International Tables for Crystallography (2013). Vol. D, Section 2.4.4, pp. 350–351.

Comparing (2.4.2.11) and (2.4.2.2), one sees that the effective elastic tensor $\mathbf{c}^{(e)}$ now depends on the propagation direction $\hat{\mathbf{Q}}$. Otherwise, all considerations of the previous section, starting from (2.4.2.6), remain, with $\mathbf{c}$ simply replaced by $\mathbf{c}^{(e)}$.

2.4.3. Coupling of light with elastic waves

2.4.3.1. *Direct coupling to displacements*

The change in the relative optical dielectric tensor $\boldsymbol{\kappa}$ produced by an elastic wave is usually expressed in terms of the strain, using the Pockels piezo-optic tensor $\mathbf{p}$, as

$$(\Delta\kappa^{-1})_{ij} = p_{ijk\ell}S_{k\ell}. \qquad (2.4.3.1)$$

The elastic wave should, however, be characterized by both strain $\mathbf{S}$ *and* rotation $\mathbf{A}$ (Nelson & Lax, 1971; see also Section 1.3.1.3):

$$A_{[k\ell]} = \tfrac{1}{2}\left(\frac{\partial u_k}{\partial x_\ell} - \frac{\partial u_\ell}{\partial x_k}\right). \qquad (2.4.3.2)$$

The square brackets on the left-hand side are there to emphasize that the component is antisymmetric upon interchange of the indices, $A_{[k\ell]} = -A_{[\ell k]}$. For birefringent crystals, the rotations induce a change of the local $\boldsymbol{\kappa}$ in the laboratory frame. In this case, (2.4.3.1) must be replaced by

$$(\Delta\kappa^{-1})_{ij} = p'_{ijk\ell}\frac{\partial u_k}{\partial x_\ell}, \qquad (2.4.3.3)$$

where $\mathbf{p}'$ is the new piezo-optic tensor given by

$$p'_{ijk\ell} = p_{ijk\ell} + p_{ij[k\ell]}. \qquad (2.4.3.4)$$

One finds for the rotational part

$$p_{ij[k\ell]} = \tfrac{1}{2}[(\kappa^{-1})_{i\ell}\delta_{kj} + (\kappa^{-1})_{\ell j}\delta_{ik} - (\kappa^{-1})_{ik}\delta_{\ell j} - (\kappa^{-1})_{kj}\delta_{i\ell}]. \qquad (2.4.3.5)$$

If the principal axes of the dielectric tensor coincide with the crystallographic axes, this gives

$$p_{ij[k\ell]} = \tfrac{1}{2}(\delta_{i\ell}\delta_{kj} - \delta_{ik}\delta_{\ell j})(1/n_i^2 - 1/n_j^2). \qquad (2.4.3.6)$$

This is the expression used in this chapter, as monoclinic and triclinic groups are not listed in the tables below.

For the calculation of the Brillouin scattering, it is more convenient to use

$$(\Delta\kappa)_{mn} = -\kappa_{mi}\kappa_{nj}p'_{ijk\ell}\frac{\partial u_k}{\partial x_\ell}, \qquad (2.4.3.7)$$

which is valid for small $\Delta\kappa$.

2.4.3.2. *Coupling via the electro-optic effect*

Piezoelectric media also exhibit an electro-optic effect linear in the applied electric field or in the field-induced crystal polarization. This effect is described in terms of the third-rank electro-optic tensor $\mathbf{r}$ defined by

$$(\Delta\kappa^{-1})_{ij} = r_{ijm}E_m. \qquad (2.4.3.8)$$

Using the same approach as in (2.4.2.10), for long waves E_m can be expressed in terms of $S_{k\ell}$, and (2.4.3.8) leads to an effective Pockels tensor $\mathbf{p}^e$ accounting for both the piezo-optic and the electro-optic effects:

$$p^e_{ijk\ell} = p_{ijk\ell} - \frac{r_{ijm}e_{nk\ell}\hat{Q}_m\hat{Q}_n}{\varepsilon_{gh}\hat{Q}_g\hat{Q}_h}. \qquad (2.4.3.9)$$

The total change in the inverse dielectric tensor is then

$$(\Delta\kappa^{-1})_{ij} = (p^e_{ijk\ell} + p_{ij[k\ell]})\frac{\partial u_k}{\partial x_\ell} = p'_{ijk\ell}\frac{\partial u_k}{\partial x_\ell}. \qquad (2.4.3.10)$$

The same equation (2.4.3.7) applies.

2.4.4. Brillouin scattering in crystals

2.4.4.1. *Kinematics*

Brillouin scattering occurs when an incident photon at frequency ν_i interacts with the crystal to either produce or absorb an acoustic phonon at $\delta\nu$, while a scattered photon at ν_s is simultaneously emitted. Conservation of energy gives

$$\delta\nu = \nu_s - \nu_i, \qquad (2.4.4.1)$$

where positive $\delta\nu$ corresponds to the anti-Stokes process. Conservation of momentum can be written

$$\mathbf{Q} = \mathbf{k}_s - \mathbf{k}_i, \qquad (2.4.4.2)$$

where $\mathbf{Q}$ is the wavevector of the emitted phonon, and $\mathbf{k}_s$, $\mathbf{k}_i$ are those of the scattered and incident photons, respectively. One can define unit vectors $\mathbf{q}$ in the direction of the wavevectors $\mathbf{k}$ by

$$\mathbf{k}_i = 2\pi\mathbf{q}n/\lambda_0, \qquad (2.4.4.3a)$$
$$\mathbf{k}_s = 2\pi\mathbf{q}'n'/\lambda_0, \qquad (2.4.4.3b)$$

where n and n' are the appropriate refractive indices, and λ_0 is the vacuum wavelength of the radiation. Equation (2.4.4.3*b*) assumes that $\delta\nu \ll \nu_i$ so that λ_0 is not appreciably changed in the scattering. The incident and scattered waves have unit polarization vectors $\mathbf{e}$ and $\mathbf{e}'$, respectively, and corresponding indices n and n'. The polarization vectors are the principal directions of vibration derived from the sections of the ellipsoid of indices by planes perpendicular to $\mathbf{q}$ and $\mathbf{q}'$, respectively. We assume that the electric vector of the light field $\mathbf{E}_{\rm opt}$ is parallel to the displacement $\mathbf{D}_{\rm opt}$. This is exactly true for many cases listed in the tables below. In the other cases (such as skew directions in the orthorhombic group) this assumes that the birefringence is sufficiently small for the effect of the angle between $\mathbf{E}_{\rm opt}$ and $\mathbf{D}_{\rm opt}$ to be negligible. A full treatment, including this effect, has been given by Nelson *et al.* (1972).

After substituting (2.4.4.3) in (2.4.4.2), the unit vector in the direction of the phonon wavevector is given by

$$\hat{\mathbf{Q}} = \frac{n'\mathbf{q}' - n\mathbf{q}}{|n'\mathbf{q}' - n\mathbf{q}|}. \qquad (2.4.4.4)$$

The Brillouin shift $\delta\nu$ is related to the phonon velocity V by

$$\delta\nu = VQ/2\pi. \qquad (2.4.4.5)$$

Since $\nu\lambda_0 = c$, from (2.4.4.5) and (2.4.4.3), (2.4.4.4) one finds

$$\delta\nu \cong (V/\lambda_0)[n^2 + (n')^2 - 2nn'\cos\theta]^{1/2}, \qquad (2.4.4.6)$$

where θ is the angle between $\mathbf{q}$ and $\mathbf{q}'$.

2.4.4.2. *Scattering cross section*

The power $\mathrm{d}P_{\rm in}$, scattered from the illuminated volume V in a solid angle $\mathrm{d}\Omega_{\rm in}$, where $P_{\rm in}$ and $\Omega_{\rm in}$ are measured inside the sample, is given by

$$\frac{\mathrm{d}P_{\rm in}}{\mathrm{d}\Omega_{\rm in}} = V\frac{k_BT\pi^2n'}{2n\lambda_0^4C}MI_{\rm in}, \qquad (2.4.4.7)$$

where $I_{\rm in}$ is the incident light intensity inside the material, $C = \rho V^2$ is the appropriate elastic constant for the observed phonon, and the factor k_BT results from taking the fluctuation–

dissipation theorem in the classical limit for $h\delta\nu \ll k_BT$ (Hayes & Loudon, 1978). The coupling coefficient M is given by

$$M = |e_m e'_n \kappa_{mi}\kappa_{nj} p'_{ijk\ell}\hat{u}_k \hat{Q}_\ell|^2. \qquad (2.4.4.8)$$

In practice, the incident intensity is defined outside the scattering volume, $I_{\rm out}$, and for normal incidence one can write

$$I_{\rm in} = \frac{4n}{(n+1)^2} I_{\rm out}. \qquad (2.4.4.9a)$$

Similarly, the scattered power is observed outside as $P_{\rm out}$, and

$$P_{\rm out} = \frac{4n'}{(n'+1)^2} P_{\rm in}, \qquad (2.4.4.9b)$$

again for normal incidence. Finally, the approximative relation between the scattering solid angle $\Omega_{\rm out}$, outside the sample, and the solid angle $\Omega_{\rm in}$, in the sample, is

$$\Omega_{\rm out} = (n')^2 \Omega_{\rm in}. \qquad (2.4.4.9c)$$

Substituting (2.4.4.9*a*,*b*,*c*) in (2.4.4.7), one obtains (Vacher & Boyer, 1972)

$$\frac{{\rm d}P_{\rm out}}{{\rm d}\Omega_{\rm out}} = \frac{8\pi^2 k_B T}{\lambda_0^4} \frac{n^4}{(n+1)^2} \frac{(n')^4}{(n'+1)^2} \beta V I_{\rm out}, \qquad (2.4.4.10)$$

where the coupling coefficient β is

$$\beta = \frac{1}{n^4 (n')^4} \frac{|e_m e'_n \kappa_{mi}\kappa_{nj} p'_{ijk\ell}\hat{u}_k \hat{Q}_\ell|^2}{C}. \qquad (2.4.4.11)$$

In the cases of interest here, the tensor $\boldsymbol{\kappa}$ is diagonal, $\kappa_{ij} = n_i^2\delta_{ij}$ without summation on i, and (2.4.4.11) can be written in the simpler form

$$\beta = \frac{1}{n^4 (n')^4} \frac{|e_i n_i^2 p'_{ijk\ell}\hat{u}_k \hat{Q}_\ell e'_j n_j^2|^2}{C}. \qquad (2.4.4.12)$$

2.4.5. Use of the tables

The tables in this chapter give information on modes and scattering geometries that are in most common use in the study of hypersound in single crystals. Just as in the case of X-rays, Brillouin scattering is not sensitive to the presence or absence of a centre of symmetry (Friedel, 1913). Hence, the results are the same for all crystalline classes belonging to the same centric group, also called Laue class. The correspondence between the point groups and the Laue classes analysed here is shown in Table 2.4.5.1. The monoclinic and triclinic cases, being too cumbersome, will not be treated here.

For tensor components $c_{ijk\ell}$ and $p_{ijk\ell}$, the tables make use of the usual contracted notation for index pairs running from 1 to 6. However, as the tensor $p'_{ijk\ell}$ is not symmetric upon interchange of (k, ℓ), it is necessary to distinguish the order (k, ℓ) and (ℓ, k). This is accomplished with the following correspondence:

$$1,1 \to 1 \quad 2,2 \to 2 \quad 3,3 \to 3$$
$$1,2 \to 6 \quad 2,3 \to 4 \quad 3,1 \to 5$$
$$2,1 \to \bar{6} \quad 3,2 \to \bar{4} \quad 1,3 \to \bar{5}.$$

Geometries for longitudinal modes (LA) are listed in Tables 2.4.5.2 to 2.4.5.8. The first column gives the direction of the scattering vector $\hat{\mathbf{Q}}$ that is parallel to the displacement $\hat{\mathbf{u}}$. The second column gives the elastic coefficient according to (2.4.2.6). In piezoelectric materials, effective elastic coefficients defined in (2.4.2.11) must be used in this column. The third column gives the direction of the light polarizations $\hat{\mathbf{e}}$ and $\hat{\mathbf{e}}'$, and the last column gives the corresponding coupling coefficient β [equation (2.5.5.11)]. In general, the strongest scattering intensity is obtained for polarized scattering ($\hat{\mathbf{e}} = \hat{\mathbf{e}}'$), which is the only situation listed in the tables. In this case, the coupling to light (β) is independent of the scattering angle θ, and thus the tables apply to any θ value.

Tables 2.4.5.9 to 2.4.5.15 list the geometries usually used for the observation of TA modes in backscattering ($\theta = 180^\circ$). In this case, $\hat{\mathbf{u}}$ is always perpendicular to $\hat{\mathbf{Q}}$ (pure transverse modes), and $\hat{\mathbf{e}}'$ is not necessarily parallel to $\hat{\mathbf{e}}$. Cases where pure TA modes with $\hat{\mathbf{u}}$ in the plane perpendicular to $\hat{\mathbf{Q}}$ are degenerate are indicated by the symbol D in the column for $\hat{\mathbf{u}}$. For the Pockels tensor components, the notation is $p_{\alpha\beta}$ if the rotational term vanishes by symmetry, and it is $p'_{\alpha\beta}$ otherwise.

Tables 2.4.5.16 to 2.4.5.22 list the common geometries used for the observation of TA modes in 90° scattering. In these tables, the polarization vector $\hat{\mathbf{e}}$ is always perpendicular to the scattering plane and $\hat{\mathbf{e}}'$ is always parallel to the incident wavevector of light $\mathbf{q}$. Owing to birefringence, the scattering vector $\hat{\mathbf{Q}}$ does not exactly bisect $\mathbf{q}$ and $\mathbf{q}'$ [equation (2.4.4.4)]. The tables are written for strict 90° scattering, $\mathbf{q}\cdot\mathbf{q}' = 0$, and in the case of birefringence the values of $\mathbf{q}^{(m)}$ to be used are listed separately in Table 2.4.5.23. The latter assumes that the birefringences are not large, so that the values of $\mathbf{q}^{(m)}$ are given only to first order in the birefringence.

2.4.6. Techniques of Brillouin spectroscopy

Brillouin spectroscopy with visible laser light requires observing frequency shifts falling typically in the range ~1 to ~100 GHz, or ~0.03 to ~3 cm^{-1}. To achieve this with good resolution one mostly employs interferometry. For experiments at very small angles (near forward scattering), photocorrelation spectroscopy can also be used. If the observed frequency shifts are ≥ 1 cm^{-1}, rough measurements of spectra can sometimes be obtained with modern grating instruments. Recently, it has also become possible to perform Brillouin scattering using other excitations, in particular neutrons or X-rays. In these cases, the coupling does not occur *via* the Pockels effect, and the frequency shifts that are observed are much larger. The following discussion is restricted to optical interferometry.

The most common interferometer that has been used for this purpose is the single-pass planar Fabry–Perot (Born & Wolf, 1993). Upon illumination with monochromatic light, the frequency response of this instrument is given by the Airy function, which consists of a regular comb of maxima obtained as the optical path separating the mirrors is increased. Successive maxima are separated by $\lambda/2$. The ratio of the maxima separation to the width of a single peak is called the finesse F, which increases as the mirror reflectivity increases. The finesse is also limited by the planarity of the mirrors. A practical limit is $F \sim 100$. The resolving power of such an instrument is $R = 2\ell/\lambda$, where ℓ is the optical thickness. Values of R around 10^6 to 10^7 can be achieved. It is impractical to increase ℓ above ~5 cm because the luminosity of the instrument is proportional to $1/\ell$. If higher

Table 2.4.5.1. *Definition of Laue classes*

Crystal system	Laue class	Point groups
Cubic	C_1 C_2	$432, \bar{4}3m, m\bar{3}m$ $23, \bar{3}m$
Hexagonal	H_1 H_2	$622, 6mm, \bar{6}2m, 6/mm$ $6, \bar{6}, 6/m$
Tetragonal	T_1 T_2	$422, 4mm, \bar{4}2m, 4/mm$ $4, \bar{4}, 4/m$
Trigonal	R_1 R_2	$32, 3m, \bar{3}m$ $3, \bar{3}$
Orthorhombic	O	$mmm, 2mm, 222$

references