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Acoustical Activity of Crystals: a Comparative Study of Three Descriptions

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Abstract

Two apparently different descriptions of acoustical activity - one due to Portigal & Burstein [*Phys. Rev.* (1968), **170**, 673-678] based on the concept of spatial dispersion of the elastic stiffness tensor and the other based on the rotation-gradient theory due to Truesdell & Toupin [*Encyclopedia of Physics*, (1960), Vol. III/1. Berlin: Springer], Mindlin & Tiersten [*Arch. Ration. Mech. Anal.* (1962), **11**, 415-447] - are analysed on the common basis of the first-gradient theory. A relation between the tensors used for describing the acoustical activity in the two earlier descriptions is obtained.

1. Introduction

Ever since the concept of acoustical activity was introduced by Andronov (1960) and independently by Silin (1960) there has been continued interest both in its experimental observation (Pine, 1970; Joffrin, Dorner & Joffrin, 1980; Bialas & Schauer, 1982; Quan, Fang, Zhigong & Zenyi, 1987) and its theoretical characterization (Truesdell & Toupin, 1960; Mindlin & Tiersten, 1962; Portigal & Burstein, 1968; Mindlin & Toupin, 1971; Vuzhva & Lyamov, 1977; Kumarswamy & Krishnamurthy, 1980). Recently the occurrence of acoustical activity in crystals of different point-group symmetries has been examined from two apparently different points of view (Bhagwat, Wadhawan & Subramanian, 1986; Bhagwat & Subramanian, 1986) - the theory of spatial dispersion (Portigal & Burstein, 1968) and the rotation-gradient theory (Mindlin & Tiersten, 1962). Even though both viewpoints lead to the same acoustically active crystal classes, a disturbing feature remains: the tensors describing acoustical activity in the two descriptions have different symmetries and appear to be completely unrelated.

The aim of the present paper is to show that both viewpoints can be reconciled on the basis of the more general strain-gradient theory of acoustical activity where one employs the strain and its first gradient to describe the elastic deformation (Toupin, 1962; Mindlin, 1972). We also obtain a rationale for the maximum number of independent non-vanishing components of the acoustical activity tensors.

The paper is organized as follows. In § 2 we briefly recapitulate the salient features of the spatial dispersion theory of acoustical activity due to Portigal & Burstein (1968) and the rotation-gradient theory due to Truesdell & Toupin (1960) and Mindlin & Tiersten (1962). In § 3 we consider the full first-strain-gradient theory of elasticity given by Toupin (1962). Here we establish the formal equivalence of this theory with the theory of spatial dispersion. Further we show that rotation-gradient theory results from the general theory under certain additional restrictions.

2. Résumé of two viewpoints

A. *Theory of spatial dispersion*

Portigal & Burstein (1968) explained the occurrence of acoustical activity on the basis of spatial dispersion of the elastic stiffness tensor, by writing the most general form of Hooke's law as

$$\alpha_{ij}(\mathbf{r}, t) = \int d\mathbf{r}' \int_{-\infty}^{t} dt' \, C_{ijkl}(\mathbf{r}-\mathbf{r}', t-t')\varepsilon_{kl}(\mathbf{r}', t'). \quad (2.1)$$

As usual, summation over repeated indices will be implied. The stress $\boldsymbol{\sigma}$ at a point $\mathbf{r}$ at a time t is a linear superposition of strains at points $\mathbf{r}'$ at earlier instants t'. When spatial dispersion is small one may

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approximate the right-hand side of (2.1) and write

$$\sigma_{ij}(\mathbf{r}, t) = \int_0^\infty ds C_{ijkl}(s)\varepsilon_{kl}(\mathbf{r}, t-s) + \int_0^\infty ds\, d_{ijklm}(s)\, \partial\varepsilon_{kl}(\mathbf{r}, t-s)/\partial x_m \quad (2.2)$$

where

$$C_{ijkl}(s) = \int d\mathbf{R} C_{ijkl}(\mathbf{R}, s) \quad (2.3)$$

and

$$d_{ijklm}(s) = \int d\mathbf{R}\, R_m C_{ijkl}(\mathbf{R}, s). \quad (2.4)$$

In the equation for acoustic wave propagation what enters, however, is the Fourier transform of the stress given by (2.2). Taking the relevant transform one gets

$$\sigma_{ij}(\mathbf{q}, \omega) = C_{ijkl}(\omega)\varepsilon_{kl}(\mathbf{q}, \omega) + iq_m d_{ijklm}(\omega)\varepsilon_{kl}(\mathbf{q}, \omega). \quad (2.5)$$

Portigal & Burstein (1968) showed that the tensor **d** describes the acoustical activity of crystals. It has the following symmetry properties:

$$d_{ijklm} = d_{jiklm} = d_{ijlkm} = -d_{klijm}. \quad (2.6)$$

It was argued (Portigal & Burstein, 1968) that the antisymmetry of d_{ijklm} with respect to the interchange of the pair of indices (ij) with (kl) follows from considerations of causality and invariance of the crystal Hamiltonian under time reversal.

Rather than working with a fifth-rank tensor **d** one may work with the fourth-rank pseudotensor **G** defined as (Bhagwat, Wadhawan & Subramanian, 1986)

$$G_{qkmn} = \tfrac{1}{2} e_{ilq} d_{iklmn} \quad (2.7)$$

where e_{ilq} is the Levi-Civita tensor. A relation inverse to (2.7) can be written as

$$d_{iklmn} = e_{ilq} G_{qkmn} + e_{kmq} G_{qiln}. \quad (2.8)$$

The symmetry properties (2.6) for **d** imply the following symmetry properties for the pseudotensor **G**:

$$G_{qkmn} = G_{qmkn} \quad (2.9)$$

$$\sum_{k=1}^{3} G_{kkmn} = 0, \quad \text{for each } m, n = 1, 2, 3. \quad (2.10)$$

If the direction of propagation of an acoustic wave is an acoustic axis which is chosen as the z axis the acoustical activity can be shown to be determined by the component G_{3333}. The maximum number of independent non-vanishing components of **G** (as well as of **d**) is 45.

B. The rotation-gradient theory

Here we describe the elastic deformation of a solid in terms of the usual strain tensor $\boldsymbol{\varepsilon}$ and the rotation gradient **X**, which is a manifestation, in the lowest order, of the presence of couple stresses. Up to first order in **X** the energy density W can be written as

$$W = \tfrac{1}{2}\boldsymbol{\varepsilon} : \mathbf{C} : \boldsymbol{\varepsilon} + \boldsymbol{\varepsilon} : \mathbf{b} : \mathbf{X}. \quad (2.11)$$

To first order in the displacement u, its gradient ∇u and its second gradient $\nabla\nabla u$, $\boldsymbol{\varepsilon}$ and **X** are given by

$$\varepsilon_{ij} = \tfrac{1}{2}(\partial u_i/\partial x_j + \partial u_j/\partial x_i) \quad (2.12)$$

$$X_{ij} = \pm\tfrac{1}{2}(\partial/\partial x_i)[\partial u_m/\partial x_l - \partial u_l/\partial x_m]_{j \neq l,m}. \quad (2.13)$$

The wave equation for the displacement **u** involves the symmetric part of the stress $\tau_s = \partial W/\partial\boldsymbol{\varepsilon}$ and the non-scalar part of the couple stress $\mu_D = \partial W/\partial\mathbf{X}$ (Mindlin & Tiersten, 1962). It follows from the study of the plane-wave-like solutions of the wave equation that the tensor **b** is responsible for acoustical activity in this theory (Bhagwat & Subramanian, 1986). It has the following symmetry

$$b_{ijkl} = b_{jikl}, \sum_k b_{ijkk} = 0, \{i, j, k, l = 1, 2, 3\}. \quad (2.14)$$

Consequently the tensor **b** has a maximum of 48 independent non-vanishing components. If the acoustic axis is chosen as the z axis, the acoustical activity for waves travelling along the acoustic axis is governed by the quantity $(b_{1331} + b_{2332})$.

Although both the descriptions have their origin in the property of elasticity, the tensors **G** and **b** have different symmetries given by (2.9), (2.10) and (2.14) and also different numbers of independent non-vanishing components.

3. Acoustical activity and full-first-gradient theory

The fundamental equations for an elastic material taking into account the full gradient of strain were given by Toupin (1962). The term in the energy density giving rise to acoustical activity may be expressed in the form (Mindlin, 1972)

$$W = \varepsilon_{ij} C_{ijklm} \varepsilon_{klm}. \quad (3.1)$$

In the above equation ε_{ij} is the usual strain tensor given by (2.12) and

$$\varepsilon_{ijk} = \partial^2 u_k/\partial x_i \partial x_j = \varepsilon_{jik}. \quad (3.2)$$

In view of the symmetry of the strain tensor ε_{ij} and of ε_{ijk} under the interchange of the indices i and j the tensor C_{ijklm} possesses the intrinsic symmetry

$$C_{ijklm} = C_{jiklm} = C_{ijlkm}. \quad (3.3)$$

Each of $\{i, j, k, l, m\}$ takes the values 1, 2 and 3. The symmetry requirement (3.3) reduces the number of independent non-vanishing components of **C** to 108. The component ε_{klm} can be expressed in terms of its symmetric part and the antisymmetric part with respect to interchange of the last two indices l and

m. Thus

$$\varepsilon_{klm} = \varepsilon_{k(lm)} + \varepsilon_{k[lm]} \tag{3.4}$$

where

$$\varepsilon_{k(lm)} = \tfrac{1}{2}(\varepsilon_{klm} + \varepsilon_{kml}) = \partial\varepsilon_{lm}/\partial x_k \tag{3.5}$$

is the first gradient of strain and

$$\varepsilon_{k[lm]} = \tfrac{1}{2}[\varepsilon_{klm} - \varepsilon_{kml}] = \pm\partial/\partial x_k(\nabla \times \mathbf{u})_{i\neq l,m} \tag{3.6}$$

is the rotation gradient. The tensor ε_{klm} which is symmetric in the first two indices has 18 independent components. The strain-gradient tensor $\varepsilon_{k(lm)}$, symmetric in the last two indices, also has 18 independent components. Furthermore, the rotation-gradient tensor $\varepsilon_{k[lm]}$ can be expressed in terms of the first-strain-gradient tensor $\varepsilon_{k(lm)}$, since

$$\varepsilon_{k[lm]} = \tfrac{1}{2}[\varepsilon_{klm} - \varepsilon_{kml}] = \varepsilon_{l(km)} - \varepsilon_{m(kl)}. \tag{3.7}$$

Thus (3.4) and (3.7) together express ε_{klm} in terms of the first gradient of strain, while (3.5) provides the inverse relation. The energy density W of (3.1) can therefore be expressed in terms of the strain ε_{ij} and its first gradient $\varepsilon_{k(lm)}$:

$$W = \varepsilon_{ij}\tilde{C}_{ijlmk}\varepsilon_{k(lm)}. \tag{3.8}$$

From (3.1), (3.4), (3.7) and (3.8) we have

$$\tilde{C}_{ijlmk} = C_{ijklm} + C_{ijkml} - C_{ijmlk}. \tag{3.9}$$

A relation inverse to (3.9) can easily be obtained. We find

$$C_{ijklm} = \tfrac{1}{2}[\tilde{C}_{ijmlk} + \tilde{C}_{ijmkl}]. \tag{3.10}$$

It follows from (3.9) that the symmetry with respect to the interchange of i and j of C_{ijklm} implies the same symmetry for $\tilde{C}_{ijlmk}$. Further, since C_{ijlmk} is symmetric with respect to the interchange of l and m, $\tilde{C}_{ijlmk}$ must also be symmetric under the same interchange. If we compare the tensor $\tilde{\mathbf{C}}$ with $\mathbf{d}$ of Portigal & Burstein (1968), the above symmetry of $\tilde{C}_{ijlmk}$ with respect to the indices i, j and l, m is the same as that of $\mathbf{d}$ implied by the first two equations of (2.6). The additional symmetry (or rather the antisymmetry) of $\mathbf{d}$ as given by the last equation in (2.6) was argued by Portigal & Burstein (1968) on considerations of causality and time-reversal symmetry as applied to the total Hamiltonian of the crystal. The total energy can be obtained by integrating the energy density (3.8) over the volume of the crystal. If the crystal surfaces are strain free, it can be shown by carrying out integration by parts that the part of $\tilde{C}_{ijlmk}$ symmetric with respect to the simultaneous interchange of (ij) with (kl) does not contribute to the total energy. Hence we can set the symmetric part equal to zero and demand that

$$\tilde{C}_{ijlmk} = -\tilde{C}_{lmijk}. \tag{3.11}$$

In addition $\tilde{\mathbf{C}}$ also has the symmetry pointed out earlier, *viz*

$$\tilde{C}_{ijlmk} = \tilde{C}_{jilmk} = \tilde{C}_{ijmlk}. \tag{3.12}$$

With properties (3.11) and (3.12) of $\tilde{\mathbf{C}}$ and those of $\mathbf{d}$ contained in (2.6) we can identify the tensor $\tilde{\mathbf{C}}$ with $\mathbf{d}$. This identification limits the maximum number of independent components of $\tilde{\mathbf{C}}$ [and in view of (3.9) and (3.10) also of $\mathbf{C}$] to 45. The above considerations show that the results based on the phenomenological theory of acoustical activity would be identical to those emerging from a theory invoking the fundamental equations of elasticity given by Toupin (1962).

To see the emergence of the rotation-gradient theory one must use the variables $\varepsilon_{\{klm\}}$ defined as (Mindlin, 1972)

$$\varepsilon_{\{klm\}} = \tfrac{1}{3}(\varepsilon_{klm} + \varepsilon_{lmk} + \varepsilon_{mkl}) \tag{3.13}$$

and the rotation gradient $\varepsilon_{k[lm]}$ in place of ε_{klm} in (3.1). It is easy to see that

$$\varepsilon_{klm} = \varepsilon_{\{klm\}} + \tfrac{2}{3}(\varepsilon_{l[km]} + \varepsilon_{k[lm]}). \tag{3.14}$$

The use of the above equation in (3.1) for W gives

$$W = \varepsilon_{ij}C_{ijklm}\varepsilon_{\{klm\}} + \tfrac{4}{3}\varepsilon_{ij}C_{ijklm}\varepsilon_{k[lm]}. \tag{3.15}$$

In the first term involving $\varepsilon_{\{klm\}}$ only that part C^S_{ijklm} of C_{ijklm} which is totally symmetric in the indices k, l and m will contribute. Thus,

$$C^S_{ijklm} = \tfrac{1}{3}(C_{ijklm} + C_{ijlmk} + C_{ijmkl}), \tag{3.16}$$

and in the second term the part C^A_{ijklm} which is antisymmetric with respect to the interchange of l with m will contribute

$$C^A_{ijklm} = \tfrac{1}{2}(C_{ijklm} - C_{ijkml}). \tag{3.17}$$

In the general case, therefore, W can be expressed as

$$W = \varepsilon_{ij}C^S_{ijklm}\varepsilon_{\{klm\}} + \tfrac{4}{3}\varepsilon_{ij}C^A_{ijklm}\varepsilon_{k[lm]}. \tag{3.18}$$

It is clear that the rotation-gradient description would result from (3.18) only if the first term on the right-hand side of it is zero, *i.e.* $C^S_{ijklm} = 0$. In terms of the components of $\mathbf{C}$ this condition reads

$$C_{ijklm} + C_{ijlmk} + C_{ijmkl} = 0. \tag{3.19}$$

The second term on the right-hand side of (3.18) can be transformed into its familiar form, *viz* the second term on the right-hand side of (2.11), by noting that $\varepsilon_{k[lm]}$ can be expressed in terms of a second-rank tensor X_{kp} by the relation

$$\varepsilon_{k[lm]} = e_{lmp}X_{kp} \tag{3.20}$$

and the identification

$$b_{ijkp} = \tfrac{4}{3}e_{lmp}C^A_{ijklm}. \tag{3.21}$$

The relation (3.19) imposes 60 constraints on the components of $\mathbf{C}$. If one disregards the constraints imposed on $\mathbf{C}$ by the relations (3.11) and (3.10), the

independent components of **C** reduce from 108 to 48, which was the number obtained for the tensor **b** in the rotation-gradient theory. However, if we were to take into account the further restrictions implied by the relation (3.11) the number of independent components of **C** (or $\tilde{\mathbf{C}}$) will be further reduced. A detailed analysis of all the constraints on **C** shows that only the six components C_{11223}, C_{11332}, C_{11132}, C_{22331}, C_{22231} and C_{33231} are independent. The other non-vanishing components are

$$C_{11232} = -C_{22131} = -2C_{13122} = 2C_{23121} = -\tfrac{1}{2}C_{11223}$$

$$C_{11233} = 2C_{23131} = -C_{33121} = -2C_{13123} = -\tfrac{1}{2}C_{11332}$$

$$C_{11123} = -2C_{13121} = -C_{11132}$$

$$C_{22133} = -C_{33122} = -2C_{23132} = 2C_{23123} = -\tfrac{1}{2}C_{22331}$$

$$C_{22123} = -2C_{23122} = -C_{22231}$$

$$C_{33132} = -2C_{23133} = -C_{33231}$$

and those related to the above by the intrinsic symmetry relation (3.3). It follows from the relations (3.21), (3.17), (3.10) and (2.8) (with the identification $\mathbf{d} = \tilde{\mathbf{C}}$) that the activity tensor **b** of the rotation-gradient theory which conforms to the restrictions from considerations of the total energy has only six non-vanishing independent components [as against 48 reported earlier (Bhagwat & Subramanian, 1986)] for the triclinic system and is related to the tensor **G** by the equation

$$b_{ijkp} = \tfrac{2}{3}[G_{pikj} - \delta_{pj}G_{qikq} + e_{lmp}e_{ikq}G_{qjml}].$$

The quantities $[b_{1331} + b_{2332}]$ and G_{3333} which govern the angle of rotation of a plane-polarized acoustic wave in the two descriptions respectively turn out to be proportional to the component C_{33231}. This shows that even this restricted tensor **b** will lead to acoustical activity in all those crystal classes which were found acoustically active in either of the two earlier descriptions. In other words, the restricted theory still retains the essential ingredients of the general theory as far as the explanation of acoustical activity is concerned.

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On the Deficiencies of the Anharmonic One-Particle Potential Model

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Abstract

The temperature factors of Cd and Zn for *h*00 reflexions calculated by numerical Fourier transformation are compared with the temperature factors determined with the help of series expansion of the anharmonic term of the probability density function (p.d.f.). The anharmonic parameters used have been derived by least-squares fit of measured Bragg intensities in the framework of the anharmonic one-particle potential (OPP) model. For Cd a deviation of up to 7% is found for the results obtained for the symmetric part and up to 50% for the antisymmetric part of the temperature factor. It is shown that numerical Fourier transformation of the p.d.f., using the anharmonic parameters given in the literature for Zn, is not always possible, because the p.d.f. is divergent for some of these parameters.

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