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The Influence of Piezoelectric Coupling on Material Constants Determining Brillouin Scattering

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The piezoelectric modifications of the tensors of elastic, optical and photoelastic constants and their consequences on spontaneous and stimulated Brillouin scattering are discussed.

1. Basic equations

Starting with the expansion of the thermodynamic potential $f(U_{lm}, E_l)$ of a lossless medium with respect to the strain U_{lm} and the electric field E_i,

$$f(U_{lm},E_l) = f_o - \alpha_i E_i - \tfrac{1}{2}\alpha_{ik}E_iE_k - \tfrac{1}{3}\alpha_{ikl}E_iE_kE_l - \ldots + \tfrac{1}{2}\beta_{iklm}U_{ik}U_{lm} + \gamma_{ilm}E_iU_{lm} + \tfrac{1}{2}\gamma_{iklm}E_iE_kU_{lm}\,, \tag{1}$$

we obtain the equations of state we need for the macroscopic description of the Brillouin scattering process

$$P_i = -\frac{\partial f}{\partial E_i} = \alpha_i + \alpha_{ik}E_k + \alpha_{ikl}E_kE_l - \gamma_{ilm}U_{lm} - \gamma_{iklm}E_kU_{lm} \tag{2}$$

$$S_{ik} = \frac{\partial f}{\partial U_{ik}} = \beta_{iklm}U_{lm} + \gamma_{sik}E_s + \tfrac{1}{2}\gamma_{lmik}E_lE_m\,. \tag{3}$$

Since we are dealing with high-frequency processes, namely ultrasonic and optical waves, in (2) and (3) the partial derivatives should be taken at constant entropy. That is the coefficients in (2) and (3) represent adiabatic constants. The optical constants α_{ik} and $\varepsilon_{ik} = \alpha_{ik} + \varepsilon_0\delta_{ik}$, the elastic constants β_{iklm}, the piezoelectric constants γ_{ikl}, the electrooptical constants α_{ikl} and the photoelastic constants γ_{iklm} give the connexions between the electric field E_i, the strain U_{lm} and the electric polarization P_i as well as the stress S_{ik}.

The linear piezoelectric coupling of the elastic and electric fields is given by the tensor of the piezoelectric constants γ_{sik}. Because of its index symmetry $\gamma_{sik} = \gamma_{ski}$ there are only 18 linearly independent components. This number is further reduced by the symmetry of the crystallographic class. For instance C_{6v} (6*mm*) (*e.g.* ZnO, CdS, ZnS, CdSe, CdTe, ZnTe) has three independent piezoelectric constants, C_{3v} (3*m*) (*e.g.* $LiNbO_3$) four constants, T_d ($\bar{4}3m$) (*e.g.* ZnS, GaAs, InSb, GaP, InAs, ZnSe) only one independent component. For media with inversion symmetry piezoelectricity does not exist.

We have to substitute the equations of state (2,3) into the field equations of Maxwell's and Newton's continuum theory

$$\left(\frac{\partial^2}{\partial x_i \partial x_k} - \frac{\partial^2}{\partial x_l \partial x_l}\delta_{ik} + \frac{\partial^2}{c_0^2\partial t^2}\delta_{ik}\right)E_k = -\mu_0\frac{\partial^2}{\partial t^2}P_i \tag{4}$$

$$\varrho\frac{\partial^2}{\partial t^2}U_i = \frac{\partial}{\partial x_k}S_{ik}\,. \tag{5}$$

2. Mixed acoustoelectromagnetic waves

At first we will consider only the linear terms and assuming plane waves with wave vectors $\mathbf{k} = k\mathbf{n}, \mathbf{K} = K\mathbf{N}$ and unit vectors $\mathbf{n}, \mathbf{N}$ of the wave normals

$$E_l = \hat{E}_l \exp\left[i(\mathbf{kx} - \omega t)\right] \tag{6}$$

$$U_l = \hat{U}_l \exp\left[i(\mathbf{Kx} - \Omega t)\right]\,, \tag{7}$$

we get the equations ($\mathbf{k} = \mathbf{K}, \omega = \Omega$):

$$(k_ik_k - k^2\delta_{ik} + \frac{\omega^2}{c_0^2}\varepsilon_{ik}^{\rm rel})E_k - i\mu_0\omega^2\gamma_{ikl}U_kk_l = 0 \tag{8}$$

$$(\varrho\Omega^2\delta_{il} - \beta_{iklm}k_mk_k)U_l + i\gamma_{sik}E_sk_k = 0\,. \tag{9}$$

If there is no piezoelectricity, we obtain the familiar two electromagnetic (eigen-)waves ($A=1,2$) propagating with the two velocities of light $c_{(A)}$ according to the dispersion relation $\omega=\omega_{(A)}(\mathbf{k})=kc_{(A)}(\mathbf{n})$ and the three acoustic (eigen-)waves ($J=1,2,3$) travelling with the three phase velocities of sound $v_{(J)}$ according to the dispersion relations $\Omega=\Omega_{(J)}(\mathbf{K})=Kv_{(J)}(\mathbf{N})$.

In the piezoelectric case each acoustic wave is accompanied by an electric wave and *vice versa*, so that there exist five mixed acoustoelectr(omagnet)ic (eigen-) waves ($Z=1,2,3,4,5$), *i.e.* we have pentrefrigence (Kyame, 1949). Their polarizations and dispersion relations $\omega=\omega_{(Z)}(\mathbf{k})=kV_{(Z)}(\mathbf{n})$ can be deduced from the linear system (8,9). The phase velocities $V_{(Z)}(\mathbf{n})$ are modified by the piezoelectric coupling, if the corresponding pure electric and acoustic modes are piezo-active, *i.e.* if $\gamma_{sik}E_sk_k\neq 0$ and $\gamma_{ikl}U_kk_l\neq 0$ respectively.

As the piezoelectric coupling is relatively weak these five waves will split into two waves propagating with phase velocities $V_{(Z)}=\bar{c}_{(A)}$ of the order of magnitude of the velocity of light (fast modes, mixed waves of electromagnetic type) and into three waves with phase velocities $V_{(Z)}=\bar{v}_{(J)}$ of the order of magnitude of the velocity of sound (slow modes, mixed waves of acoustic type).

Elimination of the elastic displacement U_l in (8) and (9) for an electromagnetic-type mode results in the familiar Maxwell wave equation

$$(k_ik_k-k^2\delta_{ik}+\frac{\omega^2}{c_0^2}\,\bar{\varepsilon}_{ik}^{\mathrm{rel}})E_k=0 \qquad (10)$$

with the effective dielectric constants ($\bar{\varepsilon}_{ik}^{\mathrm{rel}}=\bar{\varepsilon}_{ik}/\varepsilon_0$)

$$\bar{\varepsilon}_{it}(\mathbf{n})=\varepsilon_{it}-\frac{1}{\varrho c^2}\,\gamma_{irl}n_r\gamma_{tsl}n_s \qquad (11)$$

determining the polarizations and velocities $\bar{c}_{(A)}(\mathbf{n})$ of electromagnetic waves in piezoelectric media.

Eliminating the electric field of an acoustic-type mode we find in the same way an equation of elastic motion

$$(\varrho\Omega^2\delta_{il}-\bar{\beta}_{iklm}K_kK_m)U_l=0 \qquad (12)$$

with effective elastic constants $\bar{\beta}$, which are maximally altered by piezoelectricity, if the acoustic field produces a longitudinal electric field,

$$\bar{\beta}_{iklm}=\beta_{iklm}+\frac{\gamma_{sik}N_s\gamma_{rlm}N_r}{\varepsilon_{vw}N_vN_w}\,, \qquad (13)$$

and which determine the polarizations and velocities $\bar{v}_{(J)}(\mathbf{N})$ of acoustic waves in piezoelectric media.

3. Brillouin scattering in piezoelectric media

The typical macroscopic interactions of the spontaneous and stimulated Brillouin scattering processes are given by the bilinear and quadratic terms in (2) and (3) containing the photoelastic constants. As we have to treat the acoustic and the associated (longitudinal) electric field in a piezoelectric crystal on the same level we add in the electric polarization the non-linear term with the electrooptical constants. Accordingly we insert the equations of state (2,3) into the field equations (4,5).

The Brillouin scattering problem is characterized by three interacting waves which we assume to be plane and stationary. These are the mixed exciting wave of electromagnetic type $[\omega=\omega_{(A)}(\mathbf{k})=k\bar{c}_{(A)}(\mathbf{n})]$

$$E_i,U_i\sim\exp\left[i(\mathbf{kx}-\omega t)\right], \qquad (14)$$

the mixed wave of acoustic type $[\Omega=\Omega_{(J)}(\mathbf{K})=K\bar{v}_{(J)}(\mathbf{N})]$

$$\tilde{U}_i,\tilde{E}_i\sim\exp\left[i(\mathbf{Kx}-\Omega t)\right] \qquad (15)$$

causing the scattering of the exciting wave and finally the mixed scattered wave of electromagnetic type $[\omega'=\omega_{(A')}(\mathbf{k}')=k'\bar{c}_{(A')}(\mathbf{n}')]$

$$E_i',U_i'\sim\exp\left[i(\mathbf{k}'\mathbf{x}-\omega' t)\right]. \qquad (16)$$

Arranging the field equations with regard to the phase-match relations

$$\omega'=\omega\pm\Omega\,,\quad \mathbf{k}'=\mathbf{k}+\mathbf{K}\,, \qquad (17)$$

we get three pairs of equations for the mixed exciting, scattering and scattered waves. If we finally eliminate the less interesting acoustic displacement in the field equations for the mixed electromagnetic-type waves to obtain the electric field and analogously eliminate the electric field of the acoustic-type modes to obtain the acoustic displacement, we have the well known (Fourier-transformed) equations

$$\left(k_ik_k-k^2\delta_{ik}+\frac{\omega^2}{c_0^2}\,\bar{\varepsilon}_{ik}^{\mathrm{rel}}\right)E_k=-i\mu_0\omega^2\bar{\gamma}_{irtu}K_uE_r'\tilde{U}_t^* \qquad (18)$$

$$\left(k_i'k_k'-k'^2\delta_{ik}+\frac{\omega'^2}{c_0^2}\,\bar{\varepsilon}_{ik}^{\mathrm{rel}}\right)E_k'=i\mu_0\omega'^2\bar{\gamma}_{irtu}K_uE_r\tilde{U}_t \qquad (19)$$

$$(\varrho\Omega^2\delta_{il}-\bar{\beta}_{iklm}K_kK_m)\tilde{U}_l=-\tfrac{1}{2}\bar{\bar{\gamma}}_{ljit}E_l^*E_j'K_t\,, \qquad (20)$$

which describe the (anti-Stokes component of the) Brillouin scattering process (Macheleidt, 1972).

The optical, elastic and photoelastic constants are modified by the piezoelectric coupling:

$$\bar{\varepsilon}_{ik}=\varepsilon_{ik}-\frac{1}{\varrho c^2}\,\gamma_{irl}n_r\gamma_{ksl}n_s \qquad (21)$$

$$\bar{\beta}_{iklm}=\beta_{iklm}+\frac{\gamma_{sik}N_s\gamma_{rlm}N_r}{\varepsilon_{vw}N_vN_w} \qquad (22)$$

$$\bar{\gamma}_{irtu}=\gamma_{irtu}-\frac{\alpha_{irm}\gamma_{ntu}N_nN_m}{\varepsilon_{vw}N_vN_w} \qquad (23)$$

$$\bar{\bar{\gamma}}_{irtu}=\gamma_{irtu}-\frac{2\alpha_{irm}\gamma_{ntu}N_nN_m}{\varepsilon_{vw}N_vN_w}\,. \qquad (24)$$

Formula (23) was also obtained by Nelson & Lax (1971) starting with a classical microscopic theory of nonlinear electrodynamics in elastic anisotropic dielectrics. In this paper Nelson & Lax give a detailed discussion of photoelastic interaction and an extensive list of references. Besides classical papers (*e.g.* Pockels, 1889*a*,*b*, 1890) their bibliography contains works dealing with the influence of piezoelectricity on photoelasticity (*e.g.* Brody & Cummins, 1969; Chapelle & Taurel, 1955; Coquin, 1970; Wemple & Di Domenico, 1970). Compared with the nonpiezoelectric case the effective constants (21,22,23,24) exhibit changed magnitudes, additional direction dispersion and an increased number of independent components.

For representative materials the relative change in these constants can be estimated to be of the order of magnitude of 10^{-11} for the optical, 10^{-2} for the elastic and 10^{-2} for the photoelastic constants.

In addition to the (*e.g.* electronic) frequency dispersion of the pure constants, $\varepsilon_{ik},\beta_{iklm},\gamma_{irkl}$, which we are starting with as given quantities, the coupling terms introduce the frequency dependence determined by the dispersion of γ_{ikl}, α_{ikl} and ε_{ik} respectively. In our case, however, this extra dispersion is negligibly small.

As the piezoelectric coupling cannot be switched off, experimentally we measure the components of the tensors $\bar{\varepsilon},\bar{\beta}$, $\bar{\gamma}$ and $\bar{\bar{\gamma}}$, and not those of ε,β,γ. But from considerations of the symmetry of the crystal we know at least the point-group structure of the pure tensors, $\varepsilon_{ik},\beta_{iklm},\gamma_{iklm}$, which is tabulated in textbooks on crystal physics (*e.g.* Ludwig, 1970) and which in general does not coincide with that of the effective tensors, $\bar{\varepsilon},\bar{\beta},\bar{\gamma},\bar{\bar{\gamma}}$. For that reason we cannot immediately start with (the symmetry of) the pure constants $\varepsilon_{ik},\beta_{iklm},\gamma_{iklm}$ to calculate the Brillouin scattering in piezoelectric crystals, but we have first to compute the effective constants (21,22,23,24). The number of independent components of the effective material tensors is greater than that of the pure material tensors. Therefore, within the above given order of magnitude, the anisotropy of the corresponding processes (propagation of light and sound, scattering of light, generation of sound) is increased in piezoelectric media compared with a medium of the same crystallographic class, but without consideration of piezoelectricity.

The effective linear optical and elastic constants determine through the familiar electric and acoustic wave equations (10,12) the polarizations and dispersion relations of the light and sound waves participating in the Brillouin scattering process. Now it is these piezoelectrically modified phase-velocities which we have to insert into the Brillouin frequency shift formula (Macheleidt, 1972).

$$\frac{\omega'-\omega}{\omega}=\pm\bar{v}_{(J)}(\mathbf{N})\times\sqrt{\frac{4\sin^2(\theta/2)}{\bar{c}_{(A)}(\mathbf{n})\bar{c}_{(A')}(\mathbf{n}')}-\left(\frac{1}{\bar{c}_{(A)}(\mathbf{n})}-\frac{1}{\bar{c}_{(A')}(\mathbf{n}')}\right)^2} \tag{25}$$

$$\mathbf{N}=\frac{\bar{c}_{(A)}(\mathbf{n})\mathbf{n}'-c_{(A')}(\mathbf{n}')\mathbf{n}}{\sqrt{\bar{c}^2_{(A')}+\bar{c}^2_{(A)}-2\bar{c}_{(A')}\bar{c}_{(A)}\cos\theta}}\,. \tag{27}$$

Because of the relatively large piezoelectrically induced increase in the magnitude (piezoelectric stiffening) and anisotropy of the sound velocity entering into formula (25), by precise measurements of the Brillouin frequency shift in the dependence of the scattering geometry (*e.g.* the scattering angle θ) and of the polarizations, not only the pure elastic but also (certain) piezoelectric constants can be determined, (Cecchi *et al.*, 1970; O'Brien *et al.*, 1969).

Moreover the intensities of the non-linearly generated waves essentially depend on the tensor of the effective photoelastic constants. Thus the piezoelectric coupling is of consequence to the intensity of the scattered light. A glance at (19) shows that in principle it is possible to find out not only the pure photoelastic but also (certain) electrooptical constants by sufficiently exact (relative) intensity measurements.

It is very remarkable that the piezoelectrically varied photoelastic constants $\bar{\gamma}_{iklm}$ describing the scattering of light by sound differ from those photoelastic constants $\bar{\bar{\gamma}}_{iklm}$ determining the generation of acoustic waves by nonlinear interaction of two electromagnetic waves.

Since the linear constants $\bar{\varepsilon}_{ik}$, $\bar{\beta}_{iklm}$ and the product of $\bar{\gamma}_{iklm}$ and $\bar{\bar{\gamma}}_{iklm}$ enter into the relation defining the threshold of the stimulated Brillouin scattering this threshold will be modified with respect to its value and especially to its anisotropy by the piezoelectric coupling.

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