ON THE MULTIPLE SPOTS AND STREAMERS EXHIBITED BY THE (111) DYNAMIC REFLECTIONS IN DIAMOND

BY P. RAMA PISHAROTY AND R. V. SUBRAHMANIAN

(From the Department of Physics, Indian Institute of Science, Bangalore)

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1. *Introduction*

THE experiments of Raman and Nilakantan[1] on the quantum reflection of X-rays from the (111) planes of diamond have definitely established that the phase-waves of the lattice oscillations, excited by the incident X-rays are parallel to the (100) planes transverse to the plane of incidence. The present paper deals with an experimental study of the effects of the other two possible sets of phase-waves parallel to the cube faces, on the Raman reflections. One of us has shown in a previous paper[2] that these sets of phase-waves also would give rise to weaker Raman reflections, in directions considerably out of the plane of incidence of the X-rays on the static spacings. In fact, the streamers and the subsidiary spots accompanying the main Raman reflection (Raman and Nilakantan, 1940) are quantum reflections arising from the dynamic stratifications produced by the combination of these waves with the static (111) spacings.

2. *Experimental*

A Siefert tube of the demountable type with a copper anti-cathode, run at 56 k.v. peak and 22 ma. was the source of X-rays. The horizontal X-ray beam was passed through a lead slit of the pin-hole type of effective depth 95 mm. and diameter 1 mm. The emergent pencil had a divergence of 72′. This narrow pencil was then passed through a thin triangular plate of diamond, cut nearly parallel to a set of (111) planes and held nearly nomal to the incident beam. The subsidiary phenomena accompanying the main Raman reflection were studied for angles of incidence close to the Bragg angle corresponding to the (111) planes of diamond and the Cu K_α radiation.

The crystal was set initially so that the incident plane was a plane of symmetry parallel to a (110) plane. In this position the ‘ streamers ’ and the subsidiary spots are symmetrical about the horizontal line joining the direct spot and the Laue spot (Figs. 2*a*, *b*, *c*, Plate XXX). Other settings of the crystal were tried for which the plane of incidence was inclined to the symmetry plane. The two ‘streamers’ and the two subsidiary spots accompanying the main dynamic reflection are found to be dissimilar in intensity and unsymmetric with respect to the line joining the direct spot and the Laue spot. Even the primary Raman reflection lies outside this line in these cases; but this is clearly detectable only when the reflection is sufficiently away from the Laue spot and then the subsidiary phenomena become extremely weak.

The question whether the subsidiary phenomena separate from the primary quantum reflection as the glancing angle of incidence θ, is gradually decreased from the Bragg value θ_B was studied. It was found that the region of maximum intensity along the ' streamer ' gradually shifted away from the strong Raman reflection, thereby showing that the streamer was a specular reflection very much elongated by the divergence of the beam and the obliquity of the corresponding phase vector to the surface of the sphere of reflection. However, whether there is a bridge connecting this region of maximum intensity to the main Raman reflection or not can be settled only by the use of very fine beams having the divergence of only a few minutes of arc.

3. *Explanation of the Phenomena*

The direction of the Raman reflection is found with the help of the ' sphere of reflection '. The direction of incidence is represented by OI = $1/\lambda$ and the vector IC represents the reciprocal lattice vector $1/d$. When C is

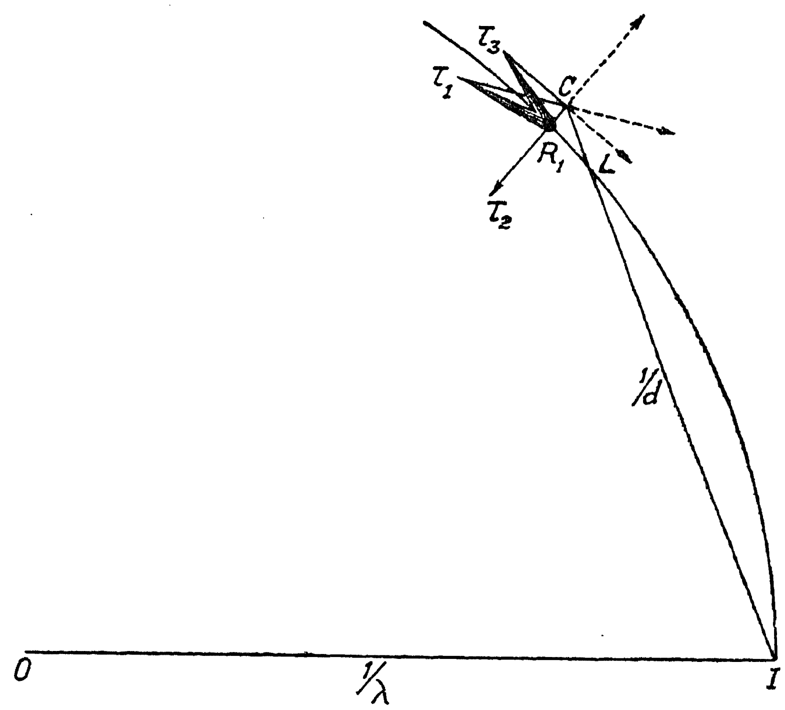

FIG. 1. Geometry of the Raman Reflections

on the sphere of reflection, the Bragg reflection is observed. No classical reflection is possible when C is either inside or outside the surface of the sphere of reflection. $C\tau_1$, $C\tau_2$ and $C\tau_3$ represent the directions of the reciprocal vectors of the phase wave-lengths of the lattice oscillations excited by the incident X-rays, and respectively parallel to the three cube edges of the unit cell of diamond. These phase-waves combine with the static lattice spacings and give rise to dynamic stratifications which produce the Raman reflections whenever the reciprocal vectors of these resultant spacings fall on the surface of the sphere of reflection.

If the plane of incidence is a symmetry plane, $C\tau_2$ is in the plane of incidence and the Raman reflection lies in the same plane as the classical reflection. The quantum reflections caused by the two vectors $C\tau_1$ and $C\tau_3$ are symmetric with respect to this plane. When C is outside the sphere, *i.e.*, $\theta < \theta_B$, $C\tau_1$ and $C\tau_3$ make only *small* angles with the surface of the sphere of reflection and they give rise to very elongated spots, which appear as 'streamers' due to the finite *divergence* of the beam (Fig. 2*a*, Plate **XXX**). On the other hand when the end C of the reciprocal lattice vector is within the sphere of reflection, *i.e.*, $\theta > \theta_B$, the vectors $C\tau_1$ and $C\tau_3$ meet the sphere at larger angles and give rise to two quantum reflections, which are drawn out into two elliptic spots (Fig. 2 *c*, Plate **XXX**) by the divergence of the beam.

In a previous paper[2] by one of us a quantitative formula was derived for the distance of the subsidiary spot from the main Raman reflection in terms of the distance between the Laue spot and the Raman reflection, for the symmetrical setting. It is approximately

$$0\cdot68 \operatorname{cosec} (35^\circ 16' - \theta).$$

Actual measurements for two different settings of the crystal giving the subsidiary spots are given below:

Plate distance 4·35 cms.

Glancing angle of incidence	Distance between Laue spot and Raman spot	Distance between the primary Raman spot and a subsidiary spot	
		Experimental	Theoretical
22° 24′	1·2 mm.	3·5 mm.	3·7 mm.
23° 28′	2·8 mm.	7 mm.	9·3 mm.

However, when the plane of incidence is not a symmetry plane, $C\tau_2$ is not in the plane of incidence and $C\tau_1$ and $C\tau_3$ are no longer symmetric with respect to the plane of the classical or Laue reflection. Hence in both the cases $\theta > \theta_B$ and $\theta < \theta_B$, the subsidiary spots and the 'streamers' are unsymmetrical, and the difference in the lengths of the reciprocal vectors $C\tau_1$ and $C\tau_3$ give different intensities for these reflections, as actually observed. Figs. 2*d*, *e* and *f* in Plate **XXX** show the 'steamers' and the subsidiary spots when the plane of incidence makes 10° with the symmetry plane.

The inclination of the vectors $C\tau_1$ and $C\tau_3$ to the surface of the sphere being small (only about 13°), a rotation of the whole configuration through 10° with IC as axis would make one of these vectors leave the surface of the sphere. Hence, only one of the ' streamers ' would be left behind and this would extend both ways if the setting is the correct Bragg setting.

The existence of these triple quantum reflections suggests that the three sets of phase-waves are not capable of combination among themselves, thus showing that the lattice oscillations are incoherent when they differ in phase-wave-lengths. Otherwise, we would have phase-waves of all possible orientations thus destroying the specular character of the quantum reflections. However, the quantum reflections are distinctly separate only when the crystal setting is off the Bragg setting by the divergence of the incident beam or more. The phase-wave-length corresponding to the quantum reflection in this position is nearly 150 A.U. or less. For nearer settings, the operative phase-wave-lengths are much greater, but due to the divergence of the beam we are not in a position to settle whether the corresponding lattice oscillations are coherent or not.

The question of coherence or incoherence of the lattice oscillations of different phase-wave-lengths in the direction of the three cube-edges of diamond, was tested out experimentally in light scattering by Nayar.[3] A crystal of diamond was illumined by a narrow parallel pencil of light incident at the correct angle so that the phase-waves parallel to a cube face may reflect the light with altered frequency. The scattered light was observed spectrographically in the proper direction of reflection and in directions upto 20° on either side of it. He found that the Raman line exhibited the same intensity in all these directions. The facts about light scattering indicate therefore, that lattice oscillations of phase wave-lengths of 2000 A.U. or more are coherent and that they combine to give phase waves of all possible orientations. In the X-ray field this means a change in the glancing angle of only a few minutes of arc from the correct Bragg setting. Hence the problem of coherence in diamond of the lattice oscillations for different phase-wave-lengths can be settled only by observing the subsidiary phenomena with X-ray beams having a divergence of 5 minutes of arc or less.

4. *Remarks on the Influence of Mosaic Structure in Diamond*

The X-ray experiments and the spectroscopic investigations in this laboratory have all been conducted with the ordinary type of diamond. The theoretical investigations have also been carried out only for the perfect crystals. The studies of Julius, Angstrom and Reinkober[4] have shown the existence of a second variety of diamond which, unlike the ordinary variety, is transparent to the infra-red radiations in the neighbourhood of $8\,\mu$.

Robertson, Fox and Martin[5] have made extensive investigations on the various properties of this type of diamond and have come to the conclusion that it possesses a definite mosaic structure. A specimen of this type has been recently procured in this laboratory, but no X-ray studies have been conducted with it as yet. While this paper was going to press, a note in *Nature* by Lonsdale and Smith[6] has reached India where it is stated that this rare type of diamond does not exhibit the subsidiary phenomena forming the subject of the present paper, and also indicating that the ordinary or Bragg X-ray reflections given by it are stronger. It is shown below that the mosaic structure readily explains these facts.

Robertson, Fox and Martin have remarked that the second type of diamond gives a higher value for the ratio of the intensity of the (111) reflection to that of the (222) reflection than the ordinary type and have put this forward as an evidence of its mosaic structure. In a paper[7] appearing in this issue of the *Proceedings*, one of us has shown that the (222) reflections can be explained quantitatively as Raman reflections produced by the lattice oscillations of *infinite* phase-wave-length produced in the crystal. In a rotation or oscillation method, the (111) planes of a mosaic crystal would give the reflection over a greater angular range so that its intensity would be more than that for the normal type. On the other hand, the (222) quantum reflection would be much weaker as the size of the mosaic would effectively prevent the co-operation between the neighbouring parts of the crystal required for such a weak reflection to manifest itself. Hence it is not surprising that the ratio of the intensity of the (111) to the (222) reflection is large for the second type of diamond.

In another paper[8] appearing in this issue, one of us has pointed out the great rapidity with which the intensity of the Raman reflection falls off with decrease in the phase-wave-length of the lattice oscillations. It means therefore, that even for a perfect crystal the structure amplitudes for these subsidiary reflections are extremely small, so that very large numbers of regular co-operating planes are needed to produce an appreciable intensity of the subsidiary reflections. In a mosaic crystal, the lack of co-operation between different mosaic blocks would therefore reduce the intensity of the subsidiary reflection to a small fraction of the value for the perfect crystal. Hence it is but natural that the subsidiary phenomena, weak even with a perfect crystal, cease to be observable with the rarer type of diamond whose mosaic structure is an accepted fact.

Lonsdale and Smith have also observed that the Raman reflections are absent, (*a*) along the (001) axis for the (220) and (113) dynamic planes, (*b*) along the (010) axis for the (202) and (131) planes, and (*c*) along the (100) axis for the (022) and (311) planes. This fits in beautifully with the concept of phase wave-normals along the three cube edges, in which case the observed inactive directions are exactly tangential to the surface of the sphere of reflection when the plane of incidencce is a symmetry plane. In this setting the other two phase vectors cut the sphere of reflection in the plane of incidence.

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5. *Summary*

The existence of three sets of phase waves of the lattice oscillations in diamond is shown to give rise to three quantum or Raman reflections from the (111) planes. Two of these reflections form very elongated spots which appear as streamers, due to the divergence of the beam and the obliquity of the corresponding reciprocal phase vectors, when the glancing angle of incidence θ is smaller than the Bragg angle θ_B. These reflections appear as discrete subsidiary spots when $\theta > \theta_B$. The behaviour of these reflections with respect to intensity and orientation, when the plane of incidence of the classical reflection is (1) parallel to, and (2) inclined to, a (110) symmetry plane of the crystal, is described and accounted for. The question as to the coherence of the lattice oscillations of differing phase-wave-lengths has been examined, and it is shown that all oscillations of phase-wave-lengths of the order of a thousand lattice spacings or more are coherent, while those with phase-wave-lengths of the order of a hundred lattice spacings or less are definitely incoherent. But the exact stages at which the incoherence sets in is still unsettled. The differences in the behaviour of the various planes in the normal type of diamond towards the subsidiary spots and 'streamers' and the absence of the subsidiary phenomena in the rare type, both observed by Lonsdale and Smith, have been accounted for. The larger value of the ratio of the intensity of the (111) to the (222) reflection as recorded by Robertson, Fox and Martin for the type II diamond is explained on the basis of its mosaic structure.

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