SCATTERING OF LIGHT IN A ROCHELLE SALT CRYSTAL.*

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Debye's theory of specific heats of solids is based on the hypothesis of the presence of sound waves of various wave-lengths associated with the thermal energy of the medium. The existence of these elastic heat waves produces in the solid continuum stratifications in the optical density. Ignoring the molecular structure in the medium, these density fluctuations are considered responsible for the scattering of light in the medium. Many years ago, Brillouin[2] pointed out that monochromatic light scattered or selectively reflected by the longitudinal sound waves should exhibit a doubling which is of the nature of a Doppler effect produced by the approaching and receding wave trains. It is known, however, that in addition to the longitudinal wave, two sets of transverse wave trains should also be considered, and that these latter should also give rise to observable optical effects. The elegant experiments of Schæfer and Bergmann[3] on the diffraction of light by ultrasonic waves in crystals have indeed demonstrated that for any given direction in a crystal there are in general three distinct sound velocities which generate an acoustic wave-surface of three sheets. Thus the longitudinal and the two transverse waves appearing in Debye's theory of specific heats should give rise to six Doppler Brillouin components—three on each side of the Rayleigh line. Gross[4] has reported that he has observed the splitting up of the Rayleigh line scattered by crystalline quartz and that his observations "warrant the conclusion that the Rayleigh line of crystals appears to have six modified components". Raman and Venkateswaran[1] working with an exceptionally clear crystal of gypsum have been able to reproduce a photograph showing the six components with the main line of unaltered

* In his valedictory lecture to the Central College Physical Society on the 25th of February 1938, Sir C. V. Raman remarked that the study of light scattering in crystals should yield valuable information regarding the solid state. The present investigation which demonstrates the physical reality of the Debye heat waves in crystals is an outcome of the suggestions made in the lecture. The work now described had been completed and a paper on the same was under preparation when the note by Raman and Venkateswaran[1] on their results with a gypsum crystal appeared in *Nature* of the 6th August 1938. The author has had much pleasure in accepting the invitation to publish the results obtained by him as a contribution to this Jubilee Number in honour of Sir C. V. Raman, to whom the author is grateful for many helpful suggestions.

frequency very much suppressed in intensity. Ornstein and van Cittert[5] have pointed out that small deviations from the ideal lattice structure in crystals will scatter the Rayleigh line of unchanged frequency. This line will therefore persist even in a crystal which may be visibly free from inclusions. But these inclusions which it is hardly possible to avoid completely in large crystals, give rise to a lot of parasitic light in scattering experiments, thus enhancing the Rayleigh line. The only crystalline solid, large and fairly clear that was readily available in this laboratory, was a Rochelle salt crystal grown by the Brush Laboratories Company and marketed by the Central Scientific Company. The present paper which sets forth the results of the study of the structure of the Rayleigh lines scattered by the Rochelle salt crystal affords a definite experimental proof of the presence of the Debye waves in solids.

The Rochelle salt crystal (measuring nearly 15 cm. × 8 cm. × 4 cm.) was enclosed in a blackened deal wood box lined with black felt and having a horizontal slot (2 cm. × 10 cm.) parallel to its longer edge in order to allow the focussing of the mercury arc light along the crystal core. A circular opening of about 2 cm. diameter with an ebonite tube of about 10 cm. length in continuation was provided for observing the scattered light at right angles. The scattered light was focussed on the slit of a spectrograph which was used in conjunction with a quartz Lummer-Gehrcke plate (20 cm. long and 3.45 mm. thick) for the analysis of the scattered mercury arc lines. The patterns were photographed with exposures of the order of 80 hours on hypersensitive panchromatic plates. These long exposures necessitate the maintenance of a constant temperature round about the high resolving power apparatus. Rochelle salt crystal ($K\,Na\,C_4H_4O_6.4H_2O$) belongs to the rhombic-hemihedral or rhombic-bisphenoidal class and possesses three digonal axes of symmetry which coincide with the three crystallographic axes, but has no planes of symmetry. The incident radiation from the mercury arc is allowed to traverse along the X-axis of the crystal, while the scattered radiation is observed along its Z-axis. The surface of the Lummer plate was adjusted parallel to the Y-axis of the crystal for obtaining good resolving power, as the electric vector of the scattered radiation is mainly along the Y-axis. The structure patterns of the scattered mercury arc lines λ 4358 Å and λ 5461 Å were examined and the displacements of the Doppler-Brillouin components were measured.

Fig. 1 gives the pattern of the scattered Rayleigh line λ 4358 Å along with the pattern of the same line directly obtained. It is clear that the pattern of the scattered line contains two new components of equal intensity which have no relation in position or intensity with the hyperfine components of λ 4358 Å. Comparing the hyperfine components with the Doppler-Brillouin components, it is readily noticed that these new components are far wider, though with the resolving power employed no structure is discernible. Assuming that the two new components observed are the Doppler-Brillouin components lying on either side of the main Rayleigh line, their wave-number separation from the main line comes out as $\pm 0\cdot 382$ cm.$^{-1}$. The width of each of the components is of the order $0\cdot 1$ cm.$^{-1}$, extending from $\pm\ 0\cdot 33$ cm.$^{-1}$ to $\pm\ 0\cdot 41$ cm.$^{-1}$ approximately. In spite of the fact that the wide components are not seen resolved, the width of the components must be considered as significant comprising as it probably does the components due to the transverse as well as the longitudinal sound waves in the medium. Yet another important result is the fact that the intensity total of the new components is greater than that of the main line, whose intensity has no doubt been enhanced by the parasitic light scattered by the obvious inclusions in the crystal. In spite of this source of error the intensity of the main line is much suppressed as is to be expected from theoretical considerations. Applying the Brillouin relation for the wave-number shift of the monochromatic light reflected by the thermal sound waves in the medium

$$d\nu = \pm 2\, \nu \frac{v}{c} \sin \frac{\theta}{2}$$

we obtain for v the value $3\cdot 53$ km. sec.$^{-1}$ Judging from the width of the components the velocity range extends from about $3\cdot 0$ km. sec.$^{-1}$ to $3\cdot 8$ km. sec.$^{-1}$ These values represent the velocities of the hypersonic sound waves (having a frequency of the order of $10^{12} - 10^{13} \sim$) in the Rochelle salt crystal mainly in a direction perpendicular to the Y-axis and at 45° with the X- and Z-axes. Since the incident light on the crystal is a convergent beam (nearly 60°) there will necessarily be a range for the angle of scattering, θ, round a mean value of 90°. This will in itself introduce a broadening in the Doppler components even if each component is sharp and single. Quite apart from this, the velocities of sound waves change with direction in the crystal. All these factors coupled with the unavoidable long exposures no doubt tend to broaden the new components and to mask their structure. No direct determination of sound velocities in Rochelle salt crystal is available for comparison with the values obtained above. The order of magnitude of the velocities can, however, be computed from a knowledge of the density (1·767 g. per c.c.) and the elastic constants of the crystal in various directions; Mandell[6] has determined the elastic moduli of Rochelle salt along different directions in the crystal. Calculating from those data, the velocity of compressional waves in a bar of Rochelle salt cut with its length perpendicular to the Y-axis and at 45° with the X- and Z-axes comes out as 2.60 km. sec.$^{-1}$ while the velocity along the Z-axis is 4·48 km. sec.$^{-1}$, the velocities in other directions lying within this range. The mercury green line λ 5461 Å also exhibits Doppler-Brillouin components (Fig. 2), whose separations from the main line are in accord with the Brillouin formula. The components in this case appear wider due to the complications arising from the superposition of the hyperfine structure lines of Hg I λ 5461 Å. The main Rayleigh component is even here much less intense than the intensity total of the new components.

In conclusion, I wish to thank Prof. A. Venkat Rao Telang for his encouragement during the course of this investigation.

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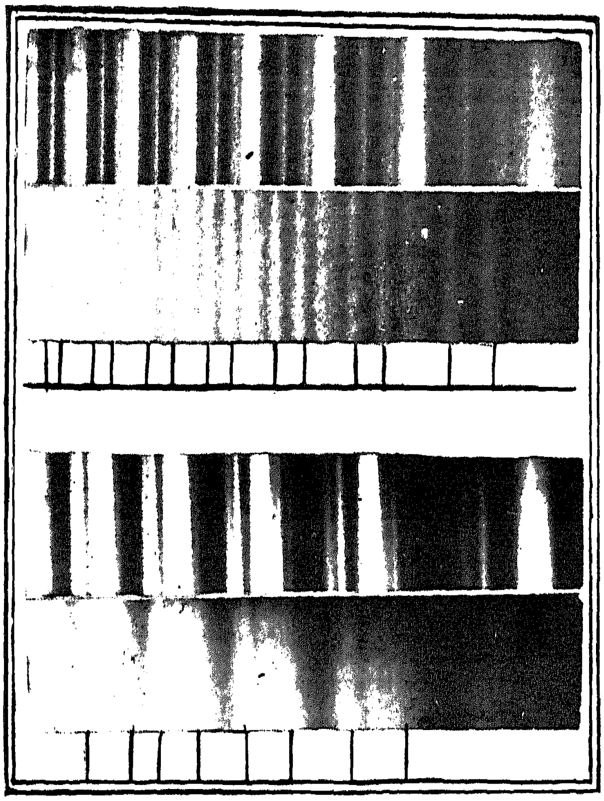

FIG. 1*a*.—The hyperfine structure pattern of HgI λ 4358 Å in the incident spectrum.
FIG. 1*b*.—The pattern of the same radiation scattered by Rochelle salt crystal, showing the Doppler-Brillouin components.

FIG. 2*a*.—The structure pattern of HgI λ 5161 Å in the incident spectrum.
FIG. 2*b*.—The pattern in the scattered radiation.