Soft-Wall Domain-Growth Kinetics of Twofold-Degenerate Ordering

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The domain growth in a two-dimensional twofold-degenerate system with soft domain walls is shown to obey dynamical scaling. The value of the growth exponent is $n \simeq 0.25$ which differs from the classical Lifshitz-Allen-Cahn prediction $n = \frac{1}{2}$, but accords with recent findings for other growth models with soft walls. The results suggest that domain-wall softness may be more important than the degeneracy of the ground state for a possible universal classification of domain-growth kinetics.

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During the search for a universal classification of domain-growth kinetics in nonequilibrium systems undergoing ordering processes, the concept has emerged that only a small number of features may be relevant for the classifying of a great variety of different growth processes. The following tentative picture is suggested by results emanating from an extensive program involving theoretical calculations,[1-7] experimental measurements,[8-12] and computer simulations[13-26] (i) The conservation laws governing the growth process are relevant. (ii) The degeneracy, p, of the ordering may be relevant. (iii) The softness of the domain walls may be relevant. (iv) Irrelevant features include spatial dimension ($d \geqslant 2$), details of the microscopic interactions, and possibly lattice structure.

The most extensively studied case is that of twofold degenerate ordering ($p = 2$) in two-dimensional systems such as binary alloys and simple Ising antiferromagnets. In the event of no conservation laws, theory,[1-3,5,6] experiments,[8,9] and computer simulations[2,13,16,18,21,26] give the unanimous result that the growth is a self-similar process and that the structure factor exhibits dynamical scaling. The characteristic exponent of the associated scaling function assumes the classical value, $n = \frac{1}{2}$, of Lifshitz-Allen-Cahn curvature-driven growth.[27,28] A fundamental property of the models and the experimental systems studied for $p = 2$ is that the domain walls, which are formed during the growth process, are *hard*, i.e., sharply confined in space. Recent computer simulation studies[17,19,23,24] of *soft*-wall models with $p > 2$ have suggested that these may belong to a different universality class characterized by $n \simeq \frac{1}{4}$, independent of the value of p.

The question then naturally arises as to the nature of the growth process in *$p = 2$ systems with soft walls.* Such systems constitute the touchstone for a dual universal classification into classes of systems with hard and soft domain walls. The present paper is the first attempt to answer this question by reporting on a computer-simulation study of the domain-growth kinetics in a two-dimensional $p = 2$ model with soft walls. The results of this study give compelling evidence in favor of the dual universal classification, and that softness, rather than the value of p, is a relevant feature.

The microscopic interaction model of the $p = 2$ soft-wall model is defined by an anisotropic planar rotor Hamiltonian

$$H = J \sum_{i>j}^{(\mathrm{nnn})} \cos(\phi_i - \phi_j) - P \sum_{i>j}^{(\mathrm{nn})} \cos\phi_i \cos\phi_j, \quad (1)$$

where ϕ_i is the polar angle of a classical rotor. The model is arrayed on a square lattice and the coupling parameters J and P are positive. The sums of Eq. (1) include the four next-nearest neighbors and the two nearest neighbors along the x axis, respectively. The uniaxial P term serves to break the cubic symmetry of the interaction resulting in a (2×1) antiferromagnetic ground state described by an $n = 1$-component Ising-type order parameter. There are $p = 2$ thermodynamically degenerate ordered (2×1) domains at low temperatures. The symmetry of the order parameter is similar to that of a binary alloy or type-III Ising antiferromagnet on a rectangular lattice.

The continuous rotor variables of Eq. (1) allow formation at low temperatures of walls of finite thickness (soft walls) between the two types of ordered domains. The value of P controls the softness and thickness of these domain walls: The smaller the value of P/J, the softer and wider are the walls. In this paper, the domain-growth kinetics is studied for a single value of the softness parameter, $P/J = 5$. Results for other values of P are reported elsewhere.[29]

The time evolution of the ordering process governed by Eq. (1) coupled with single-site Glauber dynamics is calculated by a Monte Carlo computer-simulation algorithm.[30] At time $t = 0$, a disordered configuration characteristic of infinite temperature is quenched globally to zero temperature, i.e., far below the phase transition temperature. The time is measured in Monte Carlo steps per site. During the ordering process, the order parameter is a nonconserved quantity. The simulations are carried out on lattices with periodic boundary conditions. Finite-size effects are estimated

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by studying a series of lattice sizes, including $N = 60\times 60, 100\times 100$, and 200×200 rotors. Ensemble averages at each time are obtained by averaging over a large number of independent quenches.

The dynamical evolution following the quench is monitored by calculating, as a function of time, a number of quantities:

(i) The excess energy, $\Delta E(t) = \langle H(t)\rangle - E_0$, with $E_0 = 2J + P$, accounting for the nonequilibrium internal energy stored in the entire network of domain walls.

(ii) The dynamical structure factor

$$S(q,t) = N^{-1}\left\langle \left| \sum_{j=1}^{N} \cos[\phi_j(t)] e^{i(q-\pi)r_j} \right|^2 \right\rangle \quad (2)$$

calculated in the modulated direction, $\mathbf{q} = (q,0)$, around the Bragg condition $q = 0$, as well as its two first moments

$$k_m(t) = \sum_q{}' |q|^m S(q,t) \left[\sum_q{}' S(q,t)\right]^{-1}, \quad m = 1,2, \quad (3)$$

where $q = 2\pi j/\sqrt{N}, j = -\sqrt{N}/2, \ldots, 0, \ldots, \sqrt{N}/2$. The primed sums of Eq. (3) are restricted by an ultraviolet cutoff,[31] $|q| \leq 0.3\pi$. From the moments, two measures of length scale may be obtained, $k_1^{-1}(t)$ and $k_2^{-1/2}(t)$.

(iii) The average linear domain size, $R(t)$, calculated from direct inspection of microscopic configurations.[32] $R(t)$ is a reliable measure of length scale for compact domains. Since the domain pattern for the present $p=2$ model, in analogy with that of $p=2$ Ising models,[20] is highly percolative, conclusions regarding time-dependent length scales will primarily be based on the moments of the dynamical structure factor.[33]

In Fig. 1 $S(q,t)$ is shown for a series of times as obtained from a 100×100 lattice. First of all it is noticed that there is a persistent growth at all times considered. (Some quenches have been taken up to $t = 3000$.) This result is consistent with previous observations[24] that soft-wall models on square lattices do not get pinned at zero temperature in contrast to hard-wall nearest-neighbor Ising models with no conservation laws. The absence of freezing-in behavior of the present model makes it particularly suitable for a discussion of universality.[2] Figure 1 shows, as time elapses, that $S(q,t)$ develops a rounded peak around $q=0$ which gradually becomes more intense and more narrow. $S(q,t)$, which is symmetric, has a shoulder at low $|q|$ due to the finite-size–induced partial symmetry breaking at $q \simeq 0$.[33] The nonsmooth behavior of $S(q,t)$ for small q reflects the incomplete averaging of the satellites present in $S(q,t)$ of each individual quench. A complicated pattern of satellites arises because of scattering from the inhomogeneous domain-wall network which lacks supersymmetries. Similar shoulders on $S(q,t)$ occurring at the same relative

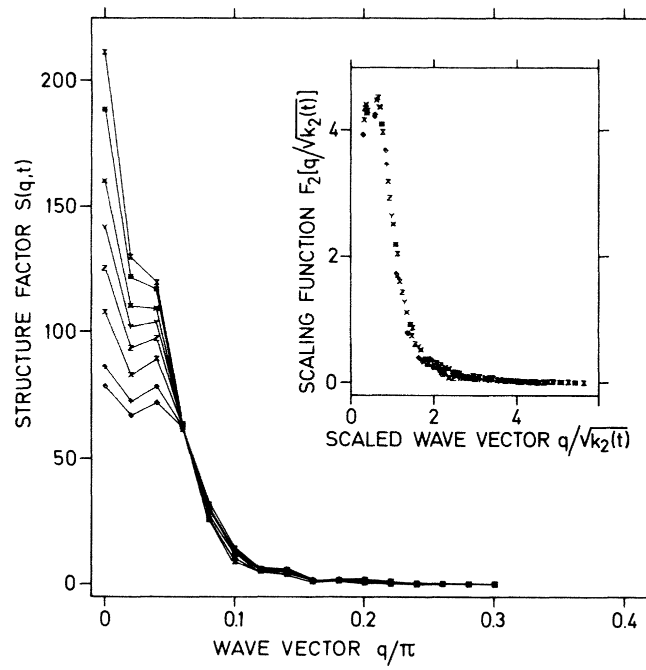

FIG. 1. Dynamical structure factor $S(q,t)$, Eq. (2), at times $t = 80$, 100, 150, 200, 250, 300, 400, and 500. Inset: The corresponding dynamical scaling function, $F_2(x)$, Eq. (4). (Data for a 100×100 lattice.)

wave vectors are seen in dynamic studies of other growth models,[13,14,18,26] including $p=2$ hard-wall Ising models.

The dynamical scaling properties of the growth process are investigated by study of the scaling functions

$$F_m(x) = k_m^{2/m} S(q,t), \quad m = 1,2, \quad (4)$$

where $x = qk_m^{-1/m}$ is the scaling variable. The inset in Fig. 1 demonstrates that, by means of the scaling variable $x = qk_2^{-1/2}(t)$, the entire set of data for $S\times(q, t \gtrsim 80)$ may be collapsed into a single function $F_2(x)$. A similar statement holds for $F_1(x)$.[29] Thus the growth process obeys dynamical scaling. Because of difficulties with the statistics at $q \approx 0$, we are unable to confirm the scaling property at small wave vectors. Similar difficulties are encountered in studies of the dynamical scaling function for $p=2$ hard-wall Ising models.[18]

Dynamical scaling of $S(q,t)$ implies that the characteristic length scales of the growth process are described by power laws $k_m^{-1/m}(t) \sim t^n$ with n being the kinetic exponent. This is clearly borne out by Fig. 2(a) which shows a log-log plot of $k_1^{-1}(t)$ and $k_2^{-1/2}(t)$ vs time. Both length scales are accurately described by power laws with approximately the same exponent, $n \simeq 0.25$. The power laws are found to hold for $t \gtrsim 20$. For comparison, Fig. 2(a) reproduces the corresponding growth data for $k_2^{-1/2}(t)$ in the case of the $p=2$ hard-wall Ising model.[34] The growth process

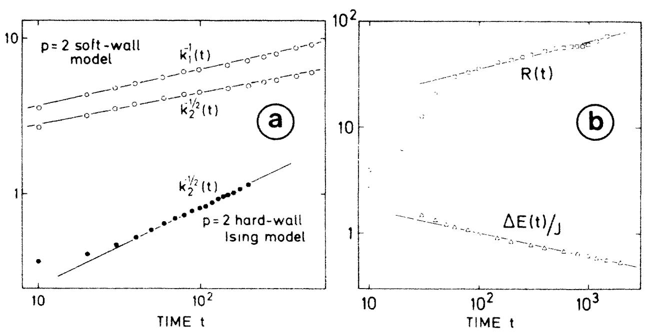

FIG. 2. (a) Log-log plot vs times of length scales derived from moments of the structure factor, Eq. (3). Results from quenches of the $p=2$ hard-wall Ising model (Ref. 18) are shown for comparison. The solid lines denote power laws, $k_m^{-1/m} \sim t^n$, with $n=0.25$ and $n=0.50$ for the soft-wall and hard-wall models, respectively. (b) Log-log plot vs time of the linear domain extension, $R(t)$, and the excess energy per rotor, $\Delta E(t)$. The solid lines denote the asymptotic power laws, $R(t) \sim t^n$ and $\Delta E(t) \sim t^{-n}$, with the same exponent $n \simeq 0.2$. (Data for a 200×200 lattice.)

in the soft-wall model may reliably be followed to later times than in the Ising model because of a much slower growth kinetics in the former model. In fact, as is apparent from Fig. 2(a), the hard-wall kinetics is nicely described by the much larger classical Lifshitz-Allen-Cahn exponent value, $n \simeq 0.50$.

When dynamical scaling holds, the excess energy, $\Delta E(t)$, measuring the total internal energy stored in the domain walls is expected to decay with time according to a power law, $\Delta E(t) \sim t^{-n}$, where the exponent assumes the same value as that describing the growth-of-length scale.[16] That this is indeed the case for the present growth process is demonstrated in Fig. 2(b). In the same figure, the data displayed for the linear domain extension, $R(t)$, are seen to corroborate the notion that also $R(t)$ is asymptotically described by the same power law, $R(t) \sim t^n$, with $n \simeq 0.25$.

In conclusion, it is found that the domain-growth process of the soft-wall model in Eq. (1) obeys dynamical scaling and that the various measures of length scale are consistently described by power laws in time with the *same* growth exponent, $n \simeq 0.25$. A fundamental property of this growth model is that during the ordering process it supports formation of soft domain walls extending over finite regions of space. The results described in this paper refer to a single value of the softness parameter. However, identical results are found for $0.5 \lesssim P \lesssim 50$,[29] i.e., $S(q,t)$ obeys dynamical scaling and the various growth laws are associated with the same value of the growth exponent, $n \simeq 0.25$. It thus appears that the class of $p=2$ soft-wall models defined by Eq. (1) gives rise to a kinetic exponent which is the same as that found for all other studied two-dimensional soft-wall models characterized by a number of different values of the order parameter degeneracy: $p=4$,[23] $p=6$,[17] and $p=48$.[20] This ensemble of findings supports the hypothesis that the kinetics of domain growth in soft-wall systems with a nonconserved order parameter belongs to a separate universality class characterized by a kinetic exponent value $n \simeq \frac{1}{4}$ which is distinctly lower than the classical Lifshitz-Allen-Cahn prediction, $n=\frac{1}{2}$. The results of the present paper suggest that the value of p is irrelevant for this classification even in the case of a binary ($p=2$) system. It should be noted that a soft domain wall in the rotor model corresponds to a spatial gradient of the order parameter. In the lattice-gas formulation of the analogous adsorption problem, a soft wall would imply a region where the adsorbed layer is out of registry with the substrate, i.e., the surface tension of the wall is lowered by relaxation in terms of translational degrees of freedom. It is therefore anticipated that soft-wall kinetics should apply for ordering phenomena in physisorbed rather than chemisorbed overlayers.

It is notoriously difficult to construct quantitive theories of domain growth for $p>2$,[4,7] whereas a number of successful theories have been advanced in the case of $p=2$.[1–3,5,6,27,28] The results of the present paper suggest that, in the search for a universal classification scheme of domain-growth kinetics, one should reexamine the theories for $p=2$ with a view to clarifying whether the kinetic behavior is changed in an essential manner when, on its way toward equilibrium, the system is allowed to relax locally, not only by boundary migration, but at the same time by domain-wall broadening.

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[1]For a comprehensive review of the field of nonequilibrium phase transformations, see J.D. Gunton, M. San Miguel, and P. S. Sahni, in *Phase Transitions and Critical Phenomena,* edited by C. Domb and J. Lebowitz (Academic, New York, 1983), Vol. 8.

[2]F. C. Zhang, O. T. Valls, and G. F. Mazenko, Phys. Rev. B **31**, 1579 (1985).

[3]G. F. Mazenko, O. T. Valls, and F. C. Zhang, Phys. Rev. B **31**, 4453 (1985).

[4]S. A. Safran, Phys. Rev. Lett. **46**, 1581 (1981).

[5]G. F. Mazenko and O. T. Valls, Phys. Rev. B **27**, 6811 (1983), and **30**, 6732 (1984).

[6]T. Ohta, D. Jasnow, and K. Kawasaki, Phys. Rev. Lett. **49**, 1223 (1982).
[7]H. Furukawa, Phys. Rev. A **30**, 1052 (1984).
[8]G.-C. Wang and T.-M. Lu, Phys. Rev. Lett. **50**, 2014 (1983).
[9]P. K. Wu, J. H. Perepezko, J. T. McKinney, and M. G. Lagally, Phys. Rev. Lett. **51**, 1577 (1983).
[10]H. Homma and R. Clarke, Phys. Rev. Lett. **52**, 629 (1984).
[11]O. Blaschko and P. Fratzl, Phys. Rev. Lett. **51**, 288 (1983).
[12]S. Katano and M. Iizumi, Phys. Rev. Lett. **52**, 835 (1983).
[13]P. S. Sahni, G. Dee, J. D. Gunton, M. Phani, J. L. Lebowitz, and M. Kalos, Phys. Rev. B **24**, 410 (1981).
[14]P. S. Sahni and J. D. Gunton, Phys. Rev. Lett. **47**, 1754 (1981).
[15]S. A. Safran, P. S. Sahni, and G. S. Grest, Phys. Rev. B **28**, 2693 (1983); P. S. Sahni, D. J. Srolovitz, G. S. Grest, M. P. Anderson, and S. A. Safran, Phys. Rev. B **28**, 2705 (1983).
[16]A. Sadiq and K. Binder, J. Stat. Phys. **35**, 517 (1984).
[17]O. G. Mouritsen, Phys. Rev. B **28**, 3150 (1983).
[18]K. Kaski, M. C. Yalabik, J. D. Gunton, and P. S. Sahni, Phys. Rev. B **28**, 5263 (1983).
[19]G. S. Grest, D. J. Srolovitz, and M. P. Anderson, Phys. Rev. Lett. **52**, 1321 (1984).
[20]G. S. Grest and D. J. Srolovitz, Phys. Rev. B **30**, 5150 (1984).
[21]E. T. Gawlinski, M. Grant, J. D. Gunton, and K. Kaski, Phys. Rev. B **31**, 281 (1985).
[22]K. Kaski, J. Nieminen, and J. D. Gunton, Phys. Rev. B **31**, 2998 (1985).
[23]O. G. Mouritsen, Phys. Rev. B **31**, 2613 (1985).
[24]O. G. Mouritsen, Phys. Rev. B **32**, 1632 (1985).
[25]E. T. Gawlinski, K. Kaski, M. Grant, and J. D. Gunton, Phys. Rev. Lett. **53**, 2266 (1985).
[26]J. Viñals and J. D. Gunton, Surf. Sci. **157**, 473 (1985).
[27]I. M. Lifshitz, Zh. Eksp. Teor. Fiz. **42**, 1354 (1962) [Sov. Phys. JETP **15**, 939 (1962)].
[28]S. M. Allen and J. W. Cahn, Acta Metall. **27**, 1085 (1979).
[29]O. G. Mouritsen, to be published.
[30]O. G. Mouritsen, *Computer Studies of Phase Transitions and Critical Phenomena* (Springer, Heidelberg, 1984).
[31]Other cutoffs have been considered to assure that the cutoff chosen does not significantly influence the values of the moments.
[32]The radius, $R(t)$, of a domain is determined as the square root of the number of rotors in the domain. A rotor is considered as part of a domain if its angle deviates less than $\pi/15$ from the ground-state angles of that domain; see also Ref. 23.
[33]A third measure of length scale may be derived from the intensity of the Bragg peak, $L(t) = [S(0,t)/N]^{1/2}$ (see Ref. 16). $L(t)$ determines the degree of symmetry breaking between the two ground states. For a finite $p=2$ system, $L(t)$ is subject to large fluctuations from quench to quench and is therefore difficult to estimate accurately. Consequently, the present model study only uses length scales obtained from nonzero wave vector components of the structure factor.
[34]The data of Ref. 18 refer to a quench of the antiferromagnetic Ising model with Glauber dynamics to a temperature 40% below the critical temperature. The hard-wall limit is pinned on the square lattice at zero temperature.