

CONSTITUTIVE THEORY FOR SOME CONSTRAINED ELASTIC CRYSTALS

BY

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CONSTITUTIVE THEORY FOR SOME CONSTRAINED ELASTIC CRYSTALS

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Abstract — Some of the observations of A-15 superconductors near cubic-tetragonal phase transformations suggest treating them as thermoelastic bodies subject to certain material constraints. Here, we begin to develop a theory of this kind.

1. INTRODUCTION

In crystals, it is not unusual to encounter phase transformations involving some change in crystal symmetry. In some of these, observations indicate that some elastic modulus becomes quite small compared to the others, near the transformation. For example, Nakanishi [1] remarks that this seems to be a common feature of alloys exhibiting the shape memory effect. A similar thing also occurs in the so-called high-temperature or A-15 superconductors, near a transformation of the cubic-tetragonal type, as is indicated by data presented by Keller and Hanak [2], for example. Indeed, a shear modulus seems to extrapolate to zero at the transformation temperature, one of several indications that these transformations might be of second-order. Landau theory indicates that this is highly improbable, that the transformation should be of first-order, with this modulus remaining positive. As is discussed in some detail by Ericksen [3], there is room to quibble about this theory and its predictions, but it would do no harm to better understand the theories of both possibilities. Move slightly away from transformation and they share the feature that the modulus is positive, but relatively small.

Such situations are not unlike the situation encountered in, say, elastomers, for which the shear modulus is small compared to the bulk modulus. There, we employ an idealization, regarding the bulk modulus as becoming infinite or, more properly, treating the materials as constrained, in this case incompressible. Here,

we propose to play a similar game with some crystals. The decision as to what constraints are appropriate depends on which ratios of moduli are small and, for crystals, numerous possibilities exist. To be definite, we will try to model the situation occurring in the A-15 superconductors. We bias the discussion a bit, in favor of transformations which are of first-order, although much of the analysis can also be applied to those of second-order.

With situations of this general kind, we have a pretty good analog or generalization of the "order parameters" which Landau [4] introduced, in his considerations of second-order transformations. From our view, they become the deformations possible in the corresponding constrained materials. Finding the general idea useful, physicists have stretched it to cover various things encountered in first-order transformations which are in some sense weak. In this respect, we are giving one interpretation of what it means to be weak.

2. CONSTRAINTS

For the crystals of the A-15 type, unloaded crystals transform from a configuration of cubic symmetry to one of tetragonal form as the temperature is lowered through a critical value T_c . With a common labelling of elastic moduli, used by Love [5, Ch. VI], for example, the linear elastic strain energy of the cubic phase can be put in the form

$$2W = (\hat{C}_{11} - \hat{C}_{12})(\eta_{11}^2 + \eta_{22}^2 + \eta_{33}^2) + [(\hat{C}_{11} + 2\hat{C}_{12})/3](\epsilon_{11} + \epsilon_{22} + \epsilon_{33})^2 + 4\hat{C}_{44}(\epsilon_{12}^2 + \epsilon_{23}^2 + \epsilon_{31}^2), \quad (1)$$

where

$$\eta_{ij} = \epsilon_{ij} - \frac{1}{3}(\epsilon_{11} + \epsilon_{22} + \epsilon_{33})\delta_{ij}, \quad (2)$$

ϵ is the infinitesimal strain tensor and \hat{C}_{ij} denotes elastic moduli. I have added carets to distinguish these from components of a tensor C , to be introduced later. This describes the energy relative to an orthonormal basis, base vectors parallel

to the orthogonal lattice vectors associated with the cubic phase. From this form, the usual conditions on moduli, conditions that $W \geq 0$, are easily read off. Observations indicate that, as the temperature is lowered to become near T_c , the crystal softens in the manner indicated by

$$\begin{aligned} (\hat{C}_{11} - \hat{C}_{12}) / (\hat{C}_{11} + 2\hat{C}_{12}) &\ll 1, \\ (\hat{C}_{11} - \hat{C}_{12}) / \hat{C}_{44} &\ll 1. \end{aligned} \tag{3}$$

The notion that these denominators are effectively infinite then gives us an estimate of likely constraints as

$$\epsilon_{11} + \epsilon_{22} + \epsilon_{33} = 0, \tag{4}$$

and

$$\epsilon_{12} = \epsilon_{23} = \epsilon_{31} = 0. \tag{5}$$

Given the continuous or nearly continuous nature of the transformation, it seems to me reasonable to consider the constraints as also applying to the tetragonal phases. When the transformation to tetragonal form takes place, the tetragonal phase tends to be twinned. Naturally, it can be tricky to interpret measurements made on twinned crystals. To cover the two phases and the twinning, we need nonlinear theory, with deformations which must be considered as finite. Those associated with the transformation and twinning are in fact quite small, so we will aim at theory appropriate for relatively small deformations. Even then, we need to extrapolate (4) and (5) to apply to finite deformations, a somewhat ambiguous matter. Various extrapolations would be reasonably consistent with observations of the deformations associated with the transformation.

From the viewpoint of nonlinear thermoelasticity theory, it is convenient to think of selecting as a reference configuration the unloaded cubic configuration, at the transition temperature T_c . Refer this to rectangular Cartesian coordinates $\mathbf{x} = (x_1, x_2, x_3)$. A deformation maps \mathbf{x} to

$$\mathbf{y} = \mathbf{y}(\mathbf{x}), \tag{6}$$

with

$$F = \nabla y , \quad \det F > 0 , \quad (7)$$

the usual deformation gradient. Then

$$C = F^T F = C^T > 0 \quad (8)$$

is one of the commonly used measures of finite deformation. My first inclination was to interpret (4) as the condition of incompressibility, implying that $\det C = 1$. After some exploration, I decided that a different extrapolation is more promising, viz.

$$C_{11} + C_{22} + C_{33} = 3 . \quad (9)$$

Infinitely many other possibilities exist, so I will try to make clear in what sense this is unique. With (5), the only reasonable extrapolation seems to be the evident choice

$$C_{12} = C_{13} = C_{23} = 0 . \quad (10)$$

Thus, we assume that deformations possible in our constrained materials are described by

$$C = \begin{pmatrix} f & 0 & 0 \\ 0 & g & 0 \\ 0 & 0 & h \end{pmatrix} , \quad (11)$$

with f , g and h positive functions, satisfying

$$f + g + h = 3 , \quad (12)$$

relative to the preferred rectangular Cartesian coordinate system described above. For C near the identity, (12) does, of course, approximate the incompressibility condition, our constrained materials then becoming nearly incompressible.

3. KINEMATICAL CONSIDERATIONS

Formally, one can interpret (6) as a change of coordinates taking Cartesian coordinates y_i to curvilinear coordinates x_j , C being then viewed as the metric tensor in the latter coordinate system. From elementary tensor analysis, we then know that

$$y^i_{,jk} = \Gamma^{\ell}_{jk} y^i_{,\ell} \quad (13)$$

where the Γ 's are Christoffel symbols based on C , considered as a metric tensor. Here and elsewhere, commas denote partial derivatives. Further, integrability conditions for this system are summarized by the condition that the Riemann tensor based on C must vanish, a set of differential equations which f , g and h must satisfy. Without using (12), this gives the following six equations:

$$\left. \begin{aligned} 2(f_{,22} + g_{,11}) - [(f_{,2})^2 + f_{,1}g_{,1}] / f - [(g_{,1})^2 + f_{,2}g_{,2}] / g + f_{,3}g_{,3} / h &= 0 , \\ 2(g_{,33} + h_{,22}) - [(g_{,3})^2 + g_{,2}h_{,2}] / g - [(h_{,2})^2 + g_{,3}h_{,3}] / h + g_{,1}h_{,1} / f &= 0 , \\ 2(h_{,11} + f_{,33}) - [(h_{,1})^2 + h_{,3}f_{,3}] / h - [(f_{,3})^2 + h_{,1}h_{,1}] / f + h_{,2}f_{,2} / g &= 0 , \end{aligned} \right\} (14)$$

and

$$\left. \begin{aligned} 2f_{,23} - f_{,2}f_{,3} / f - f_{,2}g_{,3} / g - f_{,3}h_{,2} / h &= 0 , \\ 2g_{,31} - g_{,3}g_{,1} / g - g_{,3}h_{,1} / h - g_{,1}f_{,3} / f &= 0 , \\ 2h_{,12} - h_{,1}h_{,2} / h - h_{,1}f_{,2} / f - h_{,2}g_{,1} / g &= 0 , \end{aligned} \right\} (15)$$

Thus, our three unknowns must satisfy seven equations. At first glance, it might seem unlikely that there are solutions other than the obvious homogeneous deformations, with f , g and h constant. That the impression is misleading can be seen by considering cases where f and g are independent of x_3 , with h constant, the generalized plane deformations. With the constraint (12) applying, one can represent the possibilities in the form

$$\left. \begin{aligned} f &= a(1 + \cos \alpha) , \\ g &= a(1 - \cos \alpha) , \\ h &= 3 - 2a , \quad 0 < a < 3/2 , \end{aligned} \right\} \quad (16)$$

with a constant, α a function of x_1 and x_2 satisfying

$$\cos^2 \alpha < 1 . \quad (17)$$

One then finds that the system of equations collapses to a single equation, viz.

$$\alpha_{,11} - \alpha_{,22} = 0 . \quad (18)$$

As we all know, a general solution is

$$\alpha = \beta(z_1) + \gamma(z_2) , \quad (19)$$

where

$$\sqrt{2} z_1 = x_1 + x_2 , \quad \sqrt{2} z_2 = x_1 - x_2 , \quad (20)$$

α and β being arbitrary functions, smooth enough to satisfy (18), at least in a weak sense. Of course, (17) imposes a restriction but, locally, we have a description of infinitely many possible deformations. Using (13) or the equivalent, one can get the corresponding deformations. To within a constant rotation and translation, one finds that

$$\left. \begin{aligned} y_1 &= c \int (-\sin \beta dz_1 - \sin \gamma dz_2) , \\ y_2 &= c \int (\cos \beta dz_1 - \cos \gamma dz_2) , \\ y_3 &= dx_3 , \end{aligned} \right\} \quad (21)$$

where

$$c = \pm \sqrt{a} , \quad d = \sqrt{3 - 2a} , \quad (22)$$

the sign being chosen so that

$$c \sin(\beta + \gamma) > 0 . \quad (23)$$

Obviously, the lines or, more properly, the planes $z_1 = \text{const.}$ and $z_2 = \text{const.}$ are characteristics of the hyperbolic equation (18). Physically, these planes do have a particular significance. As the cubic phase transforms to the tetragonal phase, the material planes

$$x_1 \pm x_2 = \text{const.} , \quad x_2 \pm x_3 = \text{const.} , \quad x_3 \pm x_1 = \text{const.} \quad (24)$$

become the so-called twin planes, surfaces of discontinuity which are commonly observed. Later, we will say more about twinning. As will become clear, these planes are, in different ways, related to other possible discontinuities.

In a more general way, one can explore what happens if one adopts a different extrapolation of (4), for example, $\det C = 1$. With this choice, one gets an equation somewhat like (18), a nonlinear hyperbolic equation, with different characteristics, which seem not to admit any easy physical interpretation. For the general system, one can write conditions restricting jumps in, say, second derivatives, when the functions and their first derivatives are continuous, using the kinematical conditions of compatibility

$$[f, ij] = r \nu_i \nu_j , \quad [g, ij] = s \nu_i \nu_j , \quad [h, ij] = t \nu_i \nu_j . \quad (25)$$

Here, the square bracket denotes the jumps, and ν is the unit normal to the discontinuity surface. Equations (14) and (15) give restrictions of the form

$$r \nu_2^2 + s \nu_1^2 = 0 , \quad r \nu_2 \nu_3 = 0 , \quad \text{etc.} \quad (26)$$

For a constraint of the form $\sigma(f, g, h) = 0$, the additional restrictions take the form

$$\frac{\partial \sigma}{\partial f} r \nu_1^2 + \frac{\partial \sigma}{\partial g} s \nu_2^2 + \frac{\partial \sigma}{\partial h} t \nu_3^2 = 0 . \quad (27)$$

Examination of the set indicates that, for these surfaces to be the planes given by (24), one needs

$$\frac{\partial \sigma}{\partial f} = \frac{\partial \sigma}{\partial g} = \frac{\partial \sigma}{\partial h} . \quad (28)$$

Solve these partial differential equations for σ , and you get a constraint equivalent to (12). So, it is in this sense that this extrapolation of (4) is unique. Observations seem to suggest no planes of discontinuity other than those given by (24).

It would be nice to have a good characterization of all possible deformations but, at least as yet, I have not found this. It is perhaps worth noting that (11) implies that the coordinate planes map to triply orthogonal families of surfaces. Possibly, some old theorem in differential geometry makes easy the characterization of the subset satisfying (12). If so, I haven't yet spotted it.

4. COHERENT CO-EXISTENCE

Here, we explore the possibility of having surfaces of discontinuity across which the deformation gradient $F = \nabla y$ suffers a finite discontinuity, with y remaining continuous, a kind of situation which is encountered in twinning, in particular. Various workers use the adjective "coherent" to describe discontinuities leaving the displacement continuous, to distinguish these from defects involving slip, cracking, etc. Let overbars denote quantities evaluated on one side of the surface, the same symbols without bars indicating the corresponding quantities on the other side. The usual kinematic conditions of compatibility then give

$$\bar{F} = F(1 + A \otimes N) , \quad (29)$$

where N is the unit normal to the surface of discontinuity, in the reference configuration, and A is the so-called amplitude vector. From this we get

$$\begin{aligned} \bar{C} &= \bar{F}^T \bar{F} = (1 + N \otimes A) C (1 + A \otimes N) \\ &= C + N \otimes CA + CA \otimes N + A \cdot CA N \otimes N . \end{aligned} \quad (30)$$

Since \bar{C} and C should both be compatible with (11) and (12), we must have

$$\text{tr}(\bar{C} - C) = 2N \cdot CA + A \cdot CA = 0 . \quad (31)$$

We set

$$CA = N \cdot C A N + \lambda M , \quad M \cdot M = 1 , \quad M \cdot N = 0 , \quad (32)$$

where λ is some scalar. With (31), (30) then reduces to

$$\begin{aligned} \bar{C} - C &= \lambda(N \otimes M + N \otimes M) \\ &= \lambda(E_1 \otimes E_1 - E_2 \otimes E_2) , \end{aligned} \quad (33)$$

where E_1 and E_2 are the orthogonal unit vectors given by

$$\left. \begin{aligned} \sqrt{2} E_1 &= N + M , \\ \sqrt{2} E_2 &= N - M . \end{aligned} \right\} \quad (34)$$

Clearly, this gives a spectral representation of $\bar{C} - C$. Since \bar{C} and C share the same eigenvectors, the orthonormal base vectors e_i ($i = 1, 2, 3$) in which (11) holds, E_1 and E_2 must be parallel to two of these. Analysis of any of these choices is much the same so, to be definite, we take

$$E_1 = e_1 , \quad E_2 = e_2 , \quad (35)$$

giving

$$\sqrt{2} N = e_1 + e_2 , \quad \sqrt{2} M = e_1 - e_2 . \quad (36)$$

Clearly, N is here normal to one of the planes given by (24), and we could arrange to get any other. With (36), it follows easily that

$$\left. \begin{aligned} \bar{f} - f &= \lambda , \\ \bar{g} - g &= -\lambda , \\ \bar{h} &= h . \end{aligned} \right\} \quad (37)$$

From this, we can read off one conclusion of interest. For a cubic configuration, $f = g = h \implies C = 1$. For one of tetragonal form, two eigenvalues of C should coincide, and be different from the third, with their sum being three. Try to fit

two such configurations to (37), and you conclude that cubic and tetragonal phases cannot co-exist coherently. This is consistent with observations of A-15 superconductors. It is one of the things which has supported the notion that such transformations might be of second-order. Clearly, the present theory also excludes co-existence, if the transformation is of first-order. In alloys exhibiting the shape memory effect, it is common for less and more symmetric phases to co-exist, coherently. Clearly, some modification of our assumptions must be made, to have any chance of applying to such crystals.

With the results at hand, it is a bit tedious, but not really difficult, to complete the analysis, so I will omit some of the details. To describe the results, we set

$$\begin{aligned}
 f &= k^2(1 + \cos 2\mu) , \\
 g &= k^2(1 - \cos 2\mu) , \\
 \bar{f} &= k^2(1 + \cos 2\bar{\mu}) , \\
 \bar{g} &= k^2(1 - \cos 2\bar{\mu}) , \\
 k &= \sqrt{(3-h)/2} ,
 \end{aligned} \tag{38}$$

with μ and $\bar{\mu}$ acute angles. Introduce the rotation matrix

$$\bar{\mathbf{R}} = \begin{vmatrix} \cos \theta & \sin \theta & 0 \\ -\sin \theta & \cos \theta & 0 \\ 0 & 0 & 1 \end{vmatrix} , \tag{39}$$

with

$$\theta = \mu - \bar{\mu} . \tag{40}$$

Set

$$\mathbf{F} = \mathbf{R} \begin{vmatrix} k \cos \mu & 0 & 0 \\ 0 & k \sin \mu & 0 \\ 0 & 0 & \sqrt{h} \end{vmatrix} , \tag{41}$$

\mathbf{R} being any rotation matrix, and set

$$\bar{\mathbf{F}} = \mathbf{R}\bar{\mathbf{R}} \begin{vmatrix} k \cos \bar{\mu} & 0 & 0 \\ 0 & k \sin \bar{\mu} & 0 \\ 0 & 0 & \sqrt{h} \end{vmatrix} . \quad (42)$$

With

$$\mathbf{A} = \sqrt{2} \sin \theta \left[\sin \bar{\mu} (\cos \mu)^{-1} \mathbf{e}_1 - \cos \bar{\mu} (\sin \mu)^{-1} \mathbf{e}_2 \right] , \quad (43)$$

one then has the solutions of (29) conforming to (36).

Special cases correspond to twinning. As this is commonly interpreted, the term refers to cases where

$$\bar{\mathbf{C}} = \mathbf{H}^T \mathbf{C} \mathbf{H} \neq \mathbf{C} , \quad (44)$$

with \mathbf{H} an element of the invariance group for relevant constitutive equations, such that

$$\mathbf{H}^2 = \mathbf{1} . \quad (45)$$

Note that this implies that $\det \bar{\mathbf{C}} = \det \mathbf{C}$, which would be quite compatible with the notion that the constraint of incompressibility applies, for example. We have not yet discussed such invariance, but will do so. For the moment, we note that, with \mathbf{N} given by (36), the matrix

$$\mathbf{H} = -\mathbf{1} + 2\mathbf{N} \otimes \mathbf{N} = \mathbf{H}^T \quad (46)$$

represents a 180° rotation, with \mathbf{N} as axis, so it satisfies (45). An elementary calculation then gives

$$\bar{\mathbf{C}} = \mathbf{H}^T \begin{vmatrix} f & 0 & 0 \\ 0 & g & 0 \\ 0 & 0 & h \end{vmatrix} \mathbf{H} = \begin{vmatrix} g & 0 & 0 \\ 0 & f & 0 \\ 0 & 0 & h \end{vmatrix} , \quad (47)$$

so (44) will hold, provided

$$f \neq g , \quad (48)$$

and (37)₃ holds. Comparing (38) and (47), we now have

$$\bar{\mu} = \pi/2 - \mu , \quad (49)$$

(40) then giving

$$\theta = 2\mu - \pi/2 . \quad (50)$$

It then follows from (43) that

$$A = -2M \cos 2\mu , \quad (51)$$

where M is given by (36), implying that

$$A \cdot N = 0 , \quad (52)$$

as would be expected by those familiar with twinning analyses. Similarly familiar is the fact that (44) implies the existence of a rotation matrix \hat{R} such that

$$\bar{F} = \hat{R} F H = F(1 + A \otimes N) , \quad (53)$$

and

$$\hat{R}^2 = 1 . \quad (54)$$

For the example, a calculation gives

$$\hat{R} = -1 + 2m \otimes m ,$$

where

$$m = R(\sin \mu e_1 + \cos \mu e_2) . \quad (55)$$

Clearly, (48) excludes twinning of our cubic phases, which seems to be in accord with observations of the A-15 superconductors. Twinning is observed in the tetragonal phases. Take $f = h$, and you get the analysis of such cases. Rather obviously, with slight changes in the analysis, one can take N normal to any of the planes listed in (24). If one replaces (12) by another likely extrapolation of (4), for example $fgh = 1$, one gets these same planes as twin planes, etc., so one does need to consider something other than twinning to decide between the possibilities.

Being purely kinematic, these analyses can be applied to crystals loaded in various ways, not necessarily statically. By the same token, the analyses are incomplete, involving no consideration of energies or forces.

One thing is worth noting. The constraints (10) tend to exclude deformations of the simple shearing type. However, as is clear from (52) and (53), the relative

deformation \overline{F}^{-1} is of the simple shearing type, this being characteristic of twinning.

Clearly, the discontinuities here considered are stronger than those considered in Section 2, so it is not so obvious, before doing the analysis, that they should occur on the planes described by (24).

5. ENERGETICS

We aim at developing theory good enough to cover both phases involved in the cubic-tetragonal transformations, including twinning of the latter, for cases where the deformations are not very large, with absolute temperatures T near the critical value T_c . At least for static problems, it seems reasonable to try to use nonlinear thermoelasticity theory, for which we need some constitutive equation for ϕ , the Helmholtz free energy per unit reference volume, something of the form

$$\phi = \phi(C, T) . \quad (56)$$

In principle, ϕ should be considered to be invariant under an infinite discrete group of the kind described by Ericksen [6]. However, as is discussed by Parry [7] and Pitteri [8], we need only require invariance under the point group corresponding to our cubic reference, if we are concerned with deformations meeting restrictions which they describe. It so happens that such restrictions are met by all deformations satisfying our constraints. Thus, as long as we accept the notion that the constraints apply, we need only require that

$$\phi(H^T C H, T) = \phi(C, T) = \hat{\phi}(f, g, h, T) , \quad (57)$$

with H belonging to the indicated point group. In particular, this includes orthogonal transformations interchanging pairs of our preferred base vectors, which means that $\hat{\phi}$ should be a symmetric function of f , g and h , this being enough to ensure invariance under the full group. Note that, with (47), $\hat{\phi}$ is then invariant under (46). In physical terms, it is pretty obvious that twin-related configurations should have the same energy. Physically, this supports the view that $\hat{\phi}$ should remain invariant

under the cubic group when the crystal has transformed to configurations of the tetragonal type.

Given this symmetry, we can reduce $\hat{\phi}$ to a function of elementary symmetric functions, a matter discussed carefully by Ball [9]. Bearing in mind the constraint (12), this means that ϕ is expressible in the form

$$\phi = \tilde{\phi}(J, K, T) , \quad (58)$$

where

$$6J = (f-1)^2 + (g-1)^2 + (h-1)^2 = \text{tr}(C-1)^2 , \quad (59)$$

$$2K = (f-1)(g-1)(h-1) = \det(C-1) . \quad (60)$$

Mathematically, potentials of essentially the same form and similar character arise in considerations of isotropic-nematic phase transformations in liquid crystals, so we will borrow some results occurring in Ericksen's [10] discussion of these. First, the inequality

$$K^2 \leq J^3 \quad (61)$$

always holds. When this reduces to equality, at least two of the quantities f , g and h must be equal. Refining this a bit, we have

$$f = g = h = 1 \iff J = K = 0 , \quad (62)$$

characterizing our cubic phases. Configurations of the tetragonal type are covered by

$$\left. \begin{array}{l} f = g \neq h \\ f \neq g = h \\ f = h \neq g \end{array} \right\} \iff K^2 = J^3 > 0 . \quad (63)$$

These correspond to the nematic phases in liquid crystals, the cubic phases being the analog of the isotropic phases in the latter. Also, the constraint (12), together with the condition that these functions be positive, provides another restriction. By an elementary calculation,

$$fgh = 2K - 3J + 1 > 0 , \quad (64)$$

one can show that (61) and (64), along with $J \geq 0$, cover the limitations on possible values of J and K . In general terms, we then want $\tilde{\phi}$ to have an absolute minimum of the kind indicated by (62) when $T > T_c$, switching of the kind indicated by (63) when $T < T_c$, its being possible that both retain some status near $T = T_c$, as at least relative minimizers. Expressing some of these ideas more formally, we at least want that

$$\tilde{\phi}(J, K, T) \geq \tilde{\phi}(0, 0, T) , \quad T > T_c \quad (65)$$

and, for some choice of the function $J = J_0(T)$, and for one of the two choices of algebraic signs,

$$\tilde{\phi}(J, K, T) \geq \tilde{\phi}(J_0, \pm J_0^{3/2}, T) = \psi(J_0, T) , \quad T < T_c . \quad (66)$$

When they first reported these transformations in an A-15 superconductor, Batterman and Barrett [11] opined that they might well be of second order, this being a reasonable opinion, I think. For this, it is important that $J_0 \rightarrow 0$ as $T \nearrow T_c$, and data such as are presented by Keller and Hanak [2] indicate that this might be true. To make a long story short, experimentation still seems to leave doubt as to whether such transformations are of second-order, or of first-order, with small discontinuities in J_0 , etc. masked by experimental errors.

For analyzing such small deformations, it seems natural to try to approximate ϕ by a polynomial of rather low degree in $C-1$, if you like by the first few terms in the Taylor expansion of a smooth function. One of the form

$$\tilde{\phi} = a(T) + b(T)J + c(T)K + d(T)J^2 \quad (67)$$

covers the possible quartics. There should be no danger of confusing the temperature-dependent coefficients with constants similarly labelled earlier. Assume that the temperature-dependent coefficients are smooth and similarly approximated near $T = T_c$, and you have what is sometimes called mean field theory. As is discussed by Wilson [12], for example, such assumptions go wrong in analyses of critical points in fluids, etc., situations bearing some similarity to the kinds of transformations

here considered. Still, it seems to me worthwhile to better understand what kinds of predictions are associated with such a guess, and my own understanding of this leaves much to be desired. Of course, one could try a compromise, using (67), but allowing the coefficients to have mild singularities at $T = T_c$.

With (61) and (67), we clearly have

$$\tilde{\phi} \geq \psi(J, T) = a + bJ - |c|J^{3/2} + dJ^2, \quad (68)$$

from which it is clear that if $\hat{\phi}$ has minimizers, they should be of cubic or tetragonal form, which is good, for our purposes.

Were (67) exact, we would need to have

$$d > 0, \quad (69)$$

to get the minimizers, so we'll try assuming this. Were $d < 0$ for this term in a Taylor expansion, one might still have the minima, but one would need to consider higher order terms in the expansion to sort this out, for a smooth potential. To have even a relative minimum of cubic form ($J = K = 0$), for $T > T_c$, we must have

$$b(T) > 0 \quad T > T_c. \quad (70)$$

Other extremals can be located, by setting the derivative of ψ equal to 0, giving

$$b - 3|c|J^{1/2}/2 + 2dJ = 0, \quad (71)$$

a quadratic in $J^{1/2}$. It will have real roots if

$$9c^2 \geq 32bd, \quad (72)$$

and we want this, at least for $T < T_c$. For whatever it is worth, the Landau-type argument that the transformation should not be of second-order is as follows. To have bifurcation occur at $T = T_c$, it is easy to see that one needs $b(T_c) = 0$, so $J = 0$ then satisfies (71). At T_c , ϕ should still be a minimum, for $J = K = 0$. An inspection of (67) or (71) makes clear that, for this, it is necessary that $c(T_c) = 0$. Grant that ϕ is thrice differentiable and, by essentially the same analysis, you come to this conclusion. As Landau [4] saw it, it is highly improbable that two

functions of one variable should vanish simultaneously. Nowadays, experts in bifurcation theory would, I think, agree that generically, such a transformation is not of second-order. If we argue generically, b should remain positive at and near $T = T_c$, so the cubic phase should retain some status, as a relative minimizer, for $T < T_c$. Then, as J increases, ψ must take on a local maximum before it can take on another minimum. Thus, the latter must correspond to the larger root of (71), when this is real. At this, we have $J = J_0(T)$, with

$$J_0^{1/2} = \left(3|c| + \sqrt{9c^2 - 32bd}\right)/8d . \quad (73)$$

By elementary calculation, the cubic phase $J = 0$ has the lowest energy when

$$\psi(J_0, T) > \psi(0, T) \iff c^2 < 4bd , \quad (74)$$

and we want this for $T > T_c$. Similarly, the tetragonal phase does when

$$\psi(0, T) > \psi(J_0, T) \iff c^2 > 4bd , \quad (75)$$

and we want this for $T < T_c$. Of course, T_c represents the temperature at which the two energies become equal, so

$$c^2 = 4bd \text{ at } T = T_c , \quad (76)$$

and, generically, this should be an isolated temperature. Assuming this form of $\tilde{\phi}$ applies to the A-15 superconductors, we must have $J_0(T_c)$ very small, which requires that, for T near T_c ,

$$|c|/d \ll 1 , \quad b/d \ll 1 . \quad (77)$$

So, by the indicated kind of reasoning, this gives one estimate of what $\tilde{\phi}$ might look like, near the transformation, certainly involving a bias in favor of the notion that the transformation is of first-order. One might introduce guesses about the temperature dependence of coefficients near T_c , based on ideas of smoothness. Otherwise, this seems to be the simplest kind of model which accomodates the two phases

and twinning. It could do no harm to better understand what all it predicts, and how this compares to the behavior of real crystals, but it is a somewhat naive guess.

6. EQUILIBRIUM EQUATIONS

Here, we begin by reverting to index notation. In dealing with constrained elastic materials, we follow the most common practice, which is to use the format suggested by Ericksen and Rivlin [13], to introduce kinds of Lagrange multipliers, or forces of constraint. Some possible generalizations are considered by Antman [14], who argues that they might well be of import for some kinds of theories, but not elasticity theory. Here, we write

$$\bar{\phi} = \phi - \pi(C_{11} + C_{22} + C_{33} - 3) + 2(\lambda_1 C_{23} + \lambda_2 C_{31} + \lambda_3 C_{12}) , \quad (78)$$

where π and the λ 's are arbitrary functions of position, ϕ being given by a definite constitutive equation, for example, that represented by (67). Then, ignoring the constraints, treat $\bar{\phi}$ as the potential for an unconstrained material, using any of the common formulae for calculating stresses, to be used as usual in equations of equilibrium or motion. Without really looking at such calculations, we can notice one curiosity. We have four multipliers to play with, only three equations to be satisfied. So, it seems that it should be possible to get any kinematically possible deformation to satisfy the equilibrium equations with zero body force, this still leaving only three equations to determine four unknowns. Before, we found that a naive count of equations and unknowns was misleading, so we should look more closely at the equations. For example, the Piola-Kirchhoff stress tensor is given by

$$T_i^j = \frac{\partial \phi}{\partial y^i_{,j}} = \tilde{T}^{jk} y^i_{,k} , \quad (79)$$

where

$$\tilde{T}^{jk} = \tilde{T}^{kj} = \frac{\partial \bar{\phi}}{\partial C_{jk}} + \frac{\partial \bar{\phi}}{\partial C_{kj}} , \quad (80)$$

or, in matrix form,

$$\tilde{\mathbf{T}} = \begin{vmatrix} \mu_1 & \lambda_3 & \lambda_2 \\ \lambda_3 & \mu_2 & \lambda_1 \\ \lambda_2 & \lambda_1 & \mu_3 \end{vmatrix} , \quad (81)$$

with

$$\mu_k = \frac{\partial \phi}{\partial C_{kk}} - \pi \quad (\text{no sum}) . \quad (82)$$

The equilibrium equations

$$\mathbf{T}_{i,j}^j = 0$$

can, with the help of (13), be reduced to the form

$$\tilde{\mathbf{T}}_{,l}^{i\ell} + \Gamma_{jk}^i \tilde{\mathbf{T}}^{jk} = 0 . \quad (83)$$

With C of the form (11) we get, after some calculation

$$\left. \begin{aligned} (f\lambda_3)_{,2} + (f\lambda_2)_{,3} - (f\pi)_{,1} &= \bar{\Phi}_1 , \\ (g\lambda_1)_{,2} + (g\lambda_3)_{,1} - (g\pi)_{,2} &= \bar{\Phi}_2 , \\ (h\lambda_2)_{,3} + (h\lambda_1)_{,2} - (h\pi)_{,3} &= \bar{\Phi}_3 , \end{aligned} \right\} \quad (84)$$

where

$$\begin{aligned} 2\bar{\Phi}_1 &= \left(\hat{\phi} - 2f \frac{\partial \hat{\phi}}{\partial f} \right)_{,1} , \\ 2\bar{\Phi}_2 &= \left(\hat{\phi} - 2g \frac{\partial \hat{\phi}}{\partial g} \right)_{,2} , \\ 2\bar{\Phi}_3 &= \left(\hat{\phi} - 2h \frac{\partial \hat{\phi}}{\partial h} \right)_{,3} . \end{aligned} \quad (85)$$

With the deformation given, (84) then reduces to three linear equations for the four unknown multipliers, seeming to reinforce our first impression. We do know that we have the generalized plane deformations given by (21). Assuming that the multipliers don't depend on x_3 , the above system reduces to

$$\left. \begin{aligned} (f\lambda_3)_{,2} &= (f\pi + \sigma)_{,1} \\ (g\lambda_3)_{,1} &= (g\pi + \tau)_{,2} \\ \lambda_{1,2} + \lambda_{2,1} &= 0 . \end{aligned} \right\} \quad (86)$$

with

$$\begin{aligned} 2\sigma &= \hat{\phi} - 2f \frac{\partial \hat{\phi}}{\partial f} , \\ 2\tau &= \hat{\phi} - 2g \frac{\partial \hat{\phi}}{\partial g} . \end{aligned} \quad (87)$$

Then, $(86)_1$ and $(86)_2$ can be viewed as integrability conditions for functions ξ and η such that

$$\left. \begin{aligned} f\lambda_3 &= \xi_{,1} , & f\pi + \sigma &= \xi_{,2} \\ g\lambda_3 &= \eta_{,2} , & g\pi + \tau &= \eta_{,1} . \end{aligned} \right\} \quad (88)$$

Eliminating λ_3 and π gives

$$\left. \begin{aligned} g\xi_{,1} - f\eta_{,2} &= 0 \\ g\xi_{,2} - f\eta_{,1} &= g\sigma - f\tau . \end{aligned} \right\} \quad (89)$$

One can solve for the gradient of either function, then cross-differentiate to get an equation for the other. For example, that for ξ is

$$\left[\left(\frac{g}{f} \xi \right)_{,1} \right]_{,1} - \left[\left(\frac{g}{f} \xi \right)_{,2} \right]_{,2} = (\tau - g\sigma/f)_{,2} \quad (90)$$

a linear hyperbolic equation having as characteristics our old friends, the possible twin planes. Any solution of this generates a solution of the equilibrium equations,

locally. That is, one can work back from this to get λ_3 and π , etc. Similarly, we can satisfy (86)₃ by writing λ_1 and λ_2 in terms of derivatives of an arbitrary function. Certainly, this serves to confirm the first impression.

With the several constraints, we begin to approach the situation encountered in rigid body mechanics. There, one might introduce stress as an essentially arbitrary tensor, restricted a bit by the condition that its divergence vanish, for example. The idea is more cumbersome than useful, so we use other familiar ideas to formulate and solve physical problems. It does suggest that we might need to change our thinking habits, to make effective use of these theories of highly constrained materials.

Inherently, the physical situations envisaged are complex. As should be clear from our consideration of minimizers, the simplest problem, of the equilibrium of an unloaded crystal, is nontrivial, and requires stability analyses. For various static problems, one can use the notion of minimum energy to formulate problems, building stability criteria into the formulation. Certainly, this is a sensible approach, but it is hardly a panacea. I have begun to look at some of the simplest experiments from this point of view, but find it tricky, so it seems premature to comment on this. Of course, the general format is designed to produce some agreement with a few of the observations, but not enough to firm up specific constitutive equations, or to assess the quality of predictions of such theory.

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