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Kinetics of slow domain growth in complex systems

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Abstract The domain growth after a quench to very low and to finite temperatures T is analysed by scaling theory and Monte Carlo simulation. The growth exponent for the excess energy $\Delta E(t) \sim t^{-n}$ is found to approach $n \sim 1/4$ for $T \rightarrow 0$. The scaling theory gives exactly $n = 1/4$ for cases of hierarchical movement of domain walls. This explains the existence of a newly discovered, slow growth universality class. It is shown to be a singular Allen-Cahn class, to which belong systems with domain walls of both exactly zero and finite curvature. The model studied has continuous variables, non-conserved order parameter and has two kinds of domain walls: sharp, straight stacking faults and broad, curved soliton-like walls. For quenches to higher temperatures the growth exponent is found to approach the classical Allen-Cahn exponent $n = 1/2$.

1. Introduction

The kinetics of domain growth is of relevance for the formation of polycrystalline microstructures. This is of considerable importance in surface science[1], metallurgy[2] and earth science[3]. It is in fact the complexity of the domain structures which is decisive for the physical properties.

To illustrate this let us first consider a perfect structure, a single crystal. This is not a complex system and its properties are completely predictable. It is precisely this predictability and lack of complexity which makes perfect structures "weak" in most applications, although the specific "strength" may be very high. For example a single crystal can be cleaved by a small force, just as ripping a piece of cloth, in a certain direction. Ripping a piece of patch-work is much more difficult,

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as the seams and pattern modify or absorb the applied force. The domain walls or grain boundaries have a similar effect in polystructures where several equivalent, perfect structures form a random patch-wotk. An example is demonstrated on fig. 1. To the left is shown a high temperature, disordered cubic structure. This can easily be cleaved along the indicated line. To the right is shown the corresponding low temperature hexagonal structure, where there are four equivalent domains formed by displacing the atoms regularly in the $\pm x$ or $\pm y$ directions. A fracture may now follow the indicated "unpredictable" line, which is no longer optimal for the exerted force. The complex structure is therefore stronger. Such a phase transformation between cubic and hexagonal structures after a quench from high to low temperatures is called a martensitic transformations. It is widely used in practical applications both in metallurgy and in ceramics, just for improving or tailoring the materials' strength. Of great interest is, of course, whether the obtained micro-structures are stable or metastable at low or moderate temperatures. The problem of domain growth kinetics is therefore of particular interest. A system after a rapid quench is far from equilibrium and has a complex, unpredictable arrangement of domains walls, which may change nonlinearly and irreversibly with time. Hence a theoretical analysis is exceedingly difficult, and it is a great advantage to make use of computer simulations, partly because one can perform ideal, pure experiments and partly because one can monitor much more detailed information than available in real experiments. This offers important checking points for a theoretical approach. Since the systems are complex, the only symmetry left during the domain growth is a possible selfsimilarity or another form of scaling property. The kinetics may therefore be independent of the specific forces in the system and it might to possible to classify the behavior into a few characteristic classes, in analogy with the critical behavior at a continuous phase transition.

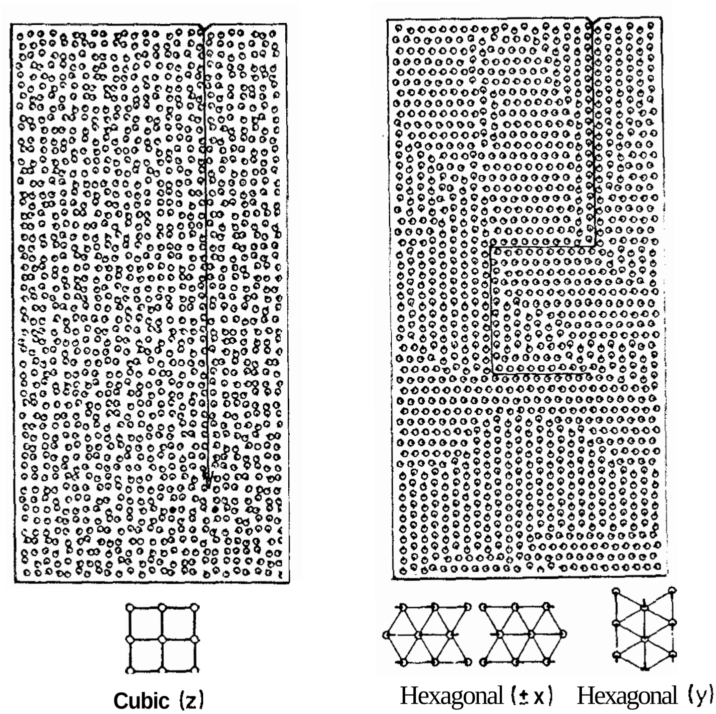


Fig. 1 - To the left is shown a high temperature, disordered, cubic phase. It will cleave easily along the indicated line. To the right is shown the four-domain, low temperature, hexagonal phase obtained after a rapid quench. It cleaves less easily, following the indicated line in the complex polystructure. The circles indicate atom positions and the small lines the displacement from an ideal cubic structure. The figure is calculated using the full dipolar model eq. (1) with $P = 2$. The figure exemplifies a martensitic transformation or a surface reconstruction.

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 non-conserved order parameter, the excess energy ΔE of the domain wall network is usually expected to decay algebraically as $\Delta E \sim t^{-n}$ with $n = 1/2$ according to the Allen-Cahn theory[5] for curvature driven growth. A possible deviation from this behavior yielding $n \sim 1/4$

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 walls for sufficiently high p also gave $n - 1/4$, and the possibility of a new universality class was proposed. The finding[6] of the small exponent $n - 1/4$ 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 $n = 1/4$ for quenches to very low, finite temperatures. It was first suggested by Mouritsen[6] that the deviation from $n = 1/2$ 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 in ref. 8 and by van Saarllos and Grant[9] who firstly showed that even if the walls were broad, the growth should follow $n = 1/2$. 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.

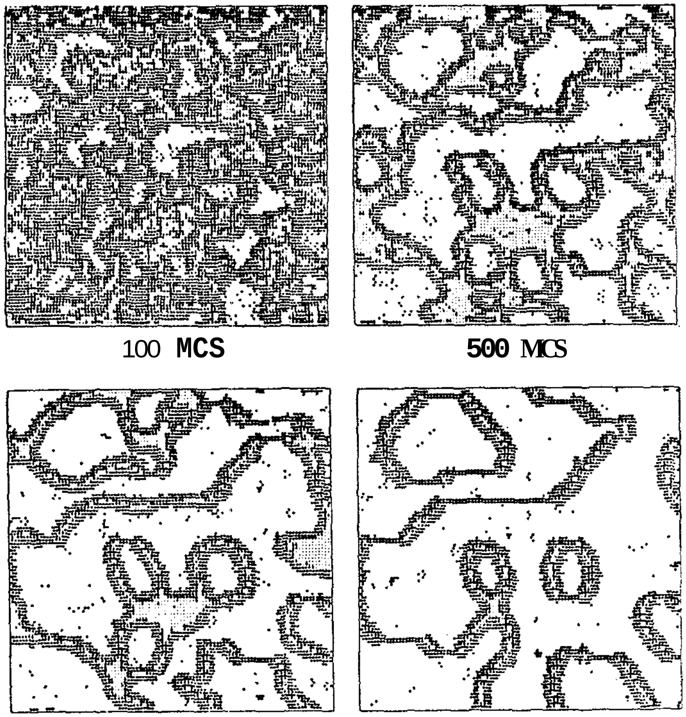
It is the aim of this work, which is described in more detail elsewhere[12], to discuss the raison d'être for the unexpected slow growth class for low temperature quenches, and further to demonstrate that the classical growth with $n = 1/2$ is recovered at higher temperatures. This insight was obtained by analysing 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 $n = 1/4$. 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 modelled by a soliton-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 depend only weaky on the phase O. 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 $n = 1/4$. The slowing down is due to a temporal pinning of the straight (zero curvature) walls, which cannot move untill 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 restricted to the $x-z$ plane on a two dimensional $x-y$ lattice ($d = 2$). We use the Hamiltonian introduced recently[13] for simulating a Martensitic transformation

$$H = \sum_{<ij>} \{ -KS_{iz}S_{jz} + J[S_i \cdot S_j - P(\hat{r}_{ij} \cdot S_i)(\hat{r}_{ij} \cdot S_j)]\} - D\sum(S_{iz}^4 + S_{iy}^4) \quad (1)$$

We have made extensive Monte Carlo computer simulations on this simple, restricted model (with $P = 3$) studying the domain growth after rapid temperature quenches from the ferromagnetic phase to the $p = 2$ phase with $\pm x$ domains only at low, finite temperature (0.01 of $T_N \sim 2J/k_B$), see snapshots on fig. 2. The details are reported elsewhere[14]. We choose to follow the behavior of the self-averaging[4] excess energy. The principal result is, as show in fig. 3, that the time evolution at late times is algebraic with a small exponent $n \sim 1/4$. This is the same result as found by Mouritsen et al.[6,10]. The simple model can be analyzed and we have shown that all assumptions made in the Allen-Cahn theory are fulfilled, yet giving a smaller exponent. The slow time evolution with an exponent exactly equal to $n = 1/4$ is explained by a scaling theory[12], 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. The straight walls cannot move and constitute temporal pinning centers. However, they can disappear by the motion of the curved walls. This slows down the domain growth, but does not stop it completely. A related problem was studied previously by dynamical scaling theory[16]. Such a hierarchy is in fact present in the models[6,10] in which the slow growth was first discovered. We believe these models systems and our model indeed form a new universality class with $n = 1/4$, 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.

Finally, we wish to demonstrate that there is a cross-over to the Cahn-Allen exponent $n = 1/2$ for quenches to higher temperatures. This was also found in the models studied by Mouritsen and Præstgård[10]. Using standard Monte Carlo simulations techniques[17], we have calculated the domain growth kinetics after quenches from $T = \infty$ to different finite temperatures. The results have been obtained on a $N = 100 \times 100$ lattice subject to periodic boundary conditions. At each temperature, the results were averaged on 15 different runs. In fig. 4 we present the best exponent obtained by fitting the averaged excess energy to the expression $\delta E(t) = t^{-n}$. The results clearly show a crossover in the kinetic exponent value from $n = 1/4$ to $n = 1/2$ as function of temperature, in agreement with previously results[10].

During the cross-over when the exponent is $1/4 < n < 1/2$ the mixture of straight and curved boundary is still present. The curved walls move with a constant velocity proportional to the curvature, which is inversely proportional to the length of the walls projected onto the y-direction, L_y, i.e. the number of solitons in the walls. The velocity increases with temperature. By studying barrel-like domains with curved walls of various lengths L_y, separated by the same length straight walls L_x, the pinning time t^* can be found as a function of temperature. The pinning time t^* is the time needed for L_x to decrease to length of the order of



100 MCS 500 MCS

1000 MCS 3000 MCS

Fig. 2 - Calculations for the restricted model eq.(1) with only two stable domains $\pm x$. Snapshots of the evolution of the ordered domains and the domains walls during a quench (we show one quarter of a 200×200 system) to $T = 0.001\ T_N$ .The white areas are the $\pm$x domains, the grey areas the z domains and the darker regions and single dots disordered spins, drawn as - o indicating their deviation. At early times $t = 100$ MCS one sees nucleation of $\pm x$ domains in a disordered z-matrix, at later time $t = 3000$ MCS one sees that two kinds of domain walls have evolved i.e. the straight stacking faults and the broad soliton walls.

an intersite distance. Therefore $t^*_{\cdot}$ is the time when the decrease of the length of the curved walls, L,, can begin. Fig. 5 shows that the pinning time decreases rapidly with increasing temperature. For T > 0.15 T_N one observes kinks being emitted from the corners to the sharp walls. This allows L, to decrease independently of the lenth L,, and this start to break the hierarchical pinning mechanism. Therefore

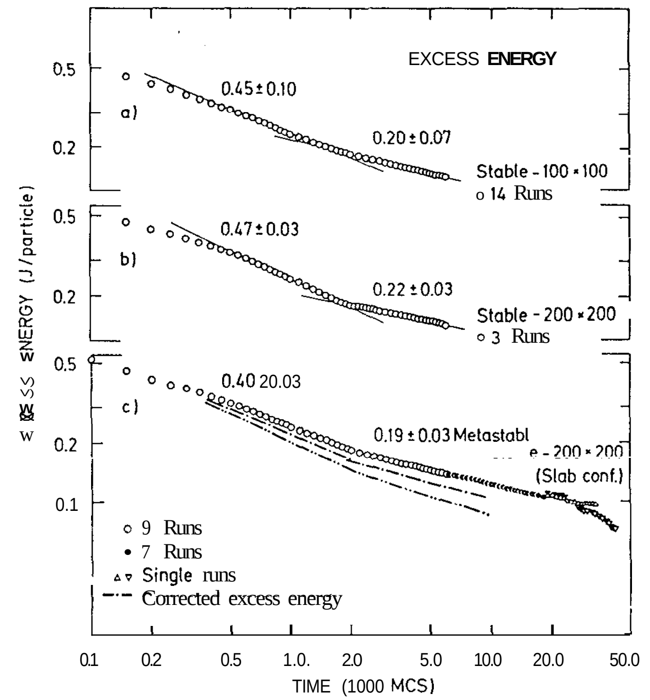


Fig. 3 - The average excess energy $\Delta E(t)$ for $T = 0.01\,T_N$ relative to the stable, single domain ground state energy $E_T(\infty)$ is plotted for two system sizes a) 100×100 for 14 runs and b) 200×200 for 3 runs. Consistently, an early regime is found with an exponent $n \sim 1/2$ and a sharp crossover to a second regime with a smaller exponent $n \sim 1/4$; c) shows the excess energy for 9 different 200×200 systems which develop into a metastable slab configuration. The signatures indicate the excess energy relative to $E_T(\infty)$ as above, yielding too low exponents. The excess energy relative to the relevant, higher slab energy $E_T(\infty)_{slab}$ is between the limits indicated by -.- and -..-. Both the exponents and the crossover for the corrected excess energy agree with the stable cases a) and b). At much later times $t > 25000$ MCS finite size effects are detected for separate runs (V).

the pinning becomes irrelevant for the late time behavior, and the system develops into a case corresponding to an all curved walls system. The scaling theory for

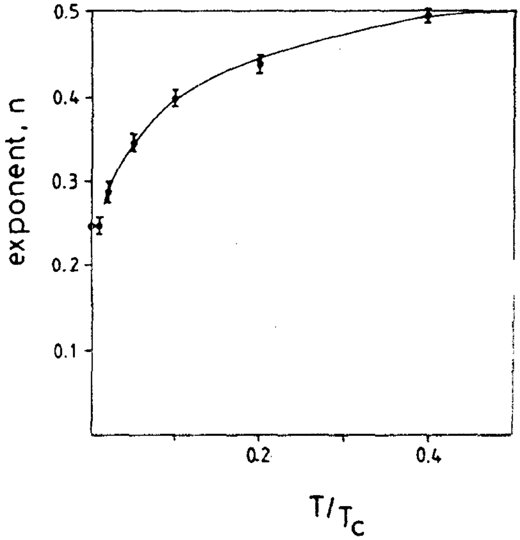


Fig. 4 - The exponent n for quenches to various finite temperatures for a $N = 100 \times 100$ system averaged over 15 Monte Carlo runs. A clear cross-over from $n = 1/4$ to $n = 1/2$ is seen.

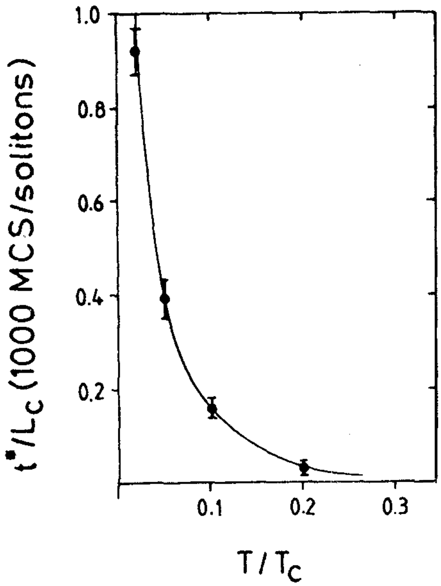


Fig. 5 - The pinning time t^* of a barrel shaped domain with curved walls of different length L,, but the same length L_s straight walls. The pinning time decreases rapidly with increasing quench temperatures.

this case gives the Cahn-Allen exponent $n = 1/2$. The decrease in the pinning time is consistent with the cross-over in the exponent shown in fig. **4.**

To obtain the algebraic growth laws it is imperative that the system has a scaling behavior. Therefore, the obtained results from the specific model are expected to have more general validity and to shed light upon the general aspects of the new, unexpected slow-domain-growth universality class.

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Resumo

O crescimento de domínios após um processo de têmpera a temperaturas T muito baixas ou finitas 6 analisado através de teoria de escala e simulações Monte Carlo. Obtém-se que o expoente de crescimento para a energia de excesso $\Delta E(t) \sim t^{-n}$ aproxima-se de $n \sim 1/4$ para $T \to 0$. A teoria de escala dá exatamente $n = 1/4$ para o caso de movimento hierárquico de paredes de domínio. Isto explica a existência de uma classe de universalidade de crescimento lento, recentemente descoberta. Mostra-se que esta é uma classe de Allen-Cahn singular, à qual pertencem sistemas com paredes de domínios de curvaturas tanto finitas quanto exatamente zero. O modelo estudado tem variáveis contínuas, parâmetro de ordem não-conservado, e dois tipos de parede de domínio: falhas de espalhamento retilineas e bem definidas, e paredes largas e curvadas, semelhantes a solitons. Para processos de têmpera a temperaturas mais altas obtém-se que o expoente de crescimento aproxima-se do valor clássico de Allen-Cahn, $n = 1/2$.