## 387 *Topochemistry. Part V.* The Crystal Structure of 2,5-Dimethyl-1,4-benzoquinone.*

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The crystal structure of 2,5-dimethyl-1,4-benzoquinone has been solved from full three-dimensional counter data by means of the Fourier-transform technique. The structure consists of two sets of symmetry-unrelated molecules, centred at (000) and ($\frac{1}{2}\frac{1}{2}\frac{1}{2}$), whose bond lengths, after correction for thermal vibrations, agree to within their e.s.d.: C=C, 1·344 Å; C–CMe, 1·500 Å; C–CH, 1·474 Å; C–Me, 1·507 Å; C=O, 1·228 Å.

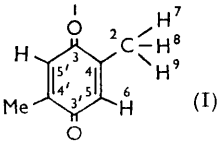
Both sets of molecules are stacked along [100]; within these stacks close parallel contacts (4·01 Å) between >C=C< groups are provided by the *a*-axis translation. The inclination of the molecular planes gives rise to a second antiparallel contact between the >C=C< bonds of stack-related molecules, across inversion centres midway between their molecular centres; these contact distances are 4·03 and 4·64 Å for the two stacks. The 40% yield in the photochemical formation of a dimer of symmetry $\bar{1}$ suggests that the short centrosymmetric contact in the first stack leads to dimerisation, and that the equivalent contact of 4·64 Å in the second stack is too long for dimerisation to occur. The formation of the cage dimer of symmetry 2/*m* in 2% yield indicates that double dimerisation along the translation contact of 4·01 Å is a rare event.

Continuing our investigation into the geometry of solid-state dimerisation we have undertaken the crystal-structure analyses of some of the *p*-quinones whose photochemistry

* Part IV, preceding paper.

has recently been described by Cookson and his co-workers.[1a,b] The present Paper presents the full three-dimensional analysis of the structure of 2,5-dimethylbenzoquinone

2,5-Dimethyl-1,4-benzoquinone. The numbering of atoms used in the crystallographic analysis.



(I)

(I); it also suggests an interpretation of the observation that this compound gives, in the solid state, on short irradiation by sunlight or ultraviolet light, two dimers formulated by Cookson as (II) and (III) (a or b) whose "optimum" yields are reported as 40 and 2%, respectively.

## Experimental

On slow evaporation from solutions in ethyl acetate 2,5-dimethylbenzoquinone crystallises in yellow, approximately equidimensional, triclinic rhombs; laths elongated along [100] can be grown by sublimation at 30°/25 mm. Crystals produced by either method are frequently twinned across (010). Cell constants were determined from G.E. diffractometer measurements of the Bragg angles of 18 high-order reflections ($0{\cdot}90 < \sin\theta < 0{\cdot}99$) and analysed by a least-squares procedure. Since these measurements were markedly temperature-dependent the ambient temperature was kept at 25 ± 1° throughout cell-dimension and intensity measurements. Crystals were sealed in Lindemann-glass capillaries to prevent sublimation. Table 1 summarises the relevant constants.

Table 1.

2,5-Dimethyl-1,4-benzoquinone, $C_8H_8O_2$, $M$ 136·14; m. p. 125°.
$a = 4{\cdot}013_2$, $\sigma(a) = 0{\cdot}0002$; $b = 9{\cdot}366_3$, $\sigma(b) = 0{\cdot}0002$; $c = 9{\cdot}738_3$, $\sigma(c) = 0{\cdot}0004$ Å.
$\alpha = 93{\cdot}50_1°$, $\sigma(\alpha) = 0{\cdot}002°$; $\beta = 101{\cdot}35_6°$, $\sigma(\beta) = 0{\cdot}005°$; $\gamma = 98{\cdot}56_8°$, $\sigma(\gamma) = 0{\cdot}004°$, at 25 ± 1°.
Axial ratio [2] $a : b : c = 0{\cdot}8626 : 1 : 0{\cdot}5382$; $\alpha = 71°\,41'$, $\beta = 93°\,22'$, $\gamma = 89°\,24'$. Our ratio (after transformation of axes) $a : b : c = 0{\cdot}8671 : 1 : 0{\cdot}5486$; $\alpha = 71°\,56'$, $\beta = 93°\,10'$, $\gamma = 89°\,14'$.
Transformation matrix: $\begin{pmatrix} \bar{1}\bar{1}\bar{1} \\ 11\bar{1} \\ 200 \end{pmatrix}$. *Systematic absences*: none; *space group* $P1$ or $P\bar{1}$; $V = 353{\cdot}3$ Å$^3$; $d$ (calc.) ($n = 2$) : 1·280 g./cm.$^3$. $F(000) = 144$.

The ($0kl$) intensities were collected by multiple-film Weissenberg photographs from an untwinned crystal of approximate dimensions 2·0 × 0·8 × 0·5 mm., mounted about [100]. Of the 224 reflections in the Cu-Kα range 182 could be measured by the usual visual technique; their intensities were placed on an approximately absolute scale by Wilson's method,[3] and corrected for Lorentz-polarisation.

The distribution of the ($0kl$) intensities indicates pseudo-orthogonal symmetry about the $b^*$ and $c^*$ axes; in addition, pseudo-absences of ($0k0$) for $k$ odd, and ($h0l$) for $h + l$ odd, suggest that the molecular arrangement approximates to space group $P2_1/n$.

On the assumption of $P\bar{1}$ as the correct space group, confirmed by the eventual refinement of the structure, two possible molecular arrangements were envisaged: the two molecules could either lie in equivalent general positions, or they could each occupy a centre of symmetry and be symmetry-unrelated to one another. The pseudo-symmetry relations noted above pointed to the second alternative, with the two molecules at the centres of symmetry (000) and $(\frac{1}{2}\frac{1}{2}\frac{1}{2})$. The Patterson projection P($yz$) confirmed this arrangement, but was insufficiently resolved to enable us to orient the two independent molecules.

The structure was solved by means of the Fourier-transform technique. On the assumption of a planar and centrosymmetric molecule the unit-cell transform $\tau$ is given by $\tau = T_1 + T_2 \cos 2\pi\left(\frac{h}{2} + \frac{k}{2} + \frac{1}{2}\right)$ where $T$ is the molecular transform and the subscripts 1 and 2 refer to

[1] (*a*) Cookson and Hudec, *Proc. Chem. Soc.*, 1959, 11; (*b*) Cookson, Cox, and Hudec, *J.*, 1961, 4499.
[2] Muthmann, *Z. Kryst.*, 1889, **15**, 394; *see* Groth, "Chemische Krystallographie," Vol. IV, p. 670, W. Engelmann, Leipzig, 1917.
[3] Wilson, *Nature*, 1942, **180**, 152.

the molecules at (000) and ($\frac{1}{2}\frac{1}{2}\frac{1}{2}$), respectively. $T$ was computed on the basis of the known dimensions of *p*-benzoquinone [4] and a C–Me bond of length 1·50 Å, bisecting the $C_1$–$C_2$–$C_3$ angle. The reciprocal ($0kl$) plane was successfully fitted to $\tau$ through the observation that $F(101)$ is outstandingly strong, and hence the molecular planes must lie nearly parallel to the (101) plane and therefore to [010]; an electron-density projection based on 13 low-order ($0kl$) reflections whose signs could be derived by means of the Fourier transform, clearly established the molecular orientations.

We checked this trial structure using Woolfson's permutation technique.[5] A set of seven strong low-order $F(0kl)$ and their mates $F(0k\bar{l})$ were chosen; the signs of the former were taken from Woolfson's table of sign permutations while the latter were given corresponding signs according to the pseudo-symmetry (*pgg*) of the projection; by assigning arbitrary signs to two further strong reflections the origin of the projection was fixed. From the resulting 16 permutation Fourier-projections we obtained one electron-density map which was clearly superior to all others as regards molecular shape and clarity of background. The trial structure based on this map agreed satisfactorily with the model derived from the Fourier transform.

Refinement was accomplished by means of a least-squares WEIZAC programme written by Dr. F. L. Hirshfeld of this Department; this programme, a modification of the procedure described by Rossman *et al.*,[6] minimises the function

$$r = \sum w(k^2F_o^2 - |F_c|^2)^2 / \sum wk^4F_o^4 \qquad (1)$$

with respect to the scale factor $k^2$ and the positional and thermal parameters of the atoms in the asymmetric unit. It uses the linear diagonal approximation for the computation of the positional-parameter shifts; the shifts in $k^2$ and the isotropic correction of the thermal parameter shifts are computed by a 2 × 2 matrix in order to allow for the interaction of these parameters.[7] Each atom in the asymmetric unit may be treated isotropically with a temperature factor $\exp(-B \sin^2\theta/\lambda^2)$ or anisotropically with a temperature factor $\exp[-(h^2\beta_{11} + k^2\beta_{22} + l^2\beta_{33} + hk\beta_{12} + kl\beta_{23} + hl\beta_{13})]$.[8] The weighting factors $w(hkl)$ were the sums of the weights assigned to each film reading of the spot $hkl$. These separate weights were a function of the visual estimates and of the factors used for translation of each such reading into $F_o^2$ (film-to-film ratio, Lorentz-polarisation). This procedure gave weights that varied roughly as $F_o^{-4}$ for most reflections, with decreased weights for the weak and the very strong reflections. The summations in eqn. (1) comprise all but those " unobserved " reflections for which $kF_t > |F_c|$ where $F_t$ is a threshold value calculated for each such reflection from $I_t$ equal to the weakest observed intensity.

The two-dimensional data were refined at first with isotropic temperature factors; after a few cycles a difference map $\delta(yz)$ was computed to locate the ring and methyl-group hydrogens which were then inserted into the structure-factor calculations. The least-squares procedure continued for a few further cycles with isotropic temperature factors of the heavy atoms until $r$ had dropped to 0·081.

At this stage the three-dimensional data were collected on a G.E. goniostat with Cu-K$\alpha$ radiation and an argon-filled counter. A crystal of suitable mosaicity was selected by a check of the diffraction profiles and widths of a few reflections recorded along several directions; an untwinned crystal of dimensions 0·25 × 0·25 × 0·20 mm. was found suitable. For $\theta < 65°$ the $\alpha_1$ and $\alpha_2$ components were recorded together, while above this range up to the instrument-set maximum of $\theta = 82·5°$ only the $\alpha_1$ component was measured. The intensities covered a range of over 200,000 to 1. As a check on the accuracy of measurement both $I(0kl)$ and $I(0k\bar{l})$ were counted: the average $\Delta F_o$ was estimated as 1%. Because of heavy iron contamination of the tube (after less than 1000 hours' use) reflections for which $h$, $k$, and $l$ are divisible by five, were eliminated from the refinement. Information on the total number of reflections is given in Table 2. Weighting factors were computed by addition of an error term proportional to the square of the measured intensity to the statistical counting error (Coppens).[9] Reflections

[4] Trotter, *Acta Cryst.*, 1960, **13**, 86.
[5] Woolfson, *Acta Cryst.*, 1954, **7**, 65.
[6] Rossman, Jacobson, Hirshfeld, and Lipscomb, *Acta Cryst.*, 1959, **12**, 530.
[7] Cruickshank, " Computing Methods and the Phase Problem," ed. Pepinsky, Robertson, and Speakman, Pergamon Press, 1961.
[8] Cruickshank, *Acta Cryst.*, 1956, **9**, 747.
[9] Coppens, Ph.D. Thesis, Amsterdam, 1960.

TABLE 2.

Structure factors.

Total number of reflections counted, 1553; number of unobserved reflections, 194; unobserved reflections included in refinement (*i.e.*, $kF_t<|F_c|$), 49; secondary extinction, 1; reflections affected by white radiation, 4; total number of reflections included in refinement, 1403.

| *h* | *k* | *l* | 100$F_o$ | 100$F_c$ | *h* | *k* | *l* | 100$F_o$ | 100$F_c$ | *h* | *k* | *l* | 100$F_o$ | 100$F_c$ | *h* | *k* | *l* | 100$F_o$ | 100$F_c$ | *h* | *k* | *l* | 100$F_o$ | 100$F_c$ |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 0 | −11 | 1 | 136 | −134 | 0 | −2 | 6 | 887 | −826 | 0 | 6 | 0 | 162 | −172 | 1 | −8 | −4 | 107 | −105 | 1 | −4 | −1 | 117 | 153 |
| | | 2 | 123 | 91 | | | 7 | 117 | 125 | | | 1 | 267 | −282 | | | −3 | 332 | 303 | | | 0 | 1116 | 1061 |
| | | 3 | 215 | −199 | | | 8 | 199 | 195 | | | 2 | 1106 | 1122 | | | −2 | 630 | 651 | | | 1 | 2795 | −2912 |
| | | 4 | 217 | 202 | | | 9 | 144 | −145 | | | 3 | 644 | 646 | | | −1 | 583 | −607 | | | 2 | 749 | −726 |
| | | 5 | 233 | −213 | | | 10 | 53 | 56 | | | 4 | 913 | 961 | | | 0 | 40 | 19 | | | 3 | 174 | 187 |
| 0 | −10 | 1 | 99 | −104 | | | 11 | 103 | −111 | | | 5 | 670 | −664 | | | 1 | 314 | −363 | | | 4 | <26 | 14 |
| | | 2 | 144 | −140 | | | 12 | 140 | −135 | | | 6 | 705 | 726 | | | 2 | 48 | 30 | | | 5 | 518 | −543 |
| | | 3 | 63 | 47 | 0 | −1 | 1 | 2831 | 3294 | | | 7 | 308 | −314 | | | 3 | 532 | −546 | | | 6 | 45 | −46 |
| | | 4 | 38 | −27 | | | 2 | 1095 | −1090 | | | 8 | 85 | −85 | | | 4 | 460 | −476 | | | 7 | 93 | 71 |
| | | 6 | 265 | −236 | | | 3 | 2139 | −2165 | | | 9 | 59 | −56 | | | 5 | 180 | 180 | | | 8 | <28 | 23 |
| | | 7 | 142 | −130 | | | 4 | 239 | 236 | 0 | 7 | 0 | 75 | −67 | | | 6 | 51 | 60 | | | 9 | 217 | −201 |
| 0 | −9 | 1 | 41 | −50 | | | 5 | 346 | −337 | | | 1 | 127 | 136 | | | 7 | 103 | −88 | | | 10 | 154 | −155 |
| | | 2 | 44 | −56 | | | 6 | 1134 | −1124 | | | 2 | 571 | 583 | | | 8 | 97 | 99 | | | 11 | 188 | −194 |
| | | 3 | 190 | −194 | | | 7 | 202 | −200 | | | 3 | 672 | 668 | | | 9 | 176 | −183 | 1 | −3 | −11 | 255 | −233 |
| | | 4 | 57 | 37 | | | 8 | 160 | −147 | | | 4 | 433 | 448 | 1 | −7 | −9 | 67 | 42 | | | −10 | 176 | −186 |
| | | 5 | 134 | 142 | | | 9 | 899 | 889 | | | 5 | 723 | 730 | | | −8 | 109 | −108 | | | −9 | 478 | 491 |
| | | 6 | 142 | 140 | | | 10 | 462 | −460 | | | 6 | 243 | −256 | | | −7 | 235 | 231 | | | −8 | 347 | −373 |
| | | 7 | 415 | −392 | | | 11 | <24 | 16 | | | 7 | 55 | 46 | | | −6 | 105 | 102 | | | −7 | 464 | −444 |
| | | 8 | 176 | 169 | | | 12 | <22 | 21 | | | 8 | 120 | 114 | | | −5 | 389 | −372 | | | −6 | 172 | 156 |
| 0 | −8 | 1 | 91 | 61 | 0 | 0 | 1 | 188 * | −22 | | | 9 | 61 | −63 | | | −4 | 996 | 986 | | | −5 | 429 | 454 |
| | | 2 | 612 | −648 | | | 2 | 2153 | 2237 | 0 | 8 | 0 | 352 | −362 | | | −3 | 808 | 798 | | | −4 | 1322 | −1179 |
| | | 3 | 528 | −552 | | | 3 | 117 * | 2 | | | 1 | 44 | 53 | | | −2 | 707 | 717 | | | −3 | 1659 | −1629 |
| | | 4 | 249 | 205 | | | 4 | 1124 | 1103 | | | 2 | 490 | −504 | | | −1 | 620 | 617 | | | −2 | 344 | 334 |
| | | 5 | 48 | 37 | | | 6 | 178 | −149 | | | 3 | 504 | 512 | | | 0 | 109 | 125 | | | −1 | 365 | 367 |
| | | 6 | 81 | −72 | | | 7 | 87 | −72 | | | 4 | 249 | 250 | | | 1 | <30 | 8 | | | 0 | 40 | 36 |
| | | 7 | 142 | 140 | | | 8 | 1002 | 992 | | | 5 | 101 | −102 | | | 2 | 99 | 95 | | | 1 | 83 | −81 |
| | | 8 | 302 | −285 | | | 9 | 130 | −63 | | | 6 | 110 | −102 | | | 3 | 427 | −446 | | | 2 | 49 | 40 |
| | | 9 | 38 | 38 | | | 11 | 44 | −32 | | | 7 | <26 | −26 | | | 4 | 612 | 602 | | | 3 | 150 | −148 |
| 0 | −7 | 1 | 40 | 49 | | | 12 | 105 | −101 | | | 8 | 132 | −137 | | | 5 | 581 | −589 | | | 4 | <26 | 44 |
| | | 2 | 423 | −451 | 0 | 1 | 0 | 48 | 50 | 0 | 9 | 0 | 38 | 20 | | | 6 | 717 | 701 | | | 5 | 640 | 634 |
| | | 3 | 598 | 620 | | | 1 | 2892 | 3151 | | | 1 | 158 | −175 | | | 7 | 221 | 225 | | | 6 | 518 | −468 |
| | | 4 | 980 | −995 | | | 2 | 976 | 962 | | | 2 | 75 | 89 | | | 8 | 48 | 50 | | | 7 | 113 | −123 |
| | | 5 | 964 | 975 | | | 3 | 1875 | −1915 | | | 3 | 67 | −56 | | | 9 | 83 | −84 | | | 8 | 57 | 66 |
| | | 6 | 320 | 315 | | | 4 | 119 | −101 | | | 4 | 73 | −59 | | | 10 | 48 | −57 | | | 9 | 61 | 49 |
| | | 7 | 110 | 99 | | | 5 | 245 | −230 | | | 5 | <22 | 7 | 1 | −6 | −10 | 34 | −52 | | | 10 | 178 | −171 |
| | | 8 | 213 | −206 | | | 6 | 990 | 964 | | | 7 | 202 | −222 | | | −9 | 77 | −95 | | | 11 | 152 | −156 |
| | | 9 | 89 | −98 | | | 7 | 117 | −107 | 0 | 10 | 1 | 85 | 90 | | | −8 | 91 | −102 | 1 | −2 | −11 | 140 | −139 |
| | | 10 | 89 | −73 | | | 8 | 166 | 147 | | | 2 | 148 | −143 | | | −7 | 107 | −112 | | | −10 | 247 | 255 |
| 0 | −6 | 1 | 367 | 376 | | | 9 | 879 | 890 | | | 3 | 63 | −58 | | | −6 | 533 | −533 | | | −9 | 206 | 208 |
| | | 2 | 1146 | 1127 | | | 10 | 385 | 385 | | | 4 | 34 | −26 | | | −5 | 982 | 1000 | | | −8 | 207 | 210 |
| | | 3 | 841 | −858 | | | 11 | <24 | 19 | 0 | 11 | 0 | <26 | 23 | | | −4 | 851 | −846 | | | −7 | 338 | 337 |
| | | 4 | 1193 | 1234 | | | 12 | <18 | 9 | | | 1 | 166 | −147 | | | −3 | 1085 | 1095 | | | −6 | 48 | −54 |
| | | 5 | 567 | 565 | 0 | 2 | 0 | 231 * | 107 | | | 2 | 140 | −115 | | | −2 | 749 | 760 | | | −5 | 1096 | −983 |
| | | 6 | 968 | 981 | | | 1 | 1794 | 1845 | | | 3 | 152 | −147 | | | −1 | 1165 | 1171 | | | −4 | 356 | 334 |
| | | 7 | 528 | 526 | | | 2 | 1806 | −1809 | 1 | −11 | −3 | 67 | −76 | | | 0 | 379 | −375 | | | −3 | 1881 | −1886 |
| | | 8 | <28 | −30 | | | 3 | 2161 | −2151 | | | −2 | 184 | −174 | | | 1 | 176 | −192 | | | −2 | 2351 | −2399 |
| | | 9 | <30 | 54 | | | 4 | 1620 | −1568 | | | −1 | 158 | −115 | | | 2 | 209 | 201 | | | −1 | 1657 | −1656 |
| | | 10 | 77 | −95 | | | 5 | 484 | 465 | | | 0 | 150 | −162 | | | 3 | 952 | 968 | | | 0 | 1559 | 1608 |
| | | 11 | <18 | −0 | | | 6 | 774 | −718 | | | 1 | 35 | 20 | | | 4 | 656 | −662 | | | 1 | 245 | 235 |
| 0 | −5 | 1 | 1936 | −1928 | | | 7 | 95 | −99 | | | 2 | 158 | −148 | | | 5 | 952 | 950 | | | 2 | 1278 | −1301 |
| | | 2 | 170 | −162 | | | 8 | 241 | 227 | | | 3 | 124 | 107 | | | 6 | 444 | 431 | | | 3 | 1124 | −1126 |
| | | 3 | 523 | 523 | | | 9 | 217 | 196 | | | 4 | 190 | −190 | | | 7 | 662 | 656 | | | 4 | 1146 | 1143 |
| | | 4 | 30 | −48 | | | 10 | 91 | 95 | | | 5 | 144 | 142 | | | 8 | 314 | 310 | | | 5 | 1102 | −1055 |
| | | 6 | 806 | 853 | | | 11 | 91 | 99 | 1 | −10 | −5 | 328 | −297 | | | 9 | 31 | −36 | | | 6 | 233 | −229 |
| | | 7 | 488 | 471 | 0 | 3 | 0 | 318 * | 193 | | | −4 | 184 | 153 | | | 10 | <24 | 36 | | | 7 | 456 | −450 |
| | | 8 | 132 | 154 | | | 1 | 328 | 307 | | | −3 | <24 | 7 | 1 | −5 | −10 | 97 | 62 | | | 8 | 49 | 50 |
| | | 9 | 484 | −506 | | | 2 | 113 | 124 | | | −2 | 75 | −55 | | | −9 | <24 | −15 | | | 9 | 132 | 120 |
| | | 11 | 41 | 41 | | | 3 | 142 | 149 | | | −1 | 182 | −162 | | | −8 | 585 | −624 | | | 10 | 115 | −124 |
| 0 | −4 | 1 | 1086 | −1050 | | | 4 | 1065 | −1063 | | | 0 | 123 | 119 | | | −7 | <30 | −54 | | | 11 | <24 | 12 |
| | | 2 | 65 | 70 | | | 5 | 857 | −744 | | | 1 | 99 | 93 | | | −6 | 528 | 500 | 1 | −1 | −12 | 69 | −57 |
| | | 3 | 223 | −216 | | | 6 | 235 | 245 | | | 2 | 83 | −93 | | | −5 | 784 | −808 | | | −11 | <24 | 18 |
| | | 4 | 879 | −867 | | | 7 | 109 | 119 | | | 3 | 132 | −143 | | | −4 | 115 | −108 | | | −10 | 83 | 25 |
| | | 5 | 178 | −191 | | | 8 | 188 | −180 | | | 4 | 81 | 60 | | | −3 | <28 | −39 | | | −9 | 670 | 654 |
| | | 6 | 154 | 150 | | | 9 | 237 | −232 | | | 5 | 55 | −28 | | | −2 | 391 | 378 | | | −8 | 1369 | 1386 |
| | | 7 | <30 | 3 | | | 10 | 261 | 269 | | | 6 | 101 | −97 | | | −1 | 225 | 202 | | | −7 | 164 | 164 |
| | | 8 | 223 | −235 | | | 11 | 134 | −130 | | | 7 | 180 | −195 | | | 0 | 1679 | −1648 | | | −6 | 450 | −403 |
| | | 9 | 288 | −287 | 0 | 4 | 0 | 3471 | −3571 | 1 | −9 | −7 | 166 | −150 | | | 1 | 41 | −39 | | | −5 | 1582 | 1576 |
| | | 10 | 389 | −388 | | | 1 | 1106 | 1054 | | | −6 | 379 | −358 | | | 2 | 1381 | −1354 | | | −4 | 79 | −91 |
| | | 11 | 97 | −104 | | | 2 | 294 | 305 | | | −5 | 148 | −149 | | | 3 | 324 | −312 | | | −3 | <41 | −10 |
| 0 | −3 | 1 | 247 | 217 | | | 3 | 41 | −29 | | | −4 | 44 | 71 | | | 4 | 314 | 301 | | | −2 | 1873 | −2002 |
| | | 2 | 514 | −533 | | | 4 | 798 | −816 | | | −3 | 83 | −76 | | | 5 | 79 | −54 | | | −1 | 1236 | 1245 |
| | | 3 | 484 | 466 | | | 5 | 152 | 163 | | | −2 | 115 | −107 | | | 6 | 87 | 51 | | | 0 | 2847 | 3262 |
| | | 4 | 1369 | 1348 | | | 6 | 170 | 147 | | | −1 | 55 | 86 | | | 7 | 488 | 485 | | | 1 | 93 | 88 |
| | | 5 | 1073 | −977 | | | 7 | <30 | 13 | | | 0 | 107 | −139 | | | 8 | 170 | 169 | | | 2 | 1881 | 1834 |
| | | 6 | 364 | −370 | | | 8 | 285 | −297 | | | 1 | <28 | −13 | | | 9 | 93 | 97 | | | 3 | 130 | −84 |
| | | 7 | 190 | 198 | | | 9 | 269 | 266 | | | 2 | 55 | −92 | | | 10 | 249 | −272 | | | 4 | 962 | −1004 |
| | | 8 | 249 | 248 | | | 10 | 241 | −272 | | | 3 | 48 | −75 | 1 | −4 | −11 | 49 | −49 | | | 5 | 115 | 190 |
| | | 9 | 344 | −347 | | | 11 | 53 | 65 | | | 4 | 138 | −134 | | | −10 | 136 | 144 | | | 6 | 302 | −301 |
| | | 10 | 350 | −359 | 0 | 5 | 1 | 1420 | −1380 | | | 5 | 49 | 31 | | | −9 | 456 | −471 | | | 7 | 482 | −453 |
| | | 11 | 168 | −169 | | | 2 | 296 | 300 | | | 6 | 45 | 63 | | | −8 | 411 | 399 | | | 8 | 65 | 53 |
| | | 12 | 150 | 147 | | | 3 | 300 | 290 | | | 7 | 87 | 108 | | | −7 | 253 | −291 | | | 9 | 156 | −141 |
| 0 | −2 | 1 | 1798 | −1862 | | | 4 | 35 | −30 | | | 8 | 247 | −263 | | | −6 | <30 | 35 | | | 10 | 458 | 443 |
| | | 2 | 1748 | −1781 | | | 6 | 532 | −539 | 1 | −8 | −8 | <24 | 4 | | | −5 | 198 | 213 | | | 11 | 188 | −192 |
| | | 3 | 2404 | 2442 | | | 7 | 215 | 202 | | | −7 | 215 | −213 | | | −4 | 318 | 344 | 1 | 0 | −12 | <22 | 9 |
| | | 4 | 1778 | −1778 | | | 8 | 48 | −34 | | | −6 | <26 | −24 | | | −3 | 996 | −954 | | | −11 | 219 | −212 |
| | | 5 | 302 | −272 | | | 9 | 340 | −380 | | | −5 | 110 | −100 | | | −2 | 170 | 154 | | | 10 | 79 | 70 |

TABLE 2. (*Continued.*)

| h | k | l | 100$F_o$ | 100$F_c$ |
|---|---|---|---|---|
| 1 | 0 | −9 | 986 | 967 |
| | | −8 | 105 | −90 |
| | | −7 | 1033 | 1056 |
| | | −6 | 107 | −105 |
| | | −5 | 302 | −329 |
| | | −4 | 192 | −184 |
| | | −3 | 1096 | 1076 |
| | | −2 | 63 | −64 |
| | | −1 | 2283 | 2475 |
| | | 0 | 124 | −100 |
| | | 1 | 6833 * | 8571 |
| | | 2 | <49 | 2 |
| | | 3 | 644 | 652 |
| | | 4 | <28 | 10 |
| | | 5 | 468 | 519 |
| | | 6 | 44 | −25 |
| | | 7 | <28 | 40 |
| | | 8 | <30 | −49 |
| | | 9 | 644 | 621 |
| | | 10 | <26 | −33 |
| | | 11 | 282 | 282 |
| 1 | 1 | −12 | 75 | −65 |
| | | −11 | <24 | 21 |
| | | −10 | 71 | 44 |
| | | −9 | 755 | −753 |
| | | −8 | 1292 | 1307 |
| | | −7 | 99 | −100 |
| | | −6 | 525 | −460 |
| | | −5 | 1579 | −1639 |
| | | −4 | 314 | −305 |
| | | −3 | 265 | 255 |
| | | −2 | 1975 | −2125 |
| | | −1 | 1209 | −1237 |
| | | 0 | 2681 | 2927 |
| | | 1 | <22 | −7 |
| | | 2 | 1562 | 1532 |
| | | 3 | 125 | 76 |
| | | 4 | 838 | −877 |
| | | 5 | 48 | −81 |
| | | 6 | 282 | −260 |
| | | 7 | 383 | 364 |
| | | 8 | 99 | 91 |
| | | 9 | 120 | 107 |
| | | 10 | 419 | 417 |
| | | 11 | 160 | 153 |
| 1 | 2 | −12 | 310 | 316 |
| | | −11 | 188 | −171 |
| | | −10 | 273 | −280 |
| | | −9 | 128 | 123 |
| | | −8 | 61 | −84 |
| | | −7 | 207 | 219 |
| | | −6 | 85 | 100 |
| | | −5 | 1085 | −1017 |
| | | −4 | 38 | 29 |
| | | −3 | 1730 | −1802 |
| | | −2 | 2035 | 2066 |
| | | −1 | 1130 | −1150 |
| | | 0 | 1339 | −1373 |
| | | 1 | 219 | 229 |
| | | 2 | 1098 | 1113 |
| | | 3 | 936 | −896 |
| | | 4 | 857 | −834 |
| | | 5 | 901 | −856 |
| | | 6 | 235 | 249 |
| | | 7 | 350 | −339 |
| | | 8 | 40 | −33 |
| | | 9 | 127 | 122 |
| | | 10 | 127 | 123 |
| | | 11 | 20 | 32 |
| 1 | 3 | −12 | 249 | −239 |
| | | −11 | 217 | 209 |
| | | −10 | 219 | −223 |
| | | −9 | 539 | −556 |
| | | −8 | 468 | −511 |
| | | −7 | 436 | 419 |
| | | −6 | 211 | 220 |
| | | −5 | 496 | −511 |
| | | −4 | 1207 | −1191 |
| | | −3 | 1491 | 1476 |
| | | −2 | 444 | 468 |
| | | −1 | 622 | −621 |
| | | 0 | 308 | −276 |
| | | 1 | 306 | 254 |
| | | 2 | 65 | −14 |
| | | 3 | 57 | −79 |
| | | 4 | 178 | −113 |
| | | 5 | 381 | −404 |
| | | 6 | 350 | −330 |
| | | 7 | 40 | 51 |
| | | 8 | <28 | 5 |
| | | 9 | <26 | −15 |
| | | 10 | 123 | −114 |
| 1 | 4 | −11 | 34 | 28 |
| | | −10 | 182 | −183 |
| | | −9 | 569 | −586 |
| | | −8 | 360 | −347 |
| | | −7 | 67 | −120 |
| | | −6 | 51 | −21 |
| | | −5 | 81 | 102 |
| | | −4 | 166 | −180 |
| | | −3 | 739 | −742 |
| | | −2 | 269 | −268 |
| | | −1 | 162 | −157 |
| | | 0 | 662 | −639 |
| | | 1 | 1975 | −2065 |
| | | 2 | 508 | 496 |
| | | 3 | 168 | 139 |
| | | 4 | 117 | −72 |
| | | 5 | 375 | −391 |
| | | 6 | <30 | 15 |
| | | 7 | 40 | 29 |
| | | 8 | <38 | 7 |
| | | 9 | 180 | −187 |
| | | 10 | 119 | 123 |
| 1 | 5 | −11 | 48 | 43 |
| | | −10 | 79 | 58 |
| | | −9 | 107 | 113 |
| | | −8 | 597 | −640 |
| | | −7 | 170 | 206 |
| | | −6 | 709 | 672 |
| | | −5 | 946 | 992 |
| | | −4 | 105 | −89 |
| | | −3 | 93 | −74 |
| | | −2 | 478 | 478 |
| | | −1 | 95 | −92 |
| | | 0 | 1460 | −1454 |
| | | 1 | 117 | 105 |
| | | 2 | 664 | −652 |
| | | 3 | 294 | 307 |
| | | 4 | 152 | 162 |
| | | 5 | <30 | −37 |
| | | 6 | 123 | 118 |
| | | 7 | 261 | −258 |
| | | 8 | <24 | 30 |
| | | 9 | <26 | −15 |
| 1 | 6 | −11 | 166 | 153 |
| | | −10 | 69 | 68 |
| | | −9 | 158 | −175 |
| | | −8 | 45 | 50 |
| | | −7 | <28 | 15 |
| | | −6 | 644 | 649 |
| | | −5 | 1003 | 1030 |
| | | −4 | 456 | 439 |
| | | −3 | 1095 | 1146 |
| | | −2 | 636 | −635 |
| | | −1 | 705 | 719 |
| | | 0 | 257 | 255 |
| | | 1 | 154 | −131 |
| | | 2 | 59 | −43 |
| | | 3 | 666 | 676 |
| | | 4 | 318 | 319 |
| | | 5 | 614 | 636 |
| | | 6 | 340 | −351 |
| | | 7 | 330 | 364 |
| | | 8 | 162 | −148 |
| 1 | 7 | −10 | 57 | 60 |
| | | −9 | 81 | −68 |
| | | −8 | 134 | −122 |
| | | −7 | 278 | −267 |
| | | −6 | 146 | 152 |
| | | −5 | 278 | 263 |
| | | −4 | 956 | 946 |
| | | −3 | 978 | −991 |
| | | −2 | 482 | 477 |
| | | −1 | 302 | −316 |
| | | 0 | 48 | 41 |
| | | 1 | 61 | −65 |
| | | 2 | 127 | 121 |
| | | 3 | 364 | 375 |
| | | 4 | 468 | 468 |
| | | 5 | 138 | 146 |
| | | 6 | 369 | 399 |
| | | 7 | 134 | −127 |
| | | 8 | 22 | 23 |
| 1 | 8 | −9 | <22 | 26 |
| | | −8 | 48 | 41 |
| | | −7 | 350 | −331 |
| | | −6 | 166 | 153 |
| | | −5 | <26 | −1 |
| | | −4 | 48 | −47 |
| | | −3 | 164 | 153 |
| | | −2 | 427 | −447 |
| | | −1 | 462 | −471 |
| | | 0 | 40 | 20 |
| | | 1 | 269 | −293 |
| | | 2 | 101 | 103 |
| 1 | 8 | 3 | 241 | −249 |
| | | 4 | 249 | 265 |
| | | 5 | 124 | 143 |
| | | 6 | 59 | −60 |
| | | 7 | 57 | −55 |
| 1 | 9 | −8 | 79 | −75 |
| | | −7 | 206 | 186 |
| | | −6 | 425 | −407 |
| | | −5 | 120 | 127 |
| | | −4 | 140 | 144 |
| | | −3 | <26 | 27 |
| | | −2 | 182 | −177 |
| | | −1 | 45 | −25 |
| | | 0 | <26 | −62 |
| | | 1 | 57 | 35 |
| | | 2 | 142 | −157 |
| | | 3 | 55 | 59 |
| | | 4 | <26 | −25 |
| | | 5 | 28 | −27 |
| 1 | 10 | −7 | 202 | −196 |
| | | −6 | 178 | −153 |
| | | −5 | 237 | −214 |
| | | −4 | 123 | −103 |
| | | −3 | 51 | −43 |
| | | −2 | 38 | 23 |
| | | −1 | 77 | −94 |
| | | 0 | 53 | −62 |
| | | 1 | <35 | 15 |
| | | 2 | 34 | 32 |
| | | 3 | 97 | −93 |
| | | 4 | 34 | −32 |
| 1 | 11 | −4 | 196 | −180 |
| | | −3 | 196 | 188 |
| | | −2 | 156 | −145 |
| | | −1 | 77 | 70 |
| | | 0 | 77 | −87 |
| | | 1 | <16 | 12 |
| 2 | −11 | −3 | 247 | −219 |
| | | −2 | 97 | −98 |
| | | −1 | 158 | −148 |
| | | 0 | 95 | −85 |
| | | 1 | 109 | −120 |
| | | 2 | <22 | 12 |
| | | 3 | 109 | −115 |
| 2 | −10 | −5 | 113 | 101 |
| | | −4 | 328 | −299 |
| | | −3 | 174 | 138 |
| | | −2 | <24 | 2 |
| | | −1 | 40 | −31 |
| | | 0 | 115 | −124 |
| | | 1 | 79 | 81 |
| | | 2 | 59 | 43 |
| | | 3 | 51 | −41 |
| | | 4 | 83 | −103 |
| | | 5 | 59 | 44 |
| 2 | −9 | −7 | <26 | −6 |
| | | −6 | 196 | −177 |
| | | −5 | 431 | −404 |
| | | −4 | 164 | −163 |
| | | −3 | 77 | 98 |
| | | −2 | 63 | −55 |
| | | −1 | 130 | −123 |
| | | 0 | 38 | 50 |
| | | 1 | 110 | −116 |
| | | 2 | 35 | −32 |
| | | 3 | 120 | −127 |
| | | 4 | 53 | −66 |
| | | 5 | 73 | −66 |
| | | 6 | <24 | 14 |
| | | 7 | <18 | −8 |
| 2 | −8 | −7 | <22 | 13 |
| | | −6 | 265 | −246 |
| | | −5 | <28 | −29 |
| | | −4 | <53 | −26 |
| | | −3 | <30 | −13 |
| | | −2 | 259 | 243 |
| | | −1 | 557 | 570 |
| | | 0 | 439 | −430 |
| | | 1 | <30 | 56 |
| | | 2 | 286 | −285 |
| | | 3 | 38 | −37 |
| | | 4 | 290 | −274 |
| | | 5 | 245 | −270 |
| | | 6 | 105 | 113 |
| | | 7 | 55 | 47 |
| | | 8 | 48 | −55 |
| 2 | −7 | −9 | 63 | 56 |
| | | −8 | 38 | 20 |
| | | −7 | 150 | −144 |
| | | −6 | 281 | 264 |
| | | −5 | 156 | 159 |
| | | −4 | 357 | −342 |
| | | −3 | 936 | 909 |
| 2 | −7 | −2 | 786 | 786 |
| | | −1 | 567 | 556 |
| | | 0 | 419 | 427 |
| | | 1 | 101 | 93 |
| | | 2 | 40 | 40 |
| | | 3 | 130 | 112 |
| | | 4 | 332 | −316 |
| | | 5 | 440 | 428 |
| | | 6 | 235 | −224 |
| | | 7 | 371 | 376 |
| | | 8 | 93 | 116 |
| 2 | −6 | −10 | 168 | 156 |
| | | −9 | 109 | −120 |
| | | −8 | 142 | −160 |
| | | −7 | 113 | −121 |
| | | −6 | 55 | −65 |
| | | −5 | 660 | −656 |
| | | −4 | 972 | 961 |
| | | −3 | 681 | −675 |
| | | −2 | 913 | 924 |
| | | −1 | 498 | 492 |
| | | 0 | 656 | 671 |
| | | 1 | 203 | −212 |
| | | 2 | 182 | −146 |
| | | 3 | 48 | −37 |
| | | 4 | 537 | 549 |
| | | 5 | 342 | −339 |
| | | 6 | 585 | 561 |
| | | 7 | 213 | 210 |
| | | 8 | 294 | 306 |
| | | 9 | 140 | 144 |
| 2 | −5 | −10 | 79 | −75 |
| | | −9 | 105 | 78 |
| | | −8 | <26 | −10 |
| | | −7 | 642 | −668 |
| | | −6 | 63 | −105 |
| | | −5 | 638 | 605 |
| | | −4 | 752 | −767 |
| | | −3 | 127 | −109 |
| | | −2 | <69 | 27 |
| | | −1 | 273 | 269 |
| | | 0 | 194 | 194 |
| | | 1 | 1067 | −1058 |
| | | 2 | 49 | −64 |
| | | 3 | 620 | −573 |
| | | 4 | 253 | −257 |
| | | 5 | 130 | 137 |
| | | 6 | <30 | −6 |
| | | 7 | 110 | 99 |
| | | 8 | 217 | 213 |
| | | 9 | 34 | 27 |
| 2 | −4 | −11 | 194 | −181 |
| | | −10 | 30 | −35 |
| | | −9 | 211 | 221 |
| | | −8 | 587 | −603 |
| | | −7 | 405 | 399 |
| | | −6 | 132 | −177 |
| | | −5 | 59 | 51 |
| | | −4 | <28 | 58 |
| | | −3 | 192 | 199 |
| | | −2 | 616 | −621 |
| | | −1 | 89 | 100 |
| | | 0 | 83 | −108 |
| | | 1 | 504 | 510 |
| | | 2 | 1276 | −1302 |
| | | 3 | 290 | −278 |
| | | 4 | <59 | 12 |
| | | 5 | <30 | 18 |
| | | 6 | 194 | −216 |
| | | 7 | <28 | 18 |
| | | 8 | <35 | 5 |
| | | 9 | <26 | 22 |
| | | 10 | 105 | −107 |
| 2 | −3 | −11 | 263 | −244 |
| | | −10 | 285 | −269 |
| | | −9 | 219 | −230 |
| | | −8 | 555 | 587 |
| | | −7 | 357 | −384 |
| | | −6 | 528 | −503 |
| | | −5 | 87 | 92 |
| | | −4 | 332 | 343 |
| | | −3 | 1029 | −924 |
| | | −2 | 1225 | −1211 |
| | | −1 | 53 | 96 |
| | | 0 | 322 | 310 |
| | | 1 | 306 | −270 |
| | | 2 | 166 | −121 |
| | | 3 | 138 | −103 |
| | | 4 | <28 | 2 |
| | | 5 | 158 | −134 |
| | | 6 | 174 | 187 |
| | | 7 | 209 | −209 |
| | | 8 | <26 | −12 |
| 2 | −3 | 9 | <24 | −17 |
| | | 10 | <20 | 16 |
| 2 | −2 | −11 | 322 | −325 |
| | | −10 | 160 | −152 |
| | | −9 | 409 | 413 |
| | | −8 | 332 | 309 |
| | | −7 | 128 | 149 |
| | | −6 | 300 | 311 |
| | | −5 | 123 | 99 |
| | | −4 | 958 | −873 |
| | | −3 | 130 | 110 |
| | | −2 | 1306 | −1260 |
| | | −1 | 992 | −979 |
| | | 0 | 624 | −597 |
| | | 1 | 739 | 720 |
| | | 2 | 211 | 217 |
| | | 3 | 401 | −424 |
| | | 4 | 500 | −480 |
| | | 5 | 381 | 361 |
| | | 6 | 504 | −494 |
| | | 7 | 61 | −59 |
| | | 8 | 182 | −173 |
| | | 9 | <24 | −5 |
| | | 10 | 53 | 58 |
| 2 | −1 | −12 | 51 | 34 |
| | | −11 | 87 | −62 |
| | | −10 | 71 | 55 |
| | | −9 | 91 | 75 |
| | | −8 | 41 | 724 |
| | | −7 | 1371 | 1409 |
| | | −6 | 233 | 228 |
| | | −5 | 484 | −450 |
| | | −4 | 1266 | 1274 |
| | | −3 | 127 | −119 |
| | | −2 | 156 | −169 |
| | | −1 | 869 | −902 |
| | | 0 | 585 | 530 |
| | | 1 | 1748 | 1737 |
| | | 2 | 57 | 67 |
| | | 3 | 737 | 676 |
| | | 4 | 110 | 104 |
| | | 5 | 352 | −351 |
| | | 6 | <28 | 46 |
| | | 7 | 172 | −160 |
| | | 8 | 166 | −139 |
| | | 9 | 120 | 109 |
| | | 10 | 89 | −77 |
| 2 | 0 | −12 | <26 | −10 |
| | | −11 | <22 | 9 |
| | | −10 | 278 | −276 |
| | | −9 | 99 | −100 |
| | | −8 | 1120 | 1123 |
| | | −7 | 105 | −95 |
| | | −6 | 749 | 777 |
| | | −5 | 115 | −109 |
| | | −4 | 120 | −107 |
| | | −3 | 107 | −123 |
| | | −2 | 488 | 510 |
| | | −1 | 51 | −29 |
| | | 0 | 1310 | 1310 |
| | | 1 | 73 | −80 |
| | | 2 | 2963 | 2965 |
| | | 3 | <28 | −8 |
| | | 4 | 206 | 277 |
| | | 5 | <28 | 5 |
| | | 6 | 97 | 106 |
| | | 7 | <26 | −11 |
| | | 8 | 91 | 75 |
| | | 9 | 31 | −28 |
| | | 10 | 308 | 284 |
| 2 | 1 | −12 | <24 | −10 |
| | | −11 | 73 | −67 |
| | | −10 | 44 | −28 |
| | | −9 | 134 | 125 |
| | | −8 | 802 | −811 |
| | | −7 | 1211 | 1255 |
| | | −6 | 132 | −133 |
| | | −5 | 492 | −450 |
| | | −4 | 1197 | −1198 |
| | | −3 | 334 | −329 |
| | | −2 | 233 | 268 |
| | | −1 | 804 | −872 |
| | | 0 | 620 | −577 |
| | | 1 | 1545 | 1482 |
| | | 2 | <28 | −35 |
| | | 3 | 581 | 557 |
| | | 4 | <28 | −46 |
| | | 5 | 318 | −312 |
| | | 6 | <30 | 15 |
| | | 7 | 166 | −139 |
| | | 8 | 127 | 103 |
| | | 9 | 123 | 107 |
| | | 10 | 59 | 48 |

TABLE 2. (*Continued.*)

| h | k | l | 100$F_o$ | 100$F_c$ |
|---|---|---|---|---|
| 2 | 2 | −12 | 152 | −141 |
| | | −11 | 409 | 402 |
| | | −10 | 172 | −169 |
| | | −9 | 401 | −409 |
| | | −8 | 168 | 158 |
| | | −7 | <40 | 7 |
| | | −6 | 132 | 137 |
| | | −5 | <28 | 1 |
| | | −4 | 855 | −806 |
| | | −3 | 130 | 142 |
| | | −2 | 1075 | −1059 |
| | | −1 | 678 | 674 |
| | | 0 | 390 | −390 |
| | | 1 | 533 | −513 |
| | | 2 | 110 | 116 |
| | | 3 | 322 | 321 |
| | | 4 | 391 | −363 |
| | | 5 | 247 | −229 |
| | | 6 | 391 | −387 |
| | | 7 | 38 | 62 |
| | | 8 | 127 | −117 |
| | | 9 | <24 | 5 |
| 2 | 3 | −11 | 286 | −270 |
| | | −10 | 196 | 181 |
| | | −9 | 251 | −255 |
| | | −8 | 537 | −547 |
| | | −7 | 419 | −459 |
| | | −6 | 350 | 339 |
| | | −5 | 119 | 130 |
| | | −4 | 239 | −257 |
| | | −3 | 798 | −743 |
| | | −2 | 838 | 853 |
| | | −1 | 55 | 106 |
| | | 0 | 336 | −319 |
| | | 1 | 302 | −299 |
| | | 2 | 109 | 112 |
| | | 3 | 69 | −70 |
| | | 4 | 48 | −60 |
| | | 5 | 152 | −142 |
| | | 6 | 81 | −115 |
| | | 7 | 132 | −146 |
| | | 8 | <22 | −7 |
| | | 9 | 38 | −33 |
| 2 | 4 | −11 | 160 | 152 |
| | | −10 | 69 | 53 |
| | | −9 | 221 | −221 |
| | | −8 | 587 | −602 |
| | | −7 | 308 | −290 |
| | | −6 | 28 | −19 |
| | | −5 | <48 | −9 |
| | | −4 | <30 | −53 |
| | | −3 | 44 | −25 |
| | | −2 | 328 | −359 |
| | | −1 | 158 | −166 |
| | | 0 | 223 | −256 |
| | | 1 | 239 | −226 |
| | | 2 | 790 | −795 |
| | | 3 | 188 | 179 |
| | | 4 | <28 | −31 |
| | | 5 | <40 | −9 |
| | | 6 | 124 | −124 |
| | | 7 | <24 | −22 |
| | | 8 | 31 | −17 |
| 2 | 5 | −11 | 105 | 96 |
| | | −10 | 71 | 60 |
| | | −9 | 34 | 20 |
| | | −8 | 120 | 123 |
| | | −7 | 476 | −505 |
| | | −6 | 227 | 253 |
| | | −5 | 561 | 544 |
| | | −4 | 699 | 712 |
| | | −3 | <28 | 33 |
| | | −2 | 65 | −74 |
| | | −1 | 203 | 205 |
| | | 0 | 69 | −55 |
| | | 1 | 757 | −710 |
| | | 2 | 91 | 104 |
| | | 3 | 249 | −241 |
| | | 4 | 154 | 166 |
| | | 5 | 97 | 92 |
| | | 6 | 57 | −44 |
| | | 7 | 97 | 88 |
| | | 8 | 87 | −100 |
| 2 | 6 | −10 | 152 | 136 |
| | | −9 | 95 | 101 |
| | | −8 | 186 | −202 |
| | | −7 | 38 | 45 |
| | | −6 | 83 | 78 |
| | | −5 | 551 | 546 |
| | | −4 | 776 | 781 |
| | | −3 | 229 | 211 |
| | | −2 | 745 | 756 |
| | | −1 | 310 | −298 |
| | | 0 | 277 | 298 |
| | | 1 | 93 | 92 |
| | | 2 | 48 | −35 |
| | | 3 | 71 | 74 |
| | | 4 | 312 | 324 |
| | | 5 | 99 | 103 |
| | | 6 | 346 | 320 |
| | | 7 | 136 | −140 |
| | | −9 | <26 | 41 |
| | | −8 | 83 | −76 |
| | | −7 | 123 | −117 |
| | | −6 | 202 | −195 |
| | | −5 | 162 | 159 |
| | | −4 | 124 | 92 |
| | | −3 | 594 | 645 |
| | | −2 | 622 | −625 |
| | | −1 | 294 | 291 |
| | | 0 | 203 | −202 |
| | | 1 | <26 | 13 |
| | | 2 | <26 | −13 |
| | | 3 | 77 | 73 |
| | | 4 | 158 | 174 |
| | | 5 | 273 | 248 |
| | | 6 | <20 | 25 |
| 2 | 8 | −8 | <22 | 19 |
| | | −7 | 53 | 36 |
| | | −6 | 281 | −265 |
| | | −5 | 103 | 113 |
| | | −4 | 44 | 52 |
| | | −3 | 93 | −100 |
| | | −2 | 83 | 86 |
| | | −1 | 265 | −256 |
| | | 0 | 227 | −232 |
| | | 1 | <24 | −5 |
| | | 2 | 168 | −176 |
| | | 3 | 91 | 79 |
| | | 4 | 87 | −77 |
| | | 5 | 75 | 93 |
| 2 | 9 | −7 | 99 | −103 |
| | | −6 | 156 | 142 |
| | | −5 | 347 | −288 |
| | | −4 | 75 | 78 |
| | | −3 | 65 | 74 |
| | | −2 | <22 | 8 |
| | | −1 | 110 | −105 |
| | | 0 | <22 | −7 |
| | | 1 | 73 | −69 |
| | | 2 | 30 | 28 |
| | | 3 | 85 | −95 |
| 2 | 10 | −4 | 160 | −152 |
| | | −3 | 67 | −57 |
| | | −2 | 44 | −48 |
| | | −1 | <24 | 8 |
| | | 0 | 61 | −55 |
| 3 | −10 | −4 | 91 | 92 |
| | | −3 | 237 | −223 |
| | | −2 | 93 | 83 |
| | | −1 | 30 | −26 |
| | | 0 | <24 | −12 |
| | | 1 | 53 | −73 |
| | | 2 | 31 | 28 |
| | | 3 | <16 | −15 |
| 3 | −9 | −6 | 44 | −34 |
| | | −5 | 154 | −142 |
| | | −4 | 322 | −300 |
| | | −3 | 117 | −115 |
| | | −2 | 49 | 64 |
| | | −1 | <24 | −13 |
| | | 0 | 83 | −84 |
| | | 1 | <24 | 24 |
| | | 2 | 79 | −90 |
| | | 3 | 40 | −25 |
| | | 4 | 81 | −95 |
| | | 5 | 26 | −39 |
| 3 | −8 | −7 | 28 | 37 |
| | | −6 | 41 | 23 |
| | | −5 | 206 | −200 |
| | | −4 | 41 | −32 |
| | | −3 | 49 | 37 |
| | | −2 | 79 | 75 |
| | | −1 | 142 | 141 |
| | | 0 | 356 | 347 |
| | | 1 | 217 | −199 |
| | | 2 | 59 | 64 |
| | | 3 | 168 | −166 |
| | | 4 | 55 | −46 |
| | | 5 | 71 | −83 |
| 3 | −7 | −8 | 49 | 36 |
| | | −7 | 61 | 33 |
| | | −6 | 136 | −136 |
| | | −5 | 176 | 167 |
| | | −4 | 166 | 164 |
| | | −3 | 194 | −184 |
| | | −2 | 620 | 590 |
| | | −1 | 466 | 476 |
| | | 0 | 347 | 325 |
| | | 1 | 257 | 245 |
| | | 2 | 45 | 37 |
| | | 3 | <26 | 12 |
| | | 4 | 67 | 75 |
| | | 5 | 156 | −167 |
| | | 6 | 168 | 226 |
| | | 7 | 44 | −63 |
| 3 | −6 | −9 | 160 | 139 |
| | | −8 | 127 | −130 |
| | | −7 | 160 | −170 |
| | | −6 | 115 | −116 |
| | | −5 | <28 | 31 |
| | | −4 | 551 | −541 |
| | | −3 | 670 | 666 |
| | | −2 | 354 | −349 |
| | | −1 | 593 | 580 |
| | | 0 | 237 | 214 |
| | | 1 | 257 | 258 |
| | | 2 | 55 | −62 |
| | | 3 | 48 | −36 |
| | | 4 | 97 | −103 |
| | | 5 | 249 | 253 |
| | | 6 | 123 | −131 |
| | | 7 | 229 | 257 |
| 3 | −5 | −10 | 63 | 45 |
| | | −9 | 75 | −79 |
| | | −8 | 51 | 30 |
| | | −7 | <26 | −4 |
| | | −6 | 444 | −448 |
| | | −5 | 136 | −161 |
| | | −4 | 421 | 396 |
| | | −3 | 472 | −483 |
| | | −2 | <30 | 1 |
| | | −1 | 48 | 25 |
| | | 0 | 124 | 90 |
| | | 1 | 117 | 117 |
| | | 2 | 500 | −478 |
| | | 3 | 48 | −56 |
| | | 4 | 237 | −196 |
| | | 5 | 119 | −128 |
| | | 6 | 63 | 66 |
| | | 7 | <26 | 12 |
| | | 8 | 49 | 62 |
| 3 | −4 | −10 | 188 | −178 |
| | | −9 | 53 | −48 |
| | | −8 | 231 | 236 |
| | | −7 | 523 | −517 |
| | | −6 | 304 | 287 |
| | | −5 | 79 | −90 |
| | | −4 | <67 | 25 |
| | | −3 | 61 | −71 |
| | | −2 | <30 | 6 |
| | | −1 | 263 | −272 |
| | | 0 | 75 | 82 |
| | | 1 | 180 | −209 |
| | | 2 | 186 | 180 |
| | | 3 | 470 | −466 |
| | | 4 | 99 | −98 |
| | | 5 | 69 | −62 |
| | | 6 | <26 | −14 |
| | | 7 | 55 | −67 |
| | | 8 | <20 | 21 |
| 3 | −3 | −11 | 136 | −143 |
| | | −10 | 239 | −232 |
| | | −9 | 192 | −177 |
| | | −8 | 217 | −228 |
| | | −7 | 425 | 436 |
| | | −6 | 217 | −233 |
| | | −5 | 298 | −288 |
| | | −4 | <30 | 6 |
| | | −3 | 85 | 89 |
| | | −2 | 510 | −453 |
| | | −1 | 514 | −496 |
| | | 0 | 95 | −77 |
| | | 1 | 124 | 130 |
| | | 2 | 136 | −146 |
| | | 3 | 55 | −43 |
| | | 4 | 67 | −72 |
| | | 5 | <28 | 10 |
| | | 6 | 113 | −112 |
| | | 7 | 41 | 45 |
| | | 8 | 81 | −91 |
| 3 | −2 | −11 | 81 | −62 |
| | | −10 | 229 | −295 |
| | | −9 | 119 | −113 |
| | | −8 | 401 | 395 |
| | | −7 | 320 | 294 |
| | | −6 | 97 | 91 |
| | | −5 | 160 | 155 |
| | | −4 | 142 | 132 |
| | | −3 | 579 | −524 |
| | | −2 | <30 | −23 |
| | | −1 | 640 | −584 |
| | | 0 | 233 | −228 |
| | | 1 | 160 | −142 |
| | | 2 | 207 | 188 |
| | | 3 | 67 | 74 |
| | | 4 | 55 | −56 |
| | | 5 | 202 | −192 |
| | | 6 | 93 | 99 |
| | | 7 | 190 | −195 |
| | | 8 | <22 | −2 |
| 3 | −1 | −11 | <41 | 15 |
| | | −10 | 87 | −70 |
| | | −9 | 110 | 103 |
| | | −8 | 166 | 154 |
| | | −7 | 567 | 547 |
| | | −6 | 950 | 925 |
| | | −5 | 278 | 264 |
| | | −4 | 245 | −223 |
| | | −3 | 628 | 583 |
| | | −2 | 192 | −173 |
| | | −1 | <28 | −34 |
| | | 0 | 190 | −197 |
| | | 1 | 243 | 198 |
| | | 2 | 717 | 696 |
| | | 3 | 61 | 50 |
| | | 4 | 263 | 239 |
| | | 5 | 53 | 47 |
| | | 6 | 115 | −118 |
| | | 7 | <22 | 6 |
| | | 8 | 63 | −54 |
| 3 | 0 | −11 | 26 | −15 |
| | | −10 | <22 | 3 |
| | | −9 | 202 | −208 |
| | | −8 | 109 | −103 |
| | | −7 | 903 | 874 |
| | | −6 | 73 | −78 |
| | | −5 | 462 | 456 |
| | | −4 | 85 | −63 |
| | | −3 | <57 | 15 |
| | | −2 | 57 | −37 |
| | | −1 | 178 | 133 |
| | | 0 | 45 | −31 |
| | | 1 | 717 | 692 |
| | | 2 | <31 | −47 |
| | | 3 | 1021 | 982 |
| | | 4 | <30 | −3 |
| | | 5 | 162 | 146 |
| | | 6 | <26 | 8 |
| | | 7 | <22 | 1 |
| | | 8 | <20 | −4 |
| 3 | 1 | −11 | <20 | 8 |
| | | −10 | 65 | −62 |
| | | −9 | 97 | −98 |
| | | −8 | 198 | 198 |
| | | −7 | 571 | −587 |
| | | −6 | 772 | 775 |
| | | −5 | 192 | −184 |
| | | −4 | 255 | −232 |
| | | −3 | 541 | −514 |
| | | −2 | 285 | −261 |
| | | −1 | <26 | 41 |
| | | 0 | 174 | −187 |
| | | 1 | 263 | −238 |
| | | 2 | 593 | 567 |
| | | 3 | <30 | −47 |
| | | 4 | 203 | 193 |
| | | 5 | <26 | −15 |
| | | 6 | 103 | −106 |
| | | 7 | 26 | 17 |
| | | 8 | 51 | −45 |
| 3 | 2 | −11 | 142 | −130 |
| | | −10 | 330 | 322 |
| | | −9 | 128 | −123 |
| | | −8 | 338 | −345 |
| | | −7 | 134 | 135 |
| | | −6 | <28 | 14 |
| | | −5 | 55 | 52 |
| | | −4 | <26 | −22 |
| | | −3 | 450 | −444 |
| | | −2 | 138 | 135 |
| | | −1 | 480 | −470 |
| | | 0 | 172 | 148 |
| | | 1 | 117 | −109 |
| | | 2 | 119 | −121 |
| | | 3 | 40 | 36 |
| | | 4 | 38 | 46 |
| | | 5 | 156 | −139 |
| | | 6 | 49 | −47 |
| | | 7 | 134 | −146 |
| 3 | 3 | −10 | 235 | −232 |
| | | −9 | 91 | 77 |
| | | −8 | 221 | −234 |
| | | −7 | 342 | −356 |
| | | −6 | 255 | −262 |
| | | −5 | 149 | 140 |
| | | −4 | <48 | −2 |
| | | −3 | <28 | −13 |
| | | −2 | 344 | −343 |
| | | −1 | 292 | 291 |
| | | 0 | 61 | −62 |
| | | 1 | 127 | −114 |
| | | 2 | 117 | −131 |
| | | 3 | <28 | 27 |
| | | 4 | 44 | −53 |
| | | 5 | <24 | −13 |
| | | 6 | 79 | −85 |
| | | 7 | <18 | −28 |
| 3 | 4 | −10 | 140 | 125 |
| | | −9 | 34 | 20 |
| | | −8 | 188 | −177 |
| | | −7 | 391 | −407 |
| | | −6 | 176 | −167 |
| | | −5 | <26 | −13 |
| | | −4 | <28 | 25 |
| | | −3 | 40 | −67 |
| | | −2 | 28 | 25 |
| | | −1 | 134 | −136 |
| | | 0 | 95 | −98 |
| | | 1 | 184 | −205 |
| | | 2 | 59 | −64 |
| | | 3 | 271 | −275 |
| | | 4 | 67 | 64 |
| | | 5 | 55 | −49 |
| | | 6 | <22 | 4 |
| 3 | 5 | −9 | 67 | 62 |
| | | −8 | <22 | −31 |
| | | −7 | 93 | 95 |
| | | −6 | 249 | −247 |
| | | −5 | 215 | 223 |
| | | −4 | 288 | 284 |
| | | −3 | 369 | 363 |
| | | −2 | 97 | 105 |
| | | −1 | <28 | −8 |
| | | 0 | 38 | 44 |
| | | 1 | <26 | −11 |
| | | 2 | 257 | −260 |
| | | 3 | 51 | 69 |
| | | 4 | 97 | −73 |
| | | 5 | 67 | 65 |
| 3 | 6 | −8 | 79 | 64 |
| | | −7 | 160 | −151 |
| | | −6 | 45 | 50 |
| | | −5 | 67 | 82 |
| | | −4 | 318 | 307 |
| | | −3 | 433 | 434 |
| | | −2 | 87 | 62 |
| | | −1 | 369 | 374 |
| | | 0 | 117 | −116 |
| | | 1 | 91 | 100 |
| | | 2 | <24 | 24 |
| | | 3 | <26 | 7 |
| | | 4 | 67 | 61 |
| 3 | 7 | −7 | 85 | −73 |
| | | −6 | 77 | −80 |
| | | −5 | 110 | −83 |
| | | −4 | 120 | 125 |
| | | −3 | <22 | −16 |
| | | −2 | 342 | 335 |
| | | −1 | 281 | −285 |
| | | 0 | 140 | 138 |
| | | 1 | 128 | −106 |
| | | 2 | <22 | −4 |
| | | 3 | <22 | 9 |
| 3 | 8 | −5 | 152 | −145 |
| | | −4 | 55 | 51 |
| | | −3 | 53 | 53 |
| | | −2 | 83 | −80 |
| | | −1 | 49 | 46 |
| | | 0 | 120 | −110 |
| | | 1 | 81 | −87 |
| 4 | −8 | −5 | <16 | 20 |
| | | −4 | 113 | −110 |
| | | −3 | 28 | −19 |
| | | −2 | 35 | 49 |
| | | −1 | 51 | 78 |
| | | 0 | 45 | 69 |
| | | 1 | 144 | 157 |
| | | 2 | 69 | −71 |
| 4 | −7 | −6 | 48 | 48 |
| | | −5 | 73 | −85 |
| | | −4 | 57 | 51 |
| | | −3 | 101 | 121 |
| | | −2 | 59 | −56 |
| | | −1 | 292 | 293 |

TABLE 2. (*Continued.*)

| h | k | l | 100$F_o$ | 100$F_c$ | h | k | l | 100$F_o$ | 100$F_c$ | h | k | l | 100$F_o$ | 100$F_c$ | h | k | l | 100$F_o$ | 100$F_c$ | h | k | l | 100$F_o$ | 100$F_c$ |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 4 | −7 | 0 | 198 | 212 | 4 | −4 | 1 | 55 | 62 | 4 | −1 | −5 | 466 | 433 | 4 | 1 | 4 | <26 | −29 | 4 | 5 | −5 | 95 | −82 |
| | | 1 | 120 | 145 | | | 2 | 128 | −137 | | | −4 | 206 | 183 | | | 5 | 65 | 64 | | | −4 | 154 | 131 |
| | | 2 | 97 | 117 | | | 3 | 65 | 55 | | | −3 | 71 | −54 | 4 | 2 | −9 | 150 | 156 | | | −3 | 140 | 115 |
| | | 3 | <22 | 10 | | | 4 | 123 | −150 | | | −2 | 243 | 209 | | | −8 | 44 | −68 | | | −2 | 172 | 146 |
| 4 | −6 | −7 | 75 | −78 | | | 5 | <22 | −35 | | | −1 | 144 | −115 | | | −7 | 164 | −200 | | | −1 | 91 | 75 |
| | | −6 | 101 | −115 | 4 | −3 | −9 | 160 | −172 | | | 0 | <30 | 42 | | | −6 | 69 | 79 | | | 0 | <22 | 10 |
| | | −5 | 99 | −95 | | | −8 | 69 | −57 | | | 1 | <26 | 8 | | | −5 | 34 | −32 | | | 1 | <18 | 3 |
| | | −4 | 49 | 61 | | | −7 | 156 | −165 | | | 2 | 93 | 72 | | | −4 | <24 | 10 | 4 | 6 | −4 | 41 | 50 |
| | | −3 | 292 | −305 | | | −6 | 229 | 223 | | | 3 | 251 | 250 | | | −3 | <26 | −3 | | | −3 | 130 | 123 |
| | | −2 | 320 | 339 | | | −5 | 117 | −103 | | | 4 | 49 | 27 | | | −2 | 174 | −197 | | | −2 | 184 | 189 |
| | | −1 | 136 | −137 | | | −4 | 87 | −82 | | | 5 | 57 | 82 | | | −1 | 67 | 71 | 5 | −4 | −4 | 20 | 47 |
| | | 0 | 253 | 274 | | | −3 | 59 | −61 | 4 | 0 | −9 | <28 | −7 | | | 0 | 188 | −176 | | | −3 | 22 | −34 |
| | | 1 | 79 | 76 | | | −2 | <28 | −29 | | | −8 | 69 | −77 | | | 1 | 41 | 21 | | | −2 | <20 | −26 |
| | | 2 | 91 | 81 | | | −1 | 206 | −205 | | | −7 | 69 | −73 | | | 2 | 31 | −24 | | | −1 | 22 | −33 |
| | | 3 | <26 | −12 | | | 0 | 120 | −144 | | | −6 | 478 | 470 | | | 3 | 34 | −30 | 5 | −3 | −4 | 38 | −43 |
| | | 4 | <20 | 6 | | | 1 | 77 | −83 | | | −5 | 38 | −45 | | | 4 | <20 | 11 | | | −3 | <22 | −11 |
| 4 | −5 | −8 | 53 | −58 | | | 2 | <26 | 41 | | | −4 | 235 | 221 | 4 | 3 | −8 | <26 | 2 | | | −2 | 44 | −52 |
| | | −7 | 31 | −23 | | | 3 | 55 | −59 | | | −3 | 38 | −27 | | | −7 | 142 | −156 | | | −1 | 24 | −29 |
| | | −6 | <41 | −5 | | | 4 | <28 | −6 | | | −2 | <59 | −14 | | | −6 | 150 | −167 | | | 0 | 69 | −83 |
| | | −5 | 194 | −193 | | | 5 | 28 | −44 | | | −1 | <28 | −18 | | | −5 | 109 | −117 | 5 | −2 | −5 | 57 | 86 |
| | | −4 | 140 | −140 | 4 | −2 | −9 | 154 | −158 | | | 0 | <28 | 34 | | | −4 | 24 | 35 | | | −4 | 59 | 64 |
| | | −3 | 182 | 175 | | | −8 | 63 | −53 | | | 1 | 35 | −28 | | | −3 | 48 | −61 | | | −3 | 34 | 25 |
| | | −2 | 225 | −237 | | | −7 | 239 | 247 | | | 2 | 308 | 305 | | | −2 | 53 | 42 | | | −2 | <22 | 33 |
| | | −1 | 75 | 60 | | | −6 | 196 | 186 | | | 3 | 45 | −24 | | | −1 | 152 | −148 | | | −1 | 85 | −81 |
| | | 0 | <26 | −7 | | | −5 | 101 | 86 | | | 4 | 278 | 304 | | | 0 | 59 | 63 | | | 0 | <40 | −6 |
| | | 1 | <26 | 3 | | | −4 | 71 | 63 | | | 5 | <22 | −1 | | | 1 | 73 | −68 | 5 | −1 | −5 | 87 | 114 |
| | | 2 | 45 | 46 | | | −3 | 61 | 76 | 4 | 1 | −9 | 53 | −41 | | | 2 | 34 | −36 | | | −4 | 154 | 161 |
| | | 3 | 170 | −168 | | | −2 | 245 | −227 | | | −8 | 85 | −111 | | | 3 | 45 | −54 | | | −3 | 99 | 80 |
| | | 4 | <24 | −34 | | | −1 | 53 | −33 | | | −7 | 154 | 174 | 4 | 4 | −7 | 97 | −92 | | | −2 | <22 | −7 |
| | | 5 | 49 | −54 | | | 0 | 198 | −221 | | | −6 | 288 | −302 | | | −6 | 199 | −189 | | | −1 | 65 | 63 |
| 4 | −4 | −8 | 61 | −72 | | | 1 | 38 | −36 | | | −5 | 346 | 354 | | | −5 | 69 | −58 | | | 0 | 59 | −45 |
| | | −7 | 160 | 170 | | | 2 | 35 | −25 | | | −4 | 138 | −138 | | | −4 | <22 | −12 | 5 | 0 | −4 | 24 | −19 |
| | | −6 | 300 | −297 | | | 3 | 61 | 45 | | | −3 | 95 | −57 | | | −3 | 38 | 42 | | | −3 | 95 | 88 |
| | | −5 | 148 | 143 | | | 4 | 30 | 20 | | | −2 | 176 | −163 | | | −2 | <22 | −25 | | | −2 | <20 | −8 |
| | | −4 | 51 | −56 | | | 5 | <30 | 6 | | | −1 | 91 | −128 | | | −1 | 30 | 24 | | | −1 | <24 | −13 |
| | | −3 | <28 | −14 | 4 | −1 | −9 | 71 | −59 | | | 0 | 67 | −34 | | | 0 | 59 | −53 | 5 | 1 | −3 | 55 | −62 |
| | | −2 | <30 | −65 | | | −8 | 89 | 103 | | | 1 | <65 | −4 | | | 1 | 49 | −39 | | | −2 | <24 | −7 |
| | | −1 | <28 | −28 | | | −7 | 160 | 160 | | | 2 | 81 | −90 | | | 2 | 95 | −98 | | | | | |
| | | 0 | 57 | −110 | | | −6 | 304 | 289 | | | 3 | 198 | 195 | 4 | 5 | −6 | 49 | 52 | | | | | |

Reflections marked < were "unobserved"; those marked * were assigned zero weight.

for which the error in intensity measurement was greater than the measurement itself, were treated as "unobserved," and assigned threshold values $I_t$ equal to this error.

Trial $x$-co-ordinates were obtained from the fit of a planar molecular model to the (0$kl$) projection, and partially refined by a least-squares analysis of the ($hk$0) data. Three-dimensional refinement proceeded through a series of isotropic cycles with a limited number of reflections, location of the hydrogen atoms from difference maps $\delta(yz)$ and $\delta(xz)$, and anisotropic (C and O only) cycles of all reflections. In the final cycles the positional and (isotropic) thermal parameters of the hydrogens were also allowed to move. The following scattering-factor curves were used throughout: $f_{C,O}$, Berghuis *et al.*,[10] $f_H$, McWeeny.[11] Refinement was stopped when the residual shifts as well as the accumulated shifts of the last four cycles, damped by a factor of $\frac{1}{2}$, were smaller than 0·001 and 0·015 Å for carbon and hydrogen atoms, respectively. The "final" agreement factor $r$ was 0·014, the conventional agreement factor $R = \Sigma(|kF_o - |F_c||)/\Sigma kF_o$ was 0·050. These agreement factors excluded all those "unobserved" terms for which $kF_t > |F_c|$, $F$(101) believed to suffer from secondary extinction, as well as $F$(001), $F$(003), $F$(020), and $F$(030) affected by white-radiation streaks of neighbouring strong reflections. Observed and calculated structure factors are listed in Table 2.

Standard deviations of parameters other than $\beta_{ii}$ and $k^2$ were estimated from the diagonal approximation:

$$\sigma^2(u_i) = \sum w(k^2F_o^2 - |F_c|^2)^2/(n - s)A_{ii} \qquad (2)$$

where $n(= 1354 + 49 = 1403)$ is the number of reflections, observed plus threshold included in the summation of the last cycle, and $s$ (= 123) the number of the parameters adjusted. $A_{ii} = \Sigma w(\partial|F_c|^2/\partial u_i)^2$ is the diagonal element in the normal equation corresponding to the parameter $u_i$. The approximate validity of the diagonal approximation follows from the near-orthogonality of the axes and the large number of observed reflections.

The e.s.d. in positional co-ordinates averaged about 0·0011 Å for the oxygen, 0·0013 Å for the ring carbons, and 0·0016 Å for the methyl carbons; the e.s.d. of the methyl and ring

[10] Berghuis, Haanappel, Potters, Loopstra, MacGillavry, and Veenendaal, *Acta Cryst.*, 1955, **8**, 478.
[11] McWeeny, *Acta Cryst.*, 1951, **4**, 513.

hydrogens averaged 0·018 and 0·015 Å, respectively. The numbering of atoms is shown in diagram I.

TABLE 3.

Atomic co-ordinates and standard deviations (in Å) referred to axes *a*,*b*,*c*.

| Atom | | *x* | *y* | *z* | $\sigma(x)$ | $\sigma(y)$ | $\sigma(z)$ | r.m.s. $\sigma$ |
|---|---|---|---|---|---|---|---|---|
| | | | | Molecule at (000) | | | | |
| O | 1 * | −0·7888 | −0·9994 | 2·1563 | 0·0013 | 0·0011 | 0·0009 | 0·0011 |
| C | 2 | 0·1822 | 2·9354 | −0·2794 | 0·0019 | 0·0014 | 0·0017 | 0·0017 |
| | 3 | −0·4243 | −0·5449 | 1·1564 | 0·0016 | 0·0013 | 0·0013 | 0·0014 |
| | 4 | 0·0716 | 1·4514 | −0·0911 | 0·0015 | 0·0012 | 0·0013 | 0·0103 |
| | 5 | −0·3267 | 0·9069 | 0·9833 | 0·0016 | 0·0013 | 0·0013 | 0·0014 |
| H | 6 | −0·524 | 1·423 | 1·670 | 0·018 | 0·013 | 0·017 | 0·016 |
| | 7 | −0·103 | 3·410 | 0·445 | 0·017 | 0·014 | 0·015 | 0·015 |
| | 8 | 1·093 | 3·306 | −0·312 | 0·017 | 0·016 | 0·018 | 0·017 |
| | 9 | −0·375 | 3·036 | −1·145 | 0·020 | 0·015 | 0·017 | 0·018 |
| | | | | Molecule at ($\frac{1}{2}\frac{1}{2}\frac{1}{2}$) | | | | |
| O | 1 | 2·7696 | 3·5881 | 2·7318 | 0·0013 | 0·0010 | 0·0009 | 0·0011 |
| C | 2 | 2·1319 | 1·7481 | 4·6476 | 0·0018 | 0·0013 | 0·0016 | 0·0016 |
| | 3 | 2·4052 | 4·0827 | 3·7156 | 0·0015 | 0·0013 | 0·0012 | 0·0013 |
| | 4 | 2·0513 | 3·2342 | 4·8087 | 0·0015 | 0·0012 | 0·0013 | 0·0013 |
| | 5 | 1·6790 | 3·8265 | 5·8756 | 0·0015 | 0·0012 | 0·0012 | 0·0013 |
| H | 6 | 1·442 | 3·305 | 6·590 | 0·015 | 0·013 | 0·015 | 0·014 |
| | 7 | 1·838 | 1·265 | 5·310 | 0·018 | 0·015 | 0·017 | 0·017 |
| | 8 | 1·489 | 1·345 | 3·841 | 0·021 | 0·016 | 0·019 | 0·019 |
| | 9 | 2·954 | 1·652 | 4·535 | 0·020 | 0·017 | 0·019 | 0·019 |

* Refer to diagram for numbering of atoms.

Table 3 lists the co-ordinates, in Å, of all atoms in the asymmetric unit referred to the unit-cell axes, together with their e.s.d. Fig. 1 shows $\rho(yz)$. The experimental molecular dimensions are given in Fig. 2. Equivalent bonds between heavy atoms in the two molecules differ in length by 0·0037 Å r.m.s., maximum 0·0050 Å, corresponding to an r.m.s. co-ordinate error of 0·0019 Å; the latter figure is slightly larger than the r.m.s. co-ordinate error of 0·0014 Å estimated by the least-squares diagonal approximation (eqn. 2). The r.m.s. difference in bond angles is 4′, maximum 7′, compared to the corresponding value of 4′ established from eqn. 2. Table 4 lists average bond lengths and angles corrected for curvilinear thermal vibrations.

TABLE 4.

Average bond lengths and bond angles after correction for curvilinear thermal vibrations.

| | | | | | | |
|---|---|---|---|---|---|---|
| 4-5 * | (C=C) | 1·344 Å | 1-3-4 | 121° 4′ | 2-4-5 | 123° 9′ |
| 3-4 | (C–C) | 1·500 | 1-3-5′ | 120° 21′ | 3-4-5 | 118° 51′ |
| 3-5′ | (C–C) | 1·474 | 4-3-5′ | 118° 35′ | 4-5-3′ | 122° 34′ |
| 2-4 | (C–Me) | 1·507 | 2-4-3 | 118° 0′ | | |
| 1-3 | (C=O) | 1·228 | | | | |

* Refer to diagram I for numbering of atoms.

The scatter of C–H bond lengths is much wider; if all C–H bonds are averaged (probably not a fully justified procedure) one obtains a value of 0·93 Å from which the individual values differ by 0·028 r.m.s., 0·060 Å maximum. This scatter is larger than estimated from eqn. 2: the diagonal approximation evidently underestimates the errors in the hydrogen co-ordinates.

Table 5 gives the equations of the best planes of the two molecules, in fractional co-ordinates, computed from the heavy-atom co-ordinates according to Schomaker *et al.*;[12] also listed are the distances, in Å, of all atoms from these planes. The exocyclic hydrogen atoms lie, in both molecules, virtually in the molecular plane; the exocyclic C–H bond (5–6) does not exactly

[12] Schomaker, Waser, Marsh, and Bergman, *Acta Cryst.*, 1959, **12**, 600.

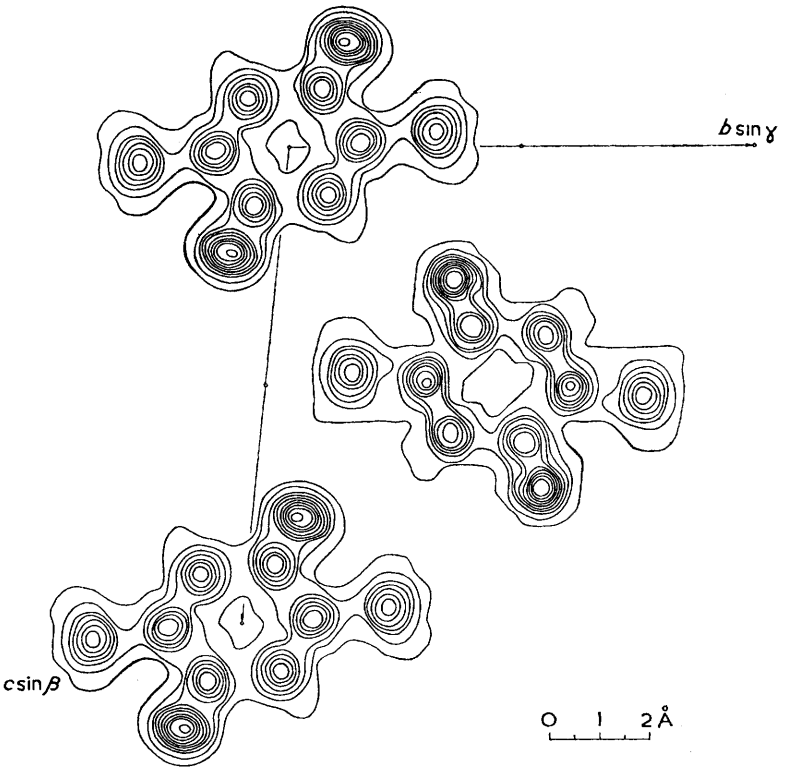



FIG. 1. 2,5-Dimethyl-1,4-benzoquinone; electron-density projection $\rho(yz)$. Contour interval 1 e/Å$^2$; lowest contour zero.

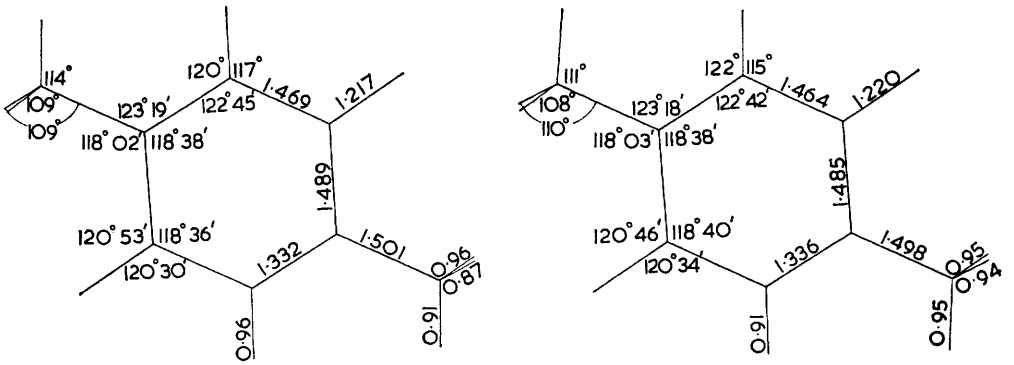



FIG. 2. 2,5-Dimethyl-1,4-benzoquinone; molecular dimensions, before correction for curvilinear thermal motion, of the two independent molecules; B stack, left, A stack, right.

bisect the opposed angle 3′–5–4, but the deviations, though similar in the two molecules, cannot be regarded as significant. The methyl groups are oriented so as to achieve maximum O • • • H separation, *i.e.*, 8-H lies in the molecular plane on the side away from the C=O group while 7-H and 9-H are symmetrically positioned above and below the plane and make equal contacts of about 2·9 Å with the oxygen atom.

TABLE 5.

Equations of best planes of the molecules (*xyz* in fractional co-ordinates); distances (in Å) of atoms from best planes.

| A molecule at (000) | | | | B molecule at $(\frac{1}{2}\frac{1}{2}\frac{1}{2})$ | | | |
|---|---|---|---|---|---|---|---|
| $3{\cdot}458x - 0{\cdot}2230y + 2{\cdot}9702z = 0$ | | | | $3{\cdot}4491x + 0{\cdot}1395y + 2{\cdot}8851z - 3{\cdot}2369 = 0$ | | | |
| O | 0·002 | H 6 | 0·023 | O | 0·006 | H 6 | 0·004 |
| C 2 | 0·002 | H 7 | −0·033 | C 2 | −0·002 | H 7 | −0·065 |
| C 3 | 0·000 | H 8 | 0·766 | C 3 | −0·008 | H 8 | −0·797 |
| C 4 | −0·001 | H 9 | −0·745 | C 4 | −0·001 | H 9 | 0·671 |
| C 5 | −0·003 | | | C 5 | 0·004 | | |

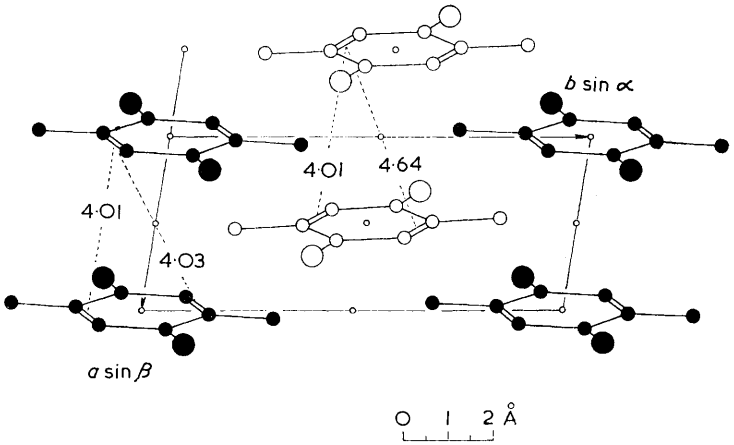



FIG. 3. 2,5-Dimethyl-1,4-benzoquinone; packing arrangement seen along [001].

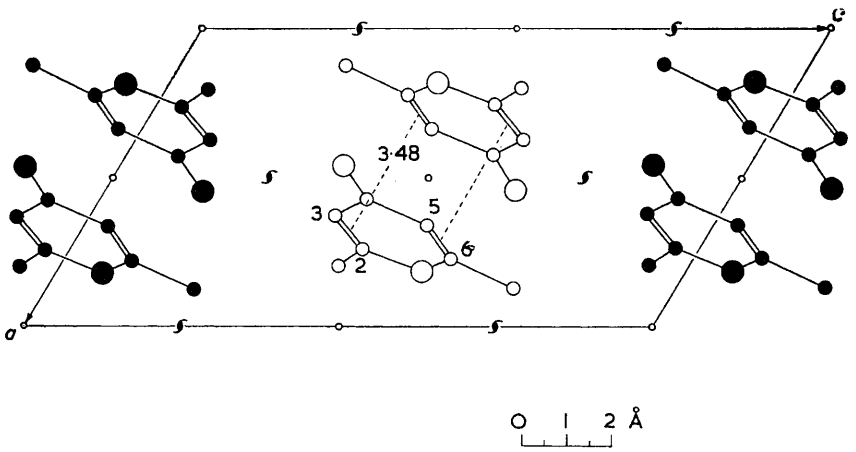



FIG. 4. 2,6-Dimethyl-γ-pyrone; packing arrangement seen along [010] (Brown, Norment, and Levy, *Acta Cryst.*, 1957, **10**, 806).

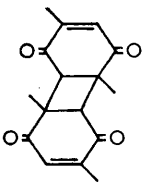
A detailed analysis of the molecular structure and of the thermal vibrations will be presented elsewhere; here we confine ourselves to a discussion of the molecular packing arrangement in so far as it relates to the interpretation of the photochemical behaviour of solid 2,5-dimethyl-1,4-benzoquinone.

The crystal structure consists of two kinds of molecular stacks, A and B, containing the molecules centred at (000) and $(\frac{1}{2}\frac{1}{2}\frac{1}{2})$, respectively; in both stacks molecules are related by the translation period of $a = 4{\cdot}013$ Å which produces parallel contact between >C=C< groups. A second set of (antiparallel) contacts between the >C=C< bonds of adjacent molecules within

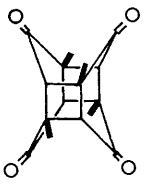
each stack is provided by the inversion centres at ($\frac{1}{2}$00) and (0$\frac{1}{2}\frac{1}{2}$), *i.e.*, midway between adjacent molecules within the two stacks. However, since the molecular orientation differs in the two stacks their centric contacts are of different lengths: **4·03** Å in stack A and **4·64** Å in stack B (Fig. 3).

We are now in a position to interpret Cookson's experimental results [1b] quoted above: the centric dimer (II) arises from the reaction across centric >C=C< contacts within a molecular stack; since it is formed in 40% yield we conclude that this reaction takes place only in the A stack at a >C=C< separation of **4·03** Å, and that molecules in the B stack whose double bonds are separated by **4·64** Å are incapable of reaction. The very low yield (2%) of the box dimer implies that double cyclisation in this crystal structure is an unlikely event; *if* it is lattice-controlled and occurs between monomers related by the *a* axis (**4·01** Å) it must give rise to the dimer of symmetry 2/*m* (IIIb) rather than to (IIIa) of symmetry 222.

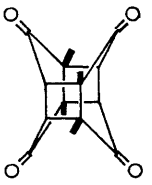
As regards reactivity and distance of reactive centres these conclusions are in accord with our results in the cinnamic acid series, namely that centric contact in the range **3·6**—**4·1** Å leads to irreversible formation of the centric dimer, and that at distances of **4·3** Å and more dimerisation no longer occurs. The structure of 2,5-dimethylbenzoquinone is further interesting in that it presents an example of *two* reaction paths, namely dimerisation to (II) and (IIIb) (possibly through the unsaturated *cis-syn*-dimer), which are equally probable from the purely geometrical point of view. The low yield of the latter is most likely due to unfavourable steric effects which bring the two pairs of methyl groups into opposition during the formation of the *cis-syn*-dimer, while in the centric dimer they are separated during dimerisation. In this connection a comparison with the crystal structure [13] and photochemistry [14] of 2,6-dimethyl-$\gamma$-pyrone (IV) is interesting. According to Levy *et al.* the pyrone (IV) crystallises as centrically related pairs of molecules (Fig. 4) such that non-equivalent (2,3 and 5′,6′) double bonds are separated by **3·48** Å while the centrically related bonds (2,3 and 2′,3′; 5,6 and 5′,6′) are **4·33** and **4·11** Å apart. All other centric >C=C< contacts are greater than **4·1** Å. Here, the alternative reaction paths are not equally probable geometrically, for the contact (**3·48** Å) leading to the box dimer (V) is significantly shorter than all centric contacts. The exclusive formation of (V) is not therefore surprising. Measurements of the quantum yield of this double-dimerisation reaction would be of considerable interest.



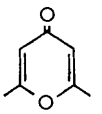
(II)



(IIIa)



(IIIb)



(IV)

(V)

The picture presented in this Paper must be regarded as preliminary. We are currently studying the crystal structures of 2,6-dimethyl-1,4-benzoquinone and of duroquinone; a discussion of the crystal chemistry of these and other quinones will be presented elsewhere.

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[13] Brown, Norment, and Levy, *Acta Cryst.*, 1957, **10**, 806.
[14] Yates and Jorgensen, *J. Amer. Chem. Soc.*, 1958, **80**, 6150.