Martensites: from Crystalline Structures to Macroscopic Properties

Johannes Zimmer

California Institute of Technology

Supported by: MURI
CIMMS

Overview

1. What are Martensites?
2. Crystalline Level: Symmetry and Material Properties

   (with Kaushik Bhattacharya, Sergio Conti, Giovanni Zanzotto)
3. Macroscopic Level
   - Regularization (Capillarity)

     (with Patrick Dondl)
   - Relaxation of the Energy

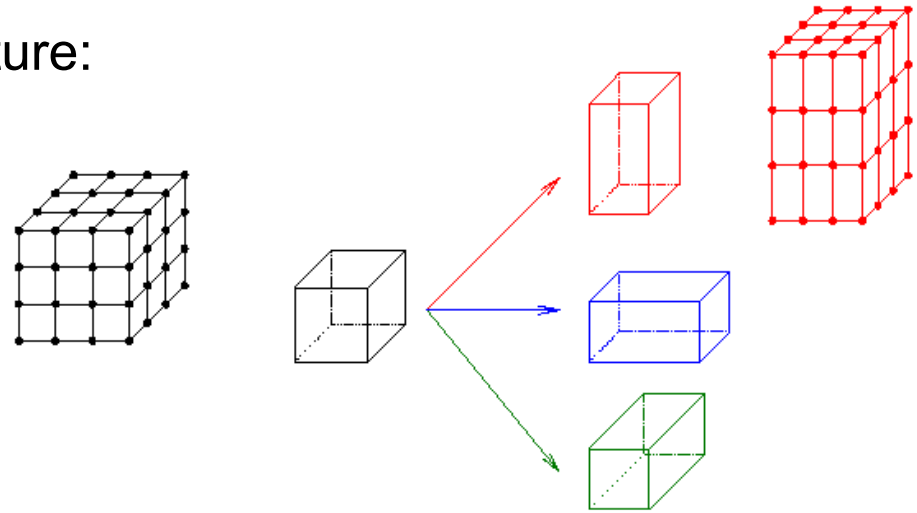
     (with Isaac Chenchiah, Carl Friedrich Kreiner)
   - Dynamics

     (with Marc Oliver Rieger)

1. What are Martensites?

Martensitic transformations: change in crystalline structure
First order phase transition, diffusionless

high temperature:
cubic



low temperature:
tetragonal

1. What are Martensites?

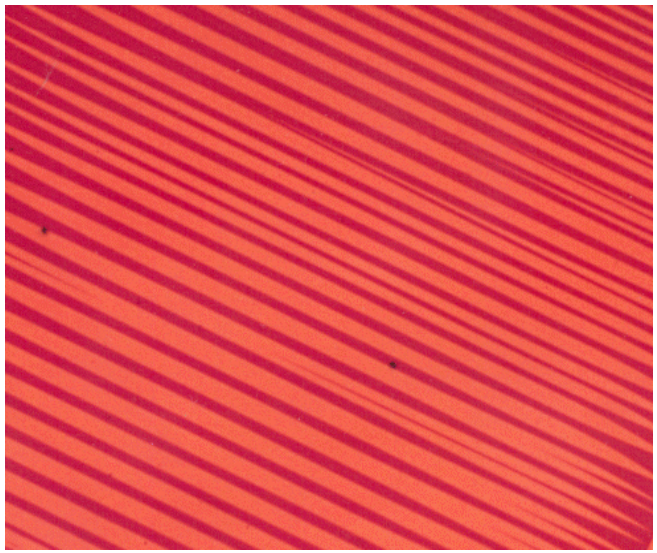


Photo by Chunhwa Chu and Richard James, University of Minnesota

Microscopic level: Formation of microstructure.

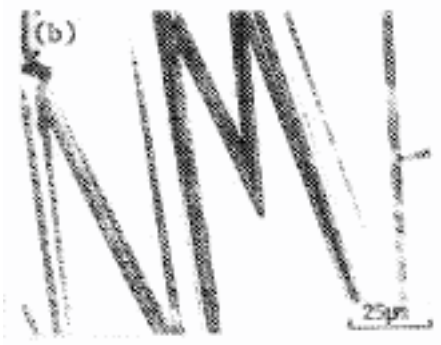
Why does this happen, and what are the consequences?

Overview

1. What are Martensites?
2. Crystalline Level: Symmetry and Material Properties
   (with Kaushik Bhattacharya, Sergio Conti, Giovanni Zanzotto)
3. Macroscopic Level
   - Regularization (Capillarity)
     (with Patrick Dondl)
   - Relaxation of the Energy
     (with Isaac Chenchiah, Carl Friedrich Kreiner)
   - Dynamics
     (with Marc Oliver Rieger)

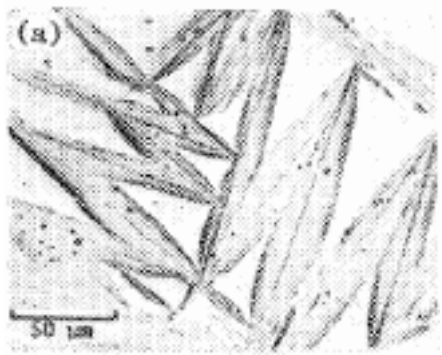
2. Crystallographic Level

1. **Weak martensitic phase transitions**
   Example: Cubic-tetragonal in shape memory alloys
   Characteristics:
   - Reversible phase transformation
   - Little or no dislocation/twinning in parent phase



Fe-31%Ni-0.23%C

2. **Non-weak martensitic phase transitions**
   Example: fcc-bcc in iron (*face-centered cubic* to *body-centered cubic*)
   Characteristics:
   - Irreversible phase transformation
   - Significant dislocation/twinning in parent phase



Fe-29%Ni-0.26%C

Fundamental question in material science:

Why this difference?

We show that group theory can provide an answer.

Crystals will be considered as ideal lattice.
Phase changes change symmetry group of the crystal.

Facts from Mathematical Crystallography: Symmetry of Bravais Lattices

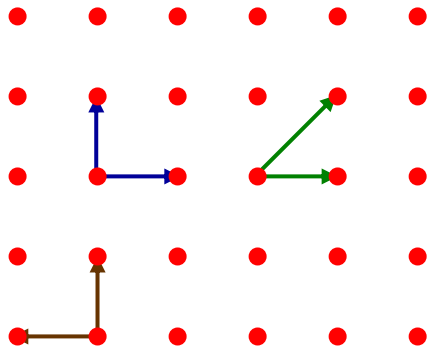
Three linearly independent vectors $\{e_1,\cdots,e_3\}$ in $\mathbb{R}^3$ generate the Bravais lattice

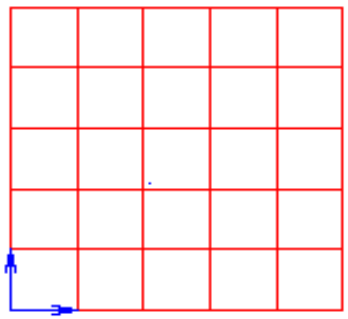
$L(e_1,\cdots,e_3) := \{v \in \mathbb{R}^3 : v = \sum_j \alpha_j e_j,\ \alpha \in \mathbb{Z}^3\}$.

Another basis $\{f_1,\cdots,f_3\}$ generates the same lattice iff $f_j = \sum_k \mu_j^k e_k$ for some $\mu \in GL(3,\mathbb{Z})$,

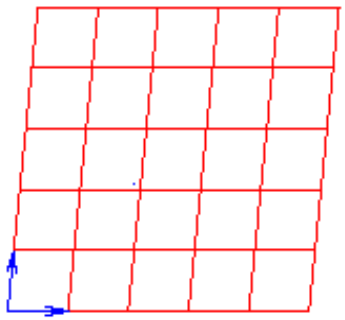
where $GL(3,\mathbb{Z}) := \{\mu \in Mat(3,\mathbb{Z}) : det(\mu) = \pm 1\}$

is the *global symmetry group* (shears and rotations + inversion).





Square reference configuration: $e_1=(1,0)$, $e_2=(0,1)$.



Deformation to f_1, f_2. Assume Cauchy-Born (still a simple lattice).

Deformation gradient is $F = f_1 \otimes e_1 + f_2 \otimes e_2$.

Energy $W = W(F)$.

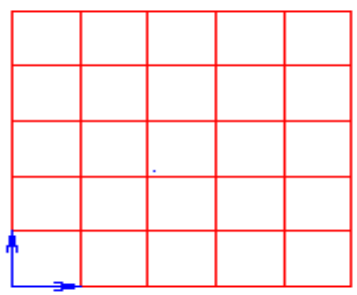
Invariance properties of the elastic energy density

- *Frame invariance*: $W(F) = W(QF)$ for all $Q \in SO(3)$.
  Consequence: Replace lattice basis by lattice metric $C_{ij} := \langle e_i, e_j \rangle$.

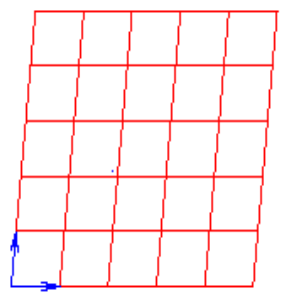
- *Invariance under composition with a change of basis:*
  $W(F) = W(F\mu)$ for all $\mu \in GL(3,\mathbb{Z})$ (for cubic reference cell).
  The action of $GL(3,\mathbb{Z})$ on C is $C^{(e)} = \mu^T C^{(f)} \mu$, where $f_i = \mu^j_i \, e_j$.

(Generic reference lattices: $W(F) = W(QA\mu A^{-1})$ for all $\mu \in GL(3,\mathbb{Z})$;
A depends on reference lattice, A = Id for square/cubic lattice.)

We need to understand orbits in $GL(3,\mathbb{Z})$, or restrict to a suitable subgroup.



Square reference configuration: $e_1=(1,0)$, $e_2=(0,1)$.



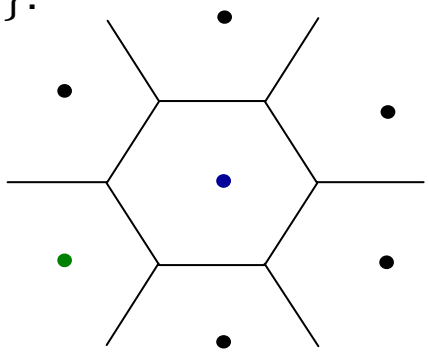
Deformation to f_1, f_2.

Restriction to Ericksen-Pitteri Neighborhoods

Symmetry group $GL(3,\mathbb{Z})$ consists of shears and rotations (+ inversion).

Rotations determine the *point group* of a lattice (self-mappings of lattice):
$P(e_1,\cdots,e_3) := \{Q \in O(3) : Qe_j = \sum_k \mu_j^k e_k \text{ for some } \mu \in GL(3,\mathbb{Z})\}$.

Exclusion of shears is possible if there
is a domain invariant under the point group P
and not under shears (*Ericksen-Pitteri Neighborhoods*, EPN).



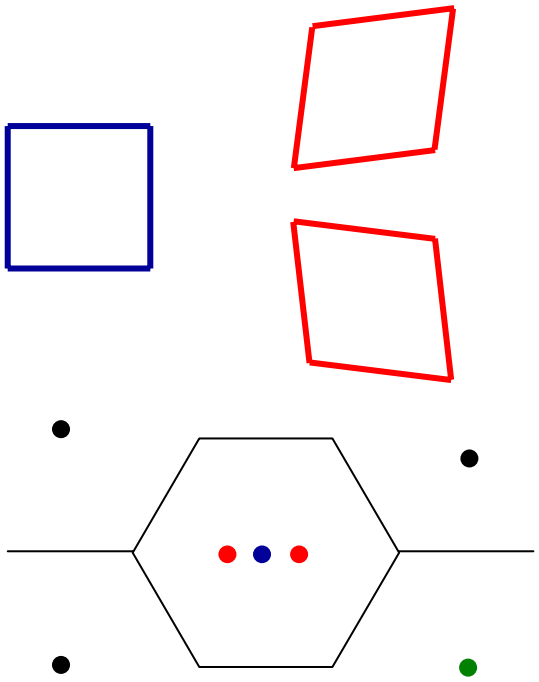
Theorem (Bhattacharya, Conti, Zanzotto, Z., '02). The integral matrices representing the point groups of any pair of phases with maximal symmetry, such as fcc and bcc, generate the entire symmetry group $GL(3,\mathbb{Z})$.

Physical consequences:

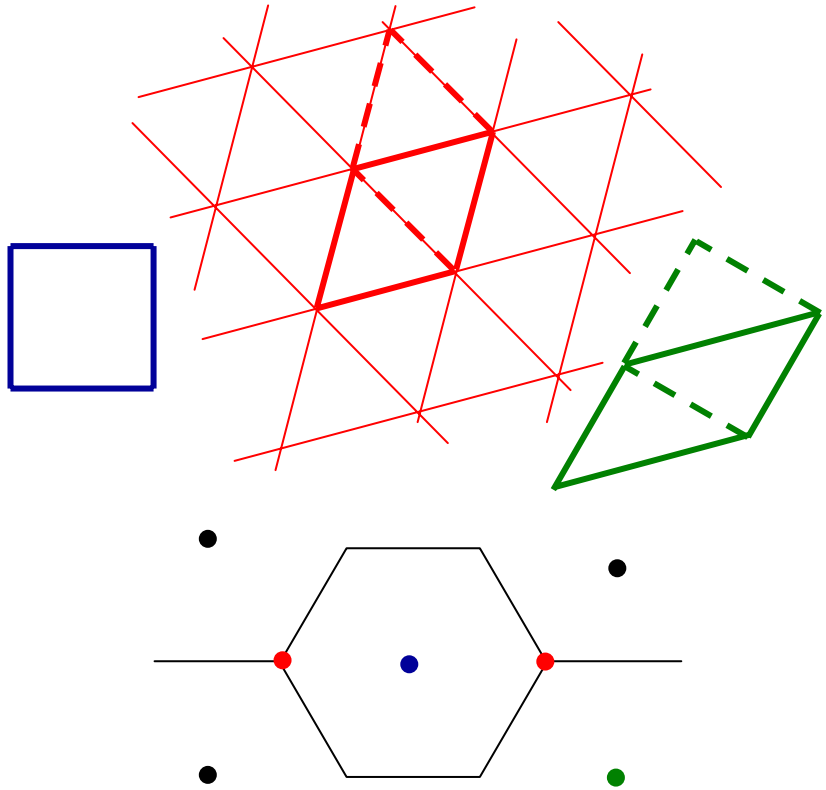
1. Materials with fcc-bcc transition are unable to resist macroscopic shear (fluid-like behavior).
2. **Theorem (Fonseca)**. In this case, the relaxed energy is $W^{qc} = f(\det F)$.

Consequences for phase transitions

Weak martensitic transformation
Product has lesser symmetry



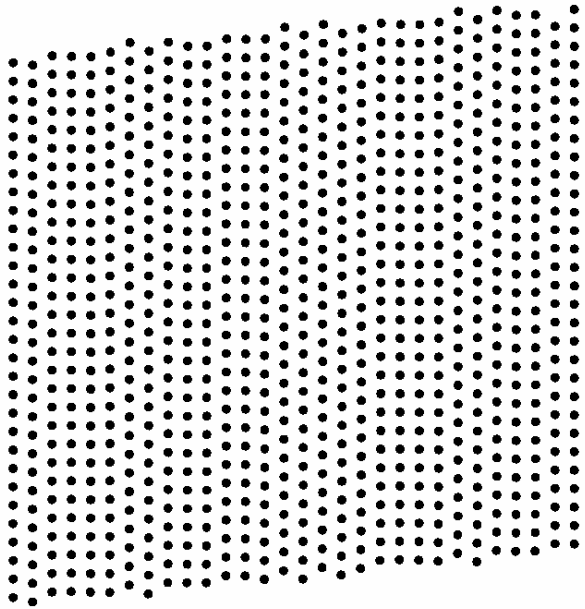
Non-weak martensitic transformation
Product has different symmetry



Recent experimental evidence for successive twinning in during repeated heating-cooling cycle (J. Kornfield et al. for fcc-bcc, Y. Liu et al. for fcc-hcp)

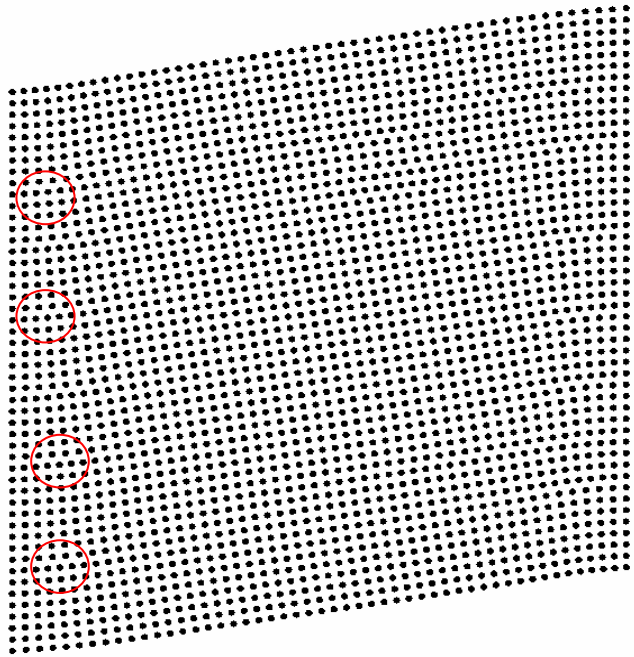
Consequences for phase transitions

Square to rhombus transformation
Product has lesser symmetry



No dislocations but phase transformation

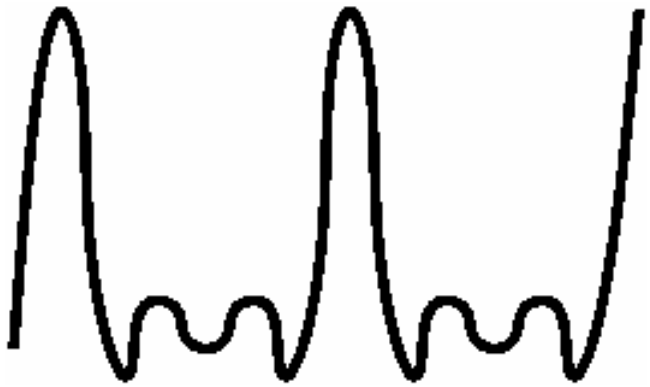
Square to triangle transformation
Product has different symmetry



Plenty of dislocations (red circles) before phase transformation

Consequences for phase transitions

Weak martensitic transformation
Product has lesser symmetry



- Ericksen-Pitteri surgery possible.
- Reversible transformation.

Non-weak martensitic transformation
Product has different symmetry

- Symmetry dictates equal energy barriers.
- Ericksen-Pitteri surgery not possible.
- Irreversible transformation.
- Continuum theory has to take full $GL(3,\mathbb{Z})$ into account.

Overview

1. What are Martensites?
2. Crystalline Level: Symmetry and Material Properties

   (with Kaushik Bhattacharya, Sergio Conti, Giovanni Zanzotto)
3. Macroscopic Level
   - Regularization (Capillarity)

     (with Patrick Dondl)
   - Relaxation of the Energy

     (with Isaac Chenchiah, Carl Friedrich Kreiner)
   - Dynamics

     (with Marc Oliver Rieger)

First Continuum Approach: Regularization

Continuum level: Energy still nonconvex (several low-symmetry phases).
Consequence: Standard analytical and numerical methods won't work.
One way out: regularize. Here: finite elasticity, diffuse interface model:

$$u_{tt} = \mathrm{Div}(\sigma(\nabla u)) + \beta \Delta u_t - \Delta^2 u + f$$

(Balance law for momentum, added capillarity and optional viscosity; u: $\mathbb{R}^n \times [0,T] \to \mathbb{R}^n$ for n=2,3 is the displacement and $\sigma(F)$ the stress tensor (derivative of energy W(F)), complemented with I.C. and B.C.)

Theorem (Dondl, Z., '02). If W(F) is C^2 and grows quadratically for large strains, then there exists a solution $u \in H^1_0(\Omega) \cap H^2(\Omega)$.

Sketch of proof:

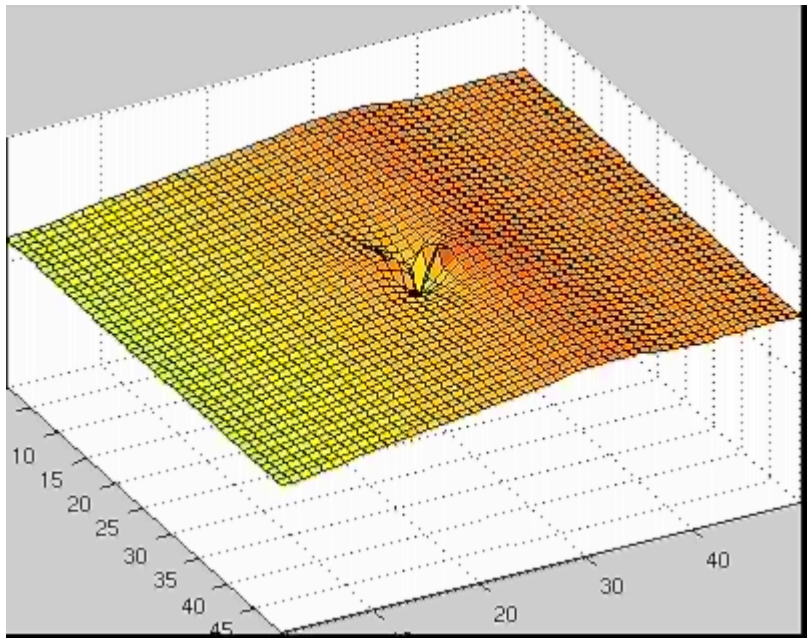
- Rewrite as semigroup for $(u(t), v(t))^T$.
- Use that the operator obtained in this way generates an analytic contraction semigroup for $\beta > 0$ (unitary group for $\beta = 0$).
- Elliptic regularity to get Lipschitz continuity of right-hand side.

Numerical simulations I

- Fully 2D computations
- Finite elements: Bogner-Fox-Schmit (C^1) to resolve higher derivatives
- Square domain
- Dynamic problem
- Parallelization ongoing

Results:

1. Interaction with elastic obstacle (Two-well potential with non-transforming defect)



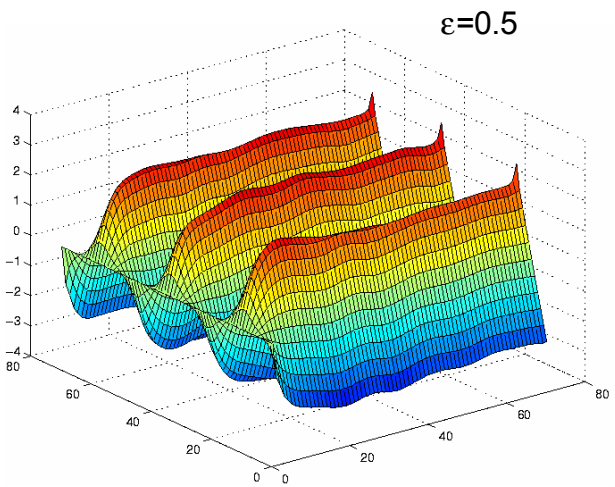
Numerical simulations II

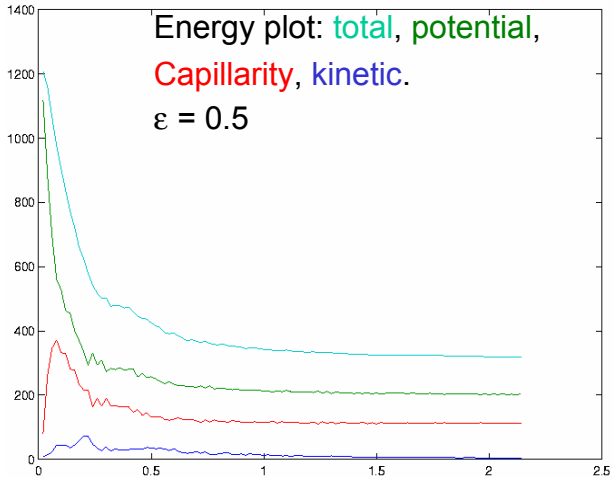
2. Scaling law: Capillarity of order ε, domain (0,1)× (0,L).

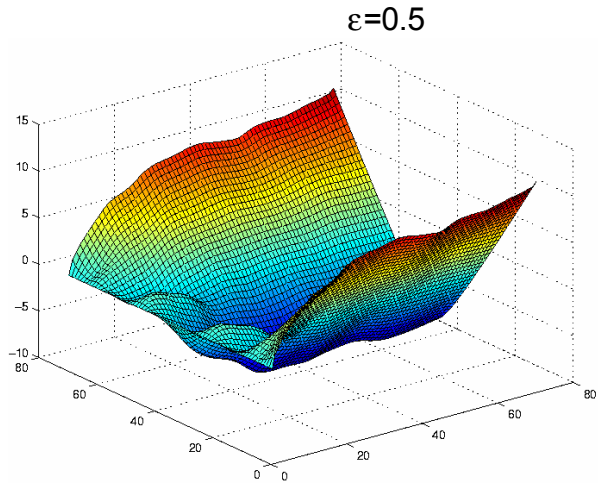
Naïve dimensional analysis: Scaling of length for phase width is $E^\epsilon \sim C\epsilon^{\frac{1}{2}} L^{\frac{1}{2}}$.

Kohn/Müller, 1994: Analytic prediction of regime with a scaling relation $E^\epsilon \sim C\epsilon^{\frac{2}{3}} L^{\frac{1}{3}}$.
(refinement at the boundary).

Simulation: 70x70 grid, u simply supported/clamped, $W(\nabla u) = (u_y^2-1)^2 + u_x^2$







Strong impact of the regularization on the (dynamics of the) system.
Insight from models without additional regularizing terms?

Overview

1. What are Martensites?
2. Crystalline Level: Symmetry and Material Properties

   (with Kaushik Bhattacharya, Sergio Conti, Giovanni Zanzotto)
3. Macroscopic Level
   - Regularization (Capillarity)

     (with Patrick Dondl)
   - Relaxation of the Energy

     (with Isaac Chenchiah, Carl Friedrich Kreiner)
   - Dynamics

     (with Marc Oliver Rieger)

Second Continuum Approach: Nonconvex Envelopes of the Energy

Fundamental problem in elasticity:
Find deformation u: $\Omega \subset \mathbb{R}^n \to \mathbb{R}^n$ that minimizes

$$I(u) := \int_\Omega W(Du)\mathrm{d}x$$

subject to suitable boundary conditions. Here, W is the (macroscopic) energy density.

The problem has a solution only if W is *quasiconvex*, that is,

$$\int_\Omega W(F)\mathrm{d}x \leq W(F + D\phi)\mathrm{d}x$$
$$\forall F \in \mathsf{Mat}(n,\mathbb{R}), \phi \in C_0^\infty(\Omega, \mathbb{R}^n).$$

Fundamental Theorem in relaxation theory: under suitable conditions,

$$\min_{u\in\mathcal{A}} \int_\Omega W^{\mathrm{qc}}(Du)\mathrm{d}x = \inf_{u\in\mathcal{A}} \int_\Omega W(Du)\mathrm{d}x.$$

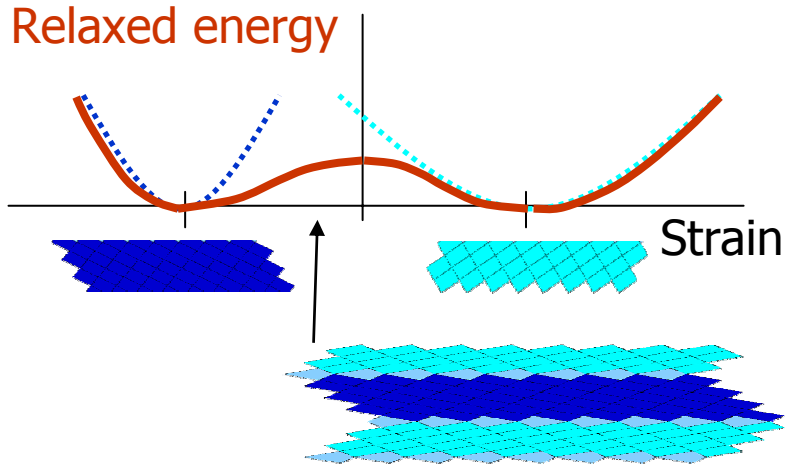
No oscillations

No attainment: oscillating minimizing sequences, weak convergence

Numerics: oscillations of scale of discretization

Problem: relaxed (effective) energy W^{qc} very difficult to compute.
Other areas of application: optimal design and homogenization.

Second Continuum Approach: Nonconvex Envelopes of the Energy



Multiscale problem:

- Microstructures on microscopic level
- Averaging on mesoscopic level, represented by relaxed energy (quasiconvex envelope)

Approximations of quasiconvex envelope:

1. Rank-1-convexity (:= convexity along rank-1-lines). It is known that $W^c \leq W^{qc} \leq W^{rc} \leq W$; often, one has $W^{qc} = W^{rc}$.

Still, rank-one convexity is hard to compute. Consider first another approximation:

2. Separate convexity (:= convexity in direction of base vectors) [Tartar, Ball, Kohn]

Goal: Compute rank-1-convex (separately convex) hull of sets A

$A^{rc} := \{B \in \mathrm{Mat}(n,\mathbb{R}) : f(B) = 0$ for every rk-1-convex f with $f(A) = 0\}$.

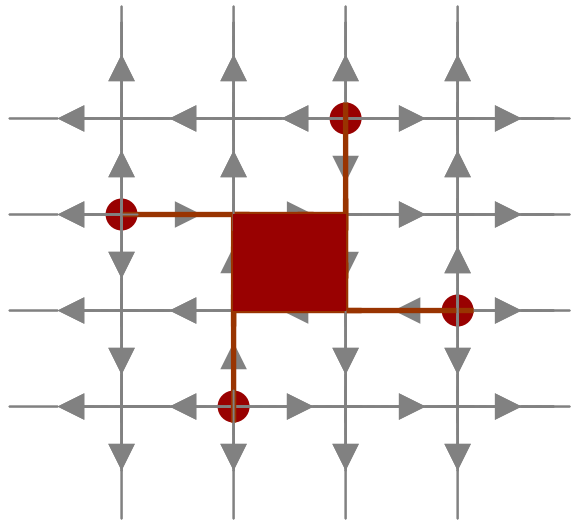
Separately Convex Hulls: A Graph-Theoretical Approach

Algorithm.

Input: Finite set A in $\mathbb{R}^n$ (think of A as set of matrices).

Output: Separately convex hull of A.

1. Define coordinate set $x_j(A) := \{x_j(A) : a \in A\}$ and grid(A) := $x_1(A) \times \cdots x_n(A)$.
2. Construct graph:
   - Vertices := grid points
   - Edges:= sep. convex lines between neighboring grid points.
   - Orientation: on edges entering or leaving points in A (pointing away from A).
3. Search for loops. Loops + interior + points with connections leading to loops belong to sep. convex hull.
4. Update orientation in 2., enlarged by points obtained in 3.



Theorem (Chenchiah, Kreiner, Z. '02). This algorithm computes the separately convex hull.

Towards Rank-1-Convex Hulls

Previous work: Dolzmann 1999, Aubry, Fago, Ortiz 2002
General strategy:

1. Guess direction in which you want to convexify (optional).
2. Discretize space and rank-1-directions. Apply successive convexifications.

Advantage: Works very well if one has information about micostructure.

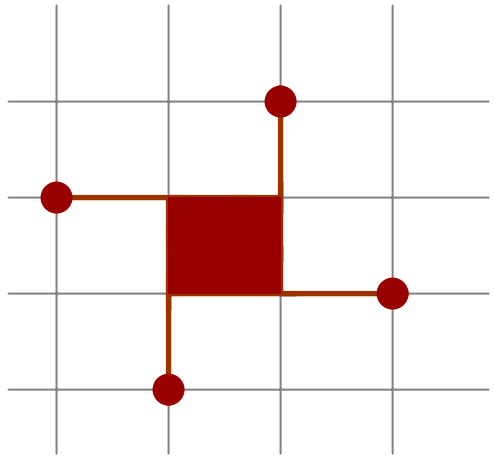
Drawbacks:

1. High complexity, computationally expensive.
2. Quality of the approximation depends on orientation of the grid.
3. Often it is hard to find starting point; algorithm might fail if crucial points are missing.

Our approach: make explicit use of mathematical structure.

One approach, ongoing:

- Rank-1 lines are algebraic objects (determinantal varieties), as are their intersections.
- Use tools from computational algebraic geometry.



Towards Rank-1-Convex Hulls

Idea: Use structure of rank-1-convexity. Here: Krein-Milman type theorems.

Definition. A point $e \in A$ is *rank-1-extreme* if it is not in the interior of a rank-1 line in A.

Lemma (Chenchiah, Kreiner, Z. '02). Suppose $A \in \mathbb{R}^d$ is compact. Then the extreme points of the rank-1-convex hull of A are in A.

Current work, inspired by work by Matousek and Plechac on separate convexity:

- Start with grid comprising A.
- Compute rank-1-extreme grid points. Discard if not in A.
- Consider intersections of (discretized) rank-1-cones emanating from extreme points.
- Examine extensibility of rank-1-convexity from cone to entire space (might require complicated interpolation).

Overview

1. What are Martensites?
2. Crystalline Level: Symmetry and Material Properties

   (with Kaushik Bhattacharya, Sergio Conti, Giovanni Zanzotto)
3. Macroscopic Level
   - Regularization (Capillarity)

     (with Patrick Dondl)
   - Relaxation of the Energy

     (with Isaac Chenchiah, Carl Friedrich Kreiner)
   - Dynamics

     (with Marc Oliver Rieger)

Third Continuum Approach: Dynamics

1. Regularized dynamics: e.g., purely thermoviscous system (no capillarity)

$$\begin{aligned} u_{tt} &= \mathrm{Div}\,(\sigma(\nabla u, \theta) + \nabla u_t) \quad \text{in } \Omega\times]0,T[, \\ \theta_t &= \Delta\theta + \theta\sigma_\theta(\nabla u, \theta)\times\nabla u_t + \nabla u_t \times \nabla u_t \quad \text{in } \Omega\times]0,T[. \end{aligned}$$

Three-dimensional thermoviscoelastic bar with constant density, nonconvex Helmholtz free energy density.

(Balance laws of momentum and energy; u is the displacement and θ the temperature)

Difficulties:

- Several space dimensions (very low regularity of the strain).
- Viscosity allows to deal with nonconvexity of the energy, but introduces strong nonlinearity in heat equation.

Theorem (Z., '00). Under suitable growth assumption on the energy, there exists a weak renormalized solution.

Third Continuum Approach: Dynamics

2. Dynamics without regularization:

Dynamics of a one-dimensional thermoelastic bar with constant density, nonconvex Helmholtz free energy density.

Balance laws of momentum and energy:

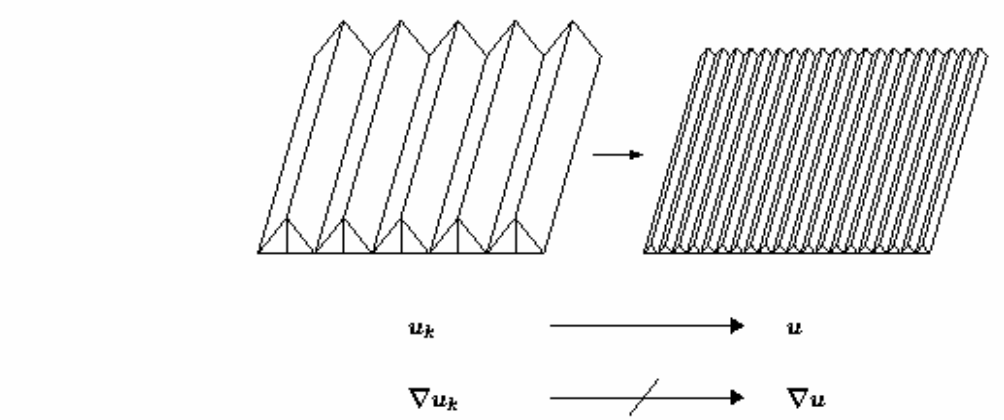
$$\begin{aligned} u_{tt} - (\phi(u_x) + \alpha_2 \theta u_x)_x &= f, \\ \alpha_1 \theta_t - \kappa \theta_{xx} - \alpha_2 \theta u_x u_{xt} &= g. \end{aligned}$$

Here u: $\mathrm{I}\times[0,\mathrm{T}] \to \mathbb{R}$ is the displacement,
θ: $\mathrm{I}\times[0,\mathrm{T}] \to \mathbb{R}$ the absolute temperature

Mathematical difficulties arise from nonconvexity of the potential and the nonlinearity of the equations.

Young measures I

The concept of (gradient) Young measures:



A *Young measure* ν gives a "one-point-statistics" for $\{\nabla u_k\}_k$.

In the example: $\nu = \frac{1}{2}(\delta_{(-1,0)} + \delta_{(+1,0)})$.

50% probability each that $\nabla u_k(x)=(-1,0)$ or $(+1,0)$.

Intuitively: Young measure solutions to nonconvex PDEs can be defined by replacing the variable with oscillations by a Young measure.

Young measures II

Definition: A *Young measure* is a family of probability measures $\{\nu_x\}_{x\in\Omega}$ on $\mathbb{R}^N$ associated with a sequence of measurable functions $\{f_j\}_j$ with f_j: $\Omega\subset \mathbb{R}^n \to \mathbb{R}^N$ such that for any continuous function $\phi:\mathbb{R}^N\to$ R the function

$$\overline{\phi}(x) = \int_{\mathbb{R}^N} \phi(F)\, d\nu_x(F) =: \langle \nu_x, \phi \rangle$$

is measurable, and such that for every weakly-converging sequence $\{f_j\}_j$ we have

$$(\phi(f_j))_j \rightharpoonup \overline{\phi}.$$

A *gradient Young measure* is a Young measure generated by a sequence $f_j=\nabla u_j$, where $u_j \in W^{1,2}(\Omega)$.

Young Measure Solution for Thermoelasticity

Definition: For thermoelasticity, we define a *Young measure solution* in the following way:

Let $T > 0$, let
$u \in W^{1,\infty}(0,T),L^2(I)) \cap L^\infty((0,T),W^{1,6}(I))$,
$\theta \in W^{1,\infty}((0,T),L^1(I)) \cap L^2(0,T),H^1(I))$.

and let ν be a gradient Young measure with $\langle Id,\nu\rangle = u_x$ a.e., then (u,ν,θ) is a *Young measure solution* if for all $\xi,\zeta \in H_0^1((0,T)\times I)$:

$$\int_0^T \int_I (u_t\xi_t - u_x\theta\xi_x - \langle\phi,\nu\rangle\xi_x)\,\mathrm{d}x\,\mathrm{d}t = -\int_0^T \int_I f\xi\,\mathrm{d}x\,\mathrm{d}t,$$

$$\int_0^T \int_I \left(\ln\theta\,\zeta_t + \theta_{xx}\frac{\zeta}{\theta} - \frac{1}{2}\langle|\cdot|^2,\nu\rangle\zeta_t\right)\mathrm{d}x\,\mathrm{d}t = -\int_0^T \int_I \frac{g}{\theta}\zeta\,\mathrm{d}x\,\mathrm{d}t.$$

Approximate Solutions

Consider a sequence of approximating systems:

$$\begin{aligned} u_{tt} - (u_x\theta - \phi(u_x))_x - \epsilon u_{xxxx} &= f, \\ \theta_t - \theta_{xx} + \theta u_x u_{xt} &= g. \end{aligned}$$

(Regularization as in First Continuum Approach!)

Assume bound $\theta(x,t) > \theta_{min} > 0$ for all $x \in I$, $t \in [0,T]$ uniform in ε.

(Exclude absolute temperatures close to zero, where classical equations of thermoelasticity do not hold).

Global Existence for Thermoelasticity

Theorem (Rieger, Z. '02). Suppose $u_0, u_1, \theta_0 \in H^1(I)$ and $f, g \in L^2(L^2(I))$. Then there exists a Young measure solution (u, ν, θ) to (1)-(2), provided the a priori assumption on the temperature bounds holds and the sequence θ^ε converges strongly in t.
The solution is global in time, and the initial data can be arbitrarily large.

Sketch of proof:

1. Prove a priori bounds for approximate systems, independent of epsilon.
2. Passage to the limit.
3. Show limiting objects give Young measure solution.

Main difficulty: no control of higher order derivatives.

Some Details of the Proof I

1. Energy estimate:

$$\max_{t\in[0,T]} \int_I \left[(u_t^\epsilon)^2 + \Phi(u_x^\epsilon) + \theta^\epsilon + \epsilon(u_{xx}^\epsilon)^2\right] \mathrm{d}x + \int_0^t \int_I |\theta_x^\epsilon|^2 \,\mathrm{d}x\,\mathrm{d}s \;\leq\; C.$$

This implies convergence of

$$\begin{aligned} u^\epsilon &\rightharpoonup u \text{ in } L^\infty(W^{1,6}(I)) \cap W^{1,\infty}(L^2(I)), \\ \theta^\epsilon &\rightharpoonup \theta \text{ in } L^2(H^1(I)) \cap L^\infty(L^1(I)), \\ \theta_{xx}^\epsilon &\rightharpoonup \theta_{xx} \text{ in } L^2(H^{-1}(I)). \end{aligned}$$

Additionally, u^ϵ_x generates gradient Young measure ν.

Some Details of the Proof II

$$u^\epsilon \rightharpoonup u \text{ in } L^\infty(W^{1,6}(I)) \cap W^{1,\infty}(L^2(I)),$$
$$u^\epsilon_x \text{ generates a gradient Young measure } \nu,$$
$$\theta^\epsilon \rightharpoonup \theta \text{ in } L^2(H^1(I)) \cap L^\infty(L^1(I)),$$
$$\theta^\epsilon_{xx} \rightharpoonup \theta_{xx} \text{ in } L^2(H^{-1}(I)).$$

2. Passage to the limit:
   - Convergence of u^ε: sufficient regularity
   - Convergence of θ^ε u^ε_x: Div-Curl-Lemma for $(\theta^\varepsilon,0)$ and $(u^\varepsilon_x, u^\varepsilon_t)$.
   - Convergence of $\ln(\theta^\varepsilon)_t$ and $\theta^\varepsilon_x/\theta^\varepsilon$ needs additional assumption (Possible improvement: parabolic regularity)

$$\int_0^T \int_I (u_t \xi_t - u_x \theta \xi_x - \langle \phi, \nu \rangle \xi_x) \, \mathrm{d}x \, \mathrm{d}t = -\int_0^T \int_I f \xi \, \mathrm{d}x \, \mathrm{d}t,$$
$$\int_0^T \int_I \left(\ln \theta \, \zeta_t + \theta_{xx} \frac{\zeta}{\theta} - \frac{1}{2} \langle |\cdot|^2, \nu \rangle \zeta_t \right) \mathrm{d}x \, \mathrm{d}t = -\int_0^T \int_I \frac{g}{\theta} \zeta \, \mathrm{d}x \, \mathrm{d}t.$$

Conclusions

1. Crystalline Level: Symmetry dictates energy landscape.
   Difference between weak and non-weak transformations:
   Phase transitions with two maximal subgroups of GL(3,$\mathbb{Z}$)
   - cannot be described locally by Ericksen-Pitteri neighborhood,
   - cannot resist shear (fluid-like).

2. Continuum level:
   - Analysis and numerical simulation for regularized system with capillarity gives some insight, but regularization heavily influences behavior.
   - Alternative approach: compute effective energy (=quasiconvex envelope). Approximate by rank-1 convexity and separate convexity.
   - Evolution of microstructures: Analysis in terms of Young measures.

3. Future work: Passage from crystalline to continuum level:
   - Traveling wave solution in bistable systems.