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Deformable dielectrics
III. A model of interactions

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THE FIELD equations and thermodynamical equations developed in the first two parts of this work are recovered by using a direct approach through the postulate of global balance laws and by considering a simple model for the interactions that occur between the material lattice and the polarization sub-lattices in deformable ferroelectrics.

Równania polowe i termodynamiczne, wyprowadzone w dwóch pierwszych częściach tej pracy, otrzymuje się powtórnie w sposób bezpośredni za pomocą postulatu równowagi globalnej oraz przez rozważenie prostego modelu oddziaływania jakie występuje między siatką materialną a polaryzacyjnymi siatkami drugiego rzędu w odkształcalnych ferroelektrykach.

Полевые и термодинамические уравнения, выведенные в двух первых частях данной работы, получаются повторно непосредственным образом при помощи постулата глобального равновесия, а также путем рассмотрения простой модели взаимодействий, какие выступают между материальной решеткой и поляризационными решетками второго порядка в деформируемых ферроэлектриках.

1. Introduction

IN THE PREVIOUS two papers of this series [1, 2][1] a theory of deformable dielectrics made of several molecular species has been constructed on rational grounds using as starting points: (i) the principle of virtual power along with the objectivity requirement for the internal forces — the latter being introduced via the duality inherent in the method; (ii) the two principles of thermodynamics. Although it clearly was a theory of interactions between deformations and electromagnetic fields, no specific model was used to introduce the new internal forces or phenomenological fields. If one desires to construct the same theory with a similar thermodynamical generality (Lagrangian and Hamiltonian principles are here excluded because of their limitation to the description of thermodynamically reversible phenomena; furthermore, they require a postulate for the functional dependence of the relevant potential, so that the behavior of the material is selected from the start), one must consider the so-called "direct approach", which consists in postulating the various global balance laws such as those of linear momentum, angular momentum and energy, but also in the present case new global balance laws that will govern the new fields introduced, such as the polarization. To do so we must consider some kind of model for the interactions that take place between the usual deformation field and these new fields. This is what is done below for the theory developed in Parts One and Two. The

[1] Equations of Parts I and II are accordingly referred to by I and II followed by their number. We refer to I and II for the main notation.

interactions will be represented by some phenomenological fields for which one needs to construct constitutive equations, and which are shown, at least for the nonlinear theory, to participate in all field equations, thus providing the necessary couplings[2]. This notion of "interaction" field is very similar, and can be traced back, to the concept of molecular field introduced by Pierre WEISS [4] in ferromagnetism. The model that follows in a certain sense is a generalization (to dynamical processes and to the case of several molecular species) of a simpler but ingenious model considered by TIERSTEN [5]. It also clearly is the dielectric counterpart of the model considered by the author [6] in his theory of deformable ferrimagnetic bodies. It is shown that all local field equations and energetic expressions obtained in Parts One and Two are recovered.

2. The model of interactions

We shall consider the following model of interactions. Although some microscopic concepts are used, there is no one-to-one correspondence between these concepts and those introduced below. That is, the model is purely phenomenological. The dielectric material is considered to be the assembly of $n+1$ co-existing and co-extensive continua, one of which being the usual material lattice, the substratum of elastic deformations, and the n remaining ones being the polarization sub-lattices $\mathbf{P}_\alpha$ that arise from the internal polarization of n molecular species, $\alpha = 1, 2, \ldots, n$, and contribute to the total volume polarization according to the formula

$$\mathbf{P} = \varrho\boldsymbol{\pi} = \sum_\alpha \mathbf{P}_\alpha, \quad \mathbf{P}_\alpha = \varrho c_\alpha \boldsymbol{\pi}_\alpha, \quad \varrho_\alpha = \varrho c_\alpha. \tag{2.1}$$

Here ϱ is the matter density at time t, $\boldsymbol{\pi}$ is the total electric polarization per unit mass, $\boldsymbol{\pi}_\alpha$ is the polarization per unit mass of α-molecular species, ϱ_α is the mass density of the α-molecular species, and c_α is the corresponding mass concentration. Assuming that no chemical processes occur between these species, we have

$$\dot{c}_\alpha = 0. \tag{2.2}$$

Hence the volumetric behavior of each molecular species is the same as that of the material lattice. Next, the electric dipole moment density associated with each molecular species has charge q_α and results from an elementary displacement $\mathbf{w}_\alpha$, so that we shall have to write an equation of balance of linear momentum to govern this displacement. In a co-moving frame, each polarization sub-lattice is subjected to the action of the local electromotive intensity $\mathscr{E}$ — see Part I — and, due to the interactions with the material lattice and the other molecular species, to the action of a local electric field ${}^L\mathbf{E}_\alpha$ — a phenomenological field for which a constitutive equation must be constructed. Each electric dipole moment is of course subjected in the same fashion to couples $\varrho_\alpha\boldsymbol{\pi}_\alpha \times \mathscr{E}$ and $\varrho_\alpha\boldsymbol{\pi}_\alpha \times {}^L\mathbf{E}_\alpha$, which tend to orient the dipole along the fields $\mathscr{E}$ and ${}^L\mathbf{E}_\alpha$, respectively. The material lattice is subjected to usual contact actions and to ponderomotive forces, thus also to surface ponderomotive forces when we consider the whole spatial volume occupied by

[2] We have tried to make clear these general features in Ref. [3].

the material lattice. We suppose however that it is not subjected to a ponderomotive couple, the latter being assumed to be directly transmitted to each polarization sub-lattice (cf. the terms $\varrho_\alpha \pi_\alpha \times \mathscr{E}$ referred to above). Nonetheless, it suffers from each molecular species the reaction of the local electric field ${}^L\mathbf{E}_\alpha$ in the form of a volume couple $\varrho_\alpha {}^L\mathbf{E}_\alpha \times \boldsymbol{\pi}_\alpha$, so that, in virtue of Newton's law of action and reaction, the two couples $\varrho_\alpha \pi_\alpha \times {}^L\mathbf{E}_\alpha$ and $\varrho_\alpha {}^L\mathbf{E}_\alpha \times \boldsymbol{\pi}_\alpha$ nullify each other when we consider globally the dielectric continuum. This last property will be used in writing the global statement of the first principle of thermodynamics for the whole dielectric continuum. This simple model is sufficient to enable us to write down the relevant global balance laws.

3. Global balance laws

Let U be the matter velocity, f the volume body force of nonelectromagnetic origin acting on the material lattice, T the prescribed mechanical surface traction, x the spatial position with respect to some fixed origin 0, e the internal energy density, E and B the electric field and magnetic induction expressed in a fixed Galilean frame, h the heat supply per unit mass, and q the heat influx vector through the regular boundary $\partial\mathscr{D}_t$ — of unit outward normal n — of the open, bounded, simply-connected spatial domain $\mathscr{D}_t$ occupied by the dielectric continuum at time t. On account of the above-described model we can set forth the following global conservation laws:

A. *Mass conservation*

$$\frac{d}{dt}\int_{\mathscr{D}_t} \varrho dv = 0. \tag{5.1}$$

B. *Balance of linear momentum for the material lattice*

$$\frac{d}{dt}\int_{\mathscr{D}_t} \varrho \mathbf{U} dv = \int_{\mathscr{D}_t} (\mathbf{f}+\mathbf{f}^{\mathrm{em}}) dv + \int_{\partial\mathscr{D}_t} (\mathbf{T}+\mathbf{T}^{\mathrm{em}}) da. \tag{3.2}$$

C. *Balance of linear momentum for each polarization sub-lattice* ($\alpha = 1, 2, \ldots, n$)

$$\frac{d}{dt}\int_{\mathscr{D}_t} \varrho_\alpha \dot{\mathbf{w}}_\alpha dv = \int_{\mathscr{D}_t} \varrho_\alpha (\mathscr{E} + {}^L\mathbf{E}_\alpha) dv. \tag{3.3}$$

D. *Balance of angular momentum for the material lattice*

$$\frac{d}{dt}\int_{\mathscr{D}_t} (\mathbf{x}\times\varrho\mathbf{U}) dv = \int_{\mathscr{D}_t} [\mathbf{x}\times(\mathbf{f}+\mathbf{f}^{\mathrm{em}})+\mathbf{c}] dv + \int_{\partial\mathscr{D}_t} [\mathbf{x}\times(\mathbf{T}+\mathbf{T}^{\mathrm{em}})] da. \tag{3.4}$$

E. *Balance of angular momentum for each polarization sub-lattice* ($\alpha = 1, 2, \ldots, n$)

$$\frac{d}{dt}\int_{\mathscr{D}_t} \varrho_\alpha \mathbf{w}_\alpha \times \dot{\mathbf{w}}_\alpha dv = \int_{\mathscr{D}_t} \varrho_\alpha \mathbf{w}_\alpha \times (\mathscr{E} + {}^L\mathbf{E}_\alpha) dv. \tag{3.5}$$

F. *First principle of thermodynamics for the whole continuum*

$$\frac{d}{dt}\int_{\mathscr{D}_t} \left[\frac{1}{2}\varrho\mathbf{U}^2 + k^{(p)} + \varrho e + \frac{1}{2}(\mathbf{E}^2+\mathbf{B}^2)\right] dv = \int_{\mathscr{D}_t} (\mathbf{f}\cdot\mathbf{U}+\varrho h) dv + \int_{\partial\mathscr{D}_t} (\mathbf{T}\cdot\mathbf{U}-\mathbf{q}\cdot\mathbf{n}) da. \tag{3.6}$$

G. *Second principle of thermodynamics for the whole continuum*

$$\frac{d}{dt}\int_{\mathscr{P}_t}\varrho\eta\,dv \geqq \int_{\mathscr{P}_t}\theta^{-1}\varrho h\,dv - \int_{\partial\mathscr{P}_t}\theta^{-1}\tilde{\mathbf{q}}\cdot\mathbf{n}\,da. \tag{3.7}$$

The new quantities introduced in these equations are: η = entropy per unit mass; θ = = thermodynamical temperature; $\tilde{\mathbf{q}} \equiv \mathbf{q} - \mathscr{S}$, where $\mathscr{S}$ is the Poynting vector in a co-moving frame (Cf. Part I). $\mathbf{c}$ is the volume couple acting upon the material lattice. The volume force $\mathbf{f}^{em}$ and the surface force $\mathbf{T}^{em}$ have the expressions given in Part I (Eqs. (I–2.11) and (I–2.24), respectively). $k^{(p)}$ is the kinetic energy associated with the dipole displacements $\mathbf{w}_\alpha$. That is, according to Appendix A of Part One (also Eqs. (I–2.4))

$$k^{(p)} = \varrho\sum_\alpha \frac{1}{2}c_\alpha d_\alpha \dot{\boldsymbol{\pi}}_\alpha^2, \quad d_\alpha \equiv \left(\frac{\varrho_\alpha}{q_\alpha}\right)^2, \quad \boldsymbol{\pi}_\alpha = \frac{q_\alpha}{\varrho_\alpha}\mathbf{w}_\alpha. \tag{3.8}$$

In supplement to Eqs. (3.1) through (3.6) must also be postulated the global form of Maxwell's equations. We shall not recall here these equations, which do not intervene in the argument.

We claim that Eqs. (3.1) through (3.6) yield the same local governing equations for the continuum and the same local energy statements as the method used in Parts One and Two. To prove this statement in the next section we however need to recall the following global energy identity satisfied by the electromagnetic fields. We identically have (cf. [7]; here the magnetization in a co-moving frame and the conduction current are zero):

$$\frac{d}{dt}\int_{\mathscr{P}_t}\frac{1}{2}(\mathbf{E}^2+\mathbf{B}^2)\,dv = -\int_{\mathscr{P}_t}(\mathbf{f}^{em}\cdot\mathbf{U}+\varrho\mathscr{E}\cdot\dot{\boldsymbol{\pi}})\,dv - \int_{\partial\mathscr{P}_t}(\mathbf{T}^{em}\cdot\mathbf{U}+\mathscr{S}\cdot\mathbf{n})\,da. \tag{3.9}$$

Combining Eqs. (3.6) and (3.9), we obtain

$$\frac{d}{dt}\int_{\mathscr{P}_t}\left(\frac{1}{2}\varrho\mathbf{U}^2+k^{(p)}+\varrho e\right)dv = \int_{\mathscr{P}_t}[(\mathbf{f}+\mathbf{f}^{em})\cdot\mathbf{U}+\varrho\mathscr{E}\cdot\dot{\boldsymbol{\pi}}+\varrho h]\,dv + \int_{\partial\mathscr{P}_t}[(\mathbf{T}+\mathbf{T}^{em})\cdot\mathbf{U}-\tilde{\mathbf{q}}\cdot\mathbf{n}]\,da, \tag{3.10}$$

which is but an equivalent statement for the first principle of thermodynamics.

4. Local field equations

Of course, Eq. (3.1) yields the local form

$$\frac{\partial\varrho}{\partial t}+\nabla\cdot(\varrho\mathbf{U}) = 0 \quad \text{or} \quad \dot{\varrho}+\varrho\nabla\cdot\mathbf{U} = 0 \quad \text{in} \quad \mathscr{D}_t. \tag{4.1}$$

Using Cauchy's principle for stresses and the usual tetrahedron argument, Eq. (3.2) enables one to show that, on $\partial\mathscr{D}_t$, $\mathbf{T}+\mathbf{T}^{em}$ is linear in $\mathbf{n}$. That is, there exists a Cauchy stress tensor t_{ij} (a general second-order tensor) such that

$$t_{ij}n_j = T_i + T_i^{em} \quad \text{on} \quad \partial\mathscr{D}_t. \tag{4.2}$$

On account of this and Eq. (4.1), Eq. (3.2) yields in the usual manner the local balance law of linear momentum:

$$\varrho \dot{U}_i = t_{ij,j} + f_i + f_i^{\mathrm{em}} \quad \text{in} \quad \mathscr{D}_t. \tag{4.3}$$

According to the comments made in Sect. 2, the couple $\mathbf{c}$ appearing in Eq. (3.4) has the expression

$$\mathbf{c} = \varrho \sum_{\alpha} c_\alpha {}^{L}\mathbf{E}_\alpha \times \boldsymbol{\pi}_\alpha. \tag{4.4}$$

Then, on account of Eqs. (4.2), (4.3) and (4.4), Eq. (3.4) yields the expression of the local balance of angular momentum for the material lattice in the form

$$t_{[ij]} = t_{[ij]}^{INT}, \tag{4.5}$$

where

$$t_{ij}^{INT} = \varrho \sum_{\alpha} c_\alpha^L E_{\alpha i} \pi_{\alpha j} \tag{4.6}$$

is the "interaction" tensor introduced in Part One.

We do not have to use the tetrahedron argument for Eq. (3.3) since there is no surface contribution[3]. On account of Eq. $(3.8)_{2-3}$ and I-$(3.5)_2$, Eqs. (3.3) yield the local equations

$$d_\alpha \ddot{\boldsymbol{\pi}}_\alpha = \mathscr{E} + {}^{L}\mathbf{E}_\alpha \quad (\alpha = 1, 2, \ldots, n) \quad \text{in} \quad \mathscr{D}_t, \tag{4.7}$$

whereas Eqs. (3.5) yield

$$\varrho_\alpha d_\alpha \boldsymbol{\pi}_\alpha \times \ddot{\boldsymbol{\pi}}_\alpha = \varrho_\alpha \boldsymbol{\pi}_\alpha \times \mathscr{E} + \varrho_\alpha \boldsymbol{\pi}_\alpha \times {}^{L}\mathbf{E}_\alpha, \tag{4.8}$$

which are trivially satisfied according to Eqs. (4.7). However, summing Eqs. (4.8) over α, writing the resulting equation in terms of skewsymmetric tensors, using Eqs. (4.5), (4.6) and (2.1), and defining the intrinsic spin angular momentum $S_{ij}^{(p)}$ due to polarization inertia and the ponderomotive couple C_{ij}^{em} by

$$S_{ij}^{(p)} = \sum_{\alpha} c_\alpha d_\alpha \dot{\pi}_{\alpha[i} \pi_{\alpha j]} = -S_{ji}^{(p)} \tag{4.9}$$

and

$$C_{ij}^{\mathrm{em}} \equiv P_{[i} \mathscr{E}_{j]} = -C_{ji}^{\mathrm{em}}, \tag{4.10}$$

we obtain the unique equation

$$\varrho \dot{S}_{ij}^{(p)} = t_{[ij]} - C_{ij}^{\mathrm{em}}. \tag{4.11}$$

This is the local statement of balance of angular momentum for the combined material-polarization continuum, whereas Eqs. (4.5) and (4.8) represented the balance laws of angular momentum for the material lattice and each separate polarization sub-lattice, respectively.

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[3] However, as it was remarked in Part II, the consideration of such a term (a surface density of polarization) would allow us, after using Cauchy's principle and the tetrahedron argument, to introduce in the resulting Eq. (4.7) terms having the form of the divergence of second-order tensors. These tensors would account for the spatial disuniformities in each polarization field, and thus for the ordering effects observed in ferroelectrics and antiferroelectrics below their critical temperature (compare [8]).

Finally, let us find the local form of Eq. (3.10). Taking the scalar product of Eq. (4.3) with **U**, the scalar product of each of Eqs. (4.7) with the corresponding $\boldsymbol{\pi}_\alpha$, summing the equations resulting from the latter over α, taking account of Eqs. (2.1) and $(3.8)_1$, substituting from these in Eq. (3.10) while accounting for the boundary condition (4.2), and finally localizing the resulting global equation, one arrives at the local statement of the energy equation in the form:

$$\varrho\dot{e} = t_{ij}U_{i,j} - \varrho\sum_\alpha c_\alpha {}^L\mathbf{E}_\alpha \cdot \dot{\boldsymbol{\pi}}_\alpha - \nabla\cdot\tilde{\mathbf{q}} + \varrho h \quad \text{in} \quad \mathscr{D}_t. \tag{4.12}$$

Now call σ_{ij} the symmetric part of the Cauchy stress tensor, and D_{ij} and Ω_{ij}, respectively, the rate-of-deformation and vorticity tensors, in such a way that

$$D_{ij} \equiv U_{(i,j)}, \quad \Omega_{ij} \equiv U_{[i,j]}. \tag{4.13}$$

Then, according to Eq. (4.5) and Eqs. (4.13), we have

$$t_{ij} = \sigma_{ij} + t^{INT}_{[ij]} \tag{4.14}$$

and

$$t_{ij}U_{i,j} = \sigma_{ij}D_{ij} + \varrho\sum_\alpha c_\alpha {}^L E_{\alpha i}\pi_{\alpha j}\Omega_{ij}. \tag{4.15}$$

Substituting from the latter result in Eq. (4.12), we finally obtain

$$\varrho\dot{e} = \sigma_{ij}D_{ij} - \varrho\sum_\alpha c_\alpha {}^L\mathbf{E}_\alpha \cdot \hat{\boldsymbol{\Pi}}_\alpha - \nabla\cdot\tilde{\mathbf{q}} + \varrho h, \tag{4.16}$$

where

$$(\hat{\boldsymbol{\Pi}}_\alpha)_i \equiv \dot{\pi}_{\alpha i} - \Omega_{ij}\pi_{\alpha j} \tag{4.17}$$

defines the co-rotational Jaumann derivative of $\boldsymbol{\pi}_\alpha$.

We need not look for the local expression of the inequality (3.7), for the initial global expression is the same as that postulated in Part Two and the agreement of all previously obtained local equations with those obtained in Parts One and Two is already complete. Indeed, Eqs. (4.2), (4.3), (4.5), (4.6), (4.7), (4.11), (4.14) and (4.16) are none other than Eqs. (I-4.10), (I-4.9), (I-4.14$_2$), (I-4.3), (I-4.11), (I-4.25), (I-4.12) and (II-3.9), respectively, QED.

5. A special case

In conclusion let us briefly consider a special case. Namely, that of a deformable dielectric with an overall description accounting only for the total volume polarization and neglecting polarization inertia. Obviously, all the above results can be specialized to agree with these hypotheses. However, one can also start with modified balance laws (3.2) through (3.6). If we retain Eq. (3.3), without inertia, then we shall get ${}^L\mathbf{E} = -\mathscr{E}$ instead of Eqs. (4.7), and Eq. (4.8) will be replaced by the equation

$$\mathbf{c} = \mathbf{c}^{em} \equiv \mathbf{P}\times\mathscr{E}, \tag{5.1}$$

as is readily checked. That is, we need not introduce the phenomenological field ${}^{L}E$, and the material lattice is directly subjected to the ponderomotive couple. Eq. (4.5) is replaced by

$$t_{[ij]} = P_{[i}\mathscr{E}_{j]}, \tag{5.2}$$

whereas Eq. (4.12) takes on the form

$$\varrho\dot{e} = t_{ij}U_{i,j} + \varrho\mathscr{E}\cdot\dot{\boldsymbol{\pi}} - \nabla\cdot\tilde{\mathbf{q}} + \varrho h. \tag{5.3}$$

Defining now the symmetric stress tensor ${}^{L}t_{ij}$ by

$${}^{E}t_{ij} \equiv t_{(ij)} + \mathscr{E}_{(i}P_{j)} \tag{5.4}$$

and noting that, according to Eqs. (5.2) and (5.4),

$$t_{ij}U_{i,j} = t_{(ij)}D_{ij} + P_{[i}\mathscr{E}_{j]}\Omega_{ij} = {}^{E}t_{ij}D_{ij} - \mathscr{E}_{i}P_{j}U_{i,j}, \tag{5.5}$$

and that

$$\begin{aligned} \varrho\dot{\boldsymbol{\pi}} &\equiv \dot{\mathbf{P}} + \mathbf{P}(\nabla\cdot\mathbf{U}), \\ \overset{*}{\mathbf{P}} &\equiv \dot{\mathbf{P}} - (\mathbf{P}\cdot\nabla)\mathbf{U} + \mathbf{P}(\nabla\cdot\mathbf{U}), \end{aligned} \tag{5.6}$$

Eq. (5.3) transforms to

$$\varrho\dot{e} = {}^{E}t_{ij}D_{ij} + \mathscr{E}\cdot\overset{*}{\mathbf{P}} - \nabla\cdot\tilde{\mathbf{q}} + \varrho h. \tag{5.7}$$

This is the local energy equation to be considered in order to deduce Toupin's theory of dielectrics [9]. The corresponding transformation of Cauchy's equation was given in Part Two.

Given the rather simple features of the above described model, one may wonder why we took the pains to develop a more formal and somewhat cumbersome energy approach in Part One. The reason is that the latter approach is easily extended to much more complicated situations while the direct approach through balance laws becomes rapidly unmanageable as the model becomes more and more complicated. Such a situation is the following one. Let us imagine that one envisages the study of wave propagation in a thin layer of ferroelectric elastic material (this would be a typical electronic component). Then a characteristically small length is involved and the usual elastic description reveals itself unsufficient to describe this size effect. A finer mechanical description must be considered. Also, one has probably to consider the effects of polarization gradients to account for the ferroelectric ordering. Then the virtual power method used in Part One will yield, in a straightforward and elegant manner, all field equations and associated boundary conditions — the latter are known to be quite involved (compare the ferromagnetic case [10]).

Note added in proof. While lecturing at the Bell Telephone Laboratories, Murray Hill, N.J., (USA) in may 1976, I was kindly handed by Dr. D. F. Nelson and Prof. M. Lax reprints of their most recent papers [11]—[12]. It appears that these authors, by using a Lagrangian variational principle and a semi-microscopical model, arrived at equations quite similar to ours in the precise case of purely elastic pyroelectrics. In particular, their internal-motion equations, which describe the evolution of internal coordinates in a primitive-unit cell in the long-wavelength approximation—Eq. (6.2) in Ref. [11]—is the same as our equation (I-4.15) in the case of hyperelastic dielectrics. They

have no general statement of the second principle of thermodynamics at their disposal to enable them to study dissipative processes (compare Part Two). As was pointed out in Part One, this is but one shortcoming of the Lagrangian approach.

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