intensity measurements cannot be obtained. Further work is in progress, and it is not yet certain whether the crystals are twinned or whether two chemically different species are co-crystallizing. The space groups given are, therefore, somewhat uncertain.

References

ABRAHAMS, S. C. (1964). *Acta Cryst.* **17**, 1327.
ABRAHAMS, S. C. & BERNSTEIN, J. L. (1965). *Acta Cryst.* **18**, 926.
ALLEN, A. D., BOTTOMLEY, F., HARRIS, R. O., REINSALU, V. P. & SENOFF, C. V. (1967). *J. Amer. Chem. Soc.* In the press.
ALLEN, A. D. & SENOFF, C. V. (1965). *Chem. Comm.* p. 621.
BOTTOMLEY, F. & NYBURG, S. C. (1966). *Chem. Comm.* p. 897.
BUSING, W. R., LEVY, H. A., MARTIN, K. O. (1962). *A Fortran Crystallographic Least Squares Program.* Report ORNL-TM-305. Oak Ridge National Laboratory, Tennessee.
DASENT, W. E. (1965). *Non-existent Compounds,* p. 159. New York: Marcel Dekker.
HODGSON, D. J. & IBERS, J. A. (1966). Private communication.
International Tables for X-ray Crystallography (1962). Vol. III. Birmingham: Kynoch Press.
JORDAN, T. H., SMITH, H. W., STREIB, W. E. & LIPSCOMB, W. N. (1964). *J. Chem. Phys.* **41**, 756.
KAPUSTINSKII, A. (1956). *Quart. Rev. Chem. Soc. Lond.* **10**, 283.
ORGEL, L. E. (1960). *An Introduction to Transition-Metal Chemistry,* p. 137. London: Methuen.
PARKES, A. S. & HUGHES, R. E. (1963). *Acta Cryst.* **16**, 734.
SHIONO, R. (1964). Private communication.
WADDINGTON, T. C. (1966). *Trans. Faraday Soc.* **62**, 1482.

X-ray Studies of C₂₅ Terpenoids. I. The Crystal Structure of Ophiobolin Methoxybromide

BY MASUO MORISAKI AND SHIGEO NOZOE

Institute of Applied Microbiology, University of Tokyo, Yayoi, Tokyo, Japan

AND YOICHI IITAKA

Faculty of Pharmaceutical Sciences, University of Tokyo, Hongo, Tokyo, Japan

The crystal structure and absolute configuration of ophiobolin methoxybromide, a derivative of the fungus metabolite ophiobolin, has been determined by three-dimensional X-ray analysis. The unit cell of ophiobolin methoxybromide, C₂₆H₃₉O₅Br, is orthorhombic with dimensions $a = 13·19$, $b = 22·27$, $c = 8·46$ Å. The space group is $P2_12_12_1$ with four molecules per cell. Refinement was carried out for 1499 observed reflexions by diagonal and then full-matrix least-squares calculations with anisotropic thermal parameters for bromine and isotropic ones for light atoms. The final R value was 0·11. The analysis revealed a novel skeletal structure of a new kind of terpenoid. The molecule consists of a five-, eight- and five-membered tricyclic ring system and a side chain containing a tetrahydrofuran group. Experiments on the biosynthesis of ophiobolin confirmed that this compound is the first example of a C₂₅ terpenoid.

Introduction

Ophiobolin is a metabolic product of the plant pathogenic fungus *Ophiobolus miyabeanus* (later it has been reclassified as *Cochliobolus miyabeanus*). Isolation of this metabolite was first reported by Ishibashi & Nakamura (1958). It has also been found that *Helminthosporium turcicum* and other *Helminthosporium* species fungi produce ophiobolin (Ishibashi, 1961, 1962*a*). Orsenigo (1957) reported the isolation of cochliobolin from *Cochliobolus miyabeanus*. After a report of the complete analysis of ophiobolin was published (Nozoe,

Morisaki, Tsuda, Iitaka, Takahashi, Tamura, Ishibashi & Shirasaka, 1965), Canonica, Fiecchi, Kienle & Scala (1966) reported the plane structure of cochliobolin. At present, it is generally accepted that cochliobolin and ophiobolin are one and the same compound (Fig. 1). To avoid confusion, it has been decided to call both the compounds 'ophiobolin *A*' (Tsuda, Nozoe, Morisaki, Hirai, Itai, Okuda, Canonica, Fiecchi, Kienle & Scala, 1967).

Ophiobolin has a pathogenic activity for plant diseases. The physiological activities of ophiobolin and its related compounds have been reported elsewhere

(Ishibashi, 1962b). Parallel with the chemical study, an X-ray analysis of the bromomethoxy derivative of ophiobolin has been carried out. A preliminary report of this work has already appeared (Nozoe, Morisaki, Tsuda, Iitaka, Takahashi, Tamura, Ishibashi & Shirasaka, 1965).

On the other hand, two compounds, named zizanin A and zizanin B, were isolated from *Helminthosporium* species fungi. The structures of these compounds have been proposed on the basis of chemical considerations (Nozoe, Hirai & Tsuda, 1966).

Experimental

Spectroscopic analysis of ophiobolin indicated the presence of hydroxyl, α,β-unsaturated carbonyl and olefinic groups and a ketone function in a five-membered ring. Preparation of a heavy atom derivative, ophiobolin methoxybromide, was then tried by direct bromination of ophiobolin at the olefinic group. A treatment of ophiobolin with bromine in methanol solutions in the presence of sodium acetate yielded the methoxybromide, the reaction proceeding quantitatively with an equi-molar amount of bromine. Spectroscopic data (infrared, ultraviolet and nuclear magnetic resonance) and elementary analysis clearly indicated that during the bromination reaction a bromine atom and a methoxyl group were added to the olefinic group while the other part of the molecule was left unchanged.

Ophiobolin methoxybromide crystallizes from ether as orthorhombic colorless prisms elongated along the c axis. The lattice constants were determined from equatorial precession photographs taken about the a and b axes with Cu Kα radiation. The density was measured by the flotation method in aqueous solutions of potassium iodide of various concentrations.

Crystal data

Ophiobolin methoxybromide, C$_{26}$H$_{39}$O$_5$Br. Mol.wt. 511·49, m.p. 155–157°C.

Orthorhombic,
$a=13\cdot19\pm0\cdot04$, $b=22\cdot27\pm0\cdot07$, $c=8\cdot46\pm0\cdot03$ Å

$U=2485\cdot0$ Å³

$D_m=1\cdot40$ g.cm^{-3}, $D_x=1\cdot37$ g.cm^{-3}

$F(000)=1080$, $Z=4$

Absent spectra, h00 when h is odd, 0k0 when k is odd, 00l when l is odd

Space group, $P2_12_12_1$.

Three-dimensional intensity data were collected from multiple-film equi-inclination Weissenberg photographs taken with Cu Kα radiation. Layer lines of zero to six around c and zero to four around a were recorded. The intensities were estimated visually with the aid of an intensity scale. Lorentz and polarization corrections were applied and a total of 1499 independent structure factors were derived. The interlayer correlation was carried out by comparing the equivalent structure factors recorded on various layers.

Determination and refinement of the structure

Sharpened Patterson maps on Harker sections at $u=\frac{1}{2}$, $v=\frac{1}{2}$ and $w=\frac{1}{2}$, respectively, were calculated and the coordinates of the bromine atom ($x=0\cdot154$, $y=0\cdot187$, $z=-0\cdot184$) were determined by referring to the coordinate system given in *International Tables for X-ray Crystallography* (1952). The R value calculated for the structure containing only the bromine atoms was 0·50. Several cycles of structure factor and Fourier or difference Fourier calculations yielded all of the thirty-two atomic positions in the electron density map. The oxygen atoms could be identified on the Fourier map on account of their predominant peak height. The molecular structure derived in this way could well be accounted for the chemical behavior and the nuclear magnetic resonance spectra of this compound. A structure factor calculation yielded an R value of 0·22.

Refinement of the structure was first carried out by three cycles of diagonal least-squares calculations which reduced R to 0·14. Finally, four cycles of full-matrix least-squares calculations with the *ORFLS* pro-
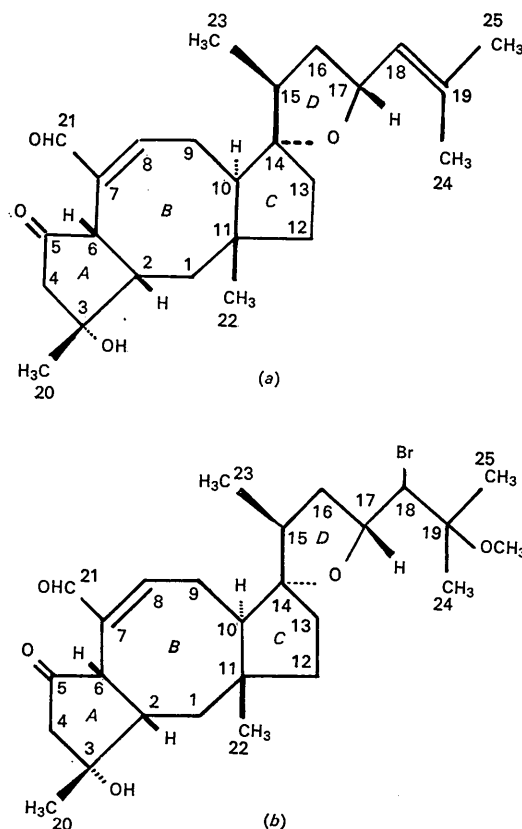
Fig. 1. Chemical formulae of (a) ophiobolin, (b) ophiobolin methoxybromide.

gram (Busing, Martin & Levy, 1962), in which anisotropic thermal vibrations for bromine and isotropic vibrations for light atoms were allowed for, gave an R value of 0·114 for 1499 observed reflexions. The weighting functions used for the full-matrix least-squares calculations were:

$$\sqrt{w} = F_0/40 \,, \quad \text{when } F_0 \le 40 \,,$$
$$\sqrt{w} = 40/F_0 \,, \quad \text{when } F_0 > 40 \,.$$

Atomic scattering factors were those of Berghuis, Haanappel, Potters, Loopstra, MacGillavry & Veenendaal (1955) for oxygen and carbon, and for bromine those cited as SX-71 in *International Tables for X-ray Crystallography* (1962, p.202).

The final atomic parameters are given in Table 1 together with their standard deviations. The observed and calculated structure factors are listed in Table 2. A composite electron density map obtained from the

Table 1. *The final atomic fractional coordinates, thermal parameters and their standard deviations*

To represent the correct absolute configuration, these coordinates should refer to the right-handed coordinate system.

	x	$\sigma(x)$	y	$\sigma(y)$	z	$\sigma(z)$	B	$\sigma(B)$
Br	0·1528	0·0003	0·1889	0·0002	−0·1838	0·0004	(see below)	
O(1)	−0·3652	0·0013	0·0144	0·0007	0·2335	0·0018	4·60	0·41
O(2)	−0·4886	0·0013	0·1711	0·0007	0·0585	0·0021	4·66	0·43
O(3)	−0·3874	0·0015	0·0586	0·0009	−0·0749	0·0023	5·05	0·45
O(4)	0·1176	0·0012	0·1168	0·0007	0·1365	0·0018	3·62	0·37
O(5)	0·2795	0·0017	0·0854	0·0009	−0·3125	0·0030	7·45	0·55
C(1)	−0·1977	0·0017	0·0832	0·0010	0·3506	0·0027	2·88	0·54
C(2)	−0·2997	0·0016	0·1109	0·0009	0·3255	0·0030	2·71	0·48
C(3)	−0·3900	0·0020	0·0648	0·0012	0·3315	0·0035	4·67	0·61
C(4)	−0·4828	0·0021	0·0995	0·0012	0·2683	0·0032	4·57	0·66
C(5)	−0·4435	0·0021	0·1414	0·0014	0·1523	0·0039	5·65	0·72
C(6)	−0·3204	0·0017	0·1459	0·0010	0·1650	0·0029	3·20	0·53
C(7)	−0·2653	0·0020	0·1268	0·0011	0·0236	0·0029	3·78	0·57
C(8)	−0·1725	0·0022	0·1471	0·0011	−0·0104	0·0029	3·62	0·53
C(9)	−0·1038	0·0018	0·1847	0·0012	0·0989	0·0027	3·62	0·53
C(10)	−0·0493	0·0016	0·1418	0·0009	0·2035	0·0027	2·27	0·47
C(11)	−0·1049	0·0017	0·1280	0·0010	0·3698	0·0025	2·50	0·49
C(12)	−0·0180	0·0019	0·0916	0·0011	0·4559	0·0027	3·13	0·55
C(13)	0·0799	0·0018	0·1286	0·0011	0·4209	0·0029	3·32	0·56
C(14)	0·0631	0·0017	0·1583	0·0011	0·2456	0·0025	2·51	0·51
C(15)	0·1125	0·0019	0·2204	0·0011	0·2171	0·0030	3·91	0·61
C(16)	0·2269	0·0017	0·2028	0·0010	0·1871	0·0032	3·52	0·53
C(17)	0·2206	0·0018	0·1369	0·0011	0·1140	0·0029	3·58	0·58
C(18)	0·2530	0·0020	0·1353	0·0012	−0·0631	0·0030	4·21	0·62
C(19)	0·2523	0·0021	0·0726	0·0012	−0·1388	0·0031	4·63	0·65
C(20)	−0·4002	0·0021	0·0352	0·0012	0·5001	0·0033	4·61	0·68
C(21)	−0·3071	0·0021	0·0863	0·0012	−0·0974	0·0033	4·27	0·66
C(22)	−0·1370	0·0019	0·1859	0·0012	0·4643	0·0024	3·39	0·49
C(23)	0·0892	0·0018	0·2688	0·0010	0·3427	0·0030	3·77	0·57
C(24)	0·1529	0·0025	0·0416	0·0012	−0·1445	0·0034	6·37	0·78
C(25)	0·3448	0·0029	0·0394	0·0014	−0·0731	0·0038	7·49	0·85
C(26)	0·3534	0·0024	0·1111	0·0012	−0·3583	0·0032	5·47	0·68

The temperature factors of the bromine atom take the form

$$\exp\{-(\beta_{11}h^2 + \beta_{22}k^2 + \beta_{33}l^2 + 2\beta_{12}hk + 2\beta_{23}kl + 2\beta_{13}hl)\}\,;$$

the parameters are

	β_{11}	β_{22}	β_{33}	β_{12}	β_{13}	β_{23}
β_{ij}	0·01196	0·00298	0·01295	0·00088	·0·00272	−0·00011
$\sigma(\beta)$	0·00029	0·00007	0·00040	0·00016	0·00041	0·00024

Mean standard deviations in positional parameters

	$\sigma(x)$	$\sigma(y)$	$\sigma(z)$	Mean value
Br	0·0003 (0·0038 Å)	0·0002 (0·0033 Å)	0·0004 (0·0030 Å)	0·0036 Å
C	0·0020 (0·025 Å)	0·0011 (0·025 Å)	0·0030 (0·025 Å)	0·025 Å
O	0·0014 (0·018 Å)	0·0008 (0·017 Å)	0·0022 (0·018 Å)	0·018 Å

Mean estimated standard deviations in bond lengths

σ(Br–C)	0·025 Å
σ(C–C)	0·035
σ(C–O)	0·031

Mean estimated standard deviations in tetrahedral bond angles
$$\sigma(\text{C–C–C}) = 2\cdot0^\circ$$

Table 2. *Observed and calculated structure factors*

h	k	l	|Fo|	A	B	|Fc|
2	0	0	114.11	-104.43	0.00	104.43
4	0	0	144.07	-135.72	0.00	135.72
6	0	0	32.03	25.00	0.00	25.90
8	0	0	53.31	46.90	0.00	46.90
1	1	0	7.94	0.00	2.72	2.72
2	1	0	164.96	-175.82	0.00	175.82
3	1	0	89.31	0.00	-80.04	80.04
4	1	0	27.46	31.69	0.00	31.69
5	1	0	19.04	0.00	-14.52	14.52
7	1	0	24.96	0.00	-12.12	12.12
9	1	0	22.09	0.00	20.45	20.45
11	1	0	43.96	0.00	-38.00	38.00
0	2	0	35.52	-43.35	0.00	43.35
1	2	0	71.68	0.00	72.21	72.21
2	2	0	20.17	-13.02	0.00	13.02
3	2	0	74.93	0.00	-73.98	73.98
4	2	0	13.31	18.81	0.00	18.81
5	2	0	69.69	0.00	-55.84	55.84
7	2	0	25.16	0.00	31.37	31.37
9	2	0	13.67	0.00	-24.00	24.00
11	2	0	17.67	0.00	-13.91	13.91
1	3	0	81.42	0.00	-88.61	88.61
3	3	0	21.92	0.00	-26.35	26.35
4	3	0	46.18	-41.60	0.00	41.60
5	3	0	69.77	0.00	66.06	66.06
6	3	0	24.54	17.42	0.00	17.42
7	3	0	48.56	0.00	33.96	33.96
8	3	0	32.60	25.73	0.00	25.73
9	3	0	23.26	0.00	-22.67	22.67
13	3	0	14.40	0.00	13.95	13.95
14	3	0	13.43	8.73	0.00	8.73
0	4	0	228.88	-229.10	0.00	229.10
1	4	0	159.61	0.00	-154.28	154.28
2	4	0	26.49	24.39	0.00	24.39
3	4	0	198.94	0.00	198.00	198.00
4	4	0	81.78	-77.23	0.00	77.23
5	4	0	79.79	0.00	47.06	47.06
6	4	0	22.42	15.25	0.00	15.25
7	4	0	19.90	0.00	-31.52	31.82
8	4	0	13.47	11.25	0.00	11.25
11	4	0	24.16	0.00	20.75	20.75
1	5	0	81.29	0.00	81.56	81.56
2	5	0	44.14	-27.92	0.00	27.92
3	5	0	29.95	0.00	-14.98	14.98
4	5	0	50.84	-39.46	0.00	39.46
5	5	0	40.47	0.00	-45.02	45.02
6	5	0	76.36	59.58	0.00	59.58
7	5	0	69.02	0.00	66.26	66.26
8	5	0	13.65	16.34	0.00	16.34
12	5	0	23.30	-20.06	0.00	20.06
0	6	0	62.21	64.29	0.00	64.29
1	6	0	18.89	0.00	-18.30	18.30
2	6	0	12.32	21.00	0.00	21.00
3	6	0	84.21	0.00	-74.04	74.04
4	6	0	68.42	-64.14	0.00	64.14
5	6	0	89.46	0.00	73.46	73.46
6	6	0	50.26	44.25	0.00	44.25
7	6	0	29.42	0.00	26.71	26.71
9	6	0	26.51	0.00	-26.97	26.97
10	6	0	36.47	-42.03	0.00	42.03
14	6	0	12.90	10.98	0.00	10.98
1	7	0	57.73	0.00	-64.27	64.27
2	7	0	80.75	-80.47	0.00	80.47
3	7	0	32.82	0.00	30.96	30.96
4	7	0	35.47	37.92	0.00	37.92
5	7	0	33.43	0.00	28.54	28.54
6	7	0	26.77	23.78	0.00	23.78
7	7	0	19.59	0.00	-27.64	27.64
8	7	0	44.21	-45.14	0.00	45.14
9	7	0	25.47	0.00	-23.13	23.13
11	7	0	24.43	0.00	22.25	22.25
0	8	0	67.85	-65.92	0.00	65.92
1	8	0	18.32	0.00	-18.24	18.24
2	8	0	28.08	26.70	0.00	26.70
3	8	0	50.62	54.03	0.00	54.03
4	8	0	64.50	-64.45	0.00	64.45
5	8	0	36.03	0.00	-26.20	26.20
6	8	0	26.10	-21.50	0.00	21.50
2	9	0	15.83	21.08	0.00	21.08
3	9	0	34.15	0.00	29.23	29.23
4	9	0	46.00	-43.82	0.00	43.82
6	9	0	26.18	-25.97	0.00	25.97
7	9	0	33.18	0.00	-23.25	23.25
8	9	0	51.76	50.94	0.00	50.94
9	9	0	46.71	43.51	0.00	43.51
1	10	0	4.61	0.00	-11.96	11.96
2	10	0	40.35	-34.12	0.00	34.12
3	10	0	60.61	0.00	56.25	56.25
4	10	0	14.28	-19.87	0.00	19.87
6	10	0	57.66	53.17	0.00	53.17
7	10	0	26.60	0.00	-25.02	25.02
8	10	0	22.66	18.58	0.00	18.58
9	10	0	21.42	0.00	21.70	21.70
1	11	0	36.32	0.00	34.42	34.42
2	11	0	40.12	-37.51	0.00	37.51
3	11	0	18.00	18.35	0.00	18.35
4	11	0	55.52	48.70	0.00	48.70
6	11	0	24.25	0.00	-20.01	20.01
1	12	0	30.02	-32.19	0.00	32.19
2	12	0	43.25	0.00	39.80	39.80
3	12	0	19.55	21.44	0.00	21.44
4	12	0	71.92	0.00	-65.57	65.57
6	12	0	38.78	37.92	0.00	37.92
8	12	0	32.87	0.00	-29.46	29.46
9	12	0	18.42	22.39	0.00	22.39
1	13	0	38.71	0.00	34.84	34.84
3	13	0	60.50	0.00	-63.48	63.48
2	13	0	5.98	4.72	0.00	4.72
5	13	0	33.10	0.00	-29.46	29.46
6	13	0	35.42	0.00	68.48	68.48
7	13	0	39.74	0.00	-36.46	36.46
8	13	0	29.85	-30.00	0.00	30.00
1	14	0	48.30	0.00	-45.85	45.85
2	14	0	15.41	-20.69	0.00	20.69
3	14	0	52.82	0.00	52.89	52.89
5	14	0	33.84	40.92	0.00	40.92
6	14	0	43.97	-39.62	0.00	39.62
7	14	0	23.75	0.00	-22.75	22.75
8	14	0	22.00	-23.64	0.00	23.64
1	15	0	19.28	0.00	19.79	19.79
2	15	0	31.28	31.69	0.00	31.69
3	15	0	16.57	0.00	-22.20	22.20
5	15	0	25.71	-26.45	0.00	26.45
6	15	0	20.83	18.53	0.00	18.53
0	16	0	50.40	50.36	0.00	50.36
1	16	0	14.20	0.00	16.59	16.59
2	16	0	22.78	-20.14	0.00	20.14
4	16	0	44.19	-49.20	0.00	49.20

h	k	l	|Fo|	A	B	|Fc|
5	16	0	24.43	25.63	0.00	25.40
1	17	0	3.50	0.00	-1.69	1.69
2	17	0	33.49	-20.63	0.30	20.63
4	17	0	4.55	0.00	14.29	14.29
4	17	0	28.72	30.52	0.00	30.52
5	17	0	21.84	0.00	-15.14	15.14
7	17	0	19.50	0.00	17.89	17.89
8	17	0	21.87	-19.12	0.00	19.12
0	18	0	51.96	-50.00	0.00	50.00
1	18	0	31.35	31.82	0.00	31.82
3	18	0	7.90	0.00	-19.34	19.34
7	18	0	16.35	0.00	13.46	13.46
8	18	0	15.92	12.60	0.30	12.60
1	19	0	7.90	0.00	-10.69	10.69
4	19	0	18.37	-19.01	0.00	19.01
1	20	0	24.21	0.00	-28.07	28.07
2	20	0	26.50	0.00	29.66	29.66
6	20	0	17.53	19.71	0.00	19.71
7	20	0	15.50	0.00	-19.92	19.92
8	20	0	14.70	-17.42	0.00	17.42
1	21	0	15.14	0.00	10.91	10.91
5	21	0	15.86	0.00	-19.18	19.18
0	22	0	26.17	26.67	0.00	26.67
3	22	0	18.00	0.00	14.36	14.36
2	23	0	13.82	-11.59	0.00	11.59
0	24	0	18.99	-18.20	0.00	18.20
1	24	0	15.19	19.09	0.00	19.09
2	24	0	15.97	13.73	0.00	13.73
1	0	1	18.57	0.00	-1.72	1.72
2	0	1	100.05	0.00	98.44	98.44
3	0	1	69.67	0.00	-55.48	55.48
4	0	1	76.01	0.00	-64.75	64.75
5	0	1	62.58	0.00	45.50	48.50
6	0	1	102.17	0.00	95.60	95.60
8	0	1	20.01	0.03	21.15	21.15
9	0	1	43.63	0.00	-47.25	47.25
10	0	1	36.49	0.00	34.18	34.18
0	1	1	116.20	0.00	130.29	130.29
1	1	1	128.62	-21.21	-120.77	122.65
2	1	1	80.75	30.22	-58.08	65.47
3	1	1	44.69	-36.67	-5.81	37.13
4	1	1	24.50	11.50	12.85	17.25
5	1	1	18.64	13.54	22.56	26.72
6	1	1	63.50	2.13	-50.35	50.39
7	1	1	25.16	-20.72	15.57	25.92
8	1	1	34.04	-3.44	-30.72	30.91
9	1	1	19.79	13.33	-15.19	20.21
10	1	1	16.12	-4.89	10.90	11.95
0	2	1	85.44	74.65	0.00	74.68
1	2	1	54.13	-43.01	12.53	44.78
2	2	1	125.77	102.35	-25.21	105.41
3	2	1	45.73	8.85	47.08	47.90
4	2	1	53.66	36.77	14.96	39.70
5	2	1	68.69	29.46	-50.24	58.24
6	2	1	44.36	46.75	10.42	47.03
7	2	1	38.10	17.71	-27.79	32.95
8	2	1	26.44	-19.68	-17.64	26.42
9	2	1	34.21	-21.92	27.03	34.81
10	2	1	40.76	32.47	-2.82	32.59
11	2	1	16.14	-8.87	-1.56	9.00
12	2	1	21.18	-15.82	9.08	18.25
0	3	1	11.25	0.00	11.89	11.89
1	3	1	102.04	4.04	-81.40	81.50
2	3	1	93.89	65.66	-45.50	81.63
3	3	1	94.41	82.56	-17.70	84.46
4	3	1	60.00	2.43	-52.39	52.45
5	3	1	72.69	-65.66	-15.42	67.44
6	3	1	21.17	-13.92	10.65	17.53
7	3	1	60.80	46.37	10.09	47.45
8	3	1	15.46	3.34	-13.84	14.03
10	3	1	4.17	11.53	0.00	11.53
1	4	1	47.72	23.49	36.60	43.49
2	4	1	63.14	-1.23	-48.76	48.77
3	4	1	52.96	-11.17	-40.60	42.11
4	4	1	71.48	-72.96	-0.37	72.96
6	4	1	56.69	-38.57	-19.52	43.22
7	4	1	77.00	69.07	0.91	69.97
9	4	1	20.21	-10.21	6.82	12.28
10	4	1	19.14	5.84	13.54	14.75
0	5	1	15.57	-13.53	-18.66	23.05
1	5	1	15.90	-12.82	-1.63	12.92
2	5	1	75.61	0.00	-78.03	78.03
3	5	1	54.82	40.50	15.93	44.70
4	5	1	111.35	-97.46	7.31	97.74
5	5	1	52.39	-29.85	36.53	47.17
6	5	1	70.99	44.17	-41.07	60.32
7	5	1	29.82	21.02	-0.43	21.02
8	5	1	17.84	-9.29	6.42	11.29
9	5	1	35.50	-27.87	-2.99	28.03
2	6	1	147.17	-145.68	0.00	145.68
3	6	1	102.75	-62.76	56.99	84.77
4	6	1	134.45	-17.32	-105.24	106.65
5	6	1	44.42	14.91	1.43	41.71
6	6	1	58.57	25.52	12.35	28.60
7	6	1	33.52	-37.03	-12.38	39.04
8	6	1	47.97	-22.67	37.50	43.82
9	6	1	22.58	19.79	14.55	24.74
10	6	1	22.40	13.58	3.85	14.11
0	7	1	29.85	26.57	-3.14	26.75
1	7	1	109.67	0.00	94.79	94.79
2	7	1	18.46	-11.00	-68.28	69.30
4	7	1	105.84	79.14	18.74	81.32
5	7	1	70.15	-16.64	-60.29	60.65
6	7	1	50.57	-38.37	-13.29	40.60
7	7	1	113.28	16.71	90.73	92.25
8	7	1	48.56	-3.67	38.52	38.69
9	7	1	23.37	21.47	0.96	21.49
1	7	1	26.89	0.73	-24.32	24.32
10	7	1	22.62	-20.53	2.59	20.69
1	8	1	36.34	35.73	0.00	35.73
2	8	1	64.50	5.50	-61.97	62.21
3	8	1	42.60	-0.35	-37.78	37.78
4	8	1	58.57	14.03	51.04	53.18
5	8	1	27.30	1.47	22.05	22.13
6	8	1	34.02	12.77	24.67	26.91
7	8	1	20.72	-7.51	-1.17	7.60
8	8	1	42.51	16.14	-42.28	45.25
9	8	1	37.75	-22.82	-25.13	33.94
10	8	1	22.82	-11.56	7.75	14.17
0	9	1	19.08	0.00	10.93	10.93
1	9	1	69.89	17.15	58.95	61.40
2	9	1	76.01	57.15	-24.03	62.00
4	9	1	53.73	-26.23	41.17	48.82
5	9	1	34.02	4.69	-35.37	35.67
7	9	1	16.21	6.05	8.30	10.28

h	k	l	|Fo|	A	B	|Fc|
10	9	1	23.00	22.32	-9.21	24.14
0	10	1	111.05	89.78	0.00	89.78
1	10	1	41.35	-3.42	37.50	37.75
2	10	1	61.42	-34.54	39.16	52.21
3	10	1	23.31	-11.71	-21.79	24.74
4	10	1	41.29	-36.61	-21.79	42.60
5	10	1	13.62	5.86	-6.55	8.86
7	10	1	25.05	14.50	22.52	26.78
8	10	1	29.25	17.57	18.47	25.78
9	10	1	22.57	12.50	-21.78	25.11
1	11	1	19.33	-2.32	-21.60	21.73
2	11	1	65.02	-57.35	-15.55	59.50
3	11	1	31.03	-23.61	-17.45	29.35
4	11	1	39.74	38.60	-8.33	39.49
5	11	1	43.03	-25.57	33.96	42.51
6	11	1	18.20	16.75	3.67	17.15
7	11	1	24.30	-1.10	-24.16	24.18
9	11	1	15.57	-13.43	2.03	13.58
10	11	1	16.41	5.38	-6.17	8.15
0	12	1	56.07	-51.76	0.00	51.76
1	12	1	33.18	-32.57	15.53	36.08
2	12	1	24.76	-23.59	10.07	25.37
4	12	1	35.42	35.17	3.80	35.27
5	12	1	53.38	32.71	-32.44	46.07
6	12	1	37.76	-27.27	16.71	31.96
7	12	1	24.98	-18.00	15.25	23.58
9	12	1	27.57	15.86	5.61	16.82
10	12	1	16.32	-9.45	20.38	22.46
11	12	1	21.18	21.15	-14.28	21.60
0	13	1	31.06	0.03	-25.13	25.13
1	13	1	39.21	-20.82	-28.09	34.96
2	13	1	64.09	50.65	27.45	57.60
3	13	1	27.10	23.24	-5.09	23.78
4	13	1	23.22	-21.86	-18.55	28.67
6	13	1	26.15	-22.10	7.92	23.48
7	13	1	15.67	-8.97	10.70	13.96
9	13	1	30.97	26.29	-0.41	26.29
0	14	1	33.69	34.93	0.00	34.93
1	14	1	37.27	21.75	-27.26	34.87
3	14	1	19.22	-7.03	17.70	19.04
5	14	1	24.27	-22.57	0.71	22.58
5	14	1	34.69	-27.25	-14.28	30.76
6	14	1	46.54	43.64	14.55	46.09
7	14	1	28.32	-3.77	-21.96	22.28
10	14	1	22.73	-16.89	2.73	17.10
0	15	1	6.57	0.00	-14.53	14.53
1	15	1	24.91	-1.16	25.59	25.61
2	15	1	16.25	-18.44	-9.10	20.29
3	15	1	18.66	4.11	14.08	14.67
5	15	1	42.00	26.09	-27.39	37.83
6	15	1	23.22	-19.19	-14.03	23.75
0	16	1	15.48	13.49	0.00	13.49
1	16	1	25.09	3.16	20.96	21.20
2	16	1	25.57	7.50	25.50	26.57
3	16	1	42.53	14.76	-34.61	37.63
4	16	1	20.34	-12.23	-11.40	16.72
5	16	1	22.92	-8.88	-21.40	23.17
6	16	1	22.21	0.00	20.23	20.23
1	17	1	44.45	1.49	-44.02	44.04
2	17	1	21.57	-9.25	-16.92	19.25
3	17	1	23.63	-5.42	-23.67	24.28
5	17	1	22.93	-14.2	23.06	23.10
7	17	1	16.39	1.85	3.47	3.94
0	18	1	9.03	-17.21	0.00	17.21
1	18	1	16.49	-6.93	-12.65	14.42
2	18	1	26.77	-2.03	21.48	21.57
3	18	1	17.09	-4.66	19.89	20.42
5	18	1	16.35	15.40	-14.35	20.94
6	18	1	16.23	0.66	-19.45	19.86
1	19	1	15.06	-1.23	15.08	15.13
2	19	1	39.11	38.52	16.05	41.73
3	19	1	15.89	-7.89	13.29	15.46
0	20	1	35.08	33.85	0.00	33.85
1	20	1	13.14	10.63	9.32	14.14
2	20	1	6.64	-13.18	-2.58	13.79
3	20	1	12.41	3.78	-12.61	13.17
0	21	1	24.72	-23.26	4.03	23.60
2	21	1	28.39	-30.02	5.66	30.55
1	23	1	17.53	-11.99	-13.55	17.94
1	24	1	15.08	5.06	-11.50	12.96
3	24	1	18.57	2.14	18.49	18.61
0	0	2	99.19	-59.58	0.00	99.58
1	0	2	10.31	-10.46	0.00	10.46
2	0	2	32.41	22.90	0.00	22.90
3	0	2	55.48	-36.66	0.00	36.66
4	0	2	57.96	56.41	0.30	56.41
5	0	2	18.51	21.01	0.00	21.01
6	0	2	26.24	-28.11	0.00	28.11
8	0	2	38.71	-33.94	0.00	33.94
10	0	2	32.57	-27.88	0.00	27.88
11	0	2	26.93	20.10	0.00	20.10
0	1	2	59.28	54.11	0.00	54.11
1	1	2	102.08	0.00	86.39	86.39
2	1	2	49.75	-71.96	-44.07	82.62
3	1	2	151.58	120.39	-34.44	125.21
4	1	2	61.56	46.46	-4.08	46.64
5	1	2	69.09	-40.35	-33.51	52.64
6	1	2	13.91	-12.46	7.37	14.48
7	1	2	36.16	-30.09	19.74	35.71
8	1	2	54.44	-39.45	8.58	40.38
9	1	2	32.98	28.11	1.87	28.36
3	1	2	34.75	32.35	-4.82	32.73
0	2	2	64.57	59.63	0.00	59.63
1	2	2	69.84	62.61	-21.28	66.13
2	2	2	56.20	-5.13	-43.77	44.07
3	2	2	64.05	50.35	15.58	53.70
4	2	2	48.92	-37.03	23.18	43.70
5	2	2	36.49	-27.62	14.53	31.35
6	2	2	24.96	0.00	20.05	20.05
8	2	2	31.26	-9.01	-18.71	21.17
9	2	2	11.94	-6.45	7.47	9.87
1	2	2	46.97	0.00	30.06	30.06
2	3	2	117.02	28.20	97.40	101.40
2	3	2	73.52	-53.66	61.25	81.25
3	3	2	43.59	-35.18	25.63	43.53
3	3	2	37.58	14.65	35.58	38.75
4	3	2	76.78	-19.07	-72.78	75.24
5	3	2	30.00	13.72	-33.00	35.92
6	3	2	23.96	19.77	8.62	21.57
7	3	2	29.51	19.22	9.33	21.36
8	3	2	36.16	2.32	32.74	32.82
0	4	2	22.21	1.63	19.39	19.46
1	4	2	25.60	-25.32	0.00	25.32
2	4	2	95.71	-60.50	62.99	87.33
3	4	2	11.28	-8.00	-5.46	9.68
4	4	2	27.58	2.21	-25.44	25.53

h	k	l	|Fo|	A	B	|Fc|
5	4	2	74.90	45.39	34.26	56.87
6	4	2	28.34	0.56	-26.50	26.50
7	4	2	28.36	4.51	25.62	26.01
8	4	2	45.93	-23.04	39.25	45.51
11	4	2	29.62	-22.02	-11.20	24.70
0	5	2	6.60	0.00	3.03	3.03
1	5	2	81.69	51.40	-59.52	78.86
2	5	2	53.18	-33.33	-26.45	42.55
3	5	2	50.10	-20.03	-32.20	37.92
4	5	2	72.03	23.17	-61.36	65.59
5	5	2	36.92	-22.12	28.84	36.35
6	5	2	46.48	7.67	47.53	48.14
7	5	2	25.96	23.86	-14.78	28.07
8	5	2	45.55	11.41	-37.78	39.46
10	5	2	2.97	-13.32	0.00	13.32
0	6	2	123.10	-100.75	-55.74	115.14
1	6	2	89.31	-5.21	-79.71	79.88
2	6	2	76.67	-35.42	58.43	68.32
3	6	2	51.07	42.79	24.00	49.07
4	6	2	50.06	38.86	-17.50	42.62
5	6	2	63.07	-18.03	37.82	41.89
6	6	2	34.21	-20.03	-17.93	26.89
7	6	2	96.49	0.00	-91.20	91.20
0	7	2	77.07	-25.96	62.21	67.40
2	7	2	92.71	70.42	17.89	72.66
3	7	2	50.48	47.35	10.50	48.50
4	7	2	61.64	-51.07	15.82	53.46
5	7	2	34.92	-27.76	-16.26	32.17
6	7	2	44.21	-27.20	-37.65	46.45
7	7	2	41.54	-44.89	0.16	44.89
9	7	2	19.96	15.10	11.35	18.89
10	7	2	18.62	-5.35	8.09	9.71
12	7	2	27.89	-22.93	5.28	23.53
0	8	2	40.75	25.92	0.00	25.92
1	8	2	75.67	48.67	-55.53	60.26
2	8	2	72.78	-57.57	5.17	57.79
3	8	2	63.32	9.35	-50.32	51.15
4	8	2	34.17	-0.43	-30.56	30.56
5	8	2	64.47	-60.44	-4.06	60.57
6	8	2	22.51	7.25	22.35	23.50
9	8	2	20.25	23.01	-7.53	24.21
10	8	2	18.73	-17.87	-7.35	19.32
0	9	2	63.46	0.00	-52.22	52.22
1	9	2	32.07	9.57	24.89	26.67
2	9	2	56.42	-39.51	20.33	44.43
3	9	2	49.64	-36.85	30.42	47.78
4	9	2	63.42	-1.71	57.60	57.63
7	9	2	39.21	36.01	-12.55	38.07
8	9	2	41.46	28.73	5.73	29.29
9	9	2	26.33	-13.82	-27.59	30.85
0	10	2	78.53	-67.12	0.00	67.12
1	10	2	24.91	6.99	21.57	22.67
2	10	2	92.23	45.28	71.58	84.78
3	10	2	28.23	-16.95	-20.60	26.92
5	10	2	25.13	20.79	-7.54	22.11
5	10	2	38.67	35.25	7.42	36.05
6	10	2	44.53	-24.69	-39.93	46.95
7	10	2	24.07	7.64	18.63	20.14
8	10	2	23.68	-13.41	-9.76	16.58
9	10	2	18.59	-5.82	-12.91	14.16
0	11	2	56.91	0.00	48.07	48.07
1	11	2	40.32	27.66	-19.80	34.01
2	11	2	29.43	17.59	-11.59	21.06
4	11	2	24.44	-26.93	-10.32	28.84
5	11	2	22.33	15.40	13.74	20.66
6	11	2	22.55	-5.35	27.07	27.59
7	11	2	24.74	-13.22	-15.02	20.46
8	11	2	20.38	11.72	-11.41	16.35
9	11	2	18.75	11.71	-12.98	17.48
0	12	2	5.10	-6.35	0.00	6.35
2	12	2	59.92	10.76	-45.41	46.67
3	12	2	43.12	-14.02	38.03	40.53
4	12	2	35.42	7.01	34.47	35.17
5	12	2	16.41	0.33	-8.97	8.97
6	12	2	24.30	22.75	-3.10	22.95
7	12	2	29.75	30.68	-13.44	33.50
0	13	2	49.24	0.00	-44.59	44.59
1	13	2	40.17	-17.04	35.49	39.36
2	13	2	18.92	19.03	5.56	19.91
3	13	2	20.01	8.62	12.79	15.42
4	13	2	32.10	10.65	22.79	22.03
5	13	2	28.27	-4.85	-32.06	32.42
6	13	2	29.36	-8.67	-25.95	27.35
7	13	2	20.46	-13.53	-2.17	13.71
1	14	2	29.53	19.00	19.07	26.92
3	14	2	40.78	-21.15	38.59	44.02
4	14	2	34.28	9.87	-30.03	32.00
5	14	2	24.92	-27.10	13.71	30.36
6	14	2	28.83	-4.19	18.71	19.07
7	14	2	13.36	3.65	13.98	14.45
8	14	2	28.83	0.00	26.53	26.53
1	15	2	18.37	6.22	-15.05	16.28
2	15	2	5.98	-11.85	1.18	12.00
3	15	2	19.06	-11.34	16.61	20.11
4	15	2	35.35	19.68	-21.49	29.14
1	16	2	24.08	-21.60	6.28	22.50
2	16	2	6.20	7.70	3.54	8.47
3	16	2	13.91	17.44	-11.39	20.83
0	17	2	33.95	-26.71	6.81	27.56
1	17	2	19.28	-15.78	9.77	18.32
2	17	2	24.14	-18.82	-5.56	19.62
3	17	2	27.68	28.35	-9.36	30.44
4	17	2	19.86	-19.74	-5.32	20.44
6	17	2	25.05	-21.64	-8.24	23.15
0	18	2	6.69	6.69	0.00	6.69
1	18	2	21.00	-1.58	21.07	21.07
3	18	2	19.50	18.14	0.10	18.14
5	18	2	21.18	-14.36	-34.39	20.33
2	18	2	21.14	0.00	-22.72	22.72
1	19	2	21.25	9.77	13.14	16.37
0	20	2	15.30	-16.92	-2.32	20.05
2	20	2	11.37	-11.89	0.00	11.89
3	20	2	19.59	-1.25	-19.28	19.32
0	21	2	11.25	-10.00	10.66	10.46
2	21	2	16.78	15.17	-11.55	19.07
4	21	2	28.76	14.13	17.56	22.53
1	0	3	88.92	0.00	-74.00	74.00
2	0	3	162.83	0.00	-149.66	149.66
3	0	3	112.56	0.00	95.89	95.89
4	0	3	11.56	0.00	96.44	96.44
5	0	3	21.64	0.00	20.78	20.78
8	0	3	38.70	0.00	33.67	33.67
1	1	3	35.21	0.00	-25.59	25.59
2	1	3	24.96	0.00	-28.16	28.16
9	0	3	18.99	0.00	-15.75	15.75

Table 2 (*cont.*)

h	k	l	$\|F_o\|$	A	B	$\|F_c\|$
10	0	3	24.60	0.00	9.75	9.75
0	1	3	97.07	0.00	-93.50	93.50
1	1	3	56.83	-52.39	6.79	52.82
2	1	3	34.40	-28.84	15.71	32.84
3	1	3	44.10	33.27	-3.60	33.46
4	1	3	70.39	-2.10	62.66	62.69
5	1	3	29.31	-9.35	-28.08	29.60
6	1	3	40.16	9.81	-38.72	39.94
7	1	3	34.37	-20.92	20.96	29.62
10	1	3	28.39	-8.23	31.77	32.82
11	1	3	17.89	16.79	-4.10	17.29
0	2	3	20.50	21.85	0.00	21.85
1	2	3	106.37	93.26	-37.28	100.43
2	2	3	34.67	13.90	29.49	32.60
3	2	3	8.79	-2.21	-8.11	8.40
4	2	3	47.46	-16.84	-33.11	37.14
5	2	3	55.52	-49.41	-0.09	49.41
7	2	3	18.24	-12.45	-3.09	12.82
8	2	3	25.13	11.92	24.10	26.89
10	2	3	22.51	12.17	-0.07	12.17
0	3	3	74.26	0.00	62.89	62.89
1	3	3	106.41	76.78	15.57	78.39
2	3	3	83.30	-33.67	-61.67	70.27
3	3	3	93.24	-64.39	29.65	71.07
4	3	3	36.71	19.80	-12.00	23.15
6	3	3	31.86	1.55	21.33	21.39
7	3	3	45.20	33.17	-12.38	35.41
8	3	3	23.68	-16.65	-0.21	16.65
11	3	3	34.35	-20.78	14.57	25.39
0	4	3	101.55	-87.43	0.00	87.43
1	4	3	76.20	-62.97	11.60	64.03
2	4	3	40.32	7.54	31.35	32.24
3	4	3	19.67	-9.79	-7.89	12.57
4	4	3	52.31	37.42	-3.53	37.58
5	4	3	49.09	45.69	-0.35	45.69
6	4	3	40.34	-29.08	-14.21	32.36
7	4	3	22.17	-15.17	-10.35	18.36
8	4	3	22.21	-19.50	-9.00	21.48
9	4	3	29.07	-19.29	20.54	28.18
11	4	3	17.91	12.71	8.35	15.20
0	5	3	36.91	0.00	33.62	33.62
1	5	3	37.46	-35.29	-0.72	35.29
2	5	3	24.05	-8.96	12.63	15.49
3	5	3	82.85	66.37	14.08	67.85
4	5	3	41.36	-25.25	-32.58	41.22
5	5	3	10.11	-0.42	11.21	11.21
6	5	3	37.62	24.10	4.42	24.50
7	5	3	29.58	-31.76	-15.79	35.47
8	5	3	26.02	22.96	-11.36	25.62
9	5	3	21.84	23.64	1.78	23.70
10	5	3	17.82	-4.53	-3.35	5.64
12	5	3	14.31	10.20	-10.82	14.86
0	6	3	28.56	-27.85	0.00	27.85
1	6	3	56.80	43.69	26.60	51.15
2	6	3	71.94	2.80	-59.63	59.69
3	6	3	62.10	14.78	45.47	50.67
4	6	3	38.04	24.70	13.00	27.91
5	6	3	46.89	-25.76	26.50	37.03
6	6	3	32.25	24.70	22.46	33.39
7	6	3	34.56	27.14	-11.10	29.32
8	6	3	26.42	-12.32	-22.39	25.55
9	6	3	17.13	12.68	-0.11	12.68
10	6	3	17.75	-11.57	4.67	12.47
11	6	3	17.89	-19.75	8.89	21.66
0	7	3	128.56	0.00	-115.89	115.89
1	7	3	63.34	52.50	-1.28	52.51
2	7	3	72.27	23.06	54.29	58.98
3	7	3	72.20	-50.94	-11.19	52.15
4	7	3	59.86	8.00	51.20	51.82
5	7	3	30.77	-9.15	-19.88	21.88
6	7	3	34.24	17.43	-25.14	30.59
7	7	3	23.50	17.40	-2.92	17.64
8	7	3	21.29	-18.12	-2.88	18.35
10	7	3	17.82	10.61	13.65	17.29
1	8	3	34.51	-28.51	3.89	28.77
2	8	3	35.86	3.10	31.36	31.51
3	8	3	31.45	-16.56	-25.66	30.53
4	8	3	58.29	18.35	-50.36	53.60
6	8	3	12.07	-4.52	12.75	13.53
8	8	3	23.68	6.50	23.15	24.04
10	8	3	17.89	14.45	-6.19	15.71
12	8	3	13.89	-0.16	-14.22	14.22
0	9	3	57.13	0.00	50.57	50.57
2	9	3	33.85	2.53	-32.96	33.05
3	9	3	19.67	-0.34	10.69	10.69
4	9	3	32.78	11.89	-31.32	33.50
5	9	3	11.70	-11.68	14.34	18.50
6	9	3	30.60	-5.39	26.94	27.47
7	9	3	22.93	25.55	-8.67	26.98
8	9	3	29.58	-13.05	23.04	26.48
10	9	3	23.17	-8.53	-20.57	22.26
0	10	3	24.01	-24.64	0.00	24.64
1	10	3	42.13	-12.60	-37.54	39.60
2	10	3	37.53	-4.75	-35.25	35.57
3	10	3	17.20	-12.60	4.34	13.32
4	10	3	23.04	5.42	19.06	19.81
5	10	3	28.67	24.20	-9.22	25.89
6	10	3	36.54	-25.87	16.89	30.89
7	10	3	25.42	-16.48	-13.52	21.50
8	10	3	22.40	-12.55	-7.37	14.55
9	10	3	20.60	-14.81	13.39	19.96
0	11	3	43.35	0.00	-45.78	45.78
1	11	3	40.75	-37.51	7.68	38.28
2	11	3	17.00	-7.25	5.27	8.96
3	11	3	40.69	34.64	7.96	35.54
4	11	3	36.17	-1.24	34.17	34.18
6	11	3	26.54	-21.82	-15.75	26.90
7	11	3	31.10	-35.23	10.65	36.80
8	11	3	17.58	5.47	-7.08	8.95
0	12	3	39.21	41.55	0.00	41.55
1	12	3	34.24	28.36	12.52	31.00
2	12	3	27.26	5.22	20.87	21.51
4	12	3	23.63	-20.65	10.57	23.20
5	12	3	24.60	-27.37	-9.28	28.90
6	12	3	21.65	-1.53	-14.84	14.92
7	12	3	22.42	29.52	-2.35	29.61
1	13	3	36.12	33.47	7.16	34.23
2	13	3	25.05	-26.07	-0.57	26.07
3	13	3	75.42	-69.87	-12.85	71.04
4	13	3	20.56	8.00	28.33	29.44
5	13	3	23.56	23.39	-8.60	24.92
7	13	3	26.97	26.02	-7.57	27.10
8	13	3	25.32	-6.56	-21.20	22.19
9	13	3	17.82	-16.64	8.00	18.46
0	14	3	26.71	-33.05	0.00	33.05
1	14	3	51.12	-44.51	14.62	46.85
2	14	3	16.45	3.17	18.45	18.71
4	14	3	21.00	24.89	7.51	26.00
5	14	3	26.17	22.42	-10.93	24.94
5	14	3	24.53	2.57	-30.94	31.04
8	14	3	14.62	3.67	12.07	12.61
0	15	3	29.10	0.00	26.16	26.16
2	15	3	16.35	7.01	-13.65	15.35
3	15	3	23.14	21.00	-6.89	22.10
4	15	3	26.18	0.82	-35.64	35.64
5	15	3	29.09	16.37	20.92	26.57
0	16	3	5.14	-6.35	0.00	6.35
1	16	3	27.56	2.80	-24.22	24.38
2	16	3	24.00	-3.58	-18.29	18.64
3	16	3	27.83	-20.87	21.70	30.10
5	16	3	22.99	-2.89	25.38	25.54
8	16	3	20.25	-9.22	-19.35	21.43
0	17	3	33.93	0.00	-32.71	32.71
1	17	3	23.39	-21.14	10.90	23.78
3	17	3	27.78	23.90	12.38	26.92
6	17	3	20.67	5.53	-20.14	20.88
0	18	3	23.46	23.63	0.00	23.63
1	18	3	20.32	19.95	9.96	22.30
2	18	3	31.03	5.75	28.66	29.23
4	18	3	24.10	-7.93	-26.53	27.69
0	19	3	24.80	0.00	28.70	28.70
3	19	3	15.19	-13.82	6.25	15.17
0	20	3	21.40	-20.67	0.00	20.67
1	20	3	13.00	-9.53	1.75	9.69
5	20	3	19.74	19.45	-9.98	21.85
0	23	3	17.38	0.00	-15.24	15.24
0	25	3	10.24	0.00	13.20	13.20
0	0	4	52.16	-47.50	0.00	47.50
1	0	4	36.28	38.47	0.00	38.47
2	0	4	45.20	36.71	0.00	36.71
3	0	4	62.29	46.86	0.00	46.86
4	0	4	36.07	24.19	0.00	24.19
5	0	4	91.38	-76.15	0.00	76.15
7	0	4	43.70	37.13	0.00	37.13
9	0	4	22.48	22.92	0.00	22.92
10	0	4	19.75	-17.97	0.00	17.97
11	0	4	21.54	-18.63	0.00	18.63
0	1	4	26.82	0.00	19.64	19.64
1	1	4	46.21	18.56	-34.17	38.88
2	1	4	22.21	18.33	6.96	19.60
3	1	4	109.60	-89.19	1.25	89.20
4	1	4	17.13	-13.90	0.27	13.90
5	1	4	10.44	-4.99	7.78	9.25
6	1	4	21.29	-0.68	-19.86	19.87
7	1	4	29.58	26.11	8.66	27.51
9	1	4	40.83	-24.55	-27.78	37.07
1	2	4	48.89	-42.85	-4.01	43.03
2	2	4	52.94	17.44	44.50	47.79
4	2	4	44.64	-9.26	-37.46	38.59
5	2	4	54.73	44.71	10.67	45.96
7	2	4	23.56	-23.35	-5.03	23.88
8	2	4	43.30	5.33	37.59	37.96
9	2	4	40.94	-4.28	-27.12	27.46
10	2	4	23.41	1.81	15.92	16.02
11	2	4	17.78	13.47	1.70	13.57
0	3	4	50.54	0.00	51.17	51.17
1	3	4	25.25	-22.81	1.50	22.85
2	3	4	19.39	10.88	-12.49	16.56
3	3	4	39.04	-9.62	-30.09	31.59
4	3	4	40.21	0.57	-37.57	37.57
6	3	4	55.61	-4.31	52.82	53.00
7	3	4	19.56	-12.35	5.05	13.36
8	3	4	21.42	0.64	18.18	18.19
9	3	4	19.14	11.23	11.11	15.80
0	4	4	55.56	41.71	0.00	41.71
1	4	4	43.56	23.17	-15.36	27.80
2	4	4	62.33	-15.78	-57.58	59.71
3	4	4	6.70	-4.78	3.50	5.92
4	4	4	63.50	-36.09	37.32	51.91
6	4	4	33.67	-10.35	28.96	30.75
7	4	4	22.58	-14.60	0.85	14.62
8	4	4	44.83	9.84	-40.41	41.59
10	4	4	19.56	13.61	2.38	13.82
0	5	4	78.52	0.00	-72.28	72.28
1	5	4	12.49	16.38	-3.17	16.67
2	5	4	15.72	-12.64	-1.61	12.74
3	5	4	24.68	16.74	1.81	16.83
4	5	4	42.95	15.35	31.82	35.32
5	5	4	27.15	4.07	27.01	27.32
6	5	4	43.70	-5.94	-41.42	41.85
7	5	4	15.50	-10.90	-11.99	16.20
10	5	4	21.82	-0.55	16.56	16.57
0	6	4	55.21	41.33	0.00	41.33
1	6	4	66.93	55.32	2.54	55.37
2	6	4	50.93	-21.04	39.21	44.50
3	6	4	4.30	6.78	0.28	6.78
4	6	4	36.96	-12.56	-26.98	29.75
5	6	4	56.98	-45.83	-13.69	47.83
6	6	4	29.47	-3.72	-24.59	24.86
7	6	4	15.53	14.38	-4.53	15.07
8	6	4	36.60	-4.32	35.24	35.50
9	6	4	30.32	8.96	24.61	26.19
10	6	4	15.41	-1.82	-18.53	18.61
0	7	4	31.11	0.00	30.82	30.82
1	7	4	61.07	43.20	-28.95	52.00
2	7	4	6.39	-9.57	-3.66	10.25
3	7	4	68.43	-60.16	-0.64	60.16
4	7	4	37.60	-17.42	-22.78	28.67
5	7	4	21.14	-7.53	14.21	16.07
6	7	4	18.50	1.64	19.53	19.60
7	7	4	30.18	28.91	6.10	29.54
8	7	4	24.01	11.00	-16.39	19.74
10	7	4	23.59	2.61	-17.32	17.51
1	8	4	63.53	-57.30	-0.77	57.30
2	8	4	13.40	-16.04	3.33	16.38
3	8	4	15.06	-10.54	13.00	16.74
4	8	4	8.76	8.05	6.32	10.23
5	8	4	56.64	49.28	8.82	50.07
6	8	4	13.47	6.23	-6.54	9.10
8	8	4	8.56	3.46	-6.99	7.80
9	8	4	23.48	-17.63	-15.41	23.41
10	8	4	12.57	-8.03	6.04	10.05
0	9	4	39.83	0.00	36.71	36.71
1	9	4	29.58	-23.27	-4.54	23.78
2	9	4	14.33	-1.46	13.35	13.43
3	9	4	55.90	48.71	-13.63	50.57
4	9	4	21.57	-4.87	-14.35	15.16
6	9	4	22.66	19.42	-0.20	19.42
7	9	4	11.86	-10.27	2.54	10.57
8	9	4	17.42	-1.75	19.22	19.30
9	9	4	19.92	18.99	-5.11	19.66
1	10	4	24.23	13.77	-13.91	19.57
2	10	4	51.54	6.61	-42.06	42.57
3	10	4	20.74	7.45	11.85	14.00
5	10	4	37.65	-35.47	10.57	37.01
5	10	4	26.00	3.02	24.64	24.82
9	10	4	23.59	16.88	-10.52	19.94
0	11	4	75.34	0.00	-74.07	74.07
2	11	4	15.44	-9.83	-2.51	10.17
3	11	4	22.24	-7.34	-22.22	23.40
4	11	4	34.24	1.47	35.46	35.49
5	11	4	18.54	6.69	-13.91	15.43
7	11	4	26.15	24.16	6.17	24.93
9	11	4	17.78	-16.14	-10.25	19.11
0	12	4	13.58	11.30	0.00	11.30
1	12	4	30.67	9.90	-33.11	34.56
2	12	4	59.92	18.14	54.74	57.66
3	12	4	7.21	0.71	8.69	8.71
4	12	4	32.51	-16.21	-25.41	30.14
5	12	4	22.09	5.17	19.00	19.68
6	12	4	8.56	3.21	-14.61	14.96
7	12	4	8.79	-16.05	-4.63	16.73
8	12	4	21.54	4.29	20.46	20.90
0	13	4	61.47	0.00	56.62	56.62
1	13	4	17.38	3.96	15.89	16.37
2	13	4	21.92	-13.62	-14.64	19.99
3	13	4	29.07	-21.28	19.27	28.71
4	13	4	23.37	-2.34	-22.82	22.94
5	13	4	24.50	-5.17	24.03	24.57
1	14	4	19.42	-12.97	13.16	18.48
2	14	4	37.03	1.75	-32.10	32.14
4	14	4	28.19	-15.50	29.89	33.67
5	14	4	21.42	17.86	-8.33	19.71
7	14	4	8.89	13.42	-8.25	15.75
8	14	4	15.25	1.82	-13.97	14.09
1	15	4	20.65	-16.61	-7.27	18.13
2	15	4	14.78	-12.78	11.70	17.32
3	15	4	21.00	14.89	13.13	19.85
4	15	4	17.65	0.60	15.77	15.78
5	15	4	15.41	11.01	-12.04	16.32
7	15	4	19.53	-22.34	0.64	22.35
0	15	4	21.18	-23.57	0.00	23.57
1	16	4	29.02	22.67	4.58	23.13
2	16	4	20.12	-14.95	-1.78	15.08
4	16	4	17.75	14.25	5.97	15.45
5	16	4	29.58	-29.10	-1.38	29.14
0	17	4	24.07	0.00	-25.40	25.40
1	17	4	20.67	17.85	-1.20	17.89
3	17	4	24.17	-24.52	2.14	24.61
1	18	4	22.09	-20.59	-11.70	23.67
2	18	4	14.95	-5.35	9.07	10.53
0	19	4	19.50	0.00	17.03	17.03
3	19	4	12.68	19.22	-3.05	19.46
2	20	4	18.50	3.89	-19.53	19.91
0	21	4	10.55	0.00	-6.00	6.00
1	0	5	29.53	0.00	-21.10	21.10
2	0	5	46.13	0.00	40.47	40.47
3	0	5	22.57	0.00	24.37	24.37
4	0	5	40.25	0.00	-32.68	32.68
5	0	5	17.98	0.00	10.96	10.96
6	0	5	31.56	0.00	-23.64	23.64
9	0	5	31.90	0.00	25.40	25.40
12	0	5	22.59	0.00	-16.49	16.49
0	1	5	52.54	0.00	48.25	48.25
1	1	5	52.57	37.20	30.92	48.37
2	1	5	19.74	10.82	-15.78	19.14
3	1	5	36.76	-23.53	12.17	26.49
4	1	5	38.28	-9.64	-31.01	32.47
5	1	5	38.33	0.38	-24.28	24.28
6	1	5	37.35	-2.32	25.83	25.93
10	1	5	32.35	-6.42	-22.12	23.03
0	2	5	17.25	20.54	0.00	20.54
1	2	5	24.54	-18.34	6.88	19.58
2	2	5	72.69	22.19	-55.00	59.30
3	2	5	42.13	-31.46	-27.60	41.85
4	2	5	27.03	11.18	17.12	20.45
5	2	5	46.60	32.09	0.07	32.09
6	2	5	34.56	16.09	18.31	24.37
10	2	5	30.00	-17.64	-0.93	17.66
0	3	5	62.06	0.00	-56.62	56.62
1	3	5	50.32	-36.94	20.87	42.42
2	3	5	38.02	-11.17	25.78	28.10
3	3	5	41.25	34.80	-1.26	34.82
4	3	5	26.66	-9.17	9.77	13.39
5	3	5	18.55	-9.92	7.96	12.72
6	3	5	20.38	2.34	-12.80	13.01
7	3	5	26.99	-22.96	-7.06	24.01
9	3	5	32.08	21.13	-20.63	29.53
0	4	5	52.72	-45.23	0.00	45.23
1	4	5	52.39	47.03	3.31	47.14
2	4	5	29.84	-7.08	21.14	22.28
3	4	5	21.24	8.65	11.23	14.17
4	4	5	51.07	40.94	-12.30	42.75
5	4	5	38.04	-37.57	-3.18	37.70
7	4	5	24.92	1.67	-13.25	13.35
9	4	5	29.50	17.89	-12.57	21.86
0	5	5	59.45	0.00	-48.95	48.95
1	5	5	19.17	14.39	2.16	14.54
2	5	5	57.66	46.52	0.25	46.52
3	5	5	51.15	-32.99	-24.24	40.93
4	5	5	28.05	-8.92	22.64	24.32
6	5	5	27.97	-16.76	-4.55	17.36
7	5	5	37.46	23.67	-15.44	28.26
9	5	5	27.30	-9.58	9.48	13.48
0	6	5	56.91	45.65	0.00	45.65
1	6	5	77.35	-66.00	-10.82	66.88
2	6	5	57.00	-16.46	46.30	49.14
3	6	5	36.35	-16.35	26.26	30.93
4	6	5	36.07	-27.23	-18.53	32.93
5	6	5	28.52	21.59	3.47	21.86
6	6	5	26.51	12.08	-21.46	24.63
9	6	5	24.43	-11.18	12.53	17.02
0	7	5	47.56	0.00	38.50	38.50
1	7	5	23.37	-14.03	13.32	19.34
2	7	5	33.35	-2.03	-26.83	26.90
3	7	5	49.47	40.48	16.97	43.89
4	7	5	21.54	14.38	-10.44	17.77
5	7	5	18.00	-11.96	-10.18	15.71
6	7	5	31.28	-5.97	33.98	34.50
1	8	5	49.47	26.94	30.85	40.96
2	8	5	49.31	-0.07	-47.23	47.23
3	8	5	56.72	-6.93	-44.08	44.62
4	8	5	34.15	-6.29	26.89	27.62
5	8	5	35.64	-3.27	-28.03	28.22
7	8	5	39.21	15.08	28.75	32.46
0	9	5	16.67	0.00	-12.46	12.46
1	9	5	37.43	-7.98	-30.07	31.11
2	9	5	21.09	-1.46	20.51	20.85
3	9	5	22.29	9.46	16.59	19.10
4	9	5	23.00	9.96	18.50	21.01
5	9	5	25.59	-7.05	-20.27	21.46
7	9	5	20.72	-17.24	8.93	19.41
0	10	5	30.56	-26.42	0.00	26.42
1	10	5	39.36	[illegible]	7.54	34.12
2	10	5	42.26	0.70	43.81	43.81
4	10	5	9.71	-4.62	-13.04	13.83
5	10	5	25.47	-23.60	-8.07	24.93
0	11	5	30.73	0.00	24.50	24.50
2	11	5	22.59	10.14	-12.13	15.81
3	11	5	28.39	-26.99	-5.38	27.52
4	11	5	13.31	-11.76	-6.51	13.44
5	11	5	26.17	28.53	-5.48	29.05
0	12	5	36.52	36.14	0.00	36.14
1	12	5	43.52	-42.48	4.31	42.69
4	12	5	17.31	-15.77	9.25	18.28
5	12	5	23.85	16.42	13.52	21.34
6	12	5	21.03	21.93	-9.28	23.81
1	13	5	7.70	-11.77	-0.50	11.78
3	13	5	36.38	24.67	-23.12	33.82
4	13	5	15.11	7.92	-12.10	14.46
1	14	5	25.51	22.23	-3.62	22.52
2	14	5	26.92	17.64	-18.36	25.46
3	14	5	22.62	5.50	-20.89	21.60
0	15	5	8.32	0.00	-5.53	5.53
1	15	5	13.14	9.20	-2.46	9.52
2	15	5	8.54	-2.77	9.29	9.69
3	15	5	15.22	-10.00	-4.84	11.10
2	16	5	24.43	-4.42	21.50	21.95
0	17	5	26.88	0.00	27.29	27.29
1	17	5	10.64	0.65	6.14	6.17
2	18	5	16.74	-9.54	-14.56	17.66
0	19	5	10.73	0.00	-10.42	10.42
1	19	5	16.00	-8.26	-5.37	9.85
1	20	5	9.65	5.45	-8.64	10.21
0	0	6	61.99	53.03	0.00	53.03
1	0	6	30.02	-32.02	0.00	32.02
2	0	6	43.79	-40.63	0.00	40.63
4	0	6	31.56	-24.39	0.00	24.39
5	0	6	29.75	23.96	0.00	23.96
6	0	6	27.74	22.11	0.00	22.11
7	0	6	14.82	-7.29	0.00	7.29
1	1	6	15.50	-17.14	4.58	17.74
2	1	6	39.53	-35.13	6.11	35.65
3	1	6	32.57	27.67	4.61	28.04
4	1	6	34.59	21.24	-10.37	23.63
5	1	6	32.41	-10.67	-21.18	23.71
6	1	6	24.07	17.28	9.09	19.52
7	1	6	27.78	-24.10	-2.33	24.21
9	1	6	24.59	3.24	17.73	18.02
0	2	6	27.33	-31.99	0.00	31.99
1	2	6	16.54	7.96	9.65	12.51
2	2	6	34.26	12.28	-27.28	29.92
3	2	6	32.45	-4.43	-23.09	23.51
4	2	6	33.20	24.46	4.13	24.80
5	2	6	26.17	-20.54	3.49	20.83
7	2	6	44.72	13.25	35.98	38.34
0	3	6	36.52	0.00	-34.87	34.87
1	3	6	4.52	6.41	-6.75	9.31
2	3	6	38.54	23.53	20.87	31.45
3	3	6	16.35	-9.73	-8.57	12.96
4	3	6	28.43	-8.28	30.43	31.53
5	3	6	44.28	2.28	30.58	30.96
6	3	6	28.25	2.53	-18.18	18.35
9	3	6	29.43	-11.81	-13.53	17.96
0	4	6	4.35	-7.67	0.00	7.67
1	4	6	25.64	18.27	-10.35	21.00
2	4	6	29.14	17.17	16.25	23.64
3	4	6	29.60	4.85	24.24	24.71
4	4	6	17.29	7.78	-12.96	15.11
7	4	6	38.57	-15.82	-21.96	27.06
8	4	6	27.03	-1.88	19.81	19.89
0	5	6	15.15	0.00	15.55	15.85
1	5	6	40.21	9.83	38.57	39.80
2	5	6	24.27	6.36	-22.51	23.39
3	5	6	23.85	-20.86	2.32	20.99
4	5	6	22.58	-18.67	-19.53	27.23
5	5	6	27.30	3.96	-22.71	23.04
0	6	6	4.63	6.29	0.00	6.29
1	6	6	33.78	-18.46	21.35	28.22
2	6	6	7.04	-5.53	9.64	11.12
3	6	6	34.70	-3.60	-33.58	33.77
4	6	6	23.10	-17.71	17.34	24.78
5	6	6	19.81	-0.21	15.08	15.08
6	6	6	31.35	22.75	-1.92	22.82
8	6	6	19.22	1.30	-13.91	13.96
9	6	6	22.07	-15.01	-10.06	18.07
1	7	6	42.71	-20.72	-36.38	41.86
2	7	6	45.84	-31.88	-18.29	36.75
3	7	6	30.67	18.78	7.67	20.28
4	7	6	27.61	16.45	12.17	20.46
6	7	6	18.42	12.19	-13.14	17.92
7	7	6	19.04	-13.99	-8.29	16.26
0	8	6	27.00	-26.57	0.00	26.57
1	8	6	4.43	-4.21	-3.00	5.17
2	8	6	15.86	14.70	5.75	15.78
4	8	6	21.09	16.57	-12.70	20.87
5	8	6	25.47	-18.96	-6.07	19.90
7	8	6	33.18	24.31	11.81	27.02
1	9	6	25.14	25.90	-8.19	27.16
2	9	6	28.07	23.51	9.09	25.21
3	9	6	7.10	-10.59	-2.26	11.12
4	9	6	21.25	-5.86	15.60	16.66
6	9	6	15.46	-15.77	4.40	16.37
7	9	6	15.67	15.00	6.60	16.39
0	10	6	27.55	21.61	0.00	21.61
1	10	6	22.22	-9.42	-19.65	21.79
2	10	6	18.17	-14.99	3.32	15.35
3	10	6	21.98	-6.15	16.53	17.63
4	10	6	18.53	-4.98	-17.04	17.75
5	10	6	26.57	24.92	-4.60	25.34
0	11	6	26.67	0.00	22.44	22.44
1	11	6	42.50	-2.32	-21.60	21.73
2	11	6	18.92	-17.02	7.71	18.68
3	11	6	20.16	11.92	14.31	18.62
4	11	6	18.64	14.41	-18.75	23.65
5	11	6	15.72	-7.63	8.37	11.32
0	12	6	8.94	-10.45	0.00	10.45
1	12	6	24.63	6.72	24.43	25.34
2	12	6	7.10	-0.89	-10.19	10.23
3	12	6	20.38	3.16	-19.43	19.68
4	12	6	15.52	-1.82	15.53	15.63
0	13	6	29.64	0.00	-30.90	30.90
1	13	6	29.27	-18.84	-22.98	29.71
3	13	6	17.57	-0.27	-14.64	14.64
5	13	6	19.22	8.36	16.68	18.66
0	14	6	13.89	-9.39	0.00	9.39
1	14	6	9.71	3.05	-7.85	8.43
2	14	6	10.57	4.73	19.03	19.60
1	15	6	15.72	13.24	6.28	14.65
2	15	6	26.67	26.68	-5.06	27.15
0	16	6	13.49	15.07	0.00	15.07
1	16	6	14.66	-14.96	0.28	14.96

Table 2 (cont.)

h	k	l	|Fo|	A	B	|Fc|	h	k	l	|Fo|	A	B	|Fc|	h	k	l	|Fo|	A	B	|Fc|	h	k	l	|Fo|	A	B	|Fc|
1	17	6	10.06	-7.76	6.10	9.87	4	5	7	37.29	27.14	16.12	31.56	3	16	7	18.20	1.07	-12.39	12.42	4	8	9	20.05	-21.17	2.62	21.33
0	18	6	12.57	-14.10	0.00	14.10	0	6	7	22.59	-20.38	0.00	20.38	1	17	7	14.03	8.10	-12.77	15.12	2	9	8	23.14	-22.46	-0.49	22.46
0	21	6	14.39	0.00	15.00	15.00	1	6	7	23.45	-0.29	23.36	23.36	0	18	7	10.51	-11.57	0.00	11.57	3	10	8	18.46	-18.57	0.00	18.57
1	0	7	34.00	0.00	31.56	31.55	3	6	7	21.51	8.05	-9.89	12.75	0	20	7	10.18	11.07	0.00	11.07	1	10	8	9.96	6.67	4.37	7.98
0	1	7	15.67	0.00	-20.08	20.08	4	6	7	18.54	12.01	12.39	17.25	0	0	8	32.16	-27.52	0.00	27.52	1	11	8	13.52	3.79	-13.46	13.96
1	1	7	43.82	9.87	-42.13	43.27	0	7	7	24.10	0.00	-20.92	20.92	1	1	8	22.71	-19.89	-10.50	22.40	1	12	8	13.43	2.40	-13.53	14.03
3	1	7	7.48	-9.05	-6.27	11.00	1	7	7	13.13	-0.49	-13.79	13.80	2	1	8	29.84	27.14	-2.78	27.28	1	13	8	11.15	-2.13	13.57	13.74
0	2	7	24.85	-21.00	0.00	21.00	3	7	7	9.82	-11.44	-17.05	20.53	4	1	8	27.97	-20.28	-3.21	20.53	0	14	8	9.24	10.05	0.00	10.05
1	2	7	16.63	-4.92	-19.37	19.99	1	8	7	22.81	4.69	-19.01	19.58	0	2	8	25.40	27.42	0.00	27.42	0	16	8	9.92	-13.31	0.00	13.31
3	2	7	13.75	-16.28	19.82	25.64	2	8	7	24.50	3.55	18.14	18.49	2	2	8	14.88	-9.46	11.32	14.75	0	2	9	20.79	20.31	0.00	20.31
4	2	7	32.10	27.04	-8.52	28.44	3	8	7	13.16	-10.38	18.03	20.81	1	3	8	26.18	-0.17	21.02	21.02	1	2	9	16.94	10.69	9.17	14.08
1	3	7	14.89	13.46	-9.37	16.40	1	9	7	24.05	-2.64	19.28	19.46	2	3	8	12.12	-10.52	0.42	10.53	2	3	9	19.00	-14.10	-7.59	16.01
2	3	7	33.03	27.50	3.23	27.68	2	9	7	14.97	4.10	-9.28	10.14	1	4	8	13.05	-1.88	11.94	12.08	0	4	9	14.46	-8.35	0.00	8.35
3	3	7	9.71	-13.50	6.35	14.92	3	10	7	20.45	-16.82	-16.17	23.34	1	5	8	15.92	0.00	13.09	13.09	1	5	9	9.47	-5.64	-10.42	11.85
0	4	7	46.26	42.10	0.00	42.10	2	11	7	24.21	-22.32	8.20	23.77	2	5	8	30.02	3.60	-26.04	26.28	2	5	9	13.10	13.47	1.91	13.60
1	4	7	28.05	-26.67	-12.17	29.31	3	11	7	7.46	13.42	4.39	14.11	2	5	8	14.75	-10.27	0.17	10.27	2	6	9	12.59	-1.25	-12.05	12.11
2	4	7	30.86	-7.60	-25.71	26.81	0	12	7	20.92	-21.07	0.00	21.07	4	5	8	23.26	9.78	-14.53	17.60	0	7	9	18.25	0.00	-16.21	16.21
3	4	7	28.17	0.82	17.00	17.02	3	12	7	10.40	2.42	5.87	6.35	0	6	8	15.83	-14.50	0.00	14.50	1	7	9	14.46	4.31	10.16	11.03
4	4	7	34.71	-28.75	-12.70	31.43	2	13	7	21.96	16.21	3.52	16.59	1	6	8	12.90	-0.44	-12.96	12.96	2	7	9	10.24	-1.04	4.44	4.56
1	5	7	21.23	-2.69	20.20	20.37	3	14	7	15.21	11.45	0.00	11.48	2	6	8	16.94	5.73	-10.11	11.62	2	8	9	10.03	0.26	15.21	15.21
2	5	7	38.44	-38.48	2.14	38.53	3	14	7	16.19	13.07	7.97	15.31	2	7	8	18.78	16.26	1.74	16.35	0	10	9	12.54	-10.92	0.00	10.92
3	5	7	7.55	10.75	1.96	10.92	1	16	7	14.52	2.22	14.39	14.55	0	8	8	35.70	32.21	0.00	32.21	0	12	9	11.30	13.92	0.00	13.92

(a)

carbon

oxygen

bromine
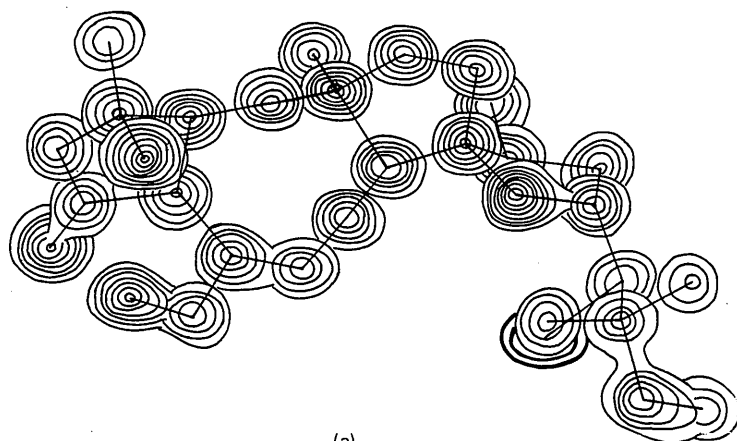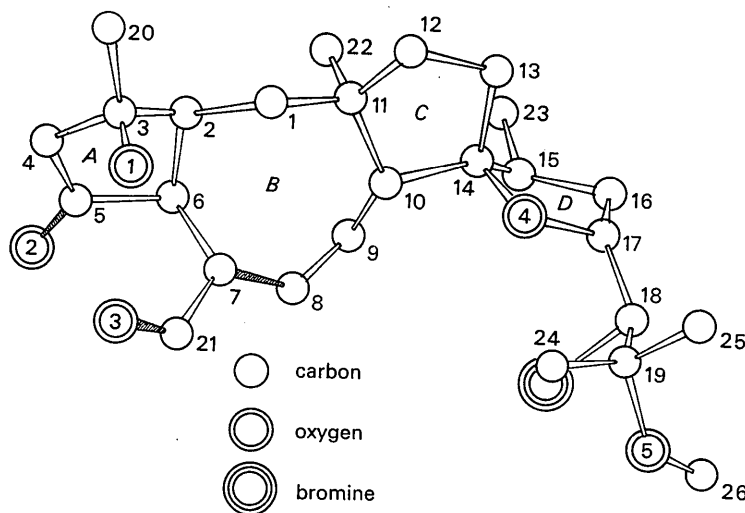
(b)

Fig. 2. (a) Composite drawing of the final electron density map viewed along the b axis. (b) Molecular structure in correct absolute configuration.

final Fourier synthesis is shown in Fig. 2 together with the perspective drawing of the molecule.

Absolute configuration

The absolute configuration was determined by the method of anomalous dispersion (Bijvoet, Peerdeman & van Bommel, 1951). The dispersion corrections of the bromine scattering factor for Cu $K\alpha$ radiation, $\Delta f' = -0.9$ and $\Delta f'' = 1.5$ (Dauben & Templeton, 1955), were applied and the structure factors for the Friedel pairs of reflexions were calculated by assuming that the atomic parameters given in Table 1 are those referred to a right-handed set of axes. The coordinate

system for space group $P2_12_12_1$ is taken from *International Tables for X-ray Crystallography* (1952). Of the 22 pairs for which the intensity difference between hkl and $h\bar{k}l$ reflexions was expected to be discernible, 21 pairs showed significant differences in the lth layer Weissenberg photographs. These results are listed in Table 3. A comparison between the observed and calculated intensities indicated that the assumed absolute configuration was really the correct one. All Figures shown in this paper are drawn with the correct absolute configuration.

Table 3. *Comparison of the observed and calculated intensity differences used for the establishment of absolute configuration*

h	k	l	$F_c(hkl)/F_c(h\bar{k}l)$	$I_o(hkl)/I_o(h\bar{k}l)$
4	1	1	0·624	<1
7	4	1	0·824	?
7	5	1	0·687	<1
11	5	1	0·744	<1
2	4	2	1·477	>1
3	4	2	2·857	>1
8	6	2	0·736	<1
6	2	2	0·749	<1
5	1	2	0·805	<1
2	1	3	0·799	<1
2	2	3	1·288	>1
3	2	3	1·557	>1
9	3	3	0·561	<1
3	4	3	0·774	<1
4	6	3	1·319	>1
4	1	4	0·746	<1
2	3	4	0·772	<1
2	5	4	0·668	<1
7	1	5	0·810	<1
4	2	5	1·416	>1
4	3	5	0·662	<1
7	4	5	1·278	>1

Discussion of the structure

The molecular structure

The molecular structure of ophiobolin methoxybromide revealed by the present analysis is shown in Fig. 1(*b*). The structure proposed for ophiobolin is therefore that shown in Fig. 1(*a*). It is essentially tricyclic and consists of five-, eight- and five-membered rings forming a novel type of ring system which has never been found in naturally occurring compounds. The juncture between the cyclopentanone ring (*A* ring) and the eight-membered *B* ring is in the *cis* form and that between *B* and *C* is in the *trans* form. A side chain, 2-isopropylidene-4-methyltetrahydrofuran, is attached to ring *C* in a *spiro* form at the 5-position of tetrahydrofuran (*D* ring). A biogenetic study of ophiobolin (Canonica, Fiecchi, Kienle, Ranzi & Scala, 1966) has shown that the ring *D* is formed after the tricyclic ring system (*A*, *B* and *C*) has been completed.

Nozoe, Morisaki, Tsuda, Iitaka, Takahashi, Tamura, Ishibashi & Shirasaka (1965) pointed out that ophiobolin is the first example of a C_{25} terpenoid. They concluded, on the basis of biogenetic considerations, that ophiobolin is probably constructed from five isoprene units which are linearly linked head to tail. They also proposed a biogenetic cyclization scheme from a hypothetical precursor, geranylfarnesyl pyrophosphate. In order to confirm this view, a biogenetic experiment has since been carried out (Nozoe, Morisaki, Tsuda & Okuda, 1967). On the other hand, Canonica, Fiecchi, Kienle, Ranzi & Scala (1966) have recently reported the results of their studies of the incorporation of [2-^{14}C]mevalonate into ophiobolin, and confirmed that this compound is really a C_{25} terpenoid.
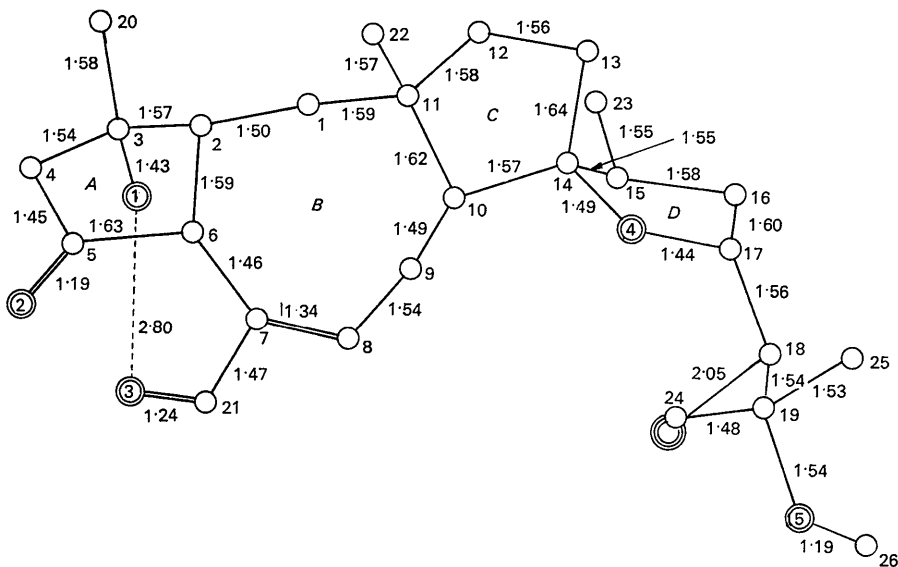
Fig. 3. Bond lengths (Å).

Bond lengths and angles found in the molecule of ophiobolin methoxybromide are shown in Figs. 3 and 4. The mean standard deviations of bond lengths and angles are calculated for each type of bond found in the molecule, and listed in Table 1.

The mean value of the twenty five C–C single bond lengths, 1·55 Å, and that of the five C–O single bond lengths, 1·42 Å, are in good agreement with the usual values, although some of the bond lengths are too long or too short. The mean C–C bond lengths and C–C–C angles averaged within each ring are listed in Table 4. Most of the unusual values are found at the junctions of two rings. The bond lengths and angles involved in the eight-membered B ring are normal except the length, 1·62 Å, found between B and C rings and the angles 119° and 118° at the junction to the cyclopentanone (A) ring.

The planarity and the conformation of each ring will be seen from Table 5. The dihedral angles between these rings are also listed in Table 5. The two double bonds C(7)=C(8) and C(21)=O(3) are arranged approximately *trans* to each other, but the bond C(21)=O(3) is twisted by 8·1° from the exact *trans* position in such a way as to bring the O(3) atom near to O(1). This may be a consequence of the intramolecular hydrogen bond of length 2·80 Å formed between O(3) and O(1). The infrared spectra measured at various concentrations in carbon tetrachloride solution also indicated the presence of the intramolecular hydrogen bond. The conformation of the molecule can be conveniently represented by indicating the internal rotation angles in a Figure of the molecule. The values have been calculated for each bond of the molecule of ophiobolin methoxybromide and are shown in Fig. 6. The definition of the internal rotation angles and the way of describing them in the Figure are illustrated in Fig. 5.

The methoxyl oxygen O(5) and the ether oxygen O(4) are both at the gauche positions to the bromine atom and not at the *trans* positions. A similar conformation is commonly observed in bromohydrins or chlorohydrins such as ethylene chlorohydrin (Bastiansen,

Table 4. *The mean C-C single bond lengths and C-C-C angles found in each ring.*

Ring	A	B	C	D
Bond length (Å)	1·56	1·54	1·59	1·57
Angle (°)	104	115	104	104

Table 5. *Perpendicular distances of the atoms from the best planes formed by the full members of each ring*

Ring A		Ring B		Ring C		Ring D	
C(2)	−0·229 Å	C(1)	−0·542 Å	C(10)	0·205 Å	C(14)	−0·215 Å
C(3)	0·235	C(2)	0·216	C(11)	−0·290	C(15)	0·213
C(4)	−0·140	C(6)	0·518	C(12)	0·260	C(16)	−0·136
C(5)	−0·003	C(7)	−0·428	C(13)	−0·124	C(17)	0·013
C(6)	0·137	C(8)	−0·353	C(14)	−0·052	O(4)	0·125
		C(9)	0·556				
		C(10)	−0·158				
		C(11)	0·190				

Dihedral angles between: ring A and ring B, 147·3°; ring B and ring C, 158·5°; ring C and ring D, 83·8°.

Fig. 4. Bond angles (°).

(a)

(b)
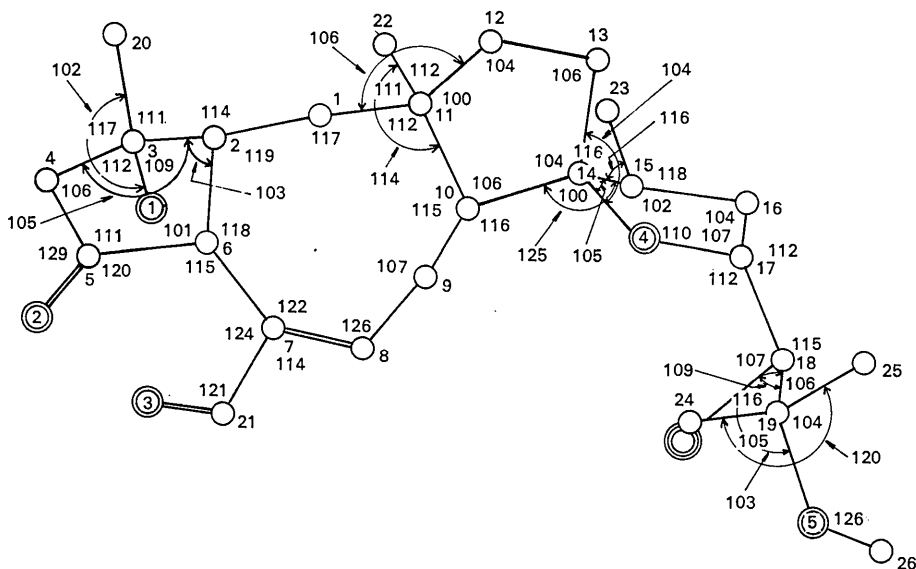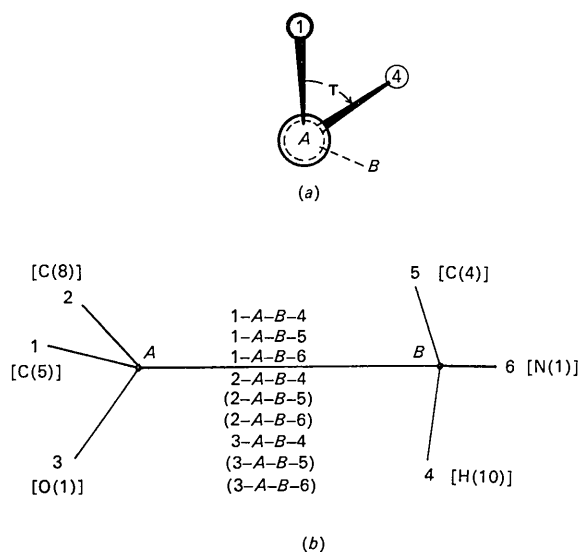
Fig. 5. (a) The internal rotation angle (torsion angle), τ, of $1 - A - B - 4$, is defined as the angle formed by the projection of the $1 - A$ bond with that of $B - 4$ when the projection is taken along the $A - B$ bond. The positive τ is taken in the same sense as that of the turning direction of a right handed screw advancing along the $A - B$ bond. It should be noted that $\tau(1 - A - B - 4) = \tau(4 - B - A - 1)$. (b) Sequence of describing the various τ values around the $A-B$ bond. The values in parentheses are not shown in Fig. 6. To ensure the correct sequence, the following conventions are adopted in this paper. (1) If A and B are different kinds of atom, the atom with greater atomic number is taken as B. (2) If both A and B atoms are the same kind, the atom with greater suffix number is taken as B. For example, for C(10)-C(11), C(10) = A and C(11) = B and for O(3)-C(10), C(10) = A and O(3) = B. (3) The assignment of atoms to 1,2,3,4,5,6, is subject to the same rule as (1) and (2). In Fig. 5(b), an example is shown in parentheses.

1949), α-methyl-D-galactoside-6-bromohydrin (Robertson & Sheldrick, 1965) and jacobin bromohydrin (Fridrichsons & Mathieson, 1963).

The absolute configuration at C(15) is opposite to that found in the sterol side chain at C_{20}, which can be explained by biogenesis of these compounds.

The crystal structure

Two projections of the crystal structure of ophiobolin methoxybromide viewed along the c and a axes are shown in Figs. 7 and 8 respectively. Intermolecular short contacts less than or equal to 3·7 Å are shown in these Figures. The molecules are packed together through van der Waals forces. There are no abnormal distances between the molecules.

References

BASTIANSEN, O. (1949). Acta Chem. Scand. 3, 415.

BERGHUIS, J., HAANAPPEL, IJ. M., POTTERS, M., LOOPSTRA, B. O., MACGILLAVRY, C. H. & VEENENDAAL, A. L. (1955). Acta Cryst. 8, 478.

BIJVOET, J. M., PEERDEMAN, A. F. & VAN BOMMEL, A. J. (1951). Nature, Lond. 168, 271.
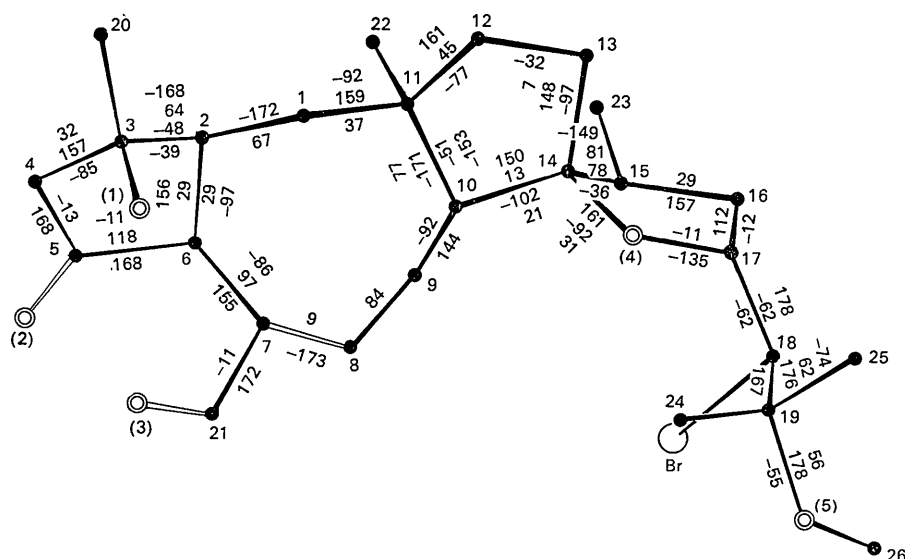
Fig. 6. Internal rotation angles found in the molecule of ophiobolin methoxybromide (°).

BUSING, W. R., MARTIN, K. O. & LEVY, H. A. (1962). ORFLS. Oak Ridge National Laboratory, Oak Ridge, Tennessee, U.S.A.

CANONICA, L., FIECCHI, A., KIENLE, M. G., RANZI, B. M. & SCALA, A. (1966). Tetrahedron Letters, p. 3035.

CANONICA, L., FIECCHI, A., KIENLE, M. G. & SCALA, A. (1966). Tetrahedron Letters, p. 1211.

DAUBEN, C. H. & TEMPLETON, D. H. (1955). Acta Cryst. 8, 841.

FRIDRICHSONS, J. & MATHIESON, A. McL. (1963). Acta Cryst. 16, 1075.

International Tables for X-ray Crystallography (1952). Vol. I. Birmingham: Kynoch Press.

International Tables for X-ray Crystallography (1962). Vol. III. Birmingham: Kynoch Press.

ISHIBASHI, K. (1961). J. Agr. Chem. Soc. Japan, 35, 323.

ISHIBASHI, K. (1962a). J. Antibiot. A15, 88.

ISHIBASHI, K. (1962b). J. Agr. Chem. Soc. Japan, 36, 649.

ISHIBASHI, K. & NAKAMURA, R. (1958). J. Agr. Chem. Soc. Japan, 32, 739.

NOZOE, S., HIRAI, K. & TSUDA, K. (1966). Tetrahedron Letters, p. 2211.
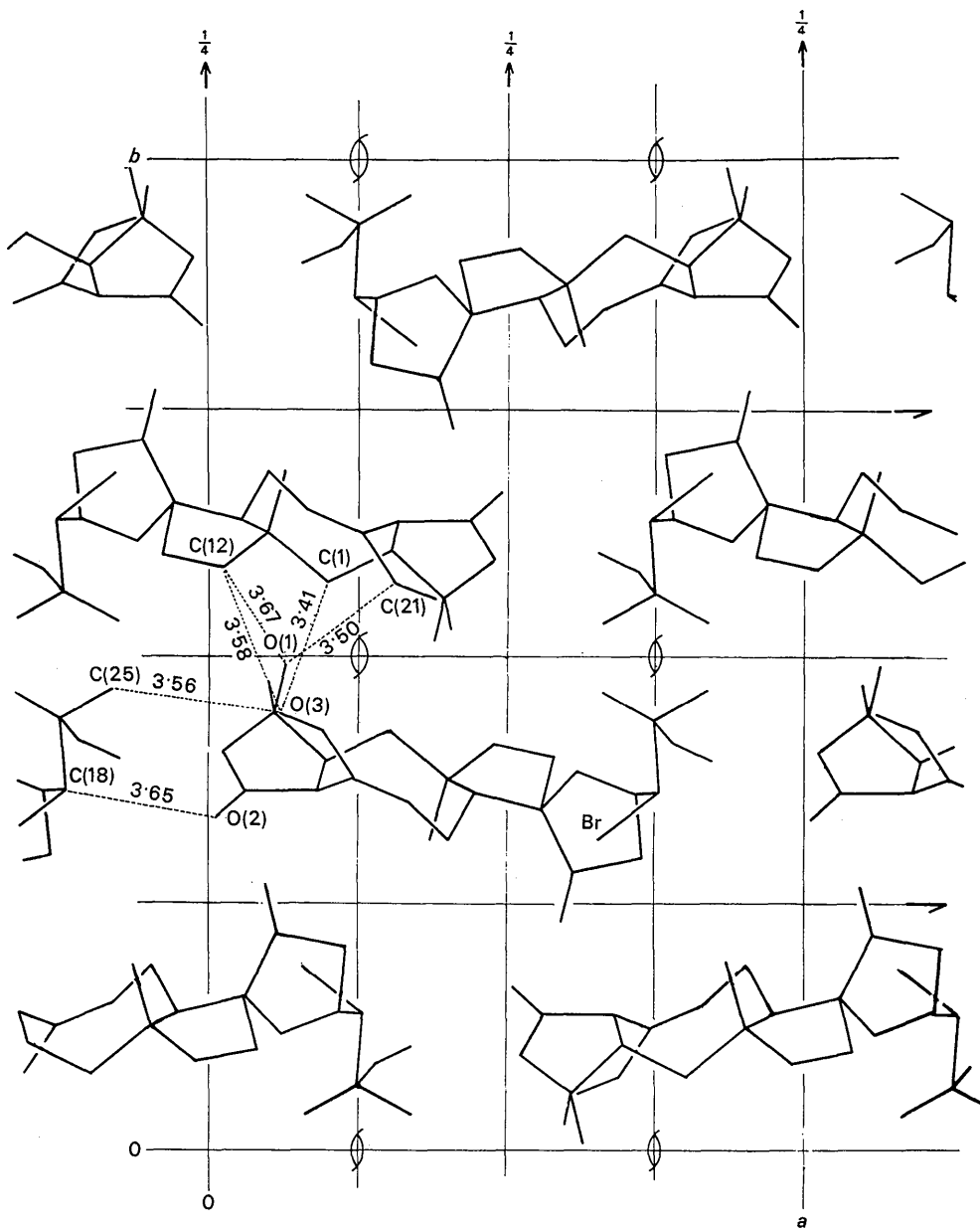
Fig. 7. The crystal structure projected along the c axis. To represent the correct absolute configuration, the positive direction of the c axis should be upwards towards the observer. Intermolecular short contacts less than or equal to 3·7 Å are shown.
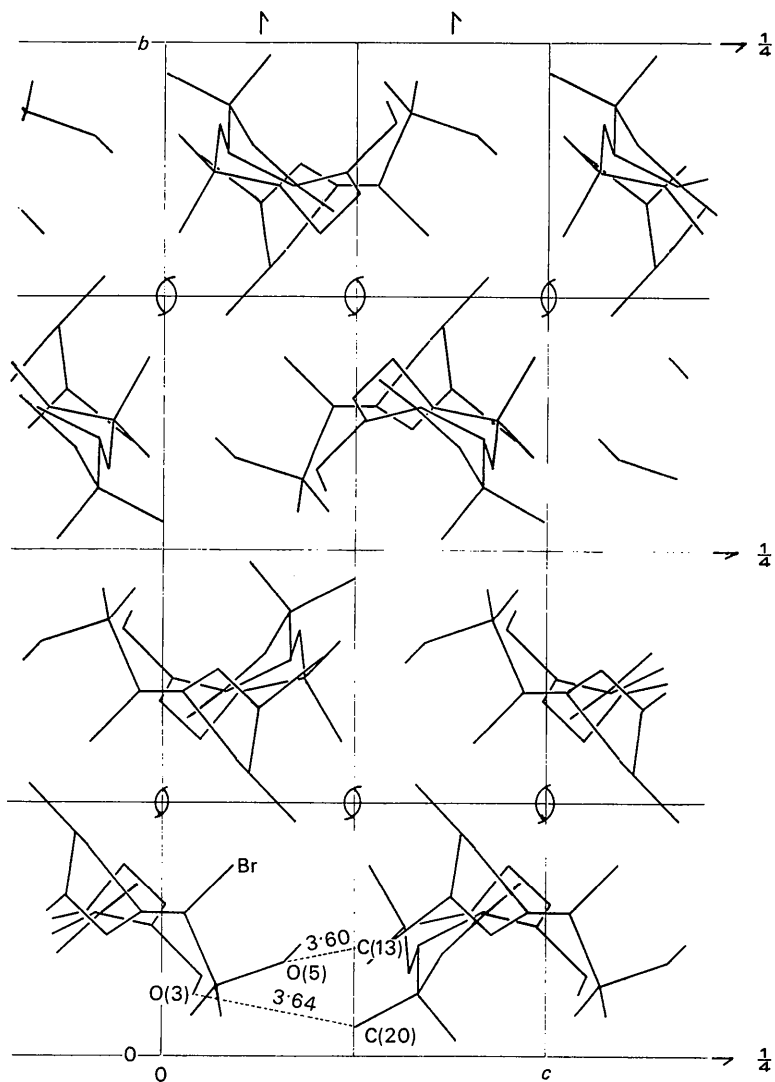
Fig. 8. The crystal structure projected along the *a* axis. To represent the correct absolute configuration, the positive direction of the *a* axis should be downwards away from the observer.

NOZOE, S., MORISAKI, M., TSUDA, K., IITAKA, Y., TAKA-HASHI, N., TAMURA, S., ISHIBASHI, K. & SHIRASAKA, M. (1965). *J. Amer. Chem. Soc.* **87**, 4968.
NOZOE, S., MORISAKI, M., TSUDA, K. & OKUDA, S. (1967). *Tetrahedron Letters*, p. 3365.

ORSENIGO, M. (1957). *Phytopathol. Z.* **29**, 189.
ROBERTSON, J. H. & SHELDRICK, B. (1965). *Acta Cryst.* **19**, 820
TSUDA, K., NOZOE, S., MORISAKI, M., HIRAI, K., ITAI, A., OKUDA, S., CANONICA, L., FIECCHI, A., KIENLE, M. G. & SCALA, A. (1967). *Tetrahedron Letters*, p. 3369.