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High-Order Multiple Diffraction in GaAs*

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Multiple diffraction patterns of a single crystal of GaAs have been recorded with Cu $K\alpha_1$ radiation. Calculations indicated that 19 peaks, all involving reflections with odd indices, should be observed in the 45° asymmetric angular range of the multiple diffraction pattern. All these were observed and indexed. In addition, six sharp but barely detectable intensity minima, were observed in the background. These result from one eight-beam, one six-beam, and four four-beam interactions, all involving reflections with *even* indices. The geometry of these interactions is discussed and their integrated intensities have been calculated. These show fair agreement with the corresponding measured intensities.

Introduction

Diffraction effects not readily detected by other methods are frequently encountered in multiple-diffraction investigations of single crystals. Several such effects, resulting from interactions involving four, six and eight beams, have been observed in 002 multiple diffraction patterns in gallium arsenide crystals. Their geometry and intensities are discussed below.

Gallium arsenide crystallizes in the cubic system with four molecules per unit cell. The space group is $F\bar{4}3m$. The 002 reflection, whose intensity equals zero in crystals with the diamond structure, is weak but readily detectable in gallium arsenide. It is due primarily to the small difference between the atomic scattering factors of the two component elements; $|F|_{002} \sim f_{As} - f_{Ga}$. As a result, multiple diffraction *minima*, which obviously cannot be observed in 002 patterns of diamond-type crystals, where the background intensity due to the 002 reflection equals zero, may be observed in corresponding patterns of gallium arsenide.

The geometry and intensities of multiple diffraction interactions have been discussed by many authors, including Renninger (1937), Cole, Chambers & Dunn (1962), Moon & Shull (1964), Zachariasen (1965), Caticha-Ellis (1969), Prager (1971) and Post (1975). A detailed bibliography is included in a review paper by Terminasov & Tuzov (1964).

Experimental

The specimen was a crystal slab about 1·8 cm long and 5 mm wide by 2 mm thick. The large face was cut normal to [001] and polished and etched in the usual way.

The experimental arrangement is similar to the one described by Renninger (1937), modified to improve resolution. It is illustrated schematically in Fig. 1.

A 'fine-focus' X-ray copper target was used; its effective size, at a 4° take-off, is 400 × 500 μm. An evacuated tube, 120 cm long, separates the source from the specimen. The incident beam passes through a 0·5 mm diameter pinhole in a brass insert at the exit end of the tube. The arrangement limits the angular divergence of the incident beam to 2′ of arc.

The crystal was set at the Bragg angle for the 002 reflection and rotated slowly about the [002] diffraction vector, taking care to avoid disturbing the precise Bragg setting. (In this manuscript, 002 refers to the 'primary reflection'.) As other ('secondary') reflections enter and leave the Ewald sphere, singly, in pairs, or in greater numbers, the resultant interactions give rise to signals received by a detector monitoring the primary reflection. *hkl* indices listed on the multiple diffraction charts refer to these secondary reflections.

Results and discussion

A 45° asymmetric portion of the indexed 002 multiple diffraction pattern of gallium arsenide, recorded with Cu $K\alpha_1$, is shown in Fig. 2. It will be noted that all the maxima in Fig. 2 are associated with reflections with odd indices. The explanation is simple. The reflection coupling the secondary reflection i, to the primary reflection j, has indices $(j-i)$ (Fig. 3). In gallium arsenide the reflections coupling the strongest secondary reflections (*i.e.* those with even indices and with $h+k+l=4n$) to the primary reflection, 002, are very weak and these interactions are therefore almost undetectable. Secondary reflections with odd indices are coupled to the primary reflection by odd-index terms; in most instances, both the secondary and the coupling reflections are then of moderate intensity and their interactions are readily detected, as seen in Fig. 2.

Closer inspection of Fig. 2 reveals a number of barely detectable intensity *minima* labelled *A* to *F*. The latter are displayed more clearly in Fig. 4 in which the data are plotted on a more sensitive scale.

Interactions *A*, *C*, *D* and *E* are four-beam types, involving *two* secondary reflections. The geometries of the eight- and six-beam interactions are shown in Figs.

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5 and 6. As the crystal is rotated about [002] all the reciprocal-lattice points involved in a given interaction reach their diffracting positions simultaneously, *i.e.*, at that instant all lie on the surface of the Ewald sphere. In the eight-beam case all those reflections lie on the circumference of the circle formed by the intersection of the $h0l$ plane and the Ewald sphere (Fig. 5). The radius of that circle is R'. The six-beam case is similar; the reciprocal lattice plane involved is $(h\bar{h}0)$. It is clear that additional high, and possibly higher, order interactions would be observed if shorter wavelengths were used.

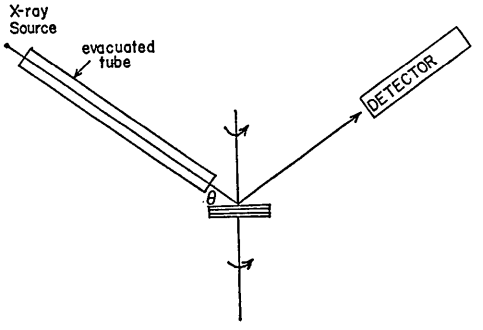

Fig. 1. Experimental setup (schematic).

Intensity calculations

The perfection of our gallium arsenide specimen was not checked in our laboratory. The supplier had informed us that it was 'essentially perfect'. It was clearly far removed from the 'ideally mosaic' category for which intensity calculations based on the kinematical theory are designed. In general, calculations of the multiple-diffraction interaction intensities for essentially perfect crystals should be based on the dynamical theory of diffraction. In the case of very weak interactions, however, Hirsch & Ramachandran (1950) have shown that the values of integrated intensities calculated on the basis of kinematical considerations on the one hand, and dynamical on the other, approach one another asymptotically as the corresponding values of the structure factors approach zero. Our calculations of the integrated intensities were therefore limited to the weakest multiple-beam interactions: the six even-index negative 'peaks' and the three weakest odd-index reflections.

We used the procedure of Moon & Shull (1964) for the calculation of integrated multiple diffraction intensities in mosaic crystals, modified to include polarization factors as described by Zachariasen (1965):

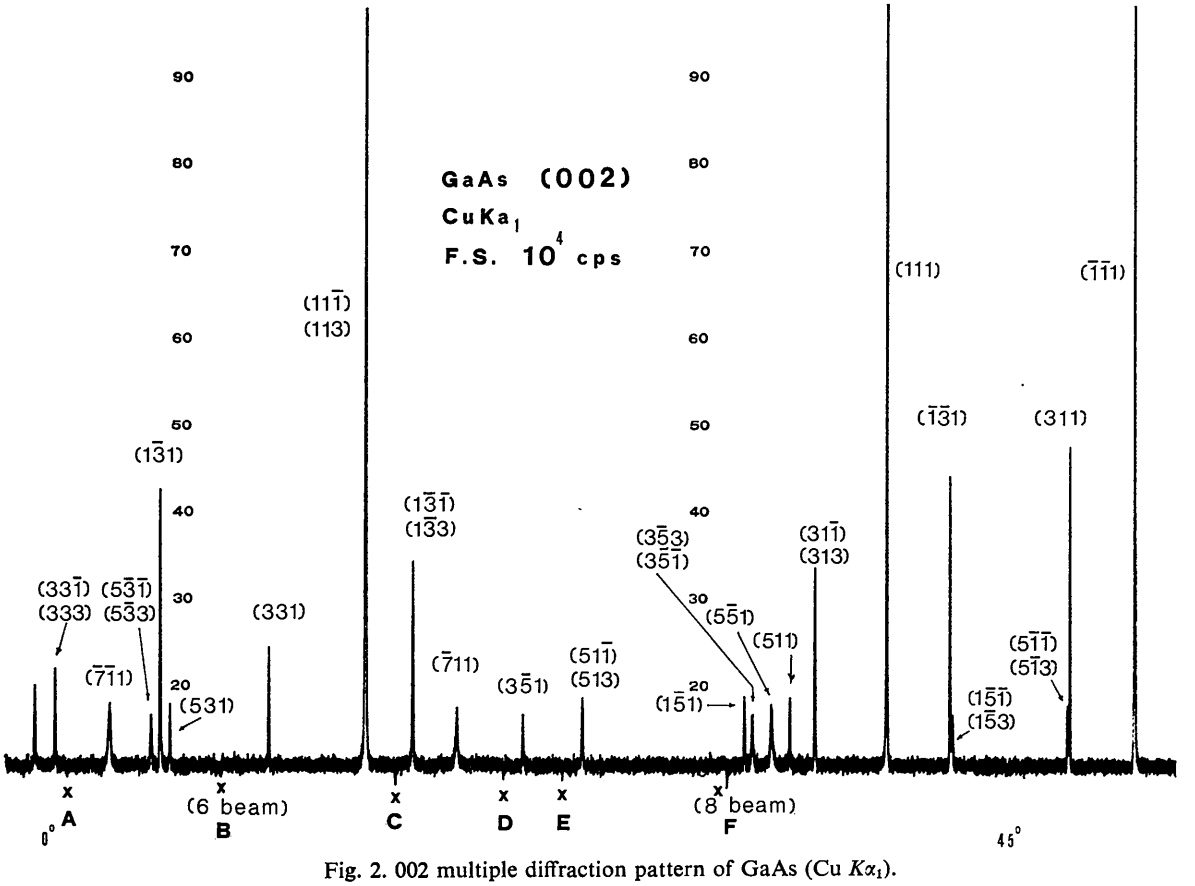

Fig. 2. 002 multiple diffraction pattern of GaAs (Cu $K\alpha_1$).

$$I_{\text{int.}} \simeq \sum_i \left\{ -\left(\frac{1}{K_{0i}}\right) Q_0 p_{1,i}[i-1] - \left(\frac{1}{K_{1i}}\right) Q_{1i} p_{1,i-1}[(i-1)-1] + (K_{0i}^2+K_{i1}^2)^{-1/2} \left(\frac{Q_{0i}Q_{i1}}{Q_{01}}\right) p_{i,i-1}[(i-1)-i] \right\}, \quad (1)$$

where

$$Q_{ij} = \left(\frac{\lambda^3 N^2 |F|^2}{\sin 2\theta}\right)_{ij}, \quad (2)$$

$$p_{i,j}(j-i) = \tfrac{1}{2}[\cos^2 2\theta_i + \cos^2 2\theta_j, + (\cos 2\theta_{i-j} - \cos 2\theta_i \cdot \cos 2\theta_j)^2] \quad (3)$$

$$K_{ij} = \left(\frac{\sin\psi \cdot \cos\chi \cdot \cos\xi}{\sin 2\theta}\right)_{ij}, \quad (4)$$

$$\sin\psi \cdot \cos\chi \cdot \cos\xi = 1/\text{Lorentz Factor (Fig. 7)}. \quad (5)$$

In equation (3), the double subscripts (i,j) refer to a beam i, incident on a set of planes with indices $(j-i)$, and reflected into direction j. i does not necessarily refer to the beam incident *on the crystal*. When integers 0 and 1 are used as subscripts they refer to the beam incident *on the crystal*, and to the primary reflected beam respectively. Q_{ij} is the reflectivity associated with the planes $(j-i)$; $P_{i,j}$ $(j-i)$ is the polarization factor; the double subscripts are defined above and $(j-i)$ refers to the term coupling i to j.

The angles used to calculate the Lorentz factor [equation (5)] are illustrated in Fig. 7. The intensity measurements were made by scanning slowly over the range of azimuthal angle within which the maxima (or minima) of interest appeared; the corresponding number of counts diffracted by the primary reflection in the same time (or angular) interval, in the absence of simultaneous diffraction, was then subtracted from the total number recorded to yield the 'integrated multiple diffraction intensities'.

Equation (1) may be used only when the intensity of the primary reflection is greater than zero. A more general expression [equation (9) of Moon & Shull], which yields the ratio of the change of the primary diffracted

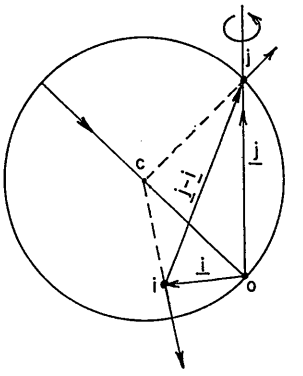

Fig. 3. Coupling reflection $(j-i)$ in multiple diffraction.

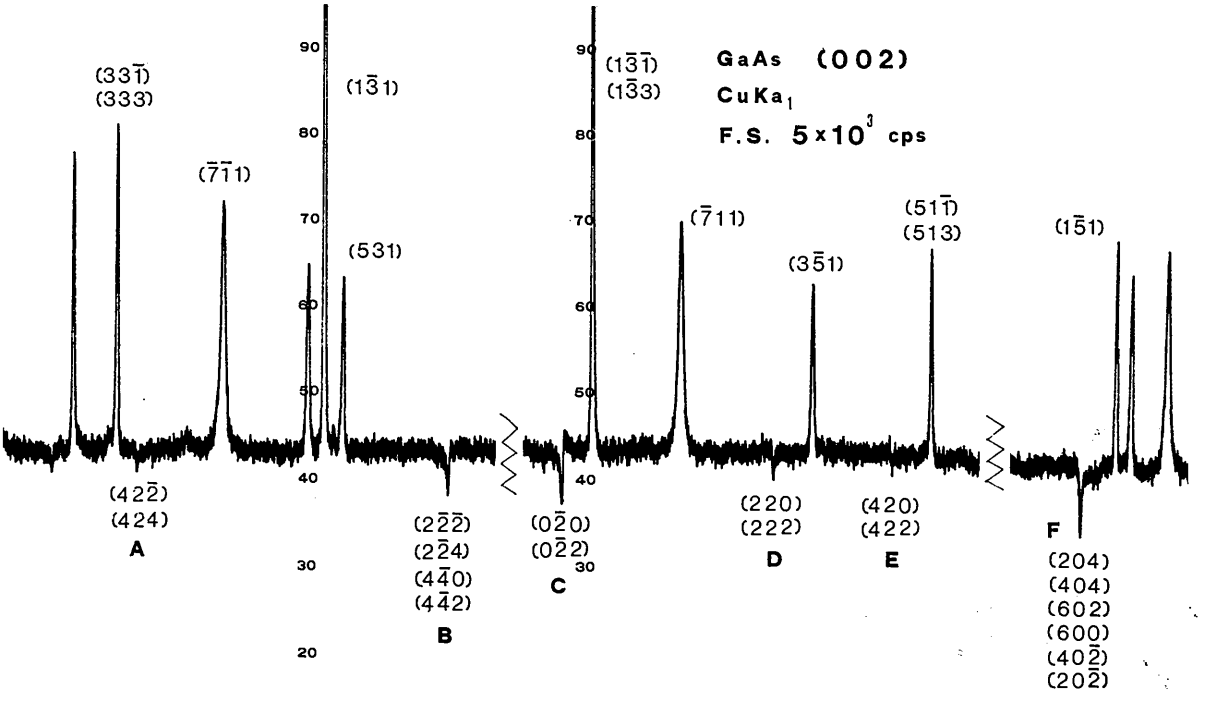

Fig. 4. Fig. 2 on a more sensitive scale.

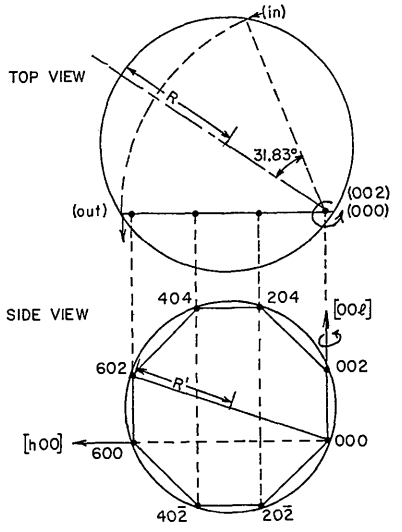

Fig. 5. Geometry of the eight-beam case.

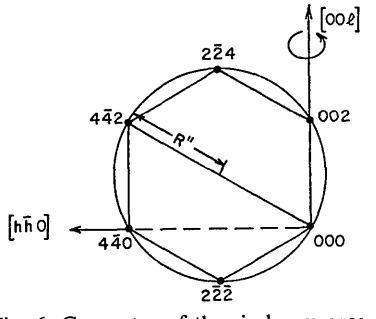

Fig. 6. Geometry of the six-beam case.

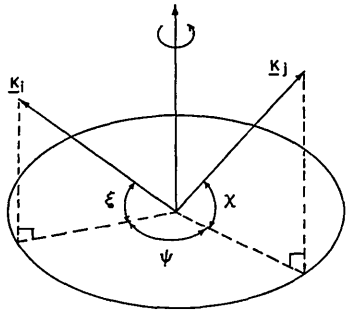

Fig. 7. Angles used for determination of Lorentz factor [based on Fig. 3 of Moon & Shull (1964)].

intensity to the intensity of the incident beam, may be used if the primary reflection intensity (Q_{01}) equals zero.

Clearly, it is not possible to claim high precision for the intensity measurements listed in Table 1. The interactions were all weak and measurements were made in the presence of a relatively high background. Relative standard deviations of the measured intensities average about 40% of the listed values for the even-index reflections and about 20% for the three odd-index terms, and, because the results can only be viewed as approximate, no effort was made to calculate the path lengths of the individual beams within the crystal and these were omitted from the expressions for calculated intensities.

Table 1. *Measured and calculated integrated intensities of weak multiple diffraction interactions*

			ΔI Integrated intensities	
hkl	*N*	φ (°)	Measured	Calculated
$42\bar{2}/424$	4	0·99	−0·8	−0·66
Six-beam*	6	8·23	−1·6	−0·99
$0\bar{2}0/0\bar{2}2$	4	16·45	−1·2	−0·82
220/222	4	21·39	−1·1	−1·05
420/422	4	24·14	−0·5	−0·33
Eight-beam†	8	31·83	−2·6	−3·95
$33\bar{1}/333$	4	0·50	7·8	7·82
$5\bar{3}1/5\bar{3}3$	4	4·93	4·8	5·10
$51\bar{1}/513$	4	25·05	5·4	5·08

* $(2\bar{2}\bar{2})$ $(2\bar{2}4)$ $(4\bar{4}0)$ $(4\bar{4}2)+(000)$ (002).
† (204) (404) (602) (600) $(40\bar{2})$ $(20\bar{2})+(000)$ (002).

Although the agreement between measured and calculated intensities is far from perfect, the two sets do show qualitative agreement; in particular, the calculated values of the contributions of each of the six sets of even-index reflections to the 002 intensity are negative, in agreement with Figs. 2 and 4.

References

CATICHA-ELLIS, S. (1969). *Acta Cryst.* A**25**, 666–673.
COLE, H., CHAMBERS, F. W. & DUNN, H. M. (1962). *Acta Cryst.* **15**, 138–144.
HIRSCH, P. B. & RAMACHANDRAN, G. N. (1950). *Acta Cryst.* **3**, 187–194.
MOON, R. M. & SHULL, C. G. (1964). *Acta Cryst.* **17**, 805–812.
POST, B. (1975). *J. Appl. Cryst.* **8**, 452–457.
PRAGER, P. R. (1971). *Acta Cryst.* A**27**, 563–569.
RENNINGER, M. (1937). *Z. Phys.* **106**, 141–176.
TERMINASOV, Y. S. & TUZOV, L. V. (1964). *Usp. Phys. Nauk*, **83**, 223–258.
ZACHARIASEN, W. H. (1965). *Acta Cryst.* **18**, 705–710.