Table 2. *Powder pattern of* Na_2SO_4 (III)

hkl	$\sin^2 \theta_c$	I_c	$\sin^2 \theta_o$	I_o
110	0·0265	27	0·0265	*m*
020	0·0298	12	0·0299	*w*
111	0·0388	168	0·0386	s^+
021	0·0421	148	0·0423	s^+
002	0·0492	103	0·0495	*s*
112	0·0757	336	0·0759	*vs*
200	0·0760	169	—	—
022	0·0790	2	—	—
130	0·0861	224	0·0861	s^+
131	0·0984	3	0·0986	*vw*
220	0·1058	95	0·1060	*s*
221	0·1181	0	—	—
040	0·1194	34	0·1194	*w*
202	0·1252	5	0·1250	w^-
041	0·1317	22	0·1317	w^+
132	0·1353	26	0·1356	*w*
113	0·1372	26	0·1373	w^+
023	0·1405	18	0·1406	*w*
222	0·1550	89	0·1548	*s*
042	0·1686	50	0·1684	*m*
310	0·1786	1	—	—
311	0·1909	25	0·1907	*w*
240	0·1954	8	—	—
004	0·1968	180	0·1967	*s*
133	0·1968	5	—	—
150	0·2055	5	—	—
241	0·2077	15	0·2078	*w*
223	0·2165	0	—	—
151	0·2178	0	—	—
114	0·2233	1	0·2237	*vw*
024	0·2266	4	0·2269	*w*
312	0·2278	10	—	—
043	0·2301	11	0·2301	w^-
330	0·2382	43	0·2378	s^-
242	0·2446	61	0·2446	s^-
331	0·2505	3	—	—
152	0·2547	23	0·2543	*m*
060	0·2686	3	—	—
204	0·2728	10	0·2725	w^-
061	0·2809	0	—	—
134	0·2829	46	0·2829	*s*
332	0·2874	11	0·2872	w^-
313	0·2893	4	—	—
224	0·3026	2	—	—
400	0·3041	27	0·3041	m^-
243	0·3061	0	—	—
.	.	.	.	.
.	.	.	.	.
.	.	.	.	.

The indexing of the powder pattern of Na_2SO_4 (III) given in Table 2 was established by comparison of diagrams from the series Na_2SO_4–Na_2CrO_4 and from the sulfate-rich part of a series Na_2SO_4–Na_2CO_3. The differential variation of the lattice dimensions with increasing chromate or carbonate content allows the unequivocal assignment of indices. It was found that the tetragonal indexing as given by Das Gupta (1954) breaks down at rather slight deformations of the cell, in contrast to the orthorhombic indexing. Moreover, intensities calculated with Frevel's parameters* agree well with the estimated intensities given in the table.

On the basis of this study, it appears evident that Na_2SO_4 (III) is isomorphous with Na_2CrO_4, the orthorhombic symmetry of which was established by Miller (1936), using single-crystal methods. In this connection, attention may be drawn to Niggli's correction (1954) of the space-group description for Na_2CrO_4, and thus for Na_2SO_4. The space group is correctly described as *Cmcm*.

All powder patterns in this study were taken with Cu $K\alpha$ radiation in a Guinier camera calibrated with NaCl on the basis of the following constants:

$$\lambda_{\text{Cu } K\alpha} = 1{\cdot}54179 \text{ Å}, \ a_{\text{NaCl}} = 5{\cdot}63984 \text{ Å} .$$

The difference in scale between the present values of $\sin^2 \theta$ and those of Das Gupta (1954) and Frevel (private communication) may be due to a different calibration standard. Film shrinkage was automatically corrected for by the procedure of Hägg (1947).

The author is indebted to Dr I. Lindqvist for suggesting the problem and for his continued interest, and to Mr A. Westlund for experimental help. The investigation was supported by a grant of the Swedish Natural Science Research Council (Statens Naturvetenskapliga Forskningsråd) which is gratefully acknowledged. Finally, the author wishes to express his gratitude to Prof. G. Hägg for the facilities placed at his disposal.

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* The author is indebted to Dr L. K. Frevel for a table of *f* values used in his original computations.

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A dynamical theory of the simultaneous reflexion by two lattice planes. III. Experimental verification.

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In previous notes (Kambe & Miyake, 1954; Miyake & Kambe, 1954; hereafter referred to as Part I and Part II), it was shown theoretically that the relative phase angle of the structure amplitudes of two lattice planes of a crystal, say $h(h_1h_2h_3)$ and $h'(h'_1h'_2h'_3)$, may be obtained from diffraction patterns in which the Bragg reflexions by these lattice planes occur simultaneously. In the

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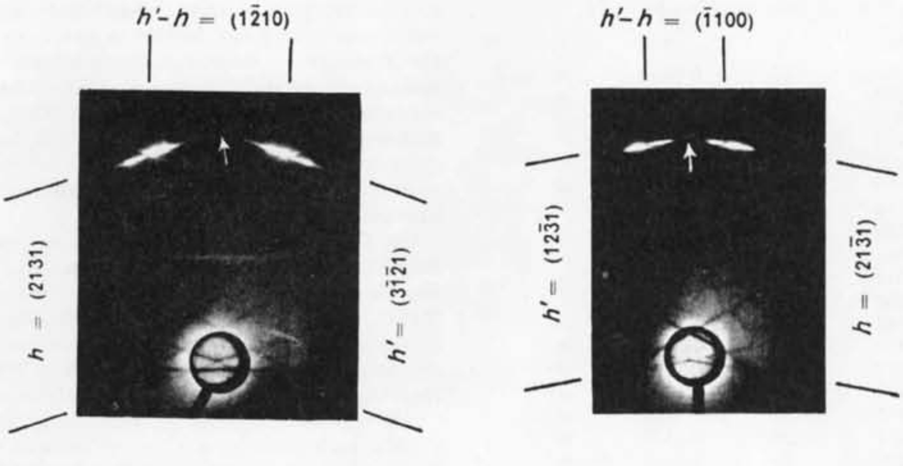


(i) (ii)

Fig. 1. Convergent-beam electron-diffraction patterns of graphite, showing the anomalies at the position of the simultaneous reflexion. (Accelerating voltage 41 kV.; crystal thickness ~ 1000 Å.) The exposure of the primary spot is diminished by covering it with a disc. Kikuchi lines appear as elongations of extinction lines in the primary spot and of the bright diffracted lines. Kikuchi bands associated with the reflexions h, h', and $h'-h$ are indicated in the margin.

(i) $h = (21\bar{3}1)$, $h' = (3\bar{1}\bar{2}1)$, $h'-h = (1\bar{2}10)$.
(ii) $h = (21\bar{3}1)$, $h' = (12\bar{3}1)$, $h'-h = (\bar{1}100)$.

present note an experimental verification of the above result is presented by taking an example in electron-diffraction patterns from a thin graphite crystal.

We have used a convergent-beam method originated by Kossel & Möllenstedt (1939). In this method, the electrons passing through the crystal produce a primary spot having a certain extended area in the pattern. The directions of incident beam satisfying the Bragg condition for a certain lattice plane are marked as an extinction line in this spot; the bright line due to this reflexion appears elsewhere in the pattern. When the extinction lines for the two reflexions h and h' cross each other in the primary spot, the simultaneous reflexion takes place. The formulations given in Part I are particularly useful in finding the intensity distribution around the crossing point.

When the structure amplitudes (or the Fourier coefficients of the periodic potential in the crystal) for the relevant reflexions satisfy $|V_{h'-h}| \gg |V_h|, |V_{h'}|$, the two extinction lines do not actually cross each other but form a hyperbola by the effect of mutual coupling of the two reflexions (Fues, 1939). When certain geometrical conditions assumed in Part II are satisfied, the particular cross-section of the dispersion surface ($c = 1$) used in Part II corresponds to a line (XX in Fig. 2) in the primary spot joining the vertices of the hyperbola (A_+, A_- in Fig. 2). Along this line the intensity (more properly, the amount of its decrease) varies in the same way as a curve shown in Fig. 1 of Part II. As proved in Part II, at the vertices of the hyperbola, corresponding to the two peaks in Fig. 1 of Part II, the intensity (integrated along the above-mentioned line) is proportional to either $|V_h + V_{h'}|$ or $|V_h - V_{h'}|$, according to whether the angle between the direction of the corresponding incident beam and the reflecting net-plane is smaller or larger than the exact Bragg angle for both reflexions; in other words, according to whether the vertex concerned lies inside or outside both of the two Kikuchi bands with the indices h and h'. (Here the term 'band' means a region between a pair of bright and dark Kikuchi lines in the pattern.)

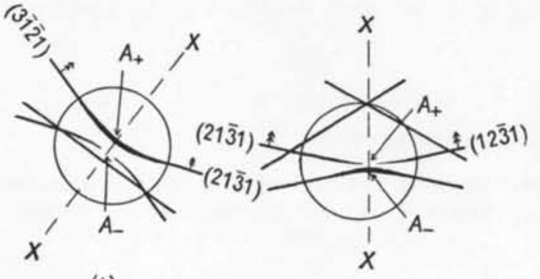
In the present observation, two reflexions participating in the simultaneous reflexion are chosen from $\{21\bar{3}1\}$ of graphite. Among various combinations of the net-planes, the following nearly satisfy $|V_{h'-h}| \gg |V_h|, |V_{h'}|$, satisfying also the geometrical conditions assumed in Part II:

(i) The combination of $(21\bar{3}1)$ and $(3\bar{1}\bar{2}1)$. $h'-h = (1\bar{2}10)$, $|V_{h'-h}| = |V_{110}| = 3{\cdot}29$ V.

(ii) The combination of $(21\bar{3}1)$ and $(12\bar{3}1)$. $h'-h = (\bar{1}100)$, $|V_{h'-h}| = |V_{100}| = 1{\cdot}47$ V.

In both cases $|V_h| = |V_{h'}| = |V_{211}| = 0{\cdot}63$ V. In the usual choice of coordinate axes in the crystal, $V_{1\bar{2}10}$ and $V_{\bar{1}100}$ are real and positive, $V_{21\bar{3}1}$, $V_{3\bar{1}\bar{2}1}$, and $V_{12\bar{3}1}$ have respectively the phase angles $+\frac{1}{2}\pi$, $+\frac{1}{2}\pi$, and $-\frac{1}{2}\pi$. In case (i), the structure amplitudes of the relevant two reflexions have the same phase angle, *i.e.* they have the *same sign*. In case (ii), they have *opposite signs*.

In the above two cases we have



(i) (ii)

Fig. 2. Sketches of the primary spots in Fig. 1. XX is a line joining the vertices A_+, A_- of the hyperbola. Double arrows attached to the Kikuchi lines indicate the inner side of the bands.

$$\text{(i)}\ |V_h+V_{h'}| = 2|V_{211}|,\ |V_h-V_{h'}| = 0\,,$$
$$\text{(ii)}\ |V_h+V_{h'}| = 0,\ |V_h-V_{h'}| = 2|V_{211}|\,,$$

so that we can expect that the intensity relation between the two vertices of the hyperbola is reversed for the two cases. This is verified experimentally very well, as shown in Figs. 1 and 2. In case (i), the vertex lying inside both of the Kikuchi bands ($21\bar{3}1$) and ($3\bar{1}\bar{2}1$) (A_+ in Fig. 2) is actually found to be strengthened. Whereas in case (ii) the outside vertex (A_- in Fig. 2) is strengthened. It is seen that the bright lines appearing in the diffracted spots behave in a corresponding manner. By observing such effects, the relative sign of the two structure amplitudes can be determined uniquely.

We see in Fig. 1 that an intensity distribution similar to that of the extinction lines in the primary spot is found at the crossing point of Kikuchi lines which lies outside the diffraction spots (indicated by an arrow). In fact, by applying our scheme given in Parts I and II to the theory of Kikuchi pattern (Laue, 1935; Kainuma, 1954) we find that a similar effect of the phase-angle relation is expected in the intensity distribution of Kikuchi patterns. This effect can be observed also at the crossing points of Kikuchi lines in reflexion patterns from natural and cleavage faces.

I wish to thank Prof. Shizuo Miyake for his guidance throughout this work.

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Steric hindrance in the dibiphenylene-ethylene molecule.

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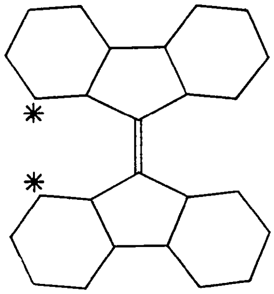
The crystal structure of dibiphenylene-ethylene (bifluorene) has been subject to partial three-dimensional analysis by Fenimore (1948), who concluded that the X-ray data were satisfactorily accounted for by centrosymmetric, essentially planar molecules.



In the planar molecule, the distance between starred carbon atoms would be only 2·5 Å and although, as pointed out by Fenimore, there is chemical and spectroscopic evidence indicative of steric interaction here, there is the more serious problem of the hydrogen atoms attached to the conflicting carbon atoms. Assuming the C–H bond to be 1·0 Å in length and coplanar with the rest of the molecule, the distance between hydrogen centres is only 0·7 Å. The van der Waals diameter for hydrogen is about 2·4 Å and it is evident that unless the C–H bonds are severely bent out of plane the steric repulsion should disturb the molecular planarity. Attention has also been drawn to this by Bell (1952) in connexion with the possibility of producing stereoisomeric derivatives.

The crystallographic data, according to Fenimore, are:

System: orthorhombic .
$a = 17{\cdot}22,\ b = 36{\cdot}9,\ c = 8{\cdot}23$ Å .
Space group: *Pcan*, uniquely determined .
$Z = 12$.

Four of the molecules are centred on special repeats of that at the origin and are necessarily centrosymmetric. The remaining eight are centred on general repeats of that at approximately ($\frac{1}{2}, \frac{1}{6}, 0{\cdot}434$) and, although not required to be centrosymmetric, were assumed to be so.

All the molecules lie close to (031) and ($0\bar{3}1$) planes so that it is the x projection which is critical in deciding the planarity question. Fenimore's analysis of this projection was two-dimensional and the resolution too poor for this question to be settled with certainty. A ($0\bar{3}1$) section of the molecule at the origin was fairly well resolved, but again this is not decisive evidence for or against strict planarity.

Accordingly, it was decided to use all Fenimore's F_o's and calculated phases to analyse the structure further by three-dimensional x sections.

Fig. 1(a) shows superimposed x sections chosen to pass through (or nearly through) the atoms in the region of the origin. As far as the (left-hand) molecule at the origin is concerned, resolution was sufficiently good to fix the coordinates with reasonable certainty. Resolution on the other molecule, which with Fenimore's parameters is centred on ($0, \frac{1}{3}, \frac{1}{2}+0{\cdot}434$), was not quite so good. This was certainly due to some extent to the fact that the orientation is not exactly that proposed. All the sections for this molecule were consistent with it being rotated slightly in the (031) plane so that the x coordinates of atoms C_{11} and C_{12} are closer to 0·040 than to 0·048 and 0·058 respectively.

Fig. 1(b) shows the positions of some of the atoms derived from the corresponding sections. Regardless of imperfect resolution in specific instances, the results can be unambiguously interpreted.

The molecules are, within the imposed limits, identical