Dynamics of a two-dimensional order-disorder transition

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We present results of a Monte Carlo study of the time development of a two-dimensional order-disorder model binary alloy following a quench to low temperature from a disordered, high-temperature state. The behavior is qualitatively quite similar to that seen in a recent study of a three-dimensional system. The structure function exhibits a scaling of the form $K^2(t)S(k,T) = G(k/K(t))$ where the moment $K(t)$ decreases with time approximately like $t^{-1/2}$. If one interprets this moment as being inversely proportional to the domain size, the characteristic domain growth rate is proportional to $t^{-1/2}$. Additional insight into this time evolution is obtained from studying the development of the short-range order, as well as from monitoring the growth of a compact ordered domain embedded in a region of opposite order. All these results are consistent with the picture of domain growth as proposed by Lifshitz and by Cahn and Allen.

I. INTRODUCTION

In a recent paper[1] the results of a computer simulation of the development of order in a quenched three-dimensional-model binary alloy were given. In this model of an order-disorder transition the order parameter is not conserved, with the result that the dynamical evolution of order from an initially disordered, high-temperature state is qualitatively different from the corresponding spinodal decomposition for a binary alloy whose order parameter is conserved. The basic theory for the time evolution of this nonconserved, order-disorder transition is due to Lifshitz[2] and to Cahn and Allen[3] (LCA); it describes the motion of curved domain walls separating regions of opposite order. These theories assume that at a given time t following the quench to a low-temperature state, domains form in which the instantaneous value of the long-range order (LRO) parameter $\eta_l(t)$ is close to the equilibrium values of $\pm\eta_l^{eq}$. The overall long-range order is, however, close to zero, with the plus and minus domains having essentially equal statistical weights. The driving force for the dynamical evolution of this system is then such as to reduce the curvature of the domain walls. The theories predict that the surface area per unit volume decreases as $t^{-1/2}$.

Several points concerning these $t^{1/2}$ predictions for a nonconserved order parameter should be made. First, the original Lifshitz theory involves the surface tension which enters as a proportionality factor in the domain growth. A major result of the Cahn-Allen analysis, however, was that their constant of proportionality did not involve the surface tension—there was therefore no critical slowing down as $T \to T_c$. They also noted that the experimental results for coarsening kinetics in Fe-Al alloys were consistent with their theory; they did not seem to depend strongly on the surface tension. Thus the Lifshitz and Cahn-Allen theories differ by a temperature-dependent term. Second, other theories also predict a $t^{1/2}$ behavior. These include a "solitary-wave" analysis by Chan[4] for a more general problem than considered by Cahn and Allen, but which yields similar results. A weak-coupling, long-time, study of a continuum version of a nonconserved dynamical model (the time-dependent Ginzburg-Landau model) by Kawasaki, Yalabik, and Gunton[5] also yields the Cahn-Allen result. In addition, such a time dependence has been observed by Binder and Billotet[6] in a Langer–Bar-on–Miller[7]–type theory of a two- and three-dimensional antiferromagnet. Finally, we note that a variational approach[8] to the late-stage domain growth for two models in which the order parameter is conserved (as for a binary alloy) or nonconserved (as for an antiferromagnet) explicitly shows why the surface tension appears in the conserved case (Lifshitz-Slyozov theory)[9] but not in the nonconserved case (Cahn-Allen theory).

In the Monte Carlo study by Phani *et al.*,[1] the au-

thors realized that the existence of a characteristic length [the domain size, $l(t)$] also suggests a scaling behavior for the order parameter (staggered) structure function, $S(k,t) = l(t)^d \mathfrak{F}(kl(t))$ where d is the dimensionality, k the wave number, and $\mathfrak{F}$ a scaling function. These ideas were confirmed in the Monte Carlo studies at early times when finite-size effects can be ignored. For later times, when the domain size becomes comparable to the size of the system, these finite-size effects must be taken into account. However, the late stages also seemed to be consistent with a theoretical interpretation based on the LCA picture.

In this paper we report an extension of these computer-simulation studies to the two-dimensional model of order disorder. We find that in general the behavior is the same as in three dimensions. In particular, there is a scaling behavior for the structure function which we have analyzed in two different ways. In the first case we have scaled our data with a length, $K^{-1}(t)$ obtained from the second moment $K^2(t)$ of the structure function. This gives a rather direct determination of a characteristic length and we find that the structure function scales reasonably well in the early time region with this choice. We also find that if we approximate $K(t)$ by a simple power-law behavior we obtain the LCA exponent of $\frac{1}{2}$. Thus we have also scaled our data using $kt^{1/2}$ as a natural variable, where k is the wave number, and have obtained possibly an even better scaling of the data. We have also attempted to extract a length scale more indirectly, as in Ref. 1, from an analysis of the time development of the short-range order. Although we obtain results consistent with the $t^{1/2}$ behavior, this is a somewhat less convincing argument, as we discuss in the text. Finally we present a novel, controlled-growth experiment to test more directly the LCA theory. Namely, we monitor the time evolution of a compact ordered domain inside a background of opposite order. According to the theory, a given isolated domain should decrease in area linearly with time. Although we find large fluc-

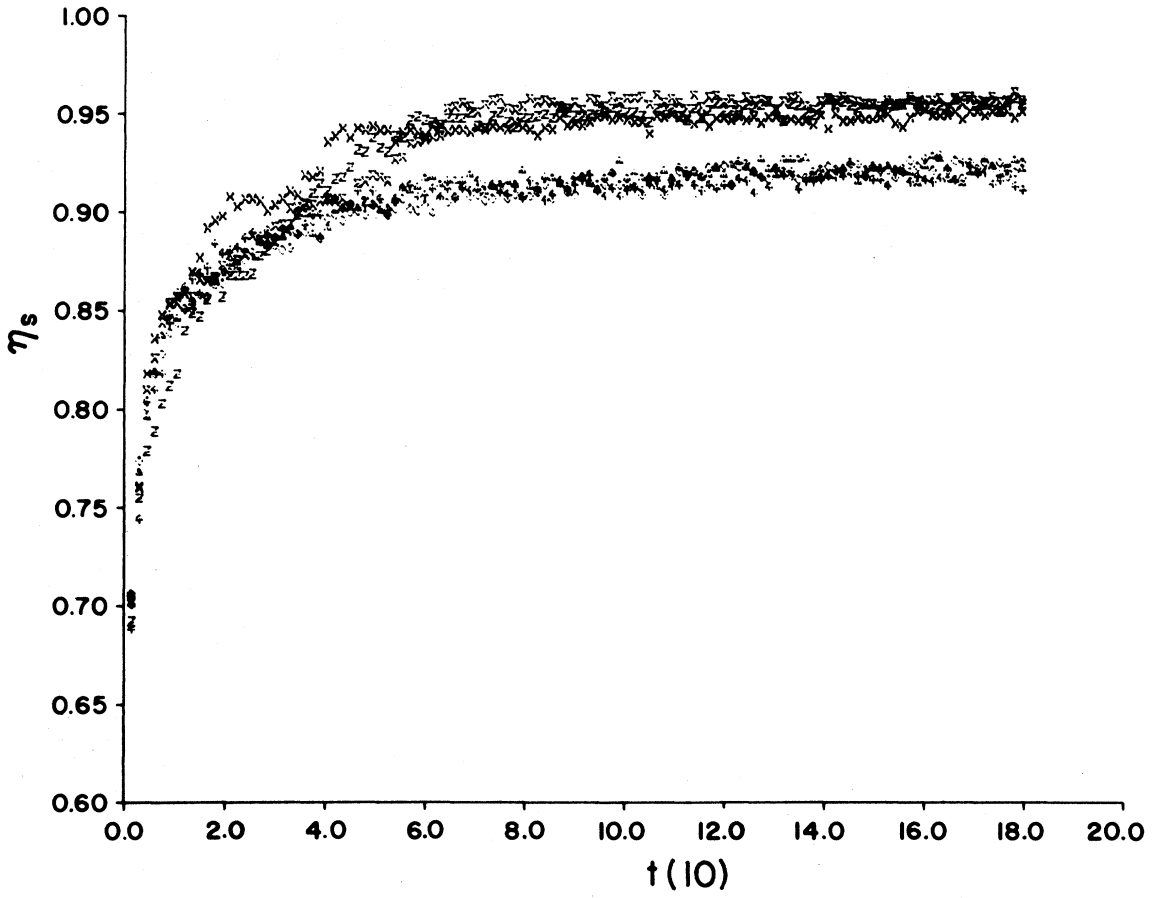

FIG. 1. SRO parameter η_s vs time for eight runs in the time interval 0–180 Monte Carlo steps.

tuations in our data, as in our study of the structure function, the results are consistent with the theory.

In Sec. II we define our model and give the results for the early time behavior of the short-range order parameter, long-range order parameter, and staggered structure function. We also present some typical configurations encountered in our Monte Carlo simulation, including one in which a rather stable "slab" forms which leads to a long-lived "metastable" state. In Sec. III we summarize the results for the late regime growth of an isolated domain, for two different initial shapes.

II. EARLY TIME BEHAVIOR

Our model alloy is a two-dimensional version of the one studied in Ref. 1. It consists of a square lattice of N sites with periodic boundary conditions. Each site $\vec{r}$ is occupied by either an A or a B atom, with an overall composition of 50% A atoms. This model is isomorphic to an Ising antiferromagnet with fixed zero total magnetization (zero magnetic field) in which the spin at each site is $\sigma(r) = \pm 1$. The Hamiltonian for the model contains only nearest-neighbor interactions, with the energy of a particular configuration given by

$$u = J \sum_{\mathrm{NN}} \sigma(\vec{r})\sigma(\vec{r}'), \quad J \geq 0 \ . \tag{1}$$

The critical point of this system is known from the Onsager solution to be given by $J/kT_c = 0.4407, \ldots$. At very low temperatures the system will be in one of two perfectly ordered states, with one sublattice being occupied by A atoms and the other by B atoms. Above the critical point, this ordered state disappears and each sublattice would then typically contain an equal number of A and B atoms.

The standard definition for the long-range order parameter η_l for this system is

$$\eta_l = N^{-1}[(N_A^e + N_B^o) - (N_A^o + N_B^e)] \ , \tag{2}$$

where $N_A^e (N_A^o)$ is the number of A atoms on the even (odd) sublattice, etc. This can also be defined in terms of σ variables as the staggered magnetization

$$\eta_l = N^{-1} \sum_{\vec{r}} (-1)^{\vec{r}} \sigma(\vec{r}) \ . \tag{3}$$

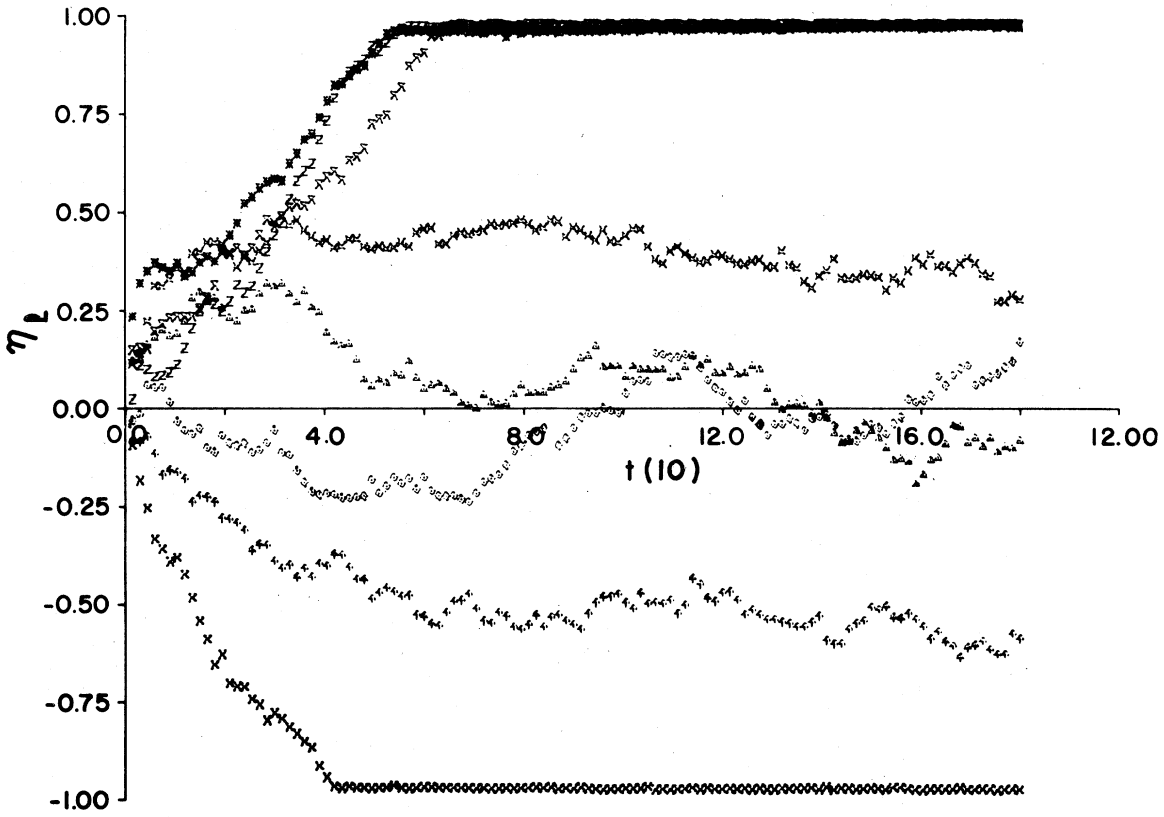

FIG. 2. LRO parameter η_l vs time for eight runs.

Another important quantity is the short-range order parameter, η_s,

$$\eta_S = [N_{AB} - (N_{AA} + N_{BB})]/2N = u/2NJ \ , \quad (4)$$

which directly measures the energy of the system. Here N_{AB}, N_{AA}, and N_{BB} denote the number of nearest-neighbor $A-B$, $A-A$, and $B-B$ bonds, respectively, with the total number of bonds being $2N$.

The dynamical properties of this model are taken to be that of Kawasaki spin exchange, in which the transition rate W for a pair of nearest neighbors to exchange is given by

$$W = \tau^{-1} e^{-\beta \Delta u}/(1 + e^{-\beta \Delta u}) \ , \quad (5)$$

where Δu denotes the change in the energy of the configuration resulting from exchanging the randomly chosen pair of opposite spins. The time scale is given by τ. We simulate the dynamical evolution of this model by a standard Monte Carlo procedure, for a system of $N = 60 \times 60$ sites. The system was quenched from an initially disordered state at an

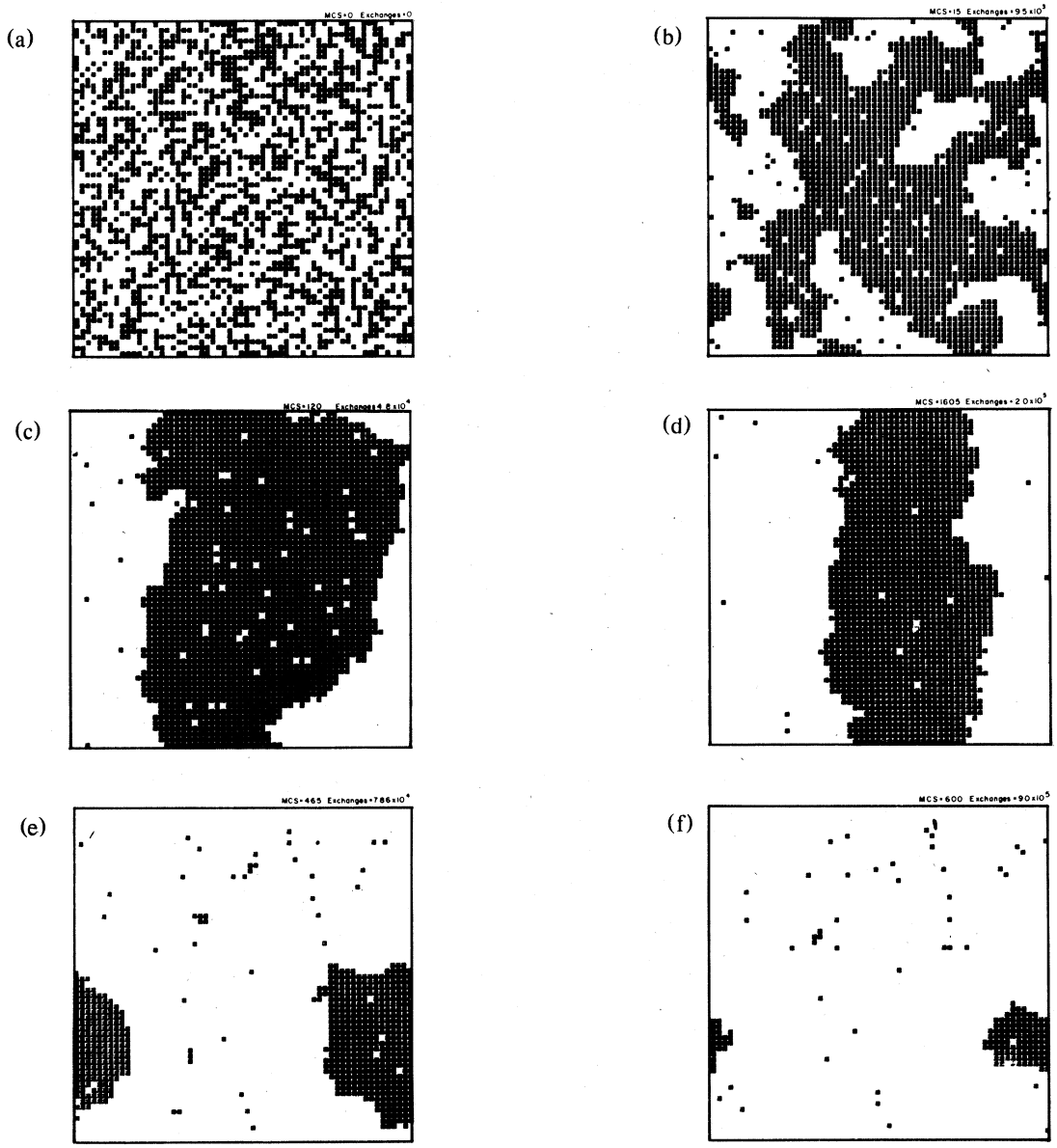

FIG. 3. The evolution of a high-temperature configuration (a) into a metastable state when quenched to a temperature $T = 0.6 T_c$ is shown in (b), (c), and (d) at the Monte Carlo times indicated. (e) and (f) correspond to a run where the LRO parameter goes to its equilibrium value.

essentially infinite temperature of $J/kT=0.000\,25$ to a low temperature of $T=0.6T_c$. We monitored $\eta_l(t)$, $\eta_s(t)$ and the circularly averaged Fourier transform $S(k,t)$ for the staggered structure function

$$S(k,t)=N^{-1}\left|\sum(-1)^{\vec{r}}e^{i\vec{k}\cdot\vec{r}}\sigma(\vec{r})\right|^2 , \qquad (6)$$

where $|\vec{k}|=2\pi j/\sqrt{N}$, with $j=0,1,2,\ldots,10$. Due to the large fluctuations encountered in the Monte Carlo studies of both the two- and three-dimensional models, we have analyzed our data using 10 runs. Plots of $\eta_s(t)$ and $\eta_l(t)$ for these runs are shown in Fig. 1 and Fig. 2, where the unit of time is the number of attempted exchanges per site. In these units the diffusion constant is $\frac{1}{8}$. Some typical stages of time evolution of the system are shown in Figs. 3(a)–3(f). As can be seen from Fig. 2, we encounter several runs in which "metastable" states persist and the system does not achieve an equilibrium. This is apparently due to the formation of very stable domains of a slablike nature which encircle our lattice. In Figs. 3(c) and 3(d) we show the evolution of a particular slab associated with one metastable run. Figures 3(e) and 3(f) show the evolution of an approximately circular domain. It should be noted that this domain is of the type described by Lifshitz and Cahn-Allen in that there is a curvature which provides the driving force for the shrinkage of this domain. On the other hand the slabs have effectively very small curvature and hence persist for much longer times than the circular domains. Since a given slab typically encircles the entire lattice, it divides the ordered region into two disjoint pieces and thereby prevents the equilibrium state from being achieved during the given run. Similar metastable effects were seen in the three-dimensional study, although they occur less often possibly due to larger size effects for our two-dimensional lattice.

In order to test the scaling behavior of the order-parameter structure function, $S(k,t)$, we have calculated the second moment, $K^2(t)$, from

$$K^2(t)=\sum k^2S(k,t)\Big/\sum S(k,t) . \qquad (7)$$

A plot of this quantity is shown in Fig. 4. If we attempt to fit this function to a power-law behavior we find a reasonable fit to $K^{-2}(t)\sim t$, as can be seen from Fig. 4. Thus we would find an exponent of approximately $\frac{1}{2}$ for the characteristic length, $K^{-1}(t)$. In Figs. 5 and 6 we show the scaling behavior of $S(k,t)$ for the different choices

$$K^2(t)S(k,t)=G(x) \qquad (8)$$

and

$$t^{-1}S(k,t)=F(y) , \qquad (9)$$

where $x=k/K(t)$ and $y=kt^{1/2}$, respectively. The fit of our data to the form $F(y)$ seems somewhat better than for the case of $G(x)$, although we would not assign any particular significance to this fact. The overall conclusion, however, would be that the structure function scales in the early time region and that it is in basic agreement with the $t^{1/2}$ prediction of LCA.

Another quantity of dynamical interest is the short-range order, $\eta_s(t)$, which form Eq. (4) is a measure of the number of like or unlike bonds, since we have

$$N_{AA}+N_{BB}=2N-N_{AB} . \qquad (10)$$

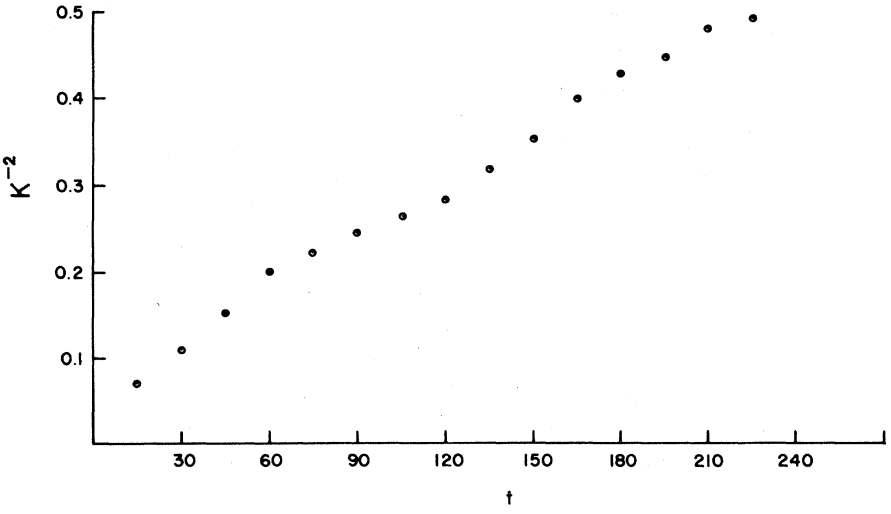

FIG. 4. A plot of K^{-2} as a function of time.

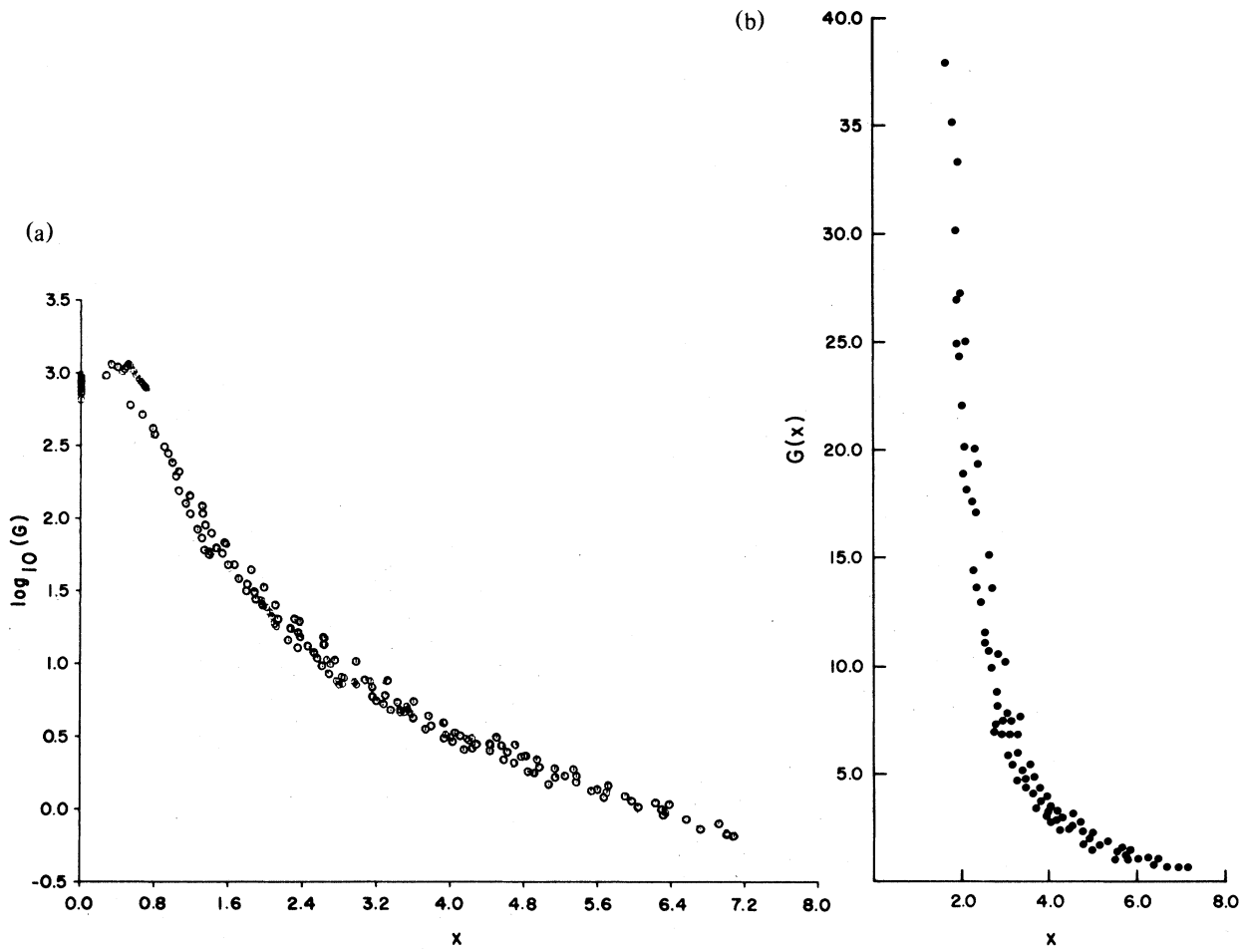

FIG. 5. The scaled structure factor $G(x)$ vs x on a (a) semilog scale and (b) regular scale.

One can attempt to fit the time dependence of this short-range order by a simple power-law behavior. If for example one considers the behavior of

$$\Delta(t) = \eta_\infty - \eta_s(t) \ , \tag{11}$$

where η_∞ is $\eta_s(\infty)$, which is essentially the Onsager value in our case (and given by $\eta_\infty = 0.9877$) one finds an exponent of 0.3, as shown in Fig. 7. On the other hand, another quantity which seems more relevant for obtaining an indirect estimate of the characteristic domain size is

$$\Delta_m(t) = \eta_m - \eta_s(t) \ , \tag{12}$$

where η_m is essentially the metastable value of the short-range order immediately after the antiphase interface has disappeared. This is not the same as η_∞ since one is still left with local "interstitial disorder," as can be seen from Figs. 3(e) and 3(f). Since it is $\eta_m - \eta_s(t)$ which is related to the "macroscopic" antiphase boundaries, one might expect that this quantity would have a time dependence which is related to the LCA theory. Indeed, if one uses the argument of Ref. 1, we would expect that a circle of radius $l(t)$ would on average intersect an antiphase boundary once (in the early time domain in which these boundaries are present), with the intersection having a length proportional to $2\pi l$. Thus the perimeter of the boundary per unit area is proportional to $(2\pi l)/\pi l^2 = 2l^{-1}$. Therefore since the perimeter in numbers of $A-A$ and $B-B$ bonds per unit area is given by $\Delta_m(t)$, one would have $l(t) \sim a\,\Delta_m^{-1}$ where a is some constant. Thus if the LCA prediction that $l(t) \sim t^{1/2}$ is correct, one should find that Δ_m^{-2} varies linearly with time. As can be seen from Fig. 7 this is in fact the case and hence gives another (indirect) confirmation of the theory. The difficulty with this argument is that the exponent one obtains for $\Delta_m^{-2}(t)$ is somewhat sensitive to the value of η_m, for which we have no unique definition. However, we have

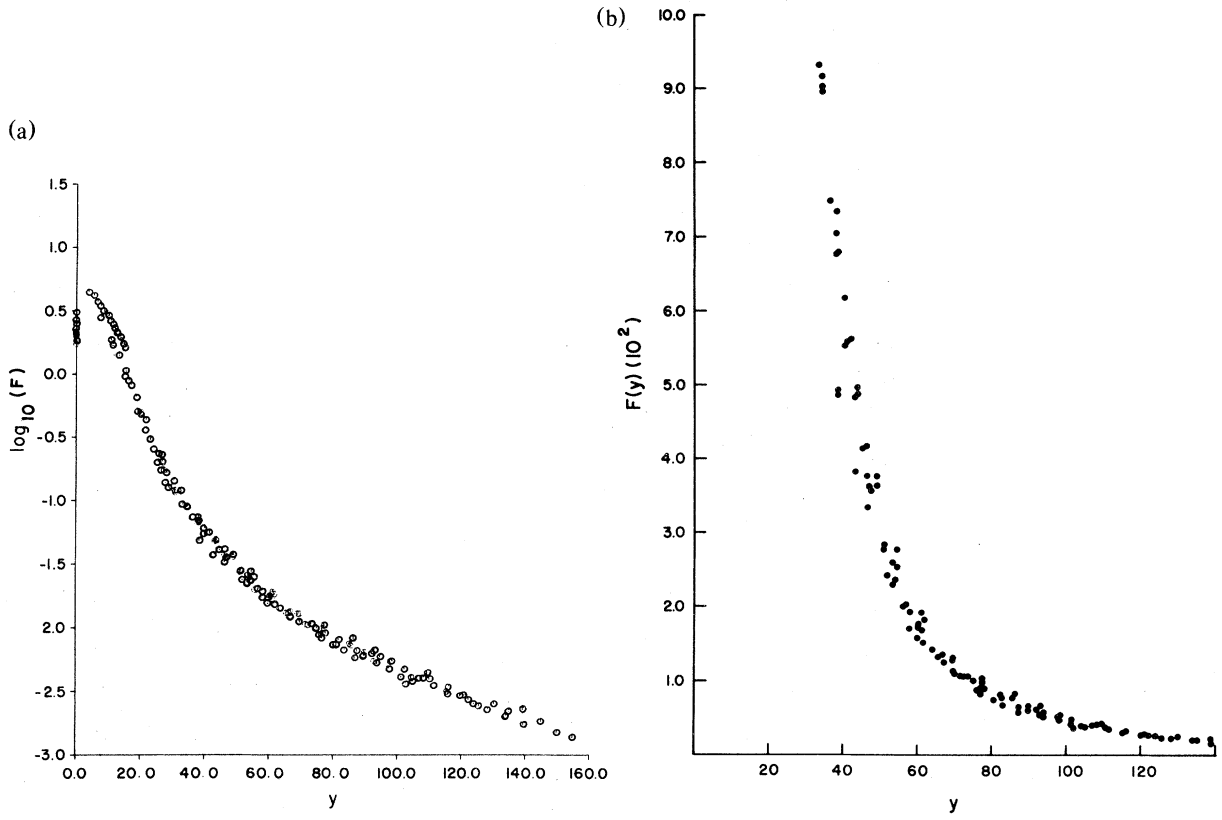

FIG. 6. The scaled structure factor $F(y)$ vs y on a (a) semilog scale and (b) regular scale.

found in practice that the values of η_m which we obtain by time averaging $\eta_s(t)$ over different time intervals following the disappearance of the antiphase boundary do not differ too much from the valued used in Fig. 7 and that the exponent remains close to the LCA value for all reasonable choices which we have considered.

III. CONTROLLED-GROWTH EXPERIMENT

As another test of the Cahn and Allen theory in two dimensions we have also monitored the time evolution of a compact ordered domain inside a sea of opposite order. According to the theory[10] a given isolated domain will decrease in area linearly with time. Assuming its shape to remain more or less fixed its perimeter would then decrease like the square root of the area, i.e., if a domain has area A_0 and perimeter S_0 at time $t=0$ then at a time t, its area $A(t)$ and perimeter $S(t)$ will be given by

$$A(t)=A_0(t_f-t) \ , \tag{13}$$

where $dA/dt=-A_0$ is a constant independent of the shape (in a continuum model which neglects fluctuations) and

$$S(t)=S_0(t_f-t)^{1/2} \ . \tag{14}$$

We started with a lattice of size 50×50 and generated a domain of one kind (which we will refer to as a "blob") of about 500 spins surrounded by the domain of the opposite kind which has a size of about 2000 spins. Figures 8(a) and 8(b) show $A(t)$ and $S(t)$ plotted versus t for eight independent runs starting from the "blob." The value of t_f differed from run to run by a considerable amount, with A_0 having a mean of 6.48 and root-mean-square deviation of 1.26. In Fig. 8(a) the solid line is the relation (14) with $S_0t_f=126$. One can see that the theory does indeed conform to the Monte Carlo data to a fair extent. In Figs. 9(a) and 9(b) similar plots for another seven independent runs starting with a "rectangular" domain of size $A_0t_f=575$ are shown. The mean $A_0=6.54\pm 1.00$. Finally it should be noted that this initial rectangular domain does not encircle the lattice, in contrast to the slabs discussed in Sec. II. Thus it has "corners" where the curvature is quite high which provide a correspondingly large driving force for shrinkage of the cluster.

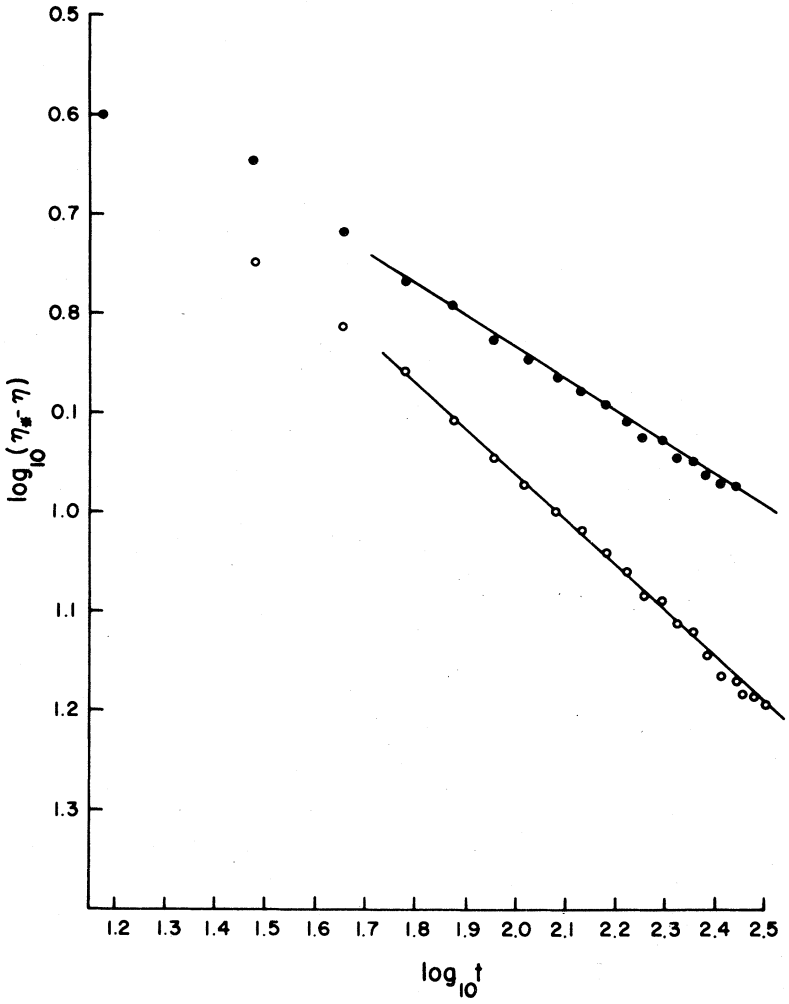

FIG. 7. Log-log plot of $\eta_* - \eta_s(t)$ vs time. The dots correspond to the Onsager value $\eta_* = \eta_\infty = 0.9877$ whereas the open circles refer to the metastable value $N_* = N_m = 0.9506$.

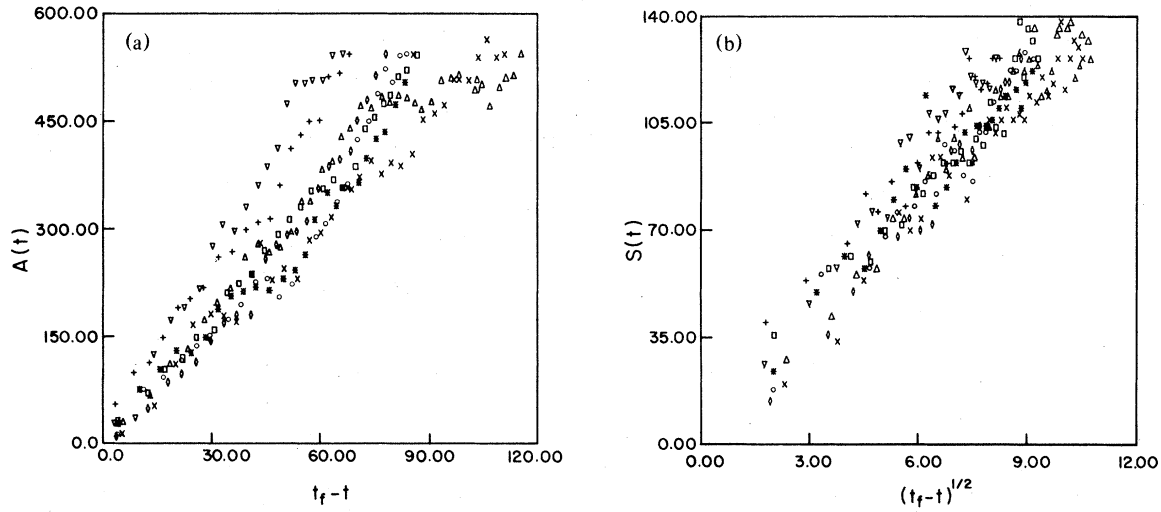

FIG. 8. The evolution of (a) area and (b) perimeter (or surface) of a circular domain as a function of time.

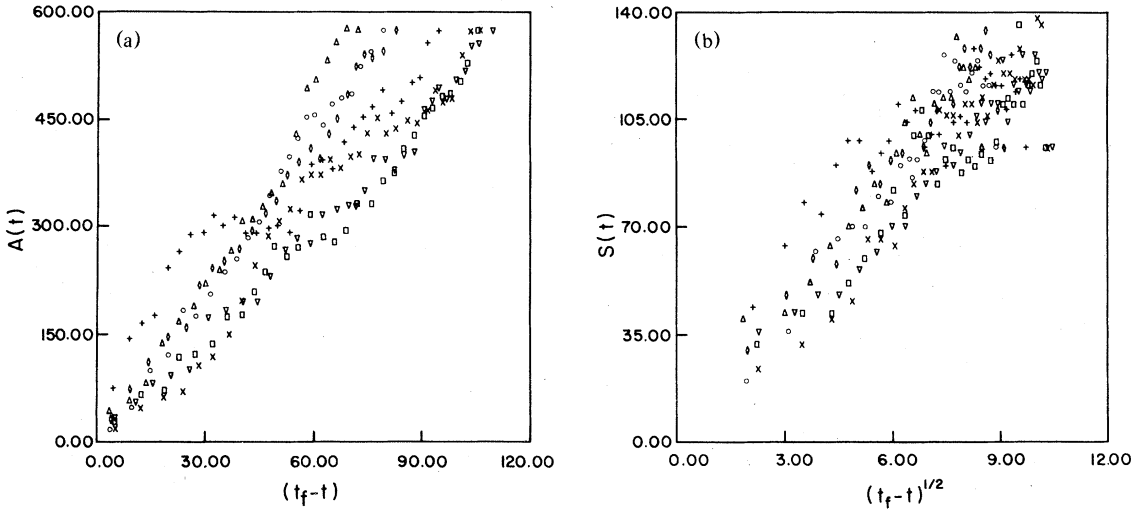

FIG. 9. The evolution of (a) area and (b) perimeter (or surface) of a square domain as a function of time.

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