Deformable dielectrics
I. Field equations for a dielectric
made of several molecular species

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USING d'Alembert's principle along with the objectivity requirement as starting point, the first part of this work develops in a rational manner both local and global field equations for the dynamical (continuum) theory of non-magnetized deformable dielectrics (ferroelectrics, *etc.*) made of several molecular species. The continuum is considered to be the assembly of a material lattice (substrate of deformations) and *n* polarization sub-lattices that account for dielectric effects. The decomposition obtained for the Cauchy stress allows a discussion and a comparison of various theories of deformable dielectrics without restriction on the thermodynamical processes.

Biorąc jako punkt wyjścia zasadę d'Alemberta oraz żądanie obiektywności, w pierwszej części pracy wyprowadzono w sposób racjonalny zarówno lokalne jak i globalne równania pola dynamicznej (kontynualnej) teorii nie magnetyzowanych odkształcalnych dielektryków (ferroelektryków itp.) wykonanych z wielu rodzai molekuł. Kontynuum rozpatruje się tu jako zbiór sieci materialnej (podstawa deformacji) i *n* spolaryzowanych podsieci odpowiadających za efekty dielektryczne. Otrzymana dekompozycja tensora Cauchy'ego pozwala na dyskusję i porównanie różnych teorii odkształcalnych dielektryków bez ograniczeń nakładanych na procesy termodynamiczne.

Принимая за исходную точку принцип Даламберта, а также требование объективности, в первой части работы выведены рациональным образом так локальные, как и глобальные уравнения поля динамической (континуальной) теории немагнитных деформируемых диэлектриков (ферроэлектриков и т. п.) состоящих из многих типов молекул. Континуум рассматривается здесь как совокупность материальной решетки (основа деформации) и *n* поляризованных подрешеток ответственных за диэлектрические эффекты. Полученное видоизменение тензора Коши позволяет обсудить и сравнить разные теории деформируемых диэлектриков без ограничений накладываемых на термодинамические процессы.

1. Introduction

THIS work grew out of the author's desire to understand the relationship that may exist between the various existing dynamical theories of non-linear elastic dielectrics (mainly the works of Toupin, Dixon and Eringen, Tiersten) and linear elastic dielectrics (Voigt's theory of piezoelectricity). Having brought these seemingly different theories within a common frame, the second aim was to try to give a more comprehensive physical description as far dielectric effects are concerned, so that the different states of dielectricity could be accounted for. To fulfill this purpose we consider herebelow that the dielectric is built up of different co-existing molecular species, each of these giving rise to an electric dipole moment, the sum of which yields the only physically observable field, the total volume polarization. For the sake of simplicity, the dielectric is assumed to be nonmagnetizable and no chemical

reactions are assumed to occur between the molecular species. Although we do not account for spatial disuniformities in the polarization sub-lattices, this multisub-lattice polarization model reproduces the ferroelectric, antiferroelectric and ferrielectric states.

The method followed in this part (compare Part III) to construct the field equations is that favoured by P. GERMAIN [6] and the author and co-workers (e.g. [1, 2]) in previous works. That is, all field equations, except Maxwell's equations, are deduced from d'Alembert's principle in which generalized internal forces which account for the interactions are formally introduced as co-factors of objective rates (for more detail see Paragraph 4.1). The form of the contributions of Maxwellian fields, especially the ponderomotive force and couple, are assumed. The intramolecular-force balance equation that governs each polarization sub-lattice is thus deduced on an equal foot with the Cauchy equations of motion. A typical decomposition of the Cauchy stress tensor is obtained in which it is shown that the interactions with the polarization sublattices participate. It is by playing with this decomposition and shifting terms either from or to the electromagnetic stress tensor (a nonuniquely defined quantity) that the relationship with other models of deformable dielectrics is exhibited.

Part two will develop the relevant thermodynamics and the constitutive equations for both the non-linear and linear thermoelastic materials.

2. Results from electrodynamics(^{1})

Let $\mathbf{E}$, $\mathbf{B}$, $\mathbf{D}$, $\mathbf{H}$, $\mathbf{P}$ and $\mathbf{M}$ be the electromagnetic fields of classical Maxwell's electromagnetism expressed in a *fixed* Galilean inertial frame $\mathcal{R}_{IG}$. Then, in Lorentz-Heaviside units,

(2.1) $$\mathbf{D} = \mathbf{E} + \mathbf{P}, \quad \mathbf{H} = \mathbf{B} - \mathbf{M}.$$

Let $\mathscr{E}$, $\mathscr{H}$ and $\mathscr{M}$ be respectively the electromotive intensity — i.e., the electric field expressed in a co-moving frame $\mathcal{R}_C(\mathbf{x}, t)$ —, the magnetic field in $\mathcal{R}_C(\mathbf{x}, t)$, and the volume magnetization in $\mathcal{R}_C(\mathbf{x}, t)$. Finally let q_f, $\mathbf{J}$ and $\mathscr{J}$ be the free-charge density, the total current (i.e. the electric current expressed in $\mathcal{R}_{IG}$), and the conduction current (i.e. the electric current expressed in $\mathcal{R}_C(\mathbf{x}, t)$, respectively. Then, up to terms of the order of $\beta^2 = (|\mathbf{U}|/c)^2$, where $\mathbf{U}$ is the matter velocity and c is the velocity of light in vacuum, one has the following relationships:

(2.2) $$\mathscr{E} = \mathbf{E} + \frac{1}{c}\mathbf{U} \times \mathbf{B},$$

(2.3) $$\mathscr{H} = \mathbf{H} - \frac{1}{c}\mathbf{U} \times \mathbf{D},$$

(2.4) $$\mathscr{M} = \mathbf{M} + \frac{1}{c}\mathbf{U} \times \mathbf{P},$$

(2.5) $$\mathbf{J} = q_f\mathbf{U} + \mathscr{J}.$$

(^{1}) As a rule we use the standard Cartesian-tensor notation in rectangular coordinate frames. The divergence of second-order tensors is taken on the last index, e.g. $(\operatorname{div}\mathbf{t})_i = t_{ij,j}$.

For the sake of simplicity, we shall consider the case of *nonmagnetizable* dielectrics, for which the magnetization vanishes identically in the co-moving frame. Then, Eq. (2.4) yields

$$(2.6) \qquad \mathcal{M} \equiv 0.$$

Furthermore, in a dielectric insulator

$$(2.7) \qquad q_f \equiv 0, \quad \mathcal{J} \equiv 0.$$

On account of these assumptions Eqs. (2.1), (2.3) and (2.5) yield

$$(2.8) \qquad \mathbf{H} = \mathbf{B} - \frac{1}{c}\mathbf{P} \times \mathbf{U},$$

$$(2.9) \qquad \mathcal{H} = \mathbf{B} - \frac{1}{c}\mathbf{U} \times \mathbf{E},$$

$$(2.10) \qquad \mathbf{J} \equiv 0.$$

Then, results set forth or established in previous papers of ours for the general electro-dynamic case [1]–[2] specialize to the following ones:

$$(2.11) \qquad \mathbf{f}^{em} = \frac{1}{c}\overset{*}{\mathbf{P}} \times \mathbf{B} + (\mathbf{P} \cdot \nabla)\mathcal{E},$$

$$(2.12) \qquad \mathbf{c}^{em} = \mathbf{P} \times \mathcal{E}$$

and

$$(2.13) \qquad \mathbf{f}^{em} \equiv \operatorname{div} \mathbf{t}^{em \cdot p} - \frac{\partial \mathbf{G}}{\partial t},$$

$$(2.14) \qquad C_{ij}^{em} \equiv -t_{[ij]}^{em \cdot p} = -\mathcal{E}_{[i}P_{j]} = -C_{ji}^{em},$$

where

$$(2.15) \qquad t_{ij}^{em \cdot p} \equiv \mathcal{E}_i P_j + t_{ij}^{em \cdot f},$$

$$(2.16) \qquad t_{ij}^{em \cdot f} \equiv E_i E_j + B_i B_j - \frac{1}{2}(E^2 + B^2)\delta_{ij},$$

$$(2.17) \qquad \mathbf{G} \equiv \frac{1}{c}\mathbf{E} \times \mathbf{B},$$

$$C_{ij}^{em} \equiv \frac{1}{2}\varepsilon_{ijk}c_k^{em}, \quad c_k^{em} = \varepsilon_{klj}C_{ij}^{em}.$$

Here, $\mathbf{f}^{em}$ is the reduced form of the volume ponderomotive force (for non-magnetizable dielectrics, and discarding electric quadrupoles) as computed by DIXON and ERINGEN [3] from a microscopic model (essentially the Lorentz theory of electrons). $\mathbf{c}^{em}$ is the corresponding volume ponderomotive couple. Equations (2.13) and (2.14) are mere *identities*, which allow us to introduce in a *non-unique* manner the electromagnetic stress tensor $t_{ij}^{em \cdot p}$ and the electromagnetic-field momentum $\mathbf{G}$. $t_{ij}^{em \cdot f}$ is none other than Maxwell's stress

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tensor for *free* electromagnetic fields. Finally, $\overset{*}{\mathbf{P}}$ denotes the convected-time derivative of **P** in such a way that

$$(2.18) \qquad \overset{*}{\mathbf{P}} \equiv \dot{\mathbf{P}} - (\mathbf{P} \cdot \nabla)\mathbf{U} + \mathbf{P}(\nabla \cdot \mathbf{U}) \equiv \frac{\partial \mathbf{P}}{\partial t} + \nabla \times (\mathbf{P} \times \mathbf{U}) + \mathbf{U}(\nabla \cdot \mathbf{P}),$$

where the superimposed dot indicates the usual material time derivative.

Remark that if one defines a new ponderomotive force (as considered by TOUPIN [4]) by

$$(2.19) \qquad \bar{\mathbf{f}}^{em} = -(\nabla \cdot \mathbf{P})\mathscr{E} + \frac{1}{c}\overset{*}{\mathbf{P}} \times \mathbf{B},$$

then, on account of Eqs. (2.15), (2.16) and (2.19), the identity (2.13) transforms to

$$(2.20) \qquad \bar{\mathbf{f}}^{em} = \operatorname{div} \mathbf{t}^{\,em\cdot f} - \frac{\partial \mathbf{G}}{\partial t}.$$

Upon comparing Eqs. (2.13) and (2.20), we obtain the following identity:

$$(2.21) \qquad \mathbf{f}^{em} - \operatorname{div} \mathbf{t}^{em\cdot p} \equiv \bar{\mathbf{f}}^{em} - \operatorname{div} \mathbf{t}^{em\cdot f},$$

that proves useful in comparing various theories of dielectrics.

The global energetic *identity* — for a regular spatial domain $\mathscr{D}_t$ of E^3 bounded by the surface $\partial \mathscr{D}_t$ of unit outward normal **n** at time t — obtained by us [2] reduces to the following one:

$$(2.22) \qquad \dot{U}^{em}(\mathscr{D}_t) = -\int_{\mathscr{D}_t} (\mathbf{f}^{em} \cdot \mathbf{U} + \varrho \mathscr{E} \cdot \dot{\boldsymbol{\pi}}) dv - \int_{\partial \mathscr{D}_t} (\mathbf{T}^{em} \cdot \mathbf{U} + \mathscr{J} \cdot \mathbf{n}) da,$$

in which

$$(2.23) \qquad U^{em}(\mathscr{D}_t) \equiv \int_{\mathscr{D}_t} \frac{1}{2}(\mathbf{E}^2 + \mathbf{B}^2) dv,$$

$$(2.24) \qquad T_i^{em} \equiv -[t_{ij}^{em\cdot p}]n_j,$$

$$(2.25) \qquad \mathscr{J} \equiv c\mathscr{E} \times \mathscr{H},$$

$$(2.26) \qquad \boldsymbol{\pi} \equiv \mathbf{P}/\varrho,$$

ϱ is the matter density at time t. T^{em} is the surface ponderomotive force, the familiar symbolism [...] indicating the jump across $\partial \mathscr{D}_t$. $\mathscr{J}$ is the Poynting vector expressed in $\mathscr{R}_c(\mathbf{x}, t)$. $\mathbf{f}^{em}$ and $t_{ij}^{em\cdot p}$ have the expressions (2.11) and (2.15), respectively. Alternate forms of Eq. (2.22) are

$$(2.27) \qquad \frac{d}{dt}\int_{\mathscr{D}_t} \frac{1}{2}(\mathbf{E}^2 + \mathbf{B}^2 + 2\mathscr{E} \cdot \mathbf{P}) dv = -\int_{\mathscr{D}_t} (\mathbf{f}^{em} \cdot \mathbf{U} + \mathbf{P} \cdot \dot{\mathscr{E}}) dv - \int_{\partial \mathscr{D}_t} (\mathbf{T}^{em} \cdot \mathbf{U} + \mathscr{J} \cdot \mathbf{n}) da,$$

and

$$(2.28) \qquad \dot{U}^{em}(\mathscr{D}_t) = \int_{\mathscr{D}_t} \left(\frac{\partial \mathbf{G}}{\partial t} \cdot \mathbf{U} - \varrho \mathscr{E} \cdot \dot{\boldsymbol{\pi}} + t_{ij}^{em\cdot p} U_{i,j} \right) dv - \int_{\partial \mathscr{D}_t} \mathscr{J} \cdot \mathbf{n} \, dz.$$

These are obtained by straightforward transformations using Eq. (2.13) and a transport theorem.

Maxwell's equations within the spatial region $\mathcal{D}_t$ read in $\mathcal{R}_{IG}$:

(2.29)
$$\nabla \times \mathbf{E} + \frac{1}{c}\frac{\partial \mathbf{B}}{\partial t} = 0, \quad \nabla \cdot \mathbf{B} = 0,$$
$$\nabla \times \mathbf{H} - \frac{1}{c}\frac{\partial \mathbf{D}}{\partial t} = 0, \quad \nabla \cdot \mathbf{D} = 0,$$

on account of Eqs. $(2.7)_1$ and (2.10). On account of Eqs. $(2.1)_1$ and (2.8) and of the second expression for $\overset{*}{\mathbf{P}}$ given in Eq. (2.18), the last two of Eqs. (2.29) can also be rewritten as

(2.30)
$$\nabla \times \mathbf{B} - \frac{1}{c}\frac{\partial \mathbf{E}}{\partial t} = \frac{1}{c}[\overset{*}{\mathbf{P}} - (\nabla \cdot \mathbf{P})\mathbf{U}], \quad \nabla \cdot \mathbf{E} = -\nabla \cdot \mathbf{P},$$

which, in agreement with Eq. (2.19), exhibit the role of induced charge density, induced convection current and induced conduction current played by $-\nabla \cdot \mathbf{P}$, $-(\nabla \cdot \mathbf{P})\mathbf{U}$ and $\overset{*}{\mathbf{P}}$, respectively. In these conditions the ponderomotive force (2.19) assumes exactly Lorentz's form.

3. The dielectric description

For the sake of simplicity we consider the following somewhat simplified description — although it is more involved than any other one heretofore considered in the non-linear theory of dielectrics. From the electrical viewpoint the dielectric is viewed as being the assembly of n co-existing molecular species[2] $\alpha = 1, 2, ..., n$. Each molecular species gives rise to a field of electric dipoles which, when suitably averaged, is represented by a volume density $\mathbf{P}'_\alpha$ of electrical polarization. The total polarization is thus (in the fixed Galilean frame $\mathcal{R}_{IG}$)

(3.1)
$$\mathbf{P} = \sum_\alpha \mathbf{P}_\alpha.$$

Let ϱ_α be the density of α molecules and c_α the corresponding concentration, i.e.,

(3.2)
$$c_\alpha \equiv \varrho_\alpha/\varrho.$$

Then we have

(3.3)
$$\boldsymbol{\pi}_\alpha \equiv \mathbf{P}_\alpha/\varrho_\alpha, \quad \mathbf{P} = \varrho \sum_\alpha c_\alpha \boldsymbol{\pi}_\alpha.$$

On the other hand, if e, Y_α, n_α and $\mathbf{W}_\alpha$ denote respectively the electronic charge, the dipole charge per α-molecule expressed in electronic units, the number of α-molecules per unit volume, and the displacement field which gives rise to electrical polarization within the α-molecule, then

(3.4)
$$\mathbf{P}_\alpha = \varrho_\alpha \boldsymbol{\pi}_\alpha = q_\alpha \mathbf{W}_\alpha, \quad q_\alpha \equiv eY_\alpha n_\alpha.$$

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[2] There is no necessary one-to-one correspondence between the present schematization and a microscopic model at the molecular level.

It is assumed that (i) no chemical reaction occurs between the different molecular species, so that there is no mass transfer; (ii) mechanically, the assembly of molecules behaves like a single continuum, the volumetric behavior being that of the classical continuum (e.g., the substratum of elastic deformations), so that there is no diffusion; (iii) the total continuum is everywhere neutral (dielectric). On account of these assumptions we have, with Eq. (3.2) and the conservation of charge equation:

$$(3.5) \qquad \dot{c}_\alpha = 0, \quad \frac{d}{dt}\left(\frac{\varrho}{q_\alpha}\right) = 0 = \frac{d}{dt}\left(\frac{\varrho_\alpha}{q_\alpha}\right), \quad \nabla \cdot \mathbf{w}_\alpha = 0,$$

of which the latter is a generalization of the condition set forth by TIERSTEN [5], but is in no way essential.

The natural time rate of π_α^{-1} is $\dot{\pi}_\alpha$. However, an *objective* time rate is obtained by considering the Jaumann co-rotational derivative:

$$(3.6) \qquad (\hat{\mathbf{\Pi}}_\alpha)_i \equiv \dot{\pi}_{\alpha i} - \Omega_{ij}\pi_{\alpha j}, \quad \Omega_{ij} \equiv U_{[i,j]}.$$

If necessary, one can also introduce the objective convected-time derivative $\overset{*}{\mathbf{P}}_\alpha$ defined as in Eq. (2.18) but for each $\mathbf{P}_\alpha$. Then it is a simple matter to show that $\overset{*}{\mathbf{P}}_\alpha$ and $\hat{\mathbf{\Pi}}_\alpha$ are related by the equation

$$(3.7) \qquad \varrho_\alpha \hat{\mathbf{\Pi}}_{\alpha i} = \overset{*}{\mathbf{P}}_{\alpha i} + \varrho_\alpha D_{ij}\pi_{\alpha j}, \quad D_{ij} \equiv U_{(i,j)}.$$

4. Field equations

4.1. The principle of virtual power

We proceed to deduce both local and global equations that govern the motion of the dielectric and the polarization fields of the molecular species from a unique principle, the virtual power principle in the form used by P. GERMAIN [6] and the author [1, 7, 8]. We do not repeat here in detail its justification and the manner in which the different virtual powers are constructed. All that need be recalled is that, in order to guarantee the *objectivity* (or material frame indifference) of the generalized internal forces for which one ultimately needs to construct constitutive equations, the virtual power of *internal* forces ought to be written as a linear functional on a set of adequate *objective* time rates. In the present case where the description is based on the classical motion and the different molecular species, the fundamental generalized velocity field is $v = \{\mathbf{U}, \dot{\pi}_\alpha; \alpha = 1, 2, ..., n\}$, whereas the objective set of generalized velocities needed to describe non-linear elastic dielectrics is

$$(4.1) \qquad \mathscr{V}_{\text{obj}} = \{D_{ij}, \hat{\mathbf{\Pi}}_\alpha; \alpha = 1, 2, ..., n\}$$

or, for instance, since such a set is not unique

$$(4.2) \qquad \overline{\mathscr{V}}_{\text{obj}} = \{D_{ij}, \overset{*}{\mathbf{P}}_\alpha; \alpha = 1, 2, ..., n\}.$$

Following previous works [7, 8], we shall use the set (4.1) to start with. Remark that there is no such requirement of objectivity as regards the volume forces, the surface or contact forces, and the inertia forces, which are therefore introduced as co-factors in linear functionals on the sets $\mathscr{V} = \{\mathbf{U}, \nabla\mathbf{U}, \dot{\boldsymbol{\pi}}_\alpha; \alpha = 1, 2, ..., n\}$, and v, respectively. Proceeding as in [7 and 8], we thus set the expressions of the virtual power of internal, volume and contact forces, respectively, $\mathscr{P}_{(i)}$, $\mathscr{P}_{(v)}$ and $\mathscr{P}_{(c)}$, for the whole region $\mathscr{D}_t$ bounded by $\partial\mathscr{D}_t$, as

$$(4.3) \qquad \mathscr{P}_{(i)}^*(\mathscr{D}_t) = -\int_{\mathscr{D}_t} \left(\sigma_{ij} D_{ij}^* - \varrho \sum_\alpha c_\alpha \, {}^L\mathbf{E}_\alpha \cdot \hat{\boldsymbol{\Pi}}_\alpha^*\right) dv,$$

$$(4.4) \qquad \mathscr{P}_{(v)}^*(\mathscr{D}_t) = \int_{\mathscr{D}_t} \left(\mathbf{f} \cdot \mathbf{U}^* - t_{ij}^{em \cdot p} U_{i,j}^* + \sum_\alpha \varrho_\alpha \mathscr{E} \cdot \dot{\boldsymbol{\pi}}_\alpha^*\right) dv,$$

$$(4.5) \qquad \mathscr{P}_{(c)}^*(\partial\mathscr{D}_t) = \int_{\partial\mathscr{D}_t} (\mathbf{T} \cdot \mathbf{U}^*) da.$$

Here an asterisk indicates a *virtual* generalized velocity field or the value of an expression in such a field. Signs are chosen, and ϱ_α is introduced, for convenience. In Eq. (4.3) the generalized internal forces are introduced via the duality inherent in the method. $\sigma_{ij} = \sigma_{ji}$ is called the *intrinsic* stress tensor, whereas the fields ${}^L\mathbf{E}_\alpha$, $\alpha = 1, 2, ..., n$, whose dimension is that of an electric field, will be referred to as the *local* electric fields. In Eq. (4.4), $\mathbf{f}$ is a volume body force of non-electromagnetic origin (e.g., gravity), $t_{ij}^{em \cdot p}$ is the electromagnetic stress tensor defined by Eq. (2.15), and $\mathscr{E}$ is given by Eq. (2.2). Finally, $\mathbf{T}$ is a purely *mechanical* surface traction (either a prescribed quantity or an unknown to be determined by solving a well-posed problem). We have anticipated the forthcoming results in discarding any polarization traction on $\partial\mathscr{D}_t$.

To express the virtual power of inertia forces one must remark that three kinds of inertia can be distinguished: the classical inertia $\varrho\dot{\mathbf{U}}$, the inertia of the electromagnetic fields which, since we chose to use the tensor (2.15), is none other than $\partial\mathbf{G}/\partial t$, and the inertia possibly associated with the internal deformation of the molecules. A simple expression for the latter is derived in the appendix. Consequently, the virtual power of inertial forces for the whole body is written down in the form

$$(4.6) \qquad \mathscr{P}_{(a)}^*(\mathscr{D}_t) = \int_{\mathscr{D}_t} \varrho\left[\left(\dot{\mathbf{U}} + \varrho^{-1}\frac{\partial\mathbf{G}}{\partial t}\right) \cdot \mathbf{U}^* + \sum_\alpha c_\alpha d_\alpha \ddot{\boldsymbol{\pi}}_\alpha \cdot \dot{\boldsymbol{\pi}}_\alpha^*\right] dv,$$

where (see the appendix)

$$(4.7) \qquad d_\alpha \equiv \left(\frac{\varrho_\alpha}{\varrho_\alpha}\right)^2 = \left(\frac{m_\alpha}{eY_\alpha}\right)^2.$$

Then, for an absolute Newtonian chronology and in a Galilean inertial frame, the total virtual power of inertial forces is balanced by that of all other types of forces, internal or external (d'Alembert's principle). That is,

$$(4.8) \qquad \mathscr{P}_{(a)}^*(\mathscr{D}_t) = \mathscr{P}_{(i)}^*(\mathscr{D}_t) + \mathscr{P}_{(v)}^*(\mathscr{D}_t) + \mathscr{P}_{(c)}^*(\partial\mathscr{D}_t)$$

at all times and for arbitrary virtual velocity fields.

4.2. Local field equations

The expression (4.8) is posited to hold true for arbitrary virtual velocity fields U^* and $\dot{\boldsymbol{\pi}}_\alpha^*$ at all points within $\mathcal{D}_t$ and on $\partial\mathcal{D}_t$, provided these manifolds are sufficiently regular. On account of Eqs. (2.13), (2.24) and (3.6), we are led to the following results:

THEOREM. *The local field equations that govern the motion and the molecular polarizations in the dynamical theory of deformbale non-magnetizable dielectrics made of several molecular species are*

$$(4.9) \qquad \varrho\dot{U} = \operatorname{div}\mathbf{t}+\mathbf{f}+\frac{1}{c}\overset{*}{\mathbf{P}}\times\mathbf{B}+(\mathbf{P}\cdot\boldsymbol{\nabla})\mathscr{E} \quad \text{in } \mathcal{D}_t,$$

$$(4.10) \qquad T_i+T_i^{em} = t_{ij}\,n_j \quad \text{on } \partial\mathcal{D}_t,$$

$$(4.11) \qquad \mathscr{E}+{}^L\mathbf{E}_\alpha = d_\alpha\ddot{\boldsymbol{\pi}}_\alpha \quad \text{in } \mathcal{D}_t, \quad \alpha = 1, 2, \ldots, n,$$

with

$$(4.12) \qquad t_{ij} \equiv \sigma_{ij}+t_{[ij]}^{INT},$$

$$(4.13) \qquad t_{ij}^{INT} \equiv \varrho\sum_\alpha c_\alpha\,{}^L E_{\alpha i}\pi_{\alpha j}.$$

Equation (4.9) is none other than the local statement of the Euler-Cauchy equation of motion. Equation (4.10) is the associated boundary condition. t_{ij} is the Cauchy stress tensor, which does not account for the electromagnetic forces. However, as is shown by Eq. (4.12), it includes a part, referred to as the *interaction*-stress tensor[3], which accounts for the presence of the different polarization fields. The interactions between these polarizations and the usual crystal lattice in fact are represented by the fields ${}^L\mathbf{E}_\alpha$. From Eq. (4.12) it follows at once that

$$(4.14) \qquad t_{(ij)} = \sigma_{ij}, \quad t_{[ij]} = t_{[ij]}^{INT}.$$

The second of these equations can be considered to be the local statement of the balance law of moment of momentum, since it defines the skew-symmetric part of the Cauchy stress. The fact that this statement here follows immediately is a clear consequence of the fact that expression (4.3) has been written while taking account of the objectivity requirement. Further comments on Eq. $(4.14)_2$ are made in Sect. 5.

Equation (4.11) resembles Newton's law of motion and can in fact be referred to as the motion equation for the "continuum" of α-molecules. Indeed, on account of d_α — the molecular polarization inertia — and of Eqs. $(3.5)_1$ and (3.4), Eq. (4.11) can be rewritten as

$$(4.15) \qquad q_\alpha(\mathscr{E}+{}^L\mathbf{E}_\alpha) = \varrho_\alpha\ddot{\mathbf{w}}_\alpha.$$

This is the equation of motion of a system of charge q_α, per unit volume, on which are acting both the electromotive intensity $\mathscr{E}$ due to external sources, and the local electric field ${}^L\mathbf{E}_\alpha$ due to the presence of the crystal lattice and the other molecular species. For historical reasons recalled in Part II, Eq. (4.15) or (4.11) is called the *intramolecular force balance* law for the α-component of the dielectric.

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[3] This notion of *interaction* in continua is dealt with by us in another paper [9].

To form the complete set of local field equations Eqs. (4.9)–(4.11) must be supplemented with Maxwell's equations (2.29) and the associated boundary conditions, the continuity equation

$$(4.16) \qquad \frac{\partial \varrho}{\partial t} + \mathbf{V} \cdot (\varrho \mathbf{U}) = 0 \quad \text{or} \quad \dot{\varrho} + \varrho D_{kk} = 0 \quad \text{in } \mathscr{D}_t,$$

and a local statement of the energy equation (which yields the heat conduction equation). The latter, as well as the constitutive theory for the fields σ_{ij} and $^L\mathbf{E}_\alpha$, will be discussed in Part Two.

4.3. Global field equations

A. *Balance of momentum.* Consider Eq. (4.8) and the following *virtual* velocity field:

$$(4.17) \qquad \mathbf{U}^*(\mathbf{x}, t) = \bar{\mathbf{U}}(t), \quad \mathbf{V}\bar{\mathbf{U}} = 0, \quad \dot{\pi}_\alpha^* = 0, \quad \forall \alpha,$$

throughout $\bar{\mathscr{D}}_t$ (= closure of $\mathscr{D}_t$). Then $\mathscr{P}_{(i)}^* (\mathscr{D}_t) \equiv 0$ since $D_{ij}^* \equiv \bar{U}_{(i,j)} \equiv 0$ and $\hat{\mathbf{\Pi}}_\alpha^* \equiv \mathbf{0}$ for all α. The resulting equation (4.8), valid for all spatially uniform fields $\bar{\mathbf{U}}$. yields after using Eq. (2.13) and a transport theorem

$$(4.18) \qquad \frac{d}{dt} \int_{\mathscr{D}_t} \varrho (\mathbf{U} + \mathbf{g}) dv = \int_{\mathscr{D}_t} \mathbf{f} dv + \int_{\partial \mathscr{D}_t} \tilde{\mathbf{T}} da,$$

where

$$(4.19) \qquad \mathbf{g} \equiv \mathbf{G}/\varrho, \quad \tilde{\mathbf{T}} = \mathbf{T} + \varrho \mathbf{g}(\mathbf{U} \cdot \mathbf{n}).$$

Equation (4.18) is the global balance law of momentum. Due to the fact that we introduced the momentum of the electromagnetic fields, $\mathbf{g}$, per unit mass in order that the left-hand side of Eq. (4.18) be written in the form of a total time derivative, the contact force had to be redefined in agreement with Eq. $(4.19)_2$. This in fact results from a redefinition of the electromagnetic stress tensor. Indeed, the following identity can be derived in lieu of Eq. (2.13) or (2.20). Using Eqs. $(4.19)_1$ and (4.16), one shows that

$$(4.20) \qquad \varrho \dot{\mathbf{g}} = \frac{\partial \mathbf{G}}{\partial t} + \text{div}(\mathbf{G} \otimes \mathbf{U}).$$

Bearing in mind that $\mathbf{f}^{em}$ is, from our viewpoint, the only meaningful quantity related to the average of Lorentz's theory of electrons, and that the notions of electromagnetic stress tensor, electromagnetic momentum, and surface ponderomotive force are only secondary notions introduced through identities, Eq. (2.13) can be rewritten as

$$(4.21) \qquad \varrho \dot{\mathbf{g}} = \text{div} \tilde{\mathbf{t}}^{em} - \mathbf{f}^{em}, \quad \tilde{\mathbf{t}}^{em} \equiv \mathbf{t}^{em \cdot p} + \mathbf{G} \otimes \mathbf{U}$$

on account of Eq. (4.20). It is then readily shown that, from Eqs. (4.18), (4.19) and (4.21), follow both Eqs. (4.9) and (4.10) if one applies the tetrahedron argument, so that $\tilde{T}_i = \tilde{t}_{ij} n_j$ on $\partial \mathscr{D}_t$ with $\tilde{t}_{ij} \equiv t_{ij} + t_{ij}^{em \cdot p} + G_i U_j$.

B. *Balance of moment of momentum*. Consider Eq. (4.8) and the following particular virtual velocity field:

(4.22)
$$U_i^*(\mathbf{x}, t) = \bar{\Omega}_{ij}(t)x_j, \quad \bar{\Omega}_{ij} = -\bar{\Omega}_{ji}, \quad \bar{\Omega}_{ij,k} = 0,$$
$$\dot{\pi}_{\alpha i}^*(\mathbf{x}, t) = \bar{\Omega}_{ij}(t)\pi_{\alpha j}, \quad \forall_\alpha,$$

throughout $\bar{\mathscr{D}}_t$. Then $\mathscr{P}_{(i)}^*(\mathscr{D}_t) = 0$ again is readily checked, whereas the remainder of Eq. (4.8) valid for all spatially uniform fields $\bar{\Omega}_{ij}$ is shown to yield the following global equation after some rearrangement:

(4.23) $$\frac{d}{dt}\int\limits_{\mathscr{D}_t} \{\varrho(U_{[i}+g_{[i})x_{j]}+\varrho S_{ij}^{(p)}\}\,dv = \int\limits_{\mathscr{D}_t} (f_{[i}x_{j]}+\varrho g_{[i}U_{j]})\,dv + \int\limits_{\partial\mathscr{D}_t} (\tilde{T}_{[i}x_{j]})\,da.$$

This is the global balance law of moment of momentum, in which

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

is the spin angular momentum tensor that results from the molecular-polarization inertia. On account of the local form of Eq. (4.18) it is easily shown that the local form of Eq. (4.23) reads thus

(4.25) $$t_{[ij]} - C_{ij}^{em} = \varrho\dot{S}_{ij}^{(p)},$$

where t_{ij} and c_{ij}^{em} are the fields defined by Eqs. (4.12) and (2.14), respectively. Here, there is no need to use the tetrahedron argument, since no surface couple participates in the statement (4.23). There is no contradiction between Eq. (4.25) and Eq. (4.14)$_2$, for the former can be deduced from the latter and Eqs. (4.11) in the following manner. For a given α take the tensor product of Eq. (4.11) with $\varrho_\alpha\pi_\alpha$, and then the skew-symmetric part of the resulting tensorial equation. One obtains by summing over α:

(4.26) $$\varrho\sum_\alpha c_\alpha{}^L E_{\alpha[i}\pi_{\alpha j]} - \mathscr{E}_{[i}\left(\sum_\alpha \varrho_\alpha\pi_{\alpha j]}\right) = \varrho\dot{S}_{ij}^{(p)}.$$

On account of Eqs. (3.2), (2.14), (4.13) and (4.12) this is nothing but Eq. (4.25), QED.

C. *Global balance laws governing the molecular polarizations*. Consider Eq. (4.8) and the following particular virtual velocity field([4])

(4.27)
$$\mathbf{U}^*(\mathbf{x}, t) = \bar{\mathbf{U}}(t), \quad \nabla\bar{\mathbf{U}} = \mathbf{0},$$
$$\dot{\boldsymbol{\pi}}_\alpha^*(\mathbf{x}, t) = \mathbf{A}_\alpha(t), \quad \nabla\mathbf{A}_\alpha = \mathbf{0},$$

throughout $\bar{\mathscr{D}}_t$, where one has selected a particular α so that all other $\dot{\boldsymbol{\pi}}_\beta^* = 0$ for $\beta \neq \alpha$. Then $\mathscr{P}_{(i)}^*$ is reduced to the form

(4.28) $$\mathscr{P}_{(i)}^*(\mathscr{D}_t) = \mathbf{A}_\alpha \cdot \int\limits_{\mathscr{D}_t} \varrho_\alpha{}^L \mathbf{E}_\alpha\,dv.$$

([4]) Of course, one can also select a field $\bar{\mathbf{U}}$ vanishing everywhere in $\bar{\mathscr{D}}_t$, so that the statement (4.8) becomes that of the „principle of virtual power" for a rigid body.

The first of Eqs. (4.27) again yields Eq. (4.18), whereas the remainder of Eq. (4.8) valid for arbitrary spatially uniform $\mathbf{A}_\alpha$ yields the global equation (α fixed):

(4.29)
$$\frac{d}{dt}\int_{\mathcal{D}_t} \varrho c_\alpha d_\alpha \dot{\pi}_\alpha \, dv = \int_{\mathcal{D}_t} \varrho_\alpha(\mathcal{E} + {}^L\mathbf{E}_\alpha)\, dv,$$

of which the local form naturally is Eq. (4.11). The same procedure is repeated for all $\alpha = 1, 2, ..., n$, so that one gets n equations of the type (4.29) for the n molecular species. Although the physical significance of the local equation (4.11) was made clear via Eq. (4.15), the meaning of Eq. (4.29) is as yet unclear.

   D. *The principle of virtual power for real velocity fields.* Consider now that the virtual fields that appear in the general expression (4.8) are none other than the present fields (no asterisk), at time t, which are the solutions of a well-posed problem. Then Eq. (4.8) can be rewritten in the following form:

(4.30)
$$\dot{K}(\mathcal{D}_t) + \int_{\partial \mathcal{D}_t} \frac{\partial \mathbf{G}}{\partial t} \cdot \mathbf{U} dv = \mathscr{P}_{(i)}(\mathcal{D}_t) + \mathscr{P}_{(v)}(\mathcal{D}_t) + \mathscr{P}_{(c)}(\partial \mathcal{D}_t),$$

where the total kinetic energy is defined as

$$K(\mathcal{D}_t) = \int_{\mathcal{D}_t} \frac{1}{2} \varrho \left(\mathbf{U}^2 + \sum_\alpha c_\alpha d_\alpha \dot{\pi}_\alpha^2 \right) dv.$$

Upon combining Eq. (4.30) with the energetic identity (2.28), one obtains the following energetic expression:

(4.31)
$$\frac{d}{dt}\{K(\mathcal{D}_t) + U^{em}(\mathcal{D}_t)\} = \mathscr{P}_{(i)}(\mathcal{D}_t) + \int_{\mathcal{D}_t} \mathbf{f} \cdot \mathbf{U} dv + \int_{\partial \mathcal{D}_t} (\mathbf{T} \cdot \mathbf{U} - \mathscr{J} \cdot \mathbf{n}) da.$$

Remark that this is not the statement of the first principle of thermodynamics. This completes the exploitation of the virtual power principle (4.8), *per se*. To proceed further one must combine it with the global statements of the first and second principles of thermodynamics. (See Part Two). To conclude this section, it can in fact be remarked that the virtual power principle used herein above is of essentially mechanistic nature and is not a general energy principle (for instance, Maxwell's equations cannot be deduced from it). However, in contrast to Lagrangian and Hamiltonian principles, d'Alembert's principle allows one to deal with general thermodynamical processes (e.g., no hypotheses have been made with regard to the thermodynamical behaviour of the fields σ_{ij} and ${}^L\mathbf{E}_\alpha$ which may present dissipative contributions).

5. Relationship with other theories of deformable dielectrics

   First, one may think of eliminating the interaction fields ${}^L\mathbf{E}_\alpha$ between Eqs. (4.9) and (4.11). To this purpose it is convenient to define the following symmetric tensor:

(5.1)
$$^E t_{ij} \equiv t_{ij} - t_{ij}^{INT} = \sigma_{ij} - t_{(ij)}^{INT} = {}^E t_{ji}.$$

From Eqs. (4.11) it follows that

(5.2)
$$t_{ij}^{INT} = \varrho \sum_\alpha c_\alpha d_\alpha \ddot{\pi}_{\alpha i} \pi_{\alpha j} - \mathcal{E}_i P_j.$$

Substituting from t_{ij} in Eq. (4.9) and taking account of Eqs. (5.1), (5.2), (2.13) and (2.15) one obtains the following expression for the Euler-Cauchy equations:

$$(5.3) \qquad \varrho\dot{\mathbf{U}} + \frac{\partial \mathbf{G}}{\partial t} = \mathrm{div}(^{E}\mathbf{t} + \mathbf{t}^{em\cdot f}) + \mathbf{f} + \varrho\sum_{\alpha} c_{\alpha}d_{\alpha}[(\boldsymbol{\pi}_{\alpha}\cdot\nabla)\ddot{\boldsymbol{\pi}}_{\alpha} + \ddot{\boldsymbol{\pi}}_{\alpha}(\nabla\cdot\boldsymbol{\pi}_{\alpha})].$$

It can be shown in the case of non-linear elastic bodies (Cf. Part 2) that $^{E}t_{ij}$ no longer contains the interaction fields $^{L}\mathbf{E}_{\alpha}$. Thus, at least for this case, the interactions $^{L}\mathbf{E}_{\alpha}$ no longer participate in the motion equation (5.3). However, when there are several molecular species, we still have to consider the equations (4.11) for, obviously, by summimg over α, we have lost much information. That is, the three scalar equations represented by Eq. (5.3) cannot replace the $3(n+1)$ scalar equations represented by Eqs. (4.9) and (4.11). One may however inquire in what conditions this equivalence holds true. Obviously, this is so if there is only *one* polarization continuum, for instance, when one considers only electronic polarization, in which case Eqs. (5.3) and (4.11) are reduced to (d^{e} = electronic polarization inertia; cf. Appendix)

$$(5.4) \qquad \varrho\dot{\mathbf{U}} + \frac{\partial \mathbf{G}}{\partial t} = \mathrm{div}(^{E}\mathbf{t} + \mathbf{t}^{em\cdot f}) + \varrho d^{e}[(\boldsymbol{\pi}\cdot\nabla)\ddot{\boldsymbol{\pi}} + \ddot{\boldsymbol{\pi}}(\nabla\cdot\boldsymbol{\pi})] + \mathbf{f},$$

and

$$(5.5) \qquad \boldsymbol{\mathcal{E}} + {}^{L}\mathbf{E} = d^{e}\ddot{\boldsymbol{\pi}},$$

where there exists only one interaction field $^{L}\mathbf{E}$. Then the relationship with theories previously set forth by several authors is made clear. If polarization inertia is discarded — $d^{e} = 0$ — then, using the identity (2.20), Eq. (5.4) takes on the form

$$(5.6) \qquad \varrho\dot{\mathbf{U}} = \mathrm{div}^{E}\mathbf{t} + \mathbf{f} + \bar{\mathbf{f}}^{em},$$

where $\bar{\mathbf{f}}^{em}$ has the expression given by Eq. (2.19). Eq. (5.6) is the Cauchy equation of Toupin's [4] theory (See also DIXON and ERINGEN [3]). If, furthermore, magnetic induction is discarded (i.e., in quasi-electrostatics), then Eq. (5.4) reduces to

$$(5.7) \qquad \varrho\dot{\mathbf{U}} = \mathrm{div}(^{E}\mathbf{t} + \mathbf{t}^{em\cdot f}) + \mathbf{f},$$

or

$$(5.8) \qquad \varrho\dot{\mathbf{U}} = \mathrm{div}\,\mathbf{t} + \mathbf{f} + (\mathbf{P}\cdot\nabla)\mathbf{E}.$$

The Kelvin force $(\mathbf{P}\cdot\nabla)\mathbf{E}$ of Eq. (5.7) can also be written as $(\nabla\mathbf{E})\cdot\mathbf{P}$ on account of Maxwell's electrostatic equation $\nabla\times\mathbf{E} = \mathbf{0}$. By the same token Eq. (5.5) is reduced to

$$(5.9) \qquad \mathbf{E} + {}^{L}\mathbf{E} = \mathbf{0}.$$

Equation (5.7) is Cauchy's equation as considered by ERINGEN [10] and SUHUBI [11], whereas Eqs. (5.8) and (5.9) are the equations of TIERSTEN [5]. Eq. (5.9) was also considered in the pioneering work of TOUPIN [13]. Thus all previously given theories of non-linear elastic dielectrics (excluding polarization gradients and electric quadrupoles) appear to be special cases of the present one (at least, as far as Cauchy's equation and the balance equation of the type (4.11) are concerned, but the statement holds true also for the thermo-dynamical considerations; cf. Part 2). A final remark, however, is in order, for the different formulations, although mathematically equivalent in their respective frame of approxi-

mation, do not bear the same conceptual and physical significance. In the formulations (5.4), (5.5) and (5.6) the notion of interaction or local electric field has disappeared. This fact yields a common misunderstanding. Explicitly, since only *symmetric* stress tensors — such as $^E t_{ij}$ and $t_{ij}^{em \cdot f}$ — remain in the formulations, one is tempted to say that the stress tensor is symmetric and that there do not exist volume couples in such dielectrics. However, the tensor $^E t_{ij}$ is not to be mistaken for the Cauchy stress([5]), the latter still being *non-symmetric* according to Eq. (4.12) or (4.14)$_2$, of which the latter is equivalent to Eq. (4.25) in the general case and reduces to the equations $t_{[ij]} = P_{[i} \mathscr{E}_{j]}$ or $t_{[ij]} = P_{[i} E_{j]}$ depending on whether one considers the simplified case of Eq. (5.5) or (5.8), respectively. In these last two cases the asymmetry of t_{ij} is caused or balanced by the ponderomotive couple, a notion absolutely necessary if one desires to understand the phenomenon of electric-dipole orientation in an applied electric field. That is, the medium considered is polar in spite of the absence of couple stresses.

Appendix

We determine the inertia associated with the internal deformations of the molecular species. If m_α is the mass set in motion within the α-molecule in the process of its polarization then, per unit volume which contains n_α α-molecules and $\alpha = 1, 2, ..., n$ different molecular species, the corresponding kinetic energy due to the dipole displacements $\mathbf{w}_\alpha$ is

$$(A.1) \qquad k^{(p)} = \sum_\alpha k_\alpha^{(p)} = \sum_\alpha \frac{1}{2} n_\alpha m_\alpha \dot{\mathbf{w}}_\alpha^2 .$$

On account of Eqs. (3.4) and (3.5), (A.1) is rewritten as

$$(A.2) \qquad k^{(p)} = \varrho \sum_\alpha \frac{1}{2} c_\alpha d_\alpha \dot{\boldsymbol{\pi}}_\alpha^2 ,$$

where the α-molecular-polarization inertia d_α is defined as

$$(A.3) \qquad d_\alpha \equiv \left(\frac{m_\alpha}{eY_\alpha}\right)^2 = \left(\frac{\varrho_\alpha}{q_\alpha}\right)^2 .$$

Equivalently, if K_α is the spring constant related to the displacement $\mathbf{w}_\alpha$ in the mechanical model of polarization, and if A_α denotes the polarizability of the α-molecular species in such a way that $A_\alpha \equiv (eY_\alpha)^2/K_\alpha$, then $d_\alpha = m_\alpha^2/A_\alpha K_\alpha$. Furthermore, if electrons only are accounted for, then $Y_\alpha = 1$ and d_α goes to $d^e \equiv (m_0/e)^2$, which is the value considered by MAUGIN and COLLET in a previous work [13]. Finally, it is clear that the virtual power associated with the kinetic energy $k_\alpha^{(p)}$ is

$$(A.4) \qquad p_{(p)\alpha}^* = \varrho c_\alpha d_\alpha \pi_\alpha \cdot \dot{\pi}_\alpha^* ,$$

so that, for a *real* field (no asterisk),

$$p_{(p)\alpha} \equiv \varrho \frac{d}{dt} (k_\alpha^{(p)}/\varrho) .$$

([5]) That, is the equation $^E t_{[ij]} = 0$ is not to be mistaken for the local statement of the balance of moment of momentum, but is only an algebraic identity resulting from the very definition (5.1).

Acknowledgments

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