Arrested States of Solids

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Solids produced as a result of a fast quench across a freezing or a structural transition get stuck in long-lived metastable configurations of distinct morphology, sensitively dependent on the processing history. Martensites are particularly well studied examples of nonequilibrium solid-solid transformations. Since there are some excellent reviews on the subject, we shall, in this brief article, mainly present our viewpoint.

I. NONEQUILIBRIUM STRUCTURES IN SOLIDS

What determines the final microstructure of a solid under changes of temperature or pressure? This is an extremely complex issue, since a rigid solid finds it difficult to flow along its free energy landscape to settle into a unique equilibrium configuration. Solids often get stuck in long-lived metastable or jammed states because the energy barriers that need to be surmounted in order to get unstuck are much larger than $k_B T$.

Such nonequilibrium solid structures may be obtained either by quenching from the liquid phase across a freezing transition (see Ref. [1] for a comprehensive review), or by cooling from the solid phase across a structural transition. Unlike the former, nonequilibrium structures resulting from structural transformations do not seem to have attracted much attention amongst physicists, apart from Refs. [2,3], possibly because the microstructures and mechanical properties obtained appear nongeneric and sensitively dependent on details of processing history.

Metallurgical studies have however classified some of the more generic nonequilibrium microstructures obtained in solid (parent/austenite) - solid (product/ferrite) transformations depending on the kind of shape change and the mobility of atoms. To cite a few:

- **Martensites** are the result of solid state transformations involving shear and no atomic transport. Martensites occur in a wide variety of alloys, polymeric solids and ceramics, and exhibit very distinct plate-like structures built from twinned variants of the product.

- **Bainites** are similar to martensites, but in addition possess a small concentration of impurities (e.g. carbon in iron) which diffuse and preferentially dissolve in the parent phase.

- **Widmanstätten ferrites** result from structural transformations involving shape changes and are accompanied by short range atomic diffusion.

- **Pearlites** are a eutectic mixture of bcc Fe and the carbide consisting of alternating stripes.

- **Amorphous** alloys, a result of a fast quench, typically possess some short range ordering of atoms.

- **Polycrystalline** materials of the product phase are a result of a slower quench across a structural transition and display macroscopic regions of ordered configurations of atoms separated by grain boundaries.

That the morphology of a solid depends on the detailed dynamics across a solid-solid transformation, has been recognised by metallurgists who routinely use time-temperature-transformation (TTT) diagrams to determine heat treatment schedules. The TTT diagram is a family of curves parametrized by a fraction $\delta$ of transformed product. Each curve is a plot of the time required to obtain $\delta$ versus temperature of the quench (Fig. 1). The TTT curves for an alloy of fixed composition may be viewed as a ‘kinetic phase diagram’. For example, starting from a hot alloy at $t = 0$ equilibrated above the transition temperature (upper left corner) one could, depending on the quench rate (obtained from the slope of a line $T(t)$), avoid the nose of the curve and go directly into the martensitic region or obtain a mixture of ferrite and carbide when cooled slowly.
Two different equilibrium crystalline phases separated by a first order boundary (Fig. 2). An adiabatically slow quench from $T_{in} \rightarrow T_{fin}$ across the phase boundary in which the cooling rate is so small that at any instant the solid is in equilibrium corresponding to the instantaneous temperature would clearly result in an equilibrium final product at $T_{fin}$. On the other hand, an instantaneous quench would result in a metastable product bearing some specific relation to the parent phase. The task is to develop a nonequilibrium theory of solid state transformations which would relate the nature of the final arrested state and the dynamics leading to it to the type of structural change, the quench rate and the mobility of atoms.

In this article we concentrate on the dynamical and structural features of a class of solid-solid transformations called Martensites. Because of its commercial importance, martensitic transformations are a well studied field in metallurgy and material science. Several classic review articles and books discuss various aspects of martensites in great detail [6]. The growing literature on the subject is a clear indication that the dynamics of solid state transformations is still not well understood. We would like to take this opportunity to present, for discussion and criticism, our point of view on this very complex area of nonequilibrium physics [6].

We next review the phenomenology of martensites and highlight generic features that need to be explained by a nonequilibrium theory of solid state transformations.

II. PHENOMENOLOGY OF MARTENSITES

One of the most studied alloys undergoing martensitic transformations is iron-carbon [5]. As the temperature is reduced, Fe with less than 0.02% C undergoes an equilibrium structural transition (Fig. 2) from fcc (austenite) to bcc (ferrite) at $T_c = 910^\circ C$. An adiabatic cooling across $T_c$ nucleates a grain of the ferrite which grows isotropically, leading to a polycrystalline bcc solid. A faster quench from $T_{in} > T_c$ to $T_{fin} < M_s < T_c$ (where $M_s$ : martensite start temperature) produces instead a rapidly transformed metastable phase called the martensite, preempting the formation of the equilibrium ferrite. It is believed that martensites form by a process of heterogeneous nucleation. On nucleation, martensite ‘plates’ grow radially with a constant front velocity $\sim 10^3 cm/s$, comparable to the speed of sound. Since the transformation is not accompanied by the diffusion of atoms, either in the parent or the product, it is called a diffusionless transformation. Electron microscopy reveals that each plate consists of an alternating array of twinned or slipped bcc regions of size $\approx 100 \mu m$. Such martensites are called acicular martensites.

![Fig. 1 TTT curves for steel AISI 1090 (0.84% C + 0.60% Mn). A: austenite (fcc), F: ferrite (bcc), C: carbide (Fe$_3$C). Curves correspond to 0, 50 and 100 % transformation. Below a temperature $M_s$, the metastable martensite (M) is formed - the transformation curves for martensites are horizontal.](image1)

![Fig. 2 Phase diagram of Fe-C (weight percent of C $< 0.02\%$). $M_s$ is the martensite start temperature.](image2)
as habit planes. Martensites, characterised by such a
configuration of jammed plates, are long lived since the
elastic energy barriers for reorganisation are much larger
than $k_B T$.

A theoretical analysis of the dynamics of the martensitic transformation in Fe-C is complicated by the fact
that the deformation is 3-dimensional (Bain strain) with
3 twin variants of the bcc phase. Alloys like In-Ti, In-Pb,
Mn-Fe and high-$T_c$ ceramics however, offer the simplest
elements of martensitic transformations having only two
twin variants. For instance, the high-$T_c$ cuprates un-
dergo a tetragonal to orthorhombic transformation [2].

The rhombic phase can be obtained from the tetrago-


cal phase by a two-dimensional deformation, essentially
a square to rhombus transition. Experiments indicate
that all along the kinetic pathway, the local configura-
tions can be obtained from a two-dimensional deformation
of the tetragonal cell. This would imply that the
movement of atoms is strongly anisotropic and confined
to the ab-plane. Thus as far as the physics of this trans-
formation is concerned, the ab-planes are in perfect reg-
istry (no variation of the strain along the c-axis). In
the next two sections we shall discuss our work on the
dynamics of the square to a rhombus transformation in
2-dimensions using a molecular dynamics simulation and
a coarse-grained mode coupling theory.

### III. MOLECULAR DYNAMICS SIMULATION OF
SOLID-SOLID TRANSFORMATIONS

Our aim in this section will be to study the simplest
molecular dynamics (MD) simulation of the square to
rhombus transformation. We would like to use the sim-
ulation results to construct the complete set of coarse
grained variables needed in a continuum description of
the dynamics of solid state transformations. We carry
out the MD simulation in the constant $NVT$ ensemble
using a Nosé-Hoover thermostat ($N = 12000$) [3].

Our MD simulation is to be thought of as a ‘coarse-
grained’ MD simulation, where the effective potential
is a result of a complicated many-body interaction. One
part of the interaction is a purely repulsive two-body
potential $V_2(r_{ij}) = v_2/r_{ij}^{12}$ where $r_{ij}$ is the distance
between particles $i$ and $j$. The two-body interaction
favours a triangular lattice ground state. In addition,
triplets of particles interact via a short range three-body
potential $V_3(r_i, r_j, r_k) = v_3w(r_{ij}, r_{jk}, r_{ik})[\sin^2(4\theta_{ijk}) + \sin^2(2\theta_{ijk})]$ where $w(r)$ is smooth short-
range function and $\theta_{ijk}$ is the bond angle at $j$ between
particles $(ijk)$. Since $V_3$ is minimised when $\theta_{ijk} = 0$ or
$\pi/2$, the three-body term favours a square lattice ground
state. Thus at sufficiently low temperatures, we can in-
duce a square to triangular lattice transformation by tun-
ing $v_3$. The phase diagram in the $T - v_3$ plane is exhibited
in Fig. 3.

We define elastic variables, coarse-grained over a spa-
tial block of size $\xi$ and a time interval $\tau$, from the in-
stantaneous positions $u$ of the particles. These include
the deformation tensor $\partial u_i/\partial x^k$, the full non linear strain
$\epsilon_{ij}$, and the vacancy field $\phi = \rho - \overline{\rho}$ ($\rho =$ coarse grained
local density, $\overline{\rho} =$ average density). We have kept track
of time dependence of these coarse-grained fields during
the MD simulation.

![Fig. 3 T – $v_3$ phase diagram from the MD simulations showing the freezing and structural transitions. The upper and lower arrows correspond to the high and low temperature quenches respectively.](image)

Consider two ‘quench’ scenarios — a high and low tem-
perature quench (upper and lower arrows in Fig. 3 respec-
tively) across the phase boundary. In both cases the solid
is initially at equilibrium in the square phase.

The high temperature quench across the phase bound-
ary, induces a homogeneous nucleation (i.e., strain inho-


cogeneities created by thermal fluctuations are sufficient
to induce critical nucleation) and growth of a triangular
region. The product nucleus grows isotropically with the
size $R \sim t^{1/2}$. A plot of the vacancies/interstitial field
shows that, at these temperatures they diffuse fast to
their equilibrium value (vacancy diffusion obeys an Ar-
renius form $D_v = D_0 \exp(-A/k_B T)$, where $A$ is an ac-
tivation energy and so is larger at higher temperatures).
The final morphology is a polycrystalline triangular solid.

The low temperature quench on the other hand, needs
defects (either vacancies or dislocations) to seed nucle-
ation in an appreciable time. This heterogeneous
ucleation initiates an embryo of triangular phase, which
grows anisotropically along specific directions (Fig. 4).
Two aspects are immediately apparent, the growing nu-
cleus is twinned and the front velocities are high. In-


deed the velocity of the front is a constant and roughly
half the velocity of longitudinal sound. A plot of the va-
cancy/interstitial field shows a high concentration at the
parent-product interface. The vacancy field now diffuses
very slowly and so appear to get stuck to the interface over
the time scale of the simulation. If we force the vacan-
cies and interstitials to annihilate each other, then

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[3]: Nosé-Hoover thermostat

[2]: Tetragonal to orthorhombic transformation

[1]: Square to rhombus transformation
the anisotropic twinned nucleus changes in the course of time to an isotropic untwinned one!

Therefore the lessons from the MD simulation are:
1. There are at least two scenarios of nucleation of a product in a parent depending on the temperature of quench. The product grows via homogeneous nucleation at high \( T \), and via heterogeneous nucleation at low \( T \). The complete set of slow variables necessary to describe the nucleation of solid-solid transformations should include the strain tensor and defects (vacancies and dislocations) which are generated at the parent-product interface at the onset of nucleation. The relaxation times of these defects dictate the final morphology. At high temperatures the defects relax fast and the grains grow isotropically with a diffusive front. The final morphology is a polycrystalline triangular solid. At low temperatures the interfacial defects (vacancies) created by the nucleating grain relax slowly and get stuck at the parent-product interface. The grains grow anisotropically along specific directions. The critical nucleus is twinned and the front grows ballistically (with a velocity comparable to the sound speed). The final morphology is a twinned martensite.

**IV. MODE COUPLING THEORY OF SOLID-SOLID TRANSFORMATIONS**

Armed with the lessons from the MD simulation, let us now construct a continuum elastic theory of solid-state nucleation. The analysis follows in part the theories of Krumhansl et. al. [2], but has important points of departure. The procedure is to define a coarse grained free energy functional in terms of all the relevant ‘slow’ variables. The tuning parameter \( a \) induces a transition from a square (described by \( \epsilon = 0 \)) to a rhombus (\( \epsilon = \pm \epsilon_0 \)).

Starting with \( \epsilon = 0 \) corresponding to the equilibrium square parent phase at a temperature \( T > T_c \), we quench across the structural transition. The initial configuration of \( \epsilon \) is now metastable at this lower temperature, and would decay towards the true equilibrium configuration by nucleating a small ‘droplet’ of the product. As we saw in the last section, as soon as a droplet of the product appears embedded in the parent matrix, atomic mismatch at the parent-product interface gives rise to interfacial defects like vacancies and dislocations.

Let us confine ourselves to solids for which the energy cost of producing dislocations is prohibitively large. This would imply that the interfacial defects consist of only vacancies and interstitials. The dynamics of nucleation now written in terms of \( \epsilon \), \( g \) (the conserved momentum density) and vacancy \( \phi \) are complicated [3]. For the present purpose, all we need to realise is that \( \phi \) couples to the strain and is diffusive with a diffusion coefficient \( D_\phi \) depending on temperature.

As in the MD simulation, we find that the morphology and growth of the droplet of the product depends critically on the diffusion of these vacancies. If the temperature of quench is high, \( \phi \) diffuses to zero before the critical nucleus size is attained and the nucleus eventually grows into an equilibrium (or polycrystalline) triangular solid. In this case, the nucleus grows isotropically with \( R \sim t^{1/2} \). However a quench to lower temperatures results in a low vacancy diffusion coefficient. In the limit \( D_\phi \rightarrow 0 \), the \( \phi \)-field remains frozen at the moving parent-product interface. In this case a constrained variational calculation of the morphology of the nucleus, shows that it is energetically favourable to form a twinned martensite rather than a uniform triangular structure. The growth of the twinned nucleus is not isotropic, but along habit planes. Lastly the growth along the longer direction is ballistic with a velocity proportional to \( \sqrt{\chi_v} \) (of the order of the sound velocity). All these results are consistent with the results of the previous section and with martensite phenomenology. Let us try and understand in more physical terms, why the growing nucleus might want to form twins.

As soon as a droplet of the triangular phase of dimension \( L \) is nucleated, it creates vacancies at the parent-product interface. The free energy of such an inclusion is \( F = F_{bulk} + F_{pp} + F_\phi \). The first term is simply the bulk free energy gain equal to \( \Delta F L^2 \) where \( \Delta F \) is the free energy difference between the square and triangular phases. The next two terms are interfacial terms. \( F_{pp} \) is the elastic contribution to the parent-product interface coming from the gradient terms in the free energy density Eq. [4] and is equal to \( 4\pi \sigma_{pp} L \), where \( \sigma_{pp} \) is the surface tension at the parent-product interface. \( F_\phi \) is the contribution from the interfacial vacancy field glued to the
parent-product interface and is proportional to $\phi^2 \sim L^2$ (since the atomic mismatch should scale with the amount of parent-product interface). This last contribution dominates at large $L$ setting a prohibitive price to the growth of the triangular nucleus. The solid gets around this by nucleating a twin with a strain opposite to the one initially nucleated, thereby reducing $\phi$. Indeed for an equal size twin, $\phi \to 0$ on the average, and leads to a much lower interfacial energy $E_\phi \sim L$. However the solid now pays the price of having created an additional twin interface whose energy cost is $F_{tw} = \sigma_{tw} L$.

Considering now an (in general) anisotropic inclusion of length $L$, width $W$ consisting of $N$ twins, the free energy calculation goes as

$$F = \Delta F LW + \sigma_{pp}(L + W) + \sigma_{tw} NW + \beta \left( \frac{L}{N} \right)^2 N$$

(2)

where the last term is the vacancy contribution. Minimization with respect to $N$ gives $L/N \sim W^{1/2}$, a relation that is known for 2-dimensional martensites like In-Tl.

Our next task is to solve the coupled dynamical equations with appropriate initial conditions numerically, to obtain the full morphology phase diagram as a function of the type of structural change, the parameters entering the free energy functional and kinetic parameters like $D_v$.

It should be mentioned that our theory takes off from the theories of Krumhansl et. al. \cite{Krumhansl}, in that we write the elastic energy in terms of the nonlinear strain tensor and its derivatives. In addition we have shown that the process of creating a solid nucleus in a parent generates interfacial defects which evolve in time. The importance of defects has been stressed by a few metallurgists \cite{Nairm}. We note also that the parent-product interface is studded with an array of vacancies with a separation equal to the twin size. This implies that the strain decays exponentially from interface over a distance of order $L/N$. This has been called ‘fringing field’ in Ref. \cite{Krumhansl}. Krumhansl et. al. obtain this by imposing boundary conditions on the parent-product interface, whereas here it appears dynamically.

V. PATTERNING IN SOLID-SOLID TRANSFORMATIONS: GROWTH AND ARREST

So far we have discussed the nucleation and growth of single grains. This description is clearly valid at very early times, for as time progresses the grains grow to a size of approximately $1 \mu m$ and start colliding, whereupon in most alloys they stop. Optical micrographs of acicular martensites reveal that the jammed plates lie along habit planes that criss-cross and partition the surrounding fcc (parent) matrix.

Can we quantify the patterning seen in martensite aggregates over a scale of a millimeter? A useful measure is the size distribution of the martensite grains embedded in a given volume of the parent. The appropriate (but difficult !) calculation at this stage would be the analogue of a Becker-Döring theory for nucleation in solids. In the absence of such a theory, we shall take a phenomenological approach.

Clearly the size distribution $P(l,t)$ depends on the spatio-temporal distribution $I$ of nucleation sites and the growth velocity $v$. We have analysed the problem explicitly in a simple 2-dimensional context. Since the nucleating martensitic grains are highly anisotropic and grow along certain directions with a uniform velocity, a good approximation is to treat the grains as lines or rays. These rays (lines) emanate from nucleation sites along certain directions, and grow with a constant velocity $v$. The rays stop on meeting other rays and eventually after a time $T$, the 2-dimensional space is fragmented by $N$ colliding rays. The size distribution of rays, expressed in terms of a scaling variable $y = y(I,v)$, has two geometrical limits — $\Gamma$-fixed point (at $y = 0$) and the $L$-fixed point (at $y = \infty$). The $\Gamma$-fixed point corresponds to the limit where the rays nucleate simultaneously with a uniform spatial distribution. The stationary distribution $P(l)$ is a Gamma distribution with an exponentially decaying tail. The $L$-fixed point, corresponds to the limit where the rays are nucleated sequentially in time (and uniformly in space) and grows with infinite velocity. By a mapping onto a multifragmentation problem, Ben Naim and Krapivsky \cite{Krapivsky} were able to derive the exact asymptotic form for the moments of $P(l)$ at the $L$-fixed point. The distribution function $P(l)$ has a multiscaling form, characterised by its moments $<l^q> \sim N^{-\mu(q)}$ where $\mu(q) = (q + 2 - \sqrt{q^2 + 4})/2$. At intermediate values of the scaling variable $y$, there is a smooth crossover from the $\Gamma$-fixed point to the $L$-fixed point with a kinematical crossover function and crossover exponents.

The emergence of scale invariant microstructures in martensites as arising out of a competition between the nucleation rate and growth is a novel feature well worth experimental investigation. There have been similar suggestions in the literature, but as far as we know there has been no direct visualization studies of the microstructure of acicular martensites using optical micrographs. Recent acoustic emission experiments \cite{Acoustic} on the thermoelastic reversible martensite Cu-Zn-Al, may be argued to provide indirect support of the above claim \cite{Krumhansl}, but the theory of acoustic emission in martensites is not understood well enough to make such an assertion with any confidence.

VI. OPEN QUESTIONS

We hope this short review makes clear how far we are in our understanding of the dynamics of solid-solid transformations. A deeper understanding of the field will only come about with systematic experiments on carefully se-
lected systems. For instance, a crucial feature of our nonequilibrium theory of martensitic transformations is the existence of a dynamical interfacial defect field. In conventional Fe based alloys, the martensitic front grows incredibly fast, making it difficult to test this using in situ transmission electron microscopy. Colloidal solutions of polystyrene spheres (polyballs) however, are excellent systems for studying materials properties. Polyballs exhibiting fcc → bcc structural transitions have been seen to undergo twinned martensitic transformations. The length and time scales associated with colloids are large, making it comfortable to study these systems using light scattering and optical microscopy.

In this article we have focussed on a small part of the dynamics of solid state transformations, namely the dynamics and morphology of martensites. Even so our presentation here is far from complete and there are crucial unresolved questions that we need to address.

Let us list the issues as they appear following a nucleation event.

The physics of heterogeneous nucleation in solids is very poorly understood. For instance, it appears from our simulations that the morphology of the growing nucleus depends on the nature of the defects seeding the nucleation process (e.g., vacancies, dislocations and grain boundaries). In addition several martensitic transformations are associated with correlated nucleation events and autocatalysis. Though these features are not central to the issue of martensites, such a study would lead to a better understanding of the origins of athermal, isothermal and burst nucleation. This in conjunction with a ‘Becker-Döring theory’ for multiple martensite grains would be a first step towards a computation of the TTT curves.

We still do not understand the details of the dynamics of twinning and how subsequent twins are added to the growing nucleus. Moreover the structure and dynamics of the parent-product interface and of the defects embedded in it have not been clearly analysed.

It would be desirable to have a more complete theory which displays a morphology phase diagram (for a single nucleus) given the type of structural transition and the kinetic, thermal and elastic parameters.

Certain new directions immediately suggest themselves. For instance, the role of carbon in interstitial alloys like Fe-C leading to the formation of bainites; the coupling of the strain field to an external stress and the associated shape memory effect; and finally the nature of tweed phases and pre-martensitic phenomena (associated with the presence of quenched impurities).

It is clear that the study of the dynamics of solid-solid transformations and the resulting long-lived morphologies lies at the intersection of metallurgy, material science and nonequilibrium statistical mechanics. The diversity and richness of phenomena make this an extremely challenging area of nonequilibrium physics.

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Figure Caption

Fig. 4  MD snapshot of (a) the nucleating grain at some intermediate time initiated by the low temperature quench across the square-triangle transition. The dark(white) region is the triangular(square) phase respectively. Notice that the nucleus is twinned and highly anisotropic. (b) the vacancy (white)/interstitial (black) density profile at the same time as (a). Notice that the vacancies and interstitials are well separated and cluster around the parent-product interface.
This figure "slide.gif" is available in "gif" format from:

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