THERMAL SCATTERING OF LIGHT IN CUBIC CRYSTALS

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SUMMARY

The theory for the intensities of Doppler components in birefringent crystals developed in Part IV (Chandrasekharan, 1952) has been applied to the case of cubic crystals and general expressions for the intensities for 11 specific orientations of crystals belonging to T_d, O_h and O classes and 6 crystal orientations of T and T_h crystal classes have been derived. The formulæ have been used to calculate the intensities in the case of diamond and other cubic crystals and the results compared with the experimental data in the former case. There is not much agreement between theory and experiment.

1. INTRODUCTION

In a series of papers on the thermal scattering of light in crystals which appeared in the *Proceedings of the Indian Academy of Sciences*, the author (Chandrasekharan, 1951, 1952) has given the theory of light scattering in birefringent crystals and the expressions for calculating the intensities of the 12 pairs of Doppler components that can arise, in general, in these crystals. The theory is based mainly on Mueller's theory of scattering for isotropic solids. In this paper the case of cubic crystals is considered and general expressions for the intensities of the components for 11 specific crystal orientations are derived.

Cubic crystals belonging to T_h, T_d and O_h classes are singly refracting, those belonging to the other two classes T and O which can exhibit optical activity, are strictly double refracting since the refractive indices of these crystals for two circularly polarised waves with opposite senses of rotation are different. As this difference is negligibly small for most crystals even in the ultraviolet, for, *e.g.*, in the case of sodium chlorate it is only 0·00002 at λ 2537, it can be ignored in calculating the Doppler shifts and all cubic crystals may be regarded as singly refracting. Therefore, as already seen in Part III, there could, in general, be only three distinct pairs of Doppler components each of which has strictly a degeneracy of four. For calculating the intensities of these in equations (10) of Part IV, we can take arbitrarily any two orthogonal vectors normal to the propagation direction of the incident wave as the vectors $\vec{A}$ and $\vec{B}$ and similarly any two arbitrary orthogonal vectors in the scattered wave front as the vectors $\vec{P}$ and $\vec{Q}$. For convenience, we choose $\vec{A}$ and $\vec{B}$ to be parallel (h) and perpendicular (v) to the plane of scattering T. Similar components of the scattered light are $\vec{H}$ and $\vec{V}$. However, even this definition becomes arbitrary when backward scattering is considered and in these cases, the chosen directions for the various vectors are specified in the calculations. Cubic crystals have only three independent elastic constants c_{11}, c_{12} and c_{44}. Now when a medium is elastically isotropic, as for example a glass, a relation

$$c_{11} - c_{12} - 2c_{44} = 0 \tag{1}$$

subsists between the three constants. In cubic crystals, therefore, the quantity

$$\gamma = c_{11} - c_{12} - 2c_{44} \tag{2}$$

may be defined as the elastic anisotropy factor since it determines the variation in the elastic behaviour of the crystal for different directions. For example, for constant scattering angle θ, the Doppler shifts should depend on the crystal orientation if γ is finite.

In the case of crystal classes T_d, O and O_h there are only three independent elasto-optic constants p_{11}, p_{12} and p_{44}. As in the case of elastic constants a relation

$$p_{11} - p_{12} - 2p_{44} = 0 \tag{3}$$

exists for amorphous solids. In the case of cubic crystals (T_d, O and O_h) a quantity K given by

$$p_{11} - p_{12} - 2p_{44} = K \tag{4}$$

may be designated as the elasto-optic anisotropy factor. The values of K and γ determine the variations in the intensity of the Doppler components with crystal orientation, for constant θ. In the case of crystal classes T_h and T, there are four elasto-optic constants p_{11}, p_{12}, p_{13} and p_{44}, where $p_{12} \neq p_{13}$ (Bhagavantam, 1942).

In cubic crystals of all classes, one of the three types of elastic waves is strictly longitudinal and the other two transverse if the direction of propagation coincides with a cubic, dodecahedral or octahedral direction in the crystal. Tables I–IV contain the expressions for the stiffness coefficient and the intensity of scattering for the various Doppler components in backward and transverse scattering in a number of particular cases. Only those cases have been chosen where the elastic wave is propagated along a cubic, dodecahedral, or octahedral direction, the plane of scattering being different. The method of evaluation is based on equations (10), Part IV. It is best to proceed step by step in any specific case rather than get a formidable expression for intensities containing the direction cosines of the incident and observation directions and the vectors $\vec{h}$, $\vec{v}$, $\vec{H}$ and $\vec{V}$ for an arbitrary crystal orientation.

2. EXPLANATION OF TABLES

The expressions for the intensities in the various cases have only relative significance and for obtaining the absolute values of I, they must be multiplied by constant factor of $(\pi^2 k\mathrm{T}/2\lambda^4)\, n^8$, where n, the refractive index, is the same for all crystal orientations. Tables I and II give the results for crystal classes T_d, O and O_h for which $p_{12} = p_{13}$ and Tables III and IV give the results for crystal classes T and T_h for which $p_{12} \neq p_{13}$ and a term $\delta = (p_{12} - p_{13})/2$ which is finite is used. Tables I and III give the results for backward scattering and Tables II and IV for transverse scattering. We shall first discuss the results for the case $p_{12} = p_{13}$.

3. COMPARISON OF THE PRESENT THEORY WITH THE EINSTEIN-SMOLUCHOWSKI THEORY

In these cases, a perusal of Tables I and II shows that when $\mathrm{K} = 0$ and $\gamma = 0$, as happens for amorphous substances, the intensity of the scattered light is the same for all orientations provided the scattering angle θ is fixed. This affords a check on the calculations. When θ is 90°, the sum of the intensities of all the components is given by

$$\mathrm{I} = \mathrm{I}_1 + \mathrm{I}_2 + \mathrm{I}_3 = \frac{\pi^2 k\mathrm{T}}{2\lambda^4}\, n^8 \left\{ \frac{(p_{12}{}^2 + p_{44}{}^2)}{c_{11}} + \frac{p_{44}{}^2}{c_{44}} \right\} \tag{5}$$

This result is identical with that of Mueller (1938). To analyse this expression let us consider a solid which cannot be made optically anisotropic, *i.e.*, $p_{44} = 0$ (for most cubic crystals and amorphous substances $p_{44} \ll p_{12}$ and this assumption is thereby justified in actual cases also). Then only the density fluctuations can produce corresponding local fluctuations in the refractive index. Hence the first term $(p_{12}{}^2/c_{11})$ inside the double brackets of equation (5) arises essentially from density scattering and the other terms are due only to anisotropy or orientation scattering. This analysis was made by Mueller (*loc. cit.*). But we can go a step further and compare the results thus obtained with the well known expression of Einstein and Smoluchowski, which was first used by Sir C. V. Raman (1922) in the study of the thermal scattering of light in quartz. According to the Einstein-Smoluchowski expression, the intensity of light scattered in a transverse direction by a unit volume per unit solid angle is given by

$$\mathrm{I} = \frac{\pi^2 k\mathrm{T}}{2\lambda^4}\, \beta\, \frac{(n^2-1)^2\,(n^2+2)^2}{9}, \tag{6}$$

TABLE I

Backward Scattering in Crystal Classes T_d, *O* and O_h

No.	Direction of Wave Normal $\vec{R}_e$	Effective Elastic Constant q_e and Vibrn. Direction	V_v	H_h	$H_v=V_h$	I
1	[100]	L c_{11} [100]	p_{12}^2	p_{12}^2	0	$2p_{12}^2/c_{11}$
	$\vec{v}=\vec{V}=[010]$	T c_{44} [010]	0	0	0	0
		T c_{44} [001]				
2	[111]	L $c_{11}-2\gamma/3$ [111]	$(p_{12}+K/3)^2$	$(p_{12}+K/3)^2$	0	$2\,(p_{12}+K/3)^2/(c_{11}-2\gamma/3)$
	$\vec{v}=\vec{V}=[11\bar{2}]$	T $c_{44}+\gamma/3$ $[11\bar{2}]$	$K^2/18$	$K^2/18$	0	$2K^2/9\,(c_{44}+\gamma/3)$
		T $c_{44}+\gamma/3$ $[1\bar{1}0]$	0	0	$K^2/18$	
3	[110]	L $c_{11}-\gamma/2$ [110]	p_{12}^2	$(p_{12}+K/2)^2$	0	$\{p_{12}^2+(p_{12}+K/2)^2\}/(c_{11}-\gamma/2)$
	$\vec{v}=\vec{V}=[001]$	T c_{44} [001]	0	0	0	0
		T $c_{44}+\gamma/2$ $[1\bar{1}0]$	0	0	0	0

TABLE II

Transverse Scattering in Crystal Classes T_d, O and O_h

No.	Direction of Wave Normal $\vec{R}_e$	Effective Elastic Constant q_e	Plane of Scattering T	V_v	H_v	V_u	H_h	I
1	[100]	Lc_{11} $\left.\begin{matrix}Tc_{44}\\Tc_{44}\end{matrix}\right\}$	Arbitrary	p_{12}^2 0	0 $p_{44}^2/2$	0 $p_{44}^2/2$	$(p_{44}+K/2)^2$ 0	$\{p_{12}^2+(p_{44}+K/2)^2\}/c_{11}$ p_{44}^2/c_{44}
2	[111]	$L\,c_{11}-2\gamma/3$ $\left.\begin{matrix}T\,c_{44}+\gamma/3\\T\,c_{44}+\gamma/3\end{matrix}\right\}$	$[\bar{1}10]$ $\vec{I}=[441]$; $\vec{S}=[11\bar{8}]$	$(p_{12}+K/3)^2$ $K^2/18$	0 $(p_{44}+K/3+K/3\sqrt{2})^2/2$	0 $(p_{44}+K/3-K/3\sqrt{2})^2/2$	p_{44}^2 $K^2/72$	$\{(p_{12}+K/3)^2+p_{44}^2\}/(c_{11}-2\gamma/3)$ $\{(p_{44}+K/3)^2+K^2/8\}/(c_{44}+\gamma/3)$
3	[111]	$L\,c_{11}-2\gamma/3$ $\left.\begin{matrix}T\,c_{44}+\gamma/3\\T\,c_{44}+\gamma/3\end{matrix}\right\}$	$[\bar{1}10]$ $\vec{I}=[11\bar{8}]$, $\vec{S}=[441]$	$(p_{12}+K/3)^2$ $K^2/18$	0 $(p_{44}+K/3-K/3\sqrt{2})^2/2$	0 $(p_{44}+K/3+K/3\sqrt{2})^2/2$	p_{44}^2 $K^2/72$	$\{(p_{12}+K/3)^2+p_{44}^2\}/(c_{11}-2\gamma/3)$ $\{(p_{44}+K/3)^2+K^2/8\}/(c_{44}+\gamma/3)$
4	[111]	$L\,c_{11}-2\gamma/3$ $\left.\begin{matrix}T\,c_{44}+\gamma/3\\T\,c_{44}+\gamma/3\end{matrix}\right\}$	[112]	$(p_{12}+K/3)^2$ $K^2/18$	0 $\{(p_{44}+K/3)^2+K^2/18\}/2$	0 $\{(p_{44}+K/3)^2+K^2/18\}/2$	p_{44}^2 $K^2/72$	$\{(p_{12}+K/3)^2+p_{44}^2\}/(c_{11}-2\gamma/3)$ $\{(p_{44}+K/3)^2+K^2/8\}/(c_{44}+\gamma/3)$
5	$[1\bar{1}0]$	$L\,c_{11}-\gamma/2$ $T\,c_{44}$ $T\,c_{44}+\gamma/2$	[001]	p_{12}^2 0 0	0 $p_{44}^2/2$ 0	0 $p_{41}^2/2$ 0	p_{44}^2 0 0	$(p_{12}^2+p_{44}^2)/(c_{11}-\gamma/2)$ p_{44}^2/c_{44} 0
6	$[1\bar{1}0]$	$L\,c_{11}-\gamma/2$ $T\,c_{44}$ $T\,c_{44}+\gamma/2$	[111]	$(p_{12}+K/3)^2$ 0 0	$K/^236$ $p_{44}^2/6$ $(p_{44}+K/2)^2/3$	$K^2/36$ $p_{44}^2/6$ $(p_{44}+K/2)^2/3$	$(p_{44}+K/6)^2$ 0 0	$\{(p_{12}^2+K/3)^2+(p_{44}+K/6)^2+K^2/18\}/(c_{11}-\gamma/2)$ $p_{44}^2/3c_{44}$ $2\,(p_{44}+K/2)^2/3\,(c_{44}+\gamma/2)$
7	$[1\bar{1}0]$	$L\,c_{11}-\gamma/2$ $T\,c_{44}$ $T\,c_{44}+\gamma/2$	[110]	$(p_{12}+K/2)^2$ 0 0	0 0 $(p_{44}+K/2)^2/2$	0 0 $(p_{44}+K/2)^2/2$	$(p_{44}+K/2)^2$ 0 0	$\{(p_{12}+K/2)^2+(p_{44}+K/4)^2\}/(c_{11}-\gamma/2)$ 0 $(p_{44}+K/2)^2/(c_{44}+\gamma/2)$
8	$[1\bar{1}0]$	$c_{11}-\gamma/2$ c_{44} $c_{44}+\gamma/2$	$[11\bar{2}]$	$(p_{12}+K/6)^2$ 0 0	$K^2/36$ $p_{44}^2/3$ $(p_{44}+K/2)^2/6$	$K^2/36$ $p_{44}^2/3$ $(p_{44}+K/2)^2/6$	$(p_{44}+K/12)^2$ 0 0	$\{(p_{12}+K/6)^2+(p_{44}+K/12)^2+K^2/18\}/(c_{11}-\gamma/2)$ $2p_{44}^2/3c_{44}$ $(p_{44}+K/2)^2/3\,(c_{41}+\gamma/2)$

TABLE III

Backward Scattering in Crystal Classes T and T_h

$\bar{p}_{12} = (p_{12} + p_{13})/2$; $\delta = (p_{12} - p_{13})/2$; $\bar{\mathrm{K}} = p_{11} - \bar{p}_{12} - 2p_{44}$

No.	Direction of Wave Normal $\vec{\mathrm{R}}_e$	Elastic Constant q_e and Vibration Direction	V_v	H_h	$\mathrm{H}_v = \mathrm{V}_h$	I
1	[100]	c_{11} [100]	p_{13}^2	p_{12}^2	0	$(p_{12}^2+p_{13}^2)/c_{11}$
	$\vec{v}=\vec{\mathrm{V}}=[010]$	c_{44} [010]	0	0	0	0
		c_{44} [001]				
2	[111]	$c_{11} - 2\gamma/3$ [111]	$(\bar{p}_{12}+\bar{\mathrm{K}}/3)^2$	$(\bar{p}_{12}+\bar{\mathrm{K}}/3)^2$	0	$2\,(\bar{p}_{12}+\bar{\mathrm{K}}/3)^2/(c_{11}-2\gamma/3)$
	$\vec{v}=\vec{\mathrm{V}}=[11\bar{2}]$	$c_{44} + \gamma/3$ $[11\bar{2}]$	$\bar{\mathrm{K}}^2/18$	$\bar{\mathrm{K}}^2/18$	$\delta^2/6$	$2\,({}^{\mathrm{K}_2}+3\delta^2)/9\,(c_{44}+\gamma/3)$
		$c_{44} + \gamma/3$ $[1\bar{1}0]$	$\delta^2/6$	$\delta^2/6$	$\bar{\mathrm{K}}^2/18$	
3	[110]	$c_{11} - \gamma/2$ [110]	$\bar{p}_{12}^2$	$(\bar{p}_{12}+\bar{\mathrm{K}}/2)^2$	0	$\{(\bar{p}_{12}^2+(\bar{p}_{12}+\bar{\mathrm{K}}/2)^2\}/(c_{11}-\gamma/2)$
	$\vec{v}=\vec{\mathrm{V}}=[001]$	c_{44} [001]	0	0	0	0
		$c_{44} + \gamma/2$ $[1\bar{1}0]$	δ^2	$(\delta/2)^2$	0	$5\delta^2/4\,(c_{44}+\gamma/2)$

TABLE IV

Transverse Scattering in Crystal Classes T and T_h

No.	Direction of Wave Normal $\vec{R}_c$	Elastic Constant q_c and Vibration Direction	Plane of Scattering T	V_v	$H_v = V_h$	H_h	I
1	[100]	c_{11} [100]	[011]	p_{13}^2	0	$(p_{11}-p_{12})^2/2$	$\{p_{13}^2+(p_{11}-p_{12})^2\}/c_{11}$
		c_{44} [010]		0	$p_{44}^2/2$	$\left.\begin{matrix}0\\0\end{matrix}\right\}$	p_{44}^2/c_{44}
		c_{44} [001]		0	0		
2	[100]	c_{11} [100]	[001]	p_{12}^2	0	$(p_{11}-p_{13})^2/2$	$\{p_{12}^2+(p_{11}-p_{13})^2\}/c_{44}$
		c_{44} [010]		0	0	$\left.\begin{matrix}0\\0\end{matrix}\right\}$	p_{44}^2/c_{44}
		c_{44} [001]		0	$p_{44}^2/2$		
3	[110]	$c_{11}-\gamma/2$ [110]	[001]	$\bar{p}_{12}^2$	0	p_{44}^2	$(\bar{p}_{12}^2+p_{44}^2)/c_{11}$
		c_{44} [001]		0	$p_{44}^2/2$	0	p_{44}^2/c_{44}
		$c_{44}+\gamma/2$ [1$\bar{1}$0]		δ^2	0	0	$\delta^2/(c_{44}+\gamma/2)$

where β is the compressibility of the medium. In the derivation of (6) it is assumed that the Lorentz-Lorenz expression

$$\frac{n^2-1}{n^2+1} = a\rho \tag{7}$$

is valid, a being a constant and ρ the density of the substance. Differentiating the expression (7) we get

$$\rho\frac{dn}{d\rho} = (n^2-1)(n^2+2)/6n \tag{8}$$

For hydrostatic pressure the principal strains are equal and are given by

$$x_x = y_y = z_z = \frac{dV}{3V} = \frac{d\rho}{3\rho}. \tag{9}$$

But from Part IV Equation (26)

$$\begin{aligned}\Delta\epsilon = 2n.\Delta n &= n^4(p_{11}x_x + p_{12}y_y + p_{13}z_z)\\ &= n^4\frac{(p_{11}+2p_{12})\Delta\rho}{3\rho}.\end{aligned} \tag{10}$$

But

$$p_{11} = p_{12} \text{ since } p_{11} - p_{12} = 2p_{44} = 0 \tag{11}$$

$$\rho\frac{dn}{d\rho} = n^4\frac{(p_{11}+2p_{12})}{6n} = \frac{n^4 \cdot p_{12}}{2n}. \tag{12}$$

Comparing equations (12) and (8) we have

$$(n^2-1)(n^2+2)/3 = n^4 p_{12} \tag{13}$$

or

$$\frac{(n^2-1)^2(n^2+2)^2}{9} = n^8 p_{12}^2 \tag{14}$$

Substituting (14) in (6) we have

$$I = \frac{\pi^2 kT}{2\lambda^4}\beta n^8 p_{12}^2. \tag{15}$$

This expression is the same as the first term of expression (5) except for the fact that in the latter $1/c_{11}$ replaces β. But these do not differ very much in order of magnitude and therefore the results of the present theory agree with those derived from the Einstein-Smoluchowski expression. But in crystals, the Lorentz-Lorenz relation is not necessarily valid (Ramachandran, 1947). Further the scattered intensity in a given direction is due only to a few elastic waves traversing the close neighbourhood of a specified direction and it is more appropriate to consider the photoelastic effects produced by them rather than consider the density fluctuations arising from all possible elastic waves in the crystal.

4. DISCUSSION OF THE RESULTS IN TABLES I AND II

Coming to particular cases, if the directions of incidence and of observation are parallel to any two cubic directions (case 5 of Table II), the sum of the intensities of the three pairs of components, in the light scattered by unit volume of the scattering substance per unit solid angle is

$$I = \frac{\pi^2 kT}{2\lambda^4} n^8 \left\{ \frac{2(p_{12}^2+p_{44}^2)}{(c_{11}+c_{12}+2c_{44})} + \frac{p_{44}^2}{c_{44}} \right\}. \tag{16}$$

This expression was first derived by Leontowitsch and Mandelstam (1932) and is thus seen to be valid only in the particular case stated. In this case the effective elastic wave coincides with a dodecahedral direction and the plane of scattering is a cubic plane. However, a perusal of Table II, cases 5 to 8, shows that whereas the velocities of the elastic waves and consequently, the shifts of the Doppler components depend only on the direction of the elastic wave normal $\vec{R}_e$, the intensity of light scattered depends also on the plane of scattering. Thus, if the wave normal is along a dodecahedral direction, the transverse wave with $q = c_{44} + \gamma/2$ is ineffective in scattering when the plane of scattering T coincides with the cubic plane (case 5), while the other transverse wave with $q = c_{44}$ is ineffective in scattering when T is the dodecahedral plane (case 7). It is also interesting to note that the sum of the intensities for cases (5) and (7) is not the same as the sum of the intensities for cases (6) and (8). That is to say the sum of the intensities of the components due to the elastic wave traversing a specific direction [110] when observed in mutually perpendicular planes, (5) and (7); or (6) and (8) of Table II, is not the same even though the Doppler shift is the same in all the four cases. All these results arise from the fact that the intensity of light scattering depends on *two fourth order* tensors, *viz.*, elastic and elasto-optic tensors.

When the elastic wave normal $\vec{R}_e$ coincides with the cubic or octahedral direction, there is complete degeneracy (both elastic and optical) in the plane at right angles to $\vec{R}_e$. Hence the shifts as well as the intensities of all the Doppler components are independent of the plane of scattering.

In the case of backward scattering along a 2-fold axis [110] or a 4-fold axis [100] the intensity of the transverse components is zero (case 3 of Table I).

5. INTERCHANGE OF DIRECTIONS OF INCIDENCE AND OF SCATTERING

When the directions of incidence and of scattering are interchanged, $\vec{h}$ and $\vec{v}$ get interchanged with $\vec{H}$ and $\vec{V}$ and consequently, while V_v and H_h

remain unaltered in the process, V_h and H_v get interchanged. But the sum of the intensities of the various components is unaltered. In general, V_h is not equal to H_v. See cases (2) and (3) of Table II. Only if the effective elastic wave vector is parallel to an even fold axis of symmetry (2, 4 or 6-fold) or if there is a plane of reflection parallel to it and perpendicular to the plane of scattering will the relation $H_v = V_h$ be valid.

6. DEPOLARISATION FACTORS

Although not given in the above tables, the depolarisation factors of the Doppler components ρ_u for unpolarised light, ρ_v and ρ_h for polarised light can be calculated from the formulæ given below, knowing the values of H_h, H_v, V_h and V_v

$$\rho_u = (H_v + H_h)/(V_v + V_h) \tag{17}$$

$$\rho_v = H_v/V_v \tag{18}$$

$$\rho_h = V_h/H_h \tag{19}$$

If the incident light is polarised, the question arises whether in the light scattered the two components V and H are coherent or not. The derivation of the results shows that the components V and H arising from any particular elastic wave q_1, q_2 or q_3 should be coherent. Consequently, the light scattered should be plane or elliptically polarised but not partially polarised. However, there is no question of any phase relationship existing between the three elastic waves giving rise to scattering since the shifts of the components due to each wave is different. Even in the case of transverse degenerate waves travelling along the cubic or octahedral directions with the same velocity, the effects of the two waves should be treated separately and the intensities added. This procedure is similar to the calculation of intensities of degenerate Raman lines in cubic crystals (Couture and Mathieu, 1948). The degeneracy of the elastic waves exists only for specific directions and for even a very small departure of the elastic wave normal from these directions the velocities of the waves are different.

7. T and T_h CRYSTAL CLASSES

For these crystals the result $p_{12} \neq p_{13}$ follows from the fact that the cubic directions are only 2-fold axes of symmetry and not 4-fold as in the other crystal classes of cubic symmetry. The definition of X, Y and Z axes are given in the paper on Photo-Elastic Constants of Sodium Chlorate (Ramachandran and Chandrasekharan, 1951). As is to be expected, the formulæ given in Tables III and IV reduce to the corresponding expressions of Tables I and II if we put $(p_{12} - p_{13})/2 = \delta = 0$. This affords a check on the calculations.

For compression or dilatation along a cubic direction, the crystal becomes biaxial and not uniaxial as happens when $p_{12}=p_{13}$. Consequently, in light scattering this leads to interesting results. As will be noticed for backward scattering along the cubic direction, say the X-axis [010], the components of light scattered parallel to OY and OZ respectively are of different intensity. For incident unpolarised light the scattered light should be polarised. The presence of the four 3-fold axes of symmetry gives rise to the equivalence of only $(OX)_Y=(OY)_Z=(OZ)_X$ and $(OX)_Z=(OY)_X=(OZ)_Y$. But $(OX)_Y \neq (OX)_Z$ nor even $(OX)_Y \neq (OY)_X$, where $(OX)_Y$, etc., mean the component of the light scattered parallel to OY for backward scattering along OX.

Again in transverse light scattering, when the elastic wave normal coincides with the cubic direction, the intensity of the longitudinal Doppler components would vary for different planes of scattering. It is interesting to remark that these results are in striking contrast to the case of Raman scattering in which all classes of cubic symmetry are equivalent and these differences should not exist.

8. CRITICAL DISCUSSION OF ASSUMPTIONS

Firstly, we have assumed following Brillouin that the scattering in a given direction is due only to elastic waves traversing a specified direction. The fact that sharp Brillouin components appear in the thermal scattering of crystals justifies this assumption. Secondly, it is assumed that the velocity of elastic waves is given by the solution of the Christoffel determinant. This is justified because the wavelength of the sound waves is comparable with that of light except for very small angles of scattering and the effect of boundaries of the crystal on the velocity of the elastic waves even in small specimens is negligible. Since $\lambda_e \gg d$, the crystal spacing, the dispersion of velocity with the wavelength is not important. Thirdly, the effect of the elastic wave is assumed to be given by photo-elastic laws which is also reasonable since the wavelength of the elastic waves is of the same order as that of the light wave.

9. APPLICATION OF FORMULÆ TO VARIOUS CRYSTALS

The expressions given in Tables I–IV have been directly applied to the following crystals—diamond, calcium fluoride, lithium fluoride, sodium chloride, potassium chloride, potassium bromide, potassium alum, ammonium alum and sodium chlorate for which elastic and elasto-optic data are available. The last three crystals belong to T_h and T classes and expressions of Tables III and IV should be used for them. But in the case of the two alums the difference $(p_{12}-p_{13})/2=\delta$ is so small that it has been ignored and calculations made using only Tables I and II. Only in the case of sodium chlorate δ is comparatively large and the calculations have therefore been made separately in this case. The data are presented in the following Tables V to VIII. In the case of diamond the intensities of the Doppler components for cases other than those specified in Tables I and II have also been made so as to correspond to the orientations employed with the particular specimens studied.

10. DISCUSSION OF RESULTS

(*a*) *Diamond.*—Diamond is exceptional in its elasto-optic properties since p_{44} is very large and greater than even p_{12}. Also the elasto-optic anisotropy factor K is very large. Now although all the constants figure in the expression for the intensity of each component, the value of p_{44} mainly determines the intensity of the transverse components, while p_{12} determines that of the longitudinal components. Therefore, in the case of diamond the transverse components should in general be brighter than the longitudinal components unlike in the case of other cubic crystals. Further, since K is also large, the ratio of the intensities of the components should vary markedly with the orientation of the crystal. In the particular case when the effective elastic wave normal coincides with or is very near to the cubic axis (cases 1 and 9 of Tables VI) the longitudinal components should be brighter than the transverse components. Again when the elastic wave normal coincides with or is very near to the octahedral direction (cases 2 and 10) of Table VI the transverse component with a separation of 4·35 cm.$^{-1}$ should be recorded when the plane of scattering coincides with [110] but the other transverse component should have zero or very small intensity. In backward scattering along the [111] direction, the transverse components should be brighter than the longitudinal components. In the case of backward scattering along [110] direction, the transverse components should not be recorded while the longitudinal components should be very weak.

(*b*) *Comparison with Experimental Results.*—Two pairs of Doppler components in diamond have been recorded by R. S. Krishnan (1947) using the E_1 spectrograph, the outer arising from longitudinal and the inner from transverse sound waves. Recently, for four different orientations, these components have been more clearly resolved using the three-metre quartz spectrograph (Part II, Chandrasekharan, 1950). The ratio of the intensities of the longitudinal components to the transverse components has also been estimated for three different crystal orientations. It has established definitely that the ratio varies with orientation as is to be expected from theory. The

TABLE V

Backward Scattering in Diamond

No.	Direction of Wave Normal $\vec{R}_e$	$q_e \times 10^{-11}$	Vibration Direction	Velocity in km./sec.	Doppler Separation in cm.$^{-1}$	Intensities				
						V_v	H_h	$V_h = H_v$	$I = \Sigma V_v \times 10^{15}$	$I = I_1 + I_2 + I_3 \times 10^{15}$
1	[100]	95	[100]	16,440	11·34	0·85	0·85	0	1·7	1·7
	$\vec{v} = \vec{V} = [010]$	43	[010]	11,060	7·63	0	0	0	0	
		43	[001]	11,060	7·63	0	0	0	0	
2	[110]	110	[110]	17,690	12·20	0·01	0·74	0	0·75	
	$\vec{v} = \vec{V} = [1\bar{1}0]$	43	[001]	11,060	7·63	0	0	0	0	0·8
		28	$[1\bar{1}0]$	8,930	6·16	0	0	0	0	
3	[111]	115	[111]	18,090	12·47	0·12	0·12	0	0·23	
	$\vec{v} = \vec{V} = [11\bar{2}]$	33	$[11\bar{2}]$	9,700	6·69	0·43	0·43	0	0·86	2·0
		33	$[1\bar{1}0]$	9,700	6·69	0	0	0·43	0·86	

TABLE VI

Transverse Scattering in Diamond

No.	Direction of Wave Normal $\vec{R}_e$	Plane of Scattering T	$q_e \times 10^{-11}$	Vibrn. Dirn.	Doppler Separation in cm.$^{-1}$	Intensities						
						$V_{\tau'}$	$H_{\tau'}$	V_h	H_h	$I=\Sigma V_{\tau'}$	I_T/I_L	$I=I_1+I_2+I_3$
1	[100]	Arbitrary	95	[100]	8·02	0·85	0	0	4·21	5·06		
			43	[010]	5·40	0	1·68	1·68	0	3·35	0·66	8·4
			43	[001]	5·40							
2	[110]	$[1\bar{1}0]$	110	[110]	8·63	0·01	0	0	2·33	2·37		
			43	[001]	5·40	0	0	0	0	0	6·12	16·6
			28	$[1\bar{1}0]$	4·35	0	7·15	7·15	0	14·29		
3	[110]	$[1\bar{1}1]$	110	[110]	8·63	0·12	0·07	0·07	1·96	2·21		
			43	[001]	5·40	0	0·56	0·56	0	1·12	4·82	12·9
			28	$[1\bar{1}0]$	4·35	0	4·76	4·76	0	9·53		
4	[110]	[001]	110	[110]	8·63	0·74	0	0	1·31	2·05		
			43	[001]	5·40	0	1·68	1·68	0	3·35	1·64	5·4
			28	$[1\bar{1}0]$	4·35	0	0	0	0	0		
5	[110]	$[\bar{1}12]$	110	[110]	8·63	0·37	0·07	0·07	1·62	2·11		
			43	[001]	5·40	0	1·12	1·12	0	2·23	3·22	8·1
			28	$[1\bar{1}0]$	4·35	0	2·38	2·38	0	4·76		

TABLE VI—(*Contd.*)

No.	Direction of Wave Normal $\vec{R}_e$	Plane of Scattering T	$q_e \times 10^{-11}$	Vibrn. Dirn.	Doppler Separation in cm.$^{-1}$	Intensities						
						V_v	H_v	V_h	H_h	$I=\Sigma V_v$	I_T/I_L	$I=I_1+I_2+I_3$
6	[111]	$[1\bar{1}0]$	115	[111]	8·82	0·12	0	0	1·25	1·37		
			33	$[1\bar{1}0]$	4·73	0·43	6·75	2·79	0·11	10·07	7·35	11·4
			33	$[11\bar{2}]$	4·73							
7	[111]	$[1\bar{1}0]$	115	[111]	8·82	0·12	0	0	1·25	1·37		
			33	$[1\bar{1}0]$	4·73	0·43	2·79	6·75	0·11	10·07	7·35	11·4
			33	$[11\bar{2}]$	4·73							
8	[111]	$[11\bar{2}]$	115	[111]	8·82	0·12	0	0	1·25	1·37		
			33	$[1\bar{1}0]$	4·73	0·43	4·76	4·76	0·11	10·07	7·35	11·4
			33	[001]	4·73							
9	[8, 1, 1] $\vec{I}=[2\bar{1}\bar{1}]$; $\vec{S}=[\bar{1}\bar{1}\bar{1}]$	$[01\bar{1}]$	97·1	*l*	8·11	0·77	0	0	3·73	4·50		
			42·6	$[0\bar{1}1]$	5·37	0	1·66	1·73	0	3·40	0·83	8·2
			41·3	*t*	5·29	0·05	0	0	0·31	0·36		
10	$[\bar{1}, 4, 4]$ $\vec{I}=[2\bar{1}\bar{1}]$; $\vec{S}=[111]$	$[01\bar{1}]$	110·86	*l*	8·66	0·01	0	0	2·14	2·15		
			41·69	*t*	5·31	0	0	0	0·1	0·1	6·47	16·1
			28·45	$[0\bar{1}1]$	4·39	0	7·83	5·97	5·0	13·80		

TABLE VII

No.	Crystal	ρ g./c.c.	n for λ_{2537}	Elastic constants in 10^{11} dynes/cm.^{2}					Photo-elastic constants					$\frac{\pi^2 KTn^8}{2\lambda^4}$
				c_{11}	c_{12}	c_{44}	Y	Ref. No.	p_{11}	p_{12}	p_{44}	K	Ref. No.	
1	Diamond .	3·51	2·624	95	39	43	−30	3	−0·31	0·09	−0·12	−0·16	15	$1\cdot1\ \times10^{6}$
2	CaF_2 ..	3·18	1·467	16·44	5·01	3·47	4·48	2	0·056	0·227	0·024	0·219	14	$1\cdot05\times10^{7}$
3	LiF ..	2·60	1·418	11·90	4·58	5·42	−3·52	18	0·02	0·128	−0·064	0·020	7	$0\cdot80\times10^{7}$
4	NaCl ..	2·18	1·652	4·85	1·23	1·26	1·1	11	0·137	0·178	−0·011	−0·019	14	$2\cdot72\times10^{7}$
5	KBr ..	2·76	1·718	3·46	0·58	0·51	1·86	10	0·220	0·171	−0·026	0·101	7	$3\cdot72\times10^{7}$
6	KCl ..	1·98	1·585	3·98	0·62	0·63	2·11	10	0·229	0·170	−0·028	0·114	14	$1\cdot96\times10^{7}$
7	NH_4 alum ..	1·64	1·512	2·50	1·06	0·80	−0·16	18	0·38	0·46	−0·009	−0·062	6	$1\cdot34\times10^{7}$
8	K-alum ..	1·76	1·508	2·56	1·09	0·86	−0·25	17	0·27	0·345	0·0056	−0·064	4	$1\cdot31\times10^{7}$
9*	$NaClO_3$ ..	2·49	1·59	5·00	1·47	1·18	1·17	5, 12	0·173	0·243	−0·0187	−0·0153	16	$2\cdot00\times10^{7}$

* $p_{13} = 0\cdot258$, $p_{11} = 0\cdot223$.

TABLE VIII

Direction of Elastic Wave Normal $\vec{R}_e$		[100] $I_2=I_3=0$			[111]				[110] $I_3=0$				[110] $I_2=0$			[111]	
Angle of Scattering θ		180°			180*				90°				90°			90°	
Plane of scattering T		..			..				[001]				[$\bar{1}$10]			[11$\bar{2}$]	
$\vec{v} = \vec{V}$		[010]			[11$\bar{2}$]				[001]				[$\bar{1}$10]			[11$\bar{2}$]	
No.	Crystal	$\Delta\nu_1$	$I_1 \times 10^6$	$\Delta\nu_{2,3}$	$\Delta\nu_1$	$I_1 \times 10^6$	$\Delta\nu_{2,3}$	$I_2+I_3 \times 10^6$	$\Delta\nu_1$	$I_1 \times 10^6$	$\Delta\nu_2$	$I_2 \times 10^6$	$I_1 \times 10^6$	$\Delta\nu_3$	$I_3 \times 10^6$	$I_1 \times 10^6$	$I_2+I_3 \times 10^6$
	Diamond ..	11·34	1·87	7·63	12·47	0·25	6·69	1·90	8·63	2·26	5·40	3·69	2·61	4·35	15·72	1·51	11·01
2	CaF_2 ..	2·77	0·66	1·28	2·51	0·37	1·52	0·23	1·82	0·39	0·90	0·02	0·11	1·16	0·14	0·19	0·18
3	LiF ..	2·52	0·22	1·70	2·76	0·20	1·51	0·002	1·91	0·12	1·20	0·06	0·13	0·99	0·06	0·13	0·08
4	NaCl ..	2·05	3·55	1·04	1·89	3·9	1·17	0·01	1·36	2·01	0·74	0·03	1·80	0·88	0·07	1·96	0·06
5	KBr ..	1·60	6·29	0·60	1·28	14·09	0·91	0·75	0·97	4·40	0·42	0·49	7·18	0·73	0·16	2·84	0·44
6	KCl ..	1·87	2·84	0·74	1·34	6·59	1·08	0·42	1·13	1·98	0·54	0·24	3·44	0·86	1·05	3·35	0·25
7	NH_4-alum ..	1·55	22·7	0·88	1·59	19·81	0·85	0·15	1·12	11·0	0·62	0	9·60	0·59	0·30	9·91	0·25
8	K-alum ..	1·51	12·2	0·88	1·56	10·69	0·84	0·15	1·10	5·82	0·62	0·05	5·65	0·58	0·26	5·35	0·21
9	$NaClO_3$ ..	1·87	4·66	0·89	1·72	5·26	0·96	0·22	1·24	2·65	0·63	0·06	..	..	..	..	..
		*V_v = 1·99 ; H_h = 2·66							$\Delta\nu_3$ = 0·79 — I_3 = 0·03								

transverse components are always brighter than the longitudinal one, by a factor of about 1·5 to 3 times for the different orientations studied. This result is also in general accord with the theoretical result stated in the previous section. However, the theoretical prediction that for case 9 of Table VI (which was studied experimentally; case 2 of Table I, Part II) the longitudinal components should be brighter than the transverse components is in striking contradiction to the experimental result that the transverse components are brighter. Again for case 10 of Table VI (case 1 of Table I, Part II) transverse components recorded had a shift of about 5·6 cm.$^{-1}$ while according to theory, the 5·4 cm.$^{-1}$ components should have very little intensity and the 4·5 cm.$^{-1}$ components should be very bright. This disagreement between theory and experiment indicates that both on the theoretical as well as on the experimental side more work has to be done. On the experimental side it would be best to work with a large specimen and to record the Doppler components for backward scattering along the [110] direction (case 2 of Table V).

(*c*) *Other Crystals.*—It is seen from Tables VII and VIII that, since p_{44} is generally small for these crystals the intensity of light scattering due to the transverse wave is very much smaller than that due to the longitudinal waves. Further since the anisotropy factors K and γ are small compared to p_{12} and c_{11} respectively, the intensities of the components do not vary very much for different crystal orientations. Further the absolute intensity of scattering in LiF and in CaF_2 is far smaller than that in other crystals since it is proportional to n^8/q_e and since their refractive indices are small and elastic constants are large.

11. NOTE ON STIFFNESS COEFFICIENTS OF CUBIC CRYSTALS

While making the calculations for the stiffness coefficients when the effective elastic wave normal is along an arbitrary direction, it was noticed that the sum of the three stiffness coefficients is always the same and equal to $c_{11} + 2c_{44}$. This result can be proved rigorously by expanding the determinantal Christoffel equation (11) of Part IV giving the stiffness coefficients in terms of the elastic constants. On expanding, we have

$$(A_{11} - q)(A_{22} - q)(A_{33} - q) - A_{23}^2(A_{11} - q) - A_{13}^2(A_{12} - q) - A_{12}(A_{33} - q) + 2A_{12}A_{13}A_{23} = 0 \tag{19}$$

In this, the coefficient of $q^2 = A_{11} + A_{22} + A_{33}$ (20)

From equations (12) of Part IV, we have for cubic crystals.

$$A_{11} = c_{11}l^2 + c_{44}(m^2 + n^2)$$
$$A_{22} = c_{11}m^2 + c_{44}(n^2 + l^2)$$
$$A_{33} = c_{11}n^2 + c_{44}(l^2 + m^2)$$

Adding, the coefficient of $q^2 = A_{11} + A_{22} + A_{33} = c_{11}(l^2 + m^2 + n^2)$

$$+ 2c_{44}(l^2 + m^2 + n^2) = c_{11} + 2c_{44}, \text{ since } l^2 + m^2 + n^2 = 1 \qquad (21)$$

Now the coefficient of the square term q^2 in a 3rd degree equation (19) gives the sum of the three roots, q_1, q_2 and q_3.

Therefore,

$$q_1 + q_2 + q_3 = c_{11} + 2c_{44}. \qquad (22)$$

This relation is extremely useful in checking the numerical calculations in cases like 9 and 10 of Table VI.

However, no such simple relation could be found for the sum of the intensities of the various Doppler components in different settings of the crystal.

ACKNOWLEDGMENTS

The author wishes to express his sincere thanks to Prof. R. S. Krishnan and to Dr. G. N. Ramachandran for their keen interest in the work. He is also indebted to the National Institute of Sciences for the tenure of the Junior Research Fellowship.

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