HAL
open science

Lagrangian effective material constants for the modellng of thermal behavior of acoustic waves in piezoelectric crystals I. Theory

Bernard Dulmet, Roger Bourquin

To cite this version:

Bernard Dulmet, Roger Bourquin. Lagrangian effective material constants for the modellng of thermal behavior of acoustic waves in piezoelectric crystals I. Theory. Journal of the Acoustical Society of America, 2001, 110 (4), pp.1792-1799. 10.1121/1.1391250 . hal-00250943

HAL Id: hal-00250943

https://hal.science/hal-00250943v1

Submitted on 12 Mar 2008

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Lagrangian effective material constants for the modeling of thermal behavior of acoustic waves in piezoelectric crystals. I. Theory

Bernard Dulmet[a)] and Roger Bourquin
Laboratoire de Chronométrie Electronique et Piézoélectricité, École Nationale Supérieure d'Ingénieurs de Micromécanique et des Microtechniques, 26 Chemin de l'Épitaphe, 25000 Besançon, France

(Received 5 June 2000; accepted for publication 18 June 2001)

After some necessary recalls on the nonlinear theory of thermoelectroelasticity in piezoelectric crystals, asserting the need of constitutive equations which derive from a rotationally invariant energy function, this paper presents the governing equations for a small vibration superimposed on a bias originated by a slow and homogeneous temperature variation from a well-defined reference state. Thereafter, the authors define the effective coefficients appearing in the linearized incremental dynamic balance equations for linear momentum and electrical charge in Lagrange configuration, not omitting associated boundary conditions. The main features of these coefficients are discussed and explicit relations with more conventionally defined coefficients are given. Determination of numerical values of the proposed effective coefficients and examples of their use in the higher order modeling of static frequency–temperature characteristics of either bulk acoustic wave or surface acoustic wave devices are given in a companion paper. © *2001 Acoustical Society of America.*
[DOI: 10.1121/1.1391250]

PACS numbers: 43.25.Dc [MFH]

I. INTRODUCTION

The subject of this paper is to present the analysis of infinitesimal vibrations in a piezoelectric crystal which can freely expand in all space directions upon submission to slow and homogeneous temperature variations. The classical treatment[1] implicitly uses the coordinates of a material point of the crystal in the slowly varying state at the current temperature prior to any vibration. In this description, the dimensions, mass density, and orientation of the solid as well as the properties of the material vary with temperature. This leads to tedious computations to rigorously fit the frequency–temperature characteristic of a piezoelectric resonator by a high order expansion while correctly considering the effect of temperature on its size and crystalline orientation. This can be avoided by using the Lagrangian description in which the field variables are the coordinates of a material point in a reference state defined at a fixed temperature. Since those coordinates are fixed and known, the boundary conditions are expressed on surfaces which do not depend on temperature. Similarly, since the equations of motion are mapped onto the fixed volume of the body in its reference state, the mass density appearing in these equations is constant and the coefficients which describe the material are the only parameters which may vary with temperature. As a consequence, useful simplifications occur for the treatment of frequency–temperature characteristics of resonators with rather complicated shapes, such as radius of contour, gratings, and so on.

This introductory presentation of the use of the Lagrangian description for a particular purpose is indeed somewhat practical. Nevertheless, this treatment also has more theoretical justifications, since it derives from the rotationally invariant and general nonlinear theory established in the case of piezoelectric solids submitted to finite deformations.[2,3] The acceptance of this theory is quite general for the study of force–frequency effects as well as for the study of frequency shifts arising from thermal transient biasing states or from applying static electric fields,[4] but is not so widespread for the study of simple biasing states such as the one arising from an homogeneous temperature variation. More specifically, the presented work defines and makes use of *effective material constants* which are the expansion coefficients of the dynamic part of the Piola–Kirchhoff stress tensor versus the dynamic part of the total strain and of the rotationally invariant electric vector expressed in a Lagragian description. The present paper (paper I) is devoted to the definition of the coefficients while the companion paper (paper II) is devoted to the calculation of their numerical values and their use in predicting the frequency–temperature dependence of commonly encountered kinds of resonators (bulk and surface acoustic waves).

As usual in this kind of problem, we shall consider three states of a solid described by three sets of respective coordinates, as illustrated in Fig. 1:

(1) a *natural* (or *reference*) state, at a fixed temperature Θ_0, in which the coordinates of a material point of the solid are X_M,
(2) an *intermediate* state prior to vibration at a current homogeneous temperature Θ, the coordinates of the material points being ξ_α,
(3) a *final* state when an adiabatic vibration is superimposed on the initial state, the associated coordinates being y_j.

This convention leads to a straightforward writing of equations provided the derivatives with respect to space co-

[a)]Electronic mail: bernard.dulmet@ens2m.fr

 © 2001 Acoustical Society of America

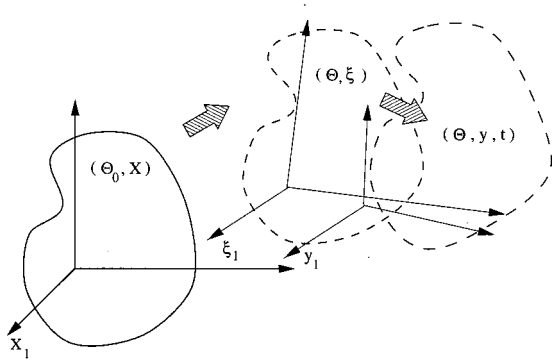

FIG. 1. A solid in successive states.

ordinates are denoted in the following manner:

$$y_{k,L} \equiv \frac{\partial y_k}{\partial X_L}, \quad \xi_{\alpha,L} \equiv \frac{\partial \xi_\alpha}{\partial X_L}, \quad X_{M,j} \equiv \frac{\partial X_M}{\partial y_j}. \tag{1}$$

In this paper we only use Cartesian coordinates and automatic summation over repeated indices is implicitly admitted. The position at time t of a material point referred to by its material coordinates X_L in the natural state is

$$y_j = (\xi_\alpha(X_L) + u_\alpha(X_L,t))\,\delta_{j\alpha}, \tag{2}$$

where u_α is the dynamic displacement and the Kronecker symbol $\delta_{j\alpha}$ is introduced to handle index correspondences from the intermediate state to final state.

II. BASIC EQUATIONS

Although most of the material presented in this section conforms to the general theory of thermoelectroelasticity of Ref. 2, the subsequent treatment involves precise definitions of notations and terms requiring a proper understanding, which makes the recall of this information necessary for our purpose.

We temporarily postpone any use of the intermediate coordinates to address the issue of Lagrange vs Euler representation. The well-known classical field equations expressing the conservation of linear momentum and electric charge are written in the final state:

$$(T_{ij}^M + T_{ij}^E)_{,i} = \rho \frac{d^2 y_j}{dt^2}, \quad D_{i,i} = 0, \tag{3}$$

where $\mathbf{D}$ denotes the electric displacement and ρ the mass density. $\mathbf{T}^M$ is the pure mechanical stress tensor which refers to the traction exerted onto a surface element taken in its *final* (deformed) state and $\mathbf{T}^E$ is Maxwell's electrostatic tensor:

$$T_{ij}^E = D_i E_j - \tfrac{1}{2}\varepsilon_0 E_k E_k \delta_{ij}, \tag{4}$$

where $\mathbf{E}$ is the macroscopic electric field. All quantities in Eq. (3) are functions of the final coordinates y_k and the boundary conditions are expressed on the external surface taken in its final state S:

$$\begin{aligned} n_i[T_{ij}^M + T_{ij}^E] &= 0 \\ n_i[D_i] &= \sigma \end{aligned} \quad \text{on } S. \tag{5}$$

σ is the electric charge density and [ ] refers to the discontinuity of an enclosed quantity across the surface the orientation of which is given by the outward unit normal $\mathbf{n}$.

The balance equation for the angular momentum is fulfilled by

$$e_{kij}(P_i E_j + T_{ij}^M) = 0, \quad k = 1,...,3, \tag{6}$$

where $\mathbf{P}$ denotes the polarization related to the field and electric displacement by the classical relation:

$$D_i = \varepsilon_0 E_i + P_i, \quad i = 1,...,3, \tag{7}$$

and e_{kij} denotes Levi-Civita's tensor which takes the value $+1$ if (k, i, j) form a circular permutation of (1, 2, 3), and the value -1 if (k, i, j) form a circular permutation of (3, 2, 1). e_{kij} vanishes in all other cases. Then, Eq. (6) is equivalent to

$$T_{ij}^{AM} = \tfrac{1}{2}(E_i P_i - P_i E_j), \tag{8}$$

where T_{ij}^{AM} denotes the antisymmetric part of the mechanical stress tensor $T_{ij}^M - T_{ji}^M$. It should be observed that all the above-mentioned equations use actual coordinates y_i as field variables. At this point, since the number of equations in (3), (4), (6), and (7) is far smaller than the number of unknowns, it is required to introduce the so-called constitutive equations. In accordance with the pertinent choice of complementary sets of dependent and independent variables for the considered problem, they express the intrinsic properties of matter as relations between those sets. More precisely, all material constants appear as partial derivatives of a thermodynamic potential with respect to the independent variables of the considered problem. A natural way of introducing an energy function consists in writing the balance between the storage of kinetic, internal, and electric field induced energies in the volume of the body, and the work performed on its boundaries:

$$\frac{d}{dt}\int_V \rho\left(\frac{1}{2}v_i v_i + \mathcal{E}\right) dV = \int_S (t_i^M v_i - n_i q_i)\, dS + \int_V \zeta\, dV, \tag{9}$$

where $\mathcal{E}$ is the internal energy per unit mass, $\mathbf{t}^M$ is the local mechanical traction applied to the boundary, $\mathbf{v} \equiv d\mathbf{y}/dt, \mathbf{q}$ is the outwardly orientated heat flow, and ζ is the rate of energy production under the action of applied electric field $\mathbf{E}$. This expression of the first principle of thermodynamics leads to the following differential form:

$$\rho \frac{d\mathcal{E}}{dt} = T_{ij}^M v_{j,i} + \rho E_i \frac{d\Pi_i}{dt} - q_{i,i}, \tag{10}$$

where $\mathbf{\Pi}$ indicates the polarization per mass unit, which is related to the polarization appearing in Eq. (7) in the following manner:

$$P_i = \rho \Pi_i. \tag{11}$$

It can be shown that in the restricted case of nondissipative behavior, the second principle of thermodynamics reduces to

$$\rho \Theta \frac{d\eta}{dt} = -q_{i,i}, \tag{12}$$

where Θ denotes absolute temperature and η is the entropy per mass unit. This result allows for easy substitution of the entropy instead of heat flow into Eq. (10), which can thus be written in two mathematically equivalent differential forms:

$$\rho\frac{d\mathcal{E}}{dt}=X_{L,i}T_{ij}^{M}\frac{dy_{i,L}}{dt}+\rho E_i\frac{d\Pi_i}{dt}+\rho\Theta\frac{d\eta}{dt},$$
$$\rho_0\frac{d\mathcal{E}}{dt}=K_{Li}^{M}\frac{dy_{i,L}}{dt}+\rho_0 E_i\frac{d\Pi_i}{dt}+\rho_0\Theta\frac{d\eta}{dt}. \tag{13}$$

Equivalence between both forms comes from the conservation of mass and the expression of Piola Kirchhoff's asymmetric mechanical tensor $\mathbf{K}^M$:[5]

$$\rho_0=\rho J,\quad J=|y_{i,L}|,\quad K_{Lj}^{M}=JX_{L,i}T_{ij}^{M}. \tag{14}$$

The above-given formula for $\mathbf{K}^M$ derives from its definition expressing the conservation of a surface traction force $\mathbf{t}^M$ along the surface representation:

$$t_j^M=T_{ij}^{M}n_i\,dS=K_{Lj}^{M}n_L^0\,dS_0, \tag{15}$$

where we denote by dS_0 the elementary surface consisting in the whole set of material points in the natural state which become the actual elementary surface dS in the deformed state. n_L^0 denotes a component of the unit normal $\mathbf{n}^0$ in the natural state, corresponding to the actual normal $\mathbf{n}$ (component n_i with notations of this paper). Equation (13b) establishes the internal energy as a function of a well-defined set of independent variables:

$$\mathcal{E}\equiv\mathcal{E}(y_{i,L};\Pi_i;\eta), \tag{16}$$

whereas the corresponding dependent variables are defined by

$$T_{ij}^{M}=\rho y_{i,L}\frac{\partial\mathcal{E}}{\partial y_{j,L}}\bigg)_{\Pi_k,\eta},\quad E_k=\frac{\partial\mathcal{E}}{\partial\Pi_k}\bigg)_{y_{i,j},\eta},\quad \theta=\frac{\partial\mathcal{E}}{\partial\eta}\bigg)_{y_{i,j},\Pi_i}. \tag{17}$$

Nevertheless, a consistent derivation of the constitutive equations requires the proper energy function for the class of considered problems to be an invariant quantity with respect to an arbitrary rotation of either the body itself or the frame used to refer the y_i coordinates.[2,3] This property is not ensured by the above-mentioned writing of internal energy. A mathematical theorem due to Cauchy[6] on the invariance of scalar functions of vectors shows that this property will be granted if $\mathcal{E}$ can be reduced to some combination of scalar products of the four vectors Π, $\partial\mathbf{y}/\partial X_1,\partial\mathbf{y}/\partial X_2,\partial\mathbf{y}/\partial X_3$ and of their determinants. Two alternative forms of $\mathcal{E}$ granting the rotational invariance are given in the following:

$$\mathcal{E}\equiv\mathcal{E}(C_{LM};\mathcal{P}_L;\eta),$$
$$C_{LM}=y_{i,L}y_{i,M}, \tag{18}$$
$$\mathcal{P}_L=y_{i,L}\Pi_i,$$

and

$$\mathcal{E}\equiv\mathcal{E}(E_{LM};p_I;\eta),$$
$$E_{LM}=y_{i,L}y_{i,M}-\delta_{LM}, \tag{19}$$
$$p_K=JX_{K,i}P_i\equiv\rho_0X_{K,i}\Pi_i.$$

In this paper, we follow the approach previously suggested by Tiersten,[2] implying the choice of the latter set (19). Thus, the differential form of internal energy per mass unit becomes

$$\rho_0\frac{d\mathcal{E}}{dt}=\tau_{LM}\frac{dE_{LM}}{dt}+W_L\frac{dp_L}{dt}+\rho_0\Theta\frac{d\eta}{dt} \tag{20}$$

instead of Eq. (13b) and the dependent variables of the problem are then defined by

$$\tau_{LM}=\rho_0\frac{\partial\mathcal{E}}{\partial E_{Lm}}\bigg)_{p_K,\eta},\quad W_K=\rho_0\frac{\partial\mathcal{E}}{\partial p_K}\bigg)_{E_{LM},\eta},$$
$$\Theta=\frac{\partial\mathcal{E}}{\partial\eta}\bigg)_{E_{LM},p_K}. \tag{21}$$

One can show, with help of rules for chain derivatives and related identities, that the above-mentioned formulas imply

$$T_{ij}^{M}=\frac{1}{J}y_{i,L}\tau_{LM}y_{j,M}-P_iE_j,$$
$$W_L=y_{k,L}E_k^M\equiv-\varphi_{,L}, \tag{22}$$

where φ denotes the electrostatic potential. Equation (22a) shows that the balance of kinetic momentum is identically satisfied:

$$T_{ij}^{AM}\equiv T_{ij}^{M}-T_{ji}^{M}=E_iP_j-E_jP_i \tag{23}$$

since the symmetry of τ_{LM} derives from relation (21a) and the symmetry of elastic deformations E_{KL}. It can readily be observed that, from Eqs. (4) and (7), the antisymmetric part of the Maxwell electostatic tensor is exactly the opposite of the antisymmetric part of the purely mechanical stress tensor:

$$T_{ij}^{AE}\equiv T_{ij}^{E}-T_{ji}^{E}=E_jP_i-E_iP_j\equiv-T_{ij}^{AM}. \tag{24}$$

Since it is known that the internal energy is not the most pertinent thermodynamic potential to formulate resonator problems, we conform to Tiersten's approach and we take Gibbs' electric enthalpy χ as the preferred thermodynamic potential for the considered class of problems, according to

$$\rho_0\chi=\rho_0\mathcal{E}-W_Lp_L-\rho_0\Theta\eta, \tag{25}$$

which establishes χ as a function of the tensor of deformation E_{LM}, the rotationally invariant electric vector W_L, and the absolute temperature Θ:

$$\chi\equiv\chi(E_{LM};W_L;\Theta). \tag{26}$$

If the variation of the independent variables of the considered problem from the given reference state to the actual state of the crystal remain finite, the corresponding variation of the thermodynamic potential χ can be represented by a generalized Taylor's expansion in terms of the independent variables $(E_{LM};W_L;\Theta)$. The coefficients of this development define the material constants characterizing the crystal, and using χ energy function defines the following quantities as "thermodynamic" dependent variables:

$$t_{KL}=\rho_0\frac{\partial\chi}{\partial E_{KL}}\Big)_{W,\Theta},\quad p_K=-\rho_0\frac{\partial\chi}{\partial W_K}\Big)_{E,\Theta},$$
$$\eta=-\frac{\partial\chi}{\partial\Theta}\Big)_{E,W}. \tag{27}$$

Although the thermodynamic stress t_{KL} and the polarization p_K differ from the usual mechanical stress and the electric induction, they are clearly connected with them. Above-mentioned relations (7), (21)–(22), and (25) are consistent with the following simple relations between the mechanical stress tensor, the electric displacement, i.e., the dependent variables of the fundamental balance equations (3), and partial derivatives of Gibbs' function:[2]

$$T^M_{ij}=\rho y_{i,M}\frac{\partial\chi}{\partial E_{MN}}\Big)_{W,\Theta}\quad y_{j,N}+\rho y_{i,M}\frac{\partial\chi}{\partial W_M}\Big)_{E,\Theta}X_{L,j}W_L,$$
$$D_i=\rho\varepsilon_0X_{M,i}W_M-y_{i,M}\frac{\partial\chi}{\partial W_M}\Big)_{E,\Theta}. \tag{28}$$

The fundamental elastic constants defined as coefficients of Taylor's expansion of $\chi(E_{LM};W_l;\Theta)$ are isothermal elastic constants. Thus, for the sake of simplicity, we shall systematically omit the notation $)_{W,\Theta}$ and $)_{E,\Theta}$ for partial derivatives of any energy function appearing in the followings.

Although conservation equations (3) and boundary conditions (5) are absolutely correct by themselves, they are not well matched with the fundamental constitutive equations which can be established by the above-mentioned expansion of χ in terms of material constants and independent variables, mainly because E_{LM} and W_L are mapped onto the coordinates of the natural state. Furthermore, recalling our introductory argument about the boundary conditions, it is also more convenient to map the balance equations onto the coordinates of the natural state because the dimensions and the orientation of the finite body in crystallographic axes are well known in this state. Thus we reformulate the problem by using new quantities, which are the *total* asymmetric Piola–Kirchhoff stress tensor $\mathbf{K}$ and the material electric displacement $\mathbf{\Delta}$:[7]

$$K_{Lj}=JX_{L,i}(T^M_{ij}+T^E_{ij})=K^M_{Lj}+K^E_{Lj},$$
$$\Delta_L=JX_{L,i}D_i. \tag{29}$$

They are related to the usual stress and electric displacement by expressing the conservation of the polarization-induced surface force $\mathbf{t}^E$ and electric charge σ per unit surface:

$$t^E_j=T^E_{ij}n_i\,dS=K^E_{Lj}n^0_L\,dS_0,$$
$$\sigma=D_in_i\,dS=\Delta_Ln^0_L\,dS_0. \tag{30}$$

Substitution of Eq. (28) into Eq. (29), together with the mutual cancellation of antisymmetric parts of $\mathbf{T}^E$ and $\mathbf{T}^M$ gives the following expression for K_{Lj} and Δ_L:

$$K_{Lj}=\rho_0y_{i,M}\frac{\partial\chi}{\partial E_{LM}}+\varepsilon_0J(X_{L,i}X_{M,i}X_{Nj}-\tfrac{1}{2}X_{L,j}X_{M,k}X_{N,k})W_MW_N,$$
$$\Delta_L=\varepsilon_0JX_{L,i}X_{M,i}W_M-\rho_0\frac{\partial\chi}{\partial W_L}. \tag{31}$$

One must note that the partial derivatives of χ in these relations are evaluated in the *final* state. The stress equation of motion and charge equation become

$$K_{Lj,L}(X_M;\Theta;t)=\rho_0\frac{d^2y_j(X_M;\Theta;t)}{dt^2},$$
$$\Delta_{L,L}(X_M;\Theta;t)=0. \tag{32}$$

The associated boundary conditions at the interface between the crystal and a perfect conductor are the following when mapped in the *natural* state:

$$\begin{matrix}N_L[K_{Lj}]=0\\ N_L[\Delta_L]=\sigma\end{matrix}\quad\text{on } S_0. \tag{33}$$

III. SMALL VIBRATION SUPERIMPOSED ON A HOMOGENEOUS THERMAL BIAS

We consider the propagation of an elastic wave in a piezoelectric medium subject to the static deformation arising from a slow and uniform temperature change. Since both static and dynamic problems are superimposed in the above-given formulation, tractable equations can readily be obtained by separating the static and the dynamic parts of the problem. Thus, the approach in this section exhibits similarities to the one of Ref. 4, which considers a quite different biasing state (static electric field). Here, since we restrict our interest to quasistatic and homogeneous temperature variation, explicitly solving the static problem is not expected to generate many difficulties.

The infinitesimal dynamic increment u_α of the mechanical displacement is defined by Eq (2). Since the transformation $X_L\rightarrow\xi_\alpha$ only depends on thermal expansion, we can arbitrarily map the dynamic displacement either as $u_\alpha(\xi_\gamma;t)$ or as $u_\alpha(X_L;\Theta-\Theta_0;t)$. The first choice corresponds to the approach followed by Bechmann, Ballato, and Lukaszek[1] and later by James,[8] for the practical determination of thermal derivatives of elastic constants of quartz. Temperature is then considered as a simple parameter acting on all material constants and on the dimensions of crystal through thermal expansion. Thus, it is also necessary to properly appreciate the small temperature-dependent change of crystalline orientation of an arbitrarily oriented device, which appears in the intermediate description due to the anisotropy of thermal expansion.[9] This effect is rather small, but is not negligible when the velocity of acoustic wave varies quickly with the direction of propagation. It is omitted in Refs. 1 and 8, but it is automatically taken into account in the present analysis which conforms to the second choice of mapping. Now, substituting Eq. (2) in Eq. (19b) with help of the chain rules for derivatives:

$$y_{j,M}=y_{j,\alpha}\xi_{\alpha,M}=\xi_{\alpha,M}\delta_{j,\alpha}+u_{\alpha,M}\delta_{j,\alpha}. \tag{34}$$

One can split the total deformation in two parts, the static one and a small dynamic increment:

$$E_{LM}=\overline{E}_{LM}+\tilde{E}_{LM},$$
$$\overline{E}_{LM}=\tfrac{1}{2}(\xi_{\alpha,L}\xi_{\alpha,M}-\delta_{LM}), \tag{35}$$
$$\tilde{E}_{LM}=\tfrac{1}{2}(\xi_{\alpha,L}u_{\alpha,M}+\xi_{\alpha,M}u_{\alpha,L}+u_{\alpha,L}u_{\alpha,M}).$$

Dynamic increments of stress tensor $\tilde{K}_{Lj}$, material electric induction $\tilde{\Delta}_L$, and electric vector $\tilde{W}_L$ are defined in the same way:

$$K_{Lj}=\overline{K}_{Lj}(\Theta)+\tilde{K}_{Lj}(u_\alpha;\tilde{W}_M),$$
$$\Delta_L=\overline{\Delta}_L(\Theta)+\tilde{\Delta}_L(u_\alpha;\tilde{W}_M), \tag{36}$$
$$W_L=\overline{W}_L(\Theta)+\tilde{W}_L(t).$$

Assuming a null electric field and a uniform temperature distribution in the intermediate state, and considering a nonpyroelectric material free to expand without external constraint, we have

$$\overline{K}_{Lj}=0,\quad \overline{W}_L=0,\quad \overline{\Delta}_L=0. \tag{37}$$

Since we are only interested in infinitesimal dynamic fields, we may use the following Taylor's expansions of partial derivatives of χ:

$$\begin{aligned}\frac{\partial\chi}{\partial E_{LM}}&=\left.\frac{\partial\chi}{\partial E_{LM}}\right)_1+\left.\frac{\partial^2\chi}{\partial E_{LM}\partial E_{NP}}\right)_1\tilde{E}_{NP}+\left.\frac{\partial^2\chi}{\partial E_{LM}\partial W_K}\right)_1\tilde{W}_K\\&\quad+\left.\frac{\partial^2\chi}{\partial E_{LM}\partial\Theta}\right)_1\Delta\tilde{\Theta}+\cdots,\\\frac{\partial\chi}{\partial W_K}&=\left.\frac{\partial\chi}{\partial W_K}\right)_1+\left.\frac{\partial^2\chi}{\partial W_K\partial E_{LM}}\right)_1\tilde{E}_{LM}+\left.\frac{\partial^2\chi}{\partial W_K\partial W_L}\right)_1\tilde{W}_L\\&\quad+\left.\frac{\partial^2\chi}{\partial W_K\partial\Theta}\right)_1\Delta\tilde{\Theta}+\cdots,\end{aligned} \tag{38}$$

where the notation $)_1$ indicates that the partial derivatives are evaluated in the intermediate state. Since the thermal exchanges are much slower than the variation of dynamic mechanical field, the high-frequency vibration can be assumed isentropic, and Eq. (27c) can be used to determine the small dynamic temperature shift created by the vibration:

$$\Delta\tilde{\Theta}=-\frac{\left.\frac{\partial^2\chi}{\partial\Theta\,\partial E_{KL}}\right)_1\tilde{E}_{KL}+\left.\frac{\partial^2\chi}{\partial\Theta\,\partial\tilde{W}_L}\right)_1\tilde{W}_L}{\left.\frac{\partial^2\chi}{\partial\Theta^2}\right)_1}. \tag{39}$$

With help of the following notations:

$$\chi_{KLMN})_1=\left.\frac{\partial^2\chi}{\partial E_{KL}\partial E_{MN}}\right)_1-\frac{\left.\frac{\partial^2\chi}{\partial E_{KL}\partial\Theta}\right)_1\left.\frac{\partial^2\chi}{\partial E_{MN}\partial\Theta}\right)_1}{\left.\frac{\partial^2\chi}{\partial\theta^2}\right)_1},$$

$$\chi_{K.LM})_1=\left.\frac{\partial^2\chi}{\partial W_K\partial E_{LM}}\right)_1-\frac{\left.\frac{\partial^2\chi}{\partial W_K\partial\Theta}\right)_1\left.\frac{\partial^2\chi}{\partial E_{MN}\partial\Theta}\right)_1}{\left.\frac{\partial^2\chi}{\partial\Theta^2}\right)_1}, \tag{40}$$

$$\chi_{KL})_1=\left.\frac{\partial^2\chi}{\partial W_K\partial W_L}\right)_1-\frac{\left.\frac{\partial^2\chi}{\partial W_K\partial\Theta}\right)_1\left.\frac{\partial^2\chi}{\partial W_L\partial\Theta}\right)_1}{\left.\frac{\partial^2\chi}{\partial\Theta^2}\right)_1}$$

and taking advantage of the possibility to exchange the order of derivation in mixed derivatives, we use Eq. (39) to eliminate $\Delta\tilde{\Theta}$ from Eq. (38), thereby obtaining

$$\begin{aligned}\frac{\partial\chi}{\partial E_{LM}}&=\left.\frac{\partial\chi}{\partial E_{LM}}\right)_1+\chi_{LMNP})_1\tilde{E}_{NP}+\chi_{K.LM})_1\tilde{W}_K+\cdots,\\\frac{\partial\chi}{\partial W_K}&=\left.\frac{\partial\chi}{\partial W_K}\right)_1+\chi_{K.LM})_1\tilde{E}_{LM}+\chi_{KL})_1\tilde{W}_L+\cdots.\end{aligned} \tag{41}$$

Substituting Eqs. (34)–(36) and (41) into Eq. (31), using the chain rules of differentation in the reverse way:

$$X_{M,i}=X_{M,\alpha}\xi_{\alpha,i}=X_{M,\alpha}(\delta_{i\alpha}-u_{\alpha,\varepsilon}\xi_{\varepsilon,i}+\cdots), \tag{42}$$

and taking into account the fact that the partial derivatives $\partial\chi/\partial E_{LM})_1$ and $\partial\chi/\partial W_K)_1$ identically vanish for the stress-free unpolarized bias of interest, we obtain the following linearized expression for $\tilde{K}_{Lj}$ and $\tilde{\Delta}_L$:

$$\begin{aligned}\tilde{K}_{Lj}&=\delta_{j\alpha}\xi_{\alpha,M}\xi_{\beta,N}\rho_0\chi_{LMNP})_1u_{\beta,P}\\&\quad+\delta_{j\alpha}\xi_{\alpha,M}\rho_0\chi_{K.LM})_1\tilde{W}_K+\cdots,\\\tilde{\Delta}_L&=[\varepsilon_0\overline{J}X_{L,\alpha}X_{M,\alpha}-\rho_0\chi_{LM})_1]\tilde{W}_M\\&\quad-\rho_0\chi_{L.MN})_1\xi_{\alpha,M}u_{\alpha,N}+\cdots,\end{aligned} \tag{43}$$

where $\overline{J}=\rho_0/\rho_1$ is the Jacobian of the transformation $X_L\rightarrow\xi_\alpha$. Thus, the incremental constitutive equations can be rewritten in the following form:

$$\begin{aligned}\tilde{K}_{Lj}&=(G_{L\gamma M\varepsilon}u_{\varepsilon,M}-R_{M\gamma L}\tilde{W}_M)\delta_{j\gamma},\\\tilde{\Delta}_L&=N_{LM}\tilde{W}_M+R_{L\alpha N}u_{\alpha,N},\end{aligned} \tag{44}$$

which define new *effective* elastic material coefficients $G_{L\gamma M\varepsilon}$, piezoelectric coefficients $R_{M\gamma L}$, and dielectric coefficients N_{LM} which govern an adiabatic vibration superposed on the bias resulting from an homogeneous temperature change accompanied by stress-free thermal expansion:

$$\begin{aligned}G_{L\gamma M\varepsilon}&=\xi_{\gamma,K}\xi_{\varepsilon,N}\rho_0\chi_{LK.MN})_1,\\R_{M\gamma L}&=-\xi_{\gamma,K}\rho_0\chi_{M.LK})_1,\\N_{LM}&=\varepsilon_0\overline{J}X_{L,\alpha}X_{M,\alpha}-\rho_0\chi_{LM})_1.\end{aligned} \tag{45}$$

These coefficients have tensorial properties and their values in a new rotated frame can be obtained by the classical rule of frame change of tensors. The associated stress equation of motion and charge equation of electrostatics take the form:

$$\tilde{K}_{Lj,L}=\rho_0\frac{\partial^2 u_\alpha}{\partial t^2}\delta_{j\alpha},$$
$$\tilde{\Delta}_{L,L}=0, \tag{46}$$

and the corresponding boundaries conditions are

$$\begin{matrix} N_L[\tilde{K}_{Lj}]=0 \\ N_L[\tilde{\Delta}_L]=\tilde{\sigma}_0 \end{matrix} \quad \text{on } S_0, \tag{47}$$

where $\tilde{\sigma}_0$ is the dynamic increment of surface charge density mapped on S_0. Equations (44)–(47) constitute the basic relations for the problem of a small vibration superimposed on a thermal bias, mapped in the *natural* state.

IV. PROPERTIES OF EFFECTIVE COEFFICIENTS

The effective coefficients defined by Eqs. (44) and (45) are related to the fundamental derivatives of energy function in a rather cumbersome though perfectly well defined way. They only depend on temperature provided that it is homogeneous and no thermally induced stress is present. All these sets of G, R, and N coefficients can be expanded up to any power of the temperature shift from Θ_0 to Θ, according to the following formula, given here for elastic coefficients:

$$G_{L\gamma M\varepsilon}(\Theta)=G_{L\gamma M\varepsilon}(\Theta_0)\times\left[1+\sum_{k=1}^{n}T^{(k)}G_{L\gamma M\varepsilon}(\Theta-\Theta_0)^k\right], \tag{48}$$

where

$$T^{(k)}G=\frac{1}{k!}\frac{\partial^k G}{\partial\Theta^k} \tag{49}$$

is the temperature coefficient of order k of the G coefficient. Similar expressions occur for piezoelectric and dielectric coefficients. When $\Theta_0=\Theta$, the intermediate and natural states are identical and the effective coefficients reduce to $\rho_0\chi_{KL})_1$, $\rho_0\chi_{K.LM})_1$, $\rho_0\chi_{KLMN})_1$. Fundamental elastic constants and their first-order temperature derivatives were previously defined from those quantities, which actually are the partial derivatives of energy under adiabatic conditions for the vibration, and were evaluated for quartz in Ref. 10. The fundamental constants must be used in case of any inhomogeneous bias inducing a field of nonzero static stresses, but no evaluation of their higher order thermal derivatives is available at this moment. In the special case when the intermediate state degenerates to the natural one, effective and fundamental constants have the same values and symmetry properties as the classical linear coefficients. But when the temperature Θ differs from the reference temperature Θ_0:

$$\xi_{\alpha,L}=\delta_{\alpha L}+\sum_n \alpha_{LM}^{(n)}\delta_{\alpha M}(\Theta-\Theta_0)^n, \tag{50}$$

and the effective coefficients have different values and lower symmetries than the classical coefficients which will be outlined in Sec. V. This holds for the G and R coefficients (but not N which exhibits the usual symmetries):

$$\begin{aligned} &G_{L\gamma M\varepsilon}(\Theta)\neq G_{P\alpha M\varepsilon}(\Theta)\delta_{P\gamma}\delta_{\alpha L},\\ &G_{L\gamma M\varepsilon}(\Theta)\neq G_{L\gamma Q\beta}(\Theta)\delta_{Q\varepsilon}\delta_{\beta M},\\ &G_{L\gamma M\varepsilon}(\Theta)\equiv G_{M\varepsilon L\gamma}(\Theta),\\ &R_{LM\varepsilon}\neq R_{LP\alpha}(\Theta)\delta_{P\varepsilon}\delta_{\alpha M}, \end{aligned} \tag{51}$$

where the Kronecker translators $\delta_{P\alpha}$ are necessary for consistency of Greek–Latin convention of indices. As a consequence, the $(L\alpha)$ couples can take nine different values. We adopt the same rule as Tiersten[4] for index compression:

$$\begin{aligned} &11\leftrightarrow 1 \quad 22\leftrightarrow 2 \quad 33\leftrightarrow 3\\ &23\leftrightarrow 4 \quad 31\leftrightarrow 5 \quad 12\leftrightarrow 6\\ &32\leftrightarrow 7 \quad 13\leftrightarrow 82 \quad 1\leftrightarrow 9. \end{aligned} \tag{52}$$

Care must be taken of index order of the deformation gradients in Eq. (44). At the temperature Θ, the structures of the matrices of the G and R coefficients for the crystallographic class 32 are the following:

$$\begin{bmatrix} G_{11} & G_{12} & G_{13} & G_{14} & 0 & 0 & G_{17} & 0 & 0\\ G_{12} & G_{11} & G_{13} & -G_{14} & 0 & 0 & -G_{17} & 0 & 0\\ G_{13} & G_{13} & G_{33} & 0 & 0 & 000 & & & 0\\ G_{14} & -G_{14} & 0 & G_{44} & 0 & 0 & G_{47} & 0 & 0\\ 0 & 0 & 0 & 0 & G_{55} & G_{17} & 0 & G_{47} & G_{17}\\ 0 & 0 & 0 & 0 & G_{17} & G_{66} & 0 & G_{14} & G_{66}\\ G_{17} & -G_{17} & 0 & G_{47} & 0 & 0 & G_{55} & 0 & 0\\ 0 & 0 & 0 & 0 & G_{47} & G_{14} & 0 & G_{44} & G_{14}\\ 0 & 0 & 0 & 0 & G_{17} & G_{66} & 0 & G_{14} & G_{66} \end{bmatrix} \tag{53}$$

$$\begin{bmatrix} R_{11} & -R_{11} & 0 & R_{14} & 0 & 0 & R_{17} & 0 & 0\\ 0 & 0 & 0 & 0 & -R_{14} & -R_{11} & 0 & -R_{17} & -R_{11}\\ 0 & 0 & 0 & 0 & 0 & 000 & & & 0 \end{bmatrix}. \tag{54}$$

These coefficients are not independent. At first, from Eq. (50) and remarking that, for the crystalline class of interest, only $\alpha_{11}^{(n)}$, $\alpha_{22}^{(n)}=\alpha_{11}^{(n)}$ and $\alpha_{33}^{(n)}$ are nonzero, it is easy to show that $G_{12}\equiv G_{11}-2G_{66},\forall\Theta$. Thus, G_{12} is eliminated, and one can choose $G_{11},G_{13},G_{14},G_{33},G_{44}$, and G_{66} as independent coefficients, considering that G_{17} depends on G_{14} while G_{47} and G_{55} depend on G_{44}. From the definition (45) with the help of (50) we find

$$\begin{aligned}
T^{(1)}G_{17}&=T^{(1)}G_{14}+\alpha_{11}^{(1)}-\alpha_{33}^{(1)},\\
T^{(1)}G_{47}&=T^{(1)}G_{44}+\alpha_{11}^{(1)}-\alpha_{33}^{(1)},\\
T^{(1)}G_{55}&=T^{(1)}G_{44}+2(\alpha_{11}^{(1)}-\alpha_{33}^{(1)}),
\end{aligned}\tag{55}$$

$$\begin{aligned}
T^{(2)}G_{17}&=T^{(2)}G_{14}+\alpha_{11}^{(1)}T^{(1)}G_{14}-\alpha_{33}^{(1)}T^{(1)}G_{17}\\
&\quad+\alpha_{11}^{(2)}-\alpha_{33}^{(2)},\\
T^{(2)}G_{47}&=T^{(2)}G_{44}+\alpha_{11}^{(1)}T^{(1)}G_{44}-\alpha_{33}^{(1)}T^{(1)}G_{47}\\
&\quad+\alpha_{11}^{(2)}-\alpha_{33}^{(2)},\\
T^{(2)}G_{55}&=T^{(2)}G_{44}+2\alpha_{11}^{(1)}T^{(1)}G_{44}-2\alpha_{33}^{(1)}T^{(1)}G_{55}\\
&\quad+2(\alpha_{11}^{(2)}-\alpha_{33}^{(2)})+(\alpha_{11}^{(1)})^2-(\alpha_{33}^{(1)})^2,
\end{aligned}\tag{56}$$

$$\begin{aligned}
T^{(3)}G_{17}&=T^{(3)}G_{14}+\alpha_{11}^{(1)}T^{(2)}G_{14}-\alpha_{33}^{(1)}T^{(2)}G_{17}\\
&\quad+\alpha_{11}^{(2)}T^{(1)}G_{14}-\alpha_{33}^{(2)}T^{(1)}G_{1}+\alpha_{11}^{(3)}-\alpha_{33}^{(3)},\\
T^{(3)}G_{47}&=T^{(3)}G_{44}+\alpha_{11}^{(1)}T^{(2)}G_{44}-\alpha_{33}^{(1)}T^{(2)}G_{17}\\
&\quad+\alpha_{11}^{(2)}T^{(1)}G_{14}-\alpha_{33}^{(2)}T^{(1)}G_{17}+\alpha_{11}^{(3)}-\alpha_{33}^{(3)},
\end{aligned}\tag{57}$$

$$\begin{aligned}
T^{(3)}G_{55}&=T^{(3)}G_{44}+2\alpha_{11}^{(1)}T^{(2)}G_{44}-2\alpha_{33}^{(1)}T^{(2)}G_{55}\\
&\quad+2[\alpha_{11}^{(2)}+(\alpha_{11}^{(1)})^2]T^{(1)}G_{44}\\
&\quad-2[\alpha_{33}^{(2)}+(\alpha_{33}^{(1)})^2]T^{(1)}G_{55}\\
&\quad+2[\alpha_{11}^{(3)}-\alpha_{33}^{(3)}+\alpha_{11}^{(1)}\alpha_{11}^{(2)}-\alpha_{33}^{(1)}\alpha_{33}^{(2)}].
\end{aligned}$$

V. CONNECTION WITH THE CLASSICAL TREATMENT

In this section, we briefly give the relation between heredefined effective coefficients and the ones used in the classical treatment[1] of the problem. The latter implicitly use the coordinates in the intermediate state as independent variables. In this state, the dynamic parts of the stress $\tilde{L}$ tensor and electric displacement vector $\tilde{D}$ can be obtained by the transformation laws:

$$\tilde{L}_{\alpha j}=\frac{1}{J}\xi_{\alpha,L}\tilde{K}_{Lj},\quad \tilde{D}_{\alpha}=\frac{1}{J}\xi_{\alpha,L}\tilde{\Delta}_{L}.\tag{58}$$

By substituting Eq. (44) into Eq. (58), with help of the chain rules of differentiation, we obtain

$$\begin{aligned}
\tilde{L}_{\alpha j}&=\delta_{j\gamma}(H^4_{\alpha\gamma\beta\delta}u_{\delta,\beta}+H^3_{\varepsilon\gamma\alpha}\tilde{\varphi}_{,\varepsilon}),\\
\tilde{D}_{\alpha}&=H^3_{\alpha\delta\beta}u_{\delta,\beta}-H^2_{\alpha\beta}\tilde{\varphi}_{,\beta},
\end{aligned}\tag{59}$$

where H^4 are the elastic coefficients of which temperature derivatives are most commonly determined,[1,8] and the following relations exist between both kinds of effective coefficients:

$$\begin{aligned}
H^4_{\alpha\gamma\beta\delta}&=\frac{1}{J}\frac{\partial\xi_\alpha}{\partial X_L}\frac{\partial\xi_\beta}{\partial X_M}G_{L\gamma M\delta},\\
H^3_{\alpha\delta\beta}&=\frac{1}{J}\frac{\partial\xi_\alpha}{\partial X_M}\frac{\partial\xi_\beta}{\partial X_L}R_{M\delta L},\\
H^2_{\alpha\beta}&=\frac{1}{J}\frac{\partial\xi_\alpha}{X_M}\frac{\partial\xi_\beta}{\partial X_N}N_{MN},
\end{aligned}\tag{60}$$

or, conversely:

$$\begin{aligned}
G_{L\gamma M\varepsilon}&=J\frac{\partial X_L}{\partial\xi_\alpha}\frac{\partial X_M}{\partial\xi_\delta}H^4_{\alpha\gamma\delta\varepsilon},\\
R_{L\gamma M}&=J\frac{\partial X_L}{\partial\xi_\varepsilon}\frac{\partial X_M}{\partial\xi_\alpha}H^3_{\varepsilon\gamma\alpha},\\
N_{MN}&=J\frac{\partial X_M}{\partial\xi_\alpha}\frac{\partial X_N}{\partial\xi_\beta}H^2_{\alpha\beta}.
\end{aligned}\tag{61}$$

These relations and (50) allow one to readily obtain the expressions between the thermal derivatives of the classical coefficients and the thermal derivatives of the effective material constants in Lagrange's formulation. Obviously, the quantities appearing in (59) are associated with the equilibrium equations:

$$\begin{aligned}
\tilde{L}_{\alpha j,\alpha}&=\rho_1\frac{\partial^2u_\beta}{\partial t^2}\delta_{j\beta},\\
\tilde{D}_{\alpha,\alpha}&=0,
\end{aligned}\tag{62}$$

and the boundary conditions:

$$\begin{aligned}
n_\alpha[\tilde{L}_{\alpha j}]&=0\\
n_\alpha[\tilde{D}_\alpha]&=\tilde{\sigma}_1
\end{aligned}\quad\text{on } S_1,\tag{63}$$

where $\tilde{\sigma}_1$ is the dynamic increment of charge mapped onto the boundary surface S_1 in the intermediate state.

VI. CONCLUSION

The formalism presented here allows one to rigorously treat the problem of acoustic wave propagation in a piezoelectric medium submitted to an homogeneous temperature variation. The effect of temperature on the actual crystalline orientation and shape of the device is automatically taken into account by using effective material constants referred to the fixed coordinates of the natural state. Although we detailed the main properties of these constants as well as their relations with other possibly defined material constants for the description of vibrations superposed on a bias, they are essentially suited for the special kind of bias considered here, thereby allowing for a consistent description of higher order static frequency–temperature characteristics of vibrating devices, which will be outlined in the companion paper, along with the obtention of numerical values of the thermal derivatives of effective constants.

[1] R. Bechmann, A. Ballato, and K. Lukaszek, ''High order temperature coefficients of the elastic stiffnesses and compliances of quartz,'' Proc. IRE **50**, 1812–1822 (1962).
[2] H. F. Tiersten, ''On the nonlinear equations of thermoelectroelasticity,'' Int. J. Eng. Sci. **9**, 587–604 (1971).
[3] R. A. Toupin, ''The elastic dielectric,'' J. Rat. Mech. Anal. **5**, 849–916 (1956).
[4] J. C. Baumhauer and H. F. Tiersten, ''Nonlinear electroelastic equations for small fields superposed on a bias,'' J. Acoust. Soc. Am. **54**, 1017–1034 (1973).
[5] C. Truesdell and R. A. Toupin, ''The classical field theories,'' vol. in *Handbuch der Physik* (Springer, Berlin, 1960), Vol. III/I, pp. 226–793.
[6] A. L. Cauchy, Mem. Acad. Sci. **XXII**, 615 (1850).
[7] R. N. Thurston, ''Waves in solids,'' in *Mechanics of Solids IV* (Springer, Berlin, 1974), pp. 109–132, Vol. VIa/4 of Encyclopedia of Physics.
[8] B. James, ''A new measurement of the basic elastic and dielectric constants of quartz,'' Proceedings of the 42nd Annual Freq. Cont. Symp., 1988, pp. 146–154.
[9] B. Dulmet and R. Bourquin, ''Méthodes de détermination des courbes fréquence-température d'un résonateur à quartz vibrant en mode d'épaisseur,'' Rev. Phys. Appl. **18**, 619–624 (1983).
[10] B. K. Sinha and H. F. Tiersten, ''First temperature derivatives of the fundamental elastic constants of quartz,'' J. Appl. Phys. **50**, 2732–2739 (1979).