Three-Dimensional Elastic Compatibility: Twinning in Martensites

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We show how the St. Venant compatibility relations for strain in three dimensions lead to twinning for the cubic to tetragonal transition in martensitic materials within a Ginzburg-Landau model in terms of the six components of the symmetric strain tensor. The compatibility constraints generate an anisotropic long-range interaction in the order parameter (deviatoric strain) components. In contrast to two dimensions, the free energy is characterized by a “landscape” of competing metastable states. We find a variety of textures, which result from the elastic frustration due to the effects of compatibility. Our results are also applicable to structural phase transitions in improper ferroelastics such as ferroelectrics and magnetoelectrics, where strain acts as a secondary order parameter.

81.30.Kf, 64.70.Kb, 61.72.Dc, 82.65.Dp

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The harmonic elastic energy contribution due to non-OP compression-shear (CS) strain components is
\[ F_{cs} = A_e e_2^2 + A_o (e_2^4 + e_3^2 + e_4^2). \]

It is also possible to add a coupling, \( F_{\text{compos}}(\eta, \nabla \eta, e_2, e_3) \), of the OP strain to compositional fluctuations \( \eta \), which are responsible for tweed, but we do not consider such effects in the present paper.

\[
\frac{-2}{\sqrt{3}} (e_{2,x} e_{3,x} - e_{2,y} e_{3,y}) + \frac{4}{3} e_{2,z}^2 \]

**Fig. 1.** Countour plot of the Landau free energy in the martensite phase depicting the three degenerate energy minima corresponding to the three tetragonal variants with tetragonal axis along the \( x, y \) and \( z \) axis, respectively. Parameters are \( A = -1.0, B = 0.83 \) and \( C = 0.04 \).

The coefficients \( A = A_o (T - T_o) \), \( B \) and \( C \) are related to the second, third and fourth order elastic constants, respectively, and can be obtained from experimental structural data. The martensitic transition temperature is denoted \( T_o \) and the gradient coefficients \( g \) and \( h \) are determined from the phonon dispersion at the appropriate high-symmetry point in the Brillouin zone of the material. \( A_e \) and \( A_o \) denote the bulk compression and shear moduli, respectively. Here we do not consider the gradient energy contribution from the non-OP components \( e_1, e_4, e_5, \) and \( e_6 \), since their contribution is of secondary nature compared to the terms reported above.

**3D compatibility and analysis.** We express the non-OP strains in terms of the OP strains \( e_2, e_3 \) using the three compatibility constraints \( e_{12,12} = e_{31,31} + e_{22,11}, e_{23,23} = e_{22,33} + e_{33,22}, e_{31,31} = e_{33,11} + e_{11,33} \). Analagous to the 2D case the constrained minimization of the compression-shear energy leads to an anisotropic long-range interaction between the OP strains in the bulk:
\[
F_{cs}^{\text{bulk}} = \int d^3r e_\alpha (\alpha) U_{\alpha \alpha'}(|r - r'|) e_{\alpha'}(r') \quad (\alpha, \alpha' = 2, 3),
\]

where the kernels \( U_{22}, U_{21}, \) and \( U_{33} \) can be expressed explicitly in Fourier space with each of the three kernels becomes a ratio of algebraic combinations of \( k_x, k_y \) and \( k_z \). All these combinations have the same order in the length, \( k \), of the wavevector \( \vec{k} \). Hence, the kernels do not have any scale dependence. That is, the kernels only depend on the angles \( \theta \) and \( \phi \), defined in terms of spherical coordinates. The minima of each kernel occur at certain \( \theta \) and \( \phi \) values. These angles vary from kernel to kernel and the system, in general, experiences frustration. Figure 2 shows contours of the kernel \( U_{22} \) and \( U_{33} \) as functions of the two angles \( \theta \) and \( \phi \). The minima define a direction in 3D in which the compression-shear energy arising from the particular kernel is minimal.

In contrast to 2D, \( F_{cs} \) can be viewed as a weighted sum of the three kernels, where the fields \( e_2 \) and \( e_3 \) act as the weights. There is thus an interplay between \( F_2 \) and the directional dependence of \( F_{cs} \) to accommodate the frustration and choose minima, which in general do not have zero energy. Thus, there exist metastable minima corresponding to different microstructures. In 2D there is no such interplay, as the directional minimum is independent of the amplitude of the field: There is only one minimum corresponding to zero energy and hence there is no frustration. Moreover, these long-range kernels in 3D fulfill the full cubic symmetry properties associated with the transformation. In particular, \( F_{cs} \) is invariant with respect to two-fold, three-fold, and four-fold rotation symmetries of the cube. We do not present here the mathematical structure underpinning this, but it is reflected in the frustration seen in 3D that is lacking in 2D.

The bulk contribution comes from a solid with periodic boundaries while the the effects of interfaces/surfaces such as habit planes (i.e., austenite-martensite or parent-product boundaries) may be included through a free energy contribution, \( F_{\text{surf}} \), where \( F_{cs} = F_{cs}^{\text{bulk}} + F_{cs}^{\text{surf}} \) and
\[
F_{cs}^{\text{surf}} = \int d^3k \Gamma(k_x, k_y, k_z) [e_2(k)^2 + e_3(k)^2].
\]

In analogy with the 2D case the function \( \Gamma(\vec{k}) \) has the form
\[
\Gamma(\vec{k}) = \nu \left[ \frac{1}{|k_x + k_y|} + \frac{1}{|k_y + k_z|} + \frac{1}{|k_z + k_x|} \right]
\]
for the \([110] \) family of habit planes.

We remark that the physics of twinning is quite different in 2D and 3D. In 2D both the gradient and the bulk compatibility terms vary as \( \sim 1/r^2 \) and thus do not lead to a length scale. However, the surface compatibility term in 2D is essential for introducing a length scale. It varies as \( \sim 1/r \) which then competes with the combined bulk compatibility and gradient terms to give rise
to a twinning width. In 3D, the gradient terms behave as $\sim 1/r^2$ and $F_{cs}^{bulk} \sim 1/r^3$. Thus, there is already a length scale present and $F_{cs}^{surf}$ merely renormalizes the length scale arising from the bulk compatibility and gradient terms. We have numerically verified the above scaling relations. On the basics of the above arguments we expect the twin width to scale as the square-root of the length in 3D to hold on the basis of above arguments.

Texture simulations. The various martensitic textures that realize minima of the energy are found from random initial conditions and relaxational dynamics for the OP strains. That is, $\dot{e}_\alpha(r) = -\nabla F(e_{\alpha}(r), e_1(r), e_2(r), e_3(r), e_4(r), e_5(r), e_6(r))$, where the time $t$ is scaled with a characteristic relaxation rate and $\alpha = 2, 3$. Depending on the details of the initial configurations, various twinning textures reflecting the metastability emerge as $\dot{e}_\alpha$ vanishes. The simulations are performed for a regular cube, and periodic boundary conditions are applied in all six directions to obtain fully relaxed textures in $e_2(r)$ and $e_3(r)$.

![FIG. 2. Three dimensional compatibility kernels $U_{22}$ (upper) and $U_{33}$ (lower) as a function of the colatitude $\theta$ and the azimuth $\phi$ angles. Dashed contours surround minima, and dashed dotted surround maxima, while solid contours show intermediate levels. The parameters are $A_s = 1.2$ and $A_c = 2.4$.](image)

![FIG. 3. (Color) Three dimensional twins in the (110) plane obtained from the time-dependent Ginzburg-Landau simulations, with representative parameters as given in Figs. 1 and 2 and, $h = 1$ and $g = 1$. The scale of the twins is determined by the competition between the interfacial energy and the bulk compression-shear energy, whereas the sharpness of the domain walls is determined by the Landau energy.](image)
layered high-temperature superconductors.

FIG. 4. (Color) Three dimensional twins with two different orientations, (110) and (1T0), in adjacent planes. Both OP fields are shown, $e_3$ in the upper panel and $e_2$ in the lower.

To illustrate the frustration that is inherent in the compatibility relations, we explain the orientation of the twins in Fig. 4. We construct an effective kernel for the red and blue twins, respectively, using the appropriate values of the strains $e_2$ and $e_3$ in the twins. Figure 5 depicts the minima of these effective kernels in the $\phi$-$\theta$ plane. The dashed line shows the minimum corresponding to the blue twins and the solid line the minimum for the red twins. We note that the minima correspond to different $\phi$ angles ($45+\delta$ and $45-\delta$). The shift $\delta$ depends on the parameters of the Landau free energy. Ideally, the red and the blue twins would not run parallel; however, the system accommodates these two conflicting directions by choosing the average angle of 45 degrees. This aspect of competing metastable states is a novel feature associated with the cubic symmetry kernels that leads to a rich landscape of microstructures.

FIG. 5. Minima of effective kernels illustrating inherent strain (orientational) frustration in 3D.

**Conclusion.** We have derived a complete symmetry based, fully 3D model that describes the cubic to tetragonal structural transitions observed in many martensitic and shape-memory alloys. We obtained analytically the compatibility-induced anisotropic long-range potential in the OP (deviatoric strain tensor components) in the bulk and at interfaces. Unlike the 2D case, there is a length scale in the system from bulk compatibility alone; the surface compatibility potential also introduces a length scale akin to 2D. Many twin orientations are possible in 3D as a result of elastic frustration and the “landscape” (probably “rugged”) of metastable energy states. Our model can be generalized straightforwardly to improper ferroelastics transitions by including symmetry allowed polarization (magnetization, etc.) nonlinear terms, and couplings to strain. Other symmetries can be handled within our approach. For example, we can study the cubic to trigonal (rhombohedral) transition in lead orthovanade, and NiTi- and AuCd-based shape-memory alloys using the three shear strains $e_4$, $e_5$, and $e_6$ as OP and with $e_1$, $e_2$, and $e_3$ expressed in terms of the shear strains via compatibility. Image reconstruction methods of systematic experimental data from HREM and neutron scattering will allow a more quantitative comparison of strain patterns with those obtained from our model.

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