Eq. (11) is violated, and the Allen-Cahn growth law is not present, for this updating procedure. Another way to update spins is as follows. One visits the spins on the lattice in random order. Doing this we find that Eq. (11) is satisfied, and the Allen-Cahn result is recovered (see Fig. 2). In addition, by interpolating times between 0 and 1 Monte Carlo steps [i.e., we call an updating of 1 spin $(1/N)$th of a Monte Carlo step] we find that Eq. (12) is satisfied.[18] On the basis of this evidence, the random updating algorithm is faithfully simulating the standard master equation.

Many other ways to update spins in an algorithm are possible. Note that in a random updating, one typically finds that widely separated spins are being updated (which is why there are negligibly few unphysical correlations there, causing history-dependent effects inside a Monte Carlo step). Of course, one can simply choose several widely separated spins and update them simultaneously. This is what is done in a multispin coding algorithm.[19] We have studied such an algorithm where we randomly update groups of 15 widely separated spins.[20] The advantage of this is that, because the orientation and energy of 15 spins can be stored in a 60-bit word, a factor of 15 is gained in computer storage space. In addition, the algorithm results in a doubling of execution speed over the random updating method discussed above. Again we find that Eq. (11) is satisfied, and that we obtain the Allen-Cahn result. In fact, as can be seen from Fig. 2, the results are indistinguishable from the random updating method. Also, interpolating between 0 and 1 Monte Carlo steps [where an updating of 15 spins is called $(15/N)$th of a Monte Carlo step] we again find that Eq. (12) is satisfied.[18] Therefore the multispin coding algorithm is also faithfully simulating the master equation, on the basis of this evidence. Thus, both the random updating and the multispin coding algorithms simulate the master equation which models the kinetics of an order-disorder transition in a binary alloy.

It is important to point out that these results and conclusions apply only to the model we are studying, and may differ significantly for other models. In general, any technique for simulating a master equation should be used with some caution, particularly when dynamics are studied. For the remainder of this paper we will study the phase-separation kinetics using the multispin coding algorithm discussed above (and in Fig. 2 the random updating algorithm).

III. RESULTS

We have considered several system sizes, always with periodic boundary conditions, 60^2, 75^2, 105^2, 150^2, and 240^2.[2] The number of runs on each lattice size was 450, 300, 180, 90, and 48, respectively. (In Fig. 2, 1000 runs for the 60^2 system are shown.) We plot our results for the two lengths $\bar{R}_M^2$ and $\bar{R}_p^2$ in Figs. 2–4. The data are represented well by the theoretical growth law, Eq. (1). Scaling is observed since they have the same time depen-

dence over $t=1$ to $t=200$ MCS. Over these times, we observe no systematic finite-size effects within the precision of our study. We find that such effects become important for $\bar{R} \gtrsim 0.4\sqrt{N}$, as was found in earlier studies.[10–14] However, it should be emphasized that these effects are primarily due to percolating clusters (slabs) which are artifacts of the periodic boundary conditions. Other more subtle finite-size effects are also possible.[14] In Fig. 5, we compare the time behavior of the two lengths.

We have studied an additional 36 quenches in the 60^2 system over longer times ($t \sim 1000$ MCS). In Fig. 5 we display results for 22 of these quenches which eventually reached equilibrium. To show the breakdown of scaling for $R \gtrsim 0.4\sqrt{N}$, we have plotted $\bar{R}_M^2$ versus $\bar{R}_p^2$ out to 525 MCS. It should be emphasized that, in contrast to the results here, in the thermodynamic limit ($N \to \infty$) the system will not equilibrate in a finite amount of time.

Finally, let us consider the criteria for minimizing statistical error in the data.[21] We have found it useful to analyze the disordered system which is present at zero time. From Eq. (12),

$$\bar{R}_M^2(t=0)=1 .$$

Further, we find that the error for $\mathcal{N}$ quenches is given by the sum rule,

$$\delta\bar{R}_M^2(t=0) \equiv \bar{R}_M^2(t=0)\Delta_0 = \sqrt{2}/\sqrt{\mathcal{N}} . \tag{13}$$

The $\sqrt{\mathcal{N}}$ factor follows from standard error analysis, while the $\sqrt{2}$ factor comes from the fourth moment of the binomial distribution.[22] In Fig. 7, we plot the observed relative error at time t, $\Delta(t)$, versus the calculated zero-time relative error Δ_0 for the different system sizes (i.e., different number of quenches) and for three times: $t=0$, $t=100$, and 200 MCS.

The approximate time and system size independence of the relative error can be understood as follows. Let the 240^2 system be broken down into 16 60^2 systems. One might think that 48 quenches on the 240^2 system would correspond to 768 quenches on the smaller 60^2 system. However the built-in $t=0$ uncertainty (calculated above) is not changed by breaking down a large system into many smaller ones.[23] Furthermore, scaling implies that the error can only depend on time through the characteristic length $\bar{R}(t)$. Thus, $\Delta(t)$ is independent of time to a good approximation, and Eq. (12) provides an error estimate which is independent of system size throughout the self-similar scaling regime. Of course one must, in addition, still consider systems large enough to encompass all the necessary physics of the model being studied.

To conclude, we have analyzed the effects of varying both system sizes and numbers of quenches on the domain growth in the ferromagnetic spin-flip Ising model. We found that the well-known results of theory and experiment, the $t^{1/2}$ law and self-similar scaling, showed no systematic finite-size deviations until domains had grown to a size $\bar{R} \gtrsim 0.4\sqrt{N}$, within the precision of our study. We

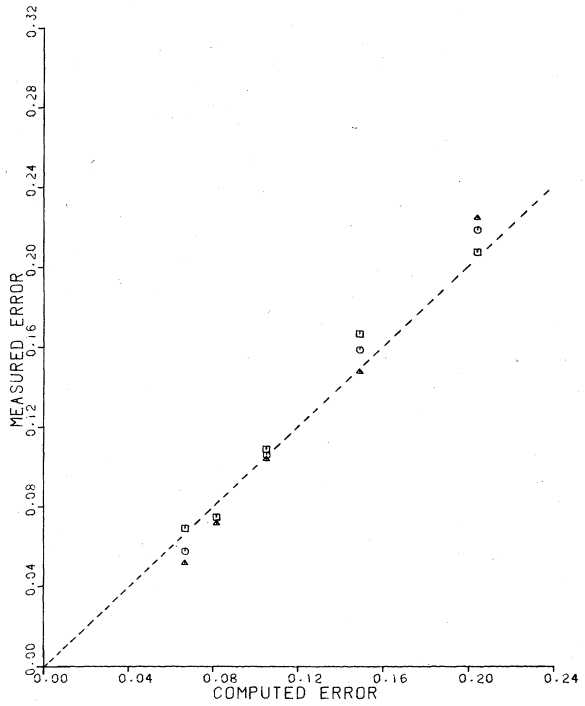

FIG. 7 Measured error (i.e., observed statistical error) versus computed error at $t=0$ [see Eq. (12), $\sqrt{2}/\sqrt{\mathcal{N}}$, for the quantity $\bar{R}_M^2$. Three times are shown for each system size: $t=0$ (⊡), $t=100$ (·), and $t=200$ (△). All times are in Monte Carlo steps. The dashed line with unit slope is the computed error, $\Delta_0(n)=\sqrt{2}/\sqrt{\mathcal{N}}$.

tested the effect of different procedures for updating the spins during simulation. We found that sequential updating led to a time evolution so fast that the Cahn-Allen growth law was no longer observed. The multispin and single-spin coding algorithms with random updating were extensively tested and found to be representative of the standard master equation associated with the kinetic Ising model. We also obtained an estimate for the statistical uncertainty in the data from a simple sum rule, which was independent of system size. In a future paper we will obtain the structure factor $S(k,t)$.

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[18]For random updating we obtain $\alpha=3.0\pm0.2$ while for the multispin coding algorithm we obtain $\alpha=2.9\pm0.3$.

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[20]We have also checked that there is no preferential direction for growth due to a spatial correlation between the 15 widely separated spins.

[21]Pointwise time correlation of our data makes the results appear somewhat better than their statistical uncertainty. This is also true of other studies.

[22]Explicitly, if m is the net displacement from the origin after a one-dimensional random walk of N steps, then $\langle m^4\rangle-\langle m^2\rangle^2=2N(N-1)$. After appropriate normalization this gives Eq. (13). Generalizations to models with more than two degenerate ground states should be straightforward.

[23]See also the discussion in Ref. 12.