# Application of the *B*2,0 and *B*3,0 formulae to the structure determination of a photodimer of *o*-distyrylbenzene, $C_{44}H_{36}$

TH. E. M. VAN DEN HARK, P. T. BEURSKENS AND W. H. LAARHOVEN Crystallography Laboratory, and Department of Organic Chemistry, University of Nijmegen, Toernooiveld, Nijmegen, The Netherlands

(Received 20 August 1973)

#### Abstract

A simple application of the B3,0 formula is described. This formula is used mainly to avoid inconsistent  $\Sigma_2$ -interactions in standard symbolic-addition or multiple-solution techniques. Computer time is reduced by a reflection selection procedure based upon the use of the B2,0 formula. The solution of a centrosymmetric structure is described. This compound, a photodimerization product of o-distyrylbenzene, is 5,6,11,12-tetraphenyl-dibenzo[2-3,8-9] tricyclo[8,2,0,0<sup>4,7</sup>] dodecadiene-2,8, C<sub>44</sub>H<sub>36</sub>. The compound crystallizes in the monoclinic space group C2/c, with unit cell parameters a = 28.047, b = 9.504, c = 12.600 Å and  $\beta = 103.4^{\circ}$ . A rather poor set of data was collected by an automatic diffractometer. Structural parameters were refined by full-matrix least-squares methods to an *R*-value of 0.06 for 970 non-zero reflections. The molecule is situated on a twofold rotation axis. It contains *cis-*, *trans-*, *cis*-substituted puckered cyclobutane rings. The dihedral angle between the benzo-groups is 61.5°.

#### Introduction

Attention is given to the solution of a phase problem that, at first sight, may be considered as easily solvable, namely a small, monoclinic, centro-

Copyright © 1974. Plenum Publishing Company Limited. No part of this publication may be reproduced, stored in a retrieval system, or transmitted, in any form or by any means, electronic, mechanical, photocopying, microfilming, recording or otherwise, without written permission of Plenum Publishing Company Limited.

# 228 TH. E. M. VAN DEN HARK, P. T. BEURSKENS AND W. H. LAARHOVEN

symmetric, light-atom structure. Standard techniques, however, failed for this example: the Patterson synthesis could not be solved because of heavy overlap and lack of resolution; attempts to solve the structure by standard multi-solution and symbolic-addition procedures failed because of inconsistent  $\Sigma_2$ -relations between reflections with large *E*-values and, probably, because of the small number of reflections available. Certainly, the structure could have been solved by repeated calculations, on modifying the computer input parameters, and also by modern probabilistic techniques (Duax et al, 1972). Nevertheless, we assume that it will be of interest to develop a simple procedure that may be used routinely for small structures when standard techniques fail at a first trial. The procedure, used to solve the present structure, is easily programmed and does not require much computer time.

On irradiation of *o*-distyrylbenzene (I) several compounds are formed. Among them, three dimeric molecules, formed by a twofold cyclization, are present. These compounds were reported independently by Müller et al (1966, 1970) and by Laarhoven et al (1970), and have the same m.p., u.v. and n.m.r. data. However, different molecular structures were assigned to these products. Müller et al (1966, 1970) proposed that these dimers were isomers of structure (II), whereas Laarhoven et al (1970) gave structures without cyclobutane rings, because the expected formation of stilbene on pyrolysis of compounds like (II) did not occur. Therefore, an X-ray analysis was carried out of one of the dimeric products (m.p. 293°).



### Experimental

5,6,11,12-tetraphenyl-dibenzo[2-3,8-9] tricyclo  $[8,2,0,0^{4,7}]$  dodecadiene-2,8 (C<sub>44</sub>H<sub>36</sub>), FW = 564.87, forms small, colourless crystals, somewhat elongated along the *b*-axis.

The crystals are monoclinic with space group C2/c (No. 15). From Pt-calibrated Weissenberg photographs taken with Ni-filtered Cu K $\alpha$  radiation ( $\lambda = 1.5418$  Å), application of a least-squares procedure yielded a = 28.047(9), b = 9.504(3), c = 12.600(4) Å and  $\beta = 103.4(1)^\circ$ ;  $V_c = 3267(3)$  Å<sup>3</sup>. The calculated density of 1.148 g cm<sup>-3</sup> with Z = 4, agrees with the measured value of 1.13 g cm<sup>-3</sup> (flotation method); F(000) = 1200.

Intensity data were collected with an automatic NONIUS diffractometer, using Zr-filtered Mo K $\alpha$  radiation up to a sin  $\theta/\lambda$ -value of 0.48. Of the 1519 attainable symmetry-independent reflections, 970 were observed above background ( $I \ge 3\sigma$ ). After every 20 reflections, a reference reflection was measured to detect and allow corrections to be made for slow fluctuations in the primary beam.

Corrections were made for Lorentz and polarization factors, the data were placed on an absolute scale by means of a Wilson plot and normalized structure factors were calculated. The experimental distribution of normalized structure factors is consistent with the centrosymmetric space group C2/c.

### Structure determination

B3,0 and B2,0 formulae. The B3,0 formula (Hauptman & Karle, 1957, 1958; Karle & Hauptman, 1957, 1958, 1959) is written as

$$E'_{\mathbf{h}}E'_{\mathbf{h}'}E'_{-\mathbf{h}-\mathbf{h}'} = AB + C \tag{1}$$

with

$$B = \langle (|E_{\mathbf{k}}|^2 - 1) (|E_{\mathbf{k}+\mathbf{h}}|^2 - 1) (|E_{\mathbf{k}+\mathbf{h}+\mathbf{h}'}|^2 - 1) \rangle_{\mathbf{k}}$$

in which the average is taken over all reflections  $\mathbf{k} = hkl$ ; A is a positive constant and C is a small positive correction term. E' is the normalized structure factor of the 'squared structure'. For large |E|-values, the approximation  $E' \approx E$  may be used. According to our experience (Kanters et al, 1966), the numerical results are rather poor. About 50% of the results, however, may be used in a sign generating procedure: the sign of the left hand side of (1) is equal to the calculated sign of AB + C provided that  $|AB + C| \ge 0$ :

$$S(E_{\mathbf{h}}E_{\mathbf{h}'}E_{-\mathbf{h}-\mathbf{h}'}) = S(AB+C)$$
<sup>(2)</sup>

Equation (2) can be used only for large values of the triple product  $E_{h}E_{h'}E_{-h-h'}$ , otherwise large deviations in the B3.0 results are to be expected. It may be noted that in the usual application of the  $\Sigma_2$ -relation, the left-hand side of (2) is supposed to be positive; the B3.0 formula provides a possibility to avoid inconsistent (non-valid) interactions.

The sigma-1 type B3,0 formulae are obtained by using symmetry-related reflections h and h'; for space group C2/c, with  $E \approx E'$ :

$$(-1)^{1} |E_{\mathbf{hkl}}|^{2} E_{0,2\mathbf{k},0} = AB + C$$

$$(-1)^{1} |E_{\mathbf{hkl}}|^{2} E_{2\mathbf{h},0,2\mathbf{l}} = AB + C$$
(3)

The B2,0 formula (Karle & Hauptman, 1959) is written as

$$|E'_{\mathbf{h}}|^2 = 1 + D\langle (|E_{\mathbf{k}}|^2 - 1) (|E_{\mathbf{k}+\mathbf{h}}|^2 - 1) \rangle_{\mathbf{k}}$$
(4)

in which D is a positive constant. This formula is used to calculate the |E'|-values of the medium-strong reflections. Where a medium-strong reflection is used in the triple product equation (1), and if  $|E'| \ll |E|$  for this reflection, many of the B3,0 results are likely to be unreliable. So the B2,0 formula is applied to select reflections in order to enhance the probability of finding useful results (large (AB + C)-values).

*Calculations.* The B3,0 formula was used to calculate (AB + C)-values for 460 interactions  $(\mathbf{h}, \mathbf{h}', \mathbf{h} + \mathbf{h}')$  among 114 reflections with  $|E| \ge 1.7$  (table 1a). The B2,0 formula (4) was used to calculate |E'|-values of the 90 reflections with  $1.4 \le |E| \le 1.7$ . For 26 reflections, we found |E'| to be greater than 2.5.

Including these reflections, the B3,0 calculations were extended to 332 additional interactions (table 1b). The sigma-1 type B3,0 formula (3) was applied to 0,2k,0 and 2h,0,2l reflections with  $|E| \ge 1.0$ .

	table 1a:	$ E  \ge 1.7$	table 1b:	extended
Range of $(AB + C)$	N(tot)	N(inv)	N(tot)	N(inv)
+200 +70	43	1	42	0
+70 +40	59	1	68	1
+40 +10	162	6	107	8
+100	96	5	37	3
020	84	68*	58	38*
-20 60	16	5**	20	4**

Table 1. Distribution of incorrect B3,0 results

N(inv) is the number of interactions contradicting eq. (2) (as observed after the structure determination).

\* In this interval, most of the negative (AB + C)-values correspond to positive triple products.

\*\* In this interval most of the negative (AB + C)-values correspond to negative triple products (invalid  $\Sigma_2$ -relations).

B3,0 results. The results were tabulated according to decreasing values of (AB + C). Eleven letter symbols were assigned to the reflections with  $|E| \ge 1.7$  occurring most often in the top half of the table. The sign correlation procedure (Beurskens, 1963) was used to generate symbolic signs from the B3,0 results. We define the following sets of reflections:

 $h_1$  are the eleven initial choices  $h_2$  are reflections  $h_1 + h'_1$  $h_3$  are reflections  $h_1 + h_2$  and  $h_2 + h'_2$ . To find the reflections  $h_2$  and  $h_3$ , only the top half of the B3,0-table was used. Several of the reflections were found at least three times with the same letter symbol. It is highly improbable that consistent results are obtained from incorrect B3,0 results, and therefore the multiple sign indications were assumed to be correct and these reflections added to the set  $h_1$ . Thereafter, new reflections  $h_2$  and  $h_3$  could be calculated, and new reflections with multiple sign indications were added to the set  $h_1$ , etc. Relations between the letter symbols were accepted as being correct where they were found at least five times, without inconsistencies. Finally the origin was fixed and two unknown letters remained to express the signs of 120 reflections  $h_1$ .

The set  $h_1$  then was used as input data to a  $\Sigma_2$ -sign generation procedure, leading to the determination of a total of 514 signs. The weaker sigma-1 type B3,0 results were used at the end of the procedure to eliminate one of the letter symbols. The most probable of the two remaining solutions clearly revealed all of the carbon atoms of the molecule.

Structure refinement. The coordinates of the carbon atoms were refined, first with isotropic and then with anisotropic thermal factors. The fourteen hydrogen atoms attached to benzene carbon atoms were placed at calculated positions (C-H distance is 1.084 Å) and were included as constants in the refinement. The four hydrogen atoms attached to the carbon atoms of the cyclobutane ring were located on a difference-Fourier map and were included as variables in the last stage of the refinement. The temperature factors of all hydrogen atoms were fixed at a value of 4.0  $Å^2$ . The full-matrix least-squares refinement was carried out on the function  $\Sigma w(|F_0| - |F_c|)^2$ , where  $1/w = \sigma_c^2 + (0.05|F_0|)^2$  with  $\sigma_c$  the standard deviation calculated from counting statistics. The final R-value was 0.06 for all non-zero reflections. Structure factor calculations for 1519 reflections, including 549 unobserved reflections, gave R = 0.12. The atomic scattering factors used are those listed in the International Tables for X-ray Crystallography Vol. III. The fractional coordinates and thermal parameters, with standard deviations, of the carbon atoms are listed in table 2. Table 3 contains the coordinates of the hydrogen atoms. The structure is illustrated in figures 1 and 2. The structure factors are listed in table 4.

## Discussion

The distribution of correct and incorrect B3,0 results is shown in table 1. About 12% of all calculated interactions do not satisfy the  $\Sigma_2$ -relationship; these invalid interactions may give rise to false solutions in standard direct methods techniques. By using only the top half of the B3,0 results we avoided about 75% of the inconsistencies, and by using a careful sign generation procedure the probablity of making mistakes is greatly reduced. It was found à postériori that the four strongest reflections, all entering into many  $\Sigma_2$ -relations, are involved in eight non-valid  $\Sigma_2$ -interactions with

	$\beta_{23}$	11(6)	2(6)	-11(6)	14(6)	0(6)	-2(6)	0(0)	-22(6)	-4(7)	-2(6)	-2(6)	4(7)	16(7)	17(10)	-2(10)	18(8)	-22(7)	-10(7)	-13(8)	-54(11)	-38(11)	-10(8)
	$\beta_{13}$	12(2)	16(2)	13(2)	13(2)	19(2)	15(2)	11(2)	24(3)	32(3)	19(2)	10(2)	14(2)	17(3)	21(3)	15(3)	17(3)	17(2)	18(2)	26(3)	28(4)	7(3)	6(3)
	$\beta_{12}$	9(3)	0(3)	-1(3)	5(3)	-2(3)	-2(3)	-8(3)	-10(3)	7(4)	2(3)	0(3)	4(3)	-3(4)	-1(4)	21(4)	17(4)	-6(3)	-7(3)	-1(4)	-24(5)	-26(5)	-13(4)
	β <sub>33</sub>	57(5)	61(5)	61(5)	79(5)	57(5)	53(5)	58(5)	91(6)	87(6)	66(5)	77(6)	96(6)	91(6)	125(8)	150(8)	146(7)	75(6)	109(7)	133(8)	179(11)	133(9)	87(7)
	$\beta_{22}$	133(9)	97(8)	115(9)	128(10)	92(9)	84(9)	111(9)	102(11)	120(11)	97(9)	118(10)	160(11)	215(14)	239(16)	277(18)	175(12)	135(12)	102(10)	137(12)	203(16)	265(18)	228(15)
	$\beta_{11}$	17(1)	18(1)	15(1)	18(1)	20(1)	18(1)	23(1)	29(2)	30(2)	24(1)	17(1)	16(1)	21(1)	21(2)	16(1)	17(1)	17(1)	22(1)	23(2)	27(2)	21(2)	19(1)
, ,	N	0.2104(4)	0.2712(4)	0.1584(4)	0.1092(5)	0.3475(4)	0.3836(4)	0.4501(5)	0.4830(5)	0.4481(6)	0.3816(5)	0.2588(5)	0.3176(5)	0.3640(5)	0.3538(7)	0.2979(7)	0.2466(6)	0.1018(5)	0.1643(6)	0.1551(6)	0.0821(9)	0.0205(7)	0.0303(6)
	у	0.0096(6)	-0.0199(6)	-0.0503(6)	-0.0771(7)	0.0893(7)	0.0746(6)	0.1763(7)	0.2929(7)	0.3047(7)	0.2037(7)	-0.0286(7)	-0.1519(8)	-0.1848(8)	-0.0948(11)	0.0275(10)	0.0624(7)	-0.2326(7)	-0.3367(8)	-0.4778(8)	-0.5148(9)	-0.4117(12)	-0.2706(10)
he key to ato	x	0.3882(2)	0.4416(2)	0.4545(2)	0.3991(2)	0.4711(3)	0.5224(3)	0.5486(2)	0.5250(3)	0.4741(3)	0.4474(2)	0.3456(2)	0.3450(2)	0.3058(3)	0.2676(3)	0.2673(3)	0.3064(3)	0.3837(2)	0.4126(2)	0.3983(3)	0.3552(4)	0.3253(3)	0.3410(3)
L	Atom	C(1)	C(2)	C(3)	C(4)	C(5)	C(6)	C(7)	C(8)	C(9)	C(10)	C(11)	C(12)	C(13)	C(14)	C(15)	C(16)	C(17)	C(18)	C(19)	C(20)	C(21)	C(22)

Table 3. Hydrogen atom coordinates. H(1)-H(4): refined positions. H(7)-H(22): calculated positions. The numbering of the hydrogens refers to the parent carbon atoms.

Atom	x	у	Z
H(1)	0.384(2)	0.128(7)	0.190(5)
H(2)	0.442(2)	-0.123(7)	0.310(5)
H(3)	0.479(2)	-0.131(7)	0.143(5)
H(4)	0.386(2)	-0.017(7)	0.041(5)
H(7)	0.588	0.166	0.478
H(8)	0.546	0.373	0.535
H(9)	0.456	0.394	0.474
H(10)	0.408	0.213	0.356
H(12)	0.376	-0.224	0.328
H(13)	0.306	-0.283	0.408
H(14)	0.237	-0.120	0.390
H(15)	0.237	0.100	0.292
H(16)	0.305	0.158	0.199
H(18)	0.446	-0.307	0.221
H(19)	0.421	-0.557	0.205
H(20)	0.345	-0.625	0.073
H(21)	0.291	-0.440	-0.034
H(22)	0.319	-0.191	-0.020



Fig. 1. Bond distances and angles, with esd in parentheses.



Fig. 2. Thermal ellipsoid plot of the molecule, as seen along the twofold rotation axis.

reflections having  $|E| \ge 1.7$ . Only one of these interactions is present in the top half of the table of B3,0 results.

The B2,0 formula appears to be very useful in saving computer time. It is seen from comparing tables 1a and 1b that the introduction of reflections with smaller |E|-values but selected for larger |E'|-values, does not reduce the reliability of the B3,0 results.

In our opinion, the procedure used in this structure investigation can never compete with standard techniques as far as simplicity and computer time is concerned. It may be useful only in cases where routine techniques have failed. As the main feature of the present procedure seems to be the avoidance of incorrect  $\Sigma_2$ -interactions, it should be possible to apply the same principles to small acentric structures.

The molecular structure is found to be in agreement with formula (II). So the argument that no cyclobutane moiety can be present in the dimer because of lack of stilbene formation on pyrolysis is wrong, obviously. This conclusion was reached recently in other investigations. Laarhoven & Cuppen (1972) prepared some compounds in which thermolysis of a 1,2-diphenylcyclobutane moiety apparently proceeds in a regiospecific way: *cis-*, *cis-*, *cis-*1,2,2a,10b-tetrahydro-1,2-diphenylcyclobuta[1]phenantrene gives stilbene on thermal decomposition, but the *trans-*, *trans-*, *trans-* isomer does not.

Meinwald & Young (1971) reported that a dimer of 1,8-distyrylnaphthalene, containing two *cis-*, *trans-*, *cis-*1,2-diphenylcyclobutane parts gives stilbene on heating, but a dimer of 2,3-distyrylnaphthalene with identically substituted cyclobutane rings does not (Ottenheijm, 1973). From these facts, it is not quite possible to predict whether or not a 1,2-diphenylcyclobutane moiety in a molecule will produce stilbene on pyrolysis. The present compound can be formed by a head-to-head dimerization of *trans-*, *trans-o*-distyrylbenzene, possibly *via* an eximer.

The geometry of the molecule is shown in figure 1. The molecule possesses a twofold rotation axis through the dodecadiene ring. The dihedral angle

$0F_c$	74	44	-111			-93	- 43			131				-136	67 - 92	-61	90			-54	-45	48	121				134	143	135	-93	-133				129	-163	-65	
F <sub>0</sub>   1	71	33.	13	6, 1 = 1		93	70	6. 1=1		25		7, <i>l</i> = 1	4	04	17	57	92		7, l=1	67	44	48	20		8, 1 = 1		21	43	42	02	31 -		8, 1 = 1		20	45	82	0 /= 1
101	-		-	= <i>¥</i>			=	<i>k</i> =				Y	Ŧ				_		- ×				-		k =		=	-	1	Ξ	=		k =		=	-		= 4
ч	-13		- 19			4	7			-			·				, ,			Ī	1	1	- <u></u>				(1	4	9	<b>x</b>	10				9	-10	-12	
$10F_{c}$	-21 -78			49	-250	-290	171-	74	67	219	144				525	-295	-123	279	119		-	152	60	111	64	48	38	64	209	-94				215	-158	200	-41	82
$10 F_0 $	36 80	, 3	k = 4, <i>l</i> =	56	262	333	141 733	69	66	226	142		k = 4, <i>l</i> =		532	286	140	288	109	k = 5   =	- 5	149	78	107	56	52	27	70	210	90		k = 5, l =		213	152	219	48	81 124
ч	-17	1		2	4	9	× 9	12	14	18	20			ç	7	1	8	$^{-10}$	-22				ŝ	S	6	11	13	15	17	19				-	-33	<b>?</b>	L	6- 1
$10F_c$	-	-34	-425 291	163	-331	164	911 84	00	1		-531	<i>LL</i>	-118	1001	-100 56	-29	64	89	-22	-	4	-90	-40	337	209	T	209	68	-76	-126				-102	167	24	362	-265
$10 F_0 $	k = 2, l =	46	423 281	155	330	164	771	4	k = 2, l =		531	72	120	32	20 54	29	69	90	33	k = 3 $l = 1$		84	25	340	225	20	207	66	89	121		k = 3, l =		110	177	29	392	266 157
ч		2.	<del>4</del> 9	80	10	4	30	07			-2	4	9 0	x c 	112	19	-18	20	-22			-	ŝ	5	L	6	Ξ	13	17	21				ς 1	-5	L-	-0	- 13
<sup>г</sup> .	32	2		4	2	5		5			2	×.	4 9	Ş		16	1	06	2 -	-	12	10				2	7	ŝ	~	9		~	~	6		5	4	×
10	ĩ-		0 =	9	ব	00 0	~ ~	1	0 =		-14	-12	1			-	Ű	6	7 -	7	-	1		-		-32	21	50	10	Ň	6(	-25	78	10	-13	- 2	<b>v</b> )	-0
$10 F_0 $ 10.	80 -8		k = 7, l = 0	82 9	63 4	78 -8	13 86	0 00	k = 8, l = 0		154 -14	128 -12	154 –15 05	с <b>к</b>	1 1=1	     	72 6	890 99	4] 4]	10- 11-	125 -12	113 -1	<	k = 1, l = 1		175 -32	187 21	519 50	96 10	30 -3	7266	262 -257	76 78	109 10	13213	42 - 2	73 5	696
$h$ 10  $F_0$   10.	10 80 -8		k = 7, l = 0	1 82 9	3 63 4	7 78 -8	9 /3 13 86	0- 00 11	k = 8, l = 0		0 154 -14	2 128 -12	4 154 –15 0 05	۲ ۲	L = 1 $I = 1$	, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1, 1,	1 72 6	3 890 99	5 41 5 7 115 1	11- GIT /	11 125 -12	17 113 -1	6 6 6 6	k = 1, l = 1		-1 175 -32	-3 187 21	-5 519 50	-7 96 10	-9 30 -3	-11 7266	-13 262 -257	-15 76 78	-17 109 10	-19 13213	-21 42 -2	-23 73 5	-25 69 -6
$10F_{\rm C}$ h $10 F_{\rm O} $ 10	0 10 80 -8	-226	-311  k = 7, l = 0 49	49 1 82 9	-106 3 63 4	-55 7 78 -8	124 9 /3 /3 / 68 13 868	52 13 000	k = 8, l = 0	0	0 154 -14	344 2 128 -12	58 4 154 -15 211 9 65	- 54 0 145 - 54 0 52 52 52 52 52 52 52 52 52 52 52 52 52	133 $k=1$ $l=1$	-7777	-123 1 72 6	37 3 890 99	131 5 41 6	11- 901 6	0 11 125 -12	17 113 -1	118	232 $k = 1, l = 1$	124	202 -1 175 -32	214 -3 187 21	87 -5 519 50	-155 -7 96 10	-244 -9 30 -3	-117 -11 7266	-13 262 -257	0 -15 76 78	-17 109 10	87 -19 132 -13	28 -21 42 -2	-165 -23 73 5	-103 $-25$ $69$ $-6$
$10 F_0 $ $10F_c$ $h$ $10 F_0 $ $10$	k = 3, l = 0 10 80 $-k$	217 -226	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	55 49 1 82 9	124 -106 3 63 4	55 -55 7 78 -8	109 124 9 /3 / 67 68 13 868	54 52 10 00	k = 8, l = 0	k = 4, l = 0	0 154 -14	375 344 2 128 -12	66 $58$ $4$ $154$ $-15$	504 - 541 8 90 50 50 50 50 50 50 50 50 50 50 50 50 50	1=1 1=1 72 CC 133 14=1	77 -77 - 77	126 -123 1 72 6	39 37 3 890 99	122 131 5 41 4 75 76 7 15 11	11 01 01 01 01 01	k = 5, l = 0 II 125 -12	17 113 -1	122 118	$235 \qquad 232 \qquad k = 1, \ l = 1$	124 124	192 202 -1 175 -32	212 214 -3 187 21	78 87 -5 519 50	140 -155 -7 96 10	227 -244 -9 30 -3	103 -117 -11 7266	-13 262 -257	c = 6, l = 0 -15 76 78	-17 109 10	95 87 -19 132 -13	26 28 -21 42 -2	156 -165 -23 73 5	$\begin{array}{rrrr} 101 & -103 & -25 & 69 & -6 \\ 74 & -70 & \end{array}$
$h$ 10 $ F_0 $ 10 $F_c$ $h$ 10 $ F_0 $ 10.	k = 3, l = 0 10 80 -8 14 111 1		3 298 -311 k=7, l=0 5 45 -49	7 55 49 1 82 9	9 124 -106 3 63 4	13 55 -55 7 78 -8	1 5/ 6 471 601 CI 87 69 13 86 13 87 15		k = 8, l = 0	k = 4, l = 0	0 154 -14	0 375 344 2 128 -12	2 66 58 4 154 -15	4 534 – 541 8 93 4	1 = 1 = 1 $1 = 22$ $25 = 22$ $1 = 1$ $1 = 1$ $1 = 1$ $1 = 1$	10 77 -77 -77 -77 -7	12 126 -123 1 72 6	16 39 37 3 890 99	18 122 131 5 41 6 20 75 76 7 15 1	11- 01 6 07 07 07	k = 5, l = 0 11 125 -12	17 113 -1	1 122 118	3 235 232 $k = 1, l = 1$	5 124 124	7 192 202 -1 175 -32	9 212 214 -3 187 21	11 78 87 -5 519 50	15 140 -155 -7 96 10	17 227 -244 -9 30 -3	19 103 -117 -11 7266	-13 262 -257	k = 6, l = 0 $-15$ 76 78	-17 109 10	0 95 87 -19 132 -13	2 26 28 -21 42 -2	4 156 -165 -23 73 5	$\begin{array}{cccccccccccccccccccccccccccccccccccc$
$10F_{\rm c}$ h $10 F_{\rm o} $ $10F_{\rm c}$ h $10 F_{\rm o} $ $10$	= 0   k = 3, l = 0   10   80   -1   -1   14   11   1   1		$\begin{array}{rrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrr$	-349 7 55 49 1 82 9	-198 9 124 $-106$ 3 63 4	-476 13 55 $-55$ 7 78 $-8$	7 57 6 477 601 CI 777- 8 90 10 63 68 13 86	148 21 54 52	-73 $k = 8, l = 0$	-146 $k = 4, l = 0$	84 0 154 -14	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	= 0 2 66 58 4 154 $-13$	2 CA 234 - 341 8 43 23 23 23 23 23 23 23 23 23 23 23 23 23	1 = 1 = 4 $2 = 2 = 2 = 2 = 2 = 2 = 2 = 2 = 2 = 2$	353 10 77 -7717	-33 12 126 -123 1 72 6	393 16 39 37 3 890 99	191 18 122 131 5 41 6 123 20 75 76 7 115 1	-129	-64 $k = 5, l = 0$ 11 125 $-12$	-65 17 113 -1	165 1 122 118	3 235 232 $k = 1, l = 1$	= 0 5 124 124	7 192 202 -1 175 -32	546 9 212 214 -3 187 21	-297 11 78 87 -5 519 50	-164 15 140 $-155$ $-7$ 96 10	-298 17 227 -244 -9 30 -3	215 19 103 -117 -11 7266	-33 -13 262 -257	$40 \qquad k = 6, l = 0 \qquad -15  76 \qquad 78$	-21 -17 109 10	-50 0 95 87 -19 132 -13	65 2 26 28 -21 42 -2	67 4 156 -165 -23 73 5	-104 6 101 $-103$ $-25$ 69 $-68 74 -70$
$10 F_0 $ $10F_c$ h $10 F_0 $ $10F_c$ h $10 F_0 $ $10$	z = 0, l = 0 $k = 3, l = 0$ 10 80 -1 14 11 11	240 -255 1 217 -226	$\begin{array}{rrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrr$	353 -349 7 55 49 1 82 9	209 -198 9 124 -106 3 63 4	494 - 476 13 55 -55 7 78 -8	7 5/ 6 471 601 CT 7/2 0/2 8/2 8/2 8/2 6/2 6/2 8/2 8/2 8/2 8/2 8/2 8/2 8/2 8/2 8/2 8	151 148 21 54 52	$69  -73  k = 8, \ l = 0$	137 - 146  k = 4, l = 0	83 84 0 154 -14	0 375 344 2 128 -12	(=1, t=0 2 66 58 4 154 $-12$	2 26 8 190 - 405 4 C3 53 50 51 201	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	351 $353$ $10$ $77$ $-77$ $-77$ $-$	32 -33 12 126 -123 1 72 6	423 393 16 39 37 3 890 99	202 191 18 122 131 5 41 6 127 122 20 75 76 7 15 1	131 - 129 - 20 - 72 - 70 - 113 - 112 - 11 - 113 - 116 - 11	k = 5, l = 0 11 125 -12	68 -65 17 113 -1	162 165 1 122 118	3 235 232 $k = 1, l = 1$	z = 2, l = 0 5 124 124	7 192 202 -1 175 -32	516 546 9 212 214 -3 187 21	285 -297 11 78 87 -5 519 50	160 -164 15 140 -155 -7 96 10	291 -298 17 227 -244 -9 30 -3	227 215 19 103 -117 -11 7266	47 –33 –13 262 –257	23 40 $k = 6, l = 0$ -15 76 78	39 -21 -17 109 10	4150 0 95 87 -19 13213	72 65 2 26 28 -21 42 -2	72 67 4 156 -165 -23 73 5	100 -104 6 101 -103 -25 69 -6 8 74 -70 -70 -6

Table 4. Observed and calculated structure factors

$10 F_0 $	$10F_c$	Ч	$10 F_0 $	$10F_{\rm c}$	Ч	$10 F_0 $	$10F_{\rm c}$	Ч	$10 F_0 $	$10F_{\rm c}$	Ч	$10 F_0 $	$10F_{\rm c}$	ч	$10 F_0 $	$10F_{c}$
137	134	17	73	82	-20	51	-37	44	123	114	-18	73	61	         	1243	1328
74	12 ·		k = 1, l =	2		k = 3, l =	- 2	⊃ ∞ I I	32	12	07-	70	0	) [- 	150	000 
								-10	160	-159		k = 7, 1	= 2	6-	322	302
k = 9, l	= ]	-	181	180	1	57	62	-12	39	17				-11	111	107
		- 3	242	270	ŝ	213	-207	-14	34	35	S	142	-136	-13	54	48
123	-122	-5	606	607	S	253	-245	-16	41	-41	15	89	69	15	96	-101
		L	202	235	2	345	-330	-18	52	-54				17	142	-142
k = 0, L	= 2	6-	298	-295	6	SI	51	-20	82	-85		k = 7, 1	= 2	-19	58	-48
			109	102	=:	114	-114	-24	89	93		i		-21	65	-67
290	316	-15	222	-215	15	49	40				ī	20	- 94	-23	47	53
1045	6011 747	- 1-	35	75		112	101		K = 5, l =	7	n <b>v</b> 	142	-152	.1	-	
202	-208		77	595	1	C11	1110	-	238	739	- 1- - 1-	11	- 00 - 72	×	- 7, 1 = .	~
165	-161	- 23	168	-160		k = 3, l =	5	- m	163	171	-	ţ	4	C	77	81
23	-12	-25	94	88				2	122	130		k = 8, l =	= 2	2	360	-385
359	331				-	319	350	6	64	70				4	190	202
57	-62		k = 2, l =	2	-3	118	-114	П	110	- 108	0	76	-73	9	61	52
139	144				-5	182	-170				4	71	-75	8	312	320
131	-122	0	1133	-1137	L –	126	-114		k = 5, l =	2				10	282	268
37	38	7	180	175	6-	266	-247					k = 8, l =	= 2	14	137	-140
110	108	4	54	20	-11	209	-195	-3	132	-129				16	62	-72
		9	49	46	-13	33	33	-5	56	47	-2	105	102	18	70	-57
k = 0, l	-5	8	140	138	-15	45	54	L –	139	130						
	000	019	141	130	-17	124	124	6,6	174	166		k = 9, l =	= 2		ζ=2, <i>l</i> =	ŝ
007	Q67-	71	707	007	-15	55	80	110	00 00	68 03	-	0	63	ſ	020	726
7771	0011	171	5	0 <del>1</del> 5	17	02	n/	15	11	011		60	70	1 -	000	007-
230 230	1420	181	18	-171 -		k = 4 1 =	6	17-	711	015-		k = 1 / =		+ 4 	202	-213
124	-121	20	161	-54		: :	1		k = 6, l =	2			1	i oc	93	16-
413	-389	22	115	123	0	110	101				-	553	596	- 10	282	266
175	-168	24	06	LL	2	213	183	0	127	-129	ŝ	84	93	- 12	92	-87
94	67				4	47	-45	2	30	29	ŝ	421	447	- 14	99	76
234	-216		k = 2, l =	2	9	33	6	4	225	225	7	164	163	16	189	181
86	-90				8	118	122	8	66	-70	6	257	-263	-18	32	-36
		2	690	-701	10	124	114	12	122	-123	11	348	-353	22	79	88
k = 1, l	= 2	-4	279	-297	12	52	56				13	61	68	-24	66	-93
0011	1.7.1	91	86 86	-54	4	37	41		k = 6, l =	5	15	4	24	-	- -	(
022	407 F	0	07 102	001	01	171	771-	ç	66	71	10	50	17	•	- 1 (0 - 1	ĉ
328	340	112	65	-68	01	C11		4 4	105	11		3	2	-	58	62
204	-201	14	231	228		k = 4, l =	5	-9-	30	37		k = 1, l =	= 3	ŝ	103	-115
474	-465	16	23	16				8	37	40				ŝ	22	-1-1
99	67	-18	41	59	2	223	-223	-12	32	-33	-	16	-18	L	220	230

Table 4-continued

35	-166	11	-66		4	171	132		4		-119	80	169	-138	-69	82		4		155	67	199	-70	-200	-93		4		231	114	-117	116	74		4		-277	-78	136		4	
33	164	71	LL		= S, <i>l</i> =	141	133		= 5, l =		120	76	168	146	LL	85		≈ 6, <i>l</i> =		156	93	185	85	195	87		= 6, <i>l</i> =		238	133	124	118	61		= 7, 1 =		274	88	134		= 1 1 =	
- 14	-16	- 18	-22		K	ŀ	15		×		-	<b>~</b>	-5	6	-13	-15		k		0	7	8	10	12	14		k		-2	4	-6	-12	-18		ķ			ę	11		K	:
96		4		-201	431	- 68	103	92	-67	85		= 4		-523	-108	-64	-169	-66	52	90	-30	181		= 4		209	16	47	17	33	-103	-31	39		= 4		45	169	62	36	-55	190
82		k = 3, 1		203	436	00	95	101	59	68		x = 3, 1		557	114	63	171	75	63	67	24	175		l = 4, l		216	29	58	26	35	93	45	35		č = 4, <i>1</i>		47	165	73	42	50	190
-26				-	ŝ	0 6	- 6	11	17	19		~		-	3	5	-1	6	-11	13	- 15	23				0	2	4	9	8	10	12	14		4		-2	4-	9-	<b>8</b> 	-10	-12
148	-210	64		= 4	200	513	778	-52	53	-82	θ	-88	-80	-124	-64	98		= 4		145	-412	-273	66		26	-147	156	231	126		= 4		-147	-182	-301	-290	11	89	-70	146	167	-53
144	212	66		c = 1, <i>l</i>	200	485	760	33	57	94	28	92	81	125	72	88		: = 2, 1		138	420	278	65	133	23	143	151	226	128		:= 2, 1		166	184	331	306	21	97	71	141	166	48
13	2	19		~		- (° 	0 Y	-7-	6-	=	-13	-15	-17	19	21	23		~		0	7	4	9	8	10	12	14	16	18		-		-7	4-	و i	81	-10	-12	-14	-16	-20	-22
	= <del>\</del>		-117	67	، ا	¢ =	108	1	= <b>4</b>		-271	37	-425	395	93	-159	114	226	72		= 4		-697	374	427	674	-299	573	145	34	-57	-143	92		= 4		72	342	-77	75	16	24
•	x = 8, l = 3		129 –117	82 67		( = Q, I = J	80 108		k = 0, l = 4		246 -271	28 37	376 -425	413 395	93 93	164 -159	115 -114	218 226	57 72		c = 0, l = 4		651 -697	401 374	446 427	703674	326 –299	608 573	148 145	38 34	63 –57	143 –143	104 · 92		c = 1, <i>l</i> = 4		65 72	343 342	73 –77	71 75	38 16	26 24
	k = 8, l = 3		2 129 -117	6 82 67	10	$k = \delta, l = J$	-2 89 108		k = 0, l = 4		0 246 -271	2 28 37	4 376 -425	6 413 395	8 93 93	10  164  -159	12 115 -114	14 218 226	18 57 72		k = 0, l = 4		-2 651 -697	4 401 374	6 446 427	-8 703674	-10 326 -299	-12 608 573	-16 148 145	-18 38 34	-20 63 -57	-22 143 $-143$	-24 104 92		k = 1, l = 4		1 6572	3 343 342	5 73 -77	7 71 71 75	9 38 16	11 26 24
-139	-106  k = 8, l = 3	84	92 2 129 -117	6 82 67		k = 0, l = 3	91 -2 89 108	52	-24 $k = 0, l = 4$	24	59 0 246 -271	-133 2 28 37	-73 4 376 -425	6 413 395	8 93 93	$10 \ 164 \ -159$	110 12 115 -114	114 14 218 226	-176 18 57 72		k = 0, l = 4		-79 -2 651 -697	474 401 374	1506 446 427	98 -8 703 -674	73 -10 326 -299	-12 608 573	-16 148 145	-18 38 34	90 -20 63 -57	38 -22 143143	-100 -24 104  92	63	k = 1, l = 4		1 6572	171 3 343 342	59 5 73 -77	57 7 71 75	-121 9 38 16	-130 11 26 24
131 -139	112 - 106 k = 8, l = 3	88 84	92 92 2 129 -117	6 82 67	t = 5, l = 3 $t = 0, 1 = 2$	100 111 $K = 5, t = 5$	88 91 -2 89 108	105 97	35 -24 k = 0, l = 4	31 24	57 59 0 246 -271	138 -133 2 28 37	71 -73 4 376 -425	6 413 395	r = 6, l = 3 8 93 93	10  164  -159	115 110 12 115 -114	107 114 14 218 226	159 -176 18 57 72		k = 6, l = 3 $k = 0, l = 4$		87 -79 -2 651 -697	57 47 -4 401 374	165 1506 446 427	101 98 -8 703 -674	73 73 -10 326 -299	-12 608 573	<i>i</i> = 7, <i>l</i> = 3 - 16 148 145	-18 38 34	99 90 -20 63 -57	4038 -22 143 -143	115 -100 -24 104 92	68 63	k = 1, l = 4	<i>i</i> = 7, <i>l</i> = 3	1 6572	163 171 3 343 342	64 59 5 73 -77	60 57 7 71 75	117 -121 9 38 16	123 -130 11 26 24
9 131 -139	11 112 -106 $k = 8, l = 3$	1.5 88 84	15 92 92 2 129 -117	6 82 67	$k = 5, l = 3$ $k = 0, \dots, 2$	$k = \delta, l = 3$ 3 100 111	-5 88 91 -2 89 108	-7 105 97	-9 35 $-24$ $k = 0$ , $l = 4$	-11 31 24	-13 57 59 0 246 -271	-17 138 -133 2 28 37	-19 71 -73 4 376 -425	6 413 395	k = 6, l = 3 8 93 93	10 164 -159	8 115 110 12 115 -114	10 107 114 14 218 226	12 159 -176 18 57 72		k = 6, l = 3 $k = 0, l = 4$		-2 87 -79 -2 651 -697	-8 57 47 -4 401 374	-10 165 150 $-6$ 446 427	-12 101 98 $-8$ 703 $-674$	-16 73 73 $-10$ 326 $-299$	-12 608 573	k = 7, l = 3 $-16$ 148 145	-18 38 34	, 1 99 90 -20 63 -57	3  40  -38  -22  143  -143	5 115 -100 -24 104 · 92	7 68 63	k = 1, l = 4	k = 7, l = 3	1 6572	-1 163 171 3 343 342	-3 64 59 5 73 -77	-5 60 57 7 71 75	-9 117 -121 9 38 16	-11 123 -130 11 26 24
= 3 9 131 -139	11 112 -106 k=8, l=3	-40 I3 88 84	-123 15 92 92 2 129 -117	67 67 682 67	20 $k = 5, l = 3$ $k = 6, l = 3$	-78 $-78$ $-7$ 100 111 $K = 5, l = 3$	-5 88 $91$ $-2$ 89 $108$	= 3 -7 105 97	-9 35 $-24$ $k = 0, l = 4$	-59 -11 31 24	61 -13 57 59 0 246 -271	305 -17 138 -133 2 28 37	-55 -19 71 -73 4 376 -425	106 6 413 395	k = 6, l = 3 8 93 93	-106 10 164 -159	-98 8 115 110 12 115 -114	-126 10 107 114 14 218 226	12 159 -176 18 57 72		k = 6, l = 3 $k = 0, l = 4$	-168	-113 -2 87 -79 -2 651 -697	488 57 474 401 374	43 -10 165 1506 446 427	-12 101 98 $-8$ 703 $-674$	= 3 $-16$ $73$ $73$ $-10$ $326$ $-299$	-12 608 573	-306 $k = 7$ , $l = 3$ $-16$ 148 145	-323 -18 38 34	-143 , 1 99 90 $-20$ 63 $-57$	-145 3 40 $-38$ $-22$ 143 $-143$	116 5 115 -100 -24 104 92	204 7 68 63	157 $k = 1, l = 4$	k = 7, l = 3	= 3 1 6572	-1 163 171 3 343 342	-103 -3 64 59 5 73 -77	113 -5 60 57 7 71 75	42 -9 117 -121 9 38 16	60 -11 123 -130 11 26 24
$\dot{c} = 3, \ l = 3$ 9 131 -139	k = 8, l = 3	50 -40 I3 88 84	118 -123 15 92 92 2 129 -117	63 67 67 682 67	31  20  k = 5, l = 3 $l = 6 l = 2$	2) 12 12 10 11 1 12 12 20 12 2	-5 88 $91$ $-2$ 89 $108$	r = 3, l = 3 $-7$ 105 $97$	-9 35 $-24$ $k = 0, l = 4$	55 -59 -11 31 24	71 61 -13 57 59 0 246 -271	317 305 -17 138 -133 2 28 37	70 -55 -19 71 -73 4 376 -425	107 106 6 413 395	64 $68$ $k = 6, l = 3$ $8$ $93$ $93$	99 -106 10 164 -159	95 –98 8 115 110 12 115 –114	125 -126 10 107 114 14 218 226	12 159 -176 18 57 72	x = 4, l = 3	k = 6, l = 3 $k = 0, l = 4$	171 -168	115 -113 -2 87 -79 -2 651 -697	54488 57 474 401 374	58 43 -10 165 1506 446 427	-12 101 98 $-8$ 703 $-674$	t = 4, l = 3 - 16 73 73 - 10 326 - 299	-12 608 573	285 - 306  k = 7, l = 3  -16  148  145	346 -323 -18 38 34	157 -143 1 99 90 -20 63 -57	141 -145 3 40 -38 -22 143 -143	103 116 5 115 -100 -24 104 92	194 204 7 68 63	162 157 $k = 1, l = 4$	k = 7, l = 3	= 5, l = 3 1 6572	-1 163 171 3 343 342	104 -103 -3 64 59 5 73 -77	109 113 -5 60 57 7 71 75	35 $42$ $-9$ $117$ $-121$ $9$ $38$ $16$	41 60 -11 123 -130 11 26 24

$10F_{\rm c}$	9	83	-95	198	-34	-38		<u> </u>	-69	-176	~280	-221	-42	128	134	126 126	-182		ú.	100	- 19U	46	-113	46	59		,	93	44	-39	132	29 1 1	DOT-	9	t	66
10/ <i>F</i> ol	= 3, <i>l</i> =	73	305 305	200	28	32		= 3, l = 1	63	177	283	225	41	132	142	114	181		= 4, <i>l</i> = 1	205	507 50	84	107	39	69	- 7 V =	-	86	47	47	140	60 76	ţ	= 5, 1 =	5	84 65
ų	k		n v		11	13		×	-	- <del>(</del>	-5	L -	6-	-	-13	01-	23		k	C	20	14	9	8	10	4	ć	44	9 - - 9	00 0   7		717	1	k	,	~ ~
10 <i>F</i> c	49	36	80	9		63	-130	-154 -218	64-	478	-87	-83	26	57	-20	6	9		-130	-32	-56 -	88	-99	85		¢	226	226	123	195	000	-187	24	- 84	171	\$71
$10 F_0 $	54 53	43	9/	k = 1, l =		55	148	158 230	46	473	73	88	24	62	32	3	k = 2, l =		142	17	54	85	105	91		(= 7, 1 = 3	228	226	118	186	200	190	34	18	164	cit
ų	6	121	0			1	Ϋ́	א ר 	- 6 -	-11	-13	-15	-17	-19	- 21	4			0 (	7 4	<b>x</b>	10	14	18		•	-2	4	9 ( 	2 0 1 -	01-	- 14	8[-	20	-22	<b>47</b> –
$10F_{\rm c}$	-93	5	138	<i>LL</i>	-105		5	-105	-118		6		169	146	20	-100	62	-161	-133	9	2	-23	317	-179	-61	142	49	122	-42	-40	100	6	<b>,</b>	133	-132	-106 38
$10 F_0 $	95	c = 7, l =	116	64	90		k = 8, <i>l</i> =	1.0	106		k = 0, l = 0		195	146 20	28	93	55	163	127	=     = 3	<b>1</b>	69	334	180	56	C/ T	57	130	48	49	107	(=1, /=		132	134	94 27
ų	L	-	ī	ί Π	-13		-	(- -	4			0	0 (	~ ·	4 V	$10^{\circ}$	12	14	16		-	-2	4-	,   	20 00   •	11	-14	-16	- 18	07-	77	1		F	τŅ Έ	- L
$10F_{c}$	-126		n	191	57	-116	-93	Ś	<b>,</b>	-105	-80	-48	40	191	- 54		5		-120	121	-200	74	-138	-73	80	v		118	L,	n	26	686	117		5 137	132
$10 F_0 $ $10F_c$	131 -126 118 108		K = 4, I = 0	192 191	64 57	105 -116	97 –93	k = 5, 1 = 5		93 -105	75 -80	50 -48	41 40	181 191	10754 10705		k = 5, l = 5		124 -120	30	200 -200	72 74	130 -138	64 –73	78 80	k = 6, l = 5		111 118		c = 1, $q = 3$	10 75	94 98	123 117		k = 7, l = 5	142 1.32 142 1.32
$h$ 10 $ F_0 $ 10 $F_c$	4 131 -126 10 118 108		K = 4, l = 5	6 192 191	8 64 57		-22 97 -93	k = 5, l = 5	, ,	1 93 ~105	3 75 -80	5 50 -48	7 41 40		11 5854 13 10705		k = 5, l = 5			-2 174 171	-7 200 -200	-11 72 74	-13 130 -138	-15 64 -73	-21 78 80	k = 6, l = 5		2 111 118		c = 1, 0 = 3	21 01 75	-8 94 98	-14 123 117		k = 7, l = 5	1 142 132 1 142 132
$10F_{\rm C}$ h $10F_{\rm O}$   $10F_{\rm C}$	-73 4 131 -126 -137 10 118 108		K = 4, 1 = 5	6 192 191	49 8 64 57	44 12 105116	197 -22 97 -93	254   k = 5   l = 5	124	20 1 93 -105	-52 3 75 -80				11 5854545454		$-166  k = 5, \ l = 5$	43	-44 $-1$ 124 $-120$	70 -5 30 -15	43 -7 200 -200	75 -11 72 74	102 -13 130 -138	-15 64 -73	5	k = 6, l = 5	-223	-286 2 111 118	-313 $t = 0$	-150  k = 0, l = 0		53 8 94 98	41 -14 123 117	212	k = 7, l = 5 1 + 42, l = 5	1 142 132
$10 F_0  = 10F_0 = h = 10 F_0  = 10F_0$	77 -73 4 131 -126 127 -137 10 118 108		k = 4, l = 5 k = 4, l = 5	6 192 191	54 49 8 64 57	28 44 12 105 -116	216 197 -22 97 -93	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	132 124	30 20 1 93 -105	48 -52 3 75 -80	149 -156 5 50 -48	115 - 129 7 41 40	80 -23 9 181 191	= 3 1 = 5 11 5854		$165 - 166  k = 5, \ l = 5$	59 43	5244 -1 124 -120	77 $70$ $-5$ $30$ $-15$	58 43 -7 200 -200	70 75 -11 72 74	90 102 -13 130 -138		= 3, l = 5 $-21$ 78 80	144 $-141$ $k = 6, l = 5$	223 -223	278 -286 2 111 118	326 - 313 303 166 1 6 1 - 6	k = 0, l = 0 k = 0, l = 0 k = 0, l = 0	132 - 130 A3 A0 6 01 75	48 538 94 98	50 41 -14 123 117	208 212	k = 7, l = 5 r = 4, l = 5 r = 1, 1, 2, 2, 2, 3, 3, 3, 3, 3, 3, 3, 3, 3, 3, 3, 3, 3,	1 142 132
$h = 10 F_0  = 10F_0 = h = 10 F_0  = 10F_0$	10 77 -73 4 131 -126 16 127 -137 10 118 108		k = 2, l = 5 $k = 4, l = 5$	6 192 191	-2 54 498 64 57	-4 28 44 -12 105 -116	-6 216 197 -22 97 -93	-6 242 233 -10 277 254 $k=5$ $l=5$	-12 132 124	-14 30 20 1 93 -105	-16 48 $-52$ 3 75 $-80$	-20 149 $-156$ 5 50 $-48$		-24 80 $-23$ 9 181 191	k = 3 $l = 5$ 11 5854 $k = 3$ 12 05		3 $165 - 166 k = 5, l = 5$	<u>5</u> 59 43	7 52 -44 -1 124 -120	11 $77$ $70$ $-5$ $30$ $-15$	13 58 43 -7 200 -200	17 70 75 -11 72 74	19 90 102 -13 130 -138		k = 3, l = 5 21 78 80	-1 144 -141 $k = 6.$ $l = 5$	-3 223 -223	-5 278 -286 2 111 118	-7 326 $-3130 202 100 t = t = t$	-11 150 $-150$ $K = 0, l = 5$	-13 $42$ $40$ $5$ $01$ $75$		-17 50 41 -14 123 117	-21 208 212	k = 7, l = 5 k = 4, l = 5 1 = 142	$n^{-4}, i^{-5}$ 1 142 132
$10F_{\rm C}$ h $10 F_{\rm ol} $ $10F_{\rm c}$ h $10 F_{\rm ol} $ $10F_{\rm c}$	-273 10 77 -73 4 131 -126 -21 16 177 -73 4 131 -126		k = 4, l = 5 k = 2, l = 5	4 6 192 191	-2 54 498 64 57	75 -4 28 44 -12 105 -116		$\begin{array}{cccccccccccccccccccccccccccccccccccc$	54 -12 132 124	-67 $-14$ 30 20 1 93 $-105$	-16 48 $-52$ 3 75 $-80$	5 -20   49 - 156   5   50 -48	-22 115 $-129$ 7 41 40	213 - 24 80 $-23$ 9 181 191	-239 $-239$ $k = 3 l = 5$ 11 58 $-5421 k = 3 l = 5 13 107 -05$	-48	-66 3 165 $-166$ $k = 5, l = 5$	-32 5 59 43	52 7 52 -44 -1 124 -120 0 124 120 7 124 -120	5 11 77 70 -5 30 -15	13 58 43 -7 200 -200	269 17 70 75 -11 72 74	34,6 19 90 102 -13 130 -138		-260 $k=3, l=5$ $-21$ 78 80	2+0 127 1 144 141 $k = 6, l = 5$	-169 -3 223 -223	-106 $-5$ $278$ $-286$ $2$ $111$ $118$	-132 -1 326 -313 36 0 303 160 1-6 1-6	-20 -20 -202 -202 -202 -202 -202 -202 -	$-10^{4}$ $-13$ $A^{2}$ $-100$ $5$ $01$ $7c$	5 -15 48 53 -8 94 98	-17 50 41 $-14$ 123 117	-79 -21 208 212	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	$93$ $n^{-4}$ , $i=3$ $1$ $142$ $132$ $132$
$10 F_0 $ $10F_C$ $h$ $10 F_0 $ $10F_C$ $h$ $10 F_0 $ $10F_C$	259         -273         10         77         -73         4         131         -126           73         -81         16         77         -73         4         131         -126	81 -72 18 105103	110 110 $k = 2, l = 5$ $k = 4, l = 5$	k = 8, l = 4 $-6$ 192 191	-2 54 498 64 57	69 75 $-4$ 28 $44$ $-12$ $105$ $-116$	-6 216 197 -22 97 -93	x = 0, y = 4 $-5, z = 42$ $z = 2.3, z = 5, y = 5, z = 5$	70 54 -12 132 124	79 -67 -14 30 20 1 93 -105	-16 48 $-52$ 3 75 $-80$	k = 1, l = 5 -20 149 -156 5 50 -48		245 $213$ $-24$ $80$ $-23$ $9$ $181$ $191$	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	41 -48	47 $-66$ 3 $165$ $-166$ $k = 5$ , $l = 5$	31 -32 5 59 43	56 52 7 52441 124120 0 124 170 2 124 0	k = 1, l = 5 11 77 70 $-5$ 30 $-15$	13 58 43 -7 200 -200	294 269 17 70 75 -11 72 74	356 346 19 90 102 -13 130 -138		$\begin{array}{cccccccccccccccccccccccccccccccccccc$	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	187 -169 -3 223 -223	104 -106 -5 278 -286 2 111 118	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	166 167 -20 -20 -20 -150 K = 0, l = 0 160 160 160 160 160 160 160 160 160 1	-13 $-13$ $-13$ $-13$ $-13$ $-13$ $-13$ $-13$ $-12$ $-13$ $-15$	k = 2, l = 5 -15 48 53 -8 94 98	-17 50 41 -14 123 117	79 -79 -21 208 212	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	92 93 $n = 1, n = 3, n = 3$

Table 4--continued

-130	-97	-28	102	125		8		-213	156	89	267	258	-65	94	118		×		58	39	82	-45		8		-7	48	13	100	126		×
116	105	36	92	134		= 1, <i>l</i> =		210	162	97	281	254	69	100	107		= 2, 1 =		60	47	86	47		= 2, 1 =		27	52	29	95	131		= 3, 1 =
5	L	6	11	13		k			L-	6-	-11	-13	-15	-17	-23		k		0	~	9	∞		k		-4	9	~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~~	-12	-14		k
	83		= 7		56		8 =		181	43	111	161	-97	-125		= 8		-5	-45	87	204	360	170	44	139	74	68		8 ::		-60	22
	90		k = 7, l		67		k = 0, l		193	54	108	165	79	133		k = 0, 1		25	46	66	183	377	172	53	133	65	67		k = 1, 1		69	32
	ŝ		-		L-				0	7	4	9	10	12				-2	4-	9-	8	-10	12	- 14	-16	-18	-22				1	ŝ
-62	- 96	-94	96	-100		= 7		29	-106		L =		-37	-137	123	64	110		= J		LL	-88		= 7 -		142	230	95	97	76		L =
76	101	95	98	94		k = 5, 1		34	112		k = 5, l		29	132	117	56	117		k = 6, l		85	87		k = 6, l		128	229	06	66	81		k = 7, 1
9-	-8	-14	-16	-18				-	S				-	۲. ۱	-5	6-	-13				9	×				-2	-4	91	-12	-14		
273	245	46	-117	- 79		1 = 7		-35	102	-85	<i>LL</i>	95		l = J		-125	-229	-139	-199	-96	-117	83	157		1 = 1		-35	70	-67		L = 1	
281	238	54	126	81		k = 3,		49	106	83	70	66		k = 3,		123	224	146	180	108	117	61	155		k = 4, .		57	92	82		k = 4,	
9-	-8	-12	-18	-72				Ι	m	5	7	13				-	÷.	5	L	- 6	-11	-13	19				0	10	12			
38	258	-31	109	82	-121				33	-30	-21	-28	250	128	-146	29	123	-67				-39	46	27	126	-86	- 33				69	395
50	250	29	112	86	121		k = 1, l = 7		33	37	39	33	254	133	147	30	126	84		k = 2, l = 7		30	26	44	133	82	30		k = 2, l = 7		72	394
-	č	S	6	11	13				-	 1	:5	L	6	11	-13	-15	-17	21				0	0	9	×	10	12				- 7	4-
= 6		-149		= 6		168	225	-137		= 6		-79	173		= 6		-102	-176	-43	87	107	52		= 6		97	-81	-136	81		= 1	
k = 5, l		147		k = 5, l		172	227	125		k = 6, l		83	177		k = 6, l		98	175	37	85	110	65		k = 7, l		95	83	142	LL		k = 1, l	
		11				-	÷.	-15				9	10				- 2	4	9-	-8	-10	-12		-		<del>د</del> .	-5	L	-13		•	
11	3																															

10F	114 -67 0	100 86 0	109 -124 0	-117 -117 1 -117 -117 -117 -104	1 114 2 -100
$ 0 F_0 $	101 89 3, /= 1	94 95 :4, <i>l</i> = 1	97 124 : 5, <i>l</i> = 1	84 105 1, <i>l</i> = 1 77 70 91	2, <i>l</i> = 1 113 0, <i>l</i> = 1 82
h 1		11 13 &=	-8 -12 k = k		k = -10 $k = -10$ $k = -10$ $k = -10$
$10F_{c}$	-144 -104 137	10 -73 -61 86	-134 -150 -100 10	124 104 10 60 69	-114 -144 101 125 125 -13
$10 F_0 $	135 108 122	r = 0, t = 65 66 27 99	134 130 77 t = 1, t =	133 107 c = 1, <i>l</i> = 66 85	107 144 96 120 (= 2, <i>l</i> = 33 33
ч И	408	4 7 7 7 7 8 9 8	-12 -12 -20	ve v.v.	
$10F_{c}$	120 61 110 - 76	67 18- 18- 18- 18- 18- 18- 18- 18- 18- 18-	139 9 - 145	-230 -277 -204 -2204 -220 -142 -220 -142 -220 -142 -220 -142 -142 -142 -142 -142 -142 -142 -142	86 866 116 159 -116 -116 10
10 F <sub>o</sub>	118 65 79	79 5 = 4, 1 = 98 80	144 : = 4, <i>l</i> = 148	230 269 202 144 : = 5, <i>l</i> =	98 86 1120 153 104 (= 0, <i>I</i> = 103
ų	$-\frac{1}{2}$	-17 4 2	-2 8	-6666666610681018	
10Fc	29 54 48	/5 140 -111	52 111 76 9	$^{22}_{-107}$ $^{-103}_{-61}$ $^{-58}_{-58}$	9 -53 -85 9 -117 117 138
$10 F_0 $		. 1 =			11 11
	5125	80 144 91 6 = 2	57 116 81 6 = 2,	40 88 88 66 51 45	<pre>k = 3, l: 53 86 k = 3, l: 139 139</pre>
ч	-11 -11 -11 -11	$\begin{array}{c} -13 & 80 \\ -15 & 144 \\ -21 & 91 \\ k = 2 \end{array}$	$\begin{array}{ccc} 0 & 57 \\ 2 & 116 \\ 8 & 81 \\ k = 2, \ 4 \end{array}$	2 40 4 88 6 113 8 66 -12 51 -12 51	$k = 3, l^{3}$ $1  53$ $3  86$ $k = 3, l^{3}$ $k = 3, l^{3}$ $\cdots 3  112$ $\cdots 5  139$
10Fc h	-94 -9 5 -67 -11 4	$\begin{array}{cccc} -13 & 80 \\ -15 & 144 \\ -21 & 91 \\ 117 & -21 & 91 \\ 141 & k = 2 \\ 161 \\ 101 \end{array}$	$\begin{array}{cccc} 0 & 57 \\ 2 & 116 \\ 8 & 81 \\ 107 & k = 2, \end{array}$	-112 -2 40 -4 88 -4 88 -125 -6 113 -125 -8 66 -12 51 -14 45	k = 3, t = 109 $k = 3, t = 130$ $k = 3, 1 = 130$ $k = 3, t = 13$ $k = 3, t = 130$ $104 = -5 = 139$
$10 F_0 $ $10F_c$ h	= 5, <i>l</i> = 8 -5 5 97 -94 -9 5 72 -67 -11 4	z = 5, l = 8 -13 80 z = 5, l = 8 -15 144 115 117 -1 91 117 -1 $k = 2$ 140 141 $k = 2$ 181 101	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	z = 6, l = 8 $-2$ 40 111 -112 -6 113 127 -125 -8 66 70 -66 -12 51 -14 45	$\begin{array}{cccccccccccccccccccccccccccccccccccc$
$h = 10 F_0  = 10F_c = h$	$k = 5, l = 8 \qquad -5 \qquad 5 \qquad -7 \qquad 12 \qquad 5 \qquad 5 \qquad 7 \qquad 7 \qquad 7 \qquad 7 \qquad 7 \qquad 7 \qquad -9 \qquad 5 \qquad 7 \qquad 7 \qquad 7 \qquad 7 \qquad 7 \qquad 7 \qquad -67 \qquad -11 \qquad 4 \qquad 5 \qquad 5$	k = 5, l = 8  -1.3  80  km k = 5, l = 8  -1.5  144  -2.1  914  -2.1  914  -2.1  914  km k = 2  -2.1  914  km k = 2  -2.1  916  +2.1  916  +2.1  916  +2.1  916  +2.1  -2.1  916  +2.1  +	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	$k = 6, l = 8 \qquad -2 \qquad 40$ $-2 \qquad 111 \qquad -112 \qquad -6 \qquad 113$ $-4 \qquad 127 \qquad -125 \qquad -8 \qquad 66$ $-10 \qquad 70 \qquad -66 \qquad -12 \qquad 51$ $k = 1 \qquad l = 5$	$\begin{array}{cccccccccccccccccccccccccccccccccccc$
$10F_{\rm c}$ $h$ $10 F_{\rm o} $ $10F_{\rm c}$ $h$	$k = 5, l = 8 \qquad -5 \qquad 5$ -111 $k = 5, l = 8 \qquad -5 \qquad 5$ -7 $12$ -04 $-9 \qquad 5$ -111 $7 \qquad 72 \qquad -67 \qquad -11 \qquad 4$	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	$\begin{array}{cccccccccccccccccccccccccccccccccccc$
$10 F_0 $ $10 F_c$ $h$ $10 F_0 $ $10 F_c$ $h$	k = 5, l = 8 $k = 5, l = 8$ $-5 = -7$ $36 = 46$ $5 = 97$ $-94 = -9$ $5$ $110 = -111$ $7 = 72$ $-67 = -11$ $4$	93     89 $k = 5, l = 8$ $-13$ 80 $r = 3, l = 8$ $k = 5, l = 8$ $-15$ 144 $r = 3, l = 8$ $-5$ $115$ $-117$ $r = 1$ $-7$ $140$ $141$ $k = 2$ $r = 2$ $-7$ $181$ $101$ $k = 2$	$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	$ \begin{array}{cccccccccccccccccccccccccccccccccccc$

Table 4-continued

between the benzo-groups is  $61.5^{\circ}$ . The cyclobutane rings are *cis*-, *trans*-, *cis*-substituted. The molecular symmetry  $(C_2)$  is far from  $mm2(C_{2v})$  because of the non-planarity of the cyclobutane rings. The deviations of the atoms C(1) through C(4) from the best plane through the cyclobutane ring are 0.139, -0.142, 0.141 and -0.138 Å, respectively, all  $\pm 0.006$  Å. The two dihedral angles formed by the pairs of planes through three carbon atoms having a diagonal in common are both  $152^{\circ}$ . This angle is in the range of 149-155°, reported for many puckered cyclobutane derivatives (Andreetti et al, 1973, and many references therein; Margulis, 1965; Adman & Margulis, 1967).

In the literature, there are two more structure determinations of *cis*, *trans*-, *cis*-substituted cyclobutane containing compounds described: tetracyanocyclobutane (Greenberg & Post, 1968) and cyclobutanetetracarboxylic acid (Margulis, 1971). In both of them, however, the cyclobutane rings are planar.

As is usually found in tetra-substituted cyclobutane rings, the *cis*substituted carbon atoms (1.573(8), 1.606(8) Å) are significantly longer than a single C–C bond of 1.537 Å; the distances between *trans*-substituted carbon atoms (1.542 and 1.554 Å) are only slightly longer.

The packing of the molecules in the unit cell is in agreement with van der Waals distances; no unusual intermolecular contacts occur.

### Acknowledgements

The authors wish to express their thanks to Mr W. P. Bosman, Dr J. H. Noordik, Dr P. Benci and Mr J. M. M. Smits for valuable assistance in the X-ray work. The investigations were supported in part by 'FOMRE' with financial aid from the Netherlands Organization for Advancement of Pure Research (ZWO).

#### References

Adman, E. & Margulis, T. N. (1967) Chem. Comm. 641.

Andreetti, G. D., Bocelli, G. & Sgarabotto, P. (1973) Cryst. Struct. Comm. 2, 115.

Beurskens, P. T. (1963) Technical Report on Sign Correlation by the Sayre Equation. The Crystallography Laboratory, Univ. of Pittsburgh, Pittsburgh, Pennsylvania.

Duax, W. L., Weeks, C. M. & Hauptman, H. (1972) Acta Cryst. B28, 1857.

Greenberg, B. & Post, B. (1968) Acta Cryst. B24, 918.

Hauptman, H. & Karle, J. (1957) Acta Cryst. 10, 267.

Hauptman, H. & Karle, J. (1958) Acta Cryst. 11, 149.

Kanters, J. A., Kroon, J., Beurskens, P. T. & Vliegenthart, J. A. (1966) Acta Cryst. 21, 990.

Karle, J. & Hauptman, H. (1957) Acta Cryst. 10, 515.

Karle, J. & Hauptman, H. (1958) Acta Cryst. 11, 264.

Karle, J. & Hauptman, H. (1959) Acta Cryst. 12, 404.

242 TH. E. M. VAN DEN HARK, P. T. BEURSKENS AND W. H. LAARHOVEN

Laarhoven, W. H., Cuppen, Th. J. H. M. & Nivard, R. J. F. (1970) Tetrahderon 26, 1069.

Laarhoven, W. H. & Cuppen, Th. J. H. M. (1972) J. Chem. Soc. Perkin I, 2074.

Margulis, T. N. (1965) Acta Cryst. 19, 857.

Margulis, T. N. (1971) J. Am. Chem. Soc. 93, 2193.

Meinwald, J. & Young, J. W. (1971) J. Am. Chem. Soc. 93, 725.

Müller, E., Sauerbier, M. & Heiss, J. (1966) Tetrahedron Letters, 2473.

Müller, E., Meier, H. & Sauerbier, M. (1970) Chem. Ber. 103, 1356.

Ottenheijm, H. C. J. (1973) Personal communication of unpublished results.