## ON THERMOELASTIC DIELECTRICS

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Abstract—Constitutive equations for a linear thermoelastic dielectric are derived from the energy balance equation assuming dependence of the stored energy function on the strain tensor, the polarization vector, the polarization gradient tensor and entropy. A method is indicated for constructing a hierarchy of constitutive equations for materials with arbitrary symmetry by introducing various thermodynamic potentials. Maxwell's relations are constructed for the thermodynamic potential  $W^L$ . The entropy inequality is used to obtain stability conditions for an elastic dielectric in equilibrium under prescribed boundary constraints. Frequencies are explicitly determined for a plane wave propagating along the  $x_1$ -axis in an infinite centro-symmetric isotropic thermoelastic dielectric.

#### I. INTRODUCTION

Mindlin[1] and Suhubi[2] recently extended Toupin's[3] work on elastic dielectrics and the equations of classical theory of piezo-electricity by assuming the stored energy function to depend on the strain tensor, the polarization vector as well as the polarization gradient tensor. This theory explains observed phenomena, otherwise not included in Eringen's[4] or Toupin's work[3] such as: (1) an electromechanical interaction in symmetric and non-symmetric materials; (2) capacitance of thin dielectric films. By including a magnetic field, Mindlin and Toupin[5] have investigated acoustical and optical activity in alpha quartz.

Pyroelectric crystals develop spontaneous polarization and successive changes in point group symmetry occur with change in temperature[8]. Thus, for example, barium-titanate (BaTiO<sub>3</sub>) with transition temperatures of 393, 278, 180°K transforms from class m3m to pyroelectric classes 4mm, mm2 and 3m, respectively, as the temperature is lowered. For potassium dihydrogen phosphate (KH<sub>2</sub>PO<sub>4</sub>) phase transition occurs at 123°K. Above this Curie point, the crystal is in paraelectric phase with tetragonal symmetry,  $\overline{4}2m$ , and below in a ferroelectric phase with orthorhombic symmetry mm2.

This paper deals with thermoelastic dielectrics where the contribution due to polarization gradient is taken into account. Constitutive equations for the stress tensor, electric vector, electric tensor and the temperature are derived from the energy balance equation by assuming the strain energy function to depend on the strain tensor, the polarization vector, the polarization gradient tensor, and entropy. Relations between the isothermal and adiabatic constants are derived. The arbitrary choice of independent and dependent variables and various types of boundary constraints suggests introduction of a number of thermodynamic potentials for each of which differentials and constitutive equations are derived. Maxwell's relations are obtained. As an example, constitutive equations for materials with 42m point group symmetry are constructed.

The entropy inequality is employed to determine stability conditions for an elastic dielectric with given boundary constraints. Finally, the frequencies are explicitly determined for a plane wave propagating along the  $x_1$ -axis in an infinite isotropic centro-symmetric thermoelastic dielectric.

## 2. BASIC EQUATIONS

Let a homogeneous linear elastic dielectric continuum with the contribution of the polarization gradient taken into account, occupy a region V in a rectangular Cartesian coordinate system.

The basic equations developed in [4] reduce to equations of motion

$$T_{ij,i} + \rho f_j = \rho \ddot{u}_j, \quad T_{ij} = T_{ji} \tag{2.1a}$$

$$\epsilon_{ij,i} + {}_L E_j + E_j^{MS} = -E_j^0 \tag{2.1b}$$

K. L. CHOWDHURY and P. G. GLOCKNER

$$\boldsymbol{\epsilon}_{0}\boldsymbol{\phi}_{,ii}+\boldsymbol{P}_{i,i}=-\boldsymbol{\rho}_{c}\quad\text{in}\quad V, \tag{2.1c}$$

$$\boldsymbol{\phi}_{,ii} = 0 \quad \text{in} \quad V^* \tag{2.2}$$

kinematic relations

$$E_i^{MS} = -\phi_{,i}, \quad S_{ij} = \frac{1}{2} \left( u_{i,j} + u_{j,i} \right)$$
(2.3)

the boundary conditions

$$n_i T_{ij} = k_j, \quad n_i \epsilon_{ij} = S_j \tag{2.4}$$

$$n_i[P_i - \epsilon_0 \| \boldsymbol{\phi}_{,i} \|] = \eta(x)$$
(2.5)

in which  $T_{ij}$ ,  $\epsilon_{ij}$  and  $S_{ij}$  denote components of the stress-tensor, electric tensor, and the strain tensor, respectively;  $u_i$ ,  $P_i$ ,  $_LE_i$ ,  $E_i^{MS}$ ,  $f_i$  and  $n_i$  designate components of displacement vector, polarization vector, local electric vector, the Maxwell self field vector, the external body force vector and the unit normal vector, respectively;  $\phi$ ,  $\|\phi_i\|$ ,  $\rho_c$  represent potential of Maxwell field, jump in  $\phi_{,i}$  across S and the charge density;  $k_i(x)$ ,  $S_i(x)$  and  $\eta(x)$  are surface loadings; V\* is the outer vacuum and  $\epsilon_0$  its permittivity.

The entropy density,  $\sigma$ , is defined by [6]

$$\dot{q}_{i,i} = -\theta \dot{\sigma} \tag{2.6}$$

and from Fourier's law of heat conduction one writes

$$-\dot{q}_i = k_{ij}\theta_{,i} \tag{2.7}$$

where  $q_i$  is the heat conduction vector,  $\theta$  is the absolute temperature, and  $k_{ij}$  are the heat conduction coefficients, symmetric in *i* and *j*.

### 3. ENERGY BALANCE AND CONSTITUTIVE EQUATIONS

The principle of conservation of energy for an elastic dielectric occupying a region V and bounded by a surface S can be stated as follows:

The rate of increase in total energy is equal to the rate at which work is done by the tractions across S, the external body and electric forces within V, less the outward flux of electrical and thermal energy across S. Thus the energy balance can be written as

$$\int_{V} (\dot{K} + \dot{U}) \, \mathrm{d}v = \int_{S} n_{i} [T_{ij}\dot{u}_{j} + \epsilon_{ij}\dot{P}_{j} - \dot{q}_{i}] + \eta(x)\dot{\phi} \, \mathrm{d}s + \int_{V} [\rho f_{j}\dot{u}_{j} + E_{j}^{0}\dot{P}_{j} + \rho_{c}\dot{\phi}] \, \mathrm{d}v \qquad (3.1)$$

where

$$K=\frac{\rho}{2}\,\dot{u}_i\dot{u}_i$$

and

$$U = W^{L}(S_{ij}, P_{i}, P_{j,i,\sigma}) + \phi_{,j}P_{j} - \frac{\epsilon_{0}}{2}\phi_{,i}\phi_{,i}$$
(3.2)

are the kinetic (K) energy and  $(W^L)$  the strain energy density functions of deformation and polarization.

Applying the divergence theorem to the surface integral in (3.1) and making use of eqns (2.1), one obtains

$$\int_{V} \dot{W}^{L} dv = \int_{V} [T_{ij} \dot{S}_{ij} - {}_{I} E_{j} \dot{P}_{j} + \epsilon_{ij} \dot{\Pi}_{ij} - \dot{q}_{i,i}] dv$$
(3.3)

where

$$\prod_{ij} = P_{j,i}$$

1174

Since (3.3) holds for any arbitrary volume V, we have

$$\dot{W}^{L} = T_{ij}\dot{S}_{ij} - {}_{L}E_{j}\dot{P}_{j} + \epsilon_{ij}\dot{\Pi}_{ij} + \theta\dot{\sigma}$$
(3.4)

where we have used (2.6). This is the first law of thermodynamics for a homogeneous elastic dielectric.

We shall treat  $(T_{ij}, S_{ij}), (-_L E_{j}, P_i), (\epsilon_{ij}, \Pi_{ij})$  and  $(\theta, \sigma)$  as a set of conjugate thermodynamic variables. Assuming  $S_{ij}, P_i, \Pi_{ij}, \sigma$  as an independent set, one can write

$$\dot{W}^{L} = \frac{\partial W^{L}}{\partial S_{ij}} \dot{S}_{ij} + \frac{\partial W^{L}}{\partial P_{i}} \dot{P}_{j} + \frac{\partial W^{L}}{\partial \Pi_{ij}} \dot{\Pi}_{ij} + \frac{\partial W^{L}}{\delta \sigma} \dot{\sigma}$$
(3.5)

and from a comparison of eqns (3.4) and (3.5) the system of constitutive equations are written as

$$T_{ij} = \frac{\partial W^L}{\partial S_{ij}}, \quad -_L E_i = \frac{\partial W^L}{\partial P_i}$$
  
$$\epsilon_{ij} = \frac{\partial W^L}{\partial \Pi_{ij}}, \quad \theta = \frac{\partial W^L}{\partial \sigma}.$$
 (3.6)

The Maxwell relations can now be derived as

$$\frac{\partial T_{ij}}{\partial P_m} = \frac{\partial_L E_m}{\partial S_{ij}}, \qquad \frac{\partial T_{ij}}{\partial \Pi_{kl}} = \frac{\partial \epsilon_{kl}}{\partial S_{ij}}, \qquad \frac{\partial T_{ij}}{\partial \sigma} = \frac{\partial \theta}{\partial S_{ij}}$$
$$-\frac{\partial_L E_i}{\partial \Pi_{kl}} = \frac{\partial \epsilon_{kl}}{\partial P_i}, \qquad -\frac{\partial_L E_i}{\partial \sigma} = \frac{\partial \theta}{\partial P_i}, \qquad \frac{\partial \epsilon_{kl}}{\partial \sigma} = \frac{\partial \theta}{\partial \Pi_{kl}}. \tag{3.7}$$

The choice of independent or dependent thermodynamic variables is suggested by the boundary constraints and is effected by a Legendre transformation. Following Mason[9], we list in Appendix A, 12 thermodynamic potentials for mechanical, electrical and thermal variables, each with definition, independent variables, differential relations and state equations.

Each thermodynamic potential contains a complete description of thermodynamic properties of the homogeneous elastic dielectric. For the thermodynamic process governed by the strain energy function of deformation and polarization  $W^L(S_{ij}, P_i, \Pi_{kl}, \sigma)$ , the stress tensor  $T_{ij}$ , the local electric vector  $_L E_m$ , the electric tensor  $\epsilon_{ij}$  and the temperature  $\theta$  are functions of  $S_{ij}$ ,  $P_m$ ,  $\Pi_{kl}$  and  $\sigma$ , respectively. Thus

$$T_{ij} = T_{ij}(S_{ij}, P_m, \Pi_{ij}, \sigma)$$
(3.8)

with similar relations for  $_{L}E_{m}$ ,  $\epsilon_{kl}$  and  $\theta$ .

Let us consider the differential

$$dT_{kl} = \left[\frac{\partial T_{kl}}{\partial S_{ij}}\right]^{P\Pi\sigma} dS_{ij} + \left[\frac{\partial T_{kl}}{\partial P_m}\right]^{S\Pi\sigma} dP_m + \left[\frac{\partial T_{kl}}{\partial \Pi_{ij}}\right]^{SP\sigma} d\Pi_{ij} + \left[\frac{\partial T_{kl}}{\partial \sigma}\right]^{SP\Pi} d\sigma$$
(3.9)

where the superscripts indicate the variables held constant. For a linear theory,  $W^L$  is a quadratic function of the independent variables and the coefficients are constants; thus eqns (3.9) may be integrated to give

$$T_{kl} = \left[\frac{\partial T_{kl}}{\partial S_{ij}}\right]^{P\Pi\sigma} S_{ij} + \left[\frac{\partial T_{kl}}{\partial P_i}\right]^{S\Pi\sigma} P_i + \left[\frac{\partial T_{kl}}{\partial \Pi_{ij}}\right]^{SP\sigma} \Pi_{ij} + \left[\frac{\partial T_{kl}}{\partial \sigma}\right]^{SP\Pi} \sigma_i$$

Similarly, one obtains

K. L. CHOWDHURY and P. G. GLOCKNER

$${}_{L}E_{k} = \left[\frac{\partial_{L}E_{k}}{\partial S_{ij}}\right]^{P\Pi\sigma}S_{ij} + \left[\frac{\partial_{L}E_{k}}{\partial P_{i}}\right]^{S\Pi\sigma}P_{i} + \left[\frac{\partial_{L}E_{k}}{\partial \pi_{ij}}\right]^{SP\sigma}\Pi_{ij} + \left[\frac{\partial_{L}E_{k}}{\partial \sigma}\right]^{SP\sigma}\sigma$$

$${}_{L}\epsilon_{kl} = \left[\frac{\partial\epsilon_{kl}}{\partial S_{ij}}\right]^{P\Pi\sigma}S_{ij} + \left[\frac{\partial\epsilon_{kl}}{\partial P_{i}}\right]^{S\Pi\sigma}P_{i} + \left[\frac{\partial\epsilon_{kl}}{\partial \Pi_{ij}}\right]^{SP\sigma}\Pi_{ij} + \left[\frac{\partial\epsilon_{kl}}{\partial \sigma}\right]^{SP\Pi}\sigma$$

$$\theta = \left[\frac{\partial\theta}{\partial S_{ij}}\right]^{P\Pi\sigma}S_{ij} + \left[\frac{\partial\theta}{\partial P_{i}}\right]^{S\Pi\sigma}P_{i} + \left[\frac{\partial\theta}{\partial \Pi_{ij}}\right]^{SP\sigma}\Pi_{ij} + \left[\frac{\partial\theta}{\partial \sigma}\right]^{SP\Pi}\sigma.$$
(3.10)

Keeping in mind Maxwell's relations, eqns (3.7), we assume

$$\begin{bmatrix} \frac{\partial T_{kl}}{\partial S_{ij}} \end{bmatrix}^{S\Pi\sigma} = \frac{\partial^{2} W^{L}}{\partial S_{ij}\partial S_{kl}} = c_{ijkl}^{P\Pi\sigma}$$

$$\begin{bmatrix} \frac{\partial T_{kl}}{\partial P_{j}} \end{bmatrix}^{S\Pi\sigma} = -\begin{bmatrix} \frac{\partial_{L} E_{j}}{\partial S_{kl}} \end{bmatrix}^{P\Pi\sigma} = \frac{\partial^{2} W^{L}}{\partial P_{j}\partial S_{kl}} = f_{jkl}^{\Pi\sigma}$$

$$\begin{bmatrix} \frac{\partial T_{kl}}{\partial \Pi_{ij}} \end{bmatrix}^{SP\sigma} = \begin{bmatrix} \frac{\partial \epsilon_{ij}}{\partial S_{kl}} \end{bmatrix}^{P\Pi\sigma} = \frac{\partial^{2} W^{L}}{\partial S_{kl}\partial \Pi_{ij}} = d_{ijkl}^{P\sigma}$$

$$\begin{bmatrix} \frac{\partial T_{kl}}{\partial \sigma} \end{bmatrix}^{SP\Pi} = \begin{bmatrix} \frac{\partial \theta}{\partial S_{kl}} \end{bmatrix}^{P\Pi\sigma} = \frac{\partial^{2} W^{L}}{\partial S_{kl}\partial \sigma} = \gamma_{kl}^{P\Pi}$$

$$-\begin{bmatrix} \frac{\partial_{L} E_{k}}{\partial P_{j}} \end{bmatrix}^{S\Pi\sigma} = -\begin{bmatrix} \frac{\partial_{L} E_{j}}{\partial P_{k}} \end{bmatrix}^{S\Pi\sigma} = \frac{\partial^{2} W^{L}}{\partial P_{j}\partial P_{k}} = a_{jk}^{S\Pi\sigma}$$

$$-\begin{bmatrix} \frac{\partial_{L} E_{k}}{\partial \Pi_{ij}} \end{bmatrix}^{SP\sigma} = \begin{bmatrix} \frac{\partial \epsilon_{ij}}{\partial P_{k}} \end{bmatrix}^{S\Pi\sigma} = \frac{\partial^{2} W^{L}}{\partial P_{k}\partial \Pi_{ij}} = j_{kij}^{S\sigma}$$

$$-\begin{bmatrix} \frac{\partial_{L} E_{k}}{\partial \sigma} \end{bmatrix}^{SP\Pi} = \begin{bmatrix} \frac{\partial \theta}{\partial P_{k}} \end{bmatrix}^{S\Pi\sigma} = \frac{\partial^{2} W^{L}}{\partial \sigma \partial P_{k}} = \eta_{k}^{S\Pi}$$

$$(3.11)$$

$$\begin{bmatrix} \frac{\partial \epsilon_{kl}}{\partial \Pi_{ij}} \end{bmatrix}^{SP\sigma} = \begin{bmatrix} \frac{\partial \epsilon_{ij}}{\partial \Pi_{kl}} \end{bmatrix}^{SP\sigma} = \frac{\partial^{2} W^{L}}{\partial \Pi_{ij}\partial \Pi_{kl}} = b_{ijkl}^{SP\sigma}$$

$$\begin{bmatrix} \frac{\partial \epsilon_{kl}}{\partial \sigma} \end{bmatrix}^{SP\Pi} = \begin{bmatrix} \frac{\partial \theta}{\partial \Pi_{kl}} \end{bmatrix}^{SP\sigma} = \frac{\partial^{2} W^{L}}{\partial \sigma \partial \Pi_{kl}} = \epsilon_{kl}^{SP}$$

$$\begin{bmatrix} \frac{\partial \theta}{\partial \sigma} \end{bmatrix}^{SP\Pi} = \begin{bmatrix} \frac{\partial \theta}{\partial H_{kl}} \end{bmatrix}^{SP\sigma} = \frac{\partial^{2} W^{L}}{\partial \sigma^{2} \Pi_{kl}} = \epsilon_{kl}^{SP}$$

Making use of (3.11) in (3.10), and recalling that the strain energy function of deformation and polarization is a quadratic, expressions for  $W^L$ ,  $\overline{T}$ ,  $_L\overline{E}$ ,  $\overline{\epsilon}$  and  $\theta$  may be constructed as

$$W^{L}(S_{p}, P_{k}, \Pi_{m}, \sigma) = b_{0m}\Pi_{m} + \frac{1}{2}c_{pq}^{P\Pi\sigma}S_{p}S_{q} + \frac{1}{2}a_{ij}^{S\Pi\sigma}P_{i}P_{j} + \frac{1}{2}b_{mn}^{SP\sigma}\Pi_{m}\Pi_{n} + \frac{1}{2}\beta^{SP\Pi}\sigma^{2} + d_{mp}^{P\sigma}S_{p}\Pi_{m} + f_{ip}^{\Pi\sigma}S_{p}P_{i} + \gamma_{p}^{P\Pi}S_{p}\sigma + j_{im}^{S\sigma}P_{i}\Pi_{m} + \eta_{k}^{SK}P_{k}\sigma + \epsilon_{m}^{SP}\Pi_{m}\sigma$$
(3.12)

$$T_{p} = c_{qp}^{P\Pi\sigma}S_{q} + f_{jp}^{\Pi\sigma}P_{j} + d_{mp}^{P\sigma}\Pi_{m} + \gamma_{p}^{P\Pi}\sigma$$

$$- {}_{L}E_{k} = f_{kq}^{\Pi\sigma}S_{q} + a_{kj}^{S\Pi\sigma}P_{j} + j_{km}^{S\sigma}\Pi_{m} + \eta_{k}^{S\Pi\sigma}$$

$$\epsilon_{m} = d_{m\sigma}^{P\sigma}S_{q} + j_{jm}^{S\sigma}P_{i} + b_{mn}^{SP}\Pi_{n} + \epsilon_{m}^{SP}\sigma + b_{0m}$$

$$\theta = \gamma_{q}^{PK}S_{q} + \eta_{i}^{S\Pi}P_{i} + \epsilon_{n}^{SP}\Pi_{n} + \beta^{SP\Pi}\sigma \qquad (3.13)$$

where the linear term  $b_{0m}\Pi_m$  has been added to  $W^L$  to account for the surface energy of deformation and polarization, and various indices here take ranges as follows: p, q = 1-6; m, n = 1-9; i, j, k = 1-3.

1176

		Scheme for $T_{ij}$ or $S_{kl}$														
	ij or kl	11	22	2 3	3 2	23, 32	31, 1	3 1	12, 21							
	p or q	1		2	3	4	5		6							
_	Scheme for $\epsilon_{ij}$ or $\prod_{kl}$															
	ij or kl	11	22	33	2	3 31	12	32	13	21						
	m or n	1	2	3	4	4 5	6	7	8	9						

Also, we have introduced the abbreviated indicial notation in which a pair of indices ij or kl is replaced by a single index p, or q; m or n according to

The constitutive equations may be written in various forms depending on the thermodynamic potential used. The relations (3.13) are represented by joining the corners of a cube as shown in Fig. 1.

Interchanging the positions of  $\theta$  and  $\sigma$ , the constitutive equations which may arise due to potential A (free energy) are given by

$$T_{p} = c_{qp}^{P\Pi\theta}S_{q} + f_{ip}^{\Pi\theta}P_{j} + d_{m\theta}^{P\theta}\Pi_{m} + \Delta_{p}^{P\Pi}\theta \qquad \epsilon_{m} = d_{mq}^{P\theta}S_{q} + j_{im}^{S\theta}P_{j} + b_{m\theta}^{SP\theta}\Pi_{n} + \xi_{m}^{SP}\theta + b_{0m}$$

$$-_{L}E_{k} = f_{kq}^{\Pi\theta}S_{q} + a_{kj}^{S\Pi\theta}P_{j} + j_{km}^{S\theta}\Pi_{m} + \rho_{k}^{S\Pi}\theta \qquad \sigma = -\Delta_{q}^{P\Pi}S_{q} - \rho_{j}^{S\Pi}P_{j} - \xi_{m}^{SP}\Pi_{m} + \nu^{SP\Pi}\theta.$$
(3.14)

The relations between isothermal and adiabatic constants are found to be

$$c_{qp}^{P\Pi\theta} = c_{qp}^{P\Pi\theta} - \nu^{SP\Pi} \gamma_{p}^{P\Pi} \gamma_{q}^{P\Pi}, \qquad f_{jp}^{\Pi\theta} = f_{jp}^{\Pi\sigma} - \nu^{SP\Pi} \gamma_{p}^{P\Pi} \eta_{j}^{S\Pi}$$

$$d_{mp}^{P\theta} = d_{mp}^{P\sigma} - \nu^{SP\Pi} \gamma_{p}^{P\Pi} \epsilon_{m}^{SP}, \qquad a_{kj}^{S\Pi\theta} = a_{kj}^{S\Pi\sigma} - \nu^{SP\Pi} \eta_{j}^{S\Pi} \eta_{k}^{S\Pi}$$

$$j_{km}^{S\theta} = j_{km}^{S\sigma} - \nu^{SP\Pi} \eta_{k}^{S\Pi} \epsilon_{m}^{SP}, \qquad b_{mn}^{SP\theta} = b_{mn}^{SP\sigma} - \nu^{SP\Pi} \epsilon_{m}^{SP} \epsilon_{n}^{SP}$$

$$\nu^{SP\Pi} \beta^{SP\Pi} = 1, \qquad \nu^{SP\Pi} \gamma_{p}^{P\Pi} = \Delta_{p}^{P\Pi} \qquad (3.15)$$

$$\nu^{SP\Pi} \eta_{p}^{S\Pi} = \rho_{p}^{S\Pi}, \qquad \nu^{SP\Pi} \epsilon_{m}^{SP} = \xi_{m}^{SP}.$$



Fig. 1. Geometric representation for constitutive relations (3.13).

Constitutive equations can be derived for any of 32 crystal classes by symmetry transformations and group theoretic methods [7]. Some of the constants become zero and other non-zero constants are found to bear mutual relations. As an example, we consider the crystal class with  $\overline{42m}$  (International) point group symmetry. Constitutive equations including thermal effects are constructed in Appendix B.

## 4. ENTROPY INEQUALITY AND STABILITY

The second law of thermodynamics permits only those transformations of the elastic dielectric system which make the total entropy change either positive or zero.

Let the system suffer infinitesimal transformations in which various field changes are denoted by  $\delta S_{ij}$ ,  $\delta P_i$ ,  $\delta \Pi_{ij}$ ,  $\delta \phi$  and  $\delta \sigma$  and let it be subjected to the boundary constraints

$$n_i T_{ij} \delta u_j = \text{const.}$$
  $n_i \epsilon_{ij} \delta P_j = \text{const.}$   
 $n_i (\epsilon_0 \phi_{,i} - P_i) \delta \phi = \text{const.}$  (4.1)

Let  $\delta W$  be the work done by the external fields  $f_i$ ,  $E_i^0$  and  $\rho_c$ . Then

$$\delta W = \int_{V} f_{i} \delta u_{i} \, \mathrm{d}v + \int_{V} E_{i}^{0} \delta P_{i} \, \mathrm{d}v + \int_{V} \rho_{c} \delta \phi \, \mathrm{d}v. \tag{4.2}$$

Making use of eqns (2.1), the divergence theorem and boundary constraints (4.1), one establishes

$$-\delta W = \delta \int_{v} \left[ -T_{ij}S_{ij} + {}_{L}E_{i}P_{j} - \epsilon_{ij}\Pi_{ij} - \phi_{,i}P_{j} \right] dv + \frac{\epsilon_{0}}{2} \delta \int_{v} \phi_{,i}\phi_{,i} dv.$$
(4.3)

The corresponding energy change is given by

$$\delta U = \delta \int_{V} \left[ W^{L}(S_{ij}, P_{i}, \Pi_{ij}, \sigma) + \phi_{,i} P_{i} - \frac{\epsilon_{0}}{2} \phi_{,i} \phi_{,i} \right] \mathrm{d}v.$$
(4.4)

Let the elastic dielectric be in thermal contact with a heat bath where the heat transferred from the dielectric to the heat bath =  $-(\delta U - \delta W)$  and the entropy change of the heat bath =  $-(\delta U - \delta W)/\theta$ . Therefore

the total entropy change 
$$= -\frac{\delta U - \delta W}{\theta} + \delta \int_{V} \sigma \, dv$$
$$= -\delta S$$

where

$$S = \int_{V} [W^{L} - T_{ij}S_{ij} + {}_{L}E_{j}P_{j} - \epsilon_{ij}\Pi_{ij} - \theta\sigma] dv \qquad (4.5)$$

and thus the entropy inequality reduces to

$$\delta S \le 0. \tag{4.6}$$

A thermodynamic system of an elastic dielectric described by the state variables  $S_{ij}$ ,  $T_{ij}$ ,  $P_{i}$ ,  $_{L}E_{i}$ ,  $\epsilon_{ij}$ ,  $\theta$ ,  $\sigma$  is said to be in a state of stable equilibrium under a prescribed set of constraints if an arbitrary set of small transformations  $\delta S_{ij}$ ,  $\delta P_i$ ,  $\delta \Pi_{ij}$ ,  $\delta \sigma$  will carry this state into an adjacent state in such a manner that

(i) imposed constraints are not violated

(ii)  $\delta S$  is positive definite (i.e. S is minimum).

In general, the constraints suggest the choice of the thermodynamic potential and stability is determined by the extremal properties of an integral similar to (4.5).

We proceed now to determine the restriction imposed on the behavior of the integrand of S in a stable state under the constraints (4.1). It is found that

$$\delta S = \frac{1}{2} \int_{v} \bar{\chi}^{t} \bar{L} \bar{\chi} \, \mathrm{d}v$$

where  $\bar{\chi}$  is the 19 dimensional vector

$$\bar{\chi} = [\delta S_p, \delta P_i, \delta \Pi_m, \delta \sigma], \quad p = 1-6; \quad i = 1-3, \quad m = 1-9.$$

superscript t stands for transpose, and  $\vec{L}$  is a 19 × 19 matrix

$$L = \begin{bmatrix} \frac{\partial f}{\partial \bar{S}} & \frac{\partial f}{\partial \bar{P}} & \frac{\partial f}{\partial \bar{\Pi}} & \frac{\partial f}{\partial \sigma} \\ -\frac{\partial_L \bar{E}}{\partial \bar{S}} - \frac{\partial_L E}{\bar{P}} & \frac{\partial_I \bar{E}}{\partial \bar{\Pi}} - \frac{\partial_L \bar{E}}{\partial \sigma} \\ \frac{\partial \bar{e}}{\partial \bar{S}} & \frac{\partial \bar{e}}{\partial \bar{P}} & \frac{\partial \bar{e}}{\partial \bar{\Pi}} & \frac{\partial \bar{e}}{\partial \sigma} \\ \frac{\partial \theta}{\partial \bar{S}} & \frac{\partial \theta}{\partial \bar{P}} & \frac{\partial \theta}{\partial \bar{\Pi}} & \frac{\partial \theta}{\partial \sigma} \end{bmatrix}$$
(4.7)

where we have used eqns (3.6) and (3.7).

Thus the necessary and sufficient conditions for stability of the elastic dielectric are that the principal minors of the matrix, (4.7), are all positive.

# 5. PLANE WAVES

For the homogeneous isotropic elastic dielectric the constitutive coefficients take the form

$$f_{ijk} = 0, \quad j_{ijk} = 0, \quad b_{ij}^{0} = b_{0}\delta_{ij}, \quad a_{ij} = a\delta_{ij}$$

$$\Delta_{ij} = \Delta\delta_{ij}, \quad \zeta_{ij} = \zeta\delta_{ij}$$

$$b_{ijkl} = b_{12}\delta_{ij}\delta_{kl} + b_{44}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) + b_{77}(\delta_{ik}\delta_{jl} - \delta_{il}\delta_{jk})$$

$$c_{ijkl} = c_{12}\delta_{ij}\delta_{kl} + c_{44}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})$$

$$d_{ijkl} = d_{12}\delta_{ij}\delta_{kl} + d_{44}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})$$
(5.1)

where  $\delta_{ij}$  is the Kronecker delta. The constitutive equations reduce to

$$T_{ij} = d_{12}\delta_{ij}P_{k,k} + d_{44}(P_{j,i} + P_{i,j}) + c_{12}\delta_{ij}S_{kk} + 2c_{44}S_{ij} + \Delta\delta_{ij}\theta$$
  
$$-_{L}E_{k} = aP_{k}$$
  
$$\epsilon_{ij} = b_{12}\delta_{ij}P_{k,k} + b_{44}(P_{j,i} + P_{i,j}) + b_{77}(P_{j,i} - P_{i,j}) + d_{12}\delta_{ij}S_{kk} + 2d_{44}S_{ij} + b_{0}\delta_{ij} + \zeta\delta_{ij}\theta$$
  
$$\sigma = -\Delta S_{k,k} - \zeta P_{k,k} + \nu\theta$$
(5.2)

where we have suppressed the superscripts on various constants.

Substituting (5.2) into (2.1), the equations of motion reduce to

$$c_{44}\nabla^{2}u_{j} + (c - c_{44})u_{i,ij} + d_{44}\nabla^{2}P_{j} + (d - d_{44})P_{i,ij} + \Delta\theta_{,j} = \rho \ddot{u}_{j}$$

$$d_{44}\nabla^{2}u_{j} + (d - d_{44})u_{i,ij} + b^{*}\nabla^{2}P_{j} + (b - b^{*})P_{i,ij} - aP_{j} - \phi_{,j} + \zeta\theta_{,j} = 0$$

$$- \epsilon_{0}\nabla^{2}\phi + P_{i,i} = 0$$

$$- \Delta u_{i,i} - \zeta P_{i,i} + \nu \dot{\theta} = \theta_{0}^{-1}K_{ij}\theta_{,ij}$$
(5.3)

K. L. CHOWDHURY and P. G. GLOCKNER

where

$$x = x_{12} + 2x_{44}$$
 ( $x = b, c, d$ ) and  $b^* = b_{44} + b_{77}$ 

Consider a plane wave in an infinite crystal, with its normal in the  $x_1$  direction; i.e. assume

$$u_{j} = A_{j} \cos \xi x_{1} e^{i\omega t}, \qquad P_{j} = B_{j} \cos \xi x_{1} e^{i\omega t}$$
  
$$\phi = C \sin \xi x_{1} e^{i\omega t}, \qquad \theta = D \sin \xi x_{1} e^{i\omega t}. \qquad (5.4)$$

Substituting (5.4) into (5.3) and setting the determinant of the coefficient matrix of vector  $(A_1, A_2, A_3, B_1, B_2, B_3, C, D)$  equal to zero, one obtains the secular equation

$$\begin{vmatrix} c - \lambda^2 & 0 & 0 & d & 0 & 0 & -\Delta \\ 0 & c_{44} - \lambda^2 & 0 & 0 & d_{44} & 0 & 0 \\ 0 & 0 & c_{44} - \lambda^2 & 0 & 0 & d_{44} & 0 \\ d & 0 & 0 & b + a + \epsilon_0^{-1} & 0 & 0 & 0 \\ 0 & d_{44} & 0 & 0 & b^* + a & 0 & 0 \\ 0 & 0 & d_{44} & 0 & 0 & b^* + a & 0 \\ \Delta & 0 & 0 & \zeta & 0 & 0 & \Omega \end{vmatrix} = 0$$
(5.5)

where

.

$$\lambda^{2} = \frac{\rho \omega^{2}}{\xi^{2}}, \quad \Omega = i \omega \nu + \theta_{0}^{-1} K_{11}.$$
 (5.6)

The determinant (5.5) is further simplified and the three values of  $\lambda^2$  are explicitly determined and found to be

$$\lambda_1^2 = \lambda_2^2 = c_{44} - \frac{d_{44}^2}{a+b^*}$$
(5.7)

$$\lambda_3^2 = c + \frac{\Delta}{\Omega} \left[ \Delta - \frac{\zeta d}{b + a + \epsilon_0^{-1}} \right].$$
 (5.8)

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#### REFERENCES

- 1. R. D. Mindlin, Polarization gradient in elastic dielectrics. Inter. J. Solids. Structures 4, 637 (1968).
- 2. E. S. Suhubi, Elastic dielectrics with polarization gradient. Inter. J. Engg. Sci. 7, 993 (1969).
- 3. R. A. Toupin, The elastic dielectric. J. Rat. Mech. Analysis 5, 849 (1956).
- 4. A. C. Eringen, On the foundations of electro-elastostatics. Inter. J. Engg. Sci. 1, 127 (1963).
- 5. R. D. Mindlin and R. A. Toupin, Acoustical and optical activity in alpha quartz. Inter. J. Solids. Structures 7, 1219 (1971).
- 6. R. D. Mindlin, On the equations of motion of piezo-electric crystals. Problems of Continuum Mechanics SIAM, Pennsylvania (1961).
- 7. J. F. Nye, Physical Properties of Crystals. Oxford Press, Oxford (1960).
- 8. J. Grindlay, An Introduction to the Phenomenological Theory of Ferroelectricity. Pergamon Press, Oxford (1970).
- 9. W. P. Mason, Crystal Physics of Interaction Processes. Academic Press, New York (1966).

#### APPENDIX A

Some useful thermodynamic potentials:

(1) Strain Energy: 
$$W^{L}(S_{ij}, P_{i}, \Pi_{ij}, \sigma)$$
:

$$\begin{split} \mathbf{d} \, \mathbf{W}^{L} &= T_{ij} \, \mathbf{d} S_{ij} - {}_{L} E_{j} \, \mathbf{d} P_{j} + \boldsymbol{\epsilon}_{ij} \, \mathbf{d} \Pi_{ij} + \boldsymbol{\theta} \, \mathbf{d} \sigma \\ T_{ij} &= \frac{\partial \mathbf{W}^{L}}{\partial S_{ij}}, \quad -{}_{L} E_{i} = \frac{\partial \mathbf{W}^{L}}{\partial P_{j}}, \quad \boldsymbol{\epsilon}_{ij} = \frac{\partial \mathbf{W}^{L}}{\partial \Pi_{ij}}, \quad \boldsymbol{\theta} = \frac{\partial \mathbf{W}^{L}}{\partial \sigma} \end{split}$$

(2) Free Energy:  $W^L - \theta \sigma = A(S_{ib}, P_c, \Pi_{ic}, \theta)$ 

$$dA = T_{ij} dS_{ij} - {}_{L}E_{j} dP_{j} + \epsilon_{ij} d\Pi_{ij} - \sigma d\theta$$
  
$$T_{ij} = \frac{\partial A}{\partial S_{ij}}, \quad -{}_{L}E_{i} = \frac{\partial A}{\partial P_{i}}, \quad \epsilon_{ij} = \frac{\partial A}{\partial \Pi_{ij}}, \quad \sigma = -\frac{\partial A}{\partial \theta}.$$

1180

(3) Enthalpy:  $H(T_{ij}, _{L}E_{j}, \epsilon_{ij}, \sigma) = W^{L} - T_{ij}S_{ij} + _{L}E_{j}P_{j} - \epsilon_{ij}\Pi_{ij}$   $dH = -S_{ij} dT_{ij} + P_{j} d_{L}E_{j} - \Pi_{ij} d\epsilon_{ij} + \theta d\sigma$  $S_{ij} = -\frac{\partial H}{\partial T_{ij}}, \quad P_{j} = \frac{\partial H}{\partial_{L}E_{j}}, \quad \Pi_{ij} = -\frac{\partial H}{\partial \epsilon_{ij}}, \quad \theta = \frac{\partial H}{\partial \sigma}.$ 

(4) Elastic Enthalpy:  $H_1(T_{ij}, P_j, \Pi_{ij}, \sigma) = W^L - S_{ij}T_{ij}$ 

$$dH_{1} = -S_{ij} dT_{ij} - {}_{L}E_{j} dP_{j} + \epsilon_{ij} d\Pi_{ij} + \theta d\sigma$$
$$S_{ij} = -\frac{\partial H_{1}}{\partial T_{ij}}, \quad {}_{L}E_{i} = -\frac{\partial H_{1}}{\partial P_{i}}, \quad \epsilon_{ij} = \frac{\partial H_{1}}{\partial \Pi_{ij}}, \quad \theta = \frac{\partial H_{1}}{\partial \sigma}$$

(5) Electric Enthalpy:  $H_2(S_{ij}, LE_i, \epsilon_{ij}, \sigma) = W^L + {}_LE_iP_j - \epsilon_{ij}\Pi_{ij}$ 

$$dH_2 = T_{ij} dS_{ij} + P_j d_L E_j - \Pi_{ij} d\epsilon_{ij} + \theta d\sigma$$
  
$$T_{ij} = \frac{\partial H_2}{\partial S_{ij}}, \quad P_j = + \frac{\partial H_2}{\partial_L E_j}, \quad \Pi_{ij} = -\frac{\partial H_2}{\partial \epsilon_{ij}}, \quad \theta = \frac{\partial H_2}{\partial \sigma}$$

(6) Electric Enthalpy:  $H_2^*(S_{ij}, E_i, \Pi_{ij}, \sigma) = W^L + {}_L E_i P_i$ 

$$dH_{2}^{*} = T_{ij} dS_{ij} + P_{j} d_{L} E_{i} + \epsilon_{ij} d\Pi_{ij} + \theta d\sigma$$
  
$$T_{ij} = \frac{\partial H_{2}^{*}}{\partial S_{ij}}, \quad P_{j} = \frac{\partial H_{2}^{*}}{\partial_{L} E_{j}}, \quad \epsilon_{ij} = \frac{\partial H_{2}^{*}}{\partial \Pi_{ij}}, \quad \theta = \frac{\partial H_{2}^{*}}{\partial \sigma}$$

(7) Electric Enthalpy:  $H_2^{**}(S_{ij}, P_p, \epsilon_{ij}, \sigma) = W^L - \epsilon_{ij} \Pi_{ij}$ 

$$\begin{split} dH_2^{**} &= T_{ij} \, dS_{ij} - {}_L E_j \, dP_j - \Pi_{ij} \, d\epsilon_{ij} + \theta \, d\sigma \\ T_{ij} &= \frac{\partial H_2^{**}}{\partial S_{ij}}, \quad {}_L E_j = -\frac{\partial H_2^{**}}{\partial P_j}, \quad \Pi_{ij} = -\frac{\partial H_2^{**}}{\partial \epsilon_{ij}}, \quad \theta = \frac{\partial H_2^{**}}{\partial \sigma} \end{split}$$

(8) Gibbs function:  $G(T_{ij}, LE_j, \epsilon_{ij}, \theta) = W^L - S_{ij}T_{ij} + LE_jP_j - \prod_{ij}\epsilon_{ij} - \theta\sigma$ 

$$\begin{split} \mathrm{d}G &= -S_{ij}\,\mathrm{d}T_{ij} + P_i\,\mathrm{d}_L E_j - \Pi_{ij}\,\mathrm{d}\epsilon_{ij} - \sigma\,\mathrm{d}\theta\\ S_{ij} &= -\frac{\partial G}{\partial T_{ij}}, \quad P_j = \frac{\partial G}{\partial_L E_j}, \quad \Pi_{ij} = -\frac{\partial G}{\partial \epsilon_{ij}}, \quad \sigma = -\frac{\partial G}{\partial \theta}. \end{split}$$

(9) Elastic Gibbs function:  $G_1(T_{ij}, P_j, \Pi_{ij}, \theta) = W^L - S_{ij}T_{ij} - \theta\sigma$ 

$$dG_{1} = -S_{ij} dT_{ij} - {}_{L}E_{j} dP_{j} + \epsilon_{ij} d\Pi_{ij} - \sigma d\theta$$
  
$$S_{ij} = -\frac{\partial G_{1}}{\partial T_{ij}}, \quad {}_{L}E_{j} = -\frac{\partial G_{1}}{\partial P_{j}}, \quad \epsilon_{ij} = \frac{\partial G_{1}}{\partial \Pi_{ij}}, \quad \sigma = -\frac{\partial G_{1}}{\partial \theta}.$$

(10) Electric Gibbs function:  $G_2(S_{ij}, LE_j, \epsilon_{ij}, \theta) = W^L + {}_LE_jP_j - \epsilon_{ij}\Pi_{ij} - \theta\sigma$ 

$$\begin{split} \mathrm{d}G_2 &= T_{ij}\,\mathrm{d}S_{ij} + P_j\,\mathrm{d}_L E_f - \Pi_{ij}\,\mathrm{d}\epsilon_{ij} - \sigma\,\mathrm{d}\theta\\ T_{ij} &= \frac{\partial G_2}{\partial S_{ij}}, \quad P_j = \frac{\partial G_2}{\partial_L E_j}, \quad \Pi_{ij} = -\frac{\partial G_2}{\partial \epsilon_{ij}}, \quad \sigma = -\frac{\partial G_2}{\partial \theta}. \end{split}$$

(11) Electric Gibbs function:  $G_2^*(S_{ij}, _LE_{j}, \Pi_{ij}, \theta) = W^L + _LE_jP_j - \theta\sigma$ 

$$dG_2^* = T_{ij} dS_{ij} + P_j d_L E_j + \epsilon_{ij} d\Pi_{ij} - \sigma d\theta$$
  
$$T_{ij} = \frac{\partial G_2^*}{\partial S_{ij}}, \quad P_j = \frac{\partial G_2^*}{\partial_L E_j}, \quad \epsilon_{ij} = \frac{\partial G_2^*}{\partial \Pi_{ij}}, \quad \sigma = -\frac{\partial G_2^*}{\partial \theta}.$$

(12) Electric Gibbs function:  $G_2^{**}(S_{ij}, P_j, \epsilon_{ij}, \theta) = W^L - \dot{\epsilon_{ij}} \prod_{ij} - \theta \sigma$ 

$$dG_{2}^{**} = T_{ij} dS_{ij} - {}_{L}E_{j} dP_{j} - \Pi_{ij} d\epsilon_{ij} - \sigma d\theta$$
  
$$T_{ij} = \frac{\partial G_{2}}{\partial S_{ij}}, \quad -{}_{L}E_{j} = \frac{\partial G_{2}^{**}}{\partial P_{j}}, \quad \Pi_{ij} = -\frac{\partial G_{2}^{**}}{\partial \epsilon_{ij}}, \quad \sigma = -\frac{\partial G_{2}^{**}}{\partial \theta}.$$

APPENDIX B

Constitutive relations (3.13) for crystal class  $\overline{4}2m(D_{2d})$ .

	s <sub>11</sub>	\$ <sub>22</sub>	s <sub>33</sub>	<sup>25</sup> 23	25 <sub>31</sub>	2512	P }	۴2	P 3	P1,1	P2,2	P3.3	P3,2	P2,3	P <sub>1,3</sub>	P <sub>3,1</sub>	P2,1	P1,2	c
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т <sub>22</sub>	°12	°11	°13	c	0	0	¢	с	6	<sup>d</sup> 12	<sup>d</sup> 11	<sup>d</sup> 31	9	0	С	0	ò	υ	۲ <sub>11</sub>
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Ŧ <sub>23</sub>	c	С	0	e <sub>44</sub>	0	e	f <sub>:4</sub>	Ç	Ģ	e	າ	0	d44	<sup>d</sup> 55	C	Ċ	С	0	0
т <sub>31</sub>	C	C	0	0	°44	C	c	۴ <sub>14</sub>	C	Û	0	э	0	c	<sup>d</sup> 55	d <sub>44</sub>	с	C	с
τ <sub>12</sub>	C	0	0	0	0	66	c	C	<sup>f</sup> 36	с	0	0	0	0	C	C	<sup>d</sup> 66	<sup>d</sup> 66	0
-L <sup>E</sup> 1	0	0	0	f <sub>14</sub>	с	0	a11	ç	0	0	0	c	j <sub>14</sub>	1 <sub>17</sub>	0	C	С	0	c
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- <sub>1.</sub> E <sub>3</sub>	0	0	0	С	0	f 36	Q	0	a 33	U	0	0	0	0	0	0	<sup>j</sup> 36	÷ 36	С
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F. 22	å <sub>12</sub>	d <sub>11</sub>	۹ <sup>13</sup>	с	0	0	c	0	С	<sup>b</sup> 12	<sup>b</sup> 11	5 <sub>13</sub>	С	0	0	С	G	C	٤11
е <sub>33</sub>	d 31	<sup>d</sup> 31	<sup>d</sup> 33	С	0	с	0	C	С	<sup>b</sup> 31	<sup>b</sup> 31	<sup>6</sup> 33	0	С	C	Ċ	6	0	¢22
E23	0	0	С	ć <sub>44</sub>	0	C	<sup>j</sup> 14	С	c	с	0	0	6 <sub>44</sub>	<sup>b</sup> 47	с	C	С	C	C
E 32	0	C	C	ċ55	0	С	J <sub>17</sub>	¢	c	c	0	С	t47	<sup>b</sup> 55	0	Ċ	с	0	U
F 31	0	0	0	Ü	a <sub>55</sub>	C	Ũ	:17	c	c	0	0	0	C	<sup>5</sup> 55	t <sub>47</sub>	¢	С	C
E 13	0	0	С	C	d <sub>44</sub>	0	¢	1 <sub>14</sub>	0	G	0	С	0	0	Ъ <sub>47</sub>	ь <sub>44</sub>	с	C	с
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F. 21	0	C	0	C	C	<sup>d</sup> 66	0	c	<sup>1</sup> 36	0	9	c	0	0	C	0	<sup>ь</sup> 69	<sup>b</sup> 6ú	C
ę	۲ <sub>11</sub>	۲ <sub>11</sub>	۲ <sub>33</sub>	С	С	с	0	0	C	٤11	٤11	٤33	C	0	с	0	0	с 	f