A quantitative study of diffracted X-ray intensities from type I natural diamond crystals by high resolution X-ray diffractometry and comparison with nearly perfect silicon single crystals

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Abstract. Results of accurate measurements of peak and integrated intensities of $\bar{2}20$, $\bar{4}40$, $\bar{2}24$, $\bar{1}13$, 111 and 333 reflections of natural diamonds of type I and nearly perfect silicon single crystals are reported. Highly monochromated and collimated MoK_{α_1} exploring beam was used. A quadrupole crystal X-ray diffractometer was employed in $(+, -, +)$ and $(+, -, +, -)$ settings. (111) platelets of diamond and silicon crystals with thicknesses of about 1 mm were selected. High resolution diffraction curves, stationary and traverse topographs were recorded. Diffraction curve half widths of diamond and silicon crystals were in the range: 45–200 arc sec and a few arc sec respectively. The experimental values of integrated intensities ρ for diamond crystals were found to lie between the theoretical values for ideally perfect and ideally imperfect crystals. Experimental values of ρ for silicon were closer to the "perfect crystal" values. This is consistent with the results of diffractometric and topographic evaluation. The peak intensities of all reflections were higher for diamond crystals in comparison to the silicon crystals. The ratio I_C/I_{Si} lies in the range 1·3 (111 reflection) to 10·5 ($\bar{1}13$) and (333) reflections. This is anomalous and cannot be accounted for by considering the degree of perfection, structure factor and difference in absorption coefficient.

Keywords. Natural diamonds; silicon single crystals; high resolution X-ray diffractometry and topography; accurate diffraction peak intensities; accurate integrated X-ray intensities.

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1. Introduction

Natural diamonds were among the first few crystals to be extensively investigated by X-ray diffraction techniques (Bragg 1914; Renninger 1935, 1937; Brill *et al* 1939; Ehrenberg *et al* 1928; Brindley 1932, 1933). The early investigations were aimed at quantitative understanding of the observed intensities in terms of a "mosaic" crystal model. Some of the crystals were found to possess reasonably high degree of perfection (Bragg 1921) while others were found to be closer to the mosaic model. Crystals with intermediate degree of perfection were also detected. Some of the investigators used double crystal diffractometers in the $(+, -)$ configuration and obtained very narrow diffraction curves (Bragg 1921). In recent times real structure of diamond has been studied by Lang and his co-workers by using X-ray diffraction topographic method (Lang 1974).

Considerable advances have been made in the techniques and equipments used for high resolution X-ray diffractometric experiments (Bonse and Hart 1965; Hart 1971;

Pinsker 1978; Patel and Battermann 1963; Lal 1982, 1988). It is possible to achieve highly monochromated and well collimated X-ray beams of appreciable intensities which can be used for precise X-ray diffraction studies (Lal *et al* 1979). High resolution X-ray diffractometric and topographic techniques developed in our group at National Physical Laboratory, have been very successfully used to study imperfections in crystals (Lal *et al* 1979; Lal and Thoma 1983; Lal and Goswami 1987). Here we describe results of a high resolution X-ray diffractometric study on natural diamond crystals and report anomalously higher diffracted intensities from these crystals in comparison to nearly perfect silicon single crystals.

2. Experimental details

2.1 *Specimen*

The diamond specimens were platelet-shaped natural crystals with surface along (111) having areas of a few mm^2. The thicknesses of the crystals were about 1 mm. Infrared and EPR absorption measurements carried out in our laboratory showed that these crystals contain nitrogen and are of type I. Silicon single crystal specimens were (111) discs cut out of a Czochralski grown n-type boule of $50\,\Omega\,cm$ resistivity.

2.2 *Experimental set-up*

A quadruple crystal X-ray diffractometer developed in our group has been employed in three-crystal and four-crystal modes in these experiments. Figures 1(a) and (b) show schematic line diagrams of these triple and the quadruple crystal X-ray diffractometers, respectively. Fine focus X-ray source of following specifications was used: Philips sealed tube; 2 kW; molybdenum and source size foreshortened to $0.4 \times 0.4\,mm^2$. X-ray beam emerging from the source is collimated with the help of a long collimator which is fitted with a fine slit at its end away from the source. The collimated X-ray beam is diffracted from two dislocation free plane (111) silicon single crystal monochromators set in the $(+, -)$ configuration (Bonse–Hart type). With this combination of source, collimator and the monochromators, $K_{\alpha 1}$ and $K_{\alpha 2}$ components of the characteristic K_{α} doublet are conveniently resolved. The more intense $K_{\alpha 1}$ beam is highly collimated and monochromated and is used as the exploring beam. All the
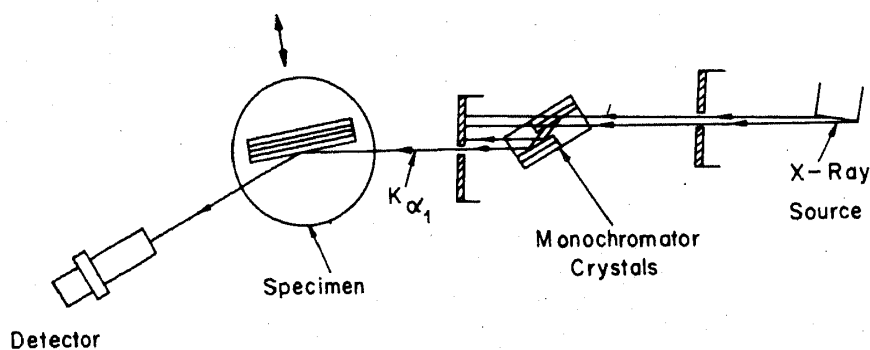

Figure 1 (a). A schematic line diagram of the quadruple crystal X-ray diffractometer when set in $(+, -, +)$ configuration.
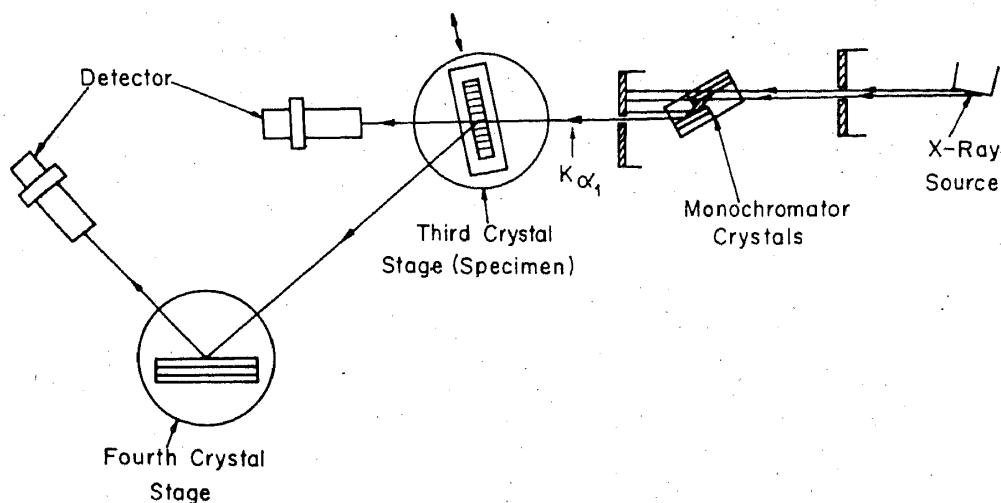
Figure 1 (b). A schematic line diagram of the quadruple crystal X-ray diffractometer when set in $(+, -, +, -)$ configuration.

experiments reported have been performed with the $MoK_{\alpha 1}$ radiation. The specimen crystal formed the third crystal of the diffractometer in the $(+, -, +)$ configuration. Some experiments with the specimen at the fourth crystal position have also been performed in the $(+, -, +, -)$ setting. The width of the exploring beam was 0·2 mm in the plane of diffraction. The height of the irradiated area of the specimen was adjusted according to different requirements. For recording traverse topographs, the height of the beam was increased to bathe the entire specimen. For quantitative diffracted X-ray intensity measurements, the height was adjusted to 1·2 mm. This is much smaller than the size of the specimen.

Specimen is mounted on a special goniometer which is fixed on top of a traversing mechanism. This assembly is mounted on a special turntable, which provides precise rotations around a vertical axis. The special goniometer provides rotations to the specimen around a horizontal axis with minimum rotation of 10 arc sec. Minimum rotations of 0·4 arc sec can be given about the vertical axis with the help of a fine angular movement attachment. Details of the quadruple crystal X-ray diffractometer are reported in Lal (1985). The specimen is aligned for diffraction from any desired set of lattice planes. Diffraction curves are recorded from different regions of the specimen. High resolution traverse topographs are recorded on photographic films by traversing the specimen and the film together across the exploring beam while maintaining the angular orientation of the crystal at the diffraction peak position. It may be mentioned that in this case the diffraction curves are significantly sharper as compared to the Lang technique. Radius of curvature of the specimen is measured from the plots of the orientation of the diffraction vector versus linear position of the specimen.

Diffraction from $(\bar{2}20)$, $(\bar{4}40)$, $(\bar{2}\bar{2}4)$ lattice planes was studied in the symmetrical Laue (transmission) geometry. 111 and 333 reflections were studied in the symmetrical Bragg (reflection) geometry. Measurements were also made with $\bar{1}\bar{1}3$ reflection in asymmetric Laue geometry.

The integrated intensities were determined by the standard method (James 1950). The area of the diffraction curve was obtained from the experimentally measured diffraction curves and the intensity of the exploring beam was accurately measured.

3. Results

3.1 *Determination of crystalline perfection of the natural diamond crystals and comparison with silicon single crystals*

First of all, natural diamond crystals were examined by high resolution X-ray diffractometry and topography to evaluate their crystalline perfection. These experiments were performed in the $(+, -, +)$ setting. Figure 2 shows a typical diffraction curve of one of the crystals. This curve is rather broad with a half width of 58 arc sec. The half width of the specimens investigated during this study were in the range of 45 to 200 arc sec. These are very large in comparison to the theoretical half widths for perfect diamond crystals for $MoK_{\alpha 1}$ radiation on the basis of the plane wave approximation of dynamical theory calculated for the reflections studied here. The theoretical values are 0·35 to 2 arc sec (Tanner 1976). A small broadening of the diffraction curve is expected due to dispersion as the interplanar spacings of the diamond crystals and those of the silicon monochromators are different. However, the observed difference is too large to be explained on this basis. Diffraction curves from some parts of these crystals show more than one peak due to the presence of low angle boundaries. Figure 3 shows one such diffraction curve. Two well resolved peaks and a shoulder on low glancing angle side are clearly seen.

A typical high resolution traverse topograph of a diamond crystal is shown in Figure 4. The diffracted intensity is not uniformly distributed in this topograph. The strain is quite large and hence individual line defects could not be resolved. These
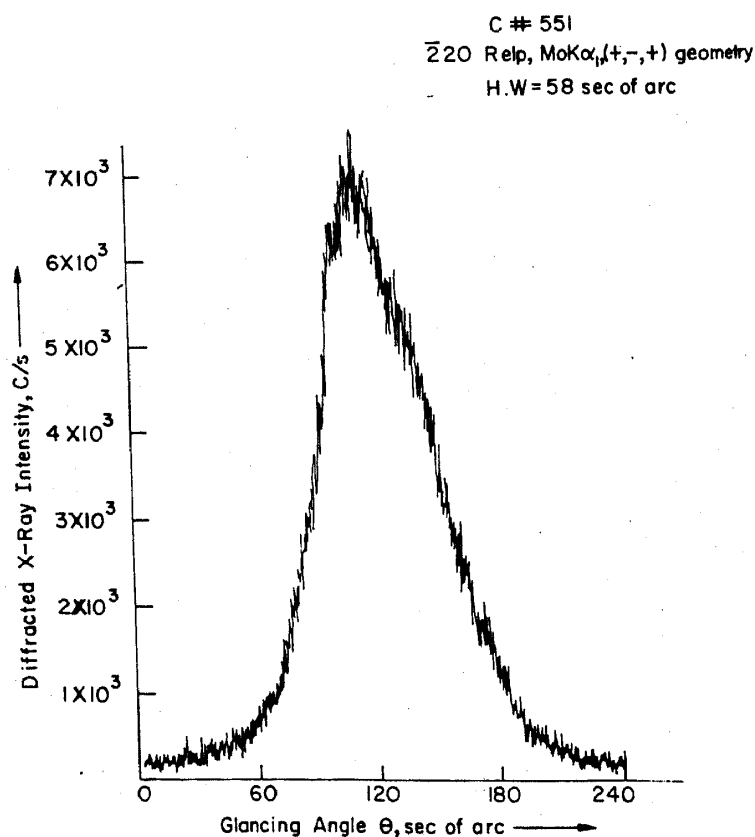
Figure 2. A typical diffraction curve of a natural diamond crystal. $\bar{2}20$ Relp; $MoK_{\alpha 1}$ beam, symmetrical Laue geometry and $(+, -, +)$ settings were used.
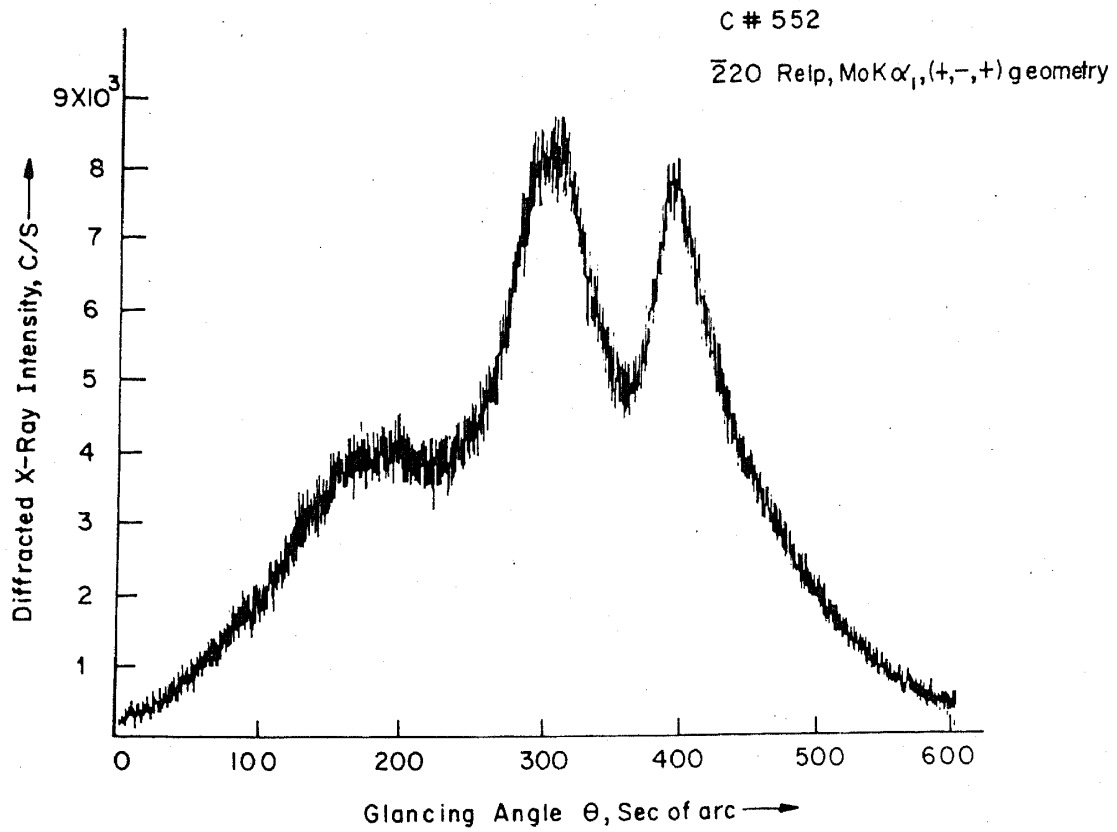
C # 552

$\overline{2}20$ Relp, MoKα'_1,(+,-,+) geometry

9X10³

8

7

6

5

4

3

2

1

0   100   200   300   400   500   600

Glancing Angle Θ, Sec of arc ⟶

Diffracted X-Ray Intensity, C/S ⟶

Figure 3. A diffraction curve of a natural diamond specimen with low angle boundaries. Experimental conditions are the same as in figure 2.

⟵$\vec{g}$ [$\overline{2}$20]

Imm

Figure 4. A high resolution traverse topograph of a natural diamond crystal. Experimental details as in figure 2.

results are understandable as these crystals have grown in nature in an uncontrolled fashion. One can observe a small inclusion in the middle of the photograph. So far, it is considered that transition metal elements like nickel promote growth of diamond crystals. The absorption of X-rays of relatively short wavelength (0·709 Å in this case) shows that this is a material with high atomic number.

Curvature plots of the diamond crystals also show the presence of many subgrains. The radii of curvature for any of these subgrains is small and lies in the range of 3 to 10 m. This result is consistent with those of the diffractometric and topographic characterizations reported above.

For comparison, diffractometric and topographic evaluation of a few nearly perfect silicon single crystals was also carried out under identical experimental conditions. Figure 5 shows a typical diffraction curve of a nearly perfect silicon single crystal recorded with ($\bar{2}20$) lattice planes, MoK$_{\alpha 1}$ radiation and ($+, -, +$) setting. The half width of this diffraction curve is about 5 arc sec. In the four crystal geometry, diffraction curves from the same set of planes have a width of only 3 arc sec (Lal and Goswami 1987). A high resolution traverse topograph of a silicon specimen is shown in Figure 6. In this topograph, no line defect is visible. Curvature plots for silicon single crystals were also recorded. The radius of curvature was determined to be about 240 m for this specimen. These results show that the silicon specimens are nearly perfect crystals.

The results reported above clearly show that the natural diamond crystals have a low degree of crystalline perfection in comparison to silicon crystals. However, the diamond crystals exhibit very high intensity of diffraction at the peak position. Figure 7 shows diffraction curves of a silicon crystal and a diamond crystal drawn on the same scale and normalized for approximately the same diffracting volume for $\bar{2}20$ reflection. The peak intensity is much higher in the case of diamond in comparison to that of the silicon. The peak intensity ratio is 5:1. This is anomalous. It is well established that with decrease in perfection of atomic arrangement the intensity of diffraction
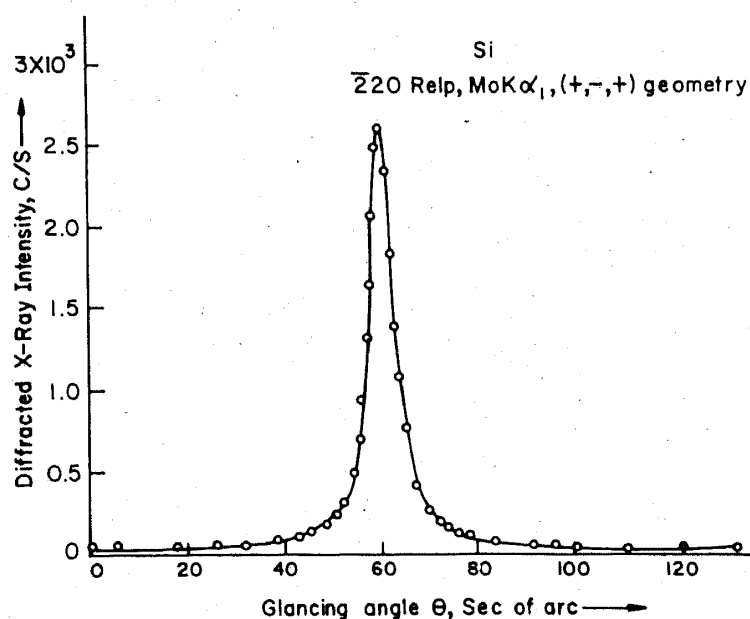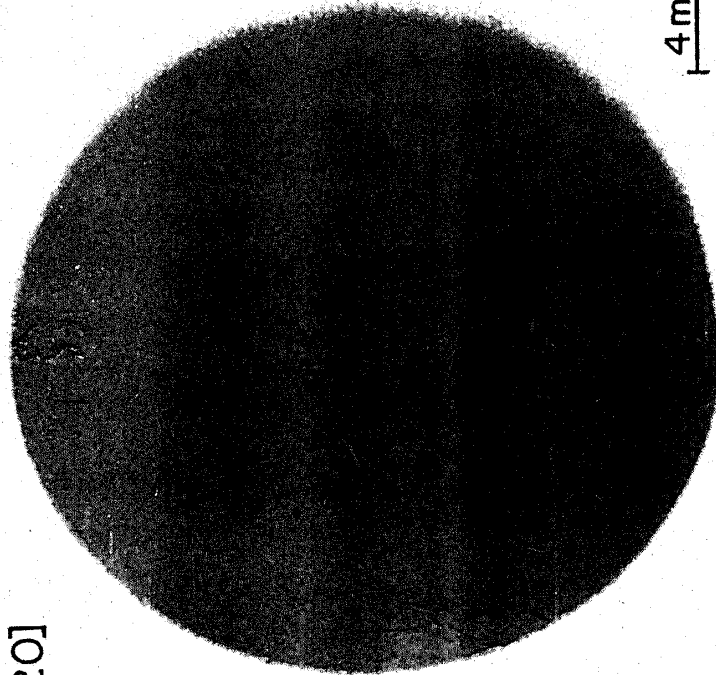

Figure 5. A typical diffraction curve of a dislocation free silicon single crystal. $\bar{2}20$ Relp, MoK$_{\alpha 1}$ and ($+, -, +$) setting were used.

Diffraction Curves, $\bar{2}20$ Relp.

$(+,-,+)$, Laue, MoKα_1

8×10^3

DIAMOND

SILICON

Diffracted Intensity, C/S

Glancing Angle Θ, Sec of Arc $\longrightarrow$

240 180 120 60 0

Figure 7. Diffraction curves of a diamond and a silicon single crystal plotted on the same scale. Diffracting volumes of both the specimens are approximately the same. $\bar{2}20$ Relp, MoK$_{\alpha 1}$ beam, symmetrical Laue geometry and $(+, -, +)$ setting were used.
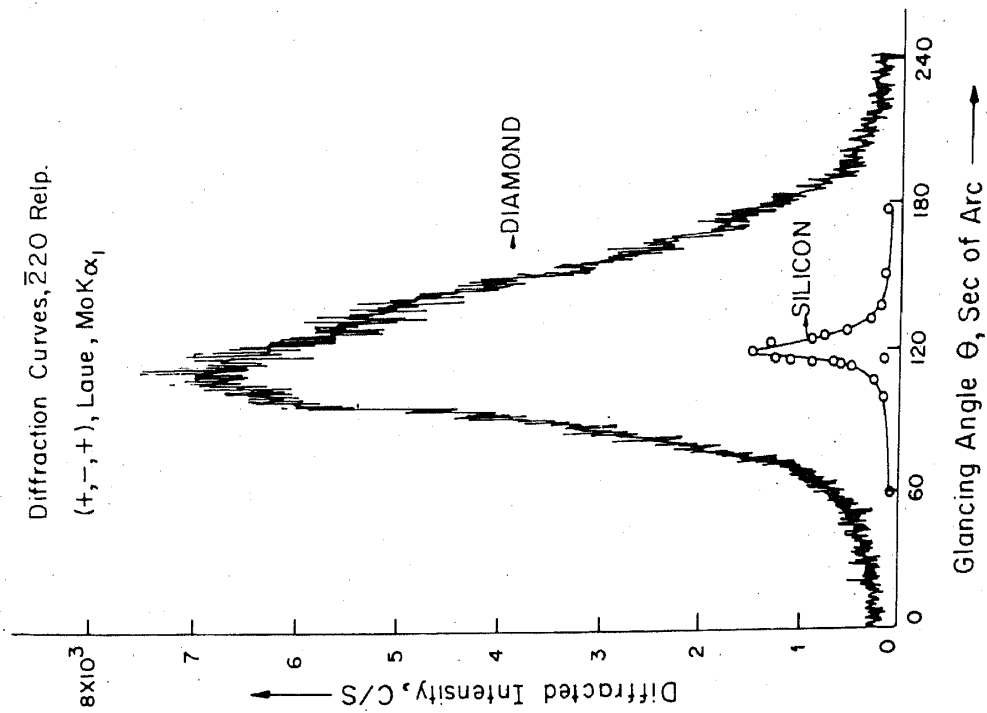
$\longrightarrow g\ [\bar{2}20]$

4mm

Figure 6. A high resolution traverse topograph of a dislocation free silicon single crystal. Experimental details are as given in figure 5.

maxima decreases (James 1950, p. 271). We shall consider this further in §3.3. Moreover, the atomic scattering factor for carbon is much smaller in comparison to that for silicon. This further enhances the discrepancy. Even when the difference in the absorption coefficients of the two crystals for $MoK_{\alpha 1}$ radiation are taken into consideration the anomaly cannot be explained. Therefore, a detailed quantitative comparative study of the diffracted X-ray intensities, both peak as well as integrated was undertaken. Results of these experiments are described in the following.

3.2 *Quantitative measurements of diffracted X-ray intensities from natural diamond and comparison with those of nearly perfect silicon single crystals*

To understand the cause of anomalous ratio of peak intensities of diamond and silicon crystals, systematic quantitative measurements of intensities of diffracted X-rays, both peak as well as integrated, for a number of reflections from natural diamond and silicon crystals have been made. The irradiated area (Bragg geometry) and volume (Laue geometry) for diamond and silicon crystals was nearly identical. It was small in comparison to the total area/volume of the specimen crystals. The results are described in the following.

3.2.1 *Results of experiments performed in the Bragg (reflection) geometry:* Experiments were performed with (111) and (333) diffracting planes. Both these reflections were studied in the symmetrical Bragg geometry with $(+, -, +)$ setting of the diffractometer. Figure 8 shows typical diffraction curves obtained with a diamond and a silicon crystal for 111 reflection. The peak intensity for diamond crystal is higher than that for silicon crystal as reported above. The peak intensity ratio is $I_C/I_{Si} = 1.3$. Here I_C and I_{Si} are the peak intensities for diamond and silicon, respectively.

Figure 9 shows results obtained with a high order reflection 333. The ratio of diffracted intensities from diamond and silicon has considerably increased when we compare this result with that reported in Figure 8. The value of I_C/I_{Si} is 10.5.
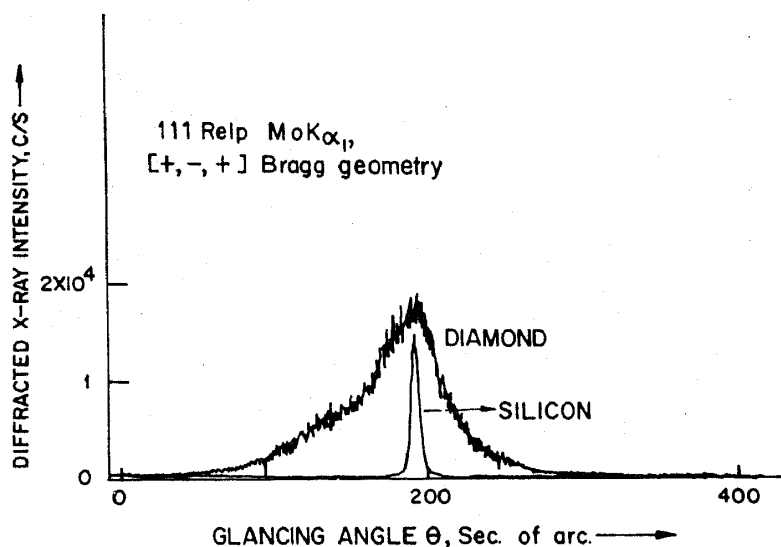

Figure 8. Diffraction curves of a diamond and a silicon single crystal. 111 Relp, $MoK_{\alpha 1}$ radiation, symmetrical Bragg geometry and $(+, -, +)$ setting were used.
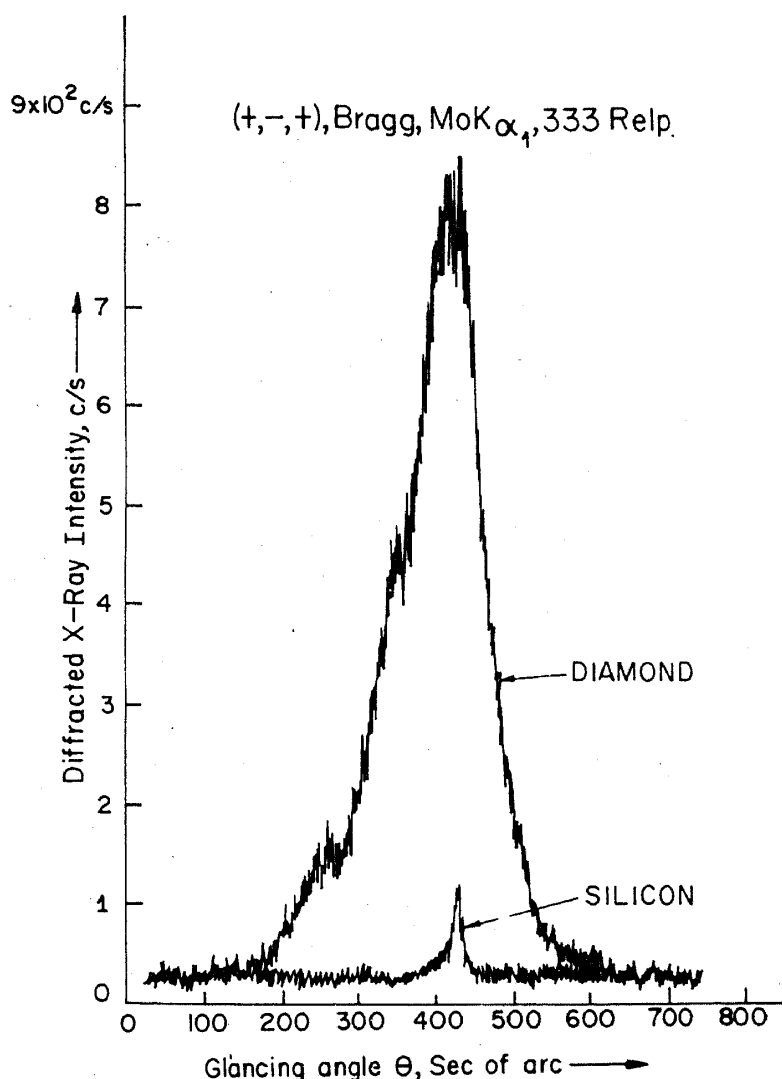
Figure 9. Diffraction curves of a diamond and a silicon single crystal. 333 Relp, $MoK_{\alpha 1}$ radiation, symmetrical Bragg geometry and $(+,-,+)$ setting were used.

3.2.2 *Results of experiments performed in the Laue (transmission) geometry:* Figure 10 shows diffraction curves of diamond and silicon crystal for $\overline{2}20$ reflection. $(+,-,+)$ setting of the diffractometer was used. The diffracted X-ray intensity is much higher for the diamond crystal in the entire angular range. The peak intensity for this diamond specimen is 4·5 times that of silicon crystal. In all the specimen crystals examined by us, peak intensity for diamond I_C was always higher than that observed for silicon I_{Si}. The ratio was in the range 4·5 to 5.

Experiments have been performed with higher order reflections. Figure 11 shows diffraction curves for diamond and silicon crystals recorded with $(\overline{4}40)$ diffracting planes. The peak intensity of $\overline{4}40$ reflection for silicon crystals with the normal size of the exploring beam, an area of $1·2 \times 0·2\,mm^2$, is rather small. It is only 90 c/s. With this small intensity, it is not possible to get good quality diffraction curves. Therefore, the vertical slit opening was increased from 1·2 mm to 5·6 mm to enhance the intensity of the exploring beam. This helped in recording a curve with good statistics, and having the peak intensity comparable to that of the diamond crystal for a beam of
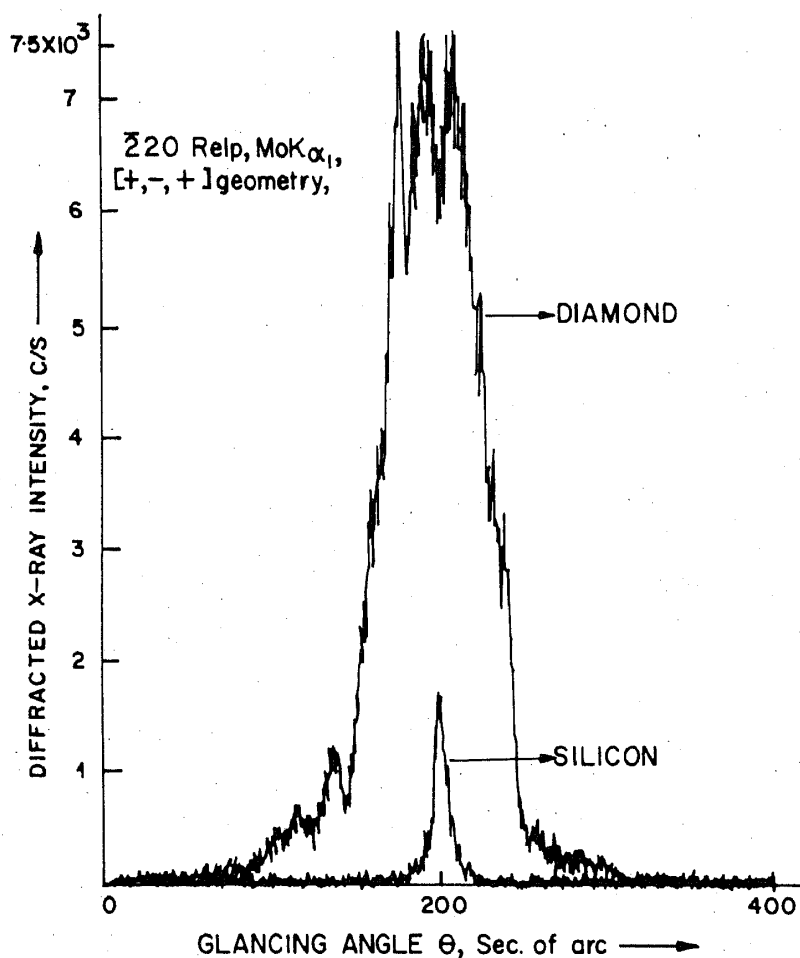
Figure 10. Diffraction curves of a diamond and a silicon single crystal recorded with ($\overline{2}$20) diffracting planes.

fixed height (1·2 mm). From the experimental curve, intensities for different glancing angles were calculated for the normal irradiated area (1·2 × 0·2 mm^2) and a normalized diffraction curve was obtained. This is the silicon curve shown in figure 11. The peak intensity for the diamond crystal is seven times higher than that of the silicon crystal.

A weak reflection like $\overline{2}$24 was also studied in the symmetrical Laue geometry. Figure 12 shows typical diffraction curves for diamond and silicon crystals for $\overline{2}$24 reflection. It is seen that even in these diffraction curves, the peak intensity for diamond is very high (7·5 times) as compared to that for silicon crystal.

In addition to the experiments described above, we have made measurements with $\overline{11}$3 reflection in the asymmetric transmission mode (Laue geometry). ($\overline{11}$3) lattice planes are inclined at an angle of 79° 58' to the surfaces of these crystals which are along (111) planes. Figure 13 shows typical diffraction curves obtained with diamond and silicon crystals for this reflection. The diffracted X-ray intensity from diamond crystal in this case also is much higher as compared to that from silicon crystal. The peak intensity ratio I_C/I_{Si} is 10·5:1.

3.2.3 Results of experiments performed with the specimen crystals at the fourth crystal position of the quadruple crystal X-ray diffractometer: To further improve upon the resolution of the experimental arrangement, the specimens were placed at the fourth

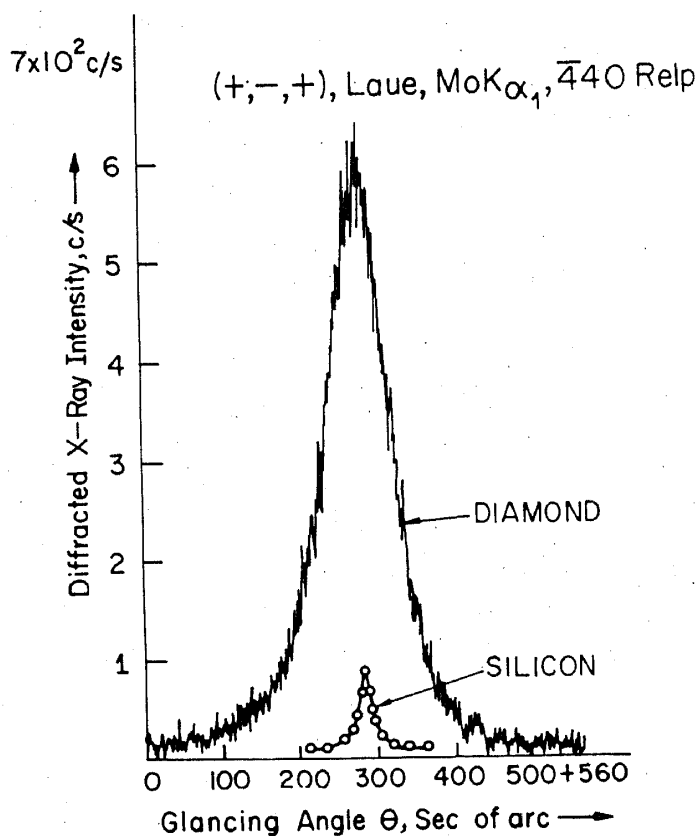**Figure 11.** Diffraction curves of a diamond and a silicon single crystal recorded with ($\bar{4}40$) diffracting planes.
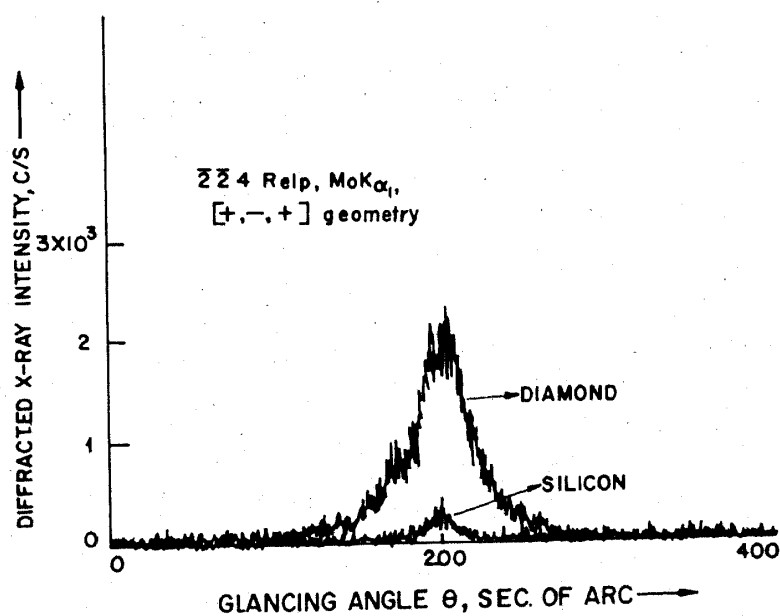

Figure 12. Diffracting curves of a diamond and a silicon single crystal recorded with ($\bar{2}\bar{2}4$) diffracting planes.
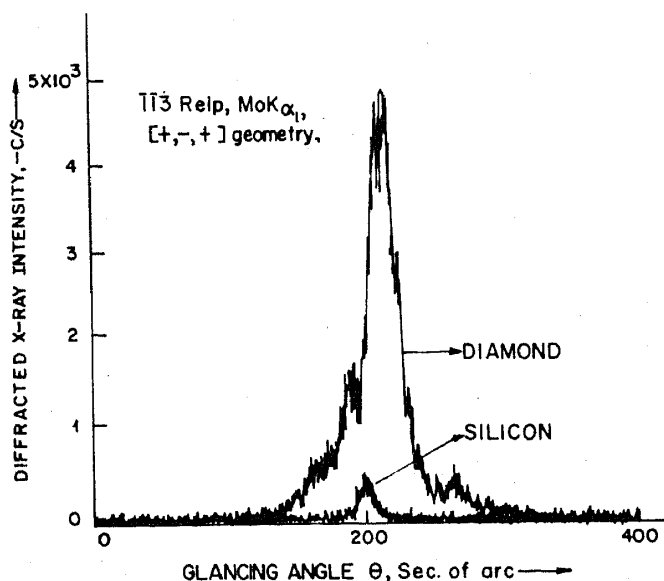
Figure 13. Diffraction curves of a diamond and a silicon single crystal recorded with ($\overline{1}\overline{1}3$) diffracting planes. MoK_{α_1} radiation; asymmetric Laue geometry and $(+,-,+)$ setting of the diffractometer were used.

crystal position and diffraction curves were recorded with the $(+, -, +, -)$ setting of the diffractometer. Symmetrical Bragg geometry was used. The third crystal was a dislocation free (111) silicon single crystal aligned for diffraction from ($\overline{2}20$) planes in the symmetrical Laue geometry. Figure 14 shows the diffraction curves recorded with the (111) diffracting planes of diamond and silicon single crystals. The difference between the peak intensities of these curves is quite small. The peak intensities are in the ratio $I_C/I_{Si} = 1\cdot1$.

3.3 A comparison of the absolute values of the integrated intensities of diamond and silicon crystals

The absolute value of the integrated intensity of a reflection can be determined experimentally by following the well known procedure (James 1950). The intensity of the exploring beam is carefully measured and maintained constant in these experiments. The area under the diffraction maxima is obtained from the curves shown in figure 8–13. The crystal was rotated with a constant velocity while recording these curves. The values of integrated intensity ρ, obtained by dividing the area of the curve by the intensity of the exploring beam, are given in tables 1 and 2, respectively, for diamond and silicon crystals.

For comparison with the experimental data, we have also calculated the values of the integrated intensities by assuming the crystals as: (i) ideally "perfect" and (ii) ideally "mosaic" crystals. For "perfect crystals" with moderate value of the absorption coefficient μ for the exploring beam, the integrated intensity is given by (James 1950)

$$\rho = \frac{8}{3\pi} N\lambda^2 |F| \frac{e^2}{mc^2} \frac{1+|\cos 2\theta|}{2\sin 2\theta} . \tag{1}$$

Here N = number of unit cells per unit volume, λ = wavelength of the exploring radiation, F = structure factor for the reflection under consideration; e, m = charge
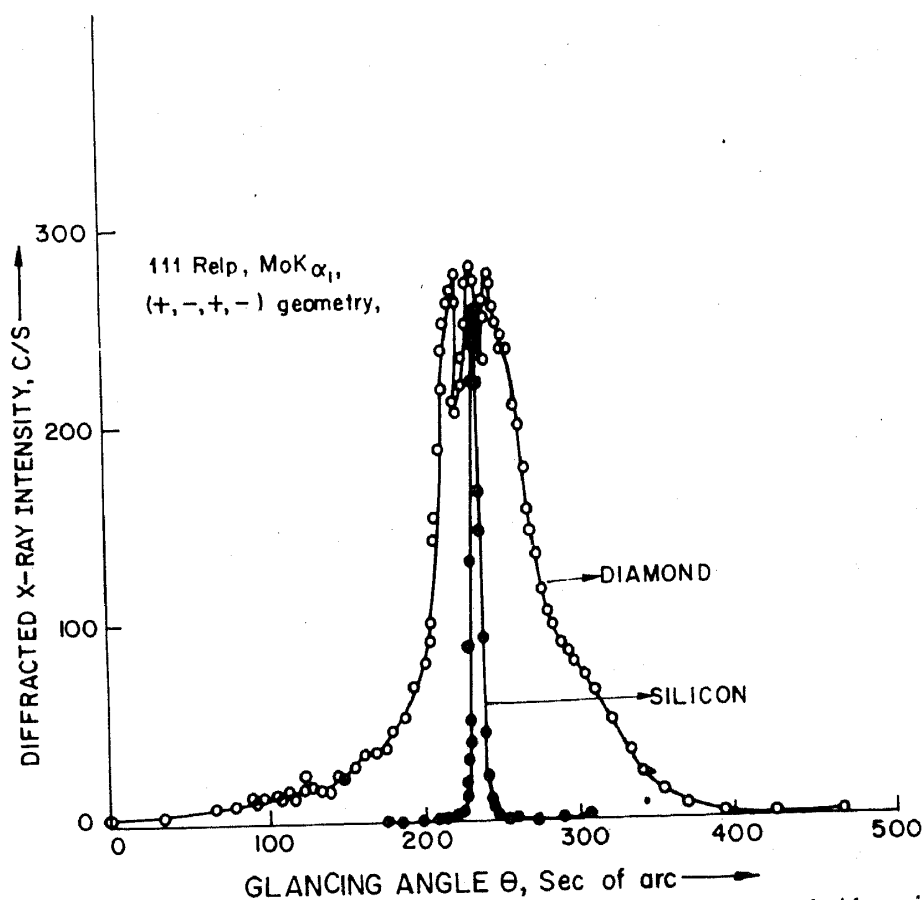
Figure 14. Diffraction curves of a diamond and a silicon single crystal recorded with specimen at the fourth crystal position of the diffractometer in the $(+,-,+,-)$ setting. 111 Relp, MoK$_{\alpha 1}$ radiation and symmetrical Bragg geometry were used. The curves were plotted after normalizing the intensities for equal area of irradiation for both types of crystals.

and mass of the electron, c = velocity of light, and θ = glancing angle = $\theta_B + \varepsilon$; θ_B being the Bragg angle and ε the angular deviation from that orientation. The exploring beam has been assumed to be unpolarized.

For an ideally imperfect or 'mosaic' crystal the integrated intensity is given by the following expression when the X-ray beam is unpolarized (James 1950):

$$\rho = \frac{N^2 \lambda^3}{2\mu} |F|^2 \left(\frac{e^2}{mc^2} \right)^2 \frac{1 + \cos^2 2\theta}{2 \sin 2\theta}. \tag{2}$$

We have used (1) and (2) for determining the theoretical values of ρ for all reflections. These are given in tables 1 and 2.

It is seen from table 1 that the experimental values of integrated intensities for diamond crystals lie between the values expected for ideally mosaic and perfect crystals. This is to be expected as all the specimens were moderately perfect (§ 3.1). From table 2 we find that the integrated intensities for silicon determined experimentally are closer to the theoretical values for a perfect crystal. In fact most of the values are even smaller than those calculated for nearly perfect crystals. This is rather surprising. It is another interesting anomaly revealed by this investigation. Further, work is in progress to understand this discrepancy.

Table 1. Absolute values of integrated intensities of different reflections from diamond single crystals at 27°C for $MoK_{\alpha 1}$ radiation.

Reflection hkl	Temperature correction $\exp[-(B\sin^2\theta/\lambda^2)]$	Theoretical values of integrated intensities		Experimental values of integrated intensities $(\times 10^{-6})$
		Perfect crystal model $(\times 10^{-6})$	Mosaic crystal model $(\times 10^{-6})$	
111	0·987	12·5	2455	229·2
333	0·925	1·61	108	83·0
$\overline{2}$20	0·977	2·01	117	90·6
$\overline{4}$40	0·911	1·92	189	50.0
$\overline{1}\overline{1}$3	0·969	3·78	403	31·3
$\overline{2}\overline{2}$4	0·933	2·57	281	16·6

Table 2. Absolute values of integrated intensities of different reflections from silicon single crystals at 27°C for $MoK_{\alpha 1}$ radiation.

Reflection hkl	Temperature correction $\exp[-(B\sin^2\theta/\lambda^2)]$	Theoretical values of integrated intensities		Experimental values of integrated intensities $(\times 10^{-6})$
		Perfect crystal model $(\times 10^{-6})$	Mosaic crystal model $(\times 10^{-6})$	
111	0·992	19·75	616·2	20·8
333	0·926	3·57	58·1	1·43
$\overline{2}$20	0·978	14·04	503·9	2·60
$\overline{4}$40	0·913	4·23	88·6	1.46
$\overline{1}\overline{1}$3	0·969	7·83	182·7	1·46
$\overline{2}\overline{2}$4	0·934	5·67	138·7	1·04

We have considered here theoretical expressions for integrated intensity from ideally perfect as well as from ideally mosaic crystals. It may be stressed that from this one cannot directly deduce the relationship between peak intensities at diffraction maxima of absolutely perfect crystals and ideally imperfect crystals. In this connection, one may quote James (1950) "... It must be borne in mind throughout this discussion (on integrated intensities) that we are considering the integrated reflection, and not the reflecting power when the crystal is set so as to reflect most strongly. This will always be greatest for the most perfect crystal, whereas, as we have just seen, the integrated reflection is increased by the introduction of imperfections."

4. Discussion

Results of experiments reported above have clearly established that the observed peak intensities of diffraction are higher for diamond crystals in comparison to those for

silicon crystals. For higher order reflections like 333 the diamond peak intensity is more than ten times that of the silicon crystals. It may be emphasized that these experiments have been performed by using recently developed high resolution techniques employing highly collimated and monochromated $K_{\alpha 1}$ beam. Therefore, there is no possibility of experimental factors being responsible for these observations.

This result is new and anomalous due to the following two reasons. The atomic scattering factor of silicon atoms is considerably higher than that of the carbon atoms due to their widely different atomic numbers. Further, the degree of perfection of silicon crystals is much higher than that of diamond crystals. This fact has been well established by the diffractometric and topographic evaluations (§ 3.1) and integrated intensity measurements (§ 3.4). For a given reflection the ratio of peak intensities for diamond and silicon — I_C/I_{Si} varies from specimen to specimen, yet the overall result is the same.

The absorption coefficient of the exploring beam is different for diamond and silicon. Correction for absorption has been applied. The observed peak intensity had already been reduced due to absorption and, therefore, (observed) $I_C = I_C^0 \exp(-\mu_C t_C')$ or $I_C^0 = I_C \exp(\mu_C t_C')$. Similarly, $I_{Si}^0 = I_{Si} \exp(\mu_{Si} \cdot t_{Si}')$. Here $t' = t \sec \theta_B$.

Hence

$$I_C^0/I_{Si}^0 = I_C/I_{Si} \exp(\mu_C t_C' - \mu_{Si} t_{Si}'). \tag{3}$$

The values of I_C/I_{Si} and I_C^0/I_{Si}^0 have been calculated and are given in table 3 for different reflections. It is very tempting to compare the experimentally observed intensities with theoretically calculated values. It is possible to make calculations of diffracted intensities at diffraction maxima for ideally perfect crystals in the case of idealised experimental conditions conforming to plane wave conditions and ideal monochromaticity (Pinsker 1978). However, it is not possible to calculate intensities at diffraction maxima in moderately perfect crystals. Even in an ideally imperfect or mosaic crystal, theoretical expressions for peak intensities are not available. Therefore, a comparison between theoretically expected peak values for perfect and imperfect crystals is not possible at present. However, even if such a quantitative comparison is made theoretically it will not explain the observed anomaly. The peak intensity for perfect crystals will always be higher than that of imperfect crystals. It may be mentioned that experimental comparison of intensities of nearly perfect and not so perfect crystals has been made several times in our group. Some of these results have been published showing that the peak intensities of perfect crystals are higher than those of less perfect crystals of the same substance (Lal *et al* 1979; Lal 1981).

Integrated intensities of diffraction from diamond crystals have been studied extensively by Raman group (Ramachandran 1944 and Hariharan 1944). It is well established that the values of ρ_C vary from specimen to specimen. The variation for

Table 3. Values of I_C/I_{Si} and I_C^0/I_{Si}^0 for different reflections.

Reflection	Observed value of I_C/I_{Si}	Value of I_C^0/I_{Si}^0
$\bar{2}20$	4·5	1·37
$\bar{4}40$	7·0	2·02
$\bar{2}\bar{2}4$	7·5	2·21
$\bar{1}\bar{1}3$	10·5	3·10

111 reflection can be in the range: 4×10^{-5} to 2×10^{-3}, a factor of 50 for CuK$_\alpha$ radiation (Lonsdale 1948). The experimental results on ρ reported in table 1 are in agreement with this keeping in view the wavelength of the exploring radiation and the structure factors of the reflections studied. However, in the present investigation emphasis has been on comparison of peak intensities from two crystals having same structure but widely different structure factors. The peak intensity is a direct manifestation of the quality of atomic ordering inside a crystal and the range over which it extends. All the experimental data reported here establish that the degree of perfection of silicon crystals is close to the ideally perfect state and yet its peak intensities are lower than those of diamond crystals which are moderately perfect. The consideration of structure factor values further enhance this anomaly.

One may argue that the observed anomaly could be partly due to large size of mosaics in diamond crystals in comparison to primary extinction distance and/or due to the resolution width of the experimental system being larger than the silicon diffraction peaks but smaller than the diamond peaks. Even if one assumes that the mosaic size in a diamond crystal is greater than the primary extinction distance, the observed anomaly cannot be accounted for due to the following reasons. In an ideally perfect crystal all mosaics are oriented identically and therefore the entire crystal can be treated as one single mosaic. In a mosaic crystal like the diamond specimens investigated here, the individual mosaics, small or big, are misoriented with respect to each other and their orientation lies in a wide angular range. Therefore, in a mosaic crystal, at any given orientation of the specimen on the diffraction curve, only a fraction of all mosaics would be oriented to satisfy the diffraction condition. Hence, the diffracted intensity will be much lower than that expected when all of them would be oriented identically, as in the case of a perfect crystal. In the present experiments, the diffraction curves of diamond crystals are broad with half widths of about 100 arc sec or so as compared to the theoretical values of a few arc sec for a perfect diamond crystal. Therefore, by these considerations, one cannot explain the observed anomaly. It is true that in all experimental systems, there is a finite divergence and wavelength spread in the exploring X-ray beam leading to a finite resolution width. This width in our experimental system has been estimated as 3 arc sec, as measured by direct experiments. This limit is comparable with the diffraction curve half widths for low order reflections of perfect crystals of silicon as well as diamond. For example, theoretical values of half widths of diamond and silicon crystals for different reflections are given below:

Theoretical half widths of diffraction curves (arc sec)

Reflection	Silicon	Diamond
111	2·97	1·96
333	0·67	0·35
220	2·19	1·14
440	0·79	0·46
224	0·99	0·54
113	1·25	0·65

These values have been calculated by us for plane polarized incident wave front (Pinsker 1978). It is seen that the resolution width is not small compared to the

theoretical half widths of diamond crystals and larger compared to that for silicon crystals. The observed broad curves for diamond crystals are due to mosaic spread. Therefore, the observed anomaly cannot be explained on the basis of different values of resolution width in relation to diffraction curve half widths of diamond and silicon crystals.

The results reported in this paper have established interesting anomalies which deserve further work for complete understanding. In this connection we have borrowed several diamond crystals from Professor C V Raman's collection at Raman Research Institute, Bangalore, for which we are grateful to the Director, Professor V Radhakrishnan and Professor S Ramaseshan.

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References

Bonse U and Hart M 1965 *Appl. Phys. Lett.* **7** 238
Bragg W H 1914 *Philos. Mag.* **12** 881
Bragg W H 1921 *Proc. Phys. Soc. London* **33** 304
Brill R, Grimm H G, Hermann C and Peters C L 1939 *Ann. d. Phys.* **34** 393
Brindley G W 1932 *Proc. Leeds Philos. Soc.* **2** 271
Brindley G W 1933 *Proc. R. Soc. (London)* **A140** 301
Ehrenberg W, Ewald P P and Mark H 1928 *Z. Kristallogr.* **66** 547
Hariharan P S 1944 *Proc. Indian Acad. Sci.* **A19** 261
Hart M 1971 *Rep. Prog. Phys.* **34** 435
James R W 1950 *The optical principles of the diffraction of X-rays* (London: G Bell and Sons) p. 271
Lal Krishan, Singh B P and Verma A R 1979 *Acta Crystallogr.* **A35** 286
Lal Krishan 1981 *Proc. Indian Natl. Sci. Acad. Suppl.* **A47** 20
Lal Krishan 1982 *Synthesis, crystal growth and characterization* (ed.) Krishan Lal (Amsterdam, New York, Oxford: North Holland) p. 287
Lal Krishan and Thoma P 1983 *Phys. Status Solidi.* **A80** 491
Lal Krishan 1985 *NPL Research Report* No. NPL-85-B4-0051
Lal Krishan 1988 *Crystal properties & preparation* **16** 143
Lal Krishan and Goswami S N N 1987 *Materials science and engineering* **85** 147
Lang A R 1974 *J. Cryst. Growth* **24/25** 108
Lonsdale K 1948 *Crystals and X-rays* (London: G. Bell and Sons) p. 138
Patel J R and Betterman B W 1963 *J. Appl. Phys.* **34** 2716
Pinsker Z G (1978) *Dynamical scattering of X-rays in crystals* (Berlin, Heidelberg, New York: Springer-Verlag)
Ramachandran G N 1944 *Proc. Indian Acad. Sci.* **A19** 304
Renninger M 1935 *Z. Tech. Phys.* **16** 440
Renninger M 1937 *Z. Phys.* **106** 141
Tanner B K 1976 *X-ray diffraction topography* (Oxford: Pergamon Press) p. 18