RELAXATION PHENOMENA IN ELECTRO-MAGNETO-ELASTICITY

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Abstract

A short review is given of the balance and constitutive equations in the theory of the interactions of electro-magnetic and elastic fields. Part of the constitutive equations is derived from thermodynamic potentials. In the classical theory these potentials are functions of the instantaneous values of the independent parameters. The theory is extended to the class of materials for which the thermodynamic potentials are functionals, defined in the domain of the histories of the parameters. This class of materials exhibits relaxation and retardation phenomena. After a general formulation for elastic materials with electro-magnetic relaxation, the theory is confined to linear relaxation, in which the functionals are quadratic. In the last section the special case of piezo-electric retardation is discussed.

1. Introduction

This paper may be considered as a generalization and continuation of a recent paper ¹ by the present author concerning the general theory of electro- and magneto-elastic interactions in continuous media. In ref. ¹ some attention was given to irreversible processes, like magnetic dissipation in ferromagnetics. Here we investigate on the basis of the modern theory of non-equilibrium thermodynamics, the relaxation phenomena which occur in dielectrics and magnetic materials.

Our considerations are relevant to polarized and magnetized materials, saturated or nonsaturated, whether conducting or not. Such a generality is not necessary for the application to special problems, e.g. dielectric relaxation, but it throws light on similarities and distinctions between electric and magnetic behavior. However, we restrict our discussion to the non-relativistic theory for the purely elastic material.

The literature on the interactions of electric, magnetic and elastic fields is extensive. In ref. ¹ we present a list of important contributions. In this paper we found our considerations on Chu’s new formulation of electro-magnetism ²). The basis of Chu’s theory is the assumption that a material body, in rest or in motion, may be considered as a set of electric and/or magnetic sources, placed in vacuum. Chu needs two electro-magnetic vectors for the description of his
field. An extensive treatment of Chu's theory with comparison to some other electro-magnetic theories and applications to relativistic and nonrelativistic problems, may be found in a book by Penfield and Haus 3).

Chu's formulation of Maxwell's equations in rationalized MKS units is

\[ \begin{align*}
\text{curl } E &= -\mu_0 \frac{\partial H}{\partial t} - \mu_0 \varrho \hat{M}, \\
\text{curl } H &= \varepsilon_0 \frac{\partial E}{\partial t} + \mathbf{J} + \varrho \hat{P}, \\
\varepsilon_0 \text{ div } E &= -\text{div} (\varrho \mathbf{P}) + \varrho_{el}, \\
\mu_0 \text{ div } H &= -\text{div} (\mu_0 \varrho \mathbf{M}),
\end{align*} \]

where \( E \) and \( H \) are the electric and magnetic field intensities, respectively, \( P \) is the polarization per unit mass, \( M \) is the magnetization per unit mass, \( \varrho \) is the mass density, \( \varepsilon_0 \) is the permittivity and \( \mu_0 \) the permeability of free space, \( \varrho_{el} \) is the density of charge and \( J \) is the current density. We have defined the time derivative \( \hat{P} \) by

\[ \hat{P} = \frac{\partial (\varrho \mathbf{P})}{\partial t} + \text{curl} (\varrho \mathbf{P} \times \mathbf{v}), \]

where \( \mathbf{v} \) is the velocity of matter. The corresponding definition for \( \hat{M} \) holds. In fact we have

\[ \hat{P} = \hat{P} - \mathbf{v} \text{ div} (\varrho \mathbf{P}), \]

where \( \hat{P} \) denotes the convected time derivative 4).

In the nonrelativistic theory the convected electro-magnetic vectors are

\[ \begin{align*}
\mathbf{H}^* &= \mathbf{H} - (\mathbf{v} \times \varepsilon_0 \mathbf{E}), \\
\mathbf{E}^* &= \mathbf{E} + (\mathbf{v} \times \mu_0 \mathbf{H}), \\
\mathbf{P}^* &= \mathbf{P}, \\
\mathbf{M}^* &= \mathbf{M},
\end{align*} \]

and these are invariant with respect to a Galilei transformation, as defined by

\[ \begin{align*}
\mathbf{v} \rightarrow \mathbf{v} + \mathbf{b}, \quad t \rightarrow t, \quad \nabla \rightarrow \nabla, \quad \frac{\partial}{\partial t} \rightarrow \frac{\partial}{\partial t} - \mathbf{b} \cdot \nabla, \quad \frac{d}{dt} \rightarrow \frac{d}{dt},
\end{align*} \]

where \( \mathbf{b} \) is a constant vector.

In matter we have to distinguish the following electric forces: \( \mathbf{E} \), the field intensity, i.e. the force on a unit charge in vacuum in a field with the prescribed sources, \( \mathbf{E}^* \), the convected field intensity, \( \mathbf{D} \), the dielectric displacement, defined in the usual way, \( G^{(e)} \), the effective field intensity, i.e. the work coefficient in the energy-balance equation. Corresponding definitions may also be given for the magnetic forces. Further we have to introduce \( f^{(em)} \), the ponderomotive force
density. We note that none of these vectors are directly accessible to measurement.

In sec. 2 we derive the balance equations from a principle, first stated by Green and Rivlin \(^5\)), that may be formulated as follows: The first law of thermodynamics, the energy-balance equation, is invariant under superposed rigid-body translations and rotations. This principle has proved its usefulness for the derivation of the known balance equations in the theory of elasticity and the theory of the Cosserat Continuum. In sec. 3 we obtain some constitutive equations by the methods of thermodynamics. In sec. 4 we confine ourselves to the class of materials with linear relaxation equations. In sec. 5 we discuss a special case, the elastic dielectric, and in sec. 6 we apply the theory to linear piezoelectricity.

2. Balance equations

We consider a body that is placed in an external electro-magnetic field and is loaded by body and surface forces. The energy-balance equation can be written in the following form:

\[
\frac{d}{dt} \int_V \rho U \, dV + \frac{d}{dt} \int_V \rho \mathbf{v}_k \cdot \mathbf{v}_k \, dV + \frac{d}{dt} \int_V W \, dV =
\]

\[
= \int_V \mathbf{f}_k^{(\text{mech})} \mathbf{v}_k \, dV + \int_S T_k \mathbf{v}_k \, dS - \int_V G_k^{(e)} \rho \mathbf{P}_k \, dV - \int_V G_k^{(m)} \rho \mathbf{M}_k \, dV +
\]

\[
+ \int_S Q_k^{(e)} \mathbf{P}_k \, dS + \int_S Q_k^{(m)} \mathbf{M}_k \, dS + \int_V \rho r \, dV - \int_S h \, dS + \int_S X \, dS. \tag{6}
\]

In (6) the energy-balance equation is applied to an arbitrary finite part of the body with volume \(V\) and bounding surface \(S\), moving in such a way that it always consists of the same particles. Thus \(d/dt\) denotes a material derivative, which is also indicated by a dot. In (6) \(U\) is the local internal energy per unit mass and \(W\) is the electro-magnetic field energy density. The mechanical body force is denoted by \(\mathbf{f}^{(\text{mech})}\) and the stress vector by \(\mathbf{T}\). The quantity \(r\) denotes the heat supply per unit mass and unit time, \(h\) is the heat flux, \(Q^{(e)}\) and \(Q^{(m)}\) are the electric and magnetic surface vectors, respectively. \(X\) is an extra energy supply due to the electro-magnetic field outside of \(V\). It contains, for instance, the Poynting energy flux and the dipole interaction energy from outside \(V\). We note that \(X\) is an unknown of the theory that has to be determined. In (6) the interaction energy is split into two parts

\[
\int_V \rho U \, dV \quad \text{and} \quad \int_V W \, dV.
\]
The first integral accounts for the short-range local energy, the exchange coupling, the spin–orbit coupling, near electric and magnetic dipole interactions and the deformation energy. The second integral comes from the long-range interaction, e.g. the dipole–dipole interaction within $V$ and the external field energy. Dipole–dipole interactions of particles within $V$ with particles outside of $V$ are included in $\int_X dS$.

According to Chu’s theory we assume for $W$ the expression

$$W = \frac{1}{2} \varepsilon_0 E^2 + \frac{1}{2} \mu_0 H^2.$$  

(7)

The condition for (6) to be invariant with respect to rigid-body transformations yields the following balance equations:

$$\dot{e} + e v_{k,k} = 0,$$

(8)

$$T_{kl,t} + e f_{k}^{(\text{mech})} + e f_{k}^{(\text{em})} = e \dot{v}_{k},$$

(9)

$$M_k = \Gamma e_{klm} M_{l} G_{m}^{(e)} + \Gamma e_{klm} P_{l} G_{m}^{(e)};$$

(10)

for mass, linear momentum and angular momentum, respectively. In (9) $f_{k}^{(\text{em})}$ is given by (if $\varepsilon_{el}$ is put equal to zero)

$$e f_{k}^{(\text{em})} = E_{k,t}^{*} e P_{t} + \mu_0 (e \hat{P} \times H^{*})_{k} + \mu_0 (J \times H^{*})_{k} +$$

$$- \varepsilon_0 \mu_0 (e \hat{M} \times E^{*})_{k} + \mu_0 H_{k,t}^{*} e M_{t};$$

(11)

which may be derived from a Maxwell stress tensor $t_{klt}$, according to

$$e f_{k}^{(\text{em})} = t_{klt},$$

(12)

with

$$t_{klt} = E_{k,t}^{*} D_{l,t}^{*} - \frac{1}{2} \varepsilon_0 E^{*2} \delta_{klt} + H_{k,t}^{*} B_{l,t}^{*} - \frac{1}{2} \mu_0 H^{*2} \delta_{klt}.$$  

(13)

In (10) $\Gamma$ is the gyromagnetic ratio. With (8), (9), and (11) the balance equation (6) simplifies to

$$\int_{V} \rho U dV = \int_{V} (E_{k}^{*} - G_{k}^{(e)}) \rho \dot{P}_{k} dV + \int_{V} (\mu_0 H_{k}^{*} - G_{k}^{(m)}) \rho \dot{M}_{k} dV +$$

$$+ \int_{V} E_{k}^{*} J_{k} dV + \int_{V} T_{klt} v_{k,t} dV + \int_{S} Q_{k}^{(m)} M_{k} dS + \int_{V} \rho r dV - \int_{S} h dS,$$

(14)

if $Q^{(e)}$ is put equal to zero.

For the derivation of (8) to (13) and many other details, see ref. 1.

3. Constitutive equations

We introduce the specific entropy $\eta$, that satisfies the Clausius–Duhan in-
equality
\[ \frac{d}{dt} \int \theta \eta \, dV - \int \frac{\theta r}{\theta} \, dV + \int \frac{h}{\theta} \, dS \geq 0, \quad (15) \]
where \( \theta \) is the (absolute) temperature, and the Helmholtz free energy \( \psi \) by
\[ \psi = U - \theta \eta. \quad (16) \]

Eliminating \( r \) and \( h \) from (14) and (15) we obtain the inequality
\[ \int \left( \theta [\dot{\eta} + \dot{\eta}_r + (G_k^{(e)} - E_k^{*}) \dot{P}_k + (G_k^{(m)} - \mu_0 H_k^{*}) \dot{M}_k] + \right. \\
- T_{kl} v_{k,l} - E_k^{*} f_k \right) dV - \int Q_k^{(m)} \dot{M}_k \, dS + \int \frac{h_k \theta_k}{\theta} \, dV \leq 0. \quad (17) \]

We assume that \( \psi \) depends on the instantaneous values of \( \theta, x_{k,a}, P_k, M_k, M_{k,a} \), but is a functional of the histories of \( P \) and \( M \). We write
\[ \psi(t) = \Psi(\theta(t), x_{k,a}(t), P_k(t), P_k(t - s), M_k(t), M_k(t - s), M_{k,a}(t)), \quad 0 < s < \infty, \quad (18) \]
where \( x_{k,a} \) and \( M_{k,a} \) are defined by
\[ x_{k,a} = \frac{dX_k}{dX_a}, \quad M_{k,a} = \frac{dM_k}{dX_a}, \quad (19) \]
with \( X_a \) the Lagrangian coordinates of the particle.

To make any further progress we have to make some assumptions concerning the functional \( \Psi \). We introduce
\[ P_k^{t}(s) = P_k(t - s), \quad M_k^{t}(s) = M_k(t - s) \quad (20) \]
and assume that the functional derivatives
\[ \delta_{P_k} \Psi(\theta, x_{k,a}, P_k, P_k^{t}, M_k, M_k^{t}, M_{k,a}, \mu_k) = \frac{d}{d\epsilon} \Psi(P_k^{t} + \epsilon \mu_k^{t}) \big|_{\epsilon = 0}, \quad (21) \]
\[ \delta_{M_k} \Psi(\theta, x_{k,a}, P_k, P_k^{t}, M_k, M_k^{t}, M_{k,a}, \mu_k) = \frac{d}{d\epsilon} (M_k^{t} + \epsilon \mu_k^{t}) \big|_{\epsilon = 0} \quad (22) \]
do exist in the domain of histories. Then we may write
\[ \dot{\psi}(t) = \frac{d\Psi}{d\theta} \dot{\theta} + \frac{d\Psi}{dX_{k,a}} x_{k,a} + \frac{d\Psi}{dP_k} \dot{P}_k + \frac{d\Psi}{dM_k} \dot{M}_k + \frac{d\Psi}{dM_{k,a}} \dot{M}_{k,a} + \]
\[ + \delta_{P_k} \Psi(\ldots/\dot{P}_k^{t}) + \delta_{M_k} \Psi(\ldots/\dot{M}_k^{t}) \quad (23) \]
Because the functional \( \Psi \) has to be invariant with respect to rigid-body rota-
tions, it must be of the form

\[ \psi = \mathcal{V}(\theta, A_{\alpha \beta}, C_{\alpha \beta}, N_{\alpha}, N_{\alpha}^t, S_{\alpha}, S_{\alpha}^t) \]  

with

\[ A_{\alpha \beta} = x_{k,\alpha} M_{k,\beta}, \quad C_{\alpha \beta} = x_{k,\alpha} x_{k,\beta}, \]
\[ N_{\alpha} = x_{k,\alpha} M_k, \quad S_{\alpha} = x_{k,\alpha} P_k. \]  

From (24), (23) and (18) we derive the constitutive equations

\[ T_{kl} = q \chi \left( \frac{\partial \psi}{\partial A_{\alpha \beta}} M_{k,\beta} + \frac{\partial \psi}{\partial C_{\alpha \beta}} M_k + 2 \frac{\partial \psi}{\partial N_{\alpha}} x_{k,\beta} + \frac{\partial \psi}{\partial S_{\alpha}} P_k \right), \]  

\[ G^{(e)}_k = E_k^* - \frac{\partial \psi}{\partial S_{\alpha}} x_{k,\alpha}, \]  

\[ R_k = \mu_0 H_k^* - \frac{\partial \psi}{\partial N_{\alpha}} x_{k,\alpha} + \frac{1}{\theta} \left( \theta \frac{\partial \psi}{\partial A_{\alpha \beta}} x_{k,\alpha} x_{l,\beta} \right), \]  

\[ Q_{k}^{(m)} = \frac{\partial \psi}{\partial A_{\alpha \beta}} x_{k,\alpha} x_{l,\beta} n_l. \]  

In (26) \( T_{kl} \) is the reversible part of \( T_{kl} \) and in (28) \( R_k \) is the nondissipative part of \( G^{(m)}_k \). Introducing magnetic dissipation, we can write \( G^{(m)}_k \) as follows:

\[ G^{(m)}_k = R_k - \eta (M_k - e_{klm} \omega_l M_m), \]  

with \( \eta \) the coefficient of "magnetic viscosity", \( e_{klm} \) the alternator and \( \omega_l \) given by

\[ \omega_l = \frac{1}{2} e_{lpq} v_{q,p}. \]  

It appears then that we also have to decompose \( T_{kl} \) according to

\[ T_{kl} = \bar{T}_{kl} + T_{kl}^*, \]  

with \( T_{kl}^* \) the dissipative part of \( T_{kl} \). We find for the skew-symmetric part of \( T_{kl}^* \) (cf. ref. 1)

\[ T_{[kl]}^* = -\theta \eta [(\dot{M}_k - e_{[kpq} \omega_p M_q)] M_{l}]. \]  

With (26) to (29) the inequality (17) simplifies to

\[ \int \left( T_{kl}^* v_{k,l} + E_k^* J_k + \theta \eta [(\dot{M}_k - (\omega \times M)_k)] \dot{M}_k - \frac{h_k \theta_k}{\theta} \right) dV + \]
\[ -\delta_{N_{\alpha}} \mathcal{V}(\ldots / N_{\alpha}^t) - \delta_{S_{\alpha}} \mathcal{V}(\ldots / S_{\alpha}^t) \geq 0. \]  

From (34) we conclude

\[ T_{kl}^* v_{(k,l)} \geq 0, \]
\[ \theta \eta [(\dot{M}_k - (\omega \times M)_k)] (\dot{M}_k - (\omega \times M)_k) \geq 0, \]
These inequalities are important. Physically they express the fact that in irreversible processes the dissipation is nonnegative. It will appear that especially (38) and (39) considerably limit the number of admissible constitutive equations.

4. Linear relaxation

The preceding theory is very general. To be able to make explicit calculations we shall confine our discussion to a smaller class of materials, the materials with linear relaxation equations. For this class we assume the free-energy functional to be quadratic in $N^t$ and $S^t$. We write

$$\psi = \psi_0(\theta, A_{\alpha \beta}, C_{\alpha \beta}, N_\alpha, S_\alpha) +$$

$$+ \int_0^\infty f_{\alpha}^{(1)}(\theta, A_{\alpha \beta}, C_{\alpha \beta}, N_\alpha; s) [N_\alpha'(s) - N_\alpha(t)] \, ds +$$

$$+ \int_0^\infty f_{\alpha}^{(2)}(\theta, A_{\alpha \beta}, C_{\alpha \beta}, N_\alpha; s) [S_\alpha'(s) - S_\alpha(t)] \, ds +$$

$$+ \frac{1}{2} \int_0^\infty \int_0^\infty f_{\alpha \beta}^{(11)}(\ldots; s_1, s_2) [N_\alpha'(s_1) - N_\alpha(t)] [N_\beta'(s_2) - N_\beta(t)] \, ds_1 \, ds_2 +$$

$$+ \frac{1}{2} \int_0^\infty \int_0^\infty f_{\alpha \beta}^{(12)}(\ldots; s_1, s_2) [N_\alpha'(s_1) - N_\alpha(t)] [S_\beta'(s_2) - S_\beta(t)] \, ds_1 \, ds_2 +$$

$$+ \frac{1}{2} \int_0^\infty \int_0^\infty f_{\alpha \beta}^{(21)}(\ldots; s_1, s_2) [N_\alpha'(s_2) - N_\alpha(t)] [N_\beta'(s_1) - N_\beta(t)] \, ds_1 \, ds_2 +$$

$$+ \frac{1}{2} \int_0^\infty \int_0^\infty f_{\alpha \beta}^{(22)}(\ldots; s_1, s_2) [S_\alpha'(s_1) - S_\alpha(t)] [S_\beta'(s_2) - S_\beta(t)] \, ds_1 \, ds_2. \quad (40)$$

In (40) we have

$$f_{\alpha \beta}^{(11)}(\ldots; s_1, s_2) = f_{\beta \alpha}^{(11)}(\ldots; s_2, s_1),$$

$$f_{\alpha \beta}^{(12)}(\ldots; s_1, s_2) = f_{\alpha \beta}^{(21)}(\ldots; s_2, s_1),$$

$$f_{\alpha \beta}^{(22)}(\ldots; s_1, s_2) = f_{\alpha \beta}^{(22)}(\ldots; s_2, s_1),$$

$$f_{\alpha \beta}^{(11)}(\ldots; s_1, s_2) = f_{\alpha \beta}^{(11)}(\ldots; s_2, s_1),$$

$$f_{\alpha \beta}^{(12)}(\ldots; s_1, s_2) = f_{\alpha \beta}^{(12)}(\ldots; s_2, s_1),$$

$$f_{\alpha \beta}^{(21)}(\ldots; s_1, s_2) = f_{\alpha \beta}^{(21)}(\ldots; s_2, s_1),$$

$$f_{\alpha \beta}^{(22)}(\ldots; s_1, s_2) = f_{\alpha \beta}^{(22)}(\ldots; s_2, s_1).$$
and corresponding equalities between the other $f$ functions. The conditions (38) and (39) yield

$$f_a^{(1)} = f_a^{(2)} = f_a^{(12)} = f_a^{(21)} = 0, \quad (\alpha, \beta = 1; 2, 3),$$

(42)

together with the dissipation inequalities

$$\int_0^\infty \int_0^\infty f_{ab}^{(11)}(\ldots; s_1, s_2) N_a(t - s_1) [N_\beta(t - s_2) - N_\beta(t)] \, ds_1 \, ds_2 \leqslant 0,$$

(43)

$$\int_0^\infty \int_0^\infty f_{ab}^{(22)}(\ldots; s_1, s_2) S_a(t - s_1) [S_\beta(t - s_2) - S_\beta(t)] \, ds_1 \, ds_2 \leqslant 0.$$

(44)

We introduce the functions

$$g_{ab}^{(pq)}(\ldots; s_1, s_2) = \int_{s_1}^{s_2} f_{ab}^{(pq)}(\ldots; s_3, s_2) \, ds_3, \quad (p, q = 1, 2)$$

(45)

and write (43) and (44) after partial integration in the form

$$\int_0^\infty \int_0^\infty g_{ab}^{(11)}(\ldots; s_1, s_2) N_a(t - s_1) \, ds_1 \, ds_2 \geqslant 0,$$

(46)

$$\int_0^\infty \int_0^\infty g_{ab}^{(22)}(\ldots; s_1, s_2) S_a(t - s_1) \, ds_1 \, ds_2 \geqslant 0.$$

(47)

To simplify the constitutive equations, we assume the $f_{ab}^{(pq)}$ and $g_{ab}^{(pq)}$ functions to be independent of $\theta, A_{ab}, C_{\alpha\beta}, N_\alpha, S_\alpha$. Then we derive, with the aid of (26) to (29), the following expressions:

$$T_{kl} = \varepsilon x_{1,\alpha} \left( \frac{\partial \psi_0}{\partial A_{ab}} M_{k,\beta} + \frac{\partial \psi_0}{\partial N_\alpha} M_k + 2 \frac{\partial \psi_0}{\partial C_{\alpha\beta}} x_{k,\beta} + \frac{\partial \psi_0}{\partial S_\alpha} P_k \right) +$$

$$- \varepsilon x_{1,\alpha} M_k \int_0^\infty g_{ab}^{(11)}(0, s) [N_\beta(t - s) - N_\beta(t)] \, ds +$$

$$- \varepsilon x_{1,\alpha} P_k \int_0^\infty g_{ab}^{(22)}(0, s) [S_\beta(t - s) - S_\beta(t)] \, ds,$$

(48)
For the complete theory we have to add eq. (29) and the balance and field equations. There results a nonlinear theory in which the partial differential equations are replaced by integral–differential equations. For a discussion of the boundary and jump conditions, see ref. 1.

5. The nonlinear elastic dielectric

For the discussion of the elastic dielectric we take in (6)

\[ G^{(m)} = M = Q^{(e)} = Q^{(m)} = \mathbf{0}. \]

It appears that for many cases of practical interest we may put

\[ G^{(e)} = J = \mathbf{0}. \]

The inequality (17) simplifies to

\[ \int \left[ \theta \left( \psi + \dot{\psi} \eta - E_k^* \dot{P}_k - T_{kl} v_{k,l} \right) \right] dV + \int \frac{h_k \theta, \psi}{\theta} dV \leq 0. \]  

Putting \( \psi \) as a function of \( \theta, C_{\alpha\beta} \) and \( S_\alpha \) and a functional of \( S_\alpha^s \):

\[ \psi = \Psi(\theta, C_{\alpha\beta}, S_\alpha, S_\alpha^s(s)), \quad 0 < s < \infty, \]

we find the constitutive equations

\[ \eta = - \frac{\partial \Psi}{\partial \theta}, \]

\[ T_{kl} = \sqrt{\rho} x_{k,\alpha} \left( 2 \frac{\partial \Psi}{\partial C_{\alpha\beta}} x_{k,\alpha} + \frac{\partial \Psi}{\partial S_\alpha} P_k \right), \]

\[ E_k^* = \frac{\partial \Psi}{\partial S_\alpha} x_{k,\alpha}. \]

The independent variables in these equations are \( x_{k,\alpha} \) and \( P_k \), from which \( C_{\alpha\beta} \) and \( S_\alpha \) are calculated. Sometimes it is expedient to change the variables and
to consider $x_{k,a}$ and $E_k^*$ as independent. To this end we introduce the electric enthalpy

$$\varrho \chi = \varrho \psi - E_k^* P_k \varrho.$$  \hfill (58)

The inequality (53) becomes

$$\int_{\mathcal{V}} \left[ \varrho \left( \dot{\chi} + \dot{\psi} \eta + \dot{E}_k^* P_k \right) - T_{kl} v_{k,l} \right] dV + \int_{\mathcal{V}} \frac{h_k \theta_k}{\theta} dV \leq 0,$$  \hfill (59)

from which we derive

$$\varrho P_k = -\varrho \frac{\partial \chi}{\partial E_k^*},$$  \hfill (60)

$$T_{kl} = \varrho \frac{\partial \chi}{\partial x_{k,a}}.$$  \hfill (61)

We consider a thermodynamic process that is isothermal and closed, i.e., there exist two times $t_1$ and $t_2$ ($> t_1$) so that for $t \leq t_1$ and for $t \geq t_2$ the states of the system are equal. Then we have the local inequality

$$\varrho \dot{\chi} + \dot{E}_k^* \varrho P_k - T_{kl} v_{k,l} \leq 0,$$  \hfill (62)

from which there results for this process

$$\int_{-\infty}^{\infty} P_k \dot{E}_k^* dt - \int_{-\infty}^{\infty} \frac{1}{\varrho} T_{kl} v_{k,l} dt \leq 0,$$  \hfill (63)

because

$$\int_{-\infty}^{\infty} \dot{\chi} dt = \chi \bigg|_{-\infty}^{\infty} = 0.$$  \hfill (64)

While a discussion based upon the \$ functional with its variables $x_{k,a}$ and $P_k$ leads to the description of the relaxation of $E_k^*$, the $\chi$ formalism yields the retardation of the polarization in dependence on the electric field.

6. Application to linear piezoelectricity

The balance equations for a linear piezo-electric body are

$$\frac{\partial \varrho}{\partial t} + \varrho_0 v_{k,k} = 0,$$  \hfill (65)

$$T_{kl,t} + \varrho_0 f_k^{(\text{mech})} = \varrho_0 \frac{\partial v_k}{\partial t},$$  \hfill (66)
where $\rho_0$ is the density of mass for the unloaded state. In (66) and (67) we have omitted the terms $\rho_0 E_{k,t} P_t$ and $\rho_0 E_{k,l} P_{13}$, respectively. Both consist of two parts: a contribution from an external electric field and one from the residual field. We take the external field uniform and constant in which case the first part vanishes. The other part is of the second order and has to be dropped for consistency.

In the linear theory $P_k$ always occurs in the combination $\rho_0 P_k$. Therefore we transform here

$$\rho_0 P_k \rightarrow P_k$$

and we shall define in this section $\mathbf{P}$ as the polarization per unit volume. We confine ourselves to isothermal processes and assume the enthalpy of the form

$$\chi = \frac{1}{2} c_{ijkl} e_{ij} e_{kl} - f_{klj} E_k e_{ij} - \frac{1}{2} e_{kl} E_k E_l +$$

$$- \frac{1}{2} \int_0^\infty \int_0^\infty K_{kl}(s_1, s_2) [E_k(t - s_1) - E_k(t)] [E_l(t - s_2) - E_l(t)] ds_1 ds_2,$$

where $e_{ij}$ is given by

$$e_{ij} = \frac{1}{2} (u_{i,j} + u_{j,i}).$$

In (70), $c_{ijkl}$ are the coefficients of elasticity, $f_{klj}$ are the piezo-electric coupling coefficients and $e_{kl}$ are the susceptibilities. We have

$$e_{kl} = e_{lk}; \quad K_{kl}(s_1, s_2) = K_{lk}(s_2, s_1).$$

In (71) $e_{ij}$ is the linear deformation tensor and $u_t$ is the displacement. From (60) and (61) we derive the constitutive equations

$$P_k(t) = f_{klj} e_{ij} + e_{kl} E_l - \int_0^\infty \int_0^\infty K_{kl}(s_1, s_2) [E_l(t - s_2) - E_l(t)] ds_1 ds_2,$$

$$T_{kl} = c_{klj} e_{ij} - f_{klj} E_l.$$

The dissipation condition (39) that takes here the form

$$\delta E_k \chi \leq 0$$

yields

$$\int_0^\infty \int_0^\infty K_{kl}(s_1, s_2) \left( \frac{d}{ds_2} E_l(t - s_2) \right) [E_k(t - s_1) - E_k(t)] ds_1 ds_2 \leq 0.$$
We introduce
\[ L_{kl}(s_1, s_2) = \int_{s_1}^{\infty} K_{kl}(s_3, s_2) \, ds_3 \] (77)
and represent (73) as
\[ P_k = f_{kij} e_{ij} + e_{kl} E_i - \int L_{kl}(0, s) \left[ E_i(t - s) - E_i(t) \right] \, ds, \] (78)
while (76) may be written as
\[ \int_{0}^{\infty} \int_{0}^{\infty} L_{kl}(s_1, s_2) \dot{E}_i(t - s_1) \dot{E}_i(t - s_2) \, ds_1 \, ds_2 \leq 0. \] (79)

The inequality (79) holds for all \( t \).

To interpret the meaning of (76), we write it in the form
\[
I(t) = \int_{0}^{\infty} \int_{0}^{\infty} K_{kl}(s_1, s_2) \frac{d}{ds_2} E_i(t - s_2) \left[ E_k(t - s_1) - E_k(t) \right] \, ds_1 \, ds_2
\]
\[ = \frac{d}{dt} \left( \frac{1}{2} \int_{0}^{\infty} \int_{0}^{\infty} K_{kl}(s_1, s_2) E_i(t - s_2) E_k(t - s_1) + \right. \]
\[ - \int_{0}^{\infty} \int_{0}^{\infty} K_{kl}(s_1, s_2) E_i(t - s_2) E_k(t) \right) + \]
\[ - \int_{0}^{\infty} \int_{0}^{\infty} K_{kl}(s_1, s_2) E_i(t - s_2) \dot{E}_k(t) \leq 0. \] (80)

We now integrate \( I(t) \) over a closed cycle starting from the virgin state. We find
\[
\int_{-\infty}^{\infty} I(t) \, dt = \frac{1}{2} \int_{0}^{\infty} \int_{0}^{\infty} K_{kl}(s_1, s_2) E_i(t - s_2) E_k(t - s_1) \, ds_1 \, ds_2 \bigg|_{r=-\infty}^{r=\infty}
\]
\[ - \int_{0}^{\infty} \int_{0}^{\infty} K_{kl}(s_1, s_2) E_i(t - s_2) E_k(t) \, ds_1 \, ds_2 \bigg|_{r=-\infty}^{r=\infty} + \]
\[ - \int \dot{E}_k(t) \, dt \int_{0}^{\infty} \int_{0}^{\infty} K_{kl}(s_1, s_2) E_i(t - s_2) \, ds_1 \, ds_2 \leq 0. \] (81)
The first four terms at the right-hand side of (81) are equal to zero. For $t = -\infty$ this is trivial, for $t = +\infty$ it is a consequence of

$$E = 0 \quad \text{for} \quad t > t_2 > 0,$$

$$K_{kl}(s_1, s_2) \to 0 \quad \text{for} \quad s_1 \to \infty \text{ or } s_2 \to \infty.$$  

(82)

It appears that we have for this kind of processes

$$\int_{-\infty}^{\infty} I(t) \, dt = -\int_{-\infty}^{\infty} \int L_{kl}(0, s) \, E_l(t - s) \, \hat{E}_k(t) \, ds \, dt.$$  

(83)

We now go back to the inequality (63). We have

$$\int_{-\infty}^{\infty} P_k \, \hat{E}_k \, dt - \int_{-\infty}^{\infty} T_{kl} \, v_{k,l} \, dt = \int_{-\infty}^{\infty} f_{kij} \, \frac{d}{dt} (e_{ij} E_k) \, dt +$$

$$+ \frac{1}{2} \int_{-\infty}^{\infty} \varepsilon_{kl} \, \frac{d}{dt} (E_k E_l) \, dt - \frac{1}{2} \int_{-\infty}^{\infty} c_{ijkl} \, \frac{d}{dt} (e_{ij} e_{kl}) \, dt +$$

$$- \int_{-\infty}^{\infty} \hat{E}_k \, dt \int_{0}^{0} \int K_{kl}(s_1, s_2) \, [E_l(t - s_2) - E_l(t)] \, ds_1 \, ds_2 \leq 0.$$  

(84)

For the closed thermodynamic process this becomes

$$\int_{-\infty}^{\infty} P_k \, \hat{E}_k \, dt - \int_{-\infty}^{\infty} T_{kl} \, v_{k,l} \, dt = \int_{-\infty}^{\infty} I(t) \, dt \leq 0.$$  

(85)

We represent the constitutive equation (78) in another form. We introduce

$$g_{kl}(s) = \int_{-\infty}^{\infty} L_{kl}(0, p) \, dp$$  

(86)

and with this function (78) becomes

$$P_k = f_{kij} e_{ij} + \varepsilon_{kl} E_l + \int_{0}^{0} \hat{g}_{kl}(s) \, [E_l(t - s) - E_l(t)] \, ds.$$  

(87)
From (87) some other expressions for $P_k$ may be obtained:

$$
P_k = f_{kij} e_{ij} + \left[ \varepsilon_{kl} + g_{kl}(0) \right] E_l + \int_0^\infty g_{kl}(s) E_l(t - s) \, ds,
$$

$$
P_k = f_{kij} e_{ij} + \varepsilon_{kl} E_l + \int_0^\infty g_{kl}(s) \dot{E}_l(t - s) \, ds,
$$

(88)

$$
P_k = f_{kij} e_{ij} + \varepsilon_{kl} E_l + \int_{-\infty}^t g_{kl}(t - s) \dot{E}_l(s) \, ds.
$$

It can easily be seen from (87) that for $t \to \infty$ the susceptibility is the symmetric tensor $\varepsilon_{kl}$.

The instantaneous susceptibility is $\varepsilon_{kl} + g_{kl}(0)$. It is the coefficient if $E$ suffers a jump. We show that $g_{kl}(0)$ is a symmetric tensor, negative definite, and that both $\varepsilon_{kl}$ and $\varepsilon_{kl} + g_{kl}(0)$ are positive.

Coleman \(^6\) has given a proof of a theorem, which states that for a viscoelastic material, all total histories ending with given values of $x_k, a$ and $\theta$, that corresponding to constant values of $x_k, a$ and $\theta$ for all times has the least free energy. Physically this means that the free energy tends to its equilibrium value and that this is a minimum. Basic for Coleman's proof is an inequality that corresponds to our (39).

We may state a corresponding theorem for the function $\varrho_0 \chi$. As a consequence of (75) it takes its minimum for constant histories, ending with given values of $e_{ij}$ and $E_k(t)$. Thus we have

$$
\int_0^\infty \int_0^\infty K_{kl}(s_1, s_2) \left[ E_k(t - s_1) - E_k(t) \right] \left[ E_l(t - s_2) - E_l(t) \right] \, ds_1 \, ds_2 \leq 0,
$$

(89)

for all admissible histories. We apply it to the history of fig. 1:

$$
E(t - s) = \begin{cases} E(t), & s = 0, \\ a, & e < s < \infty, \end{cases}
$$

(90)

Fig. 1. History of $E(t)$. 

---

**RELAXATION PHENOMENA IN ELECTRO-MAGNETO-ELASTICITY**

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while for $0 \leq s \leq \epsilon$ the function is smooth. If we let $\epsilon \to 0$, we obtain

$$
\int_0^\infty \int_0^\infty K_{kl}(s_1, s_2) a_k a_i \, ds_1 \, ds_2 = \int_0^\infty L_{kl}(0, s) a_k a_i \, ds = g_{kl}(0) a_k a_i \leq 0,
$$

$a_k$ constant, \hspace{1cm} (91)

the $g_{kl}(0)$ tensor is negative definite. It may easily be seen that $\varrho_0 \chi$ tends to its equilibrium value for $t \to \infty$ under constant continuation of the $E$ history after a finite time.

We now derive an expression for the free energy $\varrho_0 \psi$ by means of

$$
\varrho_0 \psi = \varrho_0 \chi + P_k E_k = \varrho_0 \chi - \varrho_0 \frac{\partial \chi}{\partial E_k}. \hspace{1cm} (92)
$$

We obtain

$$
\varrho_0 \psi = \frac{1}{2} c_{ijkl} e_{lj} e_{kl} + \frac{1}{2} \varepsilon_{kl} E_k E_l + \frac{1}{2} g_{kl}(0) E_k E_l +

- \frac{1}{2} \int_0^\infty \int_0^\infty K_{kl}(s_1, s_2) E_k(t - s_1) E_l(t - s_2) \, ds_1 \, ds_2. \hspace{1cm} (93)
$$

If $E(t - s)$ is a constant history we arrive at the equilibrium value of $\varrho_0 \psi$,

$$
\varrho_0 \psi = \frac{1}{2} c_{ijkl} e_{lj} e_{kl} + \frac{1}{2} \varepsilon_{kl} E_k E_l, \hspace{1cm} (94)
$$

and this has to be positive definite. Thus $\varepsilon_{kl}$ is a positive tensor. Further we see that

$$
\varepsilon_{kl} + g_{kl}(0) = \frac{\partial^2 (\varrho_0 \psi)}{\partial E_k \partial E_l}, \hspace{1cm} (95)
$$

taken with $E(t - s)$ constant. From (95) follows

$$
g_{kl}(0) = g_{lk}(0). \hspace{1cm} (96)
$$

We apply again the history, given by (90). Now (93) becomes

$$
\varrho_0 \psi = \frac{1}{2} c_{ijkl} e_{lj} e_{kl} + \frac{1}{2} \left[ \varepsilon_{kl} + g_{kl}(0) \right] E_k E_l - \frac{1}{2} g_{kl}(0) a_k a_i. \hspace{1cm} (97)
$$

Because $\varrho_0 \psi$ has to be positive definite for all values of $E$ and $a$, we conclude again that $g_{kl}(0)$ is negative definite, while $\varepsilon_{kl} + g_{kl}(0)$ is positive. Note that for sufficiently small values of $|a|$, $\varrho_0 \psi$, according to (97), is smaller than its equilibrium value (94). However, this does not violate the well-known law of physics, because $\varrho_0 \psi$ obtains its minimum for constant histories of $P$. Thus in the expression for $\varrho_0 \psi$, $E_k$ is a “wrong” variable. For the process (90) the polarization $P_k$ becomes for $t \to \infty$

$$
P_k \to f_{klj} e_{lj} + [\varepsilon_{kl} + g_{kl}(0)] E_i - g_{kl}(0) a_i \hspace{1cm} (98)$$
which is smaller for sufficiently small $|a|$ than the equilibrium value of $P_k$ corresponding to (94).

But even expressed in the wrong variable $E_k$, $\varrho_0 \psi$ has to remain positive, because it is here the internal energy of the body, loaded by mechanical and electric forces. If the excess energy with respect to the energy of the natural state ($e_{ij} = E_k = P_k = 0$), which we have taken equal to zero, changes sign, there may be loss of stability. We exclude this from our discussion.

We now derive some other properties of the tensor $g_{kl}(t)$. We write the integral (85) as

$$\int_{-\infty}^{\infty} I(t) \, dt = \int_{-\infty}^{\infty} \int_{0}^{\infty} \dot{g}_{kl}(s) \dot{E}_k(t) \dot{E}_l(t-s) \, ds \leq 0, \quad (99)$$

and apply it to some closed thermodynamic processes. We take (cf. fig. 2)

$$E = (0, 0, 0), \quad t < t_1, \; t > t_2,$$
$$E = (a_1, a_2, a_3), \quad t_1 < t < t_2. \quad (100)$$

![Fig. 2. Path of $E_1(t)$](image)

For this path we find

$$\int_{-\infty}^{\infty} \int_{0}^{\infty} \dot{g}_{kl}(s) \, ds \left[ \delta(t - t_1) - \delta(t - t_2) \right] E_k(t-s) =$$

$$= a_k a_l \int_{0}^{t_2-t_1} \dot{g}_{kl}(s) \, ds \left[ -H(t_2 - t_1 - s) \right] = -a_k a_l \int_{0}^{t_2-t_1} \dot{g}_{kl}(s) \, ds$$

$$= -a_k a_l \left[ g_{kl}(t_2 - t_1) - g_{kl}(0) \right] \leq 0. \quad (101)$$

As $t_2 - t_1$ has any positive value, we have

$$g_{kl}(t) \; a_k \; a_l \geq g_{kl}(0) \; a_k \; a_l, \quad t > 0. \quad (102)$$

In (101), $H(t-s)$ is the Heaviside step function and $\delta(t-t_1)$ is Dirac's delta.
function. Because every process may be built up of elementary processes of the kind of (100), (102) is a general statement.

Another inequality may be found with the aid of (79). We apply it to

\[ E = (0, 0, 0), \quad t < t_0, \]
\[ E = (a_1, a_2, a_3), \quad t \geq t_0, \]

(cf. fig. 3), which is not a closed process. We note that (79) holds for any thermodynamically admissible process. We find

\[
\int_{s_1}^{s_2} ds_1 \int_{s_2}^{s_3} ds_2 L_{k\ell}(s_1, s_2) a_k a_\ell \delta(t - t_0 - s_1) \delta(t - t_0 - s_2) = a_k a_\ell L_{k\ell}(t - t_0, t - t_0) \leq 0. \tag{104}
\]

If we take \( t = t_0 \), we obtain from (104)

\[ -L_{k\ell}(0, 0) a_k a_\ell = \dot{g}_{k\ell}(0) a_k a_\ell \geq 0, \tag{105} \]

from which follows that \( \dot{g}_{k\ell}(0) \) is a positive definite tensor.

It is not possible to derive more conclusions from (79) or its integrated form (85). For a closed thermodynamic process (95) may also be written as

\[
\int_{-\infty}^{\infty} dt \int_{-\infty}^{t} ds g_{k\ell}(s) \dot{E}_k(t) \dot{E}_\ell(t - s) = \int_{-\infty}^{\infty} dt \int_{-\infty}^{t} ds g_{k\ell}(t - s) \dot{E}_k(t) \dot{E}_\ell(s). \tag{106}
\]

To prove complete monotonicity for the one-dimensional retardation function \( g(s) \) we need

\[
\int_{p}^{q} \int_{p}^{q} g(s + t) a(s) a(t) ds \, dt \leq 0, \tag{107}
\]
for all continuous $a$, defined on $[p, q]$ (cf. ref. 7) and this does not follow from the inequalities we have derived. The physical meaning of (107) is that the work done on retraced paths is increased by delay.

It is also impossible to conclude that Onsager's relations

$$g_{kl}(s) = g_{lk}(s)$$

(108)

hold. It is sufficient for (108) that

$$K_{kl}(s_1, s_2) = K_{kl}(s_2, s_1),$$

(109)

but this is not required in the general theory. It may be proved that (108) holds if the work done on every closed path starting from the virgin state is invariant under time reversal 8).

The theory may be applied to the investigation of the relaxation spectrum of dielectrics. Looking for harmonic solutions, we put

$$T_{kl} = \bar{T}_{kl} \exp (i\omega t), \quad P_k = \bar{P}_k \exp (i\omega t), \text{ etc.}$$

(110)

For the amplitudes we now find from (74) and (88)

$$\bar{T}_{kl} = c_{ijkl} \bar{\epsilon}_{ij} - f_{ikl} \bar{E}_i, \quad \bar{P}_k = f_{kij} \bar{\epsilon}_{ij} + \epsilon_{kl} \bar{E}_l + i\omega \int_0^\infty g_{kl}(s) \exp (-i\omega s) \, ds.$$  

(112)

With the aid of the Laplace-transform techniques, we may also state a principle that formulates a one-to-one correspondence of the Laplace transforms of the quantities in this theory with the corresponding ones of the classical theory. The "material constants" here are functions of the Laplace parameter. This technique is well known in the theory of visco-elasticity.

REFERENCES


