

2.4. BRILLOUIN SCATTERING

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

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

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

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

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

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

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

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

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

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

where the coupling coefficient β is

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

In the cases of interest here, the tensor κ 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

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

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_{ijkl} and p_{ijkl} , the tables make use of the usual contracted notation for index pairs running from 1 to 6. However, as the tensor p'_{ijkl} 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:

$$\begin{aligned} 1, 1 &\rightarrow 1 & 2, 2 &\rightarrow 2 & 3, 3 &\rightarrow 3 \\ 1, 2 &\rightarrow 6 & 2, 3 &\rightarrow 4 & 3, 1 &\rightarrow 5 \\ 2, 1 &\rightarrow \bar{6} & 3, 2 &\rightarrow \bar{4} & 1, 3 &\rightarrow \bar{5}. \end{aligned}$$

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{Q} that is parallel to the displacement \hat{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{e} and \hat{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 ($\hat{e} = \hat{e}'$), 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{u} is always perpendicular to \hat{Q} (pure transverse modes), and \hat{e}' is not necessarily parallel to \hat{e} . Cases where pure TA modes with \hat{u} in the plane perpendicular to \hat{Q} are degenerate are indicated by the symbol D in the column for \hat{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{e} is always perpendicular to the scattering plane and \hat{e}' is always parallel to the incident wavevector of light \mathbf{q} . Owing to birefringence, the scattering vector \hat{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	432, $\bar{4}3m$, $m\bar{3}m$
	C_2	23, $\bar{3}m$
Hexagonal	H_1	622, $6mm$, $\bar{6}2m$, $6/mmm$
	H_2	6, $6/m$
Tetragonal	T_1	422, $4mm$, $\bar{4}2m$, $4/mmm$
	T_2	4, $4/m$
Trigonal	R_1	32, $3m$, $\bar{3}m$
	R_2	3, $\bar{3}$
Orthorhombic	O	mmm , $2mm$, 222