***Proc Indian Acad Sci*, 33A, 183-198, (1951)**
Thermal scattering of light in crystals.III.Theory for birefringent crystal
V. Chandrasekharan

THERMAL SCATTERING OF LIGHT IN CRYSTALS

Part III. Theory for Birefringent Crystals

BY V. CHANDRASEKHARAN
(*From the Department of Physics, Indian Institute of Science, Bangalore*)

Received December 25, 1950
(Communicated by Prof. R. S. Krishnan, F.A.SC.)

1. INTRODUCTION

THE thermal scattering of light in crystals was established as an observable phenomenon by Sir C. V. Raman in 1922. In the same year appeared a theoretical paper by L. Brillouin (1922) in which the diffusion of light in a material medium was regarded as a "coherent reflection" of light waves by the sound waves of thermal origin traversing it, and it was shown that the reflection should be accompanied by shifts of frequency in the nature of a Doppler effect, which vary with the direction of observation of the scattered light and with the frequency of the incident light. Such shifts of frequency are identical with the frequency of the sound waves which are effective in scattering along the particular direction of observation. Hence they may also be regarded as a species of Raman effect. Their magnitude is, however, very much smaller than those associated with the infra-red vibrations of the crystal and hence much more difficult of observation than the latter. Indeed, the experimental proof of their existence followed and not preceded the discovery of the Raman effect in 1928.

Ever since, several investigators, notably Mandelstam, Landsberg and Leontowitsch (1930), Tamm (1930), Mueller (1938), Gross (1938, 1940), Bhatia and Krishnan (1948) and Kastler (1949) have considered the theory of thermal scattering of light in crystals. Along any specific direction, there are three types of elastic waves travelling with different velocities and the Doppler shifts depend on these velocities. Further, since the waves effect-tive in scattering along a particular direction are either approaching or receding, the picture that is generally accepted at present appears to be that there are only three doublets or pairs of Doppler-shifted components in the light scattered in a crystal whether birefringent or not. Such a conclusion is true for singly refracting crystals (or media) but, *in general*, not true for doubly refracting or *birefringent* crystals, because the Doppler shifts depend on the velocities of the incident and the scattered light waves inside the crystal and in each direction, there are two possible velocities for the light waves.

The effect of birefringence specifically on the Doppler components does not seem to have been considered till now. In this paper, the theory of thermal scattering applicable to birefringent crystals is worked out in detail and the new and extremely interesting results obtained are presented. The results of the previous investigators are thereby derived as a special case when the birefringence is absent. The present theory is for convenience divided into two sections. The expected number, frequency shifts, and polarisation characteristics of the spectral components of the scattered light, in a general case, are dealt with in the present paper while the theoretical evaluation of the intensities of the various components will be considered in a later communication. To derive the results, one can proceed either on the basis of quantum ideas or of classical wave theory.

2. Derivation Using Quantum Theory

The treatment of scattering on the basis of quantum theory has been worked out by Tamm (1930) (applicable only to singly refracting media). Using similar ideas, we consider a collision process between the quantum of light energy or *photon* and the quantum of the elastic energy or *phonon*. Let the direction of the incident light wave be I′CI and that of the scattered wave be CS, the angle of scattering being θ (Fig. 1).

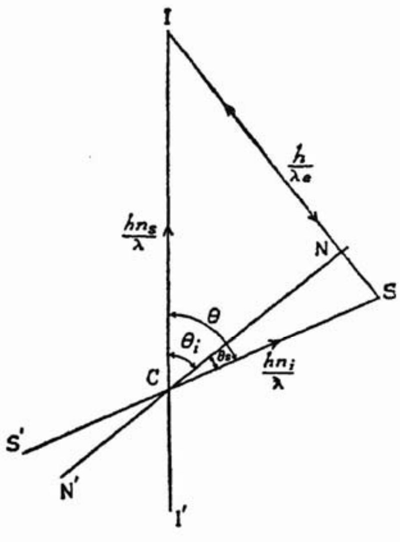


Fig. 1. Quantum picture of scattering

The momentum of a photon is given by dividing Planck's constant h by the wavelength of the light and is in the direction of the wave normal inside a birefringent crystal. For any particular direction of propagation of light, there are two possible wavelengths corresponding to the two polarised components, the wavelength of the incident (or scattered) wave being given by λ/n_i (or λ/n_s) where n_i (or n_s) is one of the two possible values of the refractive index of the crystal for the incident (or scattered direction) and λ the wavelength of light in vacuum. Therefore, the momenta of the incident and of the scattered photons are given by $h\,n_i/\lambda$ and $h\,n_s/\lambda$. Similarly the momentum of the phonon is h/λ_e where λ_e is the wavelength of the elastic wave. If now these momenta are represented vectorially by $\vec{CI}$, $\vec{CS}$ and $\vec{IS}$ the conservation of momentum in the collision process can be expressed as

$$\vec{CS}=\vec{CI}+\vec{IS}. \tag{1}$$

This vectorial equation is really equivalent to three conditions. Firstly, all the three vectors lie in a plane, *i.e.*, the wave normals of the incident and scattered light and that of the elastic waves should lie in a plane. This is analogous to the law of reflection (or refraction), *viz.*, that the incident and reflected (or refracted) wave normals lie in the plane of reflection (or refraction). The other two conditions are that the momenta resolved (*a*) parallel and (*b*) perpendicular to I S must be conserved. Let C N be drawn perpendicular to I S and make angles θ_i and θ_s with C I and C S respectively. Then we have

$$\frac{h}{\lambda_e}-\frac{h\,n_i\sin\theta_i}{\lambda}=\frac{h\,n_s\sin\theta_s}{\lambda} \tag{2}$$

$$\frac{h\,n_i\cos\theta_i}{\lambda}=\frac{h\,n_s\cos\theta_s}{\lambda}. \tag{3}$$

Therefore,

$$n_i\sin\theta_i+n_s\sin\theta_s=\lambda/\lambda_e \tag{4}$$

$$n_i\cos\theta_i-n_s\cos\theta_s=0 \tag{5}$$

Squaring and adding (4) and (5), we have

$$n_i^2+n_s^2-2n_i n_s\cos(\theta_i+\theta_s)=\lambda^2/\lambda_e^2 \tag{6}$$

$$\text{and from Fig. 1, } \theta_i+\theta_s=\theta \tag{7}$$

Therefore,

$$\lambda/\lambda_e=\sqrt{n_i^2+n_s^2-2n_i\,n_s\cos\theta} \tag{8}$$

The conservation of energy in the collision process gives

$$h\nu_s=h\nu_i\pm h\,\nu_e, \tag{9}$$

where ν_s, ν_i and ν_e are the frequencies of the scattered and incident photons and of the phonon respectively. The frequency shifts $\Delta\nu$ are, therefore, given by

$$\Delta\nu=\nu_s-\nu_i=\pm\,\nu_e \tag{10}$$

But $\quad \nu_e = v_e/\lambda_e$ and $\nu = c/\lambda$ (11)

where v_e is the velocity of the elastic wave, c is the velocity of light in vacuum, and $\nu = \nu_i \approx \nu_s$ since ν_e in equation (10) is very small compared to ν_i

From (8), (9) and (10) we have

$$\Delta \nu/\nu = \pm (v_e/c) \sqrt{n_i^2 + n_s^2 - 2\, n_i\, n_s \cos\theta}. \qquad (12)$$

(5), (7), (8) and (12) are the fundamental equations required to analyse the characteristics of the scattered radiation. However, thermal scattering is of macroscopic origin and consequently, it would be more appropriate to derive the results on the basis of the classical wave theory. We proceed to do so following a reasoning similar to that employed by Brillouin (1922) for singly refracting media.

3. Classical Wave Theoretical Derivation

Let the monochromatic radiation be incident along any specified direction I′ C I (Fig. 2) and consider how an elastic wave in the crystal disturbs the incident light wave. The elastic wave causes periodic stratifications in the crystal in which the density and anisotropy and consequently, the refractive index are also varying periodically with the frequency of the elastic wave.

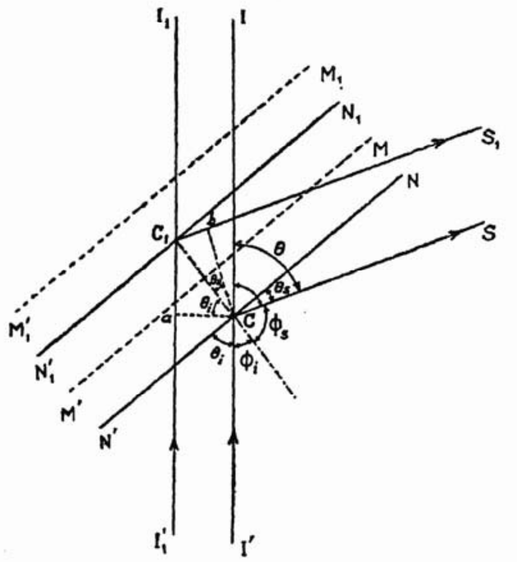


FIG. 2. Wave picture of scattering

To find the effect of these changes in refractive index on the incident light wave, let us consider a plane containing I C and C S where C S is the direction of the scattered wave and let N′ N be the intersection of a stratification with the plane of scattering T. Then the volume elements lying on this stratification will contribute to scattering in the direction C S only if there is coherence of phase of scattering by all of them. For this to happen, the conditions are (*a*) that the stratification should be normal to the plane of scattering and (*b*) that it should make angles ϕ_i and θ_s with C I and C S such that

$$n_i \cos \theta_i = n_s \cos \theta_s \tag{13}$$

$$\theta_i + \theta_s = \theta \tag{14}$$

This condition is similar to the well-known law of reflection in an anisotropic medium or of refraction which in the more familiar form is

$$n_i \sin \phi_s = n_s \sin \phi_s, \tag{15}$$

where ϕ_i and ϕ_s are the angles made by the incident and scattered wave normals with the normal to the elastic wave.

We have, however, to take into account also the light scattered by stratifications such as $N_1' N_1$, $N_2' N_2$...... Each plane will contribute very little and the nett effect will be appreciable only when the individual contributions are all in phase. Let $I_1 C_1$ be the portion of the light wave incident on the volume element C_1 lying on the next stratification $N_1' N_1$ at a distance λ_e from N′ N. Then the required condition for coherence of phase is that the difference of paths $I_1' C_1 S_1$ and I′ C S is an integral multiple of the wavelength λ of the incident light wave. Draw the normals C *a* and C *b* on $I_1 C_1$ and $C_1 S_1$ respectively.

$$a C_1 b = a C_1 + C_1 b = (n_i \sin \theta_i + n_s \sin \theta_s) \lambda_e = p \lambda, \tag{16}$$

where p is an integer.

But, there is a continuous sequence of stratifications like M′ M between $N_1' N_1$ and N′ N with the change in refractive index varying sinusoidally. The elastic wave, therefore, behaves like a sinusoidal three-dimensional grating and there can only be one maximum produced, namely, the first. That is $p = 1$ and

$$n_i \sin \theta_i + n_s \sin \theta_s = \lambda/\lambda_e. \tag{17}$$

The equations (13), (14) and (17) are the same as (5), (7) and (4) respectively and thus it is seen that the conditions for coherence of phase in the wave picture are identical with those necessary for the conservation of momentum in the quantum picture. From (13), (14) and (17) we get as before equation (8). These equations give the wavelength and the direction of propagation of the elastic wave effective in light scattering. Since the frequency of local fluctuations of refractive index of the elements on various stratifications is the same as that of the elastic wave, being v_e/c, the frequency of the scattered radiation would be altered to $\nu \pm \nu_e$. The frequency shifts of the scattered wave are given by

$$\Delta \nu/\nu = \pm v_e/\lambda_e = \pm (v_e/c) \sqrt{n_i^2 + n_s^2 - 2n_i n_s \cos\theta} \qquad (18)$$

from (8) and (11). Equation (18) is the same as (12).

The wave picture thus yields results identical with those obtained from the quantum picture and this is to be expected since the frequency shifts involved are very small. We proceed to analyse the results obtained.

4. Number of Doppler Components

For given directions of incidence and of scattering and consequently, the scattering angle θ, n_i and n_s can each take two values in general. This is generally true, for instance, in a birefringent medium or an optically active isotropic medium or in a medium possessing both birefringence and optical activity and also in crystal placed in a magnetic field, for in every one of these cases two polarised (in general, elliptically) waves are propagated in any direction. There would therefore, be four pairs of values for (n_i, n_s) and hence from equations (5), (7) and (8), four sets of values for θ_i, θ_s and λ_e. Thus the wavelengths as well as the direction of the elastic wave effective in scattering is fixed for any particular pair of values n_i and n_s. But along this direction, there are three types of elastic waves with the same wavelength λ_e but with different velocities v_e and frequencies ν_e. From equation (12), it is thus seen that there must in general be $2 \times 2 \times 3 = 12$ values for the frequency shifts $|\Delta \nu|$. Therefore, the light scattered by a birefringent crystal like calcite, must consist of 12 pairs of Doppler components. The possibility of such a large number of components has not been previously envisaged. The general characteristics of the components are given below and then it is shown how the number of components reduce in special cases.

5. Reciprocal Relations

Equations (5), (7), (8) and (12) are symmetrical with respect to the suffixes i and s. Consequently, if the directions of incidence and of scattering are interchanged, the frequency shifts of the various components would remain unaltered. Now such an interchange can take place in two ways. Referring to Fig. 1, (*a*) the direction of incidence can be S′ C and that of scattering C I or (*b*) these directions can be in the opposite senses S C and C I′. The first definition (*a*) is to be preferred since the sense of propagation is kept the same as in the original experiment and the velocities of the light waves (i and s) and hence n_i and n_s are only interchanged. On the other hand, in certain cases, *e.g.*, when the crystal is kept in a powerful magnetic field, the velocity may be different when the direction of propagation is reversed and in such a case, the second difinition (*b*) may lead to difficulties. The two experiments for which the interchange takes place according to definition (*a*) may be called reciprocal experiments and the result mentioned above may be stated in the form " Reciprocal experiments yield the same results".

6. Polarisation Characteristics

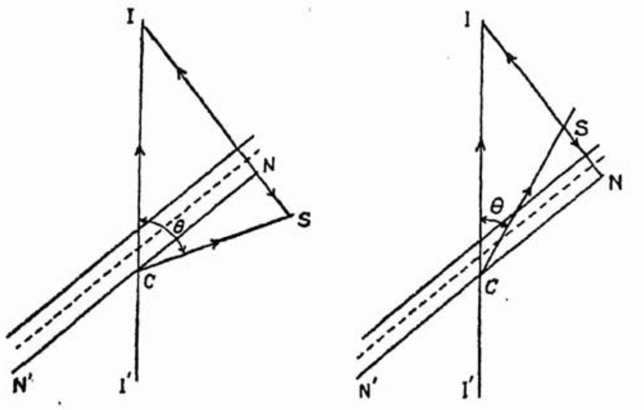
For given directions of incidence and of scattering, the two (orthogonally) polarised incident light waves may be designated A and B and the two (orthogonally) polarised scattered waves P and Q. Since either incident wave A or B can in general, give rise to either scattered wave P or Q, the scattered radiation consists of four " species " P_A, Q_A, P_B and Q_B, each with a distinctive polarisation character. For each species, the pair of values (n_i, n_s) is fixed and hence also the direction of propagation and the wave-length of the elastic wave effective in giving rise to it, and in general, both these quantatives vary for the different species. Each species consists of *three* pairs of Doppler components due to the three elastic waves. Now the different species can be separately studied by the use of proper polarising devices. For example, by the use of polarising devices in the scattered path $(P_A + P_B)$ and $(Q_A + Q_B)$ can be separated or by the use of polarising device in the incident path $(P_A + Q_A)$ and $(P_B + Q_B)$ can be separated. However, to study all the four species individually, it is necessary to use at least one polarising device in the incident and another in the scattered path. In the case of a birefringent crystal without optical activity, the proper polarising device would be a double-image prism which can separate the two plane polarised components (of the incident or scattered wave). This technique would be similar to that employed by Krishnan (1935) to study the Krishnan effect.

It is interesting to remark that the possibility of four species of Doppler components in birefringent crystals is analogous to the four reflections that can occur in a total reflection prism cut arbitrarily out of such crystals.

7. Direction of the Elastic Wave Front

For each species the direction of the normal of the elastic wave effective in scattering and hence the wave front N′ C N (Fig. 1) is uniquely determined. Now there are two distinct possibilities for the position N′ C N with respect to I′ C and CS. If $\cos\theta < n_i/n_s$ and n_s/n_i, N lies outside the internal angle of scattering $I\hat{C}S = \theta$, while if $\cos\theta > n_i/n_s$ or n_s/n_i, N′ C N lies outside the angle θ. These two cases are represented in Figs. 3*a* and 3 *b* respectively. In the first case the scattering may be regarded as " coherent reflection " of light waves from the appropriate elastic waves.

In the second case, however, the scattering of light must be regarded as "*coherent refraction*" of light waves by the effective elastic waves. Examples



(a) Coherent reflection (b) Coherent refraction

FIG. 3

of this case will be given later on. The limiting case occurs when $\cos\theta = n_i/n_s$ or n_s/n_i and in this case the elastic wave front is parallel to the incident or scattered direction and the condition is analogous to the critical angle of reflection between two different media.

When $\theta \geq 90$, $\cos\theta \geq 0$; and irrespective of the values of n_i and n_s, $\cos\theta < n_i/n_s$ and n_s/n_i. Therefore the term "coherent reflection" is always applicable. When $\theta < 90$, the condition for coherent reflection may be true for only certain species but not for others. Consequently, in the scattered radiation some species may arise from "coherent refraction" and other species from "coherent reflection" from the respective elastic waves.

It can be shown easily that all species arise from coherent refractions when $\theta < \theta_m$, where θ_m is, in general, finite and is determined by the direction of incidence. This is true for instance when $\theta \rightarrow 0$.

8. METHOD OF EVALUATION OF THE VARIOUS CHARACTERISTICS

In any specific case, it is a simple matter to calculate the frequency shifts and polarisation characteristics of the different species. Let the direction of incidence C I and that of scattering C S intersect the two sheeted index surface drawn with centre C, at I_1, I_2 and S_1, S_2 respectively. Then CI_1 and CI_2 (CS_1 and CS_2) give two values of n_i (n_s). The vectors $I_1 S_1$, I_2S_2, $I_1 S_2$ and $I_2 S_1$ represent the directions of the normals of the four possible elastic waves giving rise to the four species and their lengths represent the magnitude of the square root in equation (12), for from triangle I C S (Fig. 1)

$$IS = \sqrt{n_i^2 + n_s^2 - 2n_i\, n_s \cos\theta}. \tag{19}$$

Therefore from (12)

$$\Delta \nu = v_e (\nu/c) \text{ (I S)}$$
$$= (v_e/\lambda) \text{ (I S)}. \tag{20}$$

Thus the values of $I_1 S_1$.... when multiplied by the 3 appropriate values of v_e/λ give the frequency shifts of all the 12 pairs of Doppler components. The vibration directions of the two incident waves A and B (and of the scattered waves P and Q) are given in the case of optically inactive crystals by the principal axes of the elliptic central sections of the indicatrix parallel to the incident (and scattered) wave front. Consequently, the polarisation characteristics of all species are also specified. Further, the four perpendiculars from C on $I_1 S_1$, etc., will be parallel to the effective elastic wave fronts and depending on whether they fall inside or outside the angle θ, will show that the scattering is a coherent reflection or refraction.

9. Special Cases

In order to illustrate the procedure, we consider first the simple case of scattering in the symmetry plane (optical) of a uniaxial crystal. Let the two circles of Fig. 4 represent the section of the index surface by this plane. Fig. 4 is drawn to scale for calcite for wavelength λ 2537

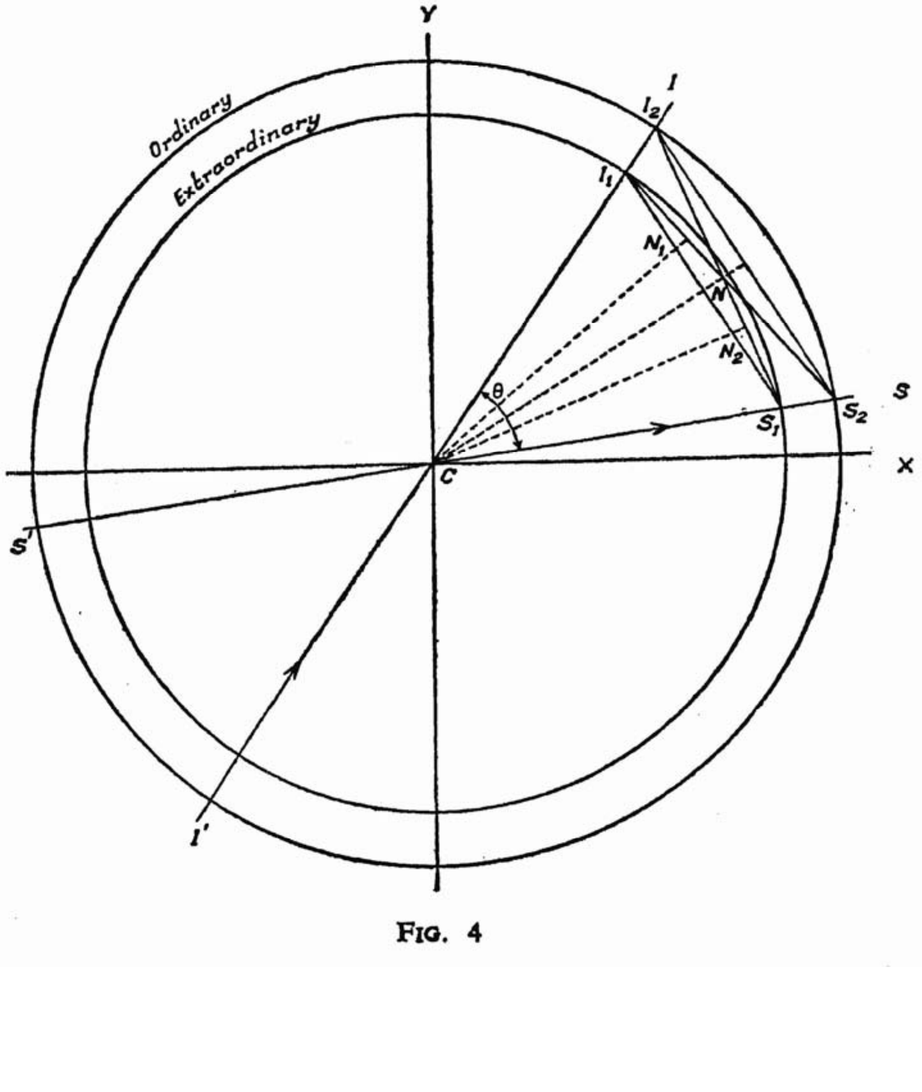
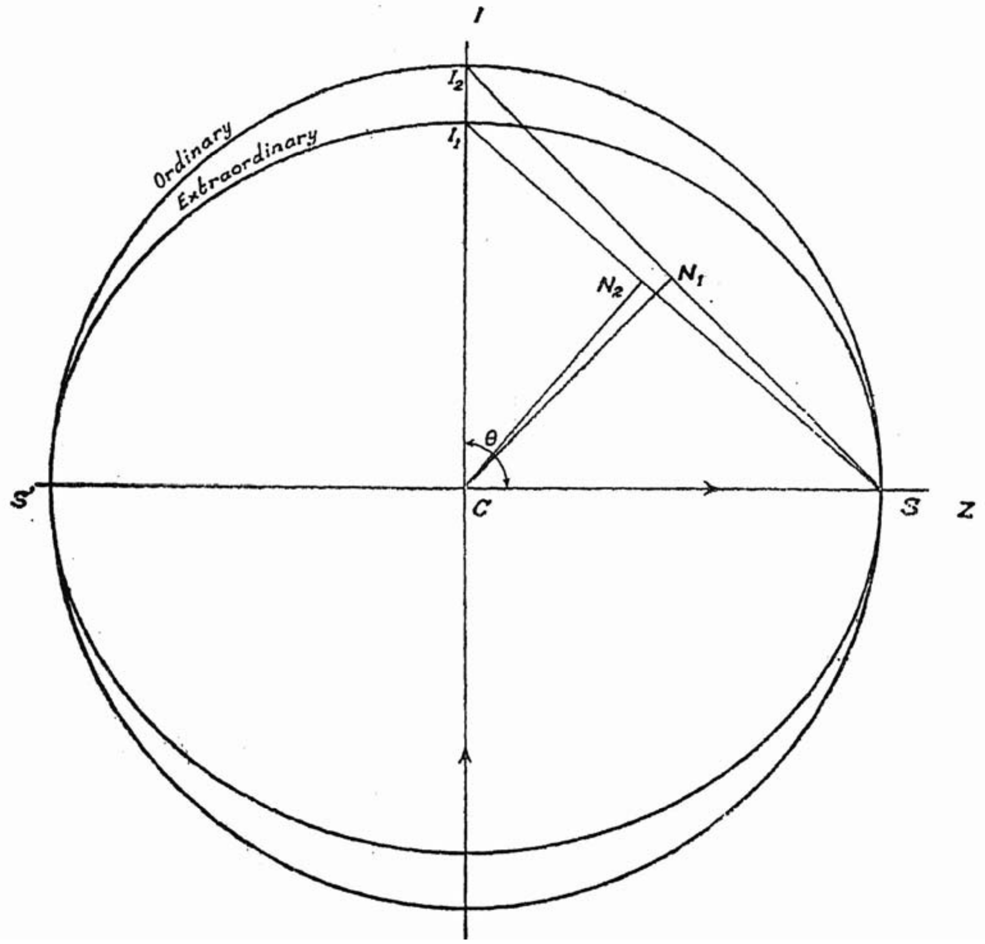


Fig. 4

($n_\omega = 1 \cdot 765$, $n_\epsilon = 1 \cdot 532$). Then both for the directions of incidence and of scattering, the refractive indices are n_ω and n_ϵ. The corresponding vibration directions of the light waves are perpendicular σ, Σ and parallel π, Π to the plane of scattering.

In Fig. 4 the directions of propagation of the different species are $I_1 S_1$ (for Σ_σ) $I_1 S_2 (\Pi_\sigma)$ $I_2 S_1 (\Sigma_\pi)$ and $I_2 S_2 (\Pi_\pi)$. For the first and the last species, the direction of elastic wave normals coincide and the wave front CN bisects always the scattering angle I C S. The scattering for these species Σ_σ and Π_π is hence a "specular reflection" from the elastic wave ($I_1 S_1$). As θ changes from 180° to 0°, C N changes from a position normal to C I′ to a position *parallel to it*. Thus the shifts are vanishingly small for small angles of θ. The situation is entirely different for the species Σ_π and Π_π when $\theta < \theta_c$, where $\cos \theta_c = n_\epsilon / n_\omega$, for example for calcite ($\lambda = 2537$) this angle is about 30°. Then these two species are coherent refractions and for very small values of θ, the wave front is *normal* to C I′. Further the shifts tend to a finite value as θ tends to zero.



10. Plane of Scattering Parallel to the Optic Axis of a Uniaxial Crystal

In this case the section of the index surface by the plane of scattering is an ellipse and a circle and Fig. 5 represents such a section for a negative crystal (drawn to scale for calcite $\lambda = 2537$). If the direction of the scattered wave (or the incident wave) coincides with the optic axis CZ then the two refractive indices for that direction become equal. Thus, in this case, there would be only two possible values for $\sqrt{n_i^2 + n_s^2 - 2n_i n_s \cos\theta}$ and also only two possible directions and wavelengths for the effective elastic waves. Therefore, there would only be 6 pairs of distinct Doppler shifts in this case.

In general however, there would be 12 pairs and the directions of the elastic waves effective in the scattering of the four species would be all different.

11. Forward Scattering

In the exactly forward direction $\theta = 0$ and hence from (12)

$$\Delta\nu/\nu = \pm (v_e/c)\sqrt{n_i^2 + n_s^2 - 2n_i n_s}$$
$$= \pm (v_e/c)\ (n_i - n_s). \qquad (21)$$

Here the two values of n_i are the same as those for n_s. Let $n_{i_1} = n_{s_1} = n_1$ and $n_{i_2} = n_{s_2} = n_2$. Then for two of the species, say P_A and Q_B, $n_i - n_s = (n_{i_1} - n_{s_1})$ or $(n_{i_2} - n_{s_2})$ both of which are zero. Hence,

$$\Delta\nu = 0.$$

For P_B and Q_A, the frequency shift is the same and given by

$$\Delta\nu/\nu = \pm (v_e/c),\ (n_1 - n_2). \qquad (22)$$

Since in general $n_1 \neq n_2$, the frequency shift is finite. We thus have the strange result of a refraction *without change of direction but with change of frequency*. The scattering in this case is due to "coherent refraction" by the elastic waves of wavelength.

$$\lambda_e = \lambda/(n_1 - n_2). \qquad (23)$$

Thus, of the 12 possible pairs of Doppler components, 6 pairs belonging to species P_A and Q_B have vanishingly small frequency shifts. Of the other 6 pairs, there will only be 3 distinct pairs of Doppler components since those produced by $n_i = n_1, n_s = n_2$ and $n_i = n_2$, $n_s = n_1$ will be superposed. In the case of a uniaxial crystal, the shifts would be a maximum when light is incident perpendicular to the optic axis. Thus for calcite using $\lambda\,2537$ incident along the Y-axis, the magnitude of the shifts are as large as $\pm 0 \cdot 22$ cm.$^{-1}$, $\pm 0 \cdot 13$ cm.$^{-1}$, $\pm 0 \cdot 10$ cm.$^{-1}$ The wavelength of the elastic waves effective in scattering in this case is, from (23), 10,880 A°.

The Doppler components with finite shifts are polarised orthogonally to the incident wave, while the transmitted light is polarised in the same way as the incident wave, and therefore by the use of a pair of polarising devices such as crossed polaroids, the incident light can be considerably reduced in intensity in the scattered path, while the scattered rays are allowed to pass unchanged. Thus the observation of the phenomenon should be rendered easy.

An analogous phenomenon should exist in the field of ultrasonics. Let the ultrasonic wave be excited inside a birefringent crystal (or even a liquid made birefringent by an electric field) and let light be incident along the direction of propagation of ultrasonic waves unlike in the case of the experimental set-up for Debye—Sears or Schaefer-Bergmann pattern. Then in the forward direction, a diffraction pattern should be capable of observation if the condition (22) is satisfied, where now $\Delta\nu$ and v_e are the frequency and the velocity of the ultrasonic waves and $(n_1 - n_2)$ is the birefringence of the crystal (or liquid) for the direction of the incident light. If say $\lambda = c/\nu = 5000 \pm 10^{-8}$ cm., and $v_e = 5 \times 10^5$ cms./sec. then $\Delta\nu = 10^{10}(n_1 - n_2)$ per sec. If $(n_1 - n_2)$ is of the order of $0 \cdot 01$ to $0 \cdot 0001$ (for example n optically active crystals) then $\Delta\nu$ would be of the order of 100 to 1 Mc. sec. Ultrasonic waves of these frequencies can easily be excited in the crystal. Further the polarisation character of the diffracted pattern should be complementary to that of the incident wave giving rise to it.

12. Backward Scattering

In this case $\theta = 180°$ and we have from (12)

$$\Delta\nu/\nu = \pm (v_e/c)\ (n_i + n_s) \tag{24}$$

and the effective elastic wave normals coincide with that of the incident direction. As in the previous case (disregarding the sense of propagation, which is irrelevant if there is no magnetic field) $n_{i_1} = n_{s_1} = n_1$ and $n_{i_2} = n_{s_2} = n_2$ and for the various species

$$P_A :- \Delta\nu/\nu = \pm (v_e/c)\, 2n_1$$

$$Q_B :- \Delta\nu/\nu = \pm (v_e/c)\, 2\, n_2$$

$$P_B \text{ or } Q_A :- \Delta\nu/\nu = \pm (v_e/c)\, (n_1 + n_2) \tag{25}$$

Thus in this case, there will be for the three possible values of v_e 9 pairs of Doppler components with different shifts.

To give an idea of the differences of the Doppler shifts for the four species, the calculated values of the frequency shifts are given below for backward scattering along the normal to the cleavage face of calcite for λ 2537 excitation. The values of $|\Delta\nu|$'s in the cm.$^{-1}$ and of λ_c in A° are given below.

				$\Delta\nu$	λ_c
P_A ($\Sigma\sigma$)	..	..	..	3·31, 1·43, 1·24	719
P_B ($\Sigma\pi$)	..	..	..	3·19, 1·39, 1·19	746
Q_A ($\Pi\sigma$)	..	..	..	3·19, 1·38, 1·19	746
Q_B ($\Pi\pi$)	..	..	..	3·06, 1·31, 1·15	776

The expected shifts (the differences for the different species being somewhat exaggerated) are represented schematically in Fig. 6.

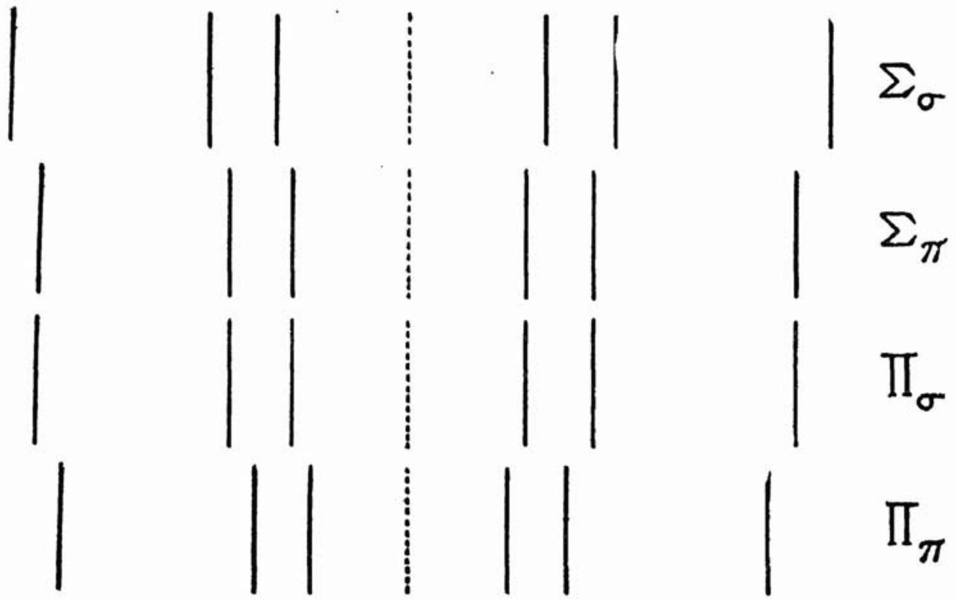


FIG. 6. Doppler components in backward scattering along the normal to the cleavage face of calcite

The actual differences are well within the limits of measurement and some experiments have been made which support these ideas. The results will be communicated in another paper.

13. Direction of Propagation of Energy

In the case of backward scattering considered above, the directions of the incident and of the scattered light waves are inclined to the optic axis. Consequently, the directions of the corresponding extraordinary rays would be different. For example, if σ and Σ refer to the ordinary incident and scattered waves and π and Π refer to extraordinary incident and scattered waves, then Fig. 7 shows how the different species of the scattered light emerge out of a negative uniaxial crystal (calcite) in the case of

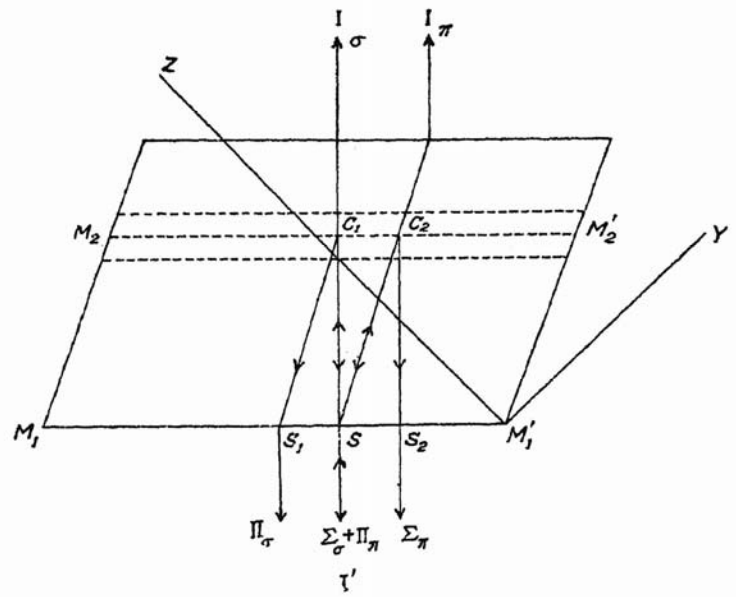


FIG. 7. Ray directions in scattering

backward scattering considered in the previous section. The incident extraordinary ray π travels in a direction inclined at 8° (shown exaggerated in the figure) to the ordinary ray. The elastic wave front $M_2 M_2'$ is normal to the incident wave normal for all the four species. But the two species arising from π and the two arising from σ are scattered from different scattering centres, C_1 and C_2 and the four species emerge out of the crystal at different points as shown. Thus the effect of the separation of the ray and wave normal directions is a lateral shift of the scattered rays. In actual experiments in which a wide pencil of rays with finite convergence is incident on the crystal, the effect of lateral shifts is unimportant.

14. Special Case

Summarising the results of the previous cases we find that in general, there are 12 possible pairs of distinct Doppler shifts in a birefringent crystal, like calcite, but in special cases of crystal orientation the number of pairs reduces to 9, 6, or 3. We consider two more very special cases, namely (1) backward scattering along the optic binormals of a uniaxial or biaxial crystal and (2) incident along one binormal and direction of scattering along the other binormal (in either of the two opposite directions). In these cases all othe four refractive indices (two for incidence and two for scattering) coincide and $n_{i_1}=n_{i_2}=n_{s_1}=n_{s_2}=n$. This last relation is always valid in the case of singly refracting cyrstals (or media) with a single refractive index, n, for all directions. The expected number of components in these cases is discussed below.

15. Single-Refracting Crystals

In these crystals, therefore, (12) reduces to the familiar expression

$$\Delta\nu/\nu = \pm (v_e/c) \sqrt{2n^2 - 2n^2 \cos\theta}$$
$$= \pm (v_e/c)\, 2n \sin\theta/2. \qquad (26)$$

Then all the four species have the same shifts and there could only be 3 pairs of Doppler components. But, in principle, each individual component can be regarded as having a degeneracy of four. This terminology would be similar to that employed for Raman lines which may however have only degeneracies of two or three in the case of first order lines in crystals.

The effective elastic wave front giving rise to the Doppler components always bisects the internal angle between the directions of incidence and of scattering and the scattering process may therefore be regarded as "specular reflection" from the elastic waves. Further, as $\theta \to 0$, $\Delta\nu \to 0$ and therefore, in the forward scattering the frequency shifts are vanishingly small.

Acknowledgment

The author is grateful to Professor R. S. Krishnan and Dr. G. N. Ramachandran for the many useful discussions. He also thanks the National Institute of Sciences for the award of the Junior Research Fellowship.

Summary

For the first time, the theory of Doppler shifts in thermal scattering of light in birefringent crystals is worked out and the magnitude of the shift $\Delta\nu$ of the components is given by $\Delta\nu/\nu = \pm v_e/c)\sqrt{n_i^2 + n_s^2 - 2n_i n_s \cos\theta}$, where $\Delta\nu$ is the frequency of the incident light, c the velocity of light in vacuum, v_e, the velocity of the elastic wave effective in scattering and n_i and n_s are either of the refractive indices of the crystal for the incident and observation directions. Since n_i and n_s can each take two values, there are four pairs of values (n_i, n_s) and further v_e takes three values. Therefore, there must *in general* be *twelve* pairs of Doppler components in the light scattered along a particular direction. The twelve pairs can be divided into four species each with a specific pair of values (n_i, n_s) and consequently specific polarisation character. They can be studied individually by the use of proper polarising devices in the incident and scattered paths. Each species consists of three pairs of components arising from the elastic waves of wavelength $\lambda_e = \lambda / \sqrt{n_i^2 + n_s^2 - 2n_i n_s \cos\theta}$, where λ is the wavelength of the incident light in vacuum and propagated along a specific direction. For

any particular species, the scattering must be appropriately regarded as "coherent reflection" or "coherent refraction" of light waves by the effective elastic waves according as $\cos\theta < n_i/n_s$ and n_s/n_i or $\cos\theta > n_i/n_s$ or n_s/n_i. There can in general be 3 pairs of Doppler components with ***finite shifts*** in the exactly *forward* scattering.

In singly refracting crystals ($n_i = n_s = n$) the expression for shift reduces to the familiar expression $\Delta\nu/\nu = \pm (v_e/c)\, 2n \sin\theta/2$ and in this case there could only be three pairs of Doppler components arising from "specular reflection" of light by elastic waves.

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