SCATTERING OF LIGHT IN SINGLE CRYSTALS

Intensity Measurements

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1. *Introduction*

Experimental observations regarding the relative intensities of the Rayleigh and Raman scattering in gases have so far been very meagre. Bhagavantam[1] showed that the intensity of the Rayleigh line in most typical gases is a few thousand times that of the vibrational Raman line. The cases of hydrogen and deuterium constitute an exception as the exciting line in them is only a few hundred times stronger than the Raman line. On the other hand, several authors[2–6] have determined the ratio of the intensity of the Rayleigh line to that of some of the prominent Raman lines in liquids like benzene, carbon tetrachloride, chloroform, etc. The results obtained by them differ to some extent amongst themselves but the general conclusion that the intensity of the Rayleigh line in liquids is a few hundred times that of the Raman lines is confirmed by all the workers. No work has at all been done so far in this direction with crystals. It will be of great interest to know the ratio of intensity of Rayleigh to that of the Raman scattering in the case of solids because such results can confidently be expected to throw light on the fundamental problem regarding the nature of the Raman scattering in solids. There are several experimental difficulties in the way of obtaining accurate results but an attempt is now made to develop a suitable technique for solving the problem.

Two typical crystals, calcite and quartz, which are available in this laboratory in the form of perfectly transparent and flawless one inch cubes with all their faces polished have been used in the present investigation. The results obtained are of great interest and are accordingly reported in this paper.

2. *Experimental Details and Results*

It is difficult to obtain the ratio of the relative intensities of Rayleigh and Raman scattering in crystals directly by photographic methods, because with the Rayleigh scattering is usually mixed up a large amount of spurious or parasitic light. As such, a special method has been adopted here to obtain an estimate of this ratio. This will be clear from Tables I and II.

The Raman spectra of the crystal (one inch cube) and a chosen liquid* contained in a glass cell of exactly the same dimensions as the crystal are obtained under identical conditions on the same plate. Great care has been taken to see that the time of exposure, the intensity of the lamp and conditions of illumination remained the same in both cases. A set of intensity marks is given on each plate by the method of varying slit widths using a quartz globe tungsten ribbon lamp as the source. The intensities of the 1085 line in calcite and of the 465 line in quartz are compared in turn with that of the 460 line in carbon tetrachloride. The results obtained are given in column 4 of each of the tables given below.

TABLE I. *Calcite*

Optic axis parallel to	I Rayleigh (liquid) / I Rayleigh (crystal)	I Rayleigh (liquid) / I Raman (liquid)	I Raman (crystal) / I Raman (liquid)	I Rayleigh (crystal) / I Raman (crystal)
OX	73		1·45	3·8
OY	83	400	2·20	2·2
OZ	65		0·87	7·1

TABLE II. *Quartz*

Optic axis parallel to	I Rayleigh (liquid) / I Rayleigh (crystal)	I Rayleigh (liquid) / I Raman (liquid)	I Raman (crystal) / I Raman (liquid)	I Rayleigh (crystal) / I Raman (crystal)
OX	77		0·96	5·4
OY	93	400	1·22	3·5
OZ	103		0·55	7·1

OX, OY and OZ represent respectively the direction of incidence, of scattering and the vertical. In column 2 of each of the tables are given figures obtained by Bhagavantam and Narayana when they compared the intensities of Rayleigh scattering in calcite and quartz with that in air by employing intermediate liquid standards. These figures may be seen to agree with those given in their paper already referred to if the conversion factor I Rayleigh (liquid CCl_4)/I Rayleigh (air) = 918 adopted by them is taken into account. In column 3 of each of the tables is given the ratio of the Rayleigh line to that of the 460 Raman line in CCl_4 as determined by Veerabhadra Rao in this laboratory. It is easily seen that the figure given in the last column is obtained by dividing 400 with the product of coulmns 2 and 4 in each case and this represents the desired ratio.

* In this paper, results obtained when the chosen liquid is carbon tetrachloride only are given. Benzene as a standard has also been experimented upon but the results are more difficult to interpret because of the large rotational wing that accompanies the Rayleigh scattering. This complication is practically absent in carbon tetrachloride.

3. *Discussion of Results*

The ratio of the intensities of Rayleigh and Raman scattering in a gas, a liquid and a crystal for one orientation are given in Table III for comparison.

TABLE III

Substance	Frequency of the Raman line	I Rayleigh / I Raman
Oxygen	1560	3330
Carbon tetrachloride	460	400
Calcite	1085	3·8

Comparatively low ratios obtained and reported in this paper for calcite and quartz are very significant. Since the intensity of Raman scattering in crystals is found to be of the same order as that observed in liquids, when equal volumes are illuminated by beams of the same intensity, we have to conclude that the Rayleigh scattering, mass per mass, has become very faint in the case of crystals whereas Raman scattering has retained its strength. This suggests that while Rayleigh scattering continues to be coherent, Raman scattering continues to be incoherent. The exact significance of such statements in relation to the mechanism of scattering in crystals is not clear and a discussion of the same will not be attempted now. The state of polarisation of the aggregate scattered light in crystals obtained by visual determinations cannot be taken as representing the character of Rayleigh scattering even approximately. Such results will be greatly influenced by the presence of depolarised Raman scattering because in the case of crystals the latter, if present, has an intensity comparable to that of the Rayleigh scattering. Only a spectroscopic investigation can lead to reliable values.

4. *Summary*

Employing carbon tetrachloride as an intermediate standard, the relative intensities of Rayleigh and Raman lines in specially cut crystals of calcite and quartz have been determined for different orientations. The Rayleigh line, in different instances, is found to possess an intensity which is only about two to seven times that of the principal Raman line.

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