Kinetics of slow domain growth: The $n = 1/4$ universality class

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The domain growth after a quench to very low, finite temperatures is analyzed by scaling theory and Monte Carlo simulation. The growth exponent for the excess energy $\Delta E(t) \sim t^{-n}$ is found to be $n \sim \frac{1}{4}$. The scaling theory gives exactly $n = \frac{1}{4}$ for cases of hierarchical movement of domain walls. This explains the existence of a slow growth universality class. It is shown to be a singular Allen-Cahn class, to which belongs systems with domain walls of both exactly zero and finite curvature. The model studied has continuous variables, nonconserved order parameter, and has two kinds of domain walls: sharp, straight, stacking faults and broad, curved, solitonlike walls.

The kinetics of domain growth is of relevance for the formation of polycrystalline microstructure which is of considerable importance in surface science,[1] metallurgy,[2] and earth science.[3] A possible universal classification of the kinetics of domain growth after a quench from high temperatures to a low temperature ordered phase has been under vivid discussion in recent years.[4] For the case of nonconserved order parameter, the excess energy ΔE of the domain wall network is usually expected to decay algebraically as $\Delta E \sim t^{-n}$ with $n = \frac{1}{2}$ according to the Allen-Cahn theory[5] for curvature driven growth. A possible deviation from this behavior yielding $n \sim \frac{1}{4}$ was first found by Mouritsen[6] by computer simulation on an anisotropic system with continuous variables and order parameter degeneracy $p = 2$. It was subsequently found by Grest *et al.*[7] that a number of generalized p-state "Potts" models with wide low angle domain wall for sufficiently high p also gave $n \sim \frac{1}{4}$, and the possibility of a new universality class was proposed. The finding[6] of the small exponent $n \sim \frac{1}{4}$ was disputed as being an artifact of inadequate data analysis[8] or a special effect of the applied zero temperature Monte Carlo method.[9] However, further extensive numerical simulations have been performed[10] on different anisotropic models with continuous variables and $p = 2$. These corroborate conclusively the existence of a new, slow growth class with $n = \frac{1}{4}$ for quenches to very low, finite temperatures. It was first suggested by Mouritsen[6] that the deviation from $n = \frac{1}{2}$ in the investigated systems indicated a breakdown of the basic assumptions in the Allen-Cahn theory[5] in the presence of broad, "soft" walls, which might screen the interaction between domains.[10] This argument was disputed by Ref. 8 and by van Saarloos and Grant[9] who firstly showed that even if the walls were broad, the growth should follow $n = \frac{1}{2}$. They pointed out that this was indeed observed experimentally.[11] Secondly they showed that in the model studied by Mouritsen the walls were in fact only partly broad, since they were sharp in some spatial directions. We agree with this observation.

It is the aim of this paper to show the *raison d'être* for the unexpected slow growth class. This insight was obtained by analyzing a model which is quite different from the ones studied by Mouritsen *et al.*[6,10] But the domain walls have the same feature, consisting of a mixture of interconnected broad and sharp walls. We shall now demonstrate that it is this *mixture* which is the cause. A scaling theory shows that the exponent is exactly $n = \frac{1}{4}$. As was also found in the previous studies[6,10] the softness of the walls is not crucial as such, except for making the walls able to curve easily. The reason is the following. A soft wall is well modeled by a solitons like shape with a width w and a phase ϕ describing the soliton maximum relative to the lattice positions. The energy and width of the soliton depends only weakly on the phase ϕ. Therefore a wall consisting of neighboring solitons can curve continuously with relatively little energy cost. In contrast a sharp straight wall can only "curve" by the introduction of a kink. This costs considerable energy. However, once formed the kink can move freely and fast, whereas the soliton wall moves slower since it involves several particles. At sufficiently low temperatures no kinks can be created by thermal fluctuations and the existing kinks will be trapped by the soliton walls. The system then consists of curved walls connected by straight walls. This situation is a singular case for the Allen-Cahn theory. Whereas the basic assumptions still hold, the exponent is nonetheless $n = \frac{1}{4}$. The slowing down is due to a temporal pinning of the straight (zero curvature) walls, which cannot move until their extent is sufficiently small. This pinning effect is already present for an order-parameter degeneracy $p = 2$, corresponding to only two types of equivalent domains. Let us consider a magnetic model with continuous spin variables with twofold order-parameter degeneracy ($p = 2$) in a two-dimensional x-y lattice ($d = 2$). We use the Hamiltonian introduced recently[12] for simulating a Martensitic transformation,

$$H = \sum_{\langle i,j \rangle} \{ -K S_{iz} S_{jz} + J [\mathbf{S}_i \cdot \mathbf{S}_j - P(\hat{\mathbf{r}}_{ij} \cdot \mathbf{S}_i)(\hat{\mathbf{r}}_{ij} \cdot \mathbf{S}_j)] \} - D \sum_i (S_{ix}^4 + S_{iy}^4) \; . \quad (1)$$

This is simplified here by confining the classical spins to only the upper half of the x-z plane; we use $D=2J$ as in Ref. 12. The dipolar term J favors an antiferromagnetic state with twofold degeneracy and the K term of ferromagnetic state. For $K/J<2.9$ the $p=2$ antiferromagnetic phase is the equilibrium state at low temperatures; at high temperature the equilibrium state is a one domain ferromagnetic state. The antiferromagnetic state consists of antiparallel ferromagnetic chains with an interaction $P-1$ times stronger along the chains than between the chains, i.e., twice when we choose the dipole parameter $P=3$. This anisotropy is essential for producing different kinds of domain walls. The wall for a mismatch along the chains is a broad soliton involving 6–7 spins canting continuously in the x-z plane. The wall parallel to the chains is sharp involving only two spins. This is a stacking fault in the sequence of chains. The energy cost for a curvature of the broad wall is small (a few percent of J) and proportional to the curvature, whereas the energy cost for a kink on the sharp wall is $4J$ at the chosen ratio $K/J=2.3$. The cost of a unit length of the broad wall (a soliton) is approximately $4J$, whereas the cost of a unit length of the sharp boundary is only $2J$.

We have made extensive Monte Carlo computer simulations on this simple model studying the domain growth after rapid temperature quenches from the ferromagnetic phase to the $p=2$ phase at low, finite temperatures (0.01 of $T_N\sim 2J/k_B$). The details are reported elsewhere.[13] We choose to follow the behavior of the self-averaging[4] excess energy. The principal results are shown in Fig. 1, proving that the time evolution at late times is algebraic with a small exponent $n\sim\frac{1}{4}$. This is the same result as found by Mouritsen *et al.*[6,10] We also observe a rapid crossover at about 2000 Monte Carlo steps per site (MCS) to an initial faster decay regime for ΔE as in Ref. 6, with an appearant exponent $n\sim\frac{1}{2}$. This has nothing to do with an Allen-Cahn behavior, but is essentially due to an exponentially fast optimization of the domain wall width. Longer runs[13] indicate that finite size effects manifest themselves at much later times at $t\sim 20\,000$ MCS for a 200×200 size system. All runs included in Fig. 1 will eventually evolve into a single domain, stable state with no domain walls (other runs, not shown, may be trapped in metastable slab configurations[14] due to finite size effects and the periodic boundary condition imposed). The conclusion is that a clear late time regime is found with an algebraic decay $\Delta E\sim t^{-n}$ with $n\sim\frac{1}{4}$.

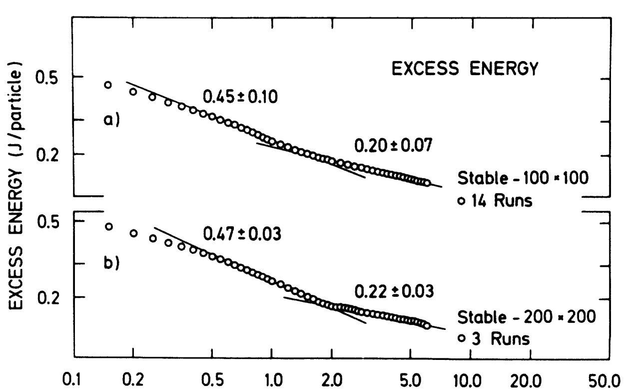

FIG. 1. Excess energy vs time in Monte Carlo steps per site (MCS).

The simple model can be analyzed and we will show that all assumptions made in the Allen-Cahn theory are fulfilled, yet giving a smaller exponent. Let us first consider the behavior of a typical domain, which is given the index "o". It is shown in the inset of Fig. 2. We notice the broad, curved walls (called C walls) predominantly in the vertical y direction in the x-y plane. The spins in the indicated solitons are in the x-z plane having a z component larger than 0.15. The ellipsoidal domain is terminated by sharp, straight walls (called S walls) shown as the limiting spins of each domain. The ordered spins are not shown. The number of solitons in the C wall is a measure for the length projected on the vertical direction, called L_C. This dominates the contribution to the excess energy, whereas the energy of the curvature is negligible. The length L_C^+ of the upper half of the ellipsoid decreases as a function of time (MCS) in a steplike fashion, but on the average as square root $(t_0-t)^{1/2}$, whereas the area (in the whole time interval) decreases linearly as $\alpha(t_0-t)$; t_0 is the time at which the domain "o" disappears. The corresponding length L_C^- of the lower part of the ellipsoid is pinned by the S wall until $t\sim 25\,000$ MCS. Up to this time the C walls move towards each other with a velocity proportional to their curvature, eliminating the S wall at $t\sim 25\,000$ MCS. In this process the area decreases again linearily as $\alpha(t_0-t)$ with the same coefficient α and consistently the length of the S wall decrease as $L_S\sim(t_0-t)$. For later times the lengths L_C^+ and L_C^- decrease both as $(t_0-t)^{1/2}$. The crucial result is that the total excess domain area A_0, i.e., the area which will disappear, decreases linearly in time, and further, when possible, the

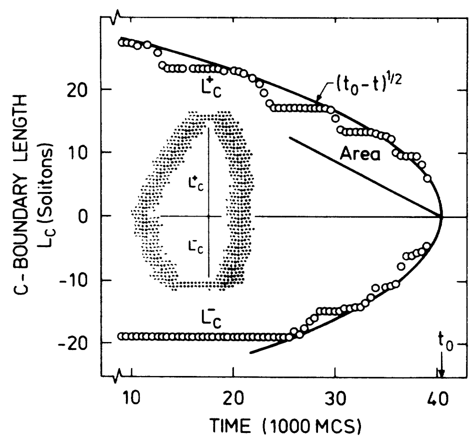

FIG. 2. The inset shows a typical domain with broad solitonlike walls and the horizontal sharp and straight walls, the stacking faults. The length projected on the vertical direction L_C is divided in the upper part L_C^+ and the lower part L_C^-. The time evolution of these are shown. Notice L_C^- is constant until the stacking fault has disappeared. The total excess area decreases linearly in time during the whole time interval. Consequently the length of the stacking fault L_S in the lower part decreases linearly in time, which is also found directly.

projected length of the C wall decreases as a square root, whereas the S wall with finite length can shorten, but not move. The recreation of short S walls is the reason for the steps on the L_c decrease in Fig. 2. The described behavior has been found for all other studied examples. It is in complete agreement with the basic assumption made by Allen and Cahn[5] for a curvature driven domain growth with the consequences:

$$L_C \sim (t_0 - t)^{1/2}, \quad A_0 = \alpha(t_0 - t) . \tag{2}$$

The next crucial assumption in the Allen-Cahn theory, generally not as strongly emphasized, is the assumption of scale invariance of the growth process. In the simplest form a simple self-similarity of the domain pattern at different times of course fulfills such a requirement. However, we shall now show that a much weaker scaling requirement is sufficient for deriving the Allen-Cahn law: Assume for simplicity a two-phase system, where the minority phase, which will disappear, forms only spherical domains. The excess area is the total area of these domains. The Allen-Cahn theory then predicts a decreasing domain radius $R_i(t) \sim (t_i - t)^{1/2}$ for each domain, now in general indexed "i". The smaller domains will disappear first, but it is important to note that all domains decrease by the same area per unit time. The system may therefore remain invariant, if the unit area A we are considering is increasing proportional to time, $A \sim t$, and the length unit L increases as $L \sim t^{1/2}$ since $A = L^2$. The excess energy $\Delta E(t)$ for a large distribution of domain sizes is proportional to the total wall length, i.e., $\Delta E(t) \sim \Sigma_i R_i(t)$. Assuming scale invariance, this length is constant when measured in the time dependent unit L, i.e., $\Sigma_i R_i(t) = \text{const}/L \sim t^{-1/2}$. From this follows the famous Allen-Cahn exponent $n = \frac{1}{2}$ and $\Delta E(t) \sim t^{-1/2}$. It is clearly sufficient that the excess area distribution is invariant when measured in the unit A, which is increasing linearly in time. However, no strict self-similarity is required.

Let us now apply the same scaling idea to our case. This is a singlular Allen-Cahn situation in which one curvature is zero, namely that of the S wall. As exemplified in Fig. 2, for each decay process of a domain "i" having an S wall, the area and consequently the length $L_{Si}(t)$ decrease linearly in time. This is contrary to the square root behavior (2). The excess energy associated with the S-decrease processes is proportional to the total S-wall length $\Delta E_S(t) \sim \Sigma_i L_{Si}(t)$. Assuming scale invariance with respect to the excess area for the S-decrease process we find $\Delta E_s(t) = \text{const}/L \sim t^{-1/2}$ in agreement with the Allen-Cahn theory. Furthermore, in the computer simulation one can separately measure the length of the S walls and explicitly show that $\Sigma_i L_{Si} \sim t^{-1/2}$. This confirms that the scaling of the area distribution is fulfilled although there is no self-similar scaling. It also shows that the unusual, linear time dependence of the individual process is not relevant.

Next we consider the decrease of the C walls, the C-decrease process. We must now distinguish between two cases: a dormant C-decrease process and an active one, which will only start when the S wall between two C walls has diminished to a length comparable to the lattice distance, i.e., when the stacking fault has a sufficiently small extent. Let us first discuss the active C-decrease process. A single process is shown in the upper part of Fig. 2, where it is proved to be a standard curvature driven Allen-Cahn process, with the area decreasing linearly in time, and the linear dimensions decrease as $(t_0 - t)^{1/2}$, in particular L_C^{ac}, where "ac" denotes active. The total excess energy for the active C-decrease processes is then

$$\Delta E_C^{\text{ac}}(t) \sim \sum_i L_{Ci}^{\text{ac}}(t) .$$

Assuming, as before, scale invariance for the area distribution ΔE_C^{ac} is constant when measured in the increasing length unit L. This leads again to $\Delta E_C^{\text{ac}}(t) \sim t^{-1/2}$. The special feature, that the energy only depends on projected length L_C, can therefore not explain the observed small exponent.

Finally let us consider the dormant C-decrease process. A single process is shown in the lower part of Fig. 2, where it is proved to be dormant until the S wall disappears at $t \sim 2500$ MCS. For a scaling argument for this process we must consider the probability that the intervening S walls of length L_S diminish to a fixed length of the order of the lattice constant a. When we consider larger and larger scales, the probability $\mathcal{P}$ for this to be the case decreases inversely as the length scale increases: $\mathcal{P}(L_S \sim a) \sim a/L \sim t^{-1/2}$. Out of the increasing area unit A, only a fraction A_{ac} of the area is available for the active C decrease process $A_{\text{ac}} = A\mathcal{P}(L_S \sim a) \sim tt^{-1/2} = t^{1/2}$. The area unit for the C-decrease process therefore increases slower, and consequently also the length unit $L_{\text{ac}} = A_{\text{ac}}^{1/2} \sim t^{1/4}$. Assuming again scale invariance for the excess area distribution we now find for the dormant C-decrease process, including the subsequent active one, that the decrease of the total excess energy is $\Delta E_c(t) \sim \Sigma_i L_{Ci}(t) = \text{const}/L_{\text{ac}}$. This gives an excess energy decrease as

$$\Delta E_c(t) \sim t^{-1/4} . \tag{3}$$

This agrees with the observed small exponent. The reason for the slow decay is simply that the dormant C-decay process has to wait for the S wall to disappear. We have also verified by the simulation that the total C length $\Sigma_i L_{Ci}$ decreases as $t^{-1/4}$. Any active C decrease process already in operation at early stages will disappear faster than the dormant one, which will dominate the late time behavior. It is interesting, however, that the equally fast S-decrease process will continue to play an important braking role for the dormant C-decrease process. This is because the prohibitive S-wall length is to be compared with the atomic scale. The excess energy is dominated by the dormant C-decrease process for energy reasons in our case, but it will always dominate at sufficiently late times. The slow time evolution with an exponent exactly equal $n = \frac{1}{4}$ is therefore now explained, not as a consequence of the softness of the walls, but as a consequence of a hierarchy of walls, where the decrease of one kind depends on the other. A related problem was studied previously by dynamical scaling theory.[15] Such a hierarchy is in fact present in the models[6,10] in which the slow growth was first discovered. We believe these model systems and our model indeed form a new universality class with $n=\frac{1}{4}$, independent of details in the models. The growth is in many respects in agreement with the Allen-Cahn theory, but we are dealing with a special case of mixed zero and finite curvature. Important examples of such straight walls are stacking faults and twin boundaries in crystals or on surfaces. We expect this class to have many members. A number of possible experiments were suggested by Mouritsen,[6] but results are yet to come. Ideally the domain wall length should be measured. However, the structure factor for scattering experiments does as well indicate a slow growth law, although being more difficult to analyze. At elevated temperatures the straight walls roughen[15] and get the possibility to move. The self-pinning then disappears and the growth approaches the standard Allen-Cahn $n=\frac{1}{2}$ behavior. This is seen in recent Monte Carlo studies.[10]

We conclude that the growth kinetics for nonconserved order parameter, even for degeneracy $p=2$, must be subdivided in at least two classes with algebraic time evolution t^{-n} with different exponents $n=\frac{1}{2}$ and $n=\frac{1}{4}$. The first represents independent domain wall movements, the latter hierarchical wall movements where the slower domain growth is due to time dependent self-pinning, operative for quenches to very low temperatures.

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