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Nucleation and interfaces in martensitic phase transformations

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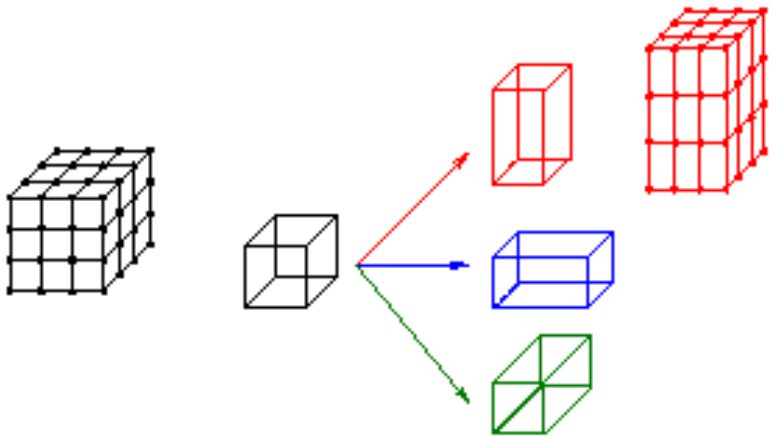
Martensitic transformations involve a change of shape of the crystal lattice of some alloy at a critical temperature.

e.g. cubic to tetragonal

$\theta > \theta_c$

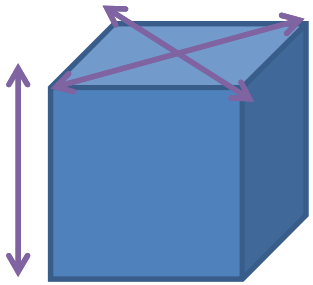
cubic austenite



$\theta < \theta_c$

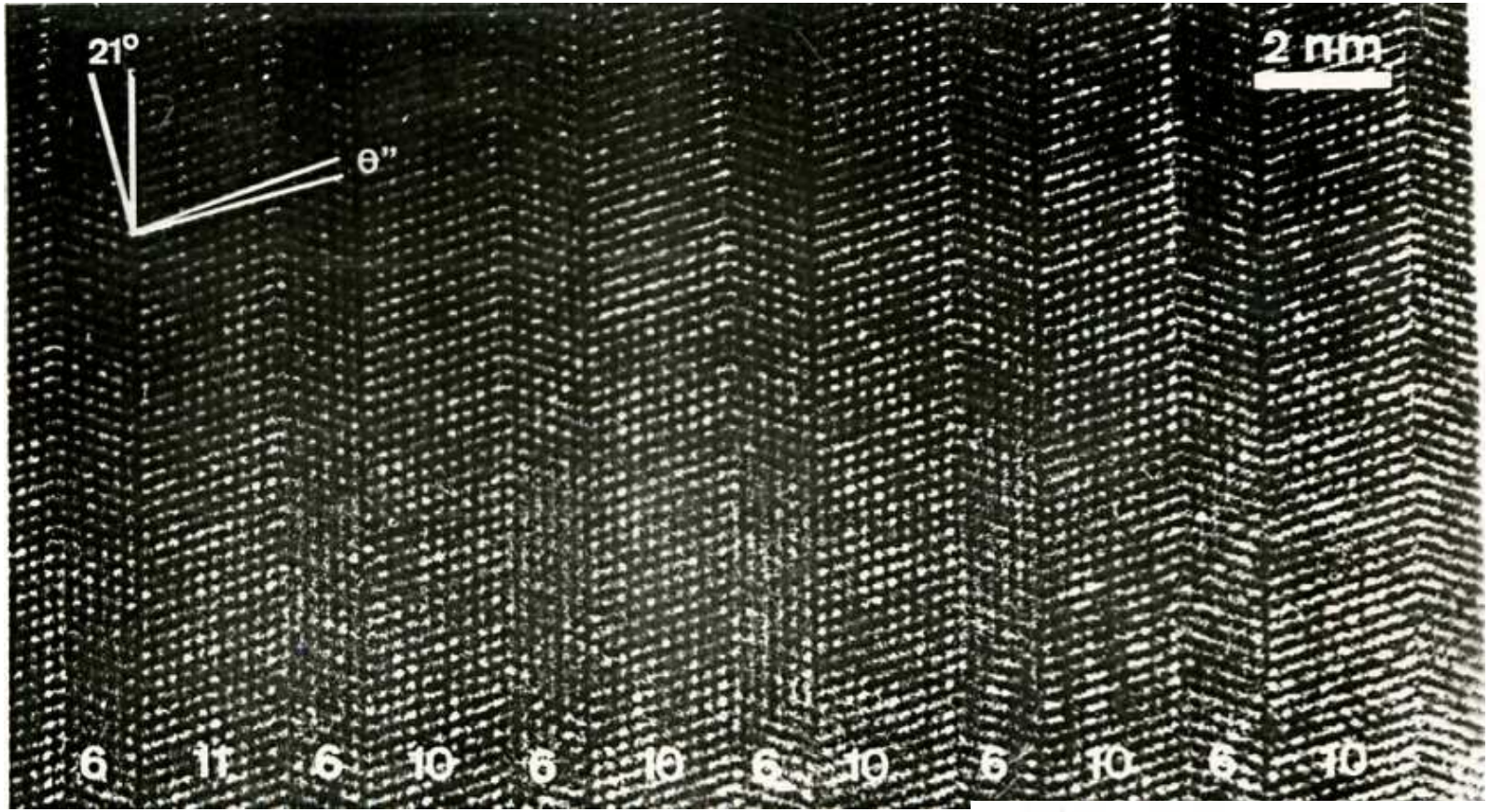
three tetragonal variants of martensite

cubic to orthorhombic (e.g. CuAlNi)



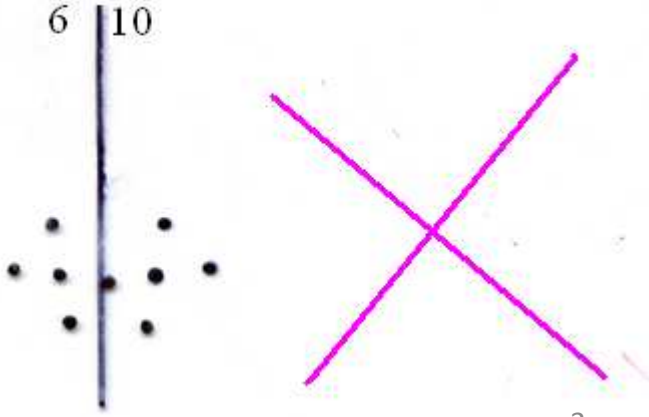
$\theta < \theta_c$

six orthorhombic variants of martensite

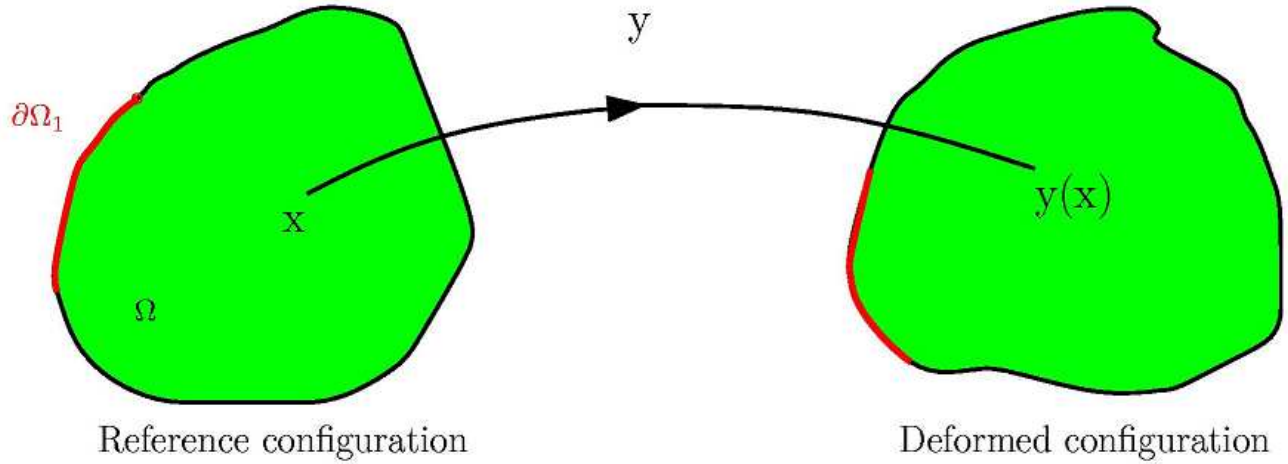


Atomistically sharp interfaces for cubic to tetragonal transformation in NiMn

Baele, van Tenderloo, Amelinckx



Energy minimization problem for single crystal



Minimize $I_\theta(y) = \int_\Omega \psi(Dy(x), \theta)\, dx$

among $y : \Omega \to \mathbb{R}^3$ subject to $y|_{\partial\Omega_1} = \bar{y}$, where $\theta =$ temperature, and the free-energy density $\psi = \psi(A, \theta)$ is defined for

$$A \in M^{3\times 3}_+ = \{A \in M^{3\times 3} : \det A > 0\}.$$

Energy-well structure

$$K(\theta) = \{A \in M^{3\times 3}_{+} \text{ that minimize } \psi(A,\theta)\}$$

Assume

austenite

$$K(\theta) = \begin{cases} \alpha(\theta)\mathrm{SO}(3) & \theta > \theta_c \\ \mathrm{SO}(3) \cup \bigcup_{i=1}^{M} \mathrm{SO}(3)U_i(\theta_c) & \theta = \theta_c \\ \bigcup_{i=1}^{M} \mathrm{SO}(3)U_i(\theta) & \theta < \theta_c, \end{cases}$$

$$\alpha(\theta_c) = 1$$

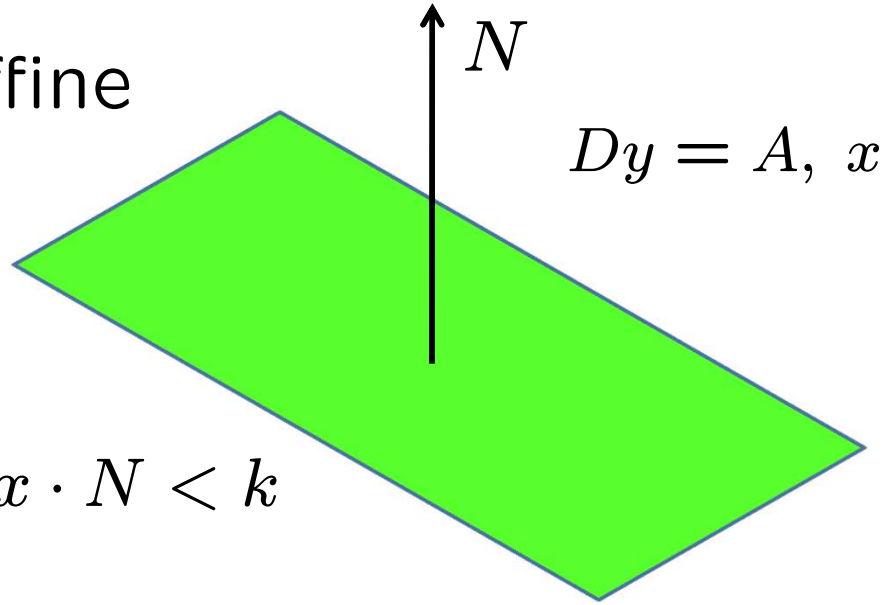
martensite

e.g. for cubic to tetragonal $M = 3$ and

$$U_1 = \mathrm{diag}\,(\eta_2, \eta_1, \eta_1),\ U_2 = \mathrm{diag}\,(\eta_1, \eta_2, \eta_1),$$
$$U_3 = \mathrm{diag}\,(\eta_1, \eta_1, \eta_2).$$

Rank-one matrices and the Hadamard jump condition

y piecewise affine



$$\boxed{A - B = a \otimes N}$$

Hadamard jump condition

Theorem

Let $U = U^T > 0$, $V = V^T > 0$. Then $\mathrm{SO}(3)\,U$, $\mathrm{SO}(3)\,V$ are rank-one connected iff

$$U^2 - V^2 = c(N \otimes \tilde{N} + \tilde{N} \otimes N) \qquad (*)$$

for unit vectors N, $\tilde{N}$ and some $c \neq 0$.

If $\tilde{N} \neq \pm N$ there are exactly two rank-one connections between V and $\mathrm{SO}(3)\,U$ given by

$$RU = V + a \otimes N, \quad \tilde{R}U = V + \tilde{a} \otimes \tilde{N},$$

for suitable $R, \tilde{R} \in SO(3)$, $a, \tilde{a} \in \mathbb{R}^3$.

Corollaries.

1. There are no rank-one connections between matrices A, B belonging to the *same* energy well.

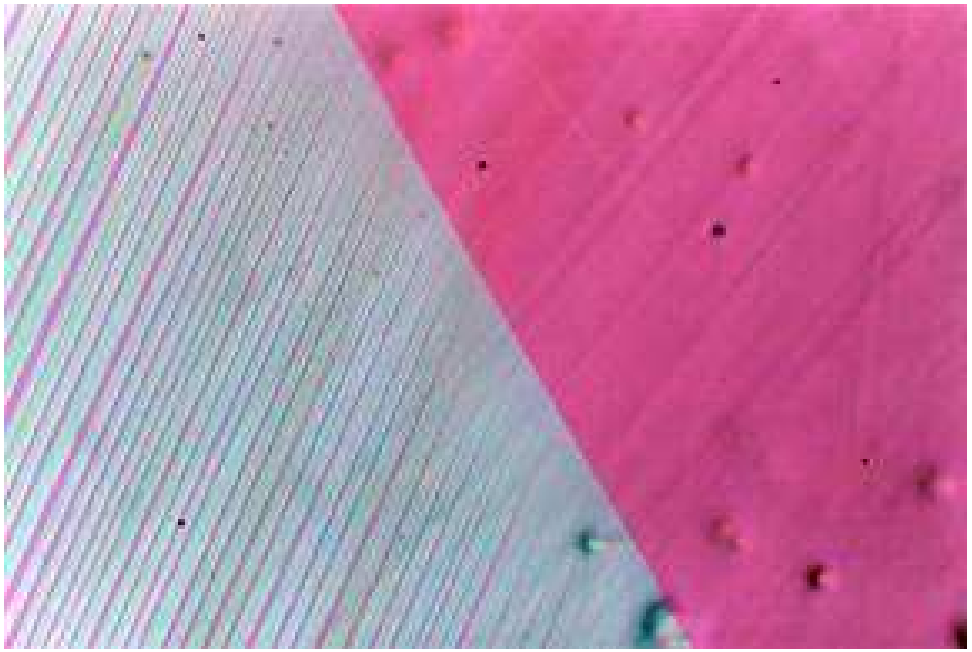
Proof. In this case $U = V$, contradicting $c \neq 0$.

2. If U_i, U_j are distinct martensitic variants then $SO(3)U_i$ and $SO(3)U_j$ are rank-one connected if and only if $\det(U_i^2 - U_j^2) = 0$, and the possible interface normals are orthogonal. Variants separated by such interfaces are called *twins*.

3. There is a rank-one connection between pairs of matrices $A \in SO(3)$ and $B \in SO(3)U_i$ *if and only if* U_i *has middle eigenvalue 1.*

When a new phase is nucleated in such a phase transformation, it has to fit geometrically onto the parent phase. This is is both an important ingredient for determining microstructure morphology, and leads to metastability when the two phases are geometrically incompatible.

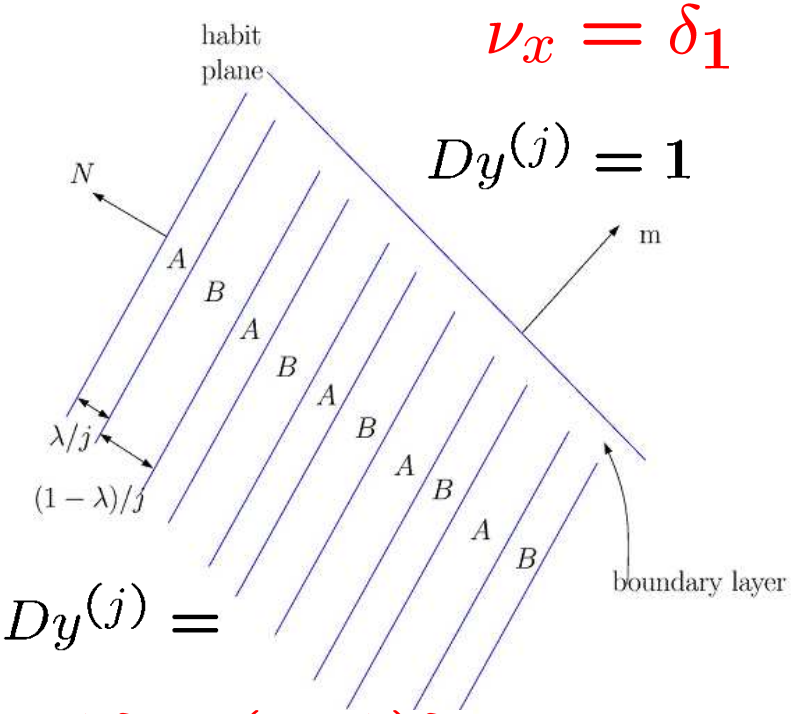
e.g. austenite-martensite interfaces



(Classical) austenite-martensite interface in CuAlNi
(courtesy C-H Chu and R.D. James)

$\nu_x = \delta_1$

$Dy^{(j)} = 1$



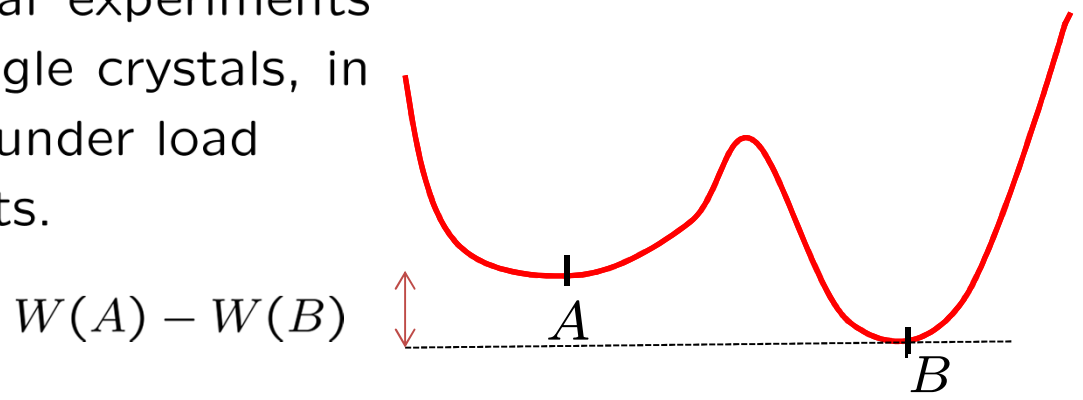
$Dy^{(j)} =$

$\nu_x = \lambda\delta_A + (1-\lambda)\delta_B$

Gives formulae of the crystallographic theory of martensite (Wechsler, Lieberman, Read)

Two examples of incompatibility-induced metastability

1. Special case of JB/James 2014 designed to explain hysteresis in the bi-axial experiments of Chu & James on CuAlNi single crystals, in which a transformation occurs under load between two martensitic variants.



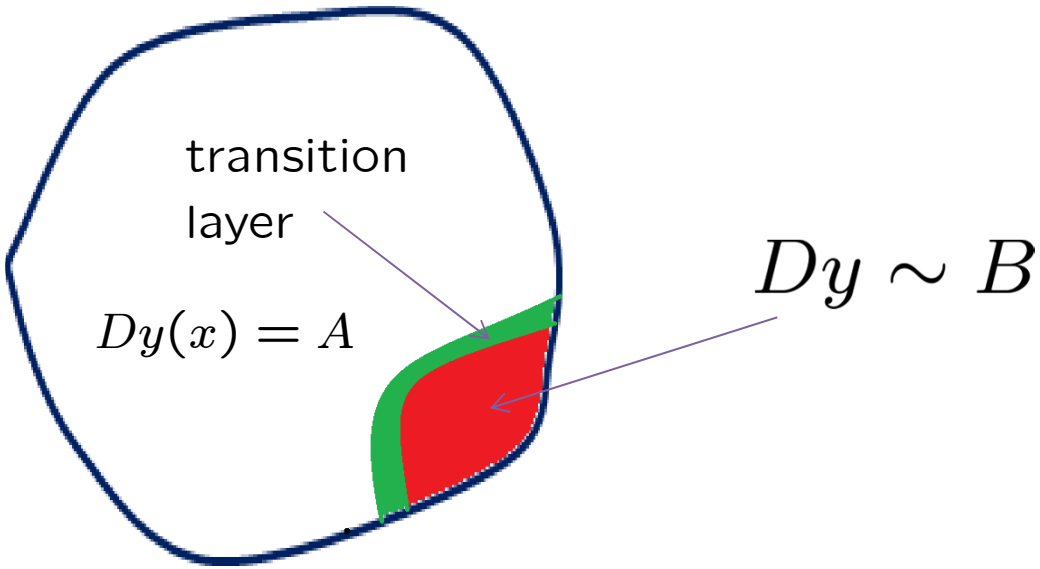
Consider the integral

$$I(y) = \int_\Omega W(Dy)\, dx, \qquad W(A) = \psi(A, \theta) - T \cdot A$$

where $W : M^{3\times 3} \to \mathbb{R}$ and W has two local minimizers at A, B with $\text{rank}\,(A - B) > 1$ and $W(A) - W(B) > 0$ sufficiently small.

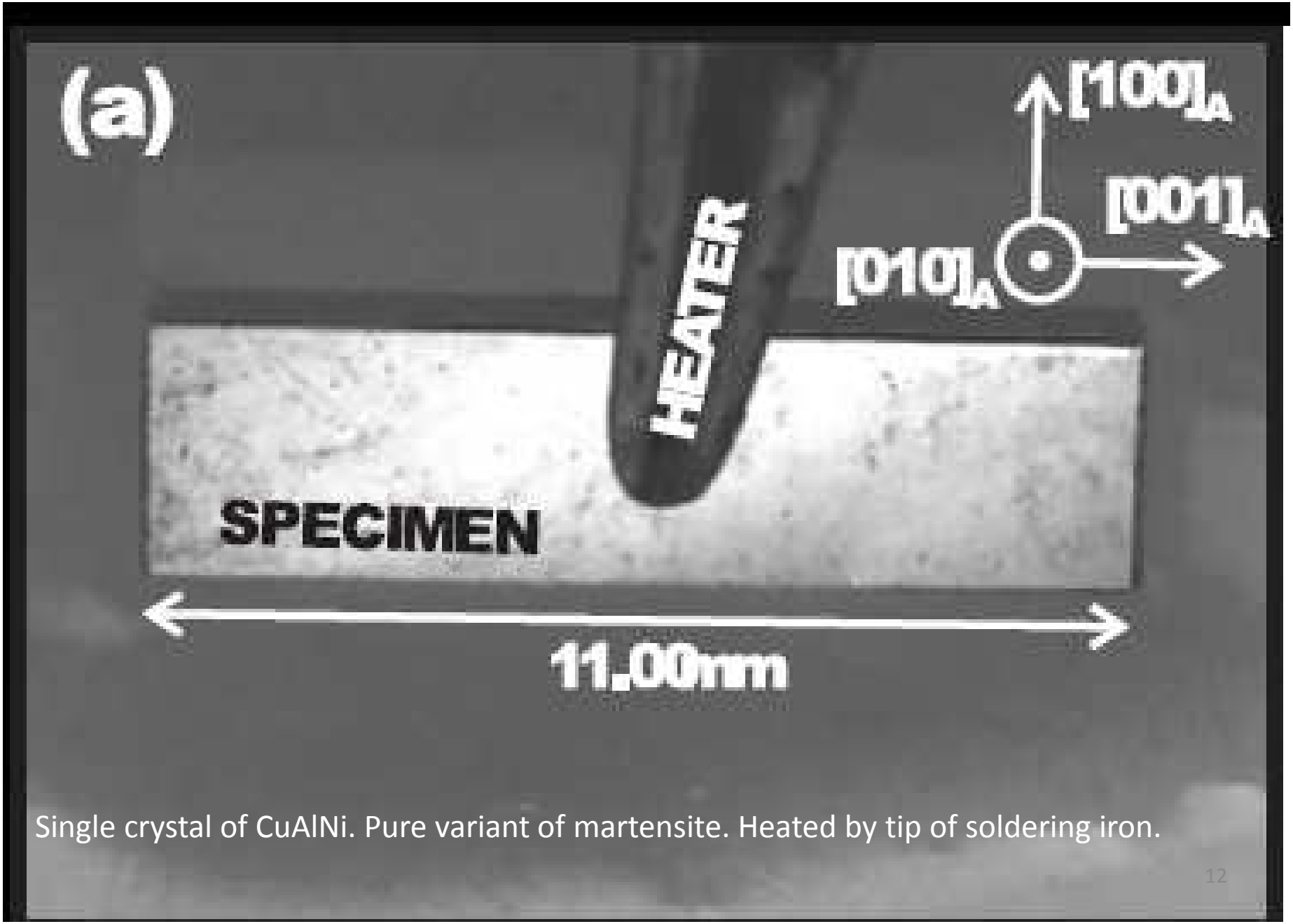
Claim. Under suitable growth hypotheses on W, $\bar{y}(x) = Ax + c$ is a local minimizer of I in $L^1(\Omega; \mathbb{R}^3)$, i.e. there exists $\varepsilon > 0$ such that $I(y) \geq I(\bar{y})$ if $\int_\Omega |y - \bar{y}|\, dx < \varepsilon$.

Idea: since A and B are incompatible, if we nucleate a region in which $Dy(x) \sim B$ there must be a transition layer in which the increase of energy is greater than the decrease of energy in the nucleus.

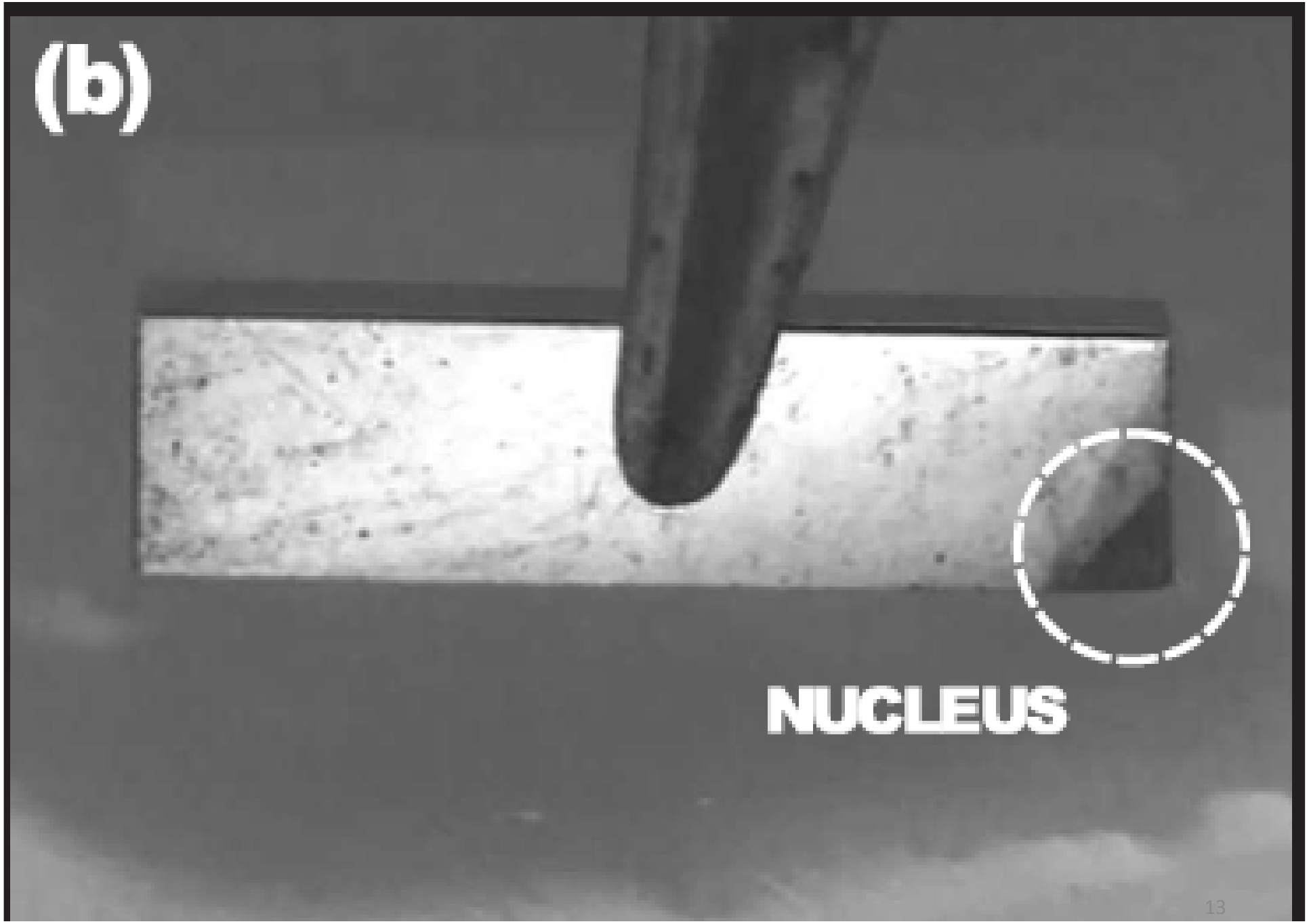


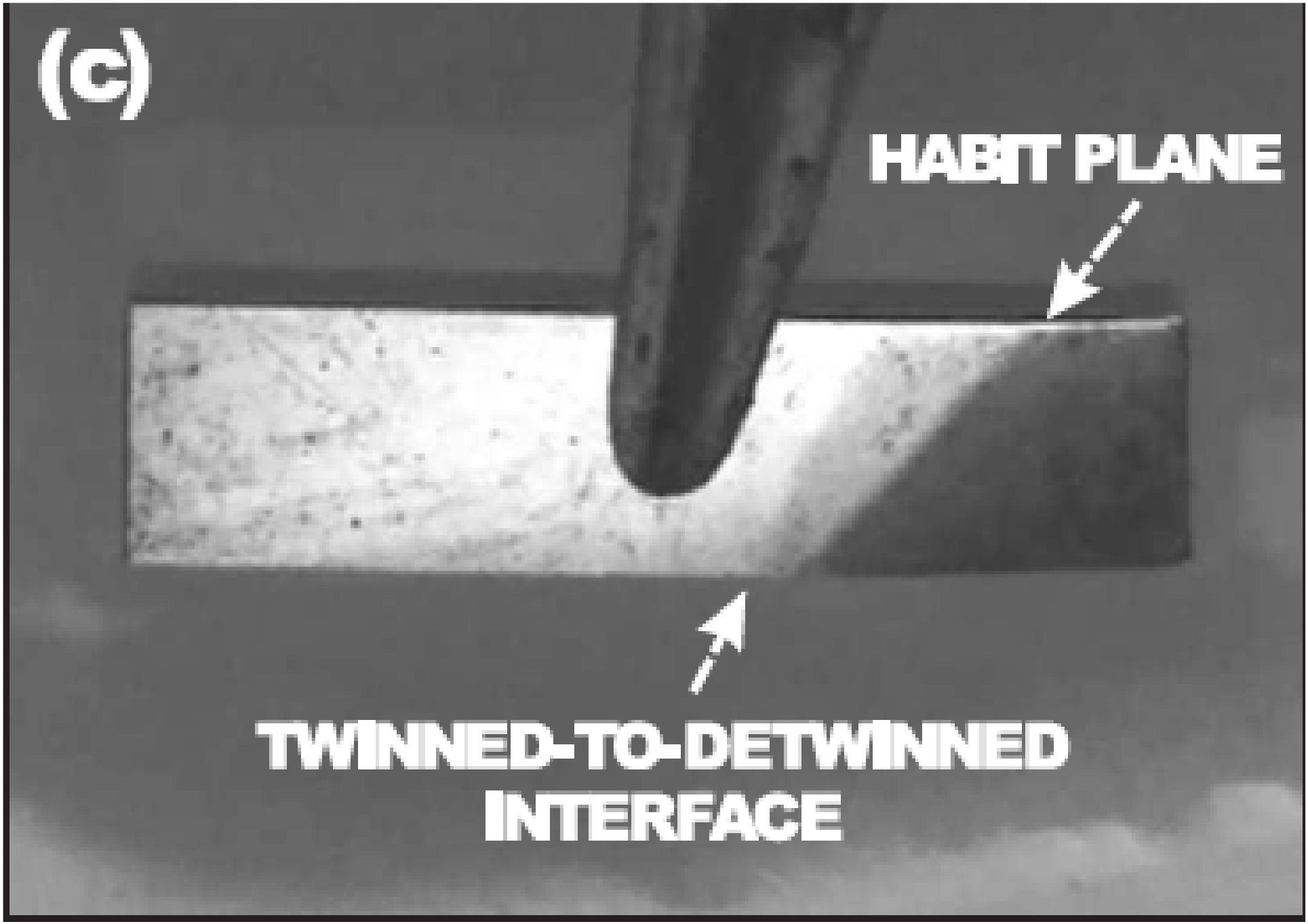
Related work:
Kohn & Sternberg 1989,
Grabovsky & Mengesha 2009

2. Nucleation of austenite in martensite (JB/K. Koumatos/H. Seiner 2013,2014)



Single crystal of CuAlNi. Pure variant of martensite. Heated by tip of soldering iron.


(b)
NUCLEUS


(c)
HABIT PLANE
TWINNED-TO-DETWINNED
INTERFACE

Proposed explanation. Nucleation is geometrically impossible in the interior, on faces and at edges, but not at a corner. We express this by proving in a simplified model that if U_s denotes the initial pure variant of martensite then at U_s the free-energy function is quasiconvex (in the interior), quasiconvex at the boundary faces, and quasiconvex at the edges, but not at a corner.

To make the problem more tractable we assume that $\psi(A, \theta) := W(A)$ is infinite outside the austenite and martensite energy wells.

Idealized model

$$I(\nu) = \int_\Omega \langle \nu_x, \psi \rangle \, dx = \int_\Omega \int_{M^{3\times 3}} \psi(A,\theta) \, d\nu_x(A) \, dx,$$

where

austenite

$$\psi(A,\theta) = \begin{cases} -\delta & A \in SO(3) \\ 0 & A \in \bigcup_{i=1}^{6} SO(3)U_i \\ +\infty & \text{otherwise} \end{cases}$$

martensite

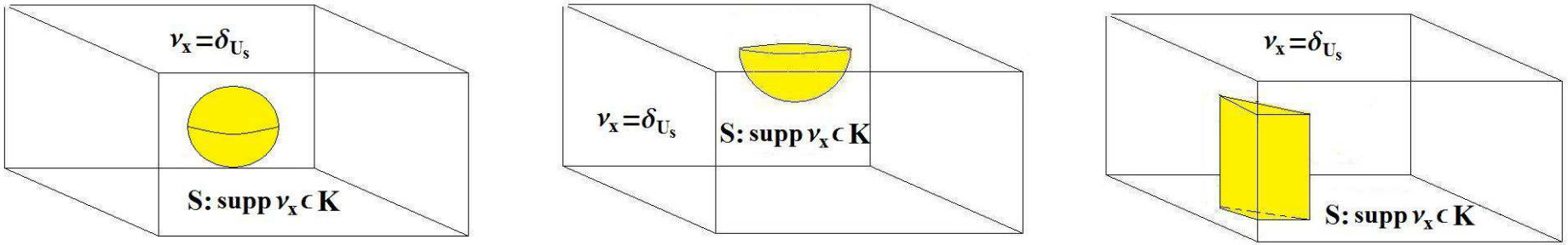
and $\delta > 0$.

So $\psi(A,\theta) < \infty$ on

$$U_1 = \begin{pmatrix} \frac{\alpha+\gamma}{2} & \frac{\alpha-\gamma}{2} & 0 \\ \frac{\alpha-\gamma}{2} & \frac{\alpha+\gamma}{2} & 0 \\ 0 & 0 & \beta \end{pmatrix} \text{ etc.}$$

$$K = SO(3) \cup \bigcup_{i=1}^{6} SO(3)U_i \, .$$

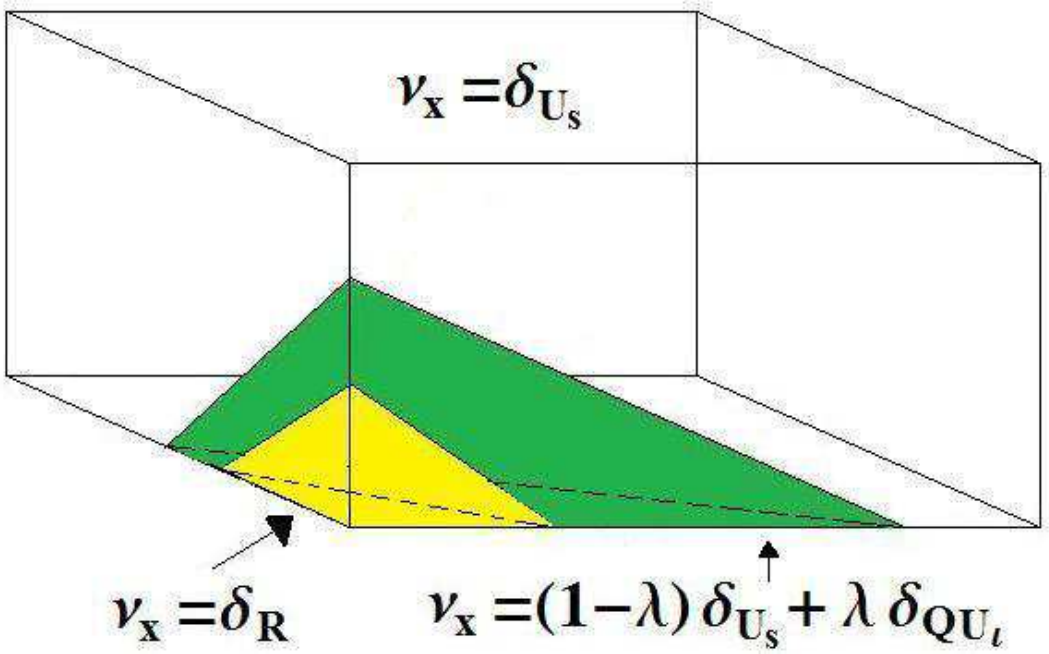
Nucleation impossible in the interior, faces or edges



Theorem $I(\nu) \geq I(\delta_{U_s})$

(quasiconvexity at U_s)

Nucleation possible at a corner



$$I(\nu) < I(\delta_{U_s})$$

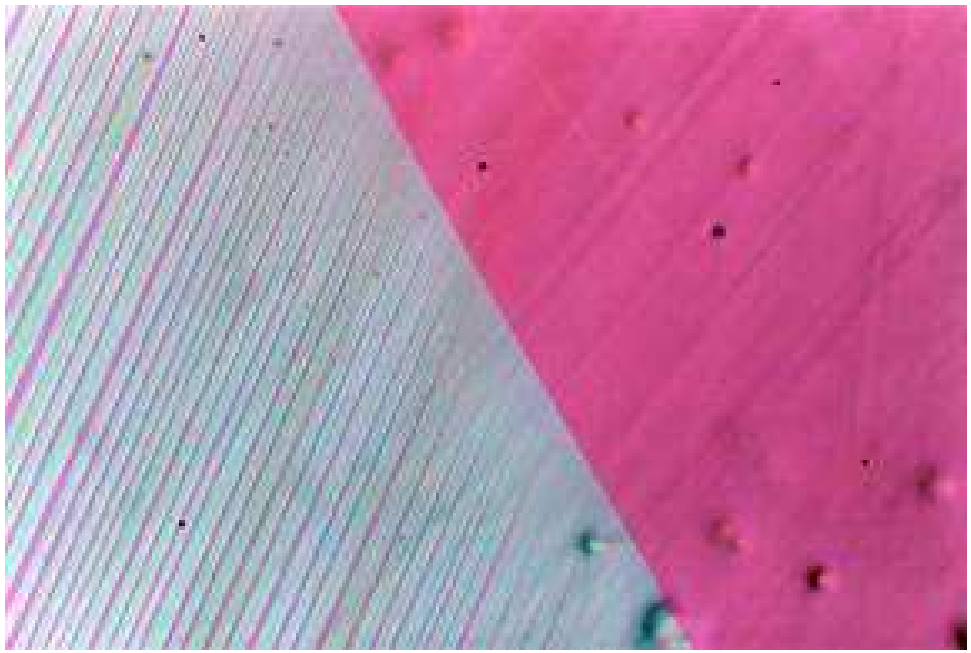
I *not* quasiconvex at such a corner.

cf Grabovsky & Mengesha (2009), Campos Cordero & Koumatos (2016)

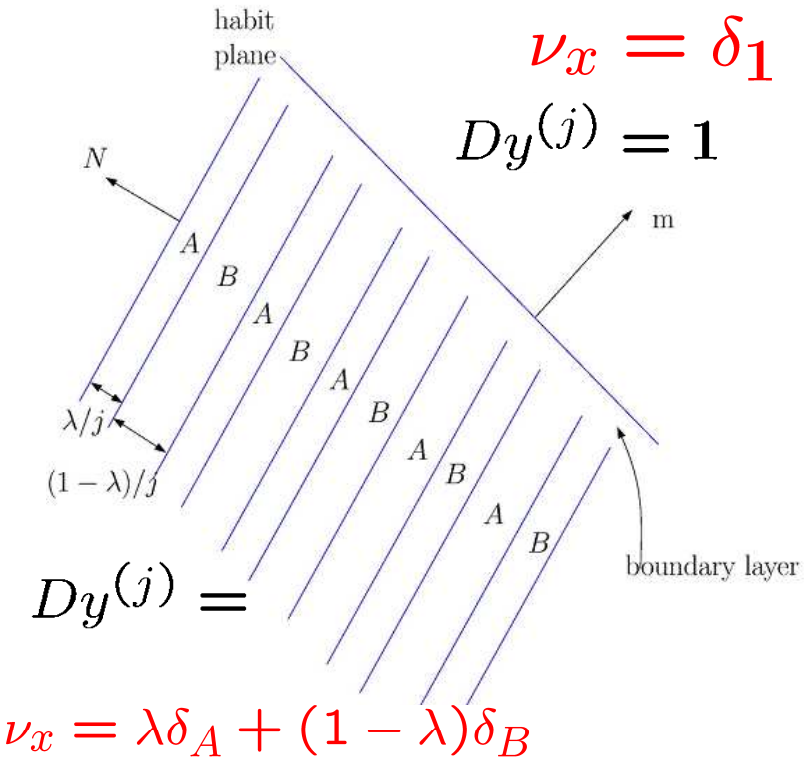
Compatibility and microstructure morphology

1. Nonclassical austenite-martensite interfaces.

Classical interfaces.



(Classical) austenite-martensite interface in CuAlNi
(courtesy C-H Chu and R.D. James)



$$\nu_x = \delta_1$$

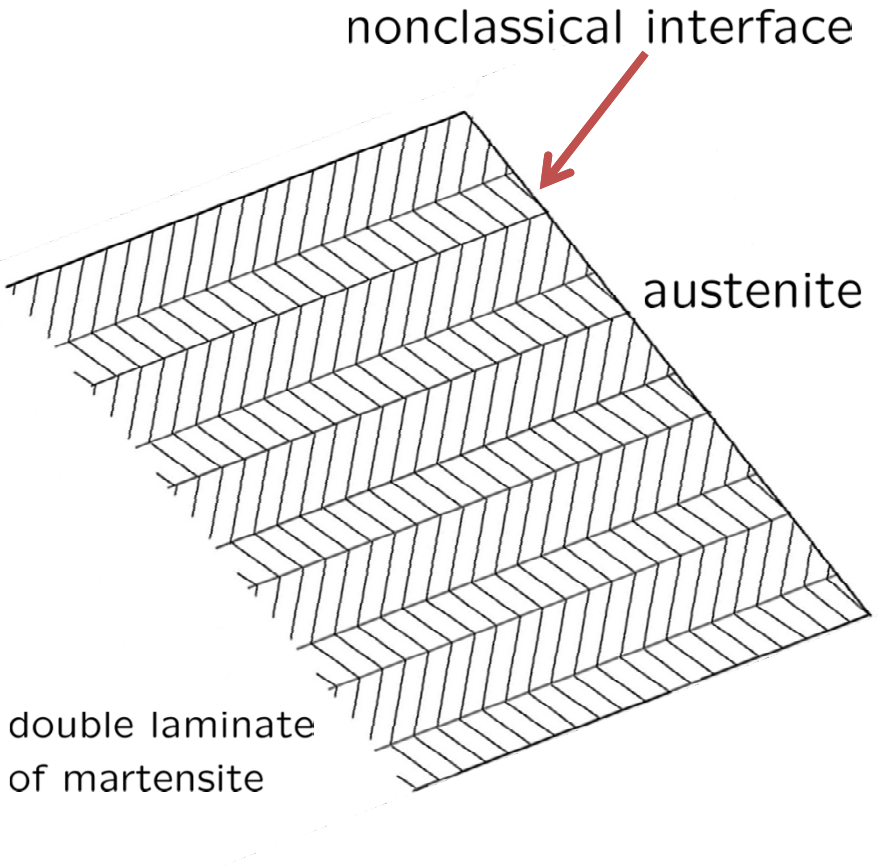
$$Dy^{(j)} = 1$$

$$Dy^{(j)} =$$

$$\nu_x = \lambda\delta_A + (1-\lambda)\delta_B$$

But why should the martensitic microstructure be a simple laminate, rather than something more complicated, such as a double laminate?

This was investigated by JB/C. Carstensen 1997.



$$Dy(x) = 1$$

m

$\nu_x = \delta_1$

$Dy(x) = F = \bar{\nu}$

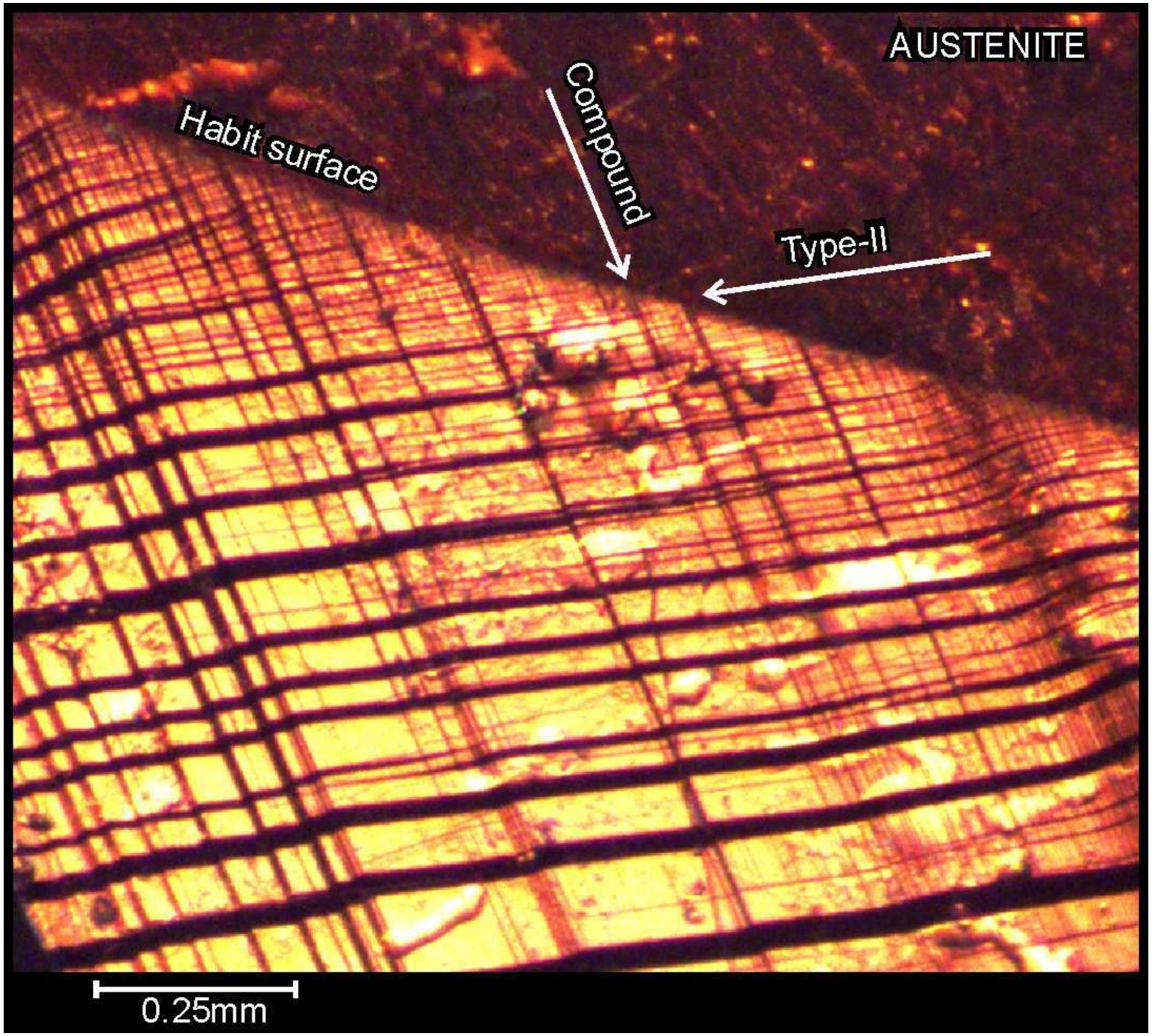
$F \in \left(\bigcup_{i=1}^{M} SO(3)U_i\right)^{\text{qc}}$

(unknown unless $M = 2$)

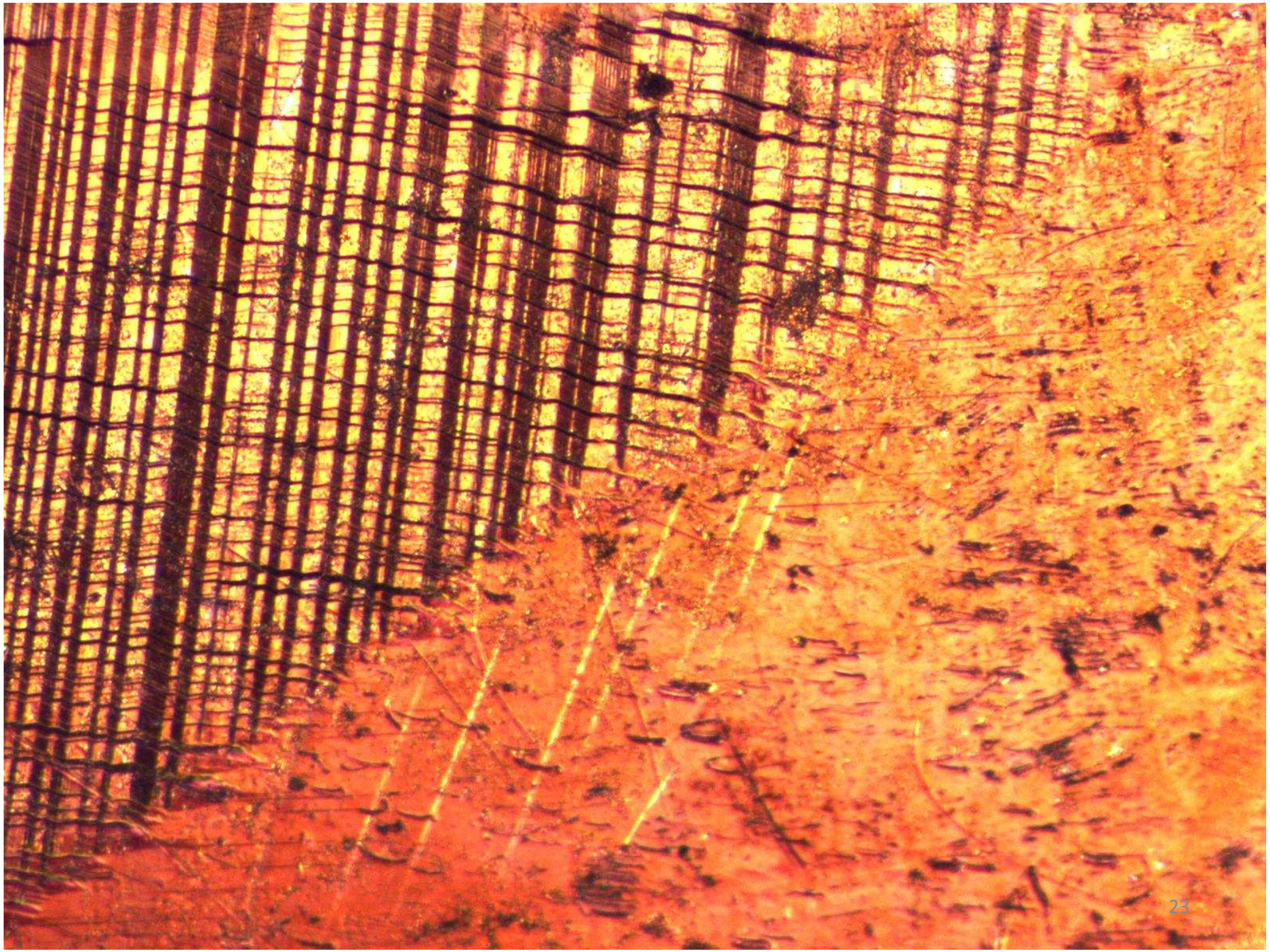
$\nu_x = \nu$

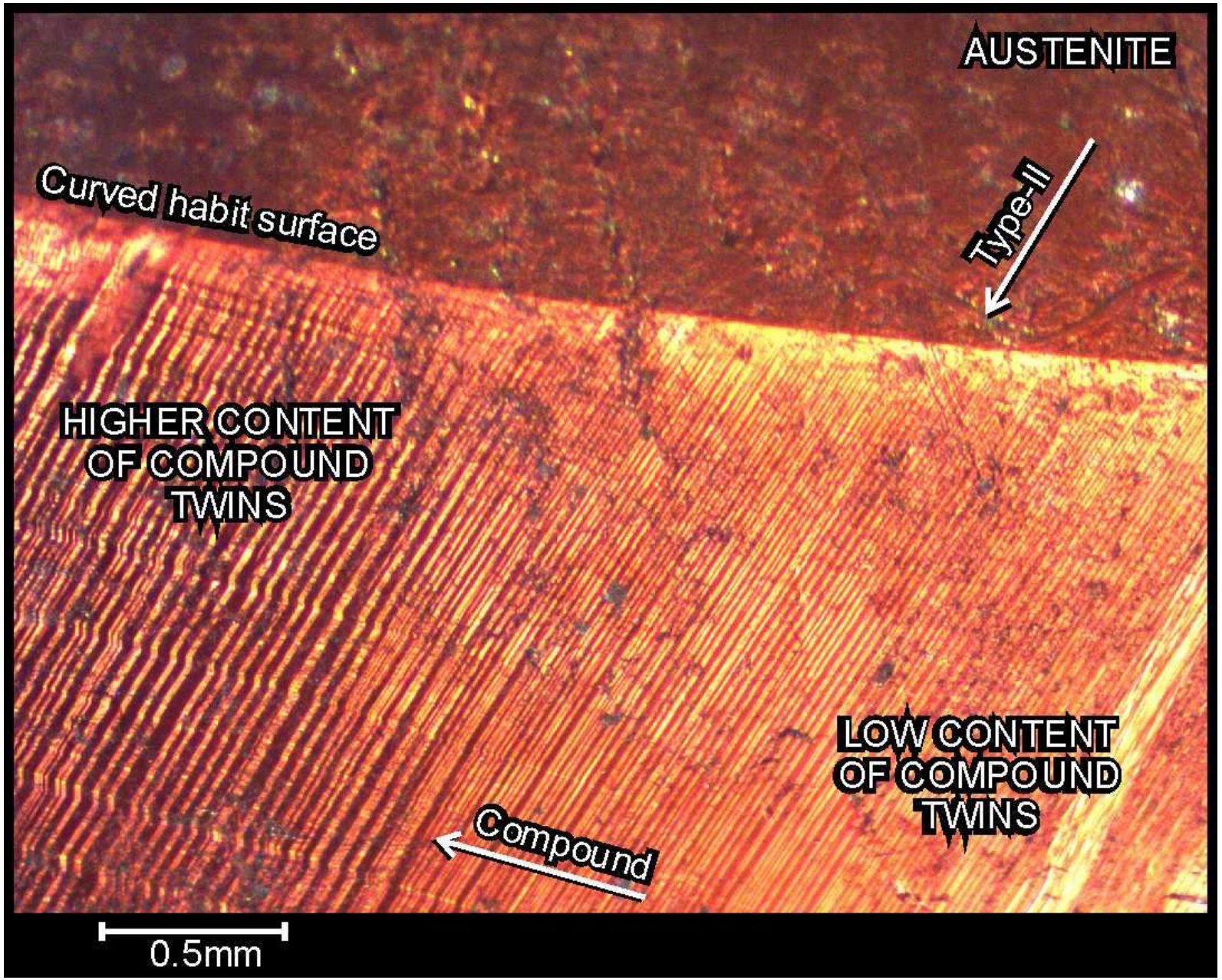
$\operatorname{supp} \nu \subset \bigcup_{i=1}^{M} SO(3)U_i$

$$F = 1 + b \otimes m$$



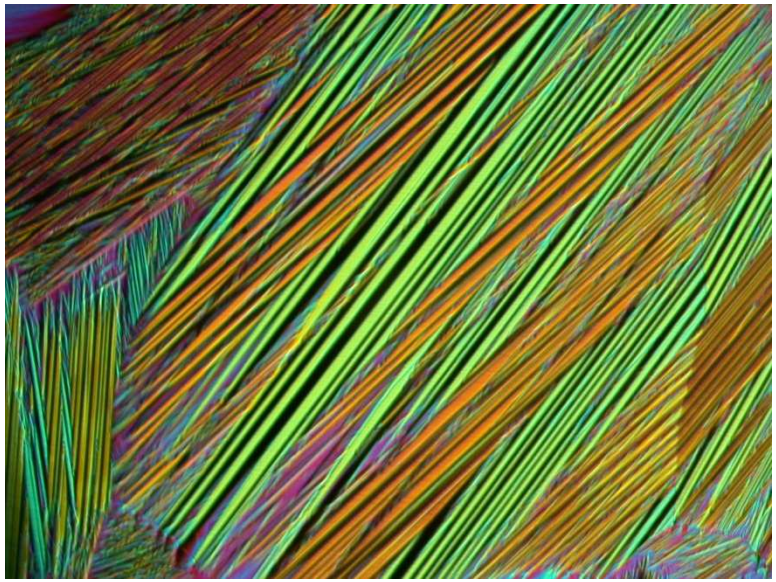
Optical micrograph (H. Seiner) of non-classical interface between austenite and a martensitic microstructure .
The arrows indicate the orientations of twinning planes of Type-II and compound twinning systems



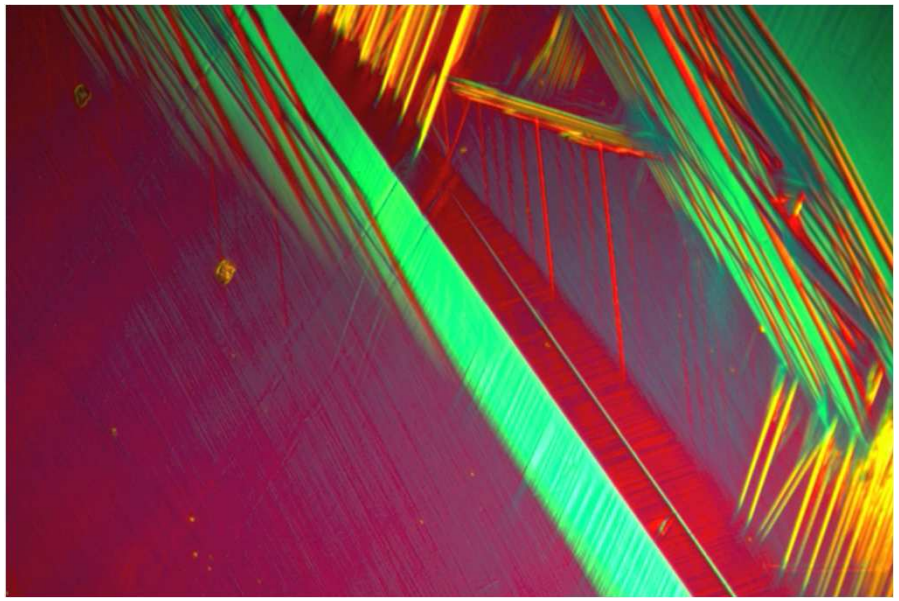


Curved interface between crossing twins and austenite resulting from the inhomogeneity of compound twinning. (Optical microscopy,H. Seiner)

More complex interfaces.



CuZnAl microstructure:
Michel Morin (INSA de Lyon)



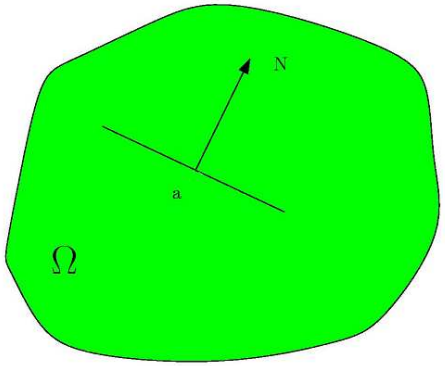
$Zn_{45}Au_{30}Cu_2$ ultra low hysteresis alloy
Song, Chen, Dabade, Shield, James, 2013

Quandt, Wuttig et al 2014

These require generalizations of the Hadamard jump condition, such as (JB/Carstensen):

Theorem

$$0 \in [Dy^{+}(a)(\mathbf{1}-N\otimes N)]^{\mathrm{qc}}-[Dy^{-}(a)(\mathbf{1}-N\otimes N)]^{\mathrm{qc}}.$$



Microstructure in polycrystals (JB/Carstensen)

General aim: to understand microstructure morphology in polycrystals arising from martensitic transformations, by taking seriously compatibility conditions both inside each grain and at grain boundaries.

For this one needs:

(a) an appropriate mathematical model (e.g. nonlinear elasticity with or without interfacial energy)

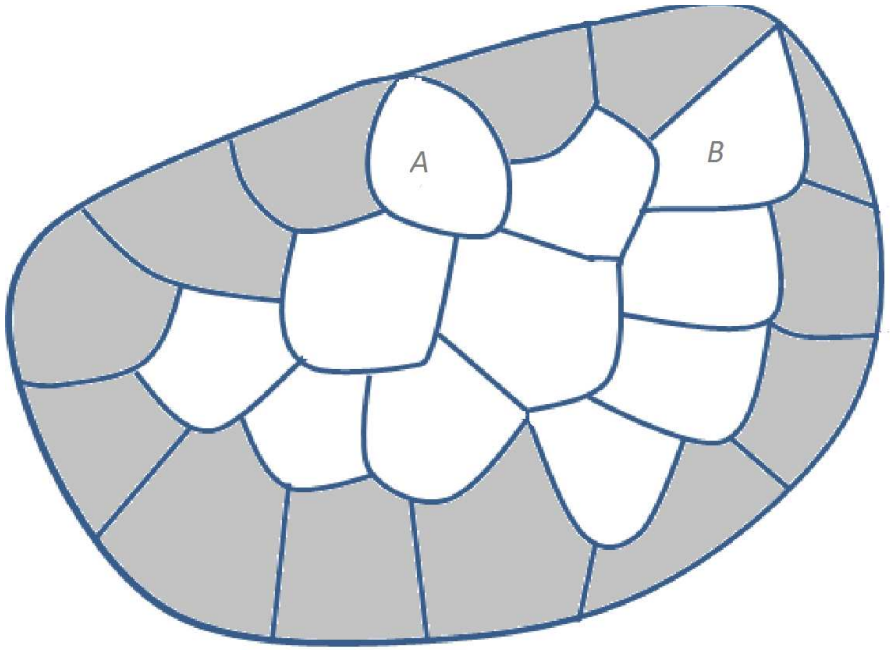
(b) a description of the geometry of grains

(c) ways of applying compatibility at a grain boundary when the microstructure on either side may be complex

Description of grain geometry

Consider a polycrystal that occupies in a reference configuration a bounded domain (open, connected set) $\Omega \subset \mathbb{R}^n$ ($n = 2$ or 3), composed of a finite number of disjoint grains Ω_j, $j = 1, \ldots, N$, where each Ω_j is a bounded domain, so that

$$\Omega = \text{int} \bigcup_{i=1}^{N} \overline{\Omega}_j.$$



Interior grains are ones for which $\partial\Omega_j \subset \bigcup_{k\neq j} \partial\Omega_k$, and the others are *boundary* grains.

A and B are interior grains but touch $\partial\Omega$.

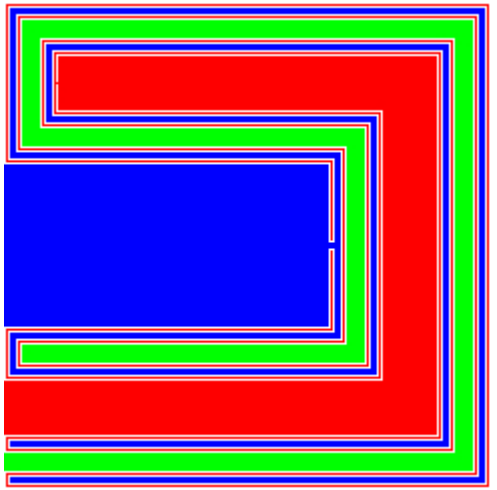
The set of *triple points* is

$$T = \bigcup_{1\leq i_1<i_2<i_3\leq N} \partial\Omega_{i_1} \cap \partial\Omega_{i_2} \cap \partial\Omega_{i_3}.$$

Theorem Suppose each grain Ω_j is convex. Then every interior grain is a convex polyhedron (i.e. an intersection of a finite number of open half-spaces).

Bounds on the set of triple points

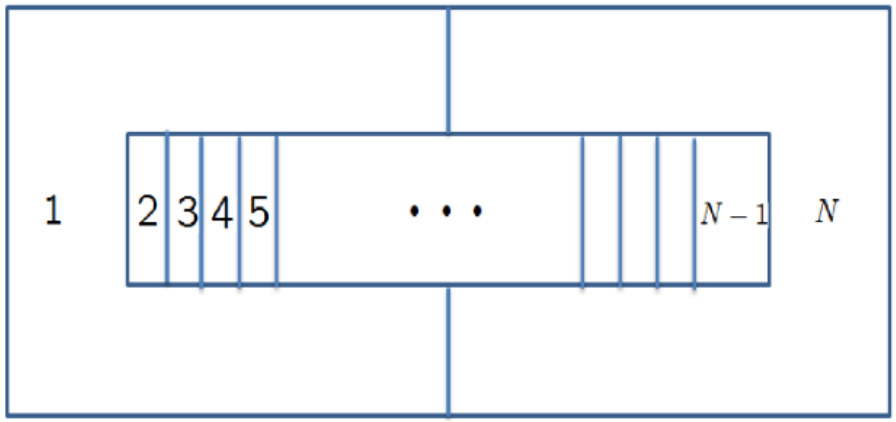
We would like to prove that most points on grain boundaries are not triple points. This is impossible without further conditions on the Ω_j because of the pathological example of the Lakes of Wada (three bounded domains in the plane having a common boundary).



(Wikipedia)

Theorem If $n = 2$ and each grain is the interior of a closed Jordan curve, then there are at most $2(N-2)$ triple points.

The bound is sharp.



The case $n = 3$ is more complicated. One such result is:

Theorem For $n \geq 2$, if each $\overline{\Omega}_j$ is a topological manifold with boundary then T is nowhere dense in $\bigcup_{j=1}^{N} \partial\Omega_j$.

Two results using the nonlinear elasticity model without interfacial energy.

In this model, at a constant temperature the total free energy of the polycrystal in a deformation $y : \Omega \to \mathbb{R}^3$ is given by

$$I(y) = \int_\Omega W(x, \nabla y(x))\, dx,$$

where $W(x, A) = \psi(AR_j)$ for $x \in \Omega_j$, $\psi = \psi(A)$ is the free-energy density corresponding to a single crystal, and $R_j \in SO(3)$.

Suppose we are at a temperature for which the free-energy of the martensite (taken to be zero) is less than that for the austenite. Then $\psi \geq 0$ and

$$K = \{A : \psi(A) = 0\} = \bigcup_{i=1}^{M} SO(3)U_i.$$

Microstructures are described by gradient Young measures $\nu = (\nu_x)_{x\in\Omega}$, with corresponding energy

$$\begin{aligned}\hat{I}(\nu) &= \int_\Omega \int_{M^{3\times 3}} W(x,A)\, d\nu_x(A)\, dx \\ &= \sum_{j=1}^{M} \int_{\Omega_j} \int_{M^{3\times 3}} \psi(AR_j)\, d\nu_x(A)\, dx.\end{aligned}$$

(Here we assume that the grains have sufficiently regular, e.g. Lipschitz, boundaries.)

Zero-energy microstructures thus correspond to ν such that $\operatorname{supp}\nu_x \subset KR_j^T$ for $x \in \Omega_j$.

For cubic-to-tetragonal (more generally for cubic austenite) a result of Bhattacharya on self-accommodation implies that in the absence of boundary conditions on $\partial\Omega$ there is always a zero-energy microstructure with uniform macroscopic deformation gradient

$$\bar{\nu}_x = \int_{M^{3\times 3}} A \, d\nu_x(A) = \nabla y(x) = (\det U_1)^{\frac{1}{3}} \mathbf{1}.$$

How complicated does ν_x have to be?

Cubic to tetragonal: $K = \bigcup_{i=1}^{3} SO(3)U_i$, where

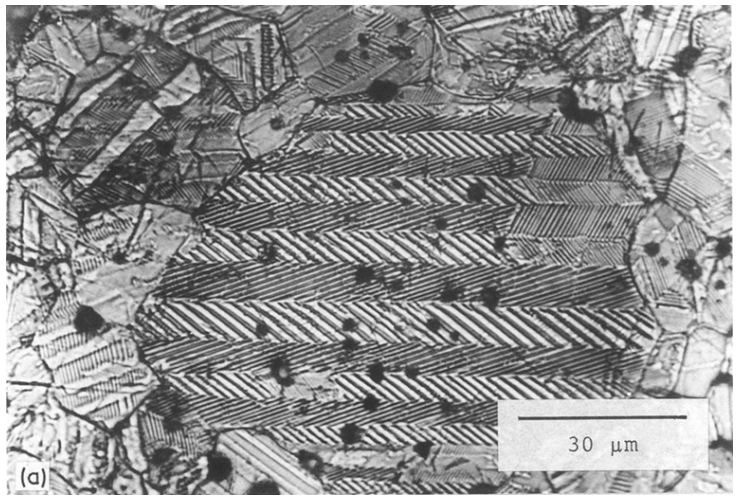
$$U_1 = \text{diag}\,(\eta_2, \eta_1, \eta_1),\ U_2 = \text{diag}\,(\eta_1, \eta_2, \eta_1),$$
$$U_3 = \text{diag}\,(\eta_1, \eta_1, \eta_2).$$

Theorem There is no homogeneous gradient Young measure

$$\nu = \sum_{i=1}^{4} \lambda_i \delta_{A_i}, \quad \lambda_i \geq 0, \sum_{i=1}^{4} \lambda_i = 1,$$

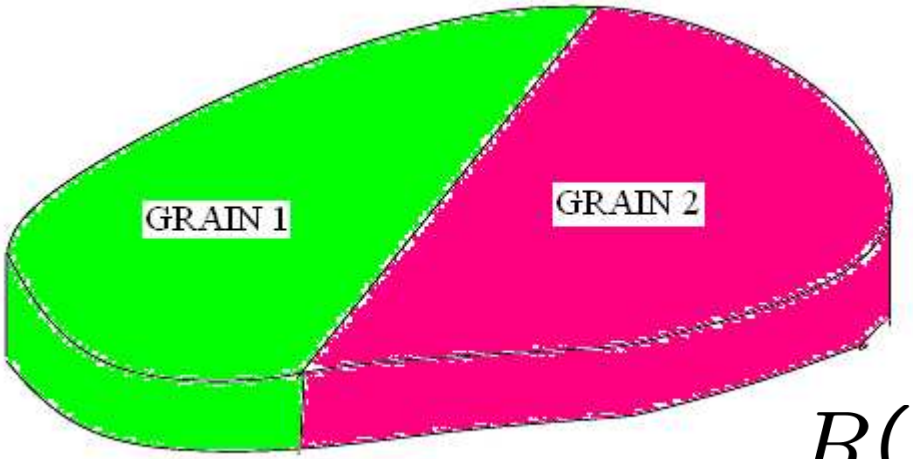
with $A_i \in K$ and $\bar{\nu} = (\eta_1^2 \eta_2)^{1/3} \mathbf{1}$.

Arlt (1990).
Microstructure with approximately four gradients in $BaTiO_3$.



Zero-energy microstructures for a bicrystal

$$K = \mathrm{SO}(3)U_1 \cup \mathrm{SO}(3)U_2$$



Grain 1

$\mathrm{supp}\,\nu_x \subset K$

Grain 2

$\mathrm{supp}\,\nu_x \subset KR(\alpha)$

$R(\alpha)e_3 = e_3$

$\alpha =$ angle of rotation.

Always possible to have zero-energy microstructure with $\nabla y = \bar{\nu}_x = (\eta_1^2\eta_2)^{1/3}\mathbf{1}$

Question: Is it true that whatever the orientation of the planar interface between the two grains there must be a nontrivial microstructure in both grains?

Result 1. Whatever the orientation there always exists a zero-energy microstructure which has a pure phase (i.e. $\nu_x = \delta_A$) in one of the grains.

Now consider the case when the boundary between the two grains has the form $S \times (0, d)$, where S is a smooth curve in the plane, so that the normal at any point is of the form $(\cos\theta, \sin\theta, 0)$.

Result 2. Suppose that $\alpha = \pi/4$. Then it is impossible to have a zero-energy microstructure with a pure phase in one of the grains if the boundary between the grains contains a normal with $\theta \in D_1$ and another normal with $\theta' \in D_2$, where

$$D_1 = (\frac{\pi}{8}, \frac{3\pi}{8}) \cup (\frac{5\pi}{8}, \frac{7\pi}{8}) \cup (\frac{9\pi}{8}, \frac{11\pi}{8}) \cup (\frac{13\pi}{8}, \frac{15\pi}{8})$$

$$D_2 = (\frac{-\pi}{8}, \frac{\pi}{8}) \cup (\frac{3\pi}{8}, \frac{5\pi}{8}) \cup (\frac{7\pi}{8}, \frac{9\pi}{8}) \cup (\frac{11\pi}{8}, \frac{13\pi}{8})$$

Proofs use:

1. A reduction to the case $m = n = 2$ using the plane strain result for the two-well problem (JB/James).

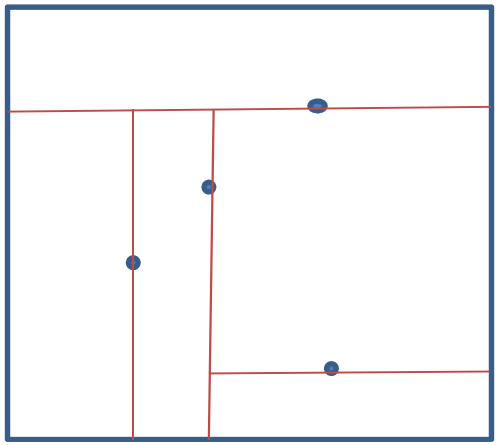
2. The characterization of the quasiconvex hull of two wells (JB/James), which equals their polyconvex hull.

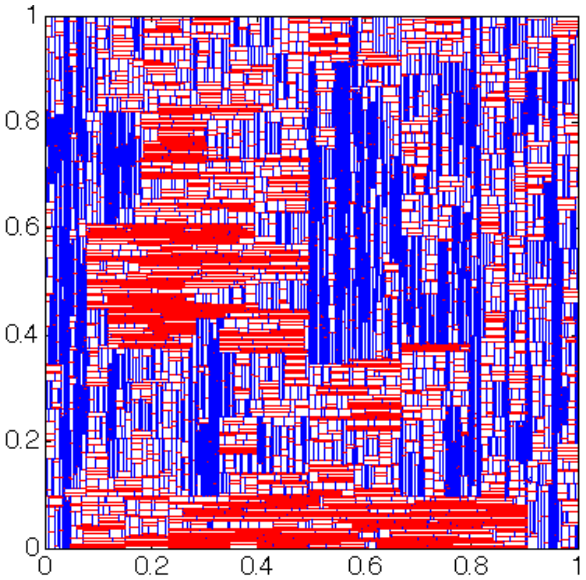
3. Use of a generalized Hadamard jump condition to show that there has to be a rank-one connection $b \otimes N$ between the polyconvex hulls for each grain.

4. Long and detailed calculations.

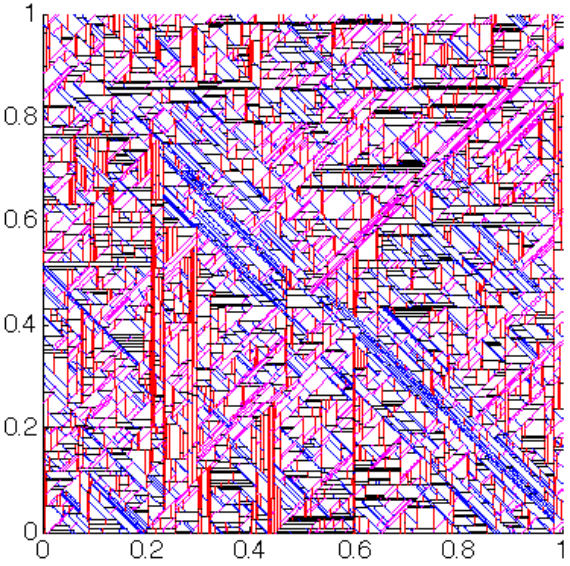
A probabilistic model for martensitic avalanches.

JB/P. Cesana/B. Hambly 2015





2 directions



4 directions

General branching random walk analysis (Cesana/Hambly) predicts approximate power laws for plate lengths, as observed for acoustic emissions.