The Crystal Structure of 1, 2, 3-Tribromo-6-(*o*-methoxyphenyl)fulvene

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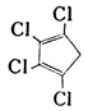
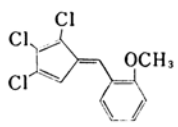
Sesquifulvalene (cycloheptatrienylidenecyclopentadiene), recently synthesized by Prinzbach and Rosswog,[1] is an interesting substance of non-alternant hydrocarbons. Kitahara, Murata, Funamizu and Asano[2] have attempted to obtain the tetrachloro derivative of this compound by the reaction of 1, 2, 3, 4-tetrachlorocyclopentadiene (I) and tropone (II) in a large volume of methanol.



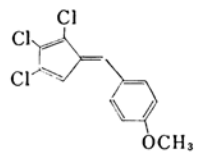
(I)

(II)

Contrary to their expectation, however, they got two kinds of products: red prisms (m. p. 117—118°C) and reddish-orange needles (m. p. 77—78°C). Elementary analysis indicated that both of these products had the molecular formula $C_{13}H_9OCl_3$. By some chemical consideration, and with the help of infrared, ultraviolet and NMR spectra, in was concluded that these isomers were trichloro-6-(*o*-methoxyphenyl)fulvene (III) and trichloro-6-(*p*-methoxyphenyl)fulvene (IV).



(III)



(IV)

Thus they have shown that the reaction of I and II in methanol results in an entirely new type of rearrangement reaction, in which the methoxy group enters the seven-membered ring (a six-membered ring after the rearrangement) and in which a chlorine atom in the five-membered ring is librated. The conformations of these molecules were also discussed on the basis of the NMR spectra.

The reaction between tropone and 1, 2, 3, 4-tetrabromocyclopentadiene followed an entirely analogous course; red plates (m. p. 128—129°C) and red prisms (m. p. 90—91°C) were obtained.[2] These compounds were supplied to us so that we could undertake an independent verification of the structures by means of X-ray.

The present paper will describe the structure determination of 1, 2, 3-tribromo-6-(*o*-methoxyphenyl)fulvene.

Experimental

Crystals of 1,2,3-tribromo-6-(*o*-methoxyphenyl)-fulvene (hereafter termed the "bromo derivative") and 1,2,3-trichloro-6-(*o*-methoxyphenyl)fulvene (hereafter the "chloro derivative") were supplied by Professor Kitahara of Tohoku University. Both of them are stable red laths elongated along the c axis.

From oscillation, Weissenberg and precession photographs of these crystals, the unit cell dimensions and space groups were determined. The crystal and physical data are summarized in Table I.

TABLE I. CRYSTAL AND PHYSICAL DATA

	Bromo derivative	Chloro derivative
M. p.	128—129°C	117—118°C
a	11.46 Å	11.65
b	14.53	14.61
c	7.85	7.64
β	96°35′	98°58′
Space group	$P2_1/c$	$P2_1/c$
Z	4	4
$\rho_{obs.}$	2.17 g.cm^{-3}	1.50
$\rho_{calcd.}$	2.15	1.49

Intensity data were collected only for the bromo derivative. Equi-inclination Weissenberg photographs around the c axis were taken with Cu$K\alpha$ radiation for l values from 0 to 6. The crystal used had a cross section of 0.1×0.2 mm. In order to determine the scale factors among the l layers, photographs for zero and first layers around the b axis were taken, also. Reflections from 2060 planes were observed out of 2900 possible (hkl)'s. In order to correlate the strong and the weak reflections, a multiple-film technique was applied, the relative intensities being ranged between 1 to 7000. The intensities were measured by a visual comparison with a standard intensity scale prepared with the same crystal. Lorentz and polarization corrections were made in the usual way. Since the main interest of this work was in determining the molecular conformation, the correction for absorption was omitted in spite of its large effect as a

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1) H. Prinzbach and W. Rosswog, *Angew. Chem.*, 73, 543 (1961).

2) Y. Kitahara, I. Murata, M. Funamizu and T. Asano, This Bulletin, 37, 1399 (1964).

result of the high bromine content. Tentative scale and temperature factors were estimated by Wilson statistics, the latter being $B=3.1$ Å^2.

Structure Determination

The three-dimensional Patterson function was synthesized on an electronic computer (IBM 7040) with a programme written by the authors. Figure 1 shows the Patterson section

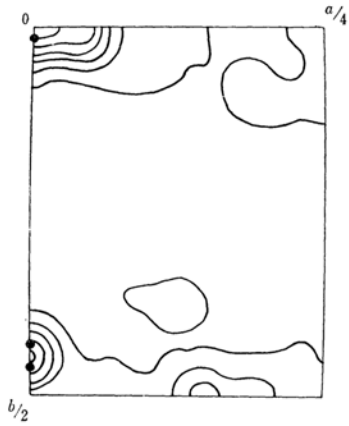

Fig. 1. Harker section $P(u,v,1/2)$. Contours are at equal intervals on an arbitrary scale. Black circles show Br-Br vectors from the final result.

$P(u, v, 1/2)$. There should appear on the $u=0$ line peaks due to vectors between atoms related to each other by the c glide operation. Only two significant peaks were observed on this line, however, at $v=0$ and at $v=0.45$. Since there should be three heavy atoms in an asymmetric unit, the peak at $v=0.45$ may be supposed to be a superimposed one due to two independent bromine atoms. Therefore, the y coordinate of one bromine atom was

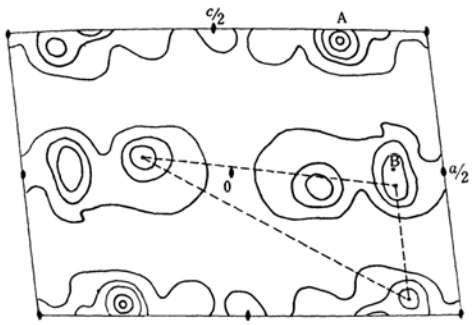

Fig. 2. Harker section $P(u,1/2,w)$. Contours are at equal intervals on an arbitrary scale. Three peaks linked by dashed lines correspond to vectors between the bromine atoms related by two-fold screw axis.

± 0.250, while those of the remaining two were ± 0.025.

The interpretation of the Harker section at $v=1/2$ was very ambiguous, because there appeared not only peaks between atoms related by a two-fold screw axis, but also peaks between independent atoms, the difference between whose y coordinates are, by accident, a half of the b axis. Figure 2 shows the Harker section $P(u, 1/2, w)$.

The first clue to the structure was obtained by examining the intramolecular heavy atom vectors in the three-dimensional Patterson function. Chemical information tells us that three bromine atoms attached to the fulvene ring form a two-lateral triangle whose sides are 3.6, 3.6 and 5.8 Å long. Figure 3 gives a

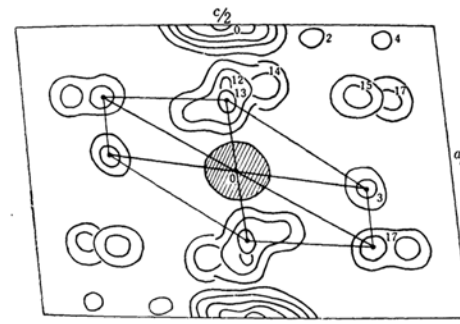

Fig. 3. Three-dimensional Patterson function. Composite diagram of sections parallel to (0 1 0). Height of section is added to each peak in unit of 1/60. Contours are at equal intervals on an arbitrary scale. Black circles show intramolecular Br-Br vector peaks.

composite three-dimensional Patterson function, showing peaks which lie within, say, 7 Å from the point of origin. Although some intermolecular vectors between the bromine atoms were observed at distances of about 4 Å from the point of origin, a constellation of Patterson peaks corresponding to the vector set of such a bromine triangle was clearly recognized, as is illustrated by the solid lines in Fig. 3. With the help of the orientation of the bromine triangle so found, the Harker section at $v=1/2$ was again examined and true Harker peaks were assigned, as is shown by the dash-line triangle in Fig. 2. The atomic coordinates of the three bromine atoms obtained were (0.605, -0.027, 0.225) for Br_1, (0.310, 0.027, 0.272) for Br_2 and (0.301, 0.250, 0.470) for Br_3. The non-Harker peak A in Fig. 2 was explained as a vector between the Br_1 of one molecule and the Br_2 of the molecule related by the c glide, and the B peak, as that between Br_3 atoms related to each other by the center of symmetry in different molecules.

Structure factors were computed with these

three bromine atoms with $B=3.1$ Å^2, the discrepancy factor, $R=\Sigma||F_o|-|F_c||/\Sigma|F_o|$, being 0.45. The first Fourier synthesis of the three-dimensional electron density distribution was carried out with signs based on the three bromine atoms. Significant peaks appearing in the Fourier map gave a reasonable molecular shape. Although some ghost peaks were observed in the map, it seemed quite certain that the molecular structure has been established by this stage. To be more precise, however, only three bromine atoms and five carbon atoms in the fulvene ring were introduced into the second calculation of the structure factors, and the R factor dropped to 0.36. The second Fourier synthesis looked very refined, and the coordinates of all the atoms could be read off from it. These coordinates of atoms gave reasonable bond lengths and angles. The third calculation of the structure factors resulted in an R factor of 0.30. At this stage, the scaling factors among the layers were corrected by the use of these calculated structure factors. All of these computations were executed on an IBM 7040 with programmes devised by the authors.

The atomic coordinates and temperature factors (anisotropic for the bromine atoms only) were refined by the successive block-diagonal matrix (9×9) least-squares method, the programmes for which were written by Tamaichi Ashida of this laboratory. Each cycle of the refinement on the IBM 7040 using all reflections took about 15 min. After four cycles, the R factor decreased to 0.126 (if non-observed reflections were omitted, the R factor was 0.113).

The final atomic coordinates and temperature factors of each atom are listed in Tables II and III respectively. The observed and calculated structure factors are shown in Table IV. Figure 4 gives the superimposed (0 0 1) sections of the final electron density distribution as taken through the atomic centers in the molecule.

TABLE II. THE FINAL ATOMIC COORDINATES

Atom	x/a	y/b	z/c
Br_1	0.6048	−0.0195	0.2265
Br_2	0.3089	0.0372	0.2778
Br_3	0.3051	0.2582	0.4716
C_1	0.5512	0.0874	0.3176
C_2	0.4434	0.1080	0.3340
C_3	0.4418	0.1975	0.4248
C_4	0.5492	0.2303	0.4586
C_5	0.6282	0.1630	0.3942
C_6	0.7470	0.1593	0.4034
C_7	0.8270	0.2338	0.4754
C_8	0.7992	0.3288	0.4509
C_9	0.8748	0.3942	0.5216
C_{10}	0.9858	0.3683	0.6090
C_{11}	1.0194	0.2752	0.6364
C_{12}	0.9346	0.2099	0.5596
C_{13}	1.0759	0.0862	0.6479
O	0.9584	0.1167	0.5790

Discussion

The very reasonable three-dimensional electron density distribution (shown in Fig. 4)

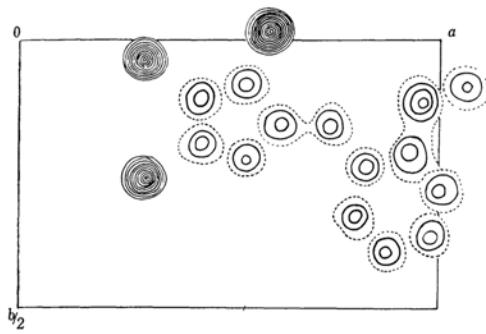

Fig. 4. The final electron density distribution. Composite diagram of sections parallel to (0 0 1). Contours are at intervals of 3 e.Å^{-3} with dashed contour of 2 e.Å^{-3}. For bromine, contours are at intervals of 6 e.Å^{-3}, beginning with 6 e.Å^{-3}.

TABLE III. THE FINAL TEMPERATURE FACTORS

For heavy atoms, temperature factors are in the form of
$\exp\{-(B_{11}h^2+B_{22}k^2+B_{33}l^2+B_{12}hk+B_{23}kl+B_{31}lh)\}$.

Atom	B_{11}	B_{22}	B_{33}	B_{12}	B_{23}	B_{31}
Br_1	0.00788	0.00346	0.01357	0.00180	−0.00473	−0.00209
Br_2	0.00627	0.00377	0.01458	−0.00255	−0.00137	−0.00375
Br_3	0.00515	0.00468	0.01179	0.00091	−0.00264	0.00172

Atom	B(Å^2)	Atom	B(Å^2)	Atom	B(Å^2)	Atom	B(Å^2)
C_1	2.40	C_5	2.09	C_9	4.10	C_{13}	4.19
C_2	2.74	C_6	2.61	C_{10}	3.75	O	3.08
C_3	2.17	C_7	2.40	C_{11}	3.69		
C_4	2.47	C_8	3.27	C_{12}	2.62		

TABLE IV. OBSERVED AND CALCULATED STRUCTURE FACTORS

h	k	l	$F_{obs.}$	$F_{calcd.}$	h	k	l	$F_{obs.}$	$F_{calcd.}$	h	k	l	$F_{obs.}$	$F_{calcd.}$	h	k	l	$F_{obs.}$	$F_{calcd.}$
2	0	0	11.11	−13.84	8	4	0	17.18	−17.01	4	9	0	19.59	−17.94	2	1	1	10.84	−11.73
3	0	0	46.60	60.38	9	4	0	8.45	− 7.70	5	9	0	8.97	8.36	2	1	−1	5.35	6.20
4	0	0	7.69	− 7.48	10	4	0	22.22	21.45	6	9	0	10.62	10.02	3	1	1	31.06	32.55
5	0	0	14.68	−14.55	11	4	0	11.47	−11.44	7	9	0	2.14	− 5.13	3	1	−1	36.25	−41.93
6	0	0	5.77	5.33	12	4	0	2.69	2.84	8	9	0	9.16	− 9.38	4	1	1	37.97	−38.81
7	0	0	24.41	24.87	13	4	0	11.51	11.91	9	9	0	19.20	16.56	4	1	−1	26.61	28.08
8	0	0	27.56	−27.26	14	4	0	3.81	− 4.77	10	9	0	4.76	− 4.35	5	1	1	10.01	8.49
9	0	0	7.69	− 5.46	1	5	0	55.18	−59.65	11	9	0	9.34	−10.44	5	1	−1	4.23	3.59
10	0	0	22.10	21.52	2	5	0	42.02	41.27	12	9	0	9.58	12.09	6	1	1	32.02	27.78
11	0	0	14.50	−12.03	3	5	0	11.63	10.81	0	10	0	13.52	−12.82	6	1	−1	25.63	−24.35
12	0	0	9.40	8.72	4	5	0	41.93	−40.44	1	10	0	4.61	3.80	7	1	1	30.24	−29.27
13	0	0	13.40	15.26	5	5	0	6.44	6.48	2	10	0	26.43	23.97	7	1	−1	33.45	30.76
14	0	0	5.25	− 6.17	6	5	0	12.67	12.90	3	10	0	16.91	−16.42	8	1	1	14.82	12.74
1	1	0	18.01	−19.64	7	5	0	12.63	−11.22	4	10	0	9.06	− 8.79	8	1	−1	1.56	− 1.22
2	1	0	28.99	38.08	8	5	0	14.71	−14.63	5	10	0	20.26	18.79	9	1	1	9.88	7.88
3	1	0	13.24	15.81	9	5	0	20.72	19.92	6	10	0	13.86	−11.92	9	1	−1	9.36	− 7.78
4	1	0	28.32	−30.78	10	5	0	8.06	− 7.04	7	10	0	9.64	− 9.19	10	1	1	6.09	− 5.18
5	1	0	6.74	5.87	11	5	0	16.48	−15.52	8	10	0	10.41	9.97	10	1	−1	15.34	12.48
6	1	0	26.18	25.66	12	5	0	11.96	13.20	10	10	0	8.55	− 9.52	13	1	1	2.09	2.14
7	1	0	12.51	−12.52	13	5	0	2.14	− 1.35	11	10	0	4.36	4.20	13	1	−1	3.13	− 3.32
8	1	0	8.09	7.37	0	6	0	8.48	− 7.11	12	10	0	2.23	2.08	14	1	1	2.94	− 3.69
9	1	0	19.96	18.84	1	6	0	14.31	−11.46	1	11	0	7.51	− 8.21	14	1	−1	2.96	3.61
10	1	0	8.03	− 7.33	2	6	0	4.79	5.53	2	11	0	19.71	16.89	0	2	1	20.03	−17.32
11	1	0	10.01	− 9.19	4	6	0	16.97	−16.05	3	11	0	12.42	−12.01	1	2	1	14.10	10.99
12	1	0	9.77	8.52	5	6	0	27.65	29.27	4	11	0	13.67	14.49	1	2	−1	15.37	−14.10
14	1	0	5.83	− 6.70	6	6	0	11.90	−11.56	5	11	0	3.02	2.63	2	2	1	15.42	13.49
0	2	0	4.61	3.92	7	6	0	12.79	−12.02	6	11	0	8.64	− 7.81	2	2	−1	1.62	− 0.28
1	2	0	26.40	−26.26	8	6	0	17.94	17.88	7	11	0	8.51	8.28	3	2	1	5.21	− 5.97
2	2	0	8.88	7.73	9	6	0	5.28	− 5.69	1	13	0	10.99	−11.25	3	2	−1	8.04	6.66
3	2	0	11.05	10.50	12	6	0	3.54	3.65	2	13	0	12.94	13.53	4	2	1	16.76	−15.42
4	2	0	20.51	−27.33	13	6	0	1.98	− 3.06	3	13	0	8.06	− 6.95	4	2	−1	1.54	1.39
5	2	0	15.78	16.74	1	7	0	9.52	10.33	4	13	0	6.78	− 5.53	5	2	1	5.19	6.71
6	2	0	13.61	−12.62	2	7	0	15.59	13.61	8	13	0	5.52	− 5.41	5	2	−1	12.95	−13.48
7	2	0	4.97	− 3.10	3	7	0	20.63	−19.04	9	13	0	5.22	6.23	7	2	1	11.19	− 8.85
8	2	0	11.57	10.27	4	7	0	6.32	6.05	0	14	0	14.53	−16.02	7	2	−1	9.36	7.96
9	2	0	8.79	− 6.55	5	7	0	3.60	2.45	1	14	0	6.74	6.87	8	2	1	1.65	2.91
10	2	0	6.07	6.19	6	7	0	5.43	− 3.95	2	14	0	9.03	10.23	8	2	−1	8.40	− 7.90
11	2	0	7.97	− 6.34	7	7	0	2.90	2.64	3	14	0	8.91	− 9.53	9	2	1	10.95	− 8.71
14	2	0	3.57	− 3.91	8	7	0	7.35	− 6.68	5	14	0	8.88	9.28	9	2	−1	5.21	3.78
1	3	0	2.26	0.91	9	7	0	3.05	1.52	6	14	0	4.61	− 4.64	10	2	1	1.56	2.20
2	3	0	3.23	2.83	10	7	0	4.21	− 2.71	7	14	0	5.46	− 6.82	10	2	−1	2.72	1.66
3	3	0	14.47	−13.43	11	7	0	2.75	− 1.45	8	14	0	7.42	8.59	11	2	1	0.	− 0.39
4	3	0	17.43	16.66	12	7	0	3.33	3.42	3	15	0	9.25	− 9.68	11	2	−1	2.72	2.15
6	3	0	10.53	− 9.54	13	7	0	3.08	− 3.80	4	15	0	5.37	6.73	12	2	1	2.44	3.06
7	3	0	9.49	9.42	0	8	0	27.89	25.76	6	15	0	9.95	−11.04	12	2	−1	0.	− 0.43
10	3	0	7.48	5.80	1	8	0	5.58	− 6.39	7	15	0	5.86	7.29	13	2	1	2.03	− 3.02
11	3	0	5.98	4.96	3	8	0	17.52	15.09	0	16	0	5.92	− 6.80	13	2	−1	0.	1.71
14	3	0	2.41	2.79	4	8	0	6.74	− 4.79	1	16	0	3.72	3.67	0	3	1	8.42	7.01
0	4	0	51.48	54.44	6	8	0	4.03	3.41	1	17	0	3.20	− 3.84	1	3	1	17.31	−14.94
1	4	0	39.25	−39.59	7	8	0	7.26	6.45	2	17	0	3.11	4.15	1	3	−1	2.85	− 1.92
2	4	0	31.04	−30.11	9	8	0	3.02	− 3.89	3	17	0	3.63	− 5.10	2	3	1	4.42	3.93
3	4	0	36.41	37.69	10	8	0	7.63	6.66	4	17	0	2.72	3.43	2	3	−1	20.52	18.21
4	4	0	14.07	−13.86	11	8	0	4.49	− 4.38	0	18	0	3.97	− 5.08	3	3	1	31.66	30.30
5	4	0	14.80	−14.39	13	8	0	2.53	3.82	1	18	0	3.02	4.48	3	3	−1	36.79	−36.40
6	4	0	10.93	8.62	1	9	0	34.97	−33.23	1	1	1	2.28	− 2.44	4	3	1	30.76	−33.24
7	4	0	15.87	15.43	2	9	0	34.82	32.30	1	1	−1	9.52	9.08	4	3	−1	25.05	24.93

TABLE IV. (Continued)

h	*k*	*l*	$F_{obs.}$	$F_{calcd.}$	*h*	*k*	*l*	$F_{obs.}$	$F_{calcd.}$	*h*	*k*	*l*	$F_{obs.}$	$F_{calcd.}$	*h*	*k*	*l*	$F_{obs.}$	$F_{calcd.}$
5	3	1	13.58	11.88	4	5	1	13.77	−12.49	10	7	−1	0.	0.07	4	10	1	14.21	−13.25
5	3	−1	0.	1.76	4	5	−1	19.65	17.62	11	7	1	8.23	7.62	4	10	−1	9.25	8.04
6	3	1	12.68	12.19	5	5	1	0.	− 0.59	11	7	−1	2.47	− 1.04	5	10	1	14.10	14.56
6	3	−1	34.05	−32.31	5	5	−1	4.53	2.98	0	8	1	2.03	− 2.50	5	10	−1	17.12	−17.48
7	3	1	25.82	−23.88	6	5	1	15.78	14.18	1	8	1	2.06	− 1.85	6	10	1	13.03	−12.53
7	3	−1	19.43	17.40	6	5	−1	13.09	−12.32	1	8	−1	21.21	17.31	6	10	−1	12.70	12.69
8	3	1	13.36	11.22	7	5	1	17.40	−16.16	2	8	1	21.84	18.88	7	10	1	6.67	− 6.56
8	3	−1	6.53	− 4.78	7	5	−1	17.31	15.49	2	8	−1	24.12	−21.70	7	10	−1	4.72	− 3.91
9	3	1	0.	0.25	8	5	1	7.87	6.43	3	8	1	20.39	−18.67	8	10	1	7.02	6.70
9	3	−1	9.49	− 7.65	8	5	−1	13.12	−11.31	3	8	−1	10.21	9.23	8	10	−1	10.07	−10.43
10	3	1	13.66	−11.58	9	5	1	0.	0.19	4	8	1	1.65	− 2.23	9	10	1	4.31	− 4.19
10	3	−1	0.	− 0.95	9	5	−1	2.69	− 3.36	4	8	−1	21.07	18.73	9	10	−1	12.02	12.44
11	3	1	2.63	3.71	10	5	1	0.	− 1.07	5	8	1	28.81	27.30	0	11	1	2.55	− 2.28
11	3	−1	0.	1.21	10	5	−1	9.82	8.16	5	8	−1	27.36	−26.13	1	11	1	12.02	11.70
12	3	1	1.65	0.84	11	5	1	2.55	1.25	6	8	1	19.81	−18.69	1	11	−1	11.44	− 9.94
12	3	−1	1.78	− 0.04	11	5	−1	6.94	− 6.20	6	8	−1	11.06	9.85	2	11	1	7.79	− 7.48
13	3	1	2.80	0.65	13	5	1	2.52	1.91	7	8	1	11.69	11.03	2	11	−1	9.30	8.57
13	3	−1	0.	− 1.88	13	5	−1	0.	− 0.60	7	8	−1	13.25	12.59	3	11	1	7.46	− 7.11
14	3	1	2.61	− 3.99	0	6	1	6.06	− 4.89	8	8	1	13.66	12.42	3	11	−1	6.94	− 6.22
14	3	−1	2.77	3.20	1	6	1	14.82	−12.41	8	8	−1	11.55	− 9.79	4	11	1	7.11	6.53
0	4	1	6.72	− 5.40	1	6	−1	2.39	0.92	9	8	1	10.40	−10.36	4	11	−1	11.66	−11.15
1	4	1	16.57	14.21	2	6	1	18.82	16.81	9	8	−1	0.	− 1.49	5	11	1	1.92	2.19
1	4	−1	13.01	10.63	2	6	−1	21.37	−18.14	10	8	1	0.	0.29	5	11	−1	6.06	5.77
2	4	1	23.82	18.59	3	6	1	17.81	−15.77	10	8	−1	2.61	3.52	6	11	1	5.46	− 5.17
2	4	−1	24.06	−21.12	3	6	−1	10.67	9.44	11	8	1	5.49	5.53	6	11	−1	0.	1.48
3	4	1	11.22	−10.24	4	6	1	16.19	−13.67	11	8	−1	0.	− 0.93	7	11	1	0.	0.61
3	4	−1	9.27	7.72	4	6	−1	13.06	11.58	0	9	1	6.28	5.76	7	11	−1	5.41	− 5.30
4	4	1	4.39	3.26	5	6	1	28.29	27.21	1	9	1	9.25	7.50	8	11	1	3.59	3.71
4	4	−1	26.31	24.71	5	6	−1	25.22	−23.02	1	9	−1	19.56	−16.46	8	11	−1	0.	− 0.42
5	4	1	24.39	23.56	6	6	1	13.86	−13.79	2	9	1	20.88	−19.89	9	11	1	5.19	− 5.76
5	4	−1	28.86	−27.19	6	6	−1	20.33	18.51	2	9	−1	2.28	2.85	9	11	−1	1.12	1.81
6	4	1	20.63	−18.79	7	6	1	6.72	− 5.82	3	9	1	10.87	9.55	11	11	1	4.12	4.76
6	4	−1	10.37	9.55	7	6	−1	4.97	3.40	3	9	−1	13.03	11.76	11	11	−1	0.	− 2.11
7	4	1	5.35	− 4.20	8	6	1	13.88	12.83	4	9	1	2.47	2.58	0	12	1	8.01	7.55
7	4	−1	9.85	8.81	8	6	−1	15.37	−14.09	4	9	−1	7.68	− 6.29	1	12	1	4.64	4.14
8	4	1	18.36	16.21	9	6	1	6.12	− 4.39	5	9	1	11.19	−10.84	1	12	−1	13.09	12.61
8	4	−1	14.87	−13.86	9	6	−1	9.03	8.05	5	9	−1	0.	− 1.18	2	12	1	3.81	3.12
9	4	1	8.94	− 7.94	11	6	1	3.51	1.98	6	9	1	0.	− 0.99	2	12	−1	9.30	− 8.26
9	4	−1	5.32	4.40	11	6	−1	4.44	− 3.81	6	9	−1	6.97	6.29	3	12	1	0.	− 0.83
10	4	1	4.72	− 4.00	1	7	1	10.32	9.08	7	9	1	3.84	4.12	3	12	−1	2.72	− 3.82
10	4	−1	2.74	2.24	1	7	−1	11.08	− 8.76	7	9	−1	3.84	4.16	4	12	1	3.87	− 4.15
11	4	1	4.53	3.71	2	7	1	2.77	− 2.98	8	9	1	7.68	6.83	4	12	−1	10.21	9.44
11	4	−1	3.76	− 2.48	2	7	−1	24.28	22.49	8	9	−1	0.	− 1.10	5	12	1	11.22	12.35
12	4	1	0.	− 0.06	3	7	1	2.06	− 0.44	9	9	1	5.84	− 6.71	5	12	−1	13.36	−13.49
12	4	−1	6.86	− 5.66	3	7	−1	19.95	−18.03	9	9	−1	7.52	6.57	6	12	1	13.33	−14.24
13	4	1	1.89	− 3.00	4	7	1	6.53	− 5.98	10	9	1	4.14	4.20	6	12	−1	0.	0.98
13	4	−1	3.57	3.55	4	7	−1	2.14	0.79	10	9	−1	6.09	5.56	7	12	1	4.42	5.12
14	4	1	1.95	1.61	5	7	1	5.21	5.41	11	9	1	3.57	2.84	7	12	−1	5.21	6.02
14	4	−1	2.09	1.99	5	7	−1	8.51	7.97	11	9	−1	7.57	− 8.04	8	12	1	5.82	6.76
0	5	1	5.41	3.58	6	7	1	2.47	3.42	0	10	1	5.35	4.54	8	12	−1	7.33	− 7.86
1	5	1	2.17	2.75	6	7	−1	13.97	−12.68	1	10	1	18.05	−17.15	9	12	1	9.38	−10.19
1	5	−1	6.06	− 4.86	7	7	1	11.72	−10.71	1	10	−1	2.41	2.46	9	12	−1	3.10	4.42
2	5	1	19.51	−17.01	7	7	−1	8.15	7.71	2	10	1	10.43	8.97	10	12	1	5.13	4.79
2	5	−1	18.49	14.72	8	7	1	8.12	7.12	2	10	−1	8.45	− 7.71	10	12	−1	0.	− 1.91
3	5	1	25.49	24.00	8	7	−1	8.92	− 8.21	3	10	1	4.36	− 4.28	0	13	1	6.09	5.69
3	5	−1	9.66	− 7.06	10	7	1	5.84	− 4.82	3	10	−1	0.	1.25	1	13	1	10.56	10.05

TABLE IV. (Continued)

h	k	l	$F_{obs.}$	$F_{calcd.}$	h	k	l	$F_{obs.}$	$F_{calcd.}$	h	k	l	$F_{obs.}$	$F_{calcd.}$	k	h	l	$F_{obs.}$	$F_{calcd.}$
1	13	−1	12.81	−13.62	1	16	−1	7.00	8.21	3	1	−2	17.80	19.77	3	3	2	11.65	−11.09
2	13	1	10.56	−10.31	2	16	1	2.28	− 2.59	4	1	2	19.17	−19.77	3	3	−2	6.06	− 6.15
2	13	−1	5.46	5.59	2	16	−1	1.78	− 1.07	4	1	−2	15.90	−17.31	4	3	2	32.58	31.66
3	13	1	4.72	4.85	3	16	1	2.17	3.14	5	1	2	2.85	2.98	4	3	−2	31.68	35.12
3	13	−1	10.56	10.57	3	16	−1	4.97	− 5.72	5	1	−2	11.62	10.67	5	3	2	0.	− 2.02
4	13	1	10.10	10.42	4	16	1	1.43	− 1.77	6	1	2	15.75	15.71	5	3	−2	12.69	−11.81
4	13	−1	7.16	− 7.00	4	16	−1	5.57	6.84	6	1	−2	18.49	16.95	6	3	2	19.77	−20.18
5	13	1	4.58	− 5.33	6	16	1	2.80	− 4.70	7	1	2	0.	− 1.36	6	3	−2	16.08	−16.59
5	13	−1	3.76	− 3.85	6	16	−1	0.	0.26	7	1	−2	15.60	−13.93	7	3	2	11.86	10.20
6	13	1	4.39	− 4.32	7	16	1	3.43	5.00	8	1	2	9.84	− 8.17	7	3	−2	11.18	10.49
6	13	−1	8.53	9.05	7	16	−1	1.40	2.11	8	1	−2	5.80	4.24	8	3	2	13.32	12.85
7	13	1	8.86	9.30	0	17	1	2.83	3.83	9	1	2	10.37	9.87	8	3	−2	8.80	8.14
7	13	−1	4.12	− 3.81	3	17	1	1.26	− 1.06	9	1	−2	9.24	7.38	9	3	2	15.60	−16.47
8	13	1	4.25	− 4.68	3	17	−1	6.67	7.68	10	1	2	0.	− 1.58	9	3	−2	19.97	−18.94
8	13	−1	3.84	− 4.94	4	17	1	4.91	5.81	10	1	−2	8.71	− 5.84	10	3	2	6.48	6.12
9	13	1	3.59	− 3.71	4	17	−1	6.15	− 4.92	11	1	2	3.92	− 4.03	10	3	−2	9.21	8.98
9	13	−1	3.84	4.66	5	17	1	4.55	− 6.14	11	1	−2	5.77	− 5.99	11	3	2	11.59	12.28
10	13	1	3.37	4.34	5	17	−1	0.82	− 0.73	12	1	2	0.	1.20	11	3	−2	11.44	10.93
10	13	−1	1.10	2.22	0	18	1	5.93	8.45	12	1	−2	6.63	7.47	12	3	2	8.32	−10.14
0	14	1	8.53	8.88	2	18	1	0.	− 0.13	13	1	2	2.82	4.17	12	3	−2	12.10	−11.77
1	14	1	12.35	−13.48	2	18	−1	3.51	5.21	13	1	−2	0.	− 1.25	14	3	−2	4.07	5.56
1	14	−1	0.	− 0.75	3	18	1	4.39	5.02	14	1	2	2.08	− 4.62	0	4	2	12.22	−10.07
2	14	1	5.98	6.52	3	18	−1	3.13	− 4.66	14	1	−2	1.19	− 3.77	1	4	2	28.15	24.12
2	14	−1	6.01	6.70	1	0	2	38.04	46.58	0	2	2	86.40	−90.78	1	4	−2	15.75	−13.00
3	14	1	4.58	4.36	1	0	−2	20.72	16.40	1	2	2	45.83	47.48	2	4	2	28.74	−26.67
3	14	−1	3.76	− 4.06	2	0	2	25.35	−25.30	1	2	−2	31.33	33.26	2	4	−2	9.12	7.53
4	14	1	5.76	− 5.86	2	0	−2	7.19	8.67	2	2	2	18.78	18.43	3	4	2	13.02	−11.01
4	14	−1	4.50	5.40	3	0	2	10.43	−10.35	2	2	−2	32.37	34.47	3	4	−2	0.	− 1.21
5	14	1	4.28	4.22	3	0	−2	16.08	−19.21	3	2	2	34.00	−36.60	4	4	2	25.89	25.17
5	14	−1	3.54	− 4.03	4	0	2	34.57	35.06	3	2	−2	36.83	−43.94	4	4	−2	10.82	9.72
6	14	1	0.	− 0.34	4	0	−2	4.79	3.98	4	2	2	5.80	6.16	5	4	2	26.78	−26.63
6	14	−1	7.90	8.04	5	0	2	16.70	−16.62	4	2	−2	16.41	17.38	5	4	−2	15.16	−15.97
7	14	1	0.	0.06	5	0	−2	15.96	−17.57	5	2	2	9.48	11.38	6	4	2	4.22	3.93
7	14	−1	3.79	− 4.60	6	0	2	0.	1.76	5	2	−2	18.70	18.52	6	4	−2	15.57	14.98
8	14	1	0.	− 0.58	6	0	−2	19.41	19.55	6	2	2	0.	0.95	7	4	2	3.75	2.79
8	14	−1	3.84	− 2.67	7	0	2	9.01	7.93	6	2	−2	13.23	−13.32	7	4	−2	9.72	9.18
9	14	1	2.39	− 3.31	7	0	−2	7.40	− 7.60	7	2	2	22.86	−20.77	8	4	2	12.28	−11.55
9	14	−1	5.38	6.09	8	0	2	0.	1.09	7	2	−2	13.70	−13.76	8	4	−2	16.47	−15.36
1	15	1	2.55	2.81	8	0	−2	18.22	−17.23	8	2	2	25.17	24.16	9	4	2	2.91	− 2.67
1	15	−1	2.52	− 3.51	9	0	2	4.96	− 4.48	8	2	−2	21.07	18.86	9	4	−2	15.13	14.12
2	15	1	0.	0.40	9	0	−2	16.67	14.84	9	2	2	9.99	8.40	10	4	2	4.99	− 4.79
2	15	−1	2.55	3.15	10	0	2	1.55	− 2.32	9	2	−2	4.84	3.64	10	4	−2	0.	− 0.31
3	15	1	6.04	− 6.46	10	0	−2	11.59	−10.97	10	2	2	19.71	−20.15	11	4	2	6.57	7.35
3	15	−1	2.50	2.60	11	0	2	10.05	10.14	10	2	−2	21.94	−20.24	11	4	−2	0.	1.04
4	15	1	5.79	6.33	11	0	−2	0.	− 1.30	11	2	2	12.36	13.64	12	4	2	6.60	− 7.44
4	15	−1	5.38	− 7.06	12	0	2	7.04	− 8.46	11	2	−2	9.96	8.82	12	4	−2	4.49	3.71
5	15	1	0.	0.77	12	0	−2	2.68	2.97	12	2	2	4.25	4.65	0	5	2	15.04	13.56
5	15	−1	4.55	5.53	13	0	2	2.85	− 3.14	12	2	−2	4.64	5.11	1	5	2	0.	0.22
6	15	1	4.06	− 3.90	13	0	−2	0.	− 0.77	13	2	2	6.81	−10.25	1	5	−2	15.04	11.63
6	15	−1	4.20	2.70	14	0	2	2.38	5.85	13	2	−2	14.98	−15.23	2	5	2	17.63	−16.20
7	15	1	3.95	4.39	14	0	−2	0.	1.00	14	2	−2	4.28	5.53	2	5	−2	6.96	− 6.36
7	15	−1	4.94	− 6.26	1	1	2	34.54	−34.45	0	3	2	5.74	− 4.84	3	5	2	25.59	24.32
8	15	1	0.	1.57	1	1	−2	26.10	26.07	1	3	2	34.57	33.60	3	5	−2	4.93	2.89
8	15	−1	4.55	3.49	2	1	2	1.81	− 0.88	1	3	−2	48.09	50.27	4	5	2	7.10	− 5.95
0	16	1	7.41	9.27	2	1	−2	32.93	39.53	2	3	2	30.02	−28.95	4	5	−2	11.06	− 9.53
1	16	1	2.33	− 2.78	3	1	2	14.03	13.51	2	3	−2	32.55	−33.80	5	5	2	13.17	−11.96

TABLE IV. (Continued)

h	*k*	*l*	$F_{obs.}$	$F_{calcd.}$	*h*	*k*	*l*	$F_{obs.}$	$F_{calcd.}$	*h*	*k*	*l*	$F_{obs.}$	$F_{calcd.}$	*h*	*k*	*l*	$F_{obs.}$	$F_{calcd.}$
5	5	−2	6.21	6.13	8	7	−2	7.61	6.62	12	9	−2	0.	− 1.55	8	12	−2	8.50	− 9.95
6	5	2	11.03	10.17	9	7	2	20.09	−20.89	0	10	2	6.93	− 5.95	9	12	2	0.	0.36
6	5	−2	0.	− 0.44	9	7	−2	16.97	−17.12	1	10	2	4.58	4.87	9	12	−2	2.29	0.98
7	5	2	3.83	3.67	10	7	2	4.67	5.67	1	10	−2	8.71	7.13	10	12	2	5.68	7.96
7	5	−2	14.65	−14.12	10	7	−2	2.82	2.91	3	10	2	3.89	4.92	10	12	−2	9.96	7.91
8	5	2	5.71	− 4.62	11	7	2	7.91	9.66	3	10	−2	9.75	− 9.26	11	12	−2	3.69	− 5.59
8	5	−2	11.00	12.01	11	7	−2	12.25	12.85	4	10	2	5.65	− 5.27	0	13	2	8.74	9.08
10	5	2	5.68	5.53	12	7	2	9.99	−12.15	4	10	−2	10.02	9.47	1	13	2	0.	− 0.83
10	5	−2	8.20	− 7.30	12	7	−2	11.29	−12.40	5	10	2	0.	− 1.46	1	13	−2	5.05	4.74
11	5	2	3.69	− 4.09	0	8	2	13.29	11.92	5	10	−2	5.71	− 4.95	2	13	2	5.02	− 5.59
11	5	−2	2.79	3.20	1	8	2	12.48	10.90	6	10	2	5.05	6.42	2	13	−2	5.05	− 5.47
12	5	2	3.86	− 4.78	1	8	−2	5.97	− 4.19	6	10	−2	5.80	− 5.92	3	13	2	12.57	14.37
12	5	−2	0.	0.30	2	8	2	21.40	−19.50	7	10	2	4.07	− 4.67	3	13	−2	5.80	6.16
13	5	2	4.34	6.62	2	8	−2	11.95	−10.32	7	10	−2	5.83	3.72	4	13	2	8.03	−10.33
13	5	−2	0.	0.18	3	8	2	14.68	13.59	9	10	2	5.08	6.12	4	13	−2	8.62	− 8.86
0	6	2	36.53	−34.83	3	8	−2	10.08	8.09	9	10	−2	1.78	− 2.23	5	13	2	0.	− 0.88
1	6	2	13.82	10.99	4	8	2	14.98	13.69	10	10	2	4.43	− 2.92	5	13	−2	4.87	− 4.83
1	6	−2	24.02	21.24	4	8	−2	7.85	6.41	10	10	−2	0.	− 0.91	6	13	2	7.82	8.95
2	6	2	12.57	11.63	5	8	2	21.07	−21.61	11	10	2	3.00	− 3.29	6	13	−2	7.67	8.85
2	6	−2	14.15	11.91	5	8	−2	23.51	−22.15	11	10	−2	0.	1.67	7	13	2	5.38	− 6.75
3	6	2	11.98	−10.18	6	8	2	13.23	12.91	1	11	2	14.86	13.87	7	13	−2	7.64	− 8.78
3	6	−2	34.36	−33.53	6	8	−2	9.10	8.59	1	11	−2	29.72	25.96	0	14	2	2.85	3.44
4	6	2	5.08	4.59	7	8	2	10.85	9.93	2	11	2	16.91	−16.83	1	14	2	6.36	− 6.72
4	6	−2	19.38	17.19	7	8	−2	10.67	9.69	2	11	−2	22.02	−21.02	1	14	−2	1.55	0.13
5	6	2	8.47	7.79	8	8	2	8.20	− 9.51	3	11	2	4.96	4.72	2	14	2	3.98	2.30
5	6	−2	6.60	5.64	8	8	−2	16.20	−15.13	3	11	−2	9.81	7.97	2	14	−2	0.	− 2.14
6	6	2	0.	− 0.90	10	8	2	5.17	5.26	4	11	2	6.48	6.37	3	14	2	3.89	4.59
6	6	−2	9.72	− 7.79	10	8	−2	3.89	4.47	4	11	−2	14.39	14.11	3	14	−2	0.	1.29
7	6	2	10.05	−10.38	11	8	2	0.	− 0.06	5	11	2	0.	− 1.69	4	14	2	3.77	− 4.33
7	6	−2	8.89	− 7.55	11	8	−2	5.53	− 6.03	5	11	−2	7.13	− 5.42	4	14	−2	0.	2.42
8	6	2	5.02	6.33	12	8	2	3.36	− 4.70	6	11	2	4.07	− 4.15	6	14	2	3.33	4.00
8	6	−2	8.00	7.03	12	8	−2	0.	− 1.66	6	11	−2	0.	− 1.70	6	14	−2	3.51	− 4.40
9	6	2	5.02	6.00	0	9	2	11.95	10.94	8	11	2	4.52	6.04	7	14	2	1.49	− 2.04
9	6	−2	5.05	5.19	1	9	2	5.35	5.43	8	11	−2	6.72	8.19	7	14	−2	3.21	3.93
10	6	2	10.05	−11.65	1	9	−2	14.45	13.03	9	11	2	5.74	− 7.33	8	14	2	2.44	− 3.67
10	6	−2	11.89	−11.71	2	9	2	25.00	−24.31	9	11	−2	8.77	−10.45	8	14	−2	0.	− 1.37
11	6	2	5.02	6.41	2	9	−2	7.85	− 6.44	10	11	2	2.76	3.96	1	15	2	0.	3.18
11	6	−2	9.01	9.18	3	9	2	22.29	21.70	10	11	−2	1.19	1.94	1	15	−2	6.57	7.26
13	6	2	2.82	− 4.94	3	9	−2	6.24	5.56	11	11	2	3.12	5.19	2	15	2	5.23	− 5.82
13	6	−2	6.84	− 8.37	4	9	2	12.10	−12.12	11	11	−2	5.62	6.73	2	15	−2	8.38	− 8.81
0	7	2	12.99	11.57	4	9	−2	5.91	− 5.59	0	12	2	14.39	14.63	3	15	2	3.60	4.77
1	7	2	40.81	41.85	5	9	2	10.14	− 8.24	1	12	2	10.40	− 9.81	3	15	−2	3.66	4.91
1	7	−2	36.47	34.55	5	9	−2	4.76	5.25	1	12	−2	1.55	− 1.95	6	15	2	0.	2.04
2	7	2	37.21	−35.54	6	9	2	6.45	6.30	2	12	2	15.34	−15.77	6	15	−2	3.77	4.29
2	7	−2	39.38	−37.87	6	9	−2	6.33	6.10	2	12	−2	15.31	−14.62	7	15	2	3.33	− 4.57
3	7	2	0.	− 0.71	7	9	2	4.13	− 3.62	3	12	2	9.21	9.83	7	15	−2	1.34	− 2.86
3	7	−2	17.89	−14.99	7	9	−2	7.10	− 5.54	3	12	−2	13.94	13.30	0	16	2	8.32	9.98
4	7	2	21.10	21.15	8	9	2	0.	0.69	4	12	2	0.	1.58	1	16	2	3.36	− 4.18
4	7	−2	25.77	24.62	8	9	−2	7.70	7.01	4	12	−2	4.13	4.37	1	16	−2	4.16	− 5.87
5	7	2	5.14	− 3.60	9	9	2	3.80	− 4.77	5	12	2	14.92	−15.77	2	16	2	2.32	− 3.80
5	7	−2	13.08	−12.26	9	9	−2	3.98	2.97	5	12	−2	8.20	− 8.81	2	16	−2	3.33	− 3.32
6	7	2	12.19	−11.87	10	9	2	4.84	5.97	6	12	2	6.24	7.24	3	16	2	6.63	8.03
6	7	−2	10.49	−10.28	10	9	−2	4.55	− 4.22	6	12	−2	6.39	7.12	3	16	−2	4.58	5.54
7	7	2	4.93	5.31	11	9	2	0.	− 2.41	7	12	2	3.72	5.18	4	16	2	0.	− 1.70
7	7	−2	10.37	9.33	11	9	−2	3.98	3.60	7	12	−2	9.10	9.81	4	16	−2	1.49	0.11
8	7	2	10.49	12.09	12	9	2	1.87	− 2.73	8	12	2	7.19	− 8.34	6	16	2	2.08	2.97

TABLE IV. (Continued)

h	k	l	$F_{obs.}$	$F_{calcd.}$	h	k	l	$F_{obs.}$	$F_{calcd.}$	h	k	l	$F_{obs.}$	$F_{calcd.}$	h	k	l	$F_{obs.}$	$F_{calcd.}$
6	16	−2	1.19	0.83	8	2	−3	10.05	10.87	3	5	3	9.81	− 9.50	7	7	−3	5.49	− 4.49
1	17	2	3.45	− 4.44	9	2	3	4.00	4.73	3	5	−3	23.48	24.64	8	7	3	4.86	− 4.70
1	17	−2	2.02	− 3.41	9	2	−3	3.91	4.23	4	5	3	21.05	21.12	8	7	−3	5.63	6.00
2	17	2	0.	0.93	11	2	3	2.55	− 3.41	4	5	−3	7.92	− 7.48	9	7	3	6.05	6.54
2	17	−2	3.39	− 2.06	11	2	−3	4.77	4.10	5	5	3	11.53	−11.40	9	7	−3	2.82	− 3.18
3	17	2	3.92	5.65	12	2	3	0.	− 0.35	5	5	−3	8.30	− 8.56	10	7	3	1.78	− 2.72
3	17	−2	3.72	5.67	12	2	−3	4.39	3.58	6	5	3	1.33	− 1.97	10	7	−3	4.68	− 5.37
4	17	2	4.84	− 7.30	0	3	3	4.86	− 4.67	6	5	−3	19.24	19.92	11	7	3	2.08	− 3.05
4	17	−2	3.39	− 4.24	1	3	3	9.84	7.07	7	5	3	12.54	13.46	11	7	−3	7.38	7.19
5	17	2	1.87	0.17	1	3	−3	12.84	9.99	7	5	−3	14.14	−13.98	12	7	3	3.94	4.87
5	17	−2	0.	− 2.31	2	3	3	11.06	10.49	8	5	3	8.48	− 8.16	12	7	−3	1.04	− 2.45
0	18	2	2.88	4.04	2	3	−3	15.98	−15.90	8	5	−3	0.	0.65	0	8	3	5.04	− 5.87
1	18	2	2.73	− 4.02	3	3	3	11.27	−12.61	9	5	3	0.	1.44	1	8	3	7.74	7.54
1	18	−2	0.	− 0.06	3	3	−3	18.15	19.83	9	5	−3	2.82	2.10	1	8	−3	0.	− 1.61
1	1	3	9.28	7.99	4	3	3	26.00	26.37	10	5	3	5.93	6.18	2	8	3	11.27	−11.36
1	1	−3	6.70	− 7.04	4	3	−3	7.35	− 7.94	10	5	−3	0.	− 0.57	2	8	−3	8.09	6.82
2	1	3	4.89	− 4.62	5	3	3	7.03	− 8.61	11	5	3	4.06	− 4.17	3	8	3	5.60	5.11
2	1	−3	11.47	−14.05	5	3	−3	12.81	−13.51	11	5	−3	0.	− 1.85	3	8	−3	7.98	6.72
3	1	3	22.92	−25.45	6	3	3	5.46	− 6.53	0	6	3	6.05	4.81	4	8	3	2.61	3.43
3	1	−3	20.76	29.83	6	3	−3	20.10	20.94	1	6	3	6.79	− 6.43	4	8	−3	15.42	−13.52
4	1	3	27.84	31.79	7	3	3	23.16	23.14	1	6	−3	18.92	−16.44	5	8	3	13.05	−13.76
4	1	−3	15.36	−19.65	7	3	−3	17.08	−18.46	2	6	3	21.70	−17.72	5	8	−3	14.29	15.05
5	1	3	22.80	−22.86	8	3	3	12.13	−12.96	2	6	−3	23.81	22.32	6	8	3	17.73	17.60
5	1	−3	10.08	−12.34	8	3	−3	0.	1.25	3	6	3	23.93	21.84	6	8	−3	7.68	− 7.43
6	1	3	15.09	−15.16	9	3	3	2.82	− 2.06	3	6	−3	3.56	− 3.30	7	8	3	0.	− 0.42
6	1	−3	24.58	27.27	9	3	−3	9.22	8.69	4	6	3	4.06	− 3.83	7	8	−3	6.85	− 6.73
7	1	3	21.85	22.46	10	3	3	4.74	5.06	4	6	−3	22.48	−21.23	8	8	3	7.32	− 8.37
7	1	−3	13.73	−13.35	10	3	−3	7.47	− 6.91	5	6	3	19.93	−20.02	8	8	−3	9.78	9.85
8	1	3	18.41	−16.95	11	3	3	2.49	− 2.64	5	6	−3	26.57	28.52	9	8	3	6.88	7.43
8	1	−3	3.65	− 3.06	11	3	−3	3.88	2.33	6	6	3	22.30	23.92	9	8	−3	9.61	− 9.80
9	1	3	0.	0.94	13	3	3	0.89	− 2.13	6	6	−3	1.78	− 2.67	10	8	3	3.26	− 3.25
9	1	−3	10.97	10.44	13	3	−3	2.97	3.15	7	6	3	5.57	− 4.87	10	8	−3	3.68	− 2.55
10	1	3	4.80	5.26	0	4	3	10.38	9.56	7	6	−3	11.30	−11.46	0	9	3	5.54	− 5.55
10	1	−3	6.32	− 5.27	1	4	3	5.93	− 5.63	8	6	3	9.37	− 9.92	1	9	3	7.09	− 6.38
11	1	3	2.55	− 1.14	1	4	−3	2.13	− 1.67	8	6	−3	11.80	10.46	1	9	−3	8.21	7.40
11	1	−3	0.	0.39	2	4	3	9.64	− 8.06	9	6	3	9.58	10.73	2	9	3	6.29	5.91
12	1	3	2.19	− 2.73	2	4	−3	9.13	7.62	9	6	−3	0.	0.91	2	9	−3	8.69	− 8.27
12	1	−3	0.	− 1.25	3	4	3	16.16	14.31	10	6	3	2.58	2.80	3	9	3	5.28	− 5.85
13	1	3	0.	0.60	3	4	−3	2.43	− 2.31	10	6	−3	5.54	− 5.61	3	9	−3	2.55	3.23
13	1	−3	2.19	2.63	4	4	3	0.	1.51	11	6	3	4.48	− 4.78	4	9	3	3.85	− 3.56
14	1	−3	1.60	− 3.01	4	4	−3	15.92	−16.22	11	6	−3	0.	1.35	4	9	−3	5.90	6.09
1	2	3	6.14	4.12	5	4	3	13.85	−14.30	12	6	3	1.72	2.54	5	9	3	7.89	− 7.68
1	2	−3	10.02	−10.10	5	4	−3	13.43	13.53	12	6	−3	4.48	4.00	5	9	−3	5.46	− 5.45
2	2	3	4.80	− 4.37	6	4	3	9.04	9.86	13	6	−3	2.43	− 3.04	6	9	3	2.82	− 2.48
2	2	−3	10.05	8.89	6	4	−3	6.91	− 7.06	0	7	3	9.43	− 8.37	6	9	−3	6.26	5.56
3	2	3	8.92	8.37	7	4	3	1.48	2.17	1	7	3	1.19	− 1.99	7	9	3	2.79	3.99
3	2	−3	3.23	− 4.03	7	4	−3	9.04	− 7.99	1	7	−3	17.23	15.15	7	9	−3	0.	− 1.62
4	2	3	3.85	− 3.55	8	4	3	6.26	− 6.46	2	7	3	11.83	11.07	8	9	3	5.99	− 6.31
4	2	−3	12.84	−14.70	8	4	−3	7.59	6.91	2	7	−3	10.76	− 8.53	8	9	−3	0.	1.49
5	2	3	15.86	−14.57	9	4	3	7.47	7.23	3	7	3	9.96	− 9.86	9	9	3	3.50	3.43
5	2	−3	9.78	10.71	9	4	−3	3.97	− 3.52	3	7	−3	3.85	− 2.72	9	9	−3	4.66	5.00
6	2	3	10.14	10.37	0	5	3	12.13	−10.65	4	7	3	4.95	4.29	10	9	3	0.	0.29
6	2	−3	1.19	− 2.22	1	5	3	4.36	2.98	4	7	−3	3.32	3.32	10	9	−3	2.46	− 2.81
7	2	3	5.81	− 5.84	1	5	−3	6.40	4.31	5	7	3	3.71	4.42	11	9	3	1.63	− 3.16
7	2	−3	5.34	− 4.88	2	5	3	11.21	− 9.03	5	7	−3	2.49	− 2.44	11	9	−3	0.	0.07
8	2	3	12.04	−11.43	2	5	−3	19.48	−19.17	7	7	3	0.	0.41	1	10	3	2.67	− 3.26

TABLE IV. (Continued)

h	*k*	*l*	$F_{obs.}$	$F_{calcd.}$	*h*	*k*	*l*	$F_{obs.}$	$F_{calcd.}$	*h*	*k*	*l*	$F_{obs.}$	$F_{calcd.}$	*h*	*k*	*l*	$F_{obs.}$	$F_{calcd.}$
1	10	−3	13.70	−12.80	8	12	−3	4.15	4.40	3	17	−3	0.	− 2.49	2	2	−4	16.20	−17.87
2	10	3	8.09	− 7.47	9	12	3	2.43	2.68	4	17	−3	0.	3.25	3	2	4	10.91	9.09
2	10	−3	14.80	13.82	9	12	−3	5.99	− 6.58	1	0	4	38.34	−33.78	3	2	−4	8.56	8.08
3	10	3	13.19	14.45	10	12	−3	2.43	3.86	1	0	−4	17.22	−17.96	4	2	4	28.32	−30.29
3	10	−3	0.	− 1.29	0	13	3	2.82	− 3.07	2	0	4	11.19	− 9.94	4	2	−4	8.19	8.56
4	10	3	0.	1.34	3	13	3	3.88	4.33	2	0	−4	23.84	−39.22	5	2	4	17.90	17.17
4	10	−3	14.29	−13.12	3	13	−3	3.94	− 3.69	3	0	4	19.94	19.65	5	2	−4	9.31	11.15
5	10	3	16.01	−17.12	4	13	3	3.77	− 4.36	3	0	−4	15.61	21.68	6	2	4	3.62	2.22
5	10	−3	18.59	18.44	4	13	−3	5.49	5.99	4	0	4	0.	− 1.42	6	2	−4	14.75	−15.93
6	10	3	14.88	17.00	5	13	3	2.55	2.68	4	0	−4	21.02	−27.12	7	2	4	5.47	− 5.23
6	10	−3	2.08	− 0.53	5	13	−3	4.63	− 5.28	5	0	4	6.89	− 5.85	7	2	−4	3.52	3.88
7	10	3	8.18	− 9.29	6	13	3	3.35	1.96	5	0	−4	20.47	−21.58	8	2	4	0.	− 1.65
7	10	−3	10.53	−10.46	6	13	−3	1.78	− 3.20	6	0	4	0.	0.43	8	2	−4	18.98	17.90
8	10	3	5.69	− 6.46	7	13	3	2.99	− 3.98	6	0	−4	14.04	13.11	9	2	4	10.08	9.28
8	10	−3	10.20	10.01	7	13	−3	0.	0.80	7	0	4	19.76	19.08	9	2	−4	10.11	−16.53
9	10	3	7.21	8.11	0	14	3	6.61	− 7.13	7	0	−4	0.	3.42	10	2	4	2.63	0.67
9	10	−3	0.	0.07	1	14	3	1.78	3.34	8	0	4	15.95	−15.34	10	2	−4	6.31	5.52
10	10	3	3.26	− 3.36	1	14	−3	4.68	− 4.68	8	0	−4	14.07	−13.10	11	2	4	9.99	−10.96
10	10	−3	0.	− 0.35	2	14	3	3.74	− 3.86	9	0	4	4.85	− 3.37	11	2	−4	6.03	5.24
0	11	3	3.91	− 4.45	2	14	−3	0.	− 0.22	9	0	−4	5.60	− 4.93	12	2	4	8.35	9.70
1	11	3	6.79	− 6.82	3	14	3	1.33	1.83	10	0	4	17.75	18.12	12	2	−4	8.01	− 7.34
1	11	−3	18.29	18.05	3	14	−3	4.57	6.15	10	0	−4	17.19	14.45	0	3	4	39.05	−32.76
2	11	3	16.07	17.79	4	14	3	0.	0.92	11	0	4	12.12	−12.94	1	3	4	17.10	13.03
2	11	−3	0.	− 0.97	4	14	−3	7.26	− 8.69	11	0	−4	9.80	− 9.60	1	3	−4	13.11	−12.90
3	11	3	4.89	− 5.35	5	14	3	3.97	− 4.92	12	0	4	0.	− 1.22	2	3	4	13.23	11.47
3	11	−3	13.11	−13.57	5	14	−3	6.94	7.86	12	0	−4	4.92	− 3.96	2	3	−4	5.22	− 4.64
4	11	3	4.00	− 5.04	6	14	3	6.58	8.43	13	0	−4	11.69	11.19	3	3	4	20.74	−20.18
4	11	−3	8.92	7.82	6	14	−3	0.	− 0.69	0	1	4	12.68	12.20	3	3	−4	5.32	5.72
5	11	3	7.41	7.89	7	14	3	2.49	− 4.19	1	1	4	29.18	−25.19	4	3	4	6.59	4.59
5	11	−3	2.67	4.16	7	14	−3	3.97	− 5.30	1	1	−4	4.95	− 9.25	4	3	−4	11.69	12.48
6	11	3	3.85	4.95	8	14	3	0.65	− 2.37	2	1	4	4.82	4.31	5	3	4	10.51	9.07
6	11	−3	6.29	− 9.07	8	14	−3	5.01	6.00	2	1	−4	23.84	31.98	5	3	−4	11.59	−11.74
7	11	3	7.32	− 8.84	9	14	−3	1.75	− 3.32	3	1	4	13.14	12.62	6	3	4	14.07	−13.77
7	11	−3	3.85	4.22	0	15	3	6.08	− 7.40	3	1	−4	5.16	6.31	6	3	−4	3.31	− 2.94
8	11	3	0.	0.47	1	15	3	3.47	− 4.15	4	1	4	17.03	−15.06	7	3	4	12.03	−10.45
8	11	−3	3.68	3.83	1	15	−3	7.86	9.67	4	1	−4	19.60	−22.21	7	3	−4	16.11	15.95
9	11	3	6.43	7.66	2	15	3	5.87	7.70	5	1	4	4.02	− 2.64	8	3	4	9.37	8.56
9	11	−3	3.35	− 3.58	2	15	−3	0.	− 0.02	5	1	−4	9.06	9.79	8	3	−4	15.61	−15.25
10	11	3	4.66	− 6.71	3	15	3	1.19	− 2.15	6	1	4	12.43	11.90	9	3	4	4.79	− 3.74
10	11	−3	3.53	− 3.79	3	15	−3	7.21	− 8.83	6	1	−4	3.15	3.28	9	3	−4	2.01	− 1.89
0	12	3	6.91	− 6.92	4	15	3	5.28	− 6.68	7	1	4	0.	0.12	10	3	4	5.78	− 5.68
1	12	3	12.96	13.89	4	15	−3	6.49	7.02	7	1	−4	10.45	−10.50	10	3	−4	8.87	6.57
1	12	−3	2.82	− 2.88	5	15	3	4.80	6.00	8	1	4	9.77	− 9.30	11	3	4	2.16	3.36
2	12	3	2.82	− 4.57	5	15	−3	1.78	3.22	8	1	−4	0.	1.11	11	3	−4	2.66	− 2.11
2	12	−3	2.82	− 2.70	6	15	3	2.91	3.83	9	1	4	8.41	8.37	12	3	4	3.40	3.92
3	12	3	0.	− 0.00	6	15	−3	4.57	− 7.21	9	1	−4	8.84	6.65	12	3	−4	7.11	− 7.07
3	12	−3	4.00	4.92	0	16	3	6.88	− 8.71	10	1	4	2.63	1.68	0	4	4	47.52	40.58
4	12	3	4.83	5.51	1	16	3	7.71	10.12	10	1	−4	7.98	− 7.19	1	4	4	24.18	−19.78
4	12	−3	8.92	− 8.63	1	16	−3	0.	0.45	11	1	4	4.02	− 5.30	1	4	−4	14.65	−16.59
5	12	3	5.43	− 6.39	2	16	3	2.91	− 3.66	11	1	−4	6.06	− 6.14	2	4	4	8.32	− 6.72
5	12	−3	2.22	3.17	2	16	−3	5.34	− 8.26	12	1	4	3.65	4.09	2	4	−4	15.40	−14.13
6	12	3	5.19	5.64	3	16	3	3.35	− 4.55	12	1	−4	8.81	8.17	3	4	4	9.80	9.08
6	12	−3	5.25	− 6.26	3	16	−3	4.60	6.54	0	2	4	18.12	13.52	3	4	−4	35.21	36.50
7	12	3	3.38	− 3.46	2	17	3	2.22	− 3.02	1	2	4	42.94	−36.96	4	4	4	0.	0.44
7	12	−3	0.	1.39	2	17	−3	0.	1.45	1	2	−4	14.28	14.62	4	4	−4	13.29	−12.62
8	12	3	1.48	0.48	3	17	3	2.37	4.02	2	2	4	33.88	31.21	5	4	4	3.52	− 3.56

TABLE IV. (Continued)

h	k	l	$F_{obs.}$	$F_{calcd.}$	h	k	l	$F_{obs.}$	$F_{calcd.}$	h	k	l	$F_{obs.}$	$F_{calcd.}$	h	k	l	$F_{obs.}$	$F_{calcd.}$
5	4	−4	6.28	− 5.90	9	6	−4	7.95	− 7.85	4	9	−4	15.46	−15.32	2	12	4	2.78	− 2.80
6	4	4	5.97	− 5.05	11	6	4	5.01	− 5.71	5	9	4	0.	− 1.49	2	12	−4	4.88	5.67
6	4	−4	11.38	10.92	11	6	−4	5.47	5.74	5	9	−4	8.81	7.52	3	12	4	6.15	− 7.22
7	4	4	11.16	11.63	12	6	−4	2.91	− 2.70	6	9	4	4.85	5.23	3	12	−4	5.60	5.15
7	4	−4	3.68	3.69	0	7	4	17.84	−16.19	6	9	−4	2.32	2.66	4	12	4	7.57	8.49
8	4	4	11.66	−11.46	1	7	4	0.	− 0.51	7	9	4	1.98	− 2.05	4	12	−4	4.79	− 5.11
8	4	−4	8.25	− 7.13	1	7	−4	20.34	−18.52	7	9	−4	2.81	− 2.88	6	12	4	6.74	− 8.28
9	4	4	6.15	− 6.27	2	7	4	25.78	23.15	8	9	4	5.60	− 6.83	6	12	−4	8.66	10.82
9	4	−4	0.	− 1.38	2	7	−4	3.25	− 3.82	8	9	−4	3.86	− 4.56	7	12	4	4.27	5.67
10	4	4	8.75	10.37	3	7	4	26.74	−25.98	9	9	4	7.95	9.80	7	12	−4	4.24	− 4.04
10	4	−4	10.42	9.56	3	7	−4	6.28	5.24	9	9	−4	7.76	8.42	8	12	4	1.76	1.21
11	4	4	6.09	− 6.60	4	7	4	6.46	6.96	10	9	4	4.98	− 5.46	8	12	−4	0.	− 0.95
11	4	−4	5.84	− 5.40	4	7	−4	2.47	2.87	10	9	−4	0.	− 0.95	9	12	−4	4.27	4.84
0	5	4	5.29	− 2.79	5	7	4	14.78	14.23	11	9	−4	7.23	− 7.83	1	13	4	2.72	− 2.49
1	5	4	20.74	−18.52	5	7	−4	9.00	− 8.98	0	10	4	11.32	−10.62	1	13	−4	10.94	−13.36
1	5	−4	32.77	−29.02	6	7	4	8.44	− 8.90	1	10	4	1.85	3.02	2	13	4	6.00	6.52
2	5	4	28.88	25.40	6	7	−4	0.	0.76	1	10	−4	3.71	3.58	2	13	−4	11.87	14.45
2	5	−4	21.36	21.45	7	7	4	0.	− 1.44	2	10	4	12.15	12.90	3	13	4	1.85	− 2.93
3	5	4	3.86	3.59	7	7	−4	12.52	13.18	2	10	−4	4.76	3.26	3	13	−4	3.80	− 4.24
3	5	−4	5.72	5.03	8	7	4	4.73	4.53	3	10	4	6.89	− 7.93	4	13	4	3.52	3.18
4	5	4	11.53	−11.59	8	7	−4	10.20	− 9.99	3	10	−4	8.78	− 7.94	4	13	−4	5.87	− 5.88
4	5	−4	18.92	−20.12	9	7	4	4.36	4.43	4	10	4	2.84	− 3.77	5	13	4	1.24	− 2.70
5	5	4	1.85	2.08	9	7	−4	2.78	− 3.51	4	10	−4	4.85	− 4.17	5	13	−4	6.21	7.05
5	5	−4	10.73	10.96	10	7	4	8.50	− 9.62	5	10	4	12.86	14.63	9	13	−4	2.54	3.55
6	5	4	15.40	15.34	10	7	−4	10.11	10.99	5	10	−4	7.48	8.10	0	14	4	9.58	−10.52
6	5	−4	9.09	9.21	11	7	4	3.37	4.44	6	10	4	6.09	− 6.73	1	14	4	3.59	4.85
7	5	4	4.88	− 4.61	11	7	−4	4.64	− 5.21	6	10	−4	3.71	− 4.05	1	14	−4	0.	1.44
7	5	−4	9.65	− 9.69	0	8	4	12.74	11.40	7	10	4	3.62	− 3.64	2	14	4	3.49	4.81
8	5	4	8.44	− 8.23	1	8	4	2.50	− 2.76	7	10	−4	7.30	− 8.76	2	14	−4	4.39	5.75
8	5	−4	2.78	− 2.85	1	8	−4	14.87	−13.34	8	10	4	8.38	8.89	3	14	4	6.71	− 7.58
9	5	4	15.24	15.99	2	8	4	2.57	− 2.75	8	10	−4	11.16	13.18	3	14	−4	4.98	− 6.52
9	5	−4	14.13	14.57	2	8	−4	8.56	− 7.37	10	10	4	3.09	− 4.47	5	14	4	4.05	5.39
10	5	4	4.88	− 5.95	3	8	4	7.48	− 7.04	10	10	−4	3.65	− 4.66	5	14	−4	3.22	4.21
10	5	−4	3.90	− 4.37	3	8	−4	16.60	16.32	11	10	−4	4.33	5.55	6	14	4	3.46	− 4.29
11	5	4	5.78	− 6.56	4	8	4	7.23	6.97	0	11	4	17.59	−17.61	6	14	−4	0.	− 1.54
11	5	−4	8.44	− 8.19	4	8	−4	17.34	−19.41	1	11	4	3.99	2.87	7	14	4	0.	− 1.61
12	5	4	5.78	8.50	6	8	4	5.66	− 6.46	1	11	−4	6.31	− 5.32	7	14	−4	5.19	− 6.36
12	5	−4	9.31	10.33	6	8	−4	6.83	5.68	2	11	4	8.93	9.18	8	14	−4	3.52	5.24
0	6	4	2.07	− 3.14	7	8	4	7.36	8.09	2	11	−4	0.	0.61	0	15	4	3.25	− 4.00
1	6	4	23.56	−20.09	7	8	−4	4.88	− 4.71	3	11	4	15.46	−17.27	1	15	4	3.18	4.74
1	6	−4	19.88	16.78	8	8	4	0.	0.34	3	11	−4	0.	0.72	1	15	−4	2.16	− 2.89
2	6	4	26.28	24.83	8	8	−4	3.96	− 4.30	4	11	4	6.25	7.35	3	15	4	6.12	− 7.13
2	6	−4	0.	− 0.21	9	8	4	4.76	− 5.76	4	11	−4	2.01	2.75	3	15	−4	3.80	− 4.40
3	6	4	4.11	− 3.77	9	8	−4	0.	1.15	5	11	4	3.83	3.73	4	15	4	7.17	9.72
3	6	−4	3.12	− 2.72	10	8	4	4.02	5.14	5	11	−4	0.	− 0.45	4	15	−4	3.62	3.65
4	6	4	17.10	−15.76	10	8	−4	0.	1.55	6	11	4	7.33	− 7.28	5	15	4	0.	− 1.20
4	6	−4	2.32	1.63	11	8	4	2.75	1.96	6	11	−4	7.27	− 7.18	5	15	−4	2.72	2.91
5	6	4	16.63	15.73	11	8	−4	6.09	− 5.91	7	11	4	1.24	1.86	6	15	−4	4.76	− 6.47
5	6	−4	12.58	12.12	0	9	4	5.22	− 4.77	7	11	−4	5.90	6.00	7	15	−4	3.06	5.88
6	6	4	6.21	− 6.14	1	9	4	17.13	−17.30	8	11	4	0.	1.13	1	16	4	5.22	6.11
6	6	−4	8.69	− 8.44	1	9	−4	25.69	−24.37	8	11	−4	5.50	− 5.81	1	16	−4	1.05	− 2.03
7	6	4	5.66	− 5.32	2	9	4	17.87	16.94	9	11	4	2.84	3.72	2	16	4	3.49	− 4.18
7	6	−4	5.50	− 5.11	2	9	−4	21.86	20.33	9	11	−4	0.93	− 2.24	2	16	−4	0.	1.73
8	6	4	0.	1.33	3	9	4	6.74	− 6.56	10	11	−4	3.65	4.36	3	16	4	2.20	− 2.86
8	6	−4	10.54	10.16	3	9	−4	0.	0.60	1	12	4	6.31	6.44	3	16	−4	0.	1.34
9	6	4	3.71	3.76	4	9	4	6.28	− 6.04	1	12	−4	2.01	− 2.40	0	1	5	2.36	1.29

TABLE IV. (Continued)

h	*k*	*l*	$F_{obs.}$	$F_{calcd.}$	*h*	*k*	*l*	$F_{obs.}$	$F_{calcd.}$	*h*	*k*	*l*	$F_{obs.}$	$F_{calcd.}$	*h*	*k*	*l*	$F_{obs.}$	$F_{calcd.}$
1	1	5	6.51	− 4.61	2	4	−5	11.57	−11.19	1	7	5	5.48	− 5.22	0	10	5	7.92	7.39
1	1	−5	0.	− 0.03	3	4	5	15.76	−15.02	1	7	−5	5.35	− 5.35	1	10	5	5.02	− 4.03
2	1	5	3.82	2.70	3	4	−5	4.23	2.03	2	7	5	0.	− 1.30	1	10	−5	0.	1.22
2	1	−5	11.37	15.09	4	4	5	4.48	4.49	2	7	−5	4.40	3.25	2	10	5	6.18	7.04
3	1	5	7.30	7.43	4	4	−5	12.94	12.98	3	7	5	6.76	7.01	2	10	−5	0.	− 0.27
3	1	−5	12.57	−15.23	5	4	5	9.54	10.70	3	7	−5	6.39	− 6.16	3	10	5	0.	0.90
4	1	5	24.85	−24.23	5	4	−5	14.10	−14.20	4	7	5	7.80	− 7.33	3	10	−5	10.08	− 9.85
4	1	−5	3.28	3.81	6	4	5	16.14	−16.49	4	7	−5	0.	− 1.31	4	10	5	0.	2.06
5	1	5	16.93	15.29	6	4	−5	0.	− 1.58	5	7	5	7.14	7.73	4	10	−5	8.71	9.02
5	1	−5	15.43	16.32	7	4	5	8.71	8.85	5	7	−5	4.85	4.92	5	10	5	4.77	5.59
6	1	5	3.44	− 2.08	7	4	−5	9.08	7.97	6	7	5	0.	− 0.96	5	10	−5	2.90	− 2.52
6	1	−5	17.01	−17.30	8	4	5	6.02	6.59	6	7	−5	7.88	− 7.88	6	10	5	8.42	−10.14
7	1	5	18.09	−17.67	8	4	−5	7.09	− 5.74	7	7	5	4.85	− 5.86	6	10	−5	0.	− 0.18
7	1	−5	9.29	7.27	9	4	5	6.43	− 6.93	7	7	−5	6.18	5.79	7	10	5	6.47	7.50
8	1	5	14.60	15.16	9	4	−5	3.57	− 3.95	8	7	5	4.52	4.83	7	10	−5	3.32	4.58
8	1	−5	6.02	5.23	10	4	5	0.	− 0.25	8	7	−5	3.53	3.14	8	10	5	0.	− 0.18
9	1	5	3.36	− 3.43	10	4	−5	7.63	6.99	9	7	5	3.94	− 4.62	8	10	−5	4.40	− 4.41
9	1	−5	10.08	− 9.07	0	5	5	8.63	7.14	9	7	−5	4.77	− 4.29	0	11	5	7.09	7.23
0	2	5	5.10	3.80	1	5	5	7.18	− 5.96	0	8	5	3.32	− 2.00	3	11	5	2.49	2.60
1	2	5	7.18	5.13	1	5	−5	14.35	−14.56	1	8	5	6.72	5.10	3	11	−5	3.53	4.95
1	2	−5	6.60	7.45	2	5	5	0.	− 1.58	1	8	−5	11.41	11.06	4	11	5	0.	1.37
2	2	5	7.72	5.00	2	5	−5	12.90	11.43	2	8	5	3.44	3.36	4	11	−5	3.53	− 4.31
2	2	−5	6.18	− 4.75	3	5	5	5.35	6.19	2	8	−5	12.40	−11.15	7	11	5	0.	1.03
3	2	5	4.60	− 3.72	3	5	−5	4.81	− 5.15	3	8	5	16.43	−17.55	7	11	−5	3.11	3.20
3	2	−5	3.65	− 3.47	4	5	5	6.55	− 6.61	3	8	−5	0.	− 0.38	0	12	5	3.44	2.36
4	2	5	8.38	7.16	4	5	−5	4.19	− 4.65	4	8	5	0.	1.10	1	12	5	2.90	− 3.46
4	2	−5	7.30	7.18	5	5	5	9.75	8.97	4	8	−5	15.43	15.92	1	12	−5	4.89	4.91
5	2	5	7.92	6.69	5	5	−5	9.46	8.59	5	8	5	10.66	12.46	2	12	5	6.72	7.67
5	2	−5	9.17	− 8.15	6	5	5	3.53	− 2.53	5	8	−5	15.35	−15.26	2	12	−5	0.	− 1.15
6	2	5	3.44	− 4.04	6	5	−5	12.03	−10.76	6	8	5	14.39	−16.90	3	12	5	7.30	− 8.00
6	2	−5	0.	1.05	7	5	5	3.57	− 3.66	6	8	−5	0.	− 0.38	3	12	−5	2.49	− 1.29
0	3	5	5.72	− 4.09	7	5	−5	3.48	3.76	7	8	5	4.69	5.50	4	12	5	0.	1.38
1	3	5	4.98	− 3.58	8	5	5	6.80	8.31	7	8	−5	11.20	10.87	4	12	−5	7.47	9.27
1	3	−5	1.04	− 1.93	8	5	−5	1.87	− 2.42	8	8	5	5.23	5.66	5	12	5	5.02	6.67
2	3	5	9.96	8.35	1	6	5	1.66	1.65	8	8	−5	10.33	− 9.83	5	12	−5	8.42	− 9.92
2	3	−5	11.74	12.61	1	6	−5	2.86	2.37	9	8	5	6.76	− 7.55	6	12	5	8.17	− 9.55
3	3	5	8.34	7.45	2	6	5	3.11	2.95	9	8	−5	0.	0.92	6	12	−5	0.	0.88
3	3	−5	15.97	−18.01	2	6	−5	2.90	− 3.27	0	8	5	6.93	7.40	7	12	5	4.19	5.94
4	3	5	19.29	−18.55	3	6	5	9.75	− 8.95	1	9	5	6.97	7.20	7	12	−5	6.26	7.35
4	3	−5	5.19	4.54	3	6	−5	10.37	− 9.73	1	9	−5	13.36	−13.12	8	12	−5	3.44	− 4.91
5	3	5	16.80	17.39	4	6	5	4.81	4.32	2	9	5	8.63	− 9.23	0	13	5	4.60	6.33
5	3	−5	11.82	12.15	4	6	−5	16.88	16.05	2	9	−5	6.02	5.43	1	13	5	3.19	2.23
6	3	5	6.02	5.75	5	6	5	4.98	5.96	3	9	5	3.57	4.40	1	13	−5	8.63	−10.06
6	3	−5	17.75	−18.24	5	6	−5	8.71	− 8.75	3	9	−5	7.01	7.73	2	13	5	6.97	− 9.07
7	3	5	11.28	−12.16	6	6	5	11.82	−12.24	4	9	5	0.	− 0.63	2	13	−5	4.56	− 2.89
7	3	−5	10.08	8.83	6	6	−5	0.	0.00	4	9	−5	4.98	− 4.98	3	13	5	1.66	3.02
8	3	5	11.62	10.71	7	6	5	3.53	4.26	5	9	5	3.53	− 4.69	3	13	−5	9.54	10.81
8	3	−5	4.98	3.45	7	6	−5	9.38	7.77	5	9	−5	0.	− 0.36	4	13	5	4.81	6.08
9	3	5	1.87	− 2.03	8	6	5	4.69	4.81	6	9	5	4.77	− 4.69	4	13	−5	7.05	− 7.33
9	3	−5	6.18	− 5.16	8	6	−5	6.18	− 6.52	6	9	−5	3.57	4.66	5	13	5	4.36	− 5.16
10	3	5	2.07	− 2.51	9	6	5	6.64	− 8.29	7	9	5	5.43	5.73	5	13	−5	1.66	− 3.09
10	3	−5	5.97	5.28	9	6	−5	0.	2.04	7	9	−ς	0.	− 1.93	7	13	−5	4.77	− 5.36
0	4	5	6.14	− 5.57	10	6	5	1.24	4.17	8	9	5	0.	− 0.54	0	14	5	5.06	7.20
1	4	5	2.49	− 1.19	10	6	−5	0.	− 2.05	8	9	−5	3.32	− 4.07	1	14	5	6.35	− 8.96
1	4	−5	11.41	12.57	11	6	−5	1.24	2.92	9	9	5	5.48	− 6.20	1	14	−5	2.95	2.01
2	4	5	9.67	8.64	0	7	5	7.59	7.33	9	9	−5	0.	1.74	2	14	5	1.66	1.54

TABLE IV. (Continued)

h	k	l	$F_{obs.}$	$F_{calcd.}$	h	k	l	$F_{obs.}$	$F_{calcd.}$	h	k	l	$F_{obs.}$	$F_{calcd.}$	h	k	l	$F_{obs.}$	$F_{calcd.}$
2	14	−5	4.98	5.70	5	2	−6	5.31	5.40	11	4	−6	7.80	− 7.63	6	7	6	3.82	− 4.28
3	14	5	0.	1.64	6	2	6	0.	1.49	12	4	−6	5.52	5.80	6	7	−6	0.	− 0.68
3	14	−5	5.68	− 7.58	6	2	−6	10.88	−10.58	0	5	6	28.72	24.78	7	7	6	3.52	3.18
0	0	6	6.21	− 9.50	7	2	6	9.08	− 9.09	1	5	6	7.42	− 5.77	7	7	−6	5.26	5.03
1	0	6	24.17	31.36	7	2	−6	0.	0.75	1	5	−6	18.86	15.71	8	7	6	3.44	4.30
1	0	−6	18.14	−22.34	8	2	6	5.62	6.05	2	5	6	18.73	−16.57	8	7	−6	2.69	2.45
2	0	6	23.33	−24.54	8	2	−6	2.82	2.57	2	5	−6	7.67	6.56	9	7	6	5.34	− 6.83
2	0	−6	8.49	19.63	9	2	6	0.	1.43	3	5	6	21.02	19.64	9	7	−6	6.11	− 5.61
3	0	6	7.98	− 6.61	9	2	−6	2.80	2.49	3	5	−6	13.29	−10.60	10	7	−6	0.	1.47
3	0	−6	3.85	− 6.51	10	2	6	7.67	− 8.46	4	5	6	4.39	− 4.21	11	7	−6	4.82	5.43
4	0	6	22.38	24.43	10	2	−6	7.72	− 6.67	4	5	−6	2.05	− 3.09	0	8	6	7.60	5.20
4	0	−6	12.29	−16.91	11	2	−6	5.52	5.27	5	5	6	10.98	−10.78	1	8	6	6.26	4.46
5	0	6	12.52	−11.97	0	3	6	5.67	− 3.75	5	5	−6	12.22	11.27	1	8	−6	11.91	−10.20
5	0	−6	2.31	− 3.90	1	3	6	10.91	9.67	6	5	6	7.70	7.79	2	8	6	13.99	−12.71
6	0	6	0.	− 3.17	1	3	−6	11.45	10.19	6	5	−6	0.	− 0.03	2	8	−6	1.98	1.82
6	0	−6	12.09	12.25	2	3	6	6.93	− 5.18	7	5	6	5.72	6.01	3	8	6	3.46	2.98
7	0	6	4.44	4.32	2	3	−6	17.89	−14.79	7	5	−6	12.83	−11.65	3	8	−6	2.80	1.37
7	0	−6	0.	0.01	3	3	6	3.49	− 2.96	8	5	6	5.75	− 6.33	4	8	6	3.44	3.93
8	0	6	5.67	5.73	3	3	−6	2.05	− 1.35	8	5	−6	12.11	10.71	4	8	−6	2.82	− 1.74
8	0	−6	12.34	−12.74	4	3	6	4.21	4.22	9	5	6	0.	− 1.67	5	8	6	8.42	− 8.25
9	0	6	6.78	− 7.84	4	3	−6	11.78	10.33	9	5	−6	3.80	3.33	5	8	−6	3.46	− 3.78
9	0	−6	16.55	15.26	5	3	6	0.	0.71	10	5	6	2.49	7.39	6	8	6	0.	2.52
10	0	6	0.	0.55	5	3	−6	10.75	− 9.18	10	5	−6	9.83	− 7.98	6	8	−6	2.82	3.09
10	0	−6	3.28	− 2.83	6	3	6	10.03	− 8.88	11	5	−6	4.16	3.73	7	8	6	0.	2.27
11	0	−6	7.57	− 6.03	6	3	−6	0.	− 0.94	12	5	−6	2.16	3.25	7	8	−6	3.88	4.17
12	0	−6	8.98	8.75	7	3	6	0.	− 1.05	0	6	6	18.09	−15.07	8	8	6	2.39	− 2.13
0	1	6	13.01	18.22	7	3	−6	7.62	6.48	1	6	6	6.80	5.01	8	8	−6	7.75	− 7.84
1	1	6	11.01	−10.54	8	3	6	3.70	4.55	1	6	−6	10.68	8.32	9	8	−6	5.21	5.41
1	1	−6	0.	0.17	8	3	−6	0.	− 1.59	2	6	6	3.77	− 2.60	0	9	6	20.63	17.32
2	1	6	8.01	− 7.03	9	3	6	6.47	− 6.41	2	6	−6	9.55	7.74	1	9	6	6.03	− 4.77
2	1	−6	8.44	9.82	9	3	−6	4.82	− 4.62	3	6	6	5.85	5.31	1	9	−6	10.96	8.76
3	1	6	12.29	11.69	10	3	6	0.	1.47	3	6	−6	19.35	−15.02	2	9	6	10.78	−10.13
3	1	−6	5.85	− 6.53	10	3	−6	4.52	3.82	4	6	6	7.72	− 7.36	2	9	−6	4.90	4.03
4	1	6	4.44	− 4.25	11	3	−6	3.98	3.57	4	6	−6	15.19	13.52	3	9	6	16.60	16.24
4	1	−6	11.55	−13.60	12	3	−6	6.29	− 6.57	5	6	6	0.	− 0.44	3	9	−6	8.75	− 7.09
5	1	6	10.44	− 9.07	0	4	6	3.90	− 1.96	5	6	−6	1.95	1.27	4	9	6	3.36	− 3.83
5	1	−6	5.70	6.22	1	4	6	28.20	25.29	6	6	6	7.70	7.65	4	9	−6	0.	0.20
6	1	6	7.21	6.51	1	4	−6	13.60	−11.07	6	6	−6	8.21	− 7.24	5	9	6	7.62	− 7.51
6	1	−6	6.54	− 6.73	2	4	6	23.74	−22.14	7	6	6	7.83	− 9.10	5	9	−6	3.44	2.66
7	1	6	5.95	5.46	2	4	−6	15.65	13.03	7	6	−6	4.49	4.19	6	9	6	7.24	7.55
7	1	−6	12.24	−11.93	3	4	6	1.82	1.61	8	6	6	0.	2.43	6	9	−6	1.80	1.66
8	1	6	9.80	− 9.74	3	4	−6	4.72	4.18	8	6	−6	2.77	2.66	7	9	6	0.	1.84
8	1	−6	9.60	9.14	4	4	6	19.04	18.37	9	6	6	3.00	4.10	7	9	−6	9.78	− 9.86
10	1	6	5.59	5.83	4	4	−6	7.57	− 5.19	9	6	−6	4.13	− 4.73	8	9	6	2.57	− 3.72
10	1	−6	9.06	− 7.76	5	4	6	8.21	− 7.44	11	6	−6	3.57	3.17	8	9	−6	7.26	6.85
12	1	−6	4.70	4.68	5	4	−6	2.57	− 2.35	0	7	6	5.34	2.99	10	9	−6	7.37	− 8.36
0	2	6	15.63	−15.88	6	4	6	0.	0.57	1	7	6	9.78	7.21	1	10	6	8.65	− 6.70
1	2	6	15.37	12.71	6	4	−6	8.60	10.47	1	7	−6	16.66	13.91	1	10	−6	4.90	5.24
1	2	−6	3.21	3.69	7	4	6	3.88	4.16	2	7	6	13.70	−11.61	2	10	6	5.18	4.03
2	2	6	1.49	0.57	7	4	−6	3.98	− 3.73	2	7	−6	14.22	−11.81	2	10	−6	4.47	− 4.14
2	2	−6	14.50	14.06	8	4	6	5.70	5.05	3	7	6	5.65	4.61	3	10	6	7.67	7.43
3	2	6	3.36	− 3.71	8	4	−6	8.73	− 8.47	3	7	−6	3.82	2.61	3	10	−6	9.32	− 9.36
3	2	−6	17.17	−20.99	9	4	6	9.47	− 9.25	4	7	6	0.	1.54	4	10	6	8.83	− 8.81
4	2	6	3.18	− 3.08	9	4	−6	10.65	9.56	4	7	−6	12.52	11.01	4	10	−6	9.21	8.71
4	2	−6	12.16	13.56	10	4	6	0.	2.22	5	7	6	3.44	2.88	6	10	6	6.57	7.38
5	2	6	0.	− 0.47	10	4	−6	3.11	− 2.67	5	7	−6	4.88	− 3.65	6	10	−6	9.75	− 9.05

TABLE IV. (Continued)

h	*k*	*l*	$F_{obs.}$	$F_{calcd.}$	*h*	*k*	*l*	$F_{obs.}$	$F_{calcd.}$	*h*	*k*	*l*	$F_{obs.}$	$F_{calcd.}$	*h*	*k*	*l*	$F_{obs.}$	$F_{calcd.}$
7	10	6	4.03	− 5.40	4	11	6	3.41	− 3.60	4	12	6	1.05	− 0.44	3	13	−6	0.	1.04
7	10	−6	6.21	5.45	4	11	−6	6.21	5.64	4	12	−6	0.	0.20	4	13	6	4.77	− 6.12
8	10	−6	3.11	2.94	5	11	6	2.69	2.79	5	12	6	2.90	− 4.50	4	13	−6	0.	− 0.15
9	10	−6	3.39	− 3.16	5	11	−6	8.47	− 8.43	5	12	−6	2.31	− 2.55	6	13	−6	3.98	4.67
0	11	6	3.34	3.17	0	12	6	7.70	7.95	7	12	−6	4.11	5.73	1	14	6	5.67	− 6.18
1	11	6	3.82	2.43	1	12	6	1.28	− 1.90	0	13	6	7.47	8.22	1	14	−6	3.08	3.57
1	11	−6	12.57	12.66	1	12	−6	1.82	− 2.10	1	13	6	4.49	− 5.03	2	14	6	2.90	4.06
2	11	6	6.75	− 6.12	2	12	6	2.98	− 2.99	1	13	−6	3.67	3.93	2	14	−6	1.92	− 3.21
2	11	−6	11.45	−10.46	2	12	−6	1.82	1.12	2	13	6	1.80	− 2.72	3	14	6	3.44	4.43
3	11	6	3.62	3.76	3	12	6	6.11	5.91	2	13	−6	0.	0.89	3	14	−6	0.	− 1.86
3	11	−6	0.	− 1.09	3	12	−6	3.98	3.46	3	13	6	5.57	8.25	4	14	−6	2.05	3.63

and the low value of the discrepancy factor obtained by the block-diagonal matrix least-squares refinement guarantee that the three-dimensional molecular structure determined here is quite reliable. Thus the molecular structure of this compound, derived by chemical reasoning, has been verified by independent X-ray analysis.

As was mentioned in the previous paragraph, we used only one chemical fact to interpret the Patterson function, the fact that three bromine atoms combine with the fulvene ring. The first Fourier synthesis, computed with signs based on the three bromine atoms, gave the whole molecular shape definitely. Two low and broad peaks were observed near the position of C_8 in the first Fourier synthesis. They were quite a bit lower than those of O and C_{13}. Although another molecular model, in which the methoxybenzene rotates by 180° about the C_6–C_7 bond, seemed to be least probable, we made a least-squares refinement for it. After three least-squares, cycles the *R* factor dropped to 0.16 without non-observed reflections ($R=0.18$ including them). A Fourier map corresponding to the last cycle of the least-squares procedure gave peaks whose heights were only 10% of those of the other carbon atoms near these assumed atomic positions (the temperature factors for these atoms became 8.1 and 7.5 Å^2 in the least-squares procedure) and rather high peaks at the positions of O and C_{13} where, in this calculation, we did not put any atom. Therefore, the reliability of the present analysis is further confirmed.

This molecular structure explains the NMR spectra well. For example, the H_6 signal of this compound (7.63 p. p. m.) shifts into a field lower by 0.41 p. p. m. than that of the *p*-methoxy compound (7.22 p. p. m.). The shift may be due to the magnetic anisotropy of the oxygen atom which is disposed near the hyrogen, H_6, in this configuration.

The bond lengths and angles are illustrated

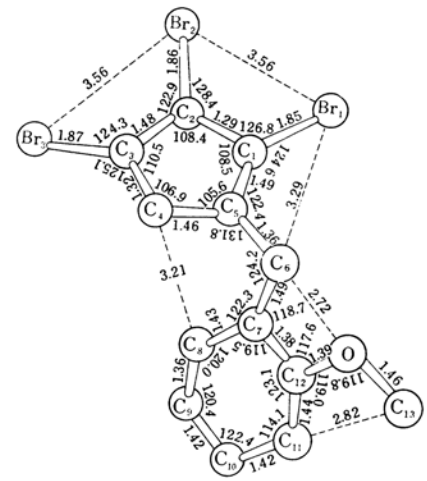

Fig. 5. Bond lengths (Å) and bond angles (°) in the molecule of 1,2,3-tribromo-6-(*o*-methoxyphenyl)fulvene.

in Fig. 5. The average estimated standard deviations of the coordinates are 0.003, 0.022 and 0.015 Å for bromine, carbon and oxygen atoms respectively. Therefore, the estimated standard deviations of the bonds are 0.022 Å for C–Br, 0.027 Å for C–O, and 0.031 Å for C–C. In the fulvene ring, C_1–C_2 and C_3–C_4 may be double bonds, the mean bond length being 1.31 Å. The bonds C_2–C_3, C_4–C_5 and C_5–C_1 are thought to be single bonds, their mean length being 1.48 Å. The bond C_5–C_6 seems to be double, and the C–C distances in the benzene ring are within the range from 1.36 to 1.44 Å. The lengths of C_{12}–O (1.39Å) and O–C_{13} (1.46 Å) are reasonable if the standard deviations in the bond lengths are allowed for. The mean C–Br distance is 1.86 Å, in good agreement with the normal value of this pair (1.85 Å).[3]

3) "Tables of Interatomic Distances and Configuration in Molecules and Ions," The Chemical Society, London (1958).

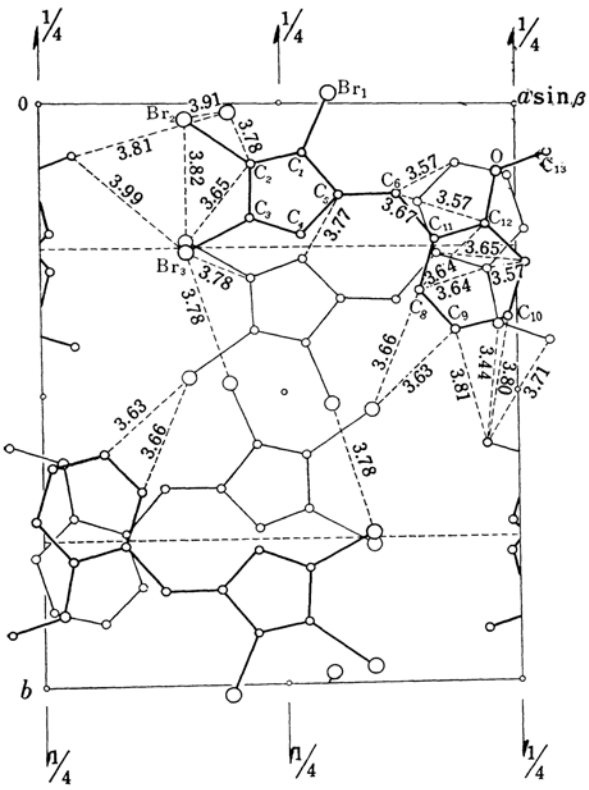

Fig. 6. Arrangement of molecules in the unit cell viewed along the c axis, with short intermolecular distances (Å).

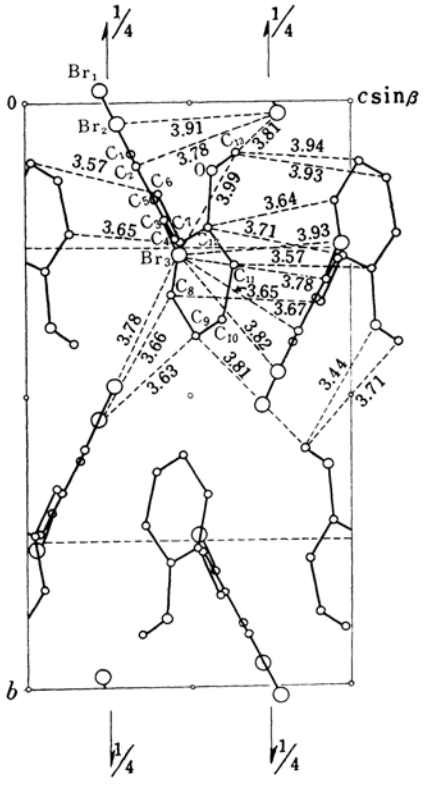

Fig. 7. Arrangement of molecules viewed along the a axis.

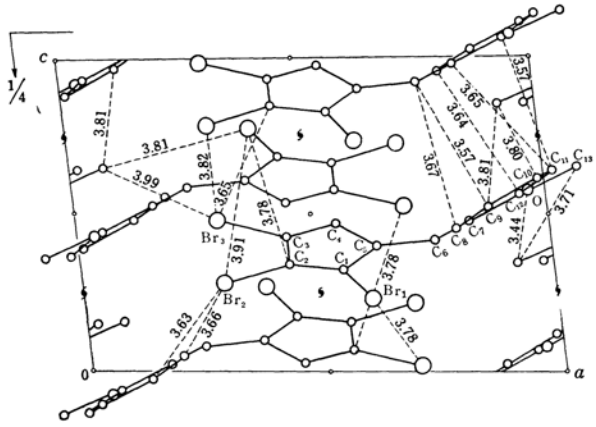

Fig. 8. Arrangement of molecules viewed along the b axis.

The benzene ring is approximately planar, the best plane being represented by the equation;

$$Y=-12.76X+27.80Z+51.01$$

where $X=x\sin\beta$, $Y=y$, and $Z=z+x\cos\beta$. The coefficients were determined by the least-squares method. The fulvene ring is also nearly planar, the best plane being described by the equation:

$$Y=0.1836X+1.859Z-3.159$$

The perpendicular displacements of the atoms from these mean planes are listed in Table V. The plane of the benzene ring makes an angle of 38°54′ with the plane of the fulvene ring. This angle between two planes mainly occurs as a result of the twisting about the bond C_6–C_7. The distance between the non-bonded atoms C_4 and C_8 is 3.21 Å; contact between

TABLE V. DEVIATIONS FROM THE LEAST-SQUARES PLANES (Å)

Atom	From the benzene plane	Atom	From the fulvene plane
C_7	0.03	C_1	0.00
C_8	−0.02	C_2	−0.01
C_9	−0.00	C_3	0.01
C_{10}	−0.03	C_4	−0.00
C_{11}	0.02	C_5	0.00
C_{12}	0.00	Br_1	0.10
C_1	0.61	Br_2	0.12
C_2	1.36	Br_3	−0.07
C_3	1.98	C_6	0.07
C_4	1.56	C_7	0.04
C_5	0.67	C_8	−0.77
Br_1	−0.30	C_9	−0.75
Br_2	1.79	C_{10}	0.01
Br_3	3.09	C_{11}	0.83
C_6	0.03	C_{12}	0.77
O	0.04	O	1.53
C_{13}	−0.15	C_{13}	2.20

them might cause this twisting. If the positions of the hydrogen atoms bonding to C_4 and C_8 are estimated with conventional C–H length and angles, the distance between them is approximately 1.9 Å; this value might be reasonable for the contact of such hydrogens.

The methyl group in the methoxy group deviates from the plane of the benzene ring, the C_{11}–C_{13} distance being 2.82 Å. Three bromine atoms and C_6 are out of the plane of the fulvene ring, probably because of the mutual repulsion among these substituted atoms and this group. The Br_1–Br_2, Br_2–Br_3 and Br_1–C_6 distances are 3.56, 3.56 and 3.29 Å respectively.

Figures 6, 7 and 8 show the crystal structures projected along the c, a and b axes respectively. If the van der Waals radii of the bromine, oxygen and methyl groups are taken to be 1.95, 1.40 and 2.0 Å respectively, and if the half thickness of the aromatic ring is 1.70 Å, as is usually accepted, all the intermolecular approaches observed are equal to or longer than the sums of the radii of the corresponding atoms within the range of errors. It might be concluded from Figs. 6, 7 and 8 that the molecules are well packed by the van der Waals interactions. The benzene rings which are related to each other by the c glide plane are nearly parallel, because the plane of any benzene ring makes a very small angle with the b axis. Therefore, these nearly parallel benzene rings are piled up at a distance of about 3.5 Å in the direction of the c axis, as may be seen from Fig. 8.

Although the dipole moment of the present compound has not been measured, it is, expected to show a considerable value, judging from the value of the related chloro derivative (4.72D). If the crystal structure of the present compound is discussed in view of the arrangement of such dipolar molecules, it may be compared to that of 3-bromo-2-methoxytropone,[4] although there is a slight difference in the direction of the glide planes, the c glide in the former and the a glide in the latter.

From the similarity of the unit cell dimensions between the present crystal and that of the chloro derivative, it may be deduced that the molecular packing of the latter is essentially the same as that of the former, which has been described here.

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4) K. Furukawa, Y. Sasada, A. Shimada and T. Watanabe, This Bulletin, 37, 1871 (1964).