PHYSICAL REVIEW B
CONDENSED MATTER

THIRD SERIES, VOLUME 31, NUMBER 5 — 1 MARCH 1985

Relevance of domain-wall softness for a universal classification of domain-growth kinetics

Ole G. Mouritsen

Department of Chemistry, Aarhus University, DK-8000 Aarhus C, Denmark

(Received 10 December 1984)

The influence of domain-wall softness on the kinetics of growth is studied within a two-dimensional microscopic model which supports domain walls of variable softness. The domain radius grows with time as $R(t)\sim t^n$, where the kinetic exponent is found to assume a value, $n\simeq 0.25$, which is independent of the degree of softness. This suggests a dual universal classification of domain-growth kinetics into classes of systems with soft and hard walls.

Formation and growth of aggregates and ordered structures from the microscopic constituents of matter are widely observed processes in nature. Among these processes is the growth of ordered domains of molecules as it takes place after a thermal quench below a phase transition temperature. The kinetics of domain growth is currently subject to a resurging interest[1–11] prompted by the indication that some unifying principle may be operative for the growth processes in a great variety of different systems. It is now anticipated that the kinetics of growth may be arranged into a small number of universality classes.[6,7,11] So far, neither has the unifying principle been discovered nor have the relevant parameters for the classification been determined unambiguously.

In some recent model studies[3,6] it has been suggested that the degeneracy p of the ordered phase as well as the conservation laws governing the growth process are important for a possible universal classification of growth. That these two parameters, however, are insufficient for a complete classification is indicated by two computer-simulation studies[4,7] which demonstrate that systems governed by the same conservation laws and described by the same value of p may have markedly different kinetic exponents. The systems employed in these two studies are distinguished by their capacity of supporting *soft* domain walls, in contrast to the commonly studied Ising[1,6] and Potts models[3] which give rise to *hard* walls only.

In this work I present the results derived from a computer-simulation study of a two-dimensional model specifically constructed to allow a systematic investigation of the influence of domain-wall softness on the domain-growth kinetics. The model, which has a fourfold-degenerate ($p=4$) ground state, contains a parameter which controls the softness and the thickness of the domain walls. The results of the study provide mounting evidence that domain-wall *softness* is relevant for a universal classification of kinetic exponents. In particular, it is shown that the kinetic exponents are independent of the *degree* of softness, except in the limit of vanishing softness where a distinct crossover to the well-known hard-wall behavior is observed. The kinetic exponents of the soft-wall and hard-wall universality classes are markedly different, i.e., $n\simeq 0.25$ and 0.50, respectively.

The two-dimensional microscopic interaction model, on which the present study is based, is defined by the anisotropic Hamiltonian

$$\mathscr{H}=J^{\mathrm{NN}}\sum_{i>j}^{(\mathrm{NN})}(\mathbf{r}_{ij}\cdot\mathbf{S}_i)(\mathbf{r}_{ij}\cdot\mathbf{S}_j)+J_{\mathrm{NNN}}\sum_{i>j}^{(\mathrm{NNN})}\mathbf{S}_i\cdot\mathbf{S}_j-P\sum_i(S_{ix}^4+S_{iy}^4)\,, \tag{1}$$

where $J^{\mathrm{NN}}=J^{\mathrm{NNN}}\equiv J>0$ and $P>0$ are model parameters. The model is arrayed on a square lattice. $\mathbf{S}_i=(S_{ix},S_{iy})=(\cos\varphi_i,\,\sin\varphi_i)$ is a classical planar spin vector. The repulsive pair interactions serve to produce (2×1) antiferromagnetic ground states of which the J^{NN} term stabilizes structures with the propagation vector parallel to the sublattice magnetization. The single-site crystal-field-like term P favors spin orientations along the two axes of the square lattice. The ordered phase of the model is described by a two-component order parameter (ψ_1,ψ_2). There are $p=4$ thermodynamically degenerate ordered (2×1) domains a low temperatures (see insert of Fig. 1). The symmetry of the model is that of the planar XY model with cubic anisotropy.[12] Several adsorbed monolayer systems are characterized by this symmetry, e.g., oxygen on W(110).

The continuous nature of the spin variables enables the system to form soft walls between domains of different types of ordering (cf. inset of Fig. 1). The P term monitors the softness and thickness of these walls: Small values of P/J facilitate the formation of very soft and thick walls; large values of P/J lead to hard and thin walls. The model is studied here over two decades of the softness parameter, $10^{-1}\leq P/J\leq 10$.

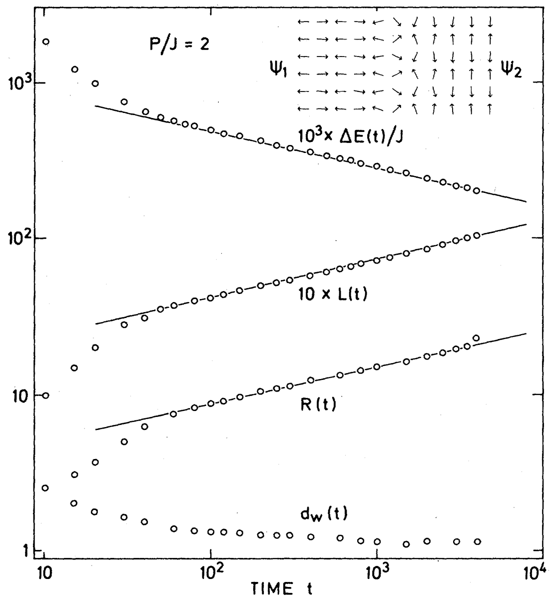

FIG. 1. Log-log plot vs time t of the linear domain extensions, $R(t)$ and $L(t)$, of the excess energy per spin $\Delta E(t)$ and of the domain-wall thickness $d_w(t)$ for softness parameter $P/J=2$. The time is in units of Monte Carlo steps per site. The solid lines denote the asymptotic power laws, Eq. (3), with the same kinetic exponent, $n=0.24$. The inset shows a low-temperature spin configuration with a soft wall between two ordered domains described by the order parameter components ψ_1 and ψ_2 of the 2×1 structure.

I have used a conventional Monte Carlo method[13] to construct the evolution of the model following a deep quench in temperature from $T\simeq\infty$ to $T\simeq 0$. The excitation mechanism is of the Glauber type involving single-site random reorientations of the spins. Thus, the order parameter is a nonconserved quantity. The main results are derived from a lattice with $N=100\times 100$ sites subjected to toroidal periodic boundary conditions. To make sure that the results are not invalidated by finite-size effects, a number of simulations have been performed on a lattice with 150×150 sites. Each quench is performed several times using different initial configurations and different random-number sequences. Ensemble averages are obtained by averaging over the independent quenches. The evolution of the system is followed for times up to 4000 Monte Carlo steps site.

Visual inspection of snapshots showing spin configurations taken at successive times after the quench reveals that the domain-wall thickness usually decreases with time. For a fixed time, the softer the potential, the wider and softer are the walls. The morphology of the domain pattern also depends on P. The softer the walls, the more spongy and elongated are the domains. A compact and regular domain pattern emerges when the walls get harder and thinner.

The domain-growth process is analyzed quantitatively by calculating, as a function of time t, two different measures of the characteristic length scale of the growth process. The first one is the average domain radius, $R(t)$. The radius of a domain is simply determined as the square root of the number of spins in the domain. A spin is defined as belonging to a domain if its directional angle deviates less than $\delta\varphi=\pi/15$ from the ground-state angles of the domain in question.[14] The second measure of length scale, $L(t)$, is obtained as[6]

$$L(t)=\{N[\psi_1^2(t)+\psi_2^2(t)]\}^{1/2}\psi(T)\ , \tag{2}$$

where $\psi(T)\simeq 1$ is the equilibrium value of the order parameter. $L(t)$ remains a useful measure of domain size only in the cases where the number of spins in the walls, $N_w(t)$, is small compared to N. From the total perimeter, $l(t)=2\sqrt{\pi}R(t)N_d(t)$, of the $N_d(t)$ domains occurring at time t, an average domain-wall thickness may be defined,

$$d_w(t)=2N_w(t)/l(t)=N_w(t)/\sqrt{\pi}R(t)N_d(t)\ .$$

Finally, the excess energy, $\Delta E(t)=\langle H(t)\rangle-\langle H(T)\rangle$, is calculated. $\Delta E(t)$ is a measure of the internal energy associated with the entire network of domain walls. If dynamical scaling holds in the late stages of the growth process, the growth laws are expected to be algebraic in time,[6,15] i.e.,

$$R(t)\sim t^a,\quad L(t)\sim t^b,\quad \Delta E(t)\sim t^{-c}\ . \tag{3}$$

Scaling implies $a=b=c\equiv n>0$.

Figure 1 shows a log-log plot of the time evolution of $R(t)$, $L(t)$, $\Delta E(t)$, and $d_w(t)$ in the case of $P/J=2$. After a transient period of about 70 Monte Carlo steps per site, the growth is seen to be described accurately by all three algebraic relations in Eq. (2) with the *same* exponent, $n\simeq 0.24$. Thus the growth process obeys dynamical scaling. The domain-wall thickness is found to decrease with time and to approach a limiting value, $d_w(\infty)\simeq 1.2$, which is characteristic for the late-time regime of the growth process for $P/J=2$. Figure 2 gives $\Delta E(t)$ for the whole range of values of the softness parameter. From these data, and the corresponding data for $R(t)$ and $L(t)$, it is observed that the growth is algebraic in time for $P/J\leq 7$ and that scaling is obeyed.

The kinetic exponent extracted from plots like those presented in Figs. 1 and 2 is given in Fig. 3 as a function of softness P. Figure 3 also shows the late-time domain-wall thickness $d_w(\infty)$ as a function of P. From this figure, the striking observation is made that, although the

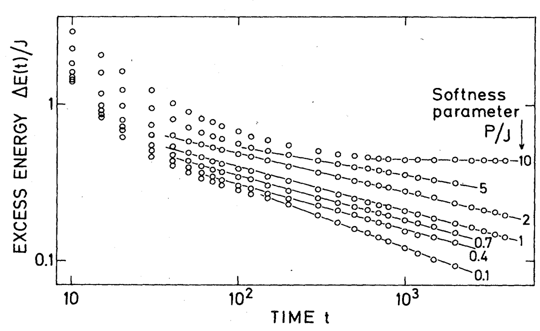

FIG. 2. Log-log plot of the excess energy per spin, $\Delta E(t)$, as a function of time t for different values of the softness parameter P/J. The time is in units of Monte Carlo steps per site. The solid lines denote asymptotic power laws with kinetic exponents as given in Fig. 3.

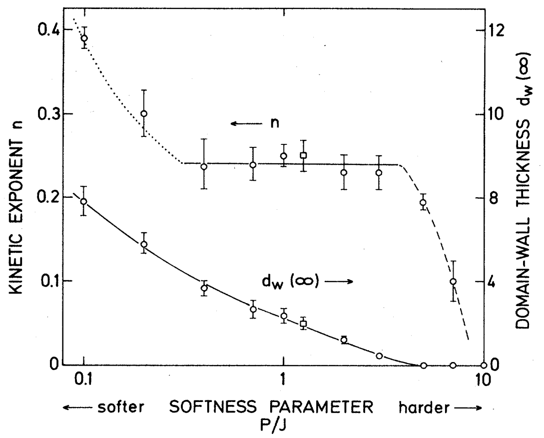

FIG. 3. Kinetic exponent n and late-time domain-wall thickness, $d_w(\infty)$, as functions of the softness parameter P/J. The solid line is a guide to the eye. The dotted line denotes early-time behavior in the extreme soft-wall limit. The dashed line indicates crossover to hard-wall low-temperature kinetic behavior. Circles indicate results for the model in Eq. (1). Squares refer to the anisotropic-planar-rotor model governing herringbone kinetics (Ref. 4). The horizontal axis is logarithmic.

late-time domain-wall thickness varies more than an order of magnitude, *the kinetic exponent is independent of P* in the decade $0.3 \lesssim P/J \lesssim 3$. The value of the exponent is $n \simeq 0.24 \pm 0.03$.

For $P/J < 0.3$, the exponent is gradually increasing. This is not likely to be the true late-time behavior since $d_w(t)$ is increasing with time in this range caused by a high frequency of coalescence processes. Much larger lattice sizes are required to obtain reliable information for $t > 2000$ when P is very small because the largest domain tends to percolate before the different domains begin to compete. The walls are so wide and soft that the domain-domain interactions are screened completely. The resulting growth is therefore fast and characteristic of "early times" where the nucleated domains are growing and coalescing independently in an "elastic medium of soft spins." Long runs, in which percolation has not yet set in, indicate that there is a crossover to a lower exponent for $t \gtrsim 2000$. This may suggest that the independence of n on the degree of softness also extends to the very soft regime.

For $P/J > 3$, a distinct crossover to a different behavior is observed. This is characteristic of the low-temperature growth of models with hard domain walls. For $P/J \gtrsim 10$, the growth is gradually slowed down and finally stops. The resulting quenched state is a metastable glasslike frozen-in configuration of domains. Similar observations have been reported for deep quenches of $p = 4$ Ising[6] and Potts[3] models on square lattices. For all three models, the lattice structure makes the systems get trapped in a metastable domain state and the true long-time kinetics is not probed.

The finding in quenched systems with soft domain walls of a kinetic exponent which is independent of details within the model [i.e., P and $d_w(\infty)$] corroborates the concept of universality in domain-growth kinetics. Obviously, it suggests that models which support soft domain walls belong to the same universality class. The general validity of this statement is supported by the recent finding of $n \simeq 0.25$ for the domain-growth kinetics of herringbone phases ($p = 6$) (Ref. 4) governed by a two-dimensional anisotropic-planar-rotor model,[16] a model which also gives rise to soft walls. The results of both n and $d_w(\infty)$ for this model fit nicely on the curves in Fig. 3.[17] Furthermore, Grest *et al.*[7] have recently studied a class of models on triangular lattices with high values of p ($p = 48$) and with interaction potentials which facilitate the formation of "wide domain boundaries" composed by a large number of small domains. These boundaries may be considered as wide and soft domain walls. For a series of models in the wide-boundary limit, Grest *et al.* also find $n \simeq 0.25$ independent of details within the models.

All this evidence suggest that the domain-growth kinetics of models with soft walls belongs to a single universality class characterized by $n \simeq 0.25$. Moreover, softness seems to be more relevant for a universal classification than the value of p, i.e., more relevant than the topology of the domain-wall network. In particular, pinning effects for $p > d$,[2,3] which on the square lattice are found to decrease dramatically the driving force for the growth in models with hard walls,[3] appear to be superseded by domain-wall softness which facilitates migration of the pinning centers.

The universal kinetic exponent of soft-wall kinetics, $n \simeq 0.25$, is markedly different from the classical exponent, $n \simeq 0.50$, found for Ising-type hard-wall kinetics with a nonconserved order parameter.[1,6] The much slower kinetic behavior for models with soft domain walls is probably due to a screening of the domain-domain interactions in the late states of the growth.[4] This screening decreases the driving force of the growth.

The author was supported by A/S De Danske Spritfabrikker Jubilaeumslegat.

[1]M. K. Phani, J. L. Lebowitz, M. H. Kalos, and O. Penrose, Phys. Rev. Lett. **45**, 366 (1980).

[2]S. A. Safran, Phys. Rev. Lett. **46**, 1581 (1980).

[3]P. S. Sahni, G. S. Grest, M. P. Anderson, and D. J. Srolovitz, Phys. Rev. Lett. **50**, 263 (1983); P. S. Sahni, D. J. Srolovitz, G. S. Grest, M. P. Anderson, and S. A. Safran, Phys. Rev. B **28**, 2705 (1983).

[4]O. G. Mouritsen, Phys. Rev. B **28**, 3150 (1983).

[5]K. Kaski, S. Kumar, J. D. Gunton, and P. A. Rikvold, Phys. Rev. B **29**, 4420 (1984).

[6]A. Sadiq and K. Binder, Phys. Rev. Lett. **51**, 674 (1983); J. Stat. Phys. **35**, 517 (1984).

[7]G. S. Grest, D. J. Srolovitz, and M. P. Anderson, Phys. Rev. Lett. **52**, 1321 (1984).

[8]S. M. Allen and J. W. Cahn, Acta Metall, **27**, 1085 (1979).

[9]G.-C. Wang and T.-M. Lu, Phys. Rev. Lett. **50**, 2014 (1983).

[10]O. Blaschko and P. Frantzl, Phys. Rev Lett. **51**, 288 (1983).

[11]J. D. Gunton, M. San Miguel, and P. S. Sahni, in *Phase Tran-*

sitions and Critical Phenomena, edited by C. Domb and J. L. Lebowitz (Academic, New York, 1984), Vol. VIII, p. 267.

[12]E. Domany and E. K. Riedel, Phys. Rev. Lett. **40**, 561 (1978).

[13]O. G. Mouritsen, *Computer Studies of Phase Transitions and Critical Phenomena* (Springer, Heidelberg, 1984), Chap. 5.

[14]The choice of a discrimination angle of $\delta\varphi=\pi/15$ is somewhat arbitrary and naturally the absolute values of $d_w(t)$ and $R(t)$ depend on $\delta\varphi$. However, it results that the asymptotic growth law for $R(t)$ is independent of $\delta\varphi$ for a wide range of small values of $\delta\varphi$.

[15]K. Binder and D. Stauffer, Phys. Rev. Lett. **33**, 1006 (1974).

[16]O. G. Mouritsen and A. J. Berlinsky, Phys. Rev. Lett. **48**, 181 (1982).

[17]There is no unambiguous way of identifying a softness parameter of the anisotropic-planar-rotor Hamiltonian of Ref. 16. One simple possibility is to equate angular derivatives of the Hamiltonian evaluated in a ground state. This leads to $P/J\simeq 5/4$.