

Quadrupole Piezoelectricity

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The crystal bodies without centre of symmetry are electrically polarized under the effect of mechanical loads. The modern theory of this phenomenon, called by Hankel piezoeffect, is developed by Toupin, Mindlin, Eringen, Tiersten etc. [1—4].

Piezoelectricity represents a complicated interaction between electric and mechanical fields and usually for the sake of simplification of the theoretical model it is assumed that:

- the considered body is elastic;
- the electric field is potential;
- the quadrupole moment as well as moments of higher order could be neglected.

These assumptions give satisfactory results in the case of nonorganic crystals which are used in technics. Recently papers appeared, however, offering more precise models of the electromechanical behaviour of the crystals. Thus in papers [2, 5, 6] are considered the polarization gradients and in papers [7, 8] the visco-elastic character of the deformation is taken into account.

Subject of the present paper is the quadrupole theory of piezoelectricity. Parallely with the polarization representing the dipole moment, the effect of the quadrupole moment is considered also. Thus, more precise information about the motion of ions, forming the crystal lattice under mechanical loads and electric fields could be obtained.

1. On the Behaviour of a System of Electrically Charged Particles in a Quasi-Static Electric Field

Let us consider a system of "n" electrically charged particles, representing the ions of a single cell of the studied body. The total force and moment of momentum by means of which the effective electric field acts on the system are:

$$(1.1) \quad F_k = \sum_{l=1}^n q^{(l)} E_k(x_m^{(l)}),$$

$$(1.2) \quad M_k = \sum_{l=1}^n e_{kij} q^{(l)} E_i(x_m^{(l)}) x_j^{(l)}.$$

$q^{(l)}$, $x_m^{(l)}$ and E_m are the electric charge of the l^{th} particle, the coordinates of the l^{th} particle and the effective electric field, respectively.

The work which is done by the electrically charged particles in the effective electric field per unit time is

$$(1.3) \quad W = \sum_{l=1}^n q^{(l)} E_k(x_m^{(l)}) \dot{x}_k^{(l)}.$$

The condition of electroneutrality of each crystal cell is satisfied

$$(1.4) \quad \sum_{l=1}^n q^{(l)} = 0.$$

Dipole Approximation

Let us denote the coordinates of the mass centre of the crystal cell with x_m and with $u_m^{(l)}$ the coordinates of the l^{th} material point in a coordinate system associated with the mass centre. Expanding $E_k(x_m^{(l)})$ in Taylor series in terms of $u_m^{(l)}$ and taking linear approximation with the help of (1.4) for F_k , M_k and W it is obtained

$$(1.5) \quad F_k = E_{k,m}(x) P_m,$$

$$(1.6) \quad M_k = e_{kij} E_i(x) P_j,$$

$$(1.7) \quad W = E_k(x) \frac{d}{dt} P_k,$$

where

$$P_k = \sum_{l=1}^n q^{(l)} u_k^{(l)}$$

is the crystal cell dipole moment.

Quadrupole Approximation

In the case of a quadratic approximation in terms of $u_m^{(l)}$ for F_k , M_k and W it is obtained

$$(1.8) \quad F_k = E_{k,m}(x) P_m + E_{k,ms}(x) P_{ms},$$

$$(1.9) \quad M_k = e_{kij} [E_i(x) P_j + E_{i,m}(x) P_{mj}],$$

$$(1.10) \quad W = E_k(x) \frac{d}{dt} P_k + E_{k,m}(x) \frac{d}{dt} P_{mk},$$

where

$$P_{km} = \frac{1}{2} \sum_{l=1}^n q^{(l)} u_k^{(l)} u_m^{(l)}$$

is the quadrupole moment of the crystal cell.

2. Kinematics

The equation of motion of dielectric body, considered as continuum medium, is

$$(2.1) \quad x_k = \chi_k(X_K, t)$$

where x_k and X_K are Cartesian coordinates by means of which the deformed and undeformed state of the body is parametrized.

The velocity field of the material points is

$$(2.2) \quad v_k = \frac{\partial}{\partial t} \chi_k(X_K, t) |_{X_K}.$$

The deformed state of the dielectric body is described by the deformation gradients

$$(2.3) \quad x_{k,K} = \frac{\partial}{\partial X_K} \chi_k(X_N, t).$$

3. Principles of Conservation

1. Mass conservation:

$$(3.1) \quad \frac{\partial}{\partial t} \varrho + (\varrho v_k)_{,k} = 0$$

ϱ is the mass density.

2. Moment conservation:

$$(3.2) \quad \varrho \frac{d}{dt} v_k = T_{mk,m} + \varrho (E_{k,m} \Pi_m + E_{k,ml} \Pi_{ml}),$$

where T_{mk} , Π_m and Π_{ml} are Cauchy stress tensor, the dipole moment and the quadrupole moment (per unit mass), respectively.

3. Moment of momentum conservation:

$$(3.3) \quad e_{ijk} [T_{jk} + \varrho (E_j \Pi_k + E_{j,m} \Pi_{mk})] = 0.$$

4. Energy conservation:

$$(3.4) \quad \varrho \frac{d}{dt} \varepsilon = T_{km} v_{m,k} + \varrho E_m \frac{d}{dt} \Pi_m + \varrho E_{mk} \frac{d}{dt} \Pi_{mk} + h_{k,k},$$

where $E_{mk} = \frac{1}{2} (E_{m,k} + E_{k,m})$.

ε and h_k are the internal energy density and heat flux.

5. Clausius-Duhem inequality

$$(3.5) \quad \varrho \frac{d}{dt} \eta - \left(\frac{h_k}{\theta} \right)_{,k} \geq 0.$$

η and θ are the entropy density and the thermodynamic temperature, respectively.

6. Maxwell Equations

It is well known that for practically interesting frequencies, the electric field could be considered as potential, i.e.

$$(3.6) \quad e_{ijk} E_{k,j} = 0.$$

In the considered dielectric body the electroneutrality condition is satisfied

$$(3.7) \quad (E_k + 4\pi \varrho \Pi_k)_{,k} = 0, \quad \pi = 3, 14, \dots$$

4. Constitutive Equations

The accounting of the quadrupole moment appears to be especially perspective when the considered dielectric body has a complicated crystal cell and every kind of ion, forming the crystal cell is characterized by

individual motion. In the present paper parallelly with the quadrupole moment, the visco-elastic character of the deformation, characteristic for greater part of the organic dielectrics, will be considered also.

The general form of the proposed constitutive equations is

$$\begin{aligned}
 (4.1) \quad & T_{kl} = T_{KL}(C_{KL}; \Pi_K; \Pi_{KL}; \theta_K; \theta; \alpha^{(m)}) x_{k,K} x_{l,L}, \\
 & E_k = E_K(C_{KL}; \Pi_K; \Pi_{KL}; \theta_K; \theta; \alpha^{(m)}) x_{k,K}, \\
 & E_{kl} = E_{KL}(C_{KL}; \Pi_K; \Pi_{KL}; \theta_K; \theta; \alpha^{(m)}) x_{k,K} x_{l,L}, \\
 & \psi = \psi(C_{KL}; \Pi_K; \Pi_{KL}; \theta_K; \theta; \alpha^{(m)}), \\
 & \eta = \eta(C_{KL}; \Pi_K; \Pi_{KL}; \theta_K; \theta; \alpha^{(m)}), \\
 & h_k = h_K(C_{KL}; \Pi_K; \Pi_{KL}; \theta_K; \theta; \alpha^{(m)}) x_{k,K}, \quad m = 1, 2, \dots, N,
 \end{aligned}$$

where $\psi = \varepsilon - \eta\theta$ is the free energy density.

$$\begin{aligned}
 (4.2) \quad & C_{KL} = x_{k,K} x_{l,L}; \Pi_{KL} = \Pi_{kl} x_{k,K} x_{l,L}, \\
 & \Pi_K = \Pi_k x_{k,K}; \theta_K = \theta_k x_{k,K}
 \end{aligned}$$

$\alpha^{(l)}$, $l = 1, 2, \dots, N$, are the internal variables, responsible for the visco-elastic character of the deformation [9, 10].

$\alpha^{(l)}$ satisfy the evolution equations:

$$(4.3) \quad \frac{d}{dt} \alpha^{(l)} = \varphi^{(l)}(C_{KL}; \Pi_L; \Pi_{KL}; \theta_K; \theta; \alpha^{(m)}), \quad l = 1, 2, \dots, N.$$

With the help of the energy conservation principle (3.4) and relations (4.1)₄ Clausius-Duhem inequality could be presented in the form:

$$\begin{aligned}
 (4.4) \quad & \varrho \left(\frac{\partial \psi}{\partial \theta} + \eta \right) \frac{d\theta}{dt} + \varrho \left(2 \frac{\partial \psi}{\partial C_{KL}} x_{l,K} + \frac{\partial \psi}{\partial \Pi_L} \Pi_l + 2 \frac{\partial \psi}{\partial \Pi_{KL}} \Pi_{lk} x_{k,K} \right. \\
 & \left. + \frac{\partial \psi}{\partial \theta_{,L}} \theta_{,l} - \frac{1}{\varrho} T_{kl} x_{l,K} \right) \frac{d}{dt} x_{l,L} + \varrho \left(\frac{\partial \psi}{\partial \Pi_L} x_{k,L} - E_k \right) \frac{d}{dt} \Pi_k \\
 & + \varrho \left(\frac{\partial \psi}{\partial \Pi_{KL}} x_{k,K} x_{l,L} + E_{kl} \right) \frac{d}{dt} \Pi_{kl} + \frac{\partial \psi}{\partial \theta_{,L}} x_{k,L} \frac{d}{dt} \theta_{,k} \\
 & + \sum_{l=1}^N \frac{\partial \psi}{\partial \alpha^{(l)}} \frac{d\alpha^{(l)}}{dt} - \frac{h_k \theta_{,k}}{\varrho} \leq 0.
 \end{aligned}$$

Since ψ , η , T_{kl} , E_k and E_{kl} do not depend, according to (4.1) on the independent thermodynamic rates

$$\frac{d}{dt} \theta; \frac{d}{dt} x_{k,K}; \frac{d}{dt} \Pi_k; \frac{d}{dt} \Pi_{kl}; \frac{d}{dt} \theta_{,k}$$

then from inequality (4.4) the following relations are obtained:

$$\begin{aligned}
 (4.5) \quad & \frac{\partial \psi}{\partial \theta_{,k}} = 0, \\
 & -\eta = \frac{\partial \psi}{\partial \theta}, \\
 & \frac{1}{\varrho} T_{kl} = 2 \frac{\partial \psi}{\partial C_{KL}} x_{k,K} x_{l,L} + \frac{\partial \psi}{\partial \Pi_{KL}} \Pi_{lm} x_{m,K} x_{k,L} + \frac{\partial \psi}{\partial \Pi_K} \Pi_k x_{l,K}, \\
 & E_k = \frac{\partial \psi}{\partial \Pi_L} x_{k,L},
 \end{aligned}$$

$$E_{kl} = \frac{\partial \psi}{\partial \Pi_{KL}} x_{k,K} x_{l,L},$$

$$\sum_{l=1}^N \frac{\partial \psi}{\partial \alpha^{(l)}} \frac{d\alpha^{(l)}}{dt} - \frac{h_k \theta_{,k}}{\theta} \leq 0.$$

Assuming that the heat conductivity processes do not depend on the internal variables $\alpha^{(l)}$ inequality (4.5)₆ is separated into the following two inequalities:

$$(4.6) \quad \sum_{l=1}^N \frac{\partial \psi}{\partial \alpha^{(l)}} \frac{d\alpha^{(l)}}{dt} \leq 0; \quad h_k \theta_{,k} \geq 0.$$

With the help of (4.5)_{4,5} equation (4.5)₃ could be written in the following equivalent form

$$\frac{1}{\varrho} T_{kl} = 2 \frac{\partial \psi}{\partial C_{KL}} x_{k,K} x_{l,L} + E_l \Pi_k + E_{km} \Pi_{ml}.$$

5. Linear Theory

In the case of small deformation and weak electric field, the free energy density could be presented as a quadratic function of the infinitesimal strain tensor ε_{ij} , the temperature θ , the polarization Π_k and the quadrupole moment Π_{kl} . If in its reference state the body is undeformed and nonpolarized, the constitutive equations, following from (4.5) and the assumptions done, are:

$$(5.1) \quad \begin{aligned} \frac{1}{\varrho} T_{kl} &= A_{kl ij} \varepsilon_{ij} + A_{kl i} \Pi_i + C_{kl ij} (\Pi_{ij} - \Pi_{ij}^0) + B_{kl} (\theta - \theta^0) + \sum_{m=1}^N C_{kl}^{(m)} \alpha^{(m)}, \\ -\eta &= B_{ij} \varepsilon_{ij} + C_i \Pi_i + D_{ij} (\Pi_{ij} - \Pi_{ij}^0) + K (\theta - \theta^0) + \sum_{m=1}^N H^{(m)} \alpha^{(m)}, \\ E_k &= A_{ijk} \varepsilon_{ij} + A_{kl} \Pi_l + D_{kij} (\Pi_{ij} - \Pi_{ij}^0) + C_k (\theta - \theta^0) + \sum_{m=1}^N H_k^{(m)} \alpha^{(m)}, \\ E_{kl} &= C_{kl ij} \varepsilon_{ij} + D_{kl i} \Pi_i + B_{kl ij} (\Pi_{ij} - \Pi_{ij}^0) + D_{kl} (\theta - \theta^0) + \sum_{m=1}^N C_{kl}^{(m)} \alpha^{(m)}, \end{aligned}$$

where

$$(5.2) \quad \begin{aligned} A_{kl ij} &= A_{l k ij} = A_{ij kl}, \\ A_{kl i} &= A_{l ki}, \\ B_{kl ij} &= B_{l k ij} = B_{ij kl}, \\ C_{kl i} &= C_{l k ij} = C_{ij kl}, \\ B_{ij} &= B_{ji}; D_{kl} = D_{lk}; C_{kl}^{(m)} = C_{lk}^{(m)} \end{aligned}$$

are constant tensors, characterizing the material medium.

$H_k^{(l)}$ are constant material vectors and $H^{(l)}$ — material constants. Π_{ij}^0 and θ^0 are the quadrupole moment and the temperature in the reference state.

From (4.1) and (4.6) the following equalities are obtained in the linear variant

$$(5.3) \quad \frac{d}{dt} \alpha^{(l)} = \sum_{m=1}^N L^{(lm)} \frac{\partial \eta}{\partial \alpha^{(m)}}; \quad h_k = \lambda_{km} \theta_{,m},$$

where the matrix $L^{(lm)}$ is negatively defined, and the tensor λ_{km} — positively.

The solution of the evolution equation (5.3) has the form:

$$(5.4) \quad \alpha^{(l)} = \int_{t'=0}^{t'=t} {}^{(l)} [\varepsilon_{ij}(t'); \Pi_i(t'); \Pi_{kl}(t'); \theta(t')],$$

where $\int_{t'=0}^{t'=t} {}^{(l)}$ are linear functionals of the variables $\varepsilon_{ij}(t')$, $\Pi_i(t')$, $\Pi_{kl}(t')$, and $\theta(t')$.

6. Isotropic elastic dielectrics

If the considered medium is elastic and isotropic

$$A_{kli}, C_{ij}^{(l)}, C_i, H_k^{(l)}, H^{(l)}, D_{kij}$$

are equal to zero.

The constitutive equations (5.1) obtain the form:

$$(6.1) \quad \begin{aligned} \frac{1}{\rho} T_{kl} &= A_{klj} \varepsilon_{ij} + C_{klj} (\Pi_{ij} - \Pi_{ij}^0) + B_{ij} (\theta - \theta^0), \\ -\eta &= B_{ij} \varepsilon_{ij} + D_{ij} (\Pi_{ij} - \Pi_{ij}^0) + K (\theta - \theta^0), \\ E_k &= A_{kl} \Pi_l, \\ E_{kl} &= C_{klj} \varepsilon_{ij} + B_{klj} (\Pi_{ij} - \Pi_{ij}^0) + D_{kl} (\theta - \theta^0), \end{aligned}$$

where the tensor coefficients on the right side of the equations have the presentation:

$$(6.2) \quad \begin{aligned} A_{klj} &= a^{(1)} \delta_{ij} \delta_{kl} + a^{(2)} (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}), \\ B_{klj} &= b^{(1)} \delta_{ij} \delta_{kl} + b^{(2)} (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}), \\ C_{klj} &= c^{(1)} \delta_{ij} \delta_{kl} + c^{(2)} (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}), \\ B_{ij} &= b \delta_{ij}; A_{kl} = a \delta_{kl}; D_{ij} = d \delta_{ij}. \end{aligned}$$

$a, b, d, a^{(1)}, a^{(2)}, b^{(1)}, b^{(2)}, c^{(1)}$ and $c^{(2)}$ are material constants.

It could be seen from Equations (6.1) that in the quadrupole variant of piezoelectricity the electromechanical interaction exists even in isotropic bodies.

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