Twin Boundaries in Ferroelastic Media without Interface Dislocations

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An elastic Ginzburg-Landau theory which includes strain and strain-gradient contributions is formulated for inhomogeneous strain fields associated with interface boundaries, heterophase inclusions, and transformation precursors. For proper purely ferroelastic materials of D_{4h} symmetry, an explicit kink-type solitary-wave solution describing a moving coherent $(1\bar{1}0)$ twin boundary is obtained.

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Among structural, displacive phase transitions there are two "distinct" classes[1]: (1) At the unit-cell level there is an intracellular ("shuffle") displacement of atoms, with little homogeneous lattice strain. (2) There is a significant strain of the unit cell (i.e., "lattice distortive"); the accompanying intracell strains are secondary. This difference plays a fundamental role in the physics and morphology of the two classes.

In class (1) long-range elastic stresses are less important; the intracell displacive transition can easily spread with the speed of propagation of the associated soft phonon [e.g., the purely ferroelectric D_{4h}-C_{4h} transition of $Ba_2NaNb_5O_{15}$ at 560 °C (Ref. 2)]. Other aspects are that it is a second-order transition with well-defined critical properties, and an appropriate soft-mode polarization vector is observed.

In class (2) the unit-cell distortion which accompanies the product phase formation necessarily implies long-range *dimensional* changes; to form and grow the new product phase in the parent phase induces large elastic strains. As a result several new considerations come into play: (1) The total free energy of the system becomes a function of not only the "intrinsic" free energies of homogeneous parent and product, but also of the macroscopic heterostructure of the system. (2) In fact, in martensites[1,3] and ferroelastic (FE) materials[4,5] the elastic component of the total free energy is consequently lowered either by plastic flow (irreversibly) or by formation of a heterogeneous array of different orientations of the product phases which usually occur in parallel twin bands. (3) The transition is first order[4,6] and mode softening never takes place completely, although macroscopically soft elastic behavior frequently accompanies the transition. (4) Precursor structures[7] ("microdomains") frequently precede the transition.

This class encompasses *martensitic* transformations which are characterized by a dominating "deviatoric" (i.e., shear) component of the transformation strain.[1] *Ferroelasticity* is the analog of ferromagnetism, where domains have differently oriented magnetization. It is defined by the existence of two or more structurally equivalent stable orientation states, which can be interconverted by mechanical stress.[5] If the spontaneous strain is the primary (secondary) order parameter in the Landau theory of phase transitions,[8] the FE transition is called "proper" ("improper") and falls into the second (first) of the above classes.[9] Many martensitic transitions are also proper FE transitions.

These lattice distortive transitions are essentially reversible, diffusionless, characterized by precise parent-product habit plane and relative unit-cell orientations, and often shape-reversible changes; though microscopically heterogeneous there are well-defined macrostates associated with this class. This situation is analogous to commensurate-incommensurate systems or epitaxy,[10] which have been modeled by nonlinear displacement fields, leading to kinks, soliton arrays,[11] and other heterostructures.

In this Letter we will address a particular aspect of heterogeneous structures in martensitic transitions: twinning in a cubic-tetragonal (O_h-D_{4h}) transformed proper purely FE material.[9] Examples are $In_{1-x}Tl_x$ alloys and the $A15$ compounds V_3Si and Nb_3Sn.[4] We provide a soliton model of twinning,[12] which constitutes an alternative to the "interface dislocation" models traditional in metallurgy.[3] The parameters of the model can be related to experimental elastic-constant data and phonon dispersion curves.

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We consider an elastic continuum, representing a cubic (O_h) prototype phase which may deform ferroelastically into any of three tetragonal (D_{4h}) variants with (nearly) orthogonal c axes. We express (a) the elastic energy functional (including terms nonlinear in *strain* up to fourth order, and *strain gradient* terms[13-15] up to second order) in terms of (b) symmetry strains appropriate for cubic symmetry[16]:

$$e_1=(\eta_{11}+\eta_{22}+\eta_{33})/\sqrt{3};\quad e_2=(\eta_{11}-\eta_{22})/\sqrt{2};$$
$$e_3=(\eta_{11}+\eta_{22}-2\eta_{33})/\sqrt{6};\quad e_4=\eta_{23}=\eta_{32};$$

e_5,e_6 by permutation. Here $\eta_{ij}=\frac{1}{2}(u_{i,j}+u_{j,i}+u_{k,i}u_{k,j})$ are the components of the Lagrangian strain tensor, and $u_{i,j}=\partial u_i/\partial x_j$ the derivatives of the displacement vector $\vec{u}(\vec{x},t)$ with respect to the material coordinates x_i in a common stationary Cartesian frame coinciding with the cubic axes of the prototype phase. We carried out a general expansion of the elastic energy with respect to the symmetry strains and their first gradients,[17] but report here only those terms that enter the particular solution for a moving $(1\bar{1}0)$ twin boundary with only (deviatoric!) (e_2,e_3) strains present[16]:

$$U(e_\alpha;e_{\alpha,i})=\Phi(e_\alpha)+\Psi(e_{\alpha,i})\quad(\alpha=2,3;\ i=1,2), \tag{1a}$$

$$\Phi=A(e_2^2+e_3^2)+Be_3(e_3^2-3e_2^2)+C(e_2^2+e_3^2)^2, \tag{1b}$$

$$\Psi=g[e_{2,1}^2+e_{2,2}^2+(e_{3,1}^2+e_{3,2}^2)/3+2(e_{2,1}e_{3,1}-e_{2,2}e_{3,2})/\sqrt{3}]$$
$$+h[e_{3,1}^2+e_{3,2}^2-\sqrt{3}(e_{2,1}e_{3,1}-e_{2,2}e_{3,2})]. \tag{1c}$$

The coefficients in (1b) are linear combinations of second- through fourth-order elastic constants.[18] The strain gradient coefficients in (1c) describe nonlocal elastic behavior.[13-15] For $C>0$ Eqs. (1a)–(1c) define the Ginzburg-Landau energy functional for an O_h-D_{4h} FE transition, provided that the only strains present are given by the two-component order parameter (e_2,e_3) and are independent of x_3. The presence of the third-order term implies a first-order transition.[6] Assuming the usual linear temperature variation of the soft shear modulus A as for a proper FE material, and constancy for the remaining coefficients, we introduce the dimensionless temperature $\tau=4AC/B^2=(T-T_c)/(T_0-T_c)$, where T_0 is the transition temperature and T_c the stability limit of the cubic phase $(A=0)$.

The Lagrange density $L=\rho_0\dot{u}_i\dot{u}_i/2-U(e_\alpha,e_{\alpha,i})$ leads to the equation of motion[13]

$$\rho_0\ddot{u}_i=\left(\frac{\partial U}{\partial u_{i,j}}\right)_{,j}-\left(\frac{\partial U}{\partial u_{i,jk}}\right)_{,jk}. \tag{2}$$

In general, the solution of (2) is very cumbersome, but a simple particular solution may be found from the *Ansatz*

$$u_i=\alpha x_i+f(\hat{n}\cdot\vec{x}\pm vt)\quad(i=1,2);$$
$$u_3=-2\alpha x_3, \tag{3}$$

with $\alpha=$const, f an unknown function, and $\hat{n}$ denoting the normal of the $(1\bar{1}0)$ twin boundary. We obtained a first-order solution [valid for small transformation ("Bain") strain] by neglecting "geometric nonlinearity" (arising from the last term in η_{ij}), but retaining "physical nonlinearity" [as represented by the anharmonic strain energy (1b)], which leads to $e_2=f'$, $e_3=\sqrt{6}\alpha$; $e_1=e_4=e_5=e_6=0$, thus justifying restriction to the elastic energy [(1a)–(1c)]. For the particular value $\tau=-9$ $(A=-9B^2/4C<0)$ Eq. (2) reduces to the ordinary differential equation

$$(\rho_0v^2-a_2)f''-3a_4(f')^2f''+gf''''=0 \tag{4}$$

for the function f, with the coefficients given by $a_2=3A/2$, $a_4=2C$, and with $\alpha=(\frac{3}{2})^{1/2}B/4C$. For $g>0$, Eq. (4) has the exact solution

$$f(s)=f_0\pm(2g/a_4)^{1/2}\ln\{\cosh[(s-s_0)/\sqrt{2}]\}\quad\text{(displacement)}, \tag{5a}$$

$$e_2(s)=\pm[(\rho_0v^2-a_2)/a_4]^{1/2}\tanh[(s-s_0)/\sqrt{2}];\quad e_3=\sqrt{6}\alpha\quad\text{(strain)}, \tag{5b}$$

in the dimensionless variable $s=(\hat{n}\cdot\vec{x}\pm vt)/\xi$, where $\xi=[g/(\rho_0v^2-a_2)]^{1/2}$ is a coherence length which determines the width of the strain kink (5b). The corresponding Lagrangian strains are $\eta_{11}=\alpha+(e_2/\sqrt{2})$, $\eta_{22}=\alpha-(e_2/\sqrt{2})$, $\eta_{33}=-2\alpha$ and describe the moving boundary between two (velocity-dependent) orthorhombically deformed variants of the cubic parent phase. The strain kink moves with the speed $\mp v$ in the $[1\bar{1}0]$ direction, but the particles move in the [110] direction (parallel to the twin boundary) with the speed $\pm\sqrt{2}e_2v$,

thus for $s \to \pm\infty$ approaching the value $\pm 6\alpha[1-(\rho_0 v^2)/a_2]^{1/2}v$. This solution represents an elastic solitary wave and provides an alternative to the usual mechanism for twin boundary motion via twinning dislocations.[3] As in other applications the global stability of such solutions requires extensive analysis, which in general confirms their stability.[11]

In the static limit $v=0$ the deformation becomes, for $s \to +\infty$ $(-\infty)$, tetragonal, with the tetragonal axes in [100] ([010]). The static kink width is $2\xi_0 = 2[g/(-a_2)]^{1/2}$, and the interfacial energy per unit area $E_0 = (-2a_2)^{3/2}g^{1/2}/a_4$. These results are best interpreted by Figs. 1 and 2. Figure 1 shows the strain energy as a function of e_2, e_3. The three equienergetic minima represent the three tetragonal strain configurations with the tetragonal axes along the three cubic $\langle 100\rangle$ directions. In Fig. 1(a) (for $T < T_c$) the cubic phase ($e_2 = e_3 = 0$) is unstable. Figure 1(b) refers to the transition temperature ($T = T_0$), and a fourth equienergetic minimum is present and corresponds to the cubic phase. Here we are concerned simply with the twin configuration below T_c which describes going from one minimum (variant) to another; for $\tau = -9$ the strain field which accomplishes this is given by (5b) or (for $\tau \neq -9$) perturbation solutions[12] derived therefrom. The static trajectory (5b) in e_2, e_3 is for $\tau = -9$ indicated in Fig. 1(a) by the dashed line. In general e_3 is not constant for arbitrary τ, and the other symmetry strains neglected in (1b) and (1c) must be included. Figure 2 shows the resulting lattice deformation pattern.

For $\tau > 0$, Eq. (3) with $\alpha = 0$ and small f and f' is a small-amplitude sinosoidal wave solution of (2) which represents the soft shear mode propagating in $[1\bar{1}0]$ with polarization in [110] associated with the O_h-D_{4h} FE transition.[4,6] This solution is obtained from the linearized form of (4) with $a_2 = 2A$ and has the dispersion relation $\rho_0\omega^2 = a_2k^2 + gk^4$. We see that for small k, ω vs k is for $T > T_c$ linear *plus* upward (downward) curvature if g is positive (negative). Thus, for $T < T_c$ static kink solutions exist *if the phonon dispersion has upward curvature at small k.* Experimental neutron scattering data above T_0 for V_3Si,[19] Nb_3Sn,[20] and $In_{0.76}Tl_{0.24}$[21] show indeed upward curvature for the soft shear mode in $\langle 110\rangle$. In brief, *the present Ginzburg-Landau theory can model static domain walls if the phonon dispersion is upward.* What is interesting and physically important is the relation of the parameters a_2, a_4, and g to experiment: a_4 is related to "fourth-order" elasticity, while a_2 and g determine energies quadratic

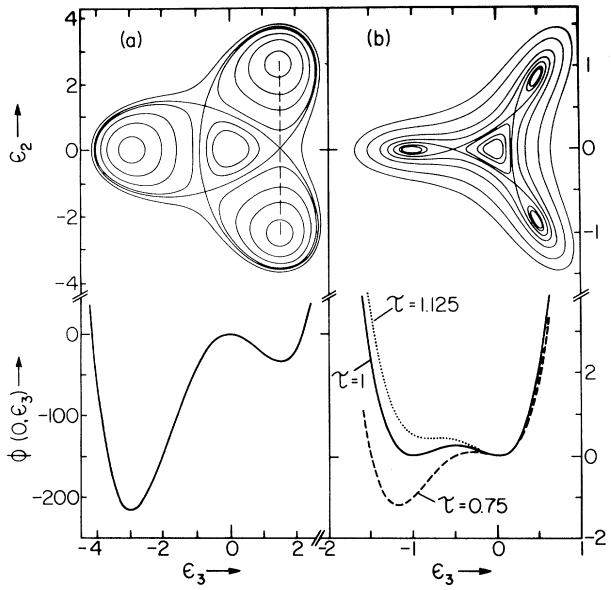

FIG. 1. Curves of constant dimensionless strain energy $\phi(\epsilon_2,\epsilon_3) = \Phi(e_2,e_3)/\Phi_0 = \phi_0$, and section $\phi(0,\epsilon_3)$ vs ϵ_3, with $\epsilon_2 = e_2/e_0$, $\epsilon_3 = e_3/e_0$, $e_0 = B/2C$, and $\Phi_0 = B^4/64C^3$. (a) For $\tau = -9$ ($\phi_0 = 0, -10, -20, -33.75, -50, -100, -150, -200$). (b) For $\tau = 1$ ($\phi_0 = 5, 2.5, 1, 0.5, 0.25, 0.1, 0.05$).

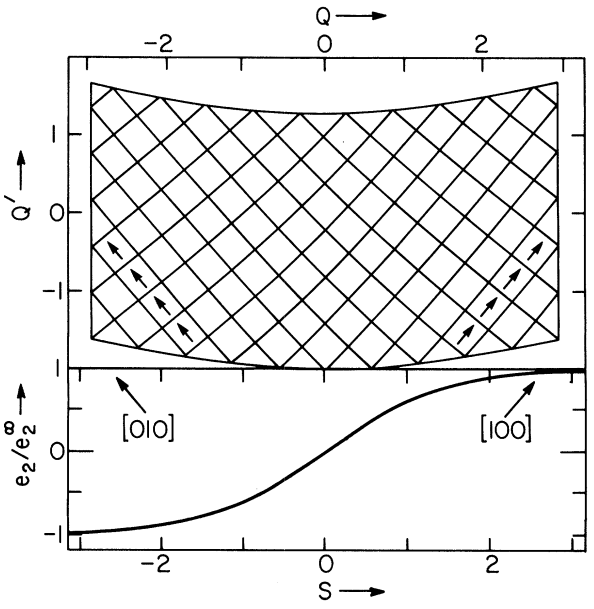

FIG. 2. Deformation pattern and strain profile of a $(1\bar{1}0)$ twin boundary in a tetragonal ferroelastic material for $\tau = -9$ and $v = 0$ with $\alpha = 0.033$. The grid $[x_1(Q,Q') = \text{const}, x_2(Q,Q') = \text{const}$, where $Q = \bar{X}\cdot\hat{n}/\xi$, $Q' = \bar{X}\cdot\hat{n}'/\xi$, $\bar{X} = \bar{x} + \bar{u}(\bar{x})$, $\hat{n}' = (1/\sqrt{2}, 1/\sqrt{2}, 0)]$ schematically represents the (001) projection of the twin-deformed cubic unit cells. The [001] axes in the two tetragonal variants are along the [010] and [100] directions of the cubic prototype phase.

in the first and second derivatives of the displacement.

The unique feature of this model is that a coherent correspondence is maintained across the twin boundary, no atoms are "lost," and artifacts such as "coherency dislocations" to form the boundary are unnecessary. However, only an extensive atomistic model could account for the Peierls barrier pertaining to twin boundary motion or the balance between core energy and elastic energy.

There is a long history of modeling of interphase interfaces, as, for example, the parent-product martensite interface discussed by Olson and Cohen[22]; they and others[15,23,24] have introduced strain gradient energies into simplified static one-dimensional models of the interface. Ours is a three-dimensional dynamic model with a two-component strain order parameter; it is particularly relevant when the strain field energy is an important part of the total boundary energy. The solutions reported here relate two tetragonal regions, but do not yet define the three-region alternating-twin to cubic interface which describes the martensite-parent habit interface. To obtain that solution it will be necessary to solve Eq. (2) for strain fields inhomogeneous in two directions.

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[1]M. Cohen, G. B. Olson, and P. C. Clapp, in *Proceedings of the International Conference on Martensitic Transformations, ICOMAT 1979* (Massachusetts Institute of Technology, Cambridge, Mass., 1979), p. 1.

[2]K. Aizu, J. Phys. Soc. Jpn. **41**, 880 (1976).

[3]J. W. Christian, *The Theory of Transformations in Metals and Alloys* (Pergamon, Oxford, 1975), 2nd ed.

[4]N. Nakanishi, A. Nagasawa, and Y. Murakami, J. Phys. (Paris) Colloq. **43**, C4-35 (1982).

[5]K. Aizu, J. Phys. Soc. Jpn. **27**, 387 (1969), and **28**, 706 (1970).

[6]R. A. Cowley, Phys. Rev. B **13**, 4877 (1976).

[7]L. E. Tanner, A. R. Pelton, and R. Gronsky, J. Phys. (Paris) Colloq. **43**, C4-169 (1982).

[8]L. D. Landau and D. M. Lifshitz, *Statistical Physics* (Pergamon, Oxford, 1968), 2nd ed.

[9]J. C. Toledano, J. Solid State Chem. **27**, 41 (1979).

[10]J. Villain, in *Ordering in Strongly Fluctuating Condensed Matter Systems,* edited by T. Riste (Plenum, New York, 1980), p. 221.

[11]A. R. Bishop, J. A. Krumhansl, and S. E. Trullinger, Physica (Utrecht) **1D**, 1 (1980).

[12]G. R. Barsch and J. A. Krumhansl, in *Proceedings of the International Conference on Solitons in Real Systems,* edited by J. C. Fernandez and G. Reinish (Centre Universitaire Mediterraneen, Nice, 1982), p. 90.

[13]R. A. Toupin, Arch. Ration. Mech. Anal. **11**, 385 (1962), and **17**, 85 (1964).

[14]J. A. Krumhansl, in *Mechanics of Generalized Continua,* edited by E. Kröner (Springer, Berlin, 1968), p. 298.

[15]A. L. Roitburd, in *Solid State Physics,* edited by H. Ehrenreich, F. Seitz, and D. Turnbull (Academic, New York, 1978), p. 317.

[16]M. W. Finnis and V. Heine, J. Phys. F **4**, 960 (1974).

[17]Reported in Ref. 12 only in brief; the full elastic energy will be given elsewhere.

[18]B. N. N. Achar and G. R. Barsch, Phys. Rev. B **19**, 3761 (1979), and **23**, 5839 (1981).

[19]G. Shirane, J. D. Axe, and R. J. Birgeneau, Solid State Commun. **9**, 397 (1971).

[20]J. D. Axe and G. Shirane, Phys. Rev. B **8**, 1965 (1973).

[21]T. R. Finlayson, M. Mostoller, W. Reichardt, and H. G. Smith, to be published.

[22]G. B. Olson and M. Cohen, J. Phys. (Paris) Colloq. **43**, C4-75 (1982).

[23]M. L Green, Ph.D. thesis, Massachusetts Institute of Technology, 1975 (unpublished); J. W. Cahn and M. L. Green, unpublished.

[24]T. Suzuki, Metall. Trans. **12A**, 709 (1981).