

1.3. ELASTIC PROPERTIES

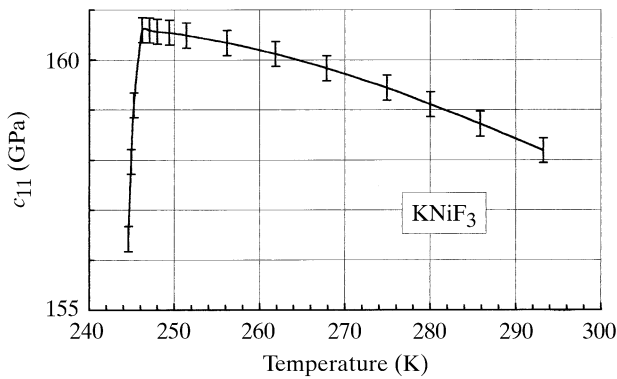


Fig. 1.3.5.4. Temperature dependence of the elastic constant c_{11} in KNiF_3 , which undergoes a para–antiferromagnetic phase transition. Reprinted with permission from *Appl. Phys. Lett.* (Nouet *et al.*, 1972). Copyright (1972) American Institute of Physics.

The softening of c_{44} when the temperature decreases starts more than 100 K before the critical temperature, Θ_c . In contrast, Fig. 1.3.5.4 shows the temperature dependence of c_{11} in KNiF_3 , a crystal that undergoes a para–antiferromagnetic phase transition at 246 K; the coupling between the elastic and the magnetic energy is weak, consequently c_{11} decreases abruptly only a few degrees before the critical temperature. We can generalize this observation and state that the softening of an elastic constant occurs over a large domain of temperature when this constant is the order parameter or is strongly coupled to the order parameter of the transformation; for instance, in the cooperative Jahn–Teller phase transition in DyVO_4 , $(c_{11} - c_{12})/2$ is the soft acoustic phonon mode leading to the phase transition and this parameter anticipates the phase transition 300 K before it occurs (Fig. 1.3.5.5).

1.3.5.3. Pressure dependence of the elastic constants

As mentioned above, anharmonic potentials are needed to explain the stress dependence of the elastic constants of a crystal. Thus, if the strain-energy density is developed in a polynomial in terms of the strain, only the first and the second elastic constants are used in linear elasticity (harmonic potentials), whereas higher-order elastic constants are also needed for nonlinear elasticity (anharmonic potentials).

Concerning the pressure dependence of the elastic constants (nonlinear elastic effect), considerable attention has been paid to their experimental determination since they are a unique source of significant information in many fields:

(i) In *geophysics*, a large part of the knowledge we have on the interior of the earth comes from the measurement of the transit time of elastic bursts propagating in the mantle and in the core (in the upper mantle, the average pressure is estimated to be about a few hundred GPa, a value which is comparable to that of the elastic stiffnesses of many materials).

(ii) In *solid-state physics*, the pressure dependence of the elastic constants gives significant indications concerning the stability of crystals. For example, Fig. 1.3.5.2 shows the pressure dependence of the elastic constants of KZnF_3 , a cubic crystal belonging to the perovskite family. As mentioned previously, this crystal is known to be stable over a wide range of temperature and the elastic stiffnesses c_{ij} depend linearly on pressure. It may be noted that, consequently, the third-order elastic constants

Table 1.3.5.2. Order of magnitude of the temperature dependence of the elastic stiffnesses for different types of crystals

Type of crystal	$(\partial \ln c_{11} / \partial \Theta)_p$ (K^{-1})	$(\partial \ln c_{44} / \partial \Theta)_p$ (K^{-1})
Ionic	-10^{-3}	-3×10^{-4}
Covalent	-10^{-4}	-8×10^{-5}
Metallic	-2×10^{-4}	-3×10^{-4}

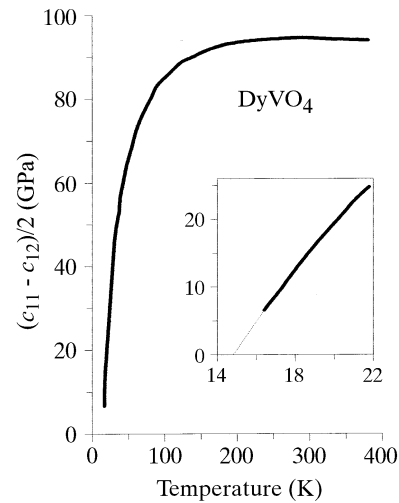


Fig. 1.3.5.5. Temperature dependence of $(c_{11} - c_{12})/2$ in DyVO_4 , which undergoes a cooperative Jahn–Teller phase transition (after Melcher & Scott, 1972).

(TOECs) are constant. On the contrary, we observe in Fig. 1.3.5.6 that the pressure dependence of the elastic constants of TlCdF_3 , a cubic crystal belonging to the same family but which is known to become unstable when the temperature is decreased to 191 K (Fischer, 1982), is nonlinear even at low pressures. In this case, the development of the strain-energy density in terms of strains cannot be stopped after the terms containing the third-order elastic constants; the contributions of the fourth- and fifth-order elastic constants are not negligible.

(iii) For practical use in the case of technical materials such as concrete or worked metals, the pressure dependence of the elastic moduli is also required for examining the effect of applied stresses or of an applied hydrostatic pressure, and for studying residual stresses resulting from loading (heating) and unloading (cooling) the materials.

1.3.6. Nonlinear elasticity

1.3.6.1. Introduction

In a solid body, the relation between the stress tensor T and the strain tensor S is usually described by Hooke's law, which postulates linear relations between the components of T and S (Section 1.3.3.1). Such relations can be summarized by (see equation 1.3.3.2)

$$T_{ij} = c_{ijkl} S_{kl},$$

where the c_{ijkl} 's are the elastic stiffnesses.

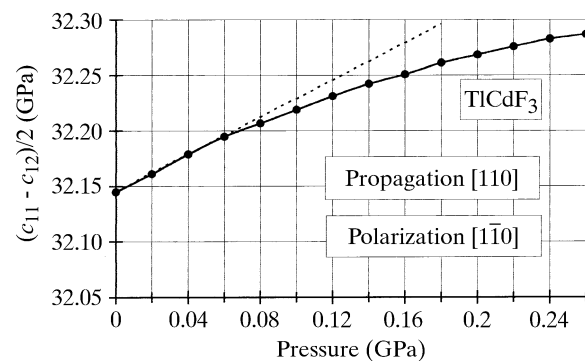


Fig. 1.3.5.6. Pressure dependence of the elastic constants $(c_{11} - c_{12})/2$ in TlCdF_3 . Reproduced with permission from *Ultrasonics Symposium Proc. IEEE* (Fischer *et al.*, 1980). Copyright (1980) IEEE.

1. TENSORIAL ASPECTS OF PHYSICAL PROPERTIES

For a solid under finite strain conditions, Hooke's law, valid for infinitesimal deformations, does not hold, and the fundamental definitions for stress and strain must be revisited.

1.3.6.2. Lagrangian and Eulerian description

Finite elastic strains may be treated from two different viewpoints using either the *Lagrangian (material)* or the *Eulerian (spatial)* descriptions.

Let us consider a fixed rectangular Cartesian coordinate system with axes x_i ($i = 1, 2, 3$). Any particular position vector \mathbf{r} of components (x_1, x_2, x_3) denotes a point in space. A point that always moves with the material is called a particle or *material point*. Let every particle be identified by its coordinates at some reference time t_0 . These reference coordinates, referred to the same Cartesian system, will be denoted by (a_1, a_2, a_3) and the corresponding position vector \mathbf{a} . A particular vector \mathbf{a} can serve as a name for the particle located at that position at the reference time t_0 .

The vectors \mathbf{r} and \mathbf{a} both specify a position in a fixed Cartesian frame of reference. At any time, we associate each \mathbf{r} with an \mathbf{a} by the rule that \mathbf{r} is the present position vector of the particle initially at \mathbf{a} . This connection between \mathbf{r} and \mathbf{a} is written symbolically as

$$\mathbf{r} = \mathbf{r}(t, \mathbf{a}) \quad \text{or} \quad x_i = x_i(t, a_1, a_2, a_3), \quad (1.3.6.1)$$

where

$$\mathbf{a} = \mathbf{r}(t_0, \mathbf{a}) \quad \text{or} \quad a_i = x_i(t_0, a_1, a_2, a_3). \quad (1.3.6.2)$$

The coordinates a_i that identify the particles are called *material coordinates*. A description that, like (1.3.6.1), uses (t, a_1, a_2, a_3) as independent variables is called a *material* or *Lagrangian* description.

The converse of (1.3.6.1) and (1.3.6.2) may be written

$$\mathbf{a} = \mathbf{a}(t, \mathbf{r}) \quad \text{or} \quad a_i = a_i(t, x_1, x_2, x_3), \quad (1.3.6.3)$$

where

$$\mathbf{r} = \mathbf{a}(t_0, \mathbf{r}) \quad \text{or} \quad x_i = a_i(t_0, x_1, x_2, x_3). \quad (1.3.6.4)$$

A spatial description or *Eulerian* description uses the independent variables (t, x_1, x_2, x_3) , the x_i being called *spatial coordinates*.

Now, for the sake of simplicity, we shall work with the Lagrangian formulation exclusively. For more details see, for instance, Thurston (1964) and Wallace (1970, 1972).

1.3.6.3. Strain and stress tensors

The displacement vector from the reference position of a particle to its new position has as components

$$u_i = x_i - a_i. \quad (1.3.6.5)$$

The term strain refers to a change in the relative positions of the material points in a body. Let a final configuration be described in terms of the reference configuration by setting t equal to a constant in (1.3.6.1). Then t no longer appears as a variable and (1.3.6.1) can be written

$$x_i = x_i(a_1, a_2, a_3),$$

where the a_i are the independent variables. It follows that

$$dx_j = \frac{\partial x_j}{\partial a_i} da_i = \left(\frac{\partial u_j}{\partial a_i} + \delta_{ij} \right) da_i. \quad (1.3.6.6)$$

Let now the particle initially at (a_1, a_2, a_3) move to (x_1, x_2, x_3) . The square of the initial distance to a neighbouring particle whose initial coordinates were $a_j + da_j$ is

$$ds^2 = da_j da_j.$$

The square of the final distance to the same neighbouring particle is

$$ds^2 = dx_j dx_j.$$

In a material description, the strain components S_{ik} are defined by the following equations:

$$dx_j dx_j - da_j da_j = 2S_{ik} da_i da_k. \quad (1.3.6.7)$$

Substituting (1.3.6.6) into (1.3.6.7), it follows that

$$\left(\frac{\partial u_j}{\partial a_i} + \delta_{ji} \right) \left(\frac{\partial u_j}{\partial a_k} + \delta_{jk} \right) da_i da_k - da_j da_j = 2S_{ik} da_i da_k.$$

Hence

$$S_{ik} = \frac{1}{2} \left(\frac{\partial u_k}{\partial a_i} + \frac{\partial u_i}{\partial a_k} + \frac{\partial u_j}{\partial a_i} \frac{\partial u_j}{\partial a_k} \right).$$

If the products and squares of the displacement derivatives are neglected, the strain components reduce to the usual form of 'infinitesimal elasticity' [see equation (1.3.1.8)]:

$$S_{ik} = \frac{1}{2} \left(\frac{\partial u_i}{\partial a_k} + \frac{\partial u_k}{\partial a_i} \right).$$

It is often useful to introduce the Jacobian matrix associated with the transformation (\mathbf{a}, \mathbf{x}) . The components of this matrix are

$$J = \begin{pmatrix} \alpha_{11} & \alpha_{12} & \alpha_{13} \\ \alpha_{21} & \alpha_{22} & \alpha_{23} \\ \alpha_{31} & \alpha_{32} & \alpha_{33} \end{pmatrix},$$

where

$$\alpha_{ik} = \frac{\partial x_i}{\partial a_k} = \frac{\partial u_j}{\partial a_k} + \delta_{jk}.$$

From the definition of matrix J , one has

$$d\mathbf{x} = J d\mathbf{a}$$

and

$$dx^2 - da^2 = (d\mathbf{x})^T d\mathbf{x} - (d\mathbf{a})^T d\mathbf{a} = (d\mathbf{a})^T (J^T J - \delta) d\mathbf{a},$$

where $(d\mathbf{a})^T$, $(d\mathbf{x})^T$ and J^T are the transpose matrices of $d\mathbf{a}$, $d\mathbf{x}$ and J , respectively, and δ is the Kronecker matrix.

The Lagrangian strain matrix S may then be written symbolically:

$$S = \frac{1}{2} (J^T J - \delta). \quad (1.3.6.8)$$

When finite strains are concerned, we have to distinguish three states of the medium: the natural state, the initial state and the final or present state: The *natural* state is a state free of stress. The *initial* state is deduced from the natural state by a homogeneous strain. The *final* state is deduced from the initial state by an arbitrary strain.

Concerning the stress tensor, as pointed out by Thurston (1964), the stress-deformation relation is complicated in nonlinear elasticity because 'the strain is often referred to a natural unstressed state, whereas the stress T_{ij} is defined per unit area of the deformed body'. For this reason, the differential of work done by the stress is not equal to the stress components times the differentials of the corresponding strain components. So, following Truesdell & Toupin (1960), we shall introduce a *thermodynamic tension tensor* t_{ij} defined as the first derivative of the energy with respect to strain. If the internal energy U per unit

1.3. ELASTIC PROPERTIES

mass is considered, the *thermodynamic tension* refers to an isentropic process. Then

$$t_{ij}^{\sigma} = \rho_0 \left(\frac{\partial U}{\partial S_{ij}} \right)_{\sigma},$$

where σ is the entropy and ρ_0 the volumic mass in the initial state.

If the Helmholtz free energy F is considered, the thermodynamic tension refers to an isothermal process. Then

$$t_{ij}^{\Theta} = \rho_0 \left(\frac{\partial U}{\partial S_{ij}} \right)_{\Theta},$$

where Θ is the temperature. It will be shown in Section 1.3.7.2 that

$$T_{ij} = (1/J) \alpha_{ik} \alpha_{jl} t_{kl}.$$

1.3.6.4. Second-order and higher-order elastic stiffnesses

Following Brugger (1964), the strain-energy density, or strain energy per unit volume Φ , is assumed to be a polynomial in the strain:

$$\Phi = \Phi_0 + c_{ij} S_{ij} + \frac{1}{2!} c_{ijkl} S_{ij} S_{kl} + \frac{1}{3!} c_{ijklmn} S_{ij} S_{kl} S_{mn}, \quad (1.3.6.9)$$

where $\Phi = \rho_0 U(X, S_{ij})$, $\Phi_0 = \rho_0 U(X, 0)$, X denotes the configuration of the initial state and the S_{ij} 's are the Lagrangian finite strain-tensor components.

If the initial energy and the deformation of the body are both zero, the first two terms in (1.3.6.9) are zero. Note that c_{ij} is a stress and not an intrinsic characteristic of the material. In this expression, the elastic stiffnesses c_{ijkl} and c_{ijklmn} are the second- and third-order stiffnesses, respectively. Since the strain tensor is symmetric, pairs of subscripts can be interchanged [see equation (1.3.3.4)]:

$$\begin{aligned} c_{ijkl} &= c_{jikl} = c_{ijlk} = c_{jilk}, \\ c_{ijklmn} &= c_{jiklmn} = c_{ijlkmn} = c_{jilknm} = c_{ijklnm} \\ &= c_{jiklnm} = c_{ijlknm} = c_{jilknm}. \end{aligned}$$

More accurately, the isentropic and the isothermal elastic stiffnesses are defined as the n th partial derivatives of the internal energy and the Helmholtz free energy, respectively. For example, the third-order isentropic and isothermal stiffnesses are, respectively,

$$\begin{aligned} c_{ijklmn}^{\sigma} &= \rho_0 \frac{\partial^3 U}{\partial S_{ij} \partial S_{kl} \partial S_{mn}}, \\ c_{ijklmn}^{\Theta} &= \rho_0 \frac{\partial^3 F}{\partial S_{ij} \partial S_{kl} \partial S_{mn}}, \end{aligned}$$

where the internal energy, U , is a function of X , S_{ij} and σ , and the Helmholtz free energy, F , is a function of X , S_{ij} and Θ .

From these definitions, it follows that the Brugger stiffness coefficients depend on the initial state. *When no additional information is given, the initial state is the natural state.*

The third-order stiffnesses form a sixth-rank tensor containing $3^6 = 729$ components, of which 56 are independent for a triclinic crystal and 3 for isotropic materials (the independent components of a sixth-rank tensor can be obtained for any point group using the accompanying software to this volume). The three independent constants for isotropic materials are often taken as c_{123} , c_{144} and c_{456} and denoted respectively by ν_1 , ν_2 , ν_3 , the 'third-order Lamé constants'.

The 'third-order Murnaghan constants' (Murnaghan, 1951), denoted by l , m , n , are given in terms of the Brugger constants by the relations

$$l = \frac{1}{2} c_{112}; \quad m = c_{155}; \quad n = 4c_{456}.$$

Similarly, the fourth-order stiffnesses form an eighth-rank tensor containing $3^8 = 6561$ components, 126 of which are independent for a triclinic crystal and 11 for isotropic materials (the independent components of a sixth-rank tensor can be obtained for any point group using the accompanying software to this volume).

For a solid under finite strain conditions, the definition of the elastic compliance tensor has to be reconsidered. In linear elasticity, the second-order elastic compliances s_{ijkl} were defined through the relations (1.3.3.2):

$$S_{ij} = s_{ijkl} T_{kl} \quad \text{or} \quad s_{ijkl} = \frac{\partial S_{ij}}{\partial T_{kl}},$$

while, in nonlinear elasticity, one has

$$s_{ijkl} = \frac{\partial S_{ij}}{\partial t_{kl}},$$

where

$$t_{kl} = \rho_0 \frac{\partial U}{\partial S_{kl}}.$$

1.3.6.5. Expansion of elastic constants for small initial stress

In most experiments, the initial stress is small compared with the second-order elastic constants (for example, 1 GPa hydrostatic pressure compared with the usual value $c_{ijkl} = 100$ GPa). Consequently, the deformation between the initial (stressed) state and the natural (unstressed) state is small compared with 1. For this reason, it is convenient to expand the elastic constants in the initial state as a power series in the strain about the natural state. *To avoid confusion, we introduce new notations:* \bar{X} now represents the coordinates in the *natural* or unstressed state; X represents the coordinates in the *initial* or homogeneously strained state; $u_i = x_i - X_i$ are the components of displacement. All letters with superscript bar refer to the natural state; for example, \bar{S}_{ij} denotes the Lagrangian strain in the natural state; $\bar{U} = U(\bar{X}, \bar{S}_{ij})$.

Now, in order to relate the properties at X to those at \bar{X} , we need to specify the strain from \bar{X} to X . Let

$$a_{ij} = \frac{\partial X_i}{\partial \bar{X}_j} = \bar{\alpha}_{ij}.$$

Consequently,

$$\frac{\partial \bar{S}_{ij}}{\partial S_{mn}} = a_{mi} a_{nj}.$$

The second-order elastic constants at X can be expressed in terms of the second- and third-order elastic constants at \bar{X} :

$$c_{ijkl} = \rho_0 \frac{\partial^2 U}{\partial S_{ij} \partial S_{kl}} = \rho_0 \frac{\partial^2 \bar{U}}{\partial \bar{S}_{mn} \partial \bar{S}_{pq}} a_{im} a_{jn} a_{kp} a_{lq}$$

or

$$c_{ijkl} = \frac{\rho_0}{\rho_0} \left(\bar{c}_{mnpq} + \bar{c}_{mnpqrs} S_{rs} + \frac{1}{2!} \bar{c}_{mnpqrstuv} S_{rs} S_{tu} + \dots \right) a_{im} a_{jn} a_{kp} a_{lq}.$$

This expression holds for both isentropic and isothermal elastic constants.

1. TENSORIAL ASPECTS OF PHYSICAL PROPERTIES

1.3.6.6. Elastic strain-energy density

The elastic strain-energy density has appeared in the literature in various forms. Most of the authors use the Murnaghan constants as long as isotropic solids are concerned. However, most of the literature uses Brugger's thermodynamic definition when anisotropic media are under consideration (Brugger, 1964).

The elastic strain-energy density for an isotropic medium, including third-order terms but omitting terms independent of strain, may be expressed in terms of three strain invariants, since an isotropic material is invariant with respect to rotation:

$$\Phi = \frac{\lambda + 2\mu}{2}(I_1)^2 - 2\mu I_2 + \frac{l + 2m}{3}(I_1)^3 - 2mI_1 I_2 + nI_3,$$

where λ and μ are the second-order Lamé constants, l, m, n are the third-order Murnaghan constants, and I_1, I_2, I_3 are the three invariants of the Lagrangian strain matrix. These invariants may be written in terms of the strain components as

$$\begin{aligned} I_1 &= S_{11} + S_{22} + S_{33} \\ I_2 &= \begin{vmatrix} S_{11} & S_{12} \\ S_{21} & S_{22} \end{vmatrix} + \begin{vmatrix} S_{22} & S_{23} \\ S_{32} & S_{33} \end{vmatrix} + \begin{vmatrix} S_{33} & S_{31} \\ S_{13} & S_{11} \end{vmatrix} \\ I_3 &= \begin{vmatrix} S_{11} & S_{12} & S_{13} \\ S_{21} & S_{22} & S_{23} \\ S_{31} & S_{32} & S_{33} \end{vmatrix}. \end{aligned}$$

The elastic strain-energy density for an *anisotropic* medium (for example a medium belonging to the most symmetrical groups of cubic crystals) is (Green, 1973)

$$\begin{aligned} \Phi &= \frac{1}{2}c_{11}[(S_{11})^2 + (S_{22})^2 + (S_{33})^2] + c_{12}[S_{11}S_{22} + S_{22}S_{33} + S_{33}S_{11}] \\ &+ c_{44}[(S_{12})^2 + (S_{21})^2 + (S_{23})^2 + (S_{32})^2 + (S_{31})^2 + (S_{13})^2] \\ &+ c_{111}[(S_{11})^3 + (S_{22})^3 + (S_{33})^3] \\ &+ c_{112}[(S_{11})^2(S_{22} + S_{33}) + (S_{22})^2(S_{33} + S_{11}) \\ &+ (S_{33})^2(S_{11} + S_{22})] \\ &+ \frac{1}{2}c_{144}\{S_{11}[(S_{23})^2 + (S_{32})^2] + S_{22}[(S_{31})^2 + (S_{13})^2] \\ &+ S_{33}[(S_{12})^2 + (S_{21})^2]\} \\ &+ \frac{1}{2}c_{166}\{[(S_{12})^2 + (S_{21})^2](S_{11} + S_{22}) \\ &+ [(S_{23})^2 + (S_{32})^2](S_{22} + S_{33}) \\ &+ [(S_{13})^2 + (S_{31})^2](S_{11} + S_{33})\} \\ &+ c_{123}S_{11}S_{22}S_{33} + c_{456}[S_{12}S_{23}S_{31} + S_{21}S_{32}S_{13}]. \end{aligned}$$

1.3.7. Nonlinear dynamic elasticity

1.3.7.1. Introduction

In recent years, the measurements of ultrasonic wave velocities as functions of stresses applied to the sample and the measurements of the amplitude of harmonics generated by the passage of an ultrasonic wave throughout the sample are in current use. These experiments and others, such as the interaction of two ultrasonic waves, are interpreted from the same theoretical basis, namely nonlinear dynamical elasticity.

A first step in the development of nonlinear dynamical elasticity is the derivation of the general equations of motion for elastic waves propagating in a solid under nonlinear elastic conditions. Then, these equations are restricted to elastic waves propagating either in an isotropic or in a cubic medium. The next step is the examination of two important cases:

(i) the generation of harmonics when *finite amplitude* ultrasonic waves travel throughout an *unstressed* medium;

(ii) the propagation of *small amplitude* ultrasonic waves when they travel throughout a *stressed* medium.

Finally, the concept of natural velocity is introduced and the experiments that can be used to determine the third- and higher-order elastic constants are described.

1.3.7.2. Equation of motion for elastic waves

For generality, these equations will be derived in the X configuration (initial state). It is convenient to obtain the equations of motion with the aid of Lagrange's equations. In the absence of body forces, these equations are

$$\frac{d}{dt} \frac{\partial L}{\partial x'_i} + \frac{\partial}{\partial X_i} \frac{\partial L}{\partial (\partial x_i / \partial X_j)} = 0 \quad (1.3.7.1)$$

or

$$\frac{d}{dt} \frac{\partial L}{\partial x'_i} + \frac{\partial}{\partial X_i} \frac{\partial L}{\partial \alpha_{ij}} = 0, \quad (1.3.7.2)$$

where L is the Lagrangian per unit initial volume and $\alpha_{ij} = \partial x_i / \partial X_j$ are the elements of the Jacobian matrix.

For adiabatic motion

$$L = \frac{1}{2} \rho_0 x_i'^2 - \rho_0 U, \quad (1.3.7.3)$$

where U is the internal energy per unit mass.

Combining (1.3.7.2) and (1.3.7.3), it follows that

$$\rho_0 x_i'' = \frac{\partial}{\partial X_j} \left(\rho_0 \frac{\partial U}{\partial S_{lm}} \frac{\partial S_{lm}}{\partial \alpha_{ij}} \right),$$

which can be written

$$\rho_0 x_i'' = \frac{\partial}{\partial X_j} \left(\alpha_{il} \alpha_{jm} \rho_0 \frac{\partial U}{\partial S_{lm}} \right)$$

since

$$\frac{\partial S_{lm}}{\partial \alpha_{ij}} = \frac{1}{2} (\alpha_{im} \delta_{jl} + \alpha_{il} \delta_{jm}).$$

Using now the equation of continuity or conservation of mass:

$$\frac{\rho_0}{\rho} = J = \det(a_{ij}),$$

and the identity of Euler, Piola and Jacobi:

$$\frac{\partial}{\partial x_j} \left(\frac{1}{J} \frac{\partial x_j}{\partial X_i} \right) = 0,$$

we get an expression of Newton's law of motion:

$$\rho x_i'' = \frac{dT_{ij}}{dX_j} \quad \text{or} \quad \rho u_i'' = \frac{dT_{ij}}{dX_j} \quad (1.3.7.4)$$

with

$$T_{ij} = \frac{\rho_0}{J} \alpha_{ik} \alpha_{jl} \frac{\partial U}{\partial S_{kl}} = \rho \alpha_{ik} \alpha_{jl} \frac{\partial U}{\partial S_{kl}}.$$

T_{ij} becomes

$$T_{ij} = \frac{1}{J} \alpha_{ik} \alpha_{jl} t_{kl}$$

since

$$t_{kl} = \rho_0 \frac{\partial U}{\partial S_{kl}}.$$