*J. Cryst. Mol. Struct.* (1974) 4, 227-242

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

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*(Received 20 August 1973)*

## Abstract

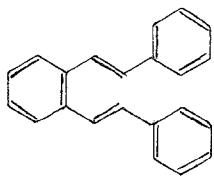
A simple application of the *B*3,0 formula is described. This formula is used mainly to avoid inconsistent $\Sigma_2$-interactions in standard symbolic-addition or multiple-solution techniques. Computer time is reduced by a reflection selection procedure based upon the use of the *B*2,0 formula. The 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,$0^{4,7}$]dodecadiene-2,8, $C_{44}H_{36}$. The compound crystallizes in the monoclinic space group *C*2/*c*, with unit cell parameters $a = 28.047$, $b = 9.504$, $c = 12.600$ Å and $\beta = 103.4°$. 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

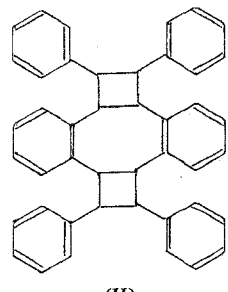
Attention is given to the solution of a phase problem that, at first sight, may be considered as easily solvable, namely a small, monoclinic, centro-

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°).



(I)



(II)

## Experimental

5,6,11,12-tetraphenyl-dibenzo[2-3,8-9] tricyclo [8,2,0,$0^{4,7}$]dodecadiene-2,8 ($C_{44}H_{36}$), $FW$ = 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)°;

$V_c = 3267(3)$ Å$^3$. The calculated density of 1.148 g cm$^{-3}$ with $Z = 4$, agrees with the measured value of 1.13 g cm$^{-3}$ (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 ($I \geqslant 3\sigma$). 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 $|AB + C| \gg 0$:

$$S(E_{\mathbf{h}} E_{\mathbf{h}'} E_{-\mathbf{h}-\mathbf{h}'}) = S(AB + C) \tag{2}$$

Equation (2) can be used only for large values of the triple product $|E_{\mathbf{h}} E_{\mathbf{h}'} E_{-\mathbf{h}-\mathbf{h}'}|$, 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 $\mathbf{h}$ and $\mathbf{h}'$; for space group $C2/c$, with $E \approx E'$:

$$\begin{aligned} (-1)^l |E_{\mathbf{hkl}}|^2 E_{0,2k,0} &= AB + C \\ (-1)^l |E_{\mathbf{hkl}}|^2 E_{2h,0,2l} &= AB + C \end{aligned} \tag{3}$$

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

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

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 $|E| \geqslant 1.7$ (table 1a). The $B2,0$ formula (4) was used to calculate $|E'|$-values of the 90 reflections with $1.4 < |E| < 1.7$. 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 $|E| \geqslant 1.0$.

Table 1. *Distribution of incorrect* B*3,0 results*

| | table 1a: $\|E\| \geqslant 1.7$ | | 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 |
| +10 . . . 0 | 96 | 5 | 37 | 3 |
| 0 . . . −20 | 84 | 68* | 58 | 38* |
| −20 . . . −60 | 16 | 5** | 20 | 4** |

$N$(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).

B*3,0 results.* The results were tabulated according to decreasing values of $(AB + C)$. Eleven letter symbols were assigned to the reflections with $|E| \geqslant 1.7$ 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:

$\mathbf{h}_1$ are the eleven initial choices
$\mathbf{h}_2$ are reflections $\mathbf{h}_1 + \mathbf{h}'_1$
$\mathbf{h}_3$ are reflections $\mathbf{h}_1 + \mathbf{h}_2$ and $\mathbf{h}_2 + \mathbf{h}'_2$.

To find the reflections $\mathbf{h}_2$ and $\mathbf{h}_3$, 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 $\mathbf{h}_1$. Thereafter, new reflections $\mathbf{h}_2$ and $\mathbf{h}_3$ could be calculated, and new reflections with multiple sign indications were added to the set $\mathbf{h}_1$, 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 $\mathbf{h}_1$.

The set $\mathbf{h}_1$ 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 $\Sigma w(|F_o| - |F_c|)^2$, where $1/w = \sigma_c^2 + (0.05|F_o|)^2$ 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

Table 2. *Fractional coordinates and thermal parameters ($\times 10^4$), with esd in parentheses. The anisotropic thermal parameters are in the form:*

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

*The key to atomic numbering is given in figure 1*

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

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* | *y* | *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 |

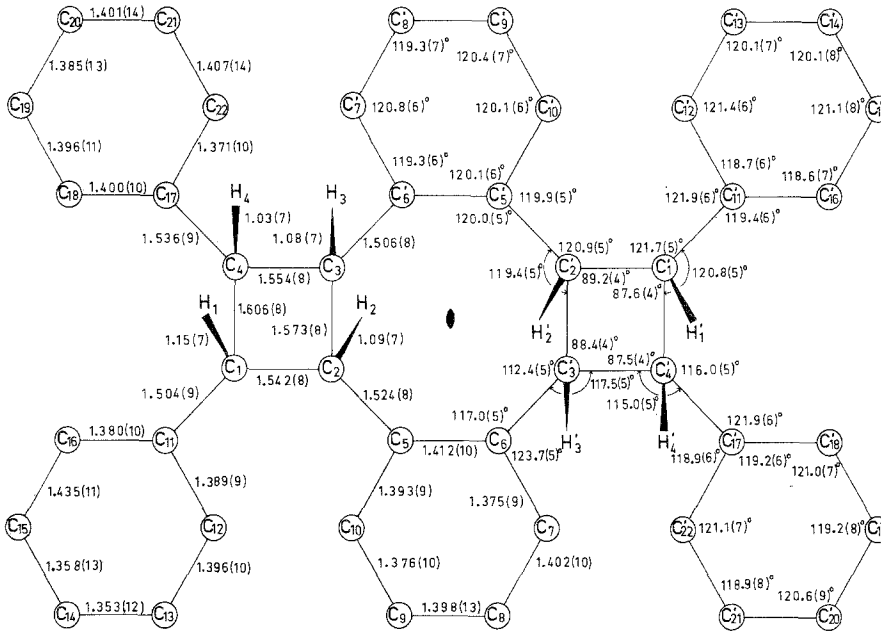



Fig. 1. Bond distances and angles, with esd in parentheses.

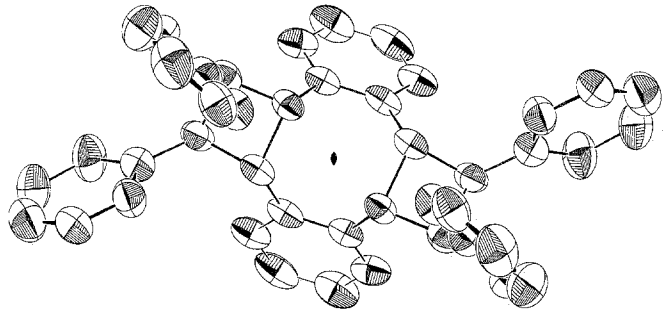


Fig. 2. Thermal ellipsoid plot of the molecule, as seen along the twofold rotation axis.

reflections having $|E| \geqslant 1.7$. 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

Table 4. *Observed and calculated structure factors*

| $h$ | $10\|F_o\|$ | $10F_c$ | $h$ | $10\|F_o\|$ | $10F_c$ | $h$ | $10\|F_o\|$ | $10F_c$ | $h$ | $10\|F_o\|$ | $10F_c$ | $h$ | $10\|F_o\|$ | $10F_c$ | $h$ | $10\|F_o\|$ | $10F_c$ |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| $k = 0,\ l = 0$ | | | $k = 3,\ l = 0$ | | | 10 | 80 | −82 | $k = 2,\ l = 1$ | | | −17 | 36 | −21 | −13 | 71 | 74 |
| | | | | | | 14 | 111 | 116 | | | | −19 | 80 | −78 | −15 | 121 | 122 |
| 4 | 240 | −255 | 1 | 217 | −226 | | | | 2 | 46 | −34 | | | | −17 | 33 | 44 |
| 6 | 172 | 155 | 3 | 298 | −311 | $k = 7,\ l = 0$ | | | 4 | 423 | −425 | $k = 4,\ l = 1$ | | | −19 | 113 | −111 |
| 8 | 192 | −208 | 5 | 45 | −49 | | | | 6 | 281 | 291 | | | | | | |
| 10 | 353 | −349 | 7 | 55 | 49 | 1 | 82 | 94 | 8 | 155 | 163 | 2 | 56 | 49 | $k = 6,\ l = 1$ | | |
| 12 | 209 | −198 | 9 | 124 | −106 | 3 | 63 | 42 | 10 | 330 | −331 | 4 | 262 | −250 | | | |
| 14 | 494 | −476 | 13 | 55 | −55 | 7 | 78 | −85 | 14 | 164 | 164 | 6 | 333 | −290 | 4 | 93 | −93 |
| 16 | 270 | −277 | 15 | 109 | 124 | 9 | 73 | 70 | 18 | 122 | 118 | 8 | 141 | −127 | 10 | 102 | −93 |
| 18 | 78 | 80 | 19 | 62 | 68 | 13 | 86 | −80 | 20 | 72 | 66 | 10 | 233 | −220 | | | |
| 20 | 151 | 148 | 21 | 54 | 52 | | | | | | | 12 | 69 | 74 | $k = 6,\ l = 1$ | | |
| 22 | 69 | −73 | | | | $k = 8,\ l = 0$ | | | $k = 2,\ l = 1$ | | | 14 | 99 | 97 | | | |
| 24 | 137 | −146 | $k = 4,\ l = 0$ | | | | | | | | | 18 | 226 | 219 | −2 | 125 | 131 |
| 26 | 83 | 84 | | | | 0 | 154 | −142 | −2 | 531 | −531 | 20 | 142 | 144 | | | |
| | | | 0 | 375 | 344 | 2 | 128 | −128 | −4 | 72 | 77 | | | | $k = 7,\ l = 1$ | | |
| $k = 1,\ l = 0$ | | | 2 | 66 | 58 | 4 | 154 | −154 | −6 | 120 | −118 | $k = 4,\ l = 1$ | | | | | |
| | | | 4 | 354 | −341 | 8 | 95 | 90 | −8 | 25 | 47 | | | | 1 | 140 | −136 |
| 3 | 105 | −120 | 6 | 53 | −52 | | | | −10 | 96 | −100 | −2 | 332 | 323 | 3 | 27 | −29 |
| 5 | 326 | −349 | 8 | 145 | 133 | $k = 1,\ l = 1$ | | | −12 | 54 | 56 | −4 | 53 | −51 | 5 | 84 | 76 |
| 7 | 351 | 353 | 10 | 77 | −77 | | | −16 | −16 | 29 | −29 | −6 | 286 | −295 | 7 | 57 | −61 |
| 9 | 32 | −33 | 12 | 126 | −123 | 1 | 72 | 67 | −18 | 69 | 64 | −8 | 140 | −123 | 9 | 92 | 90 |
| 11 | 423 | 393 | 16 | 39 | 37 | 3 | 890 | 990 | −20 | 90 | 89 | −10 | 288 | −279 | | | |
| 13 | 202 | 191 | 18 | 122 | 131 | 5 | 41 | 42 | −22 | 33 | −22 | −22 | 109 | 119 | $k = 7,\ l = 1$ | | |
| 15 | 127 | 123 | 20 | 75 | 76 | 7 | 115 | −111 | | | | | | | | | |
| 17 | 131 | −129 | | | | 9 | 106 | −111 | $k = 3,\ l = 1$ | | | $k = 5,\ l = 1$ | | | −1 | 67 | −54 |
| 19 | 69 | −64 | $k = 5,\ l = 0$ | | | 11 | 125 | −122 | | | | | | | −5 | 44 | −45 |
| 21 | 68 | −65 | | | | 17 | 113 | −110 | 1 | 84 | −90 | 1 | 149 | 152 | −7 | 48 | 48 |
| 23 | 162 | 165 | 1 | 122 | 118 | | | | 3 | 25 | −40 | 3 | 78 | 60 | −9 | 120 | 121 |
| | | | 3 | 235 | 232 | $k = 1,\ l = 1$ | | | 5 | 340 | 337 | 5 | 107 | 111 | | | |
| $k = 2,\ l = 0$ | | | 5 | 124 | 124 | | | | 7 | 225 | 209 | 9 | 56 | 64 | $k = 8,\ l = 1$ | | |
| | | | 7 | 192 | 202 | −1 | 175 | −325 | 9 | 20 | −1 | 11 | 52 | 48 | | | |
| 0 | 516 | 546 | 9 | 212 | 214 | −3 | 187 | 212 | 11 | 207 | 209 | 13 | 27 | 38 | 2 | 121 | 134 |
| 2 | 285 | −297 | 11 | 78 | 87 | −5 | 519 | 503 | 13 | 66 | −68 | 15 | 70 | −64 | 4 | 143 | 143 |
| 4 | 160 | −164 | 15 | 140 | −155 | −7 | 96 | 108 | 17 | 89 | −76 | 17 | 210 | −209 | 6 | 142 | 135 |
| 6 | 291 | −298 | 17 | 227 | −244 | −9 | 30 | −36 | 21 | 121 | −126 | 19 | 90 | −94 | 8 | 102 | −93 |
| 8 | 227 | 215 | 19 | 103 | −117 | −11 | 72 | −66 | | | | | | | 10 | 131 | −133 |
| 10 | 47 | −33 | | | | −13 | 262 | −257 | $k = 3,\ l = 1$ | | | $k = 5,\ l = 1$ | | | | | |
| 12 | 23 | 40 | $k = 6,\ l = 0$ | | | −15 | 76 | 78 | | | | | | | $k = 8,\ l = 1$ | | |
| 14 | 39 | −21 | | | | −17 | 109 | 109 | −3 | 110 | −102 | −1 | 213 | −215 | | | |
| 16 | 41 | −50 | 0 | 95 | 87 | −19 | 132 | −131 | −5 | 177 | 167 | −3 | 152 | −158 | −6 | 120 | 129 |
| 18 | 72 | 65 | 2 | 26 | 28 | −21 | 42 | −25 | −7 | 29 | 24 | −5 | 219 | 200 | −10 | 145 | −163 |
| 20 | 72 | 67 | 4 | 156 | −165 | −23 | 73 | 54 | −9 | 392 | 362 | −7 | 48 | −41 | −12 | 82 | −65 |
| 24 | 100 | −104 | 6 | 101 | −103 | −25 | 69 | −68 | −13 | 266 | −265 | −9 | 81 | 82 | | | |
| | | | 8 | 74 | −70 | | | | −15 | 157 | 153 | −11 | 124 | 124 | $k = 9,\ l = 1$ | | |

Table 4–*continued*

| $h$ | $10\|F_o\|$ | $10F_c$ | $h$ | $10\|F_o\|$ | $10F_c$ | $h$ | $10\|F_o\|$ | $10F_c$ | $h$ | $10\|F_o\|$ | $10F_c$ | $h$ | $10\|F_o\|$ | $10F_c$ | $h$ | $10\|F_o\|$ | $10F_c$ |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | 17 | 73 | 82 | −20 | 51 | −37 | −4 | 123 | 114 | −18 | 73 | −61 | −3 | 1243 | 1328 |
| 1 | 137 | −134 | | | | | | | −6 | 372 | 369 | −20 | 82 | 48 | −5 | 840 | 838 |
| 3 | 74 | −71 | | $k = 1,\ l = 2$ | | | $k = 3,\ l = 2$ | | −8 | 32 | 12 | | | | −7 | 150 | −148 |
| | | | | | | | | | −10 | 160 | −159 | | $k = 7,\ l = 2$ | | −9 | 322 | −302 |
| | $k = 9,\ l = 1$ | | −1 | 181 | 180 | 1 | 57 | 62 | −12 | 39 | 17 | | | | −11 | 111 | −107 |
| | | | −3 | 242 | −270 | 3 | 213 | −207 | −14 | 34 | 35 | 5 | 142 | −136 | −13 | 54 | 48 |
| −1 | 123 | −122 | −5 | 606 | 607 | 5 | 253 | −245 | −16 | 41 | −41 | 15 | 89 | 69 | −15 | 96 | −101 |
| | | | −7 | 202 | 235 | 7 | 345 | −330 | −18 | 52 | −54 | | | | −17 | 142 | −142 |
| | $k = 0,\ l = 2$ | | −9 | 298 | −295 | 9 | 51 | 51 | −20 | 82 | −85 | | $k = 7,\ l = 2$ | | −19 | 58 | −48 |
| | | | −11 | 109 | 102 | 11 | 114 | −114 | −24 | 89 | 93 | | | | −21 | 65 | −67 |
| 0 | 290 | −316 | −15 | 222 | −215 | 15 | 49 | 40 | | | | −1 | 70 | −94 | −23 | 47 | 53 |
| 2 | 1045 | 1109 | −17 | 55 | 52 | 17 | 102 | 107 | | $k = 5,\ l = 2$ | | −3 | 142 | −152 | | | |
| 4 | 717 | 747 | −19 | 72 | 67 | 19 | 113 | 110 | | | | −5 | 77 | −80 | | $k = 2,\ l = 3$ | |
| 6 | 207 | −208 | −21 | 65 | 56 | | | | 1 | 238 | 239 | −17 | 84 | 72 | | | |
| 8 | 165 | −161 | −23 | 168 | −160 | | $k = 3,\ l = 2$ | | 3 | 163 | 171 | | | | 0 | 77 | 81 |
| 10 | 23 | −12 | −25 | 94 | 88 | | | | 7 | 122 | 130 | | $k = 8,\ l = 2$ | | 2 | 360 | −385 |
| 12 | 359 | 331 | | | | −1 | 319 | 350 | 9 | 64 | −70 | | | | 4 | 190 | 202 |
| 14 | 57 | −62 | | $k = 2,\ l = 2$ | | −3 | 118 | −114 | 11 | 110 | −108 | 0 | 76 | −73 | 6 | 61 | 52 |
| 16 | 139 | −144 | | | | −5 | 182 | −170 | | | | 4 | 71 | −75 | 8 | 312 | 320 |
| 18 | 131 | −122 | 0 | 1133 | −1137 | −7 | 126 | −114 | | $k = 5,\ l = 2$ | | | | | 10 | 282 | 268 |
| 20 | 37 | −38 | 2 | 180 | 175 | −9 | 266 | −247 | | | | | $k = 8,\ l = 2$ | | 14 | 137 | −140 |
| 24 | 110 | 108 | 4 | 54 | 20 | −11 | 209 | −195 | −3 | 132 | −129 | | | | 16 | 62 | −72 |
| | | | 6 | 49 | 46 | −13 | 33 | 33 | −5 | 56 | 47 | −2 | 105 | −102 | 18 | 70 | −57 |
| | $k = 0,\ l = 2$ | | 8 | 140 | 138 | −15 | 45 | 54 | −7 | 139 | 130 | | | | | | |
| | | | 10 | 141 | 130 | −17 | 124 | 124 | −9 | 174 | 166 | | $k = 9,\ l = 2$ | | | $k = 2,\ l = 3$ | |
| −2 | 256 | −298 | 12 | 252 | 250 | −19 | 35 | 38 | −13 | 68 | 68 | | | | | | |
| −4 | 423 | −430 | 14 | 39 | 40 | −21 | 96 | 90 | −19 | 97 | 93 | −1 | 69 | 62 | −2 | 250 | −256 |
| −6 | 1475 | 1490 | 16 | 57 | 52 | | | | −21 | 112 | −118 | | | | −4 | 239 | −254 |
| −8 | 230 | 237 | 18 | 18 | −171 | | $k = 4,\ l = 2$ | | | | | | $k = 1,\ l = 3$ | | −6 | 327 | −313 |
| −10 | 124 | −121 | 20 | 161 | −54 | | | | | $k = 6,\ l = 2$ | | | | | −8 | 93 | −91 |
| −12 | 413 | −389 | 22 | 115 | 123 | 0 | 110 | −101 | | | | 1 | 553 | 596 | −10 | 282 | 266 |
| −14 | 175 | −168 | 24 | 90 | 77 | 2 | 213 | 183 | 0 | 127 | −129 | 3 | 84 | 93 | −12 | 92 | −87 |
| −18 | 94 | 97 | | | | 4 | 47 | −45 | 2 | 30 | 29 | 5 | 421 | −447 | −14 | 66 | 76 |
| −20 | 234 | −216 | | $k = 2,\ l = 2$ | | 6 | 33 | −9 | 4 | 225 | 225 | 7 | 164 | 163 | −16 | 189 | 181 |
| −26 | 86 | −90 | | | | 8 | 118 | 122 | 8 | 66 | −70 | 9 | 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 | 99 | −93 |
| | | | −6 | 86 | −54 | 14 | 37 | 41 | | $k = 6,\ l = 2$ | | 15 | 41 | 24 | | | |
| 1 | 1175 | 1264 | −8 | 89 | −95 | 16 | 127 | −122 | | | | 17 | 35 | −21 | | $k = 3,\ l = 3$ | |
| 3 | 779 | −828 | −10 | 193 | 190 | 18 | 115 | −119 | −2 | 77 | 71 | 19 | 35 | 36 | | | |
| 5 | 328 | 340 | −12 | 65 | −68 | | | | −4 | 105 | 111 | | | | 1 | 58 | 62 |
| 9 | 204 | −201 | −14 | 231 | 228 | | $k = 4,\ l = 2$ | | −6 | 30 | 37 | | $k = 1,\ l = 3$ | | 3 | 103 | −115 |
| 11 | 474 | −465 | −16 | 23 | 16 | | | | −8 | 37 | 40 | | | | 5 | 22 | −11 |
| 15 | 66 | 67 | −18 | 41 | 59 | −2 | 223 | −223 | −12 | 32 | −33 | −1 | 16 | −18 | 7 | 220 | 230 |

$k = 3,\ l = 3$

| | | |
|---|---|---|
| 9 | 50 | −40 |
| 11 | 118 | −123 |
| 13 | 63 | 67 |
| 15 | 31 | 20 |
| 19 | 73 | 72 |
| 21 | 80 | −78 |

$k = 3,\ l = 3$

| | | |
|---|---|---|
| −1 | 55 | −59 |
| −3 | 71 | 61 |
| −7 | 317 | 305 |
| −9 | 70 | −55 |
| −13 | 107 | 106 |
| −15 | 64 | 68 |
| −17 | 99 | −106 |
| −19 | 95 | −98 |
| −21 | 125 | −126 |

$k = 4,\ l = 3$

| | | |
|---|---|---|
| 4 | 171 | −168 |
| 6 | 115 | −113 |
| 10 | 54 | −48 |
| 12 | 58 | 43 |

$k = 4,\ l = 3$

| | | |
|---|---|---|
| −2 | 285 | −306 |
| −6 | 346 | −323 |
| −8 | 157 | −143 |
| −12 | 141 | −145 |
| −18 | 103 | 116 |
| −20 | 194 | 204 |
| −22 | 162 | 157 |

$k = 5,\ l = 3$

| | | |
|---|---|---|
| 1 | 104 | −103 |
| 3 | 109 | 113 |
| 5 | 35 | 42 |
| 7 | 41 | 60 |
| 9 | 131 | −139 |
| 11 | 112 | −106 |
| 13 | 88 | 84 |
| 15 | 92 | 92 |

$k = 5,\ l = 3$

| | | |
|---|---|---|
| −3 | 109 | 111 |
| −5 | 88 | 91 |
| −7 | 105 | 97 |
| −9 | 35 | −24 |
| −11 | 31 | 24 |
| −13 | 57 | 59 |
| −17 | 138 | −133 |
| −19 | 71 | −73 |

$k = 6,\ l = 3$

| | | |
|---|---|---|
| 8 | 115 | 110 |
| 10 | 107 | 114 |
| 12 | 159 | −176 |

$k = 6,\ l = 3$

| | | |
|---|---|---|
| −2 | 87 | −79 |
| −8 | 57 | 47 |
| −10 | 165 | 150 |
| −12 | 101 | 98 |
| −16 | 73 | 73 |

$k = 7,\ l = 3$

| | | |
|---|---|---|
| 1 | 99 | 90 |
| 3 | 40 | −38 |
| 5 | 115 | −100 |
| 7 | 68 | 63 |

$k = 7,\ l = 3$

| | | |
|---|---|---|
| −1 | 163 | 171 |
| −3 | 64 | 59 |
| −5 | 60 | 57 |
| −9 | 117 | −121 |
| −11 | 123 | −130 |

$k = 8,\ l = 3$

| | | |
|---|---|---|
| 2 | 129 | −117 |
| 6 | 82 | 67 |

$k = 8,\ l = 3$

| | | |
|---|---|---|
| −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 | 703 | −674 |
| −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 | 65 | −72 |
| 3 | 343 | 342 |
| 5 | 73 | −77 |
| 7 | 71 | 75 |
| 9 | 38 | 16 |
| 11 | 26 | 24 |
| 13 | 144 | −148 |
| 15 | 215 | −210 |
| 19 | 66 | −64 |

$k = 1,\ l = 4$

| | | |
|---|---|---|
| −1 | 357 | 386 |
| −3 | 485 | 513 |
| −5 | 760 | 778 |
| −7 | 33 | −52 |
| −9 | 57 | 53 |
| −11 | 94 | −82 |
| −13 | 28 | −3 |
| −15 | 92 | −88 |
| −17 | 81 | −80 |
| −19 | 125 | −124 |
| −21 | 72 | −64 |
| −23 | 88 | 98 |

$k = 2,\ l = 4$

| | | |
|---|---|---|
| 0 | 138 | 145 |
| 2 | 420 | −412 |
| 4 | 278 | −273 |
| 6 | 65 | 66 |
| 8 | 133 | −140 |
| 10 | 23 | 26 |
| 12 | 143 | −147 |
| 14 | 151 | 156 |
| 16 | 226 | 231 |
| 18 | 128 | 126 |

$k = 2,\ l = 4$

| | | |
|---|---|---|
| −2 | 166 | −147 |
| −4 | 184 | −182 |
| −6 | 331 | −301 |
| −8 | 306 | −290 |
| −10 | 21 | 11 |
| −12 | 97 | 89 |
| −14 | 71 | −70 |
| −16 | 141 | 146 |
| −20 | 166 | 167 |
| −22 | 48 | −53 |
| −26 | 82 | 96 |

$k = 3,\ l = 4$

| | | |
|---|---|---|
| 1 | 203 | −201 |
| 3 | 436 | 431 |
| 5 | 86 | −88 |
| 7 | 63 | 61 |
| 9 | 95 | 103 |
| 11 | 101 | 92 |
| 17 | 59 | −67 |
| 19 | 68 | −85 |

$k = 3,\ l = 4$

| | | |
|---|---|---|
| −1 | 557 | −523 |
| −3 | 114 | −108 |
| −5 | 63 | −64 |
| −7 | 171 | −169 |
| −9 | 75 | −66 |
| −11 | 63 | 52 |
| −13 | 97 | 90 |
| −15 | 24 | −30 |
| −23 | 175 | 181 |

$k = 4,\ l = 4$

| | | |
|---|---|---|
| 0 | 216 | 209 |
| 2 | 29 | 16 |
| 4 | 58 | 47 |
| 6 | 26 | 17 |
| 8 | 35 | 33 |
| 10 | 93 | −103 |
| 12 | 45 | −31 |
| 14 | 35 | 39 |

$k = 4,\ l = 4$

| | | |
|---|---|---|
| −2 | 47 | 45 |
| −4 | 165 | 169 |
| −6 | 73 | 79 |
| −8 | 42 | 36 |
| −10 | 50 | −55 |
| −12 | 190 | 190 |
| −14 | 33 | 35 |
| −16 | 164 | −166 |
| −18 | 72 | −77 |
| −22 | 77 | −66 |

$k = 5,\ l = 4$

| | | |
|---|---|---|
| 1 | 141 | −141 |
| 15 | 133 | 132 |

$k = 5,\ l = 4$

| | | |
|---|---|---|
| −1 | 120 | −119 |
| −3 | 76 | 80 |
| −5 | 168 | 169 |
| −9 | 146 | −138 |
| −13 | 77 | −69 |
| −15 | 85 | 82 |

$k = 6,\ l = 4$

| | | |
|---|---|---|
| 0 | 156 | 155 |
| 2 | 93 | 97 |
| 8 | 185 | 199 |
| 10 | 85 | −70 |
| 12 | 195 | −200 |
| 14 | 87 | −93 |

$k = 6,\ l = 4$

| | | |
|---|---|---|
| −2 | 238 | 231 |
| −4 | 133 | 114 |
| −6 | 124 | −117 |
| −12 | 118 | 116 |
| −18 | 79 | 74 |

$k = 7,\ l = 4$

| | | |
|---|---|---|
| 1 | 274 | −277 |
| 3 | 88 | −78 |
| 11 | 134 | 136 |

$k = 7,\ l = 4$

Table 4—*continued*

| $h$ | $10\|F_o\|$ | $10F_c$ | $h$ | $10\|F_o\|$ | $10F_c$ | $h$ | $10\|F_o\|$ | $10F_c$ | $h$ | $10\|F_o\|$ | $10F_c$ | $h$ | $10\|F_o\|$ | $10F_c$ | $h$ | $10\|F_o\|$ | $10F_c$ |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| −1 | 259 | −273 | 10 | 77 | −73 | 4 | 131 | −126 | 7 | 95 | −93 | 9 | 54 | 49 | | $k = 3,\ l = 6$ | |
| −3 | 73 | −81 | 16 | 122 | −137 | 10 | 118 | −108 | | | | 11 | 52 | 42 | | | |
| −11 | 81 | −72 | 18 | 105 | −103 | | | | | $k = 7,\ l = 5$ | | 13 | 43 | 36 | 1 | 73 | 83 |
| −15 | 110 | 116 | | | | | $k = 4,\ l = 5$ | | | | | 15 | 76 | 80 | 3 | 95 | −95 |
| | | | | $k = 2,\ l = 5$ | | | | | −1 | 116 | 138 | | | | 5 | 305 | 316 |
| | $k = 8,\ l = 4$ | | | | | −6 | 192 | 191 | −3 | 64 | 77 | | $k = 1,\ l = 6$ | | 7 | 200 | 198 |
| | | | −2 | 54 | 49 | −8 | 64 | 57 | −13 | 90 | −105 | | | | 11 | 28 | −34 |
| 0 | 69 | 75 | −4 | 28 | 44 | −12 | 105 | −116 | | | | −1 | 55 | 63 | 13 | 32 | −38 |
| | | | −6 | 216 | 197 | −22 | 97 | −93 | | $k = 8,\ l = 5$ | | −3 | 148 | −130 | | | |
| | $k = 8,\ l = 4$ | | −8 | 242 | 233 | | | | | | | −5 | 158 | −154 | | $k = 3,\ l = 6$ | |
| | | | −10 | 277 | 254 | | $k = 5,\ l = 5$ | | −2 | 91 | −105 | −7 | 230 | −218 | | | |
| −2 | 70 | 54 | −12 | 132 | 124 | | | | −4 | 106 | −118 | −9 | 46 | −43 | −1 | 63 | −69 |
| −8 | 79 | −67 | −14 | 30 | 20 | 1 | 93 | −105 | | | | −11 | 473 | 478 | −3 | 177 | −176 |
| | | | −16 | 48 | −52 | 3 | 75 | −80 | | $k = 0,\ l = 6$ | | −13 | 73 | −87 | −5 | 283 | −280 |
| | $k = 1,\ l = 5$ | | −20 | 149 | −156 | 5 | 50 | −48 | | | | −15 | 88 | −83 | −7 | 225 | −221 |
| | | | −22 | 115 | −129 | 7 | 41 | 40 | 0 | 195 | 169 | −17 | 24 | 26 | −9 | 41 | −42 |
| 1 | 216 | 213 | −24 | 80 | −23 | 9 | 181 | 191 | 2 | 146 | 146 | −19 | 62 | 57 | −11 | 132 | 128 |
| 3 | 245 | −239 | | | | 11 | 58 | −54 | 4 | 28 | 20 | −21 | 32 | −20 | −13 | 142 | 134 |
| 5 | 28 | 21 | | $k = 3,\ l = 5$ | | 13 | 107 | −95 | 6 | 67 | 65 | −23 | 83 | −89 | −15 | 66 | 56 |
| 7 | 41 | −48 | | | | | | | 10 | 93 | −100 | | | | −19 | 114 | 126 |
| 11 | 47 | −66 | 3 | 165 | −166 | | $k = 5,\ l = 5$ | | 12 | 55 | 62 | | $k = 2,\ l = 6$ | | −23 | 181 | −182 |
| 13 | 31 | −32 | 5 | 59 | 43 | | | | 14 | 163 | −161 | | | | | | |
| 15 | 56 | 52 | 7 | 52 | −44 | −1 | 124 | −120 | 16 | 127 | −133 | 0 | 142 | −130 | | $k = 4,\ l = 6$ | |
| | | | 9 | 164 | 170 | −3 | 124 | 121 | | | | 2 | 27 | −32 | | | |
| | $k = 1,\ l = 5$ | | 11 | 77 | 70 | −5 | 30 | −15 | | $k = 0,\ l = 6$ | | 6 | 387 | −399 | 0 | 205 | −190 |
| | | | 13 | 58 | 43 | −7 | 200 | −200 | | | | 8 | 54 | −56 | 2 | 50 | 46 |
| −1 | 294 | 269 | 17 | 70 | 75 | −11 | 72 | 74 | −2 | 69 | −23 | 10 | 85 | 88 | 4 | 48 | 46 |
| −3 | 356 | 346 | 19 | 90 | 102 | −13 | 130 | −138 | −4 | 334 | 317 | 14 | 105 | −99 | 6 | 107 | −113 |
| −5 | 133 | 135 | | | | −15 | 64 | −73 | −6 | 180 | −179 | 18 | 91 | 85 | 8 | 39 | 46 |
| −7 | 291 | −260 | | $k = 3,\ l = 5$ | | −21 | 78 | 80 | −8 | 56 | −61 | | | | 10 | 69 | 59 |
| −9 | 247 | 240 | | | | | | | −10 | 175 | 178 | | $k = 2,\ l = 6$ | | | | |
| −11 | 119 | 127 | −1 | 144 | −141 | | $k = 6,\ l = 5$ | | −12 | 131 | 142 | | | | | $k = 4,\ l = 6$ | |
| −13 | 187 | −169 | −3 | 223 | −223 | | | | −14 | 57 | 49 | −2 | 228 | 226 | | | |
| −15 | 104 | −106 | −5 | 278 | −286 | 2 | 111 | 118 | −16 | 130 | −122 | −4 | 226 | 226 | −4 | 86 | 93 |
| −17 | 137 | −132 | −7 | 326 | −313 | | | | −18 | 48 | −42 | −6 | 118 | 123 | −6 | 47 | 44 |
| −19 | 30 | −26 | −9 | 202 | −188 | | $k = 6,\ l = 5$ | | −20 | 49 | −40 | −8 | 186 | 195 | −8 | 47 | −39 |
| −21 | 166 | 162 | −11 | 152 | −150 | | | | −22 | 109 | 100 | −10 | 56 | −50 | −10 | 140 | 132 |
| | | | −13 | 43 | 49 | −6 | 81 | 75 | | | | −12 | 317 | −310 | −12 | 63 | −65 |
| | $k = 2,\ l = 5$ | | −15 | 48 | 53 | −8 | 94 | 98 | | $k = 1,\ l = 6$ | | −14 | 190 | −187 | −14 | 94 | −100 |
| | | | −17 | 50 | 41 | −14 | 123 | 117 | | | | −18 | 34 | 24 | | | |
| 0 | 79 | −79 | −21 | 208 | 212 | | | | 1 | 132 | 133 | −20 | 81 | −84 | | $k = 5,\ l = 6$ | |
| 2 | 621 | 619 | | | | | $k = 7,\ l = 5$ | | 3 | 134 | −132 | −22 | 164 | 171 | | | |
| 6 | 170 | −164 | | $k = 4,\ l = 5$ | | 1 | 142 | 132 | 7 | 94 | −106 | −24 | 115 | 125 | 1 | 84 | 76 |
| 8 | 92 | 93 | | | | 1 | 142 | 132 | 7 | 27 | 38 | | | | 5 | 65 | −66 |

$k = 5,\ l = 6$

| | | |
|---|---|---|
| 11 | 147 | −149 |

$k = 5,\ l = 6$

| | | |
|---|---|---|
| −1 | 172 | 168 |
| −3 | 227 | 225 |
| −15 | 125 | −137 |

$k = 6,\ l = 6$

| | | |
|---|---|---|
| 6 | 83 | −79 |
| 10 | 177 | 173 |

$k = 6,\ l = 6$

| | | |
|---|---|---|
| −2 | 98 | −102 |
| −4 | 175 | −176 |
| −6 | 37 | −43 |
| −8 | 85 | 87 |
| −10 | 110 | 107 |
| −12 | 65 | −52 |

$k = 7,\ l = 6$

| | | |
|---|---|---|
| −3 | 95 | 97 |
| −5 | 83 | −81 |
| −7 | 142 | −136 |
| −13 | 77 | 81 |

$k = 1,\ l = 7$

| | | |
|---|---|---|
| 1 | 50 | 38 |
| 3 | 250 | 258 |
| 5 | 29 | −31 |
| 9 | 112 | 109 |
| 11 | 86 | −82 |
| 13 | 121 | −121 |

$k = 1,\ l = 7$

| | | |
|---|---|---|
| −1 | 33 | 33 |
| −3 | 37 | −30 |
| −5 | 39 | −21 |
| −7 | 33 | −28 |
| −9 | 254 | 250 |
| −11 | 133 | 128 |
| −13 | 147 | −146 |
| −15 | 30 | 29 |
| −17 | 126 | 123 |
| −21 | 84 | −67 |

$k = 2,\ l = 7$

| | | |
|---|---|---|
| 0 | 30 | −39 |
| 2 | 26 | 46 |
| 6 | 44 | 27 |
| 8 | 133 | −126 |
| 10 | 82 | −86 |
| 12 | 30 | −33 |

$k = 2,\ l = 7$

| | | |
|---|---|---|
| −2 | 72 | 69 |
| −4 | 394 | 395 |

| | | |
|---|---|---|
| −6 | 281 | 273 |
| −8 | 238 | 245 |
| −12 | 54 | −46 |
| −18 | 126 | −117 |
| −72 | 81 | −79 |

$k = 3,\ l = 7$

| | | |
|---|---|---|
| 1 | 49 | −35 |
| 3 | 106 | 102 |
| 5 | 83 | −85 |
| 7 | 70 | 77 |
| 13 | 99 | 95 |

$k = 3,\ l = 7$

| | | |
|---|---|---|
| −1 | 123 | −125 |
| −3 | 224 | −229 |
| −5 | 146 | −139 |
| −7 | 180 | −199 |
| −9 | 108 | −96 |
| −11 | 117 | −117 |
| −13 | 79 | 83 |
| −19 | 155 | 157 |

$k = 4,\ l = 7$

| | | |
|---|---|---|
| 2 | 57 | −35 |
| 10 | 92 | 70 |
| 12 | 82 | −67 |

$k = 4,\ l = 7$

| | | |
|---|---|---|
| −6 | 76 | −62 |
| −8 | 101 | −96 |
| −14 | 95 | −94 |
| −16 | 98 | 96 |
| −18 | 94 | −100 |

$k = 5,\ l = 7$

| | | |
|---|---|---|
| 1 | 34 | 29 |
| 5 | 112 | −106 |

$k = 5,\ l = 7$

| | | |
|---|---|---|
| −1 | 29 | −37 |
| −3 | 132 | −137 |
| −5 | 117 | −123 |
| −9 | 56 | 64 |
| −13 | 117 | 110 |

$k = 6,\ l = 7$

| | | |
|---|---|---|
| 6 | 85 | −77 |
| 8 | 87 | −88 |

$k = 6,\ l = 7$

| | | |
|---|---|---|
| −2 | 128 | 142 |
| −4 | 229 | 230 |
| −6 | 90 | 95 |
| −12 | 99 | −97 |
| −14 | 81 | −76 |

$k = 7,\ l = 7$

| | | |
|---|---|---|
| 3 | 90 | 83 |

$k = 7,\ l = 7$

| | | |
|---|---|---|
| −7 | 67 | 56 |

$k = 0,\ l = 8$

| | | |
|---|---|---|
| 0 | 193 | 181 |
| 2 | 54 | 43 |
| 4 | 108 | 111 |
| 6 | 165 | 161 |
| 10 | 79 | −97 |
| 12 | 133 | −125 |

$k = 0,\ l = 8$

| | | |
|---|---|---|
| −2 | 25 | −5 |
| −4 | 46 | −45 |
| −6 | 99 | −87 |
| −8 | 183 | −204 |
| −10 | 377 | 360 |
| −12 | 172 | 170 |
| −14 | 53 | −44 |
| −16 | 133 | 139 |
| −18 | 65 | 74 |
| −22 | 67 | −68 |

$k = 1,\ l = 8$

| | | |
|---|---|---|
| 1 | 69 | −60 |
| 3 | 32 | 22 |

| | | |
|---|---|---|
| 5 | 116 | −130 |
| 7 | 105 | −97 |
| 9 | 36 | −28 |
| 11 | 92 | 102 |
| 13 | 134 | 125 |

$k = 1,\ l = 8$

| | | |
|---|---|---|
| −1 | 210 | −213 |
| −7 | 162 | 156 |
| −9 | 97 | 89 |
| −11 | 281 | −267 |
| −13 | 254 | −258 |
| −15 | 69 | −65 |
| −17 | 100 | 94 |
| −23 | 107 | 118 |

$k = 2,\ l = 8$

| | | |
|---|---|---|
| 0 | 60 | 58 |
| 2 | 47 | 39 |
| 6 | 86 | 82 |
| 8 | 47 | −45 |

$k = 2,\ l = 8$

| | | |
|---|---|---|
| −4 | 27 | −2 |
| −6 | 52 | 48 |
| −8 | 29 | 13 |
| −12 | 95 | 100 |
| −14 | 131 | 126 |

$k = 3,\ l = 8$

Table 4–*continued*

| $h$ | $10\|F_o\|$ | $10F_c$ | $h$ | $10\|F_o\|$ | $10F_c$ | $h$ | $10\|F_o\|$ | $10F_c$ | $h$ | $10\|F_o\|$ | $10F_c$ | $h$ | $10\|F_o\|$ | $10F_c$ | $h$ | $10\|F_o\|$ | $10F_c$ |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | $k = 5,\ l = 8$ | | −5 | 53 | 29 | −7 | 118 | 120 | 4 | 135 | −144 | −8 | 101 | 114 |
| 1 | 114 | −111 | | | | −7 | 129 | 116 | −9 | 65 | 61 | 6 | 108 | −104 | −18 | 89 | −67 |
| 3 | 36 | 46 | 5 | 97 | −94 | −9 | 52 | 54 | −11 | 106 | 110 | 8 | 122 | 137 | | | |
| 7 | 110 | −111 | 7 | 72 | −67 | −11 | 43 | 48 | −13 | 79 | 76 | | | | | $k = 3,\ l = 10$ | |
| 11 | 93 | 89 | | | | −13 | 80 | 75 | −17 | 79 | −87 | | $k = 0,\ l = 10$ | | | | |
| | | | | $k = 5,\ l = 8$ | | −15 | 144 | 140 | | | | | | | −11 | 94 | 100 |
| | $k = 3,\ l = 8$ | | | | | −21 | 91 | −111 | | $k = 4,\ l = 9$ | | −2 | 65 | −73 | −13 | 95 | 86 |
| | | | −5 | 115 | 117 | | | | | | | −4 | 66 | −61 | | | |
| −1 | 51 | 47 | −7 | 140 | 141 | | $k = 2,\ l = 9$ | | 2 | 98 | 96 | −6 | 27 | 27 | | $k = 4,\ l = 10$ | |
| −3 | 54 | 36 | −9 | 181 | 191 | | | | 4 | 80 | 79 | −8 | 99 | 86 | | | |
| −9 | 132 | 135 | | | | 0 | 57 | −52 | 8 | 144 | 139 | −12 | 134 | −134 | −8 | 97 | 109 |
| −11 | 58 | −54 | | $k = 6,\ l = 8$ | | 2 | 116 | −111 | | | | −14 | 130 | −150 | −12 | 124 | −124 |
| −13 | 119 | −129 | | | | 8 | 81 | 76 | | $k = 4,\ l = 9$ | | −20 | 77 | −100 | | | |
| −15 | 61 | 56 | 0 | 139 | −136 | | | | | | | | | | | $k = 5,\ l = 10$ | |
| −17 | 92 | 77 | 4 | 118 | 107 | | $k = 2,\ l = 9$ | | −2 | 148 | −145 | | $k = 1,\ l = 10$ | | | | |
| −19 | 104 | −104 | | | | | | | −4 | 230 | −230 | | | | −5 | 84 | −76 |
| | | | | $k = 6,\ l = 8$ | | −2 | 40 | 22 | −6 | 269 | −277 | 5 | 133 | 124 | −7 | 105 | −117 |
| | $k = 4,\ l = 8$ | | | | | −4 | 88 | 107 | −8 | 202 | −204 | 9 | 107 | −104 | | | |
| | | | −2 | 111 | −112 | −6 | 113 | −103 | −10 | 210 | −220 | | | | | $k = 1,\ l = 11$ | |
| 0 | 52 | 52 | −4 | 127 | −125 | −8 | 66 | −61 | −18 | 144 | 142 | | $k = 1,\ l = 10$ | | | | |
| 2 | 51 | 55 | −10 | 70 | −66 | −12 | 51 | −58 | | | | | | | −3 | 77 | 66 |
| 6 | 114 | 109 | | | | −14 | 45 | 58 | | $k = 5,\ l = 9$ | | −3 | 66 | 60 | −13 | 70 | 75 |
| | | | | $k = 1,\ l = 5$ | | | | | | | | −5 | 85 | 69 | −15 | 91 | 104 |
| | $k = 4,\ l = 8$ | | | | | | $k = 3,\ l = 9$ | | −3 | 98 | 86 | −7 | 107 | −114 | | | |
| | | | 1 | 118 | 109 | | | | −5 | 86 | 86 | −9 | 144 | −144 | | $k = 2,\ l = 11$ | |
| −2 | 28 | 19 | 3 | 119 | 130 | 1 | 53 | −53 | −7 | 120 | 116 | −13 | 96 | 101 | | | |
| −4 | 77 | −80 | 5 | 66 | −52 | 3 | 86 | −85 | −9 | 153 | 159 | −15 | 120 | 125 | −10 | 113 | 114 |
| −6 | 119 | −124 | | | | | | | −13 | 104 | −116 | | | | | | |
| −8 | 141 | −153 | | $k = 1,\ l = 9$ | | | $k = 3,\ l = 9$ | | | | | | $k = 2,\ l = 10$ | | | $k = 0,\ l = 12$ | |
| −10 | 75 | −88 | | | | | | | | $k = 0,\ l = 10$ | | | | | | | |
| −12 | 66 | 69 | −1 | 52 | 35 | −3 | 112 | 117 | | | | −2 | 33 | 18 | 4 | 73 | −46 |
| −18 | 89 | 80 | −3 | 103 | 104 | −5 | 139 | 138 | 0 | 103 | −96 | −6 | 40 | −13 | −10 | 82 | −100 |

between the benzo-groups is 61.5°. 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 ±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°. 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).

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