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Crystallographic Studies of Iodine-Containing Amino Acids. I. Di-iodo-L-tyrosine Dihydrate

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Di-iodo-L-tyrosine dihydrate crystallizes in the monoclinic system with space group $P2_1$ and cell dimensions $a=11\cdot162\pm0\cdot014$, $b=11\cdot086\pm0\cdot023$, $c=5\cdot668\pm0\cdot014$ Å and $\beta=96°33'\pm15'$. The crystal structure has been determined by X-ray diffraction from three-dimensional intensity data recorded on Weissenberg films with molybdenum radiation and estimated visually. The structure (without hydrogen atoms) has been refined by the block diagonal least-squares method, the iodine atoms having anisotropic temperature factors, others isotropic, to an R index of 0·078. The iodine–iodine distance of 6·05 Å is shorter than expected. The di-iodotyrosine molecules are held together in the crystal by means of a net of extensive hydrogen bonding involving the carboxyl, amino, and phenolic groups of the molecule and the water molecules.

Introduction

This structure determination was undertaken to provide information for protein crystallography. The direct iodination of proteins is a possible means of introducing heavy atoms to be used as isomorphous replacements. Among the more interesting results of such iodination would be mono- and di-iodotyrosine, mono- and di-iodohistidine, and possibly iodinated cystine or cysteine residues. In particular, the intramolecular iodine–iodine vector in di-iodotyrosine and di-iodohistidine residues should be a prominent vector in the Patterson maps of iodinated proteins. To help in the identification of such vectors, accurate structure determinations should be available for di-iodotyrosine and di-iodohistidine. The subject of this paper, di-iodotyrosine, is also of interest because of its similarity to the thyroid hormone, thyroxine.

Experimental

Cystals were prepared by slow evaporation of di-iodotyrosine in strong aqueous ammonia solution. Small, pale yellow prisms were obtained which were kept in the solution until needed.

Crystal data

$C_9H_{19}NO_5I_2$ F.W. 471·090 .

Crystal system: monoclinic, space group $P2_1$ with b chosen as the unique axis.*

$a=11\cdot162\pm0\cdot014$ Å

$b=11\cdot086\pm0\cdot023$

$c=\ 5\cdot668\pm0\cdot014$

$\beta=96°33'\pm15'$

$V=697\pm4$ Å^3

$\varrho_{\text{calc}}=2\cdot243$ g.cm^{-3} with $Z=2$

$\varrho_{\text{obs}}=2\cdot20$ g.cm^{-3} (flotation using xylene and bromoform) .

Linear absorption coefficient for Mo radiation, 42·01 cm^{-1}.

Crystal sizes

0·35 × 0·15 × 0·02 mm.	Three-dimensional Cu data
0·20 × 0·15 × 0·15 mm.	Three-dimensional Mo data
0·35 × 0·17 × 0·15 mm.	Mo data retake levels $hk3$ and $hk4$
0·35 × 0·17 × 0·15 mm.	Mo data retake levels $hk5$ and $hk6$

Chemical analysis

	Theory (for dihydrate)	Found
Nitrogen	2·99%	2·96%
Oxygen	17·06%	16·46%

Chemical analysis showed the presence of two molecules of water of crystallization. The cell dimensions were measured from Weissenberg films taken with molybdenum radiation and calibrated with aluminum powder lines.

Structure analysis

Intensity data were estimated visually from equi-inclination Weissenberg films of crystals sealed in thin-walled glass capillary tubes and rotated about the c axis. The first three-dimensional data set was collected with Cu $K\alpha$ radiation; no absorption corrections were applied, and the copper data were processed to see how far the structure analysis could be completed. It proved possible to determine the iodine positions from a three-dimensional Patterson map, but the first Fourier map based on phases calculated from these positions proved uninterpretable. In space group $P2_1$ the origin along the screw axis is not defined by symmetry and hence it was fixed by holding constant the y coordinate of iodine atom I(1).

Meanwhile, with Mo $K\alpha$ radiation, data had been collected from Weissenberg films taken with a fresh

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* The choice of $P2_1$ over $P2/m$ was made after confirming by optical rotation measurements that the crystals were of the L-amino acid.

crystal, again rotating about the *c* axis. These data were corrected for Lorentz and polarization effects, but not for absorption, and structure factors were calculated with the iodine positions obtained from the copper data. Individual scale factors for the levels were found by comparing intensities of spots common to these levels and two intersecting zero level precession film series. The atomic scattering curve for iodine was taken from a paper by Cromer, Larson & Waber (1964) and those for the other elements from *International Tables for X-ray Crystallography* (1962). The overall *R* value was 24%. A first three-dimensional Fourier map was calculated and from it the position of the benzene ring was obtained, but with some difficulty. The positions of the other atoms could not be determined with any certainty. The ring atoms were included in the phasing calculations and a second three-dimensional Fourier synthesis was calculated. The phenolic oxygen was now placed, but the configuration of the amino and carboxyl atoms was still very uncertain. Least-squares refinement (block-diagonal) was used to improve the positions of the known atoms; during this refinement the temperature factors of the iodine atoms were allowed to vary anisotropically, the lighter atoms isotropically. The overall *R* value was 16% when the two iodine atoms, six benzene ring atoms, and the phenolic oxygen atom were included in this refinement. The block-diagonal least-squares program is a much modified version of the Sparks, Trueblood, Okaya program. Most of the changes were made by D. R. Harris and a few by ourselves (*World List of Crystallographic Computer Programs*, 1966).

Trial and error methods were used to find the atoms in the side chain, choosing the highest peaks to calculate structure factors and then trying to refine the positions by the least-squares method. After several cycles a three-dimensional difference map appeared to be quite flat except for two peaks which proved to be the water molecules, and for some rippling around the iodine atoms. At this stage, on examination of the scale factor between the observed and calculated structure factors for each of the initial individual film levels, it was found that one of them (the zero level) was not unity. This level was rescaled to the calculated values and on a subsequent three-dimensional difference map the rippling had disappeared. The *R* value at this point was 11%; however, the temperature factors of the atoms in the side chain still appeared to be unusually high. Bond lengths were calculated for the structure, and atom C(9) seemed to be in a wrong position. This atom was omitted from phasing calculations and the position redetermined from a difference Fourier synthesis. However, it reappeared in almost exactly the same position making the bond with C(6) abnormally long (1·72 Å). Also, the benzene ring bonds seemed quite varied in length. Changing the weighting factor in the least-squares program gave no significant improvement (until this stage the weights had been unity).

Some difficulty in indexing the upper-level films had been encountered during the data estimation. In order to check on the possibility of such errors, *R* was calculated for each film level. As shown in Table 1, this was increasingly alarming as the levels progressed. Decomposition of the crystal seemed the most likely cause of this behavior in spite of precautions taken, such as mounting the crystal in a sealed capillary tube with a little mother liquor. A retake of levels five and six with a fresh crystal gave greatly improved data. Yet another fresh crystal was used to re-collect levels three and four. The number of reflections thus obtained for levels three through six was approximately twice that given by the old crystal.

Three cycles of least-squares refinement using the old parameters and including the new data reduced *R* to 9·3%. The weighting scheme, $w=(F_o/12)^2$ if $F_o<12$, and $w=(12/F_o)^2$ if $F_o>12$, was introduced (12 being eight times the minimum value of F_o) and three more cycles of refinement reduced *R* to the final value of 7·8% (excluding unobserved reflections). The quantity minimized in all least-squares refinements was $\Sigma w(F_o-F_c)^2$.

All bond lengths for the structure were now reasonable. In the initial refinement of this structure no corrections were made for the anomalous dispersion effects of the iodine atoms. At the suggestion of the editor we investigated our indexing thoroughly and reassigned indices to some of the initial data levels in accordance with a systematic set of axes. Previously about half of the data had been indexed with respect to a right-handed and half to a left-handed axial set, possibly producing a cancellation of the anomalous scattering effects. The correctly indexed data were then refined further by three cycles of least squares using the block-diagonal program which has facilities for anomalous

Table 1. *R indices*

Level	*R* index	Number of unique reflections	*R* index after inclusion of new Mo data	Number of unique reflections after inclusion of new Mo data
*hk*0	9·7	132	4·7	132
*hk*1	10·1	216	4·9	216
*hk*2	13·1	185	4·9	185
*hk*3	13·6	186	11·0	360
*hk*4	17·2	148	13·0	326
*hk*5	21·5	143	16·2	286
*hk*6	15·6	105	15·0	257

dispersion corrections (*World List of Crystallographic Computer Programs*, 1966). The $\Delta f'$ and $\Delta f''$ terms were taken from *International Tables for X-ray Crystallography* (1962). The R value did not improve and only very slight changes in the atomic parameters were obtained, certainly within a maximum value of three times the standard deviation. We believe these results follow from the fact that the iodine atoms contribute such a large part to the total scattering of the molecule that consequently we have approximately a two atom structure. In this case we would expect anomalous scattering but no differences in the Friedel pairs. Unobserved reflections were only included in the final structure factor calculation, being assigned intensities equal to half of the minimum observed value.

The final atomic parameters listed in Table 2 are those from the refinement including anomalous dispersion corrections. Tables 3, 4 and 5 include bond lengths, angles and intermolecular distances calculated both before and after anomalous dispersion corrections, and Table 7 contains the final observed and calculated structure factors calculated from the atomic parameters in Table 2. However, data given on the figures and in the discussion refer to the parameters after the anomalous dispersion corrections. Fig. 1 contains the bond lengths and angles. A diagram of the three-dimensional Fourier synthesis projected on (010) is shown in Fig. 2 and a packing diagram of the molecules in four unit cells viewed down the b axis in Fig. 3.

Results of the analysis

Discussion of the structure

The average deviation of a carbon–carbon bond is 0·03 Å, a carbon–nitrogen bond 0·02 Å, and a carbon–iodine bond 0·02 Å, the deviations being calculated

Table 3. *Intramolecular bond lengths and angles and standard deviations*

Column A refers to the atomic parameters before anomalous dispersion corrections and column B to those after anomalous dispersion corrections.

Bond lengths	A	B	
I(1) —C(4)	2·00 Å	2·02 Å	0·02 Å
I(2) —C(8)	2·09	2·11	0·02
C(3)—C(4)	1·40	1·37	0·03
C(3)—C(8)	1·46	1·45	0·02
C(3)—O(13)	1·26	1·25	0·02
C(4)—C(5)	1·45	1·45	0·03
C(5)—C(6)	1·45	1·45	0·03
C(6)—C(7)	1·44	1·46	0·03
C(6)—C(9)	1·55	1·56	0·02
C(7)—C(8)	1·33	1·31	0·02
C(9)—C(10)	1·50	1·49	0·02
C(10)-N(12)	1·59	1·58	0·02
C(10)-C(11)	1·59	1·57	0·02
C(11)-O(14)	1·23	1·25	0·02
C(11)-O(15)	1·22	1·21	0·02
Angles	A	B	
C(4)—C(3)—O(13)	117·9°	118·7°	1·3°
C(4)—C(3)—C(8)	119·0	116·8	1·1
C(8)—C(3)—O(13)	122·6	124·2	1·6
I(1) —C(4)—C(3)	125·1	124·8	0·9
I(1) —C(4)—C(5)	117·7	115·9	1·3
C(3)—C(4)—C(5)	117·2	119·3	1·9
C(4)—C(5)—C(6)	123·5	122·7	2·2
C(5)—C(6)—C(7)	115·3	115·0	1·7
C(5)—C(6)—C(9)	122·8	123·7	1·8
C(7)—C(6)—C(9)	121·9	121·2	1·6
C(6)—C(7)—C(8)	121·6	120·5	1·7
I(2) —C(8)—C(3)	117·6	116·2	1·0
I(2) —C(8)—C(7)	118·8	118·1	0·8
C(3)—C(8)—C(7)	123·4	125·5	1·8
C(6)—C(9)—C(10)	106·7	107·2	0·9
C(9)—C(10)—C(11)	112·9	112·8	0·9
C(9)—C(10)—N(12)	102·7	102·9	1·4
C(11)-C(10)-N(12)	104·5	105·1	1·3
C(10)-C(11)-O(14)	115·4	119·0	1·6
C(10)-C(11)-O(15)	118·7	116·4	1·0
O(14)-C(11)-O(15)	124·8	123·0	1·2

Table 2. *Final atomic coordinates with standard deviations and temperature parameters*

These values were obtained after correction for anomalous dispersion.

The anisotropic temperature factor is $\exp\left[-(\beta_{11}h^2+\beta_{22}k^2+\beta_{33}l^2+2\beta_{12}hk+2\beta_{13}hl+2\beta_{23}kl)\right]$ and $B_{11}=2\pi^2a^{*2}U_{11}$ *etc.*
$B_{12}=2\pi^2a^*b^*U_{12}$ *etc.*

	x/a	$\sigma(x/a)$	y/b	$\sigma(y/b)$	z/c	$\sigma(z/c)$	B
I(1)	0·7735	0·0001	0·0000*		0·0425	0·0001	†
I(2)	0·4839	0·0001	0·3337	0·0003	0·6041	0·0002	†
C(3)	0·6132	0·0014	0·1644	0·0017	0·2929	0·0025	2·6
C(4)	0·6202	0·0017	0·0822	0·0018	0·1146	0·0032	3·0
C(5)	0·5107	0·0021	0·0450	0·0021	−0·0294	0·0040	3·7
C(6)	0·3927	0·0016	0·0913	0·0017	0·0069	0·0029	2·9
C(7)	0·3916	0·0014	0·1754	0·0017	0·2041	0·0026	2·8
C(8)	0·4924	0·0016	0·2061	0·0017	0·3317	0·0030	3·0
C(9)	0·2747	0·0015	0·0595	0·0016	−0·1557	0·0029	2·9
C(10)	0·2235	0·0014	−0·0508	0·0015	−0·0560	0·0026	2·4
C(11)	0·1767	0·0013	−0·0278	0·0014	0·1917	0·0025	2·3
N(12)	0·1063	0·0015	−0·0749	0·0018	−0·2335	0·0031	3·7
O(13)	0·7072	0·0016	0·1917	0·0017	0·4222	0·0030	4·8
O(14)	0·1958	0·0010	−0·1070	0·0010	0·3383	0·0018	2·5
O(15)	0·1041	0·0013	0·0562	0·0013	0·2112	0·0024	3·4
O(16)	0·0262	0·0012	0·2680	0·0013	−0·0076	0·0024	3·6
O(17)	0·9763	0·0014	0·1337	0·0016	0·5612	0·0025	4·1

† Thermal vibration tensor components (Å²) for the anisotropic atoms.

* Fixed to define origin.

	U_{11}	U_{22}	U_{33}	U_{12}	U_{13}	U_{23}
I(1)	0·0388	0·0905	0·0738	0·00019	0·0064	−0·0135
I(2)	0·0672	0·0416	0·0443	−0·0013	0·0062	−0·0047

Table 4. *The more interesting intramolecular and intermolecular distances* ($\leq 4{\cdot}0$ Å) *with standard deviations*

Column *A* refers to values obtained before, and column *B* to those obtained after, anomalous dispersion corrections.

i	x	y	z
ii	x	y	$1+z$
iii	$1-x$	$\frac{1}{2}+y$	$1-z$
iv	$x-1$	$y+1$	z
v	x	$1+y$	$1+z$
vi	$-x$	$\frac{1}{2}+y$	$1-z$
vii	$-x$	$y-\frac{1}{2}$	$1-z$
viii	$x-1$	y	z
ix	x	$1+y$	z
x	x	$y-1$	$1+z$
xi	x	$y-1$	z
xii	$-x$	$\frac{1}{2}+y$	$-z$

	A	*B*	
O(17^{viii})–O(15^{i})	2·72 Å	2·71 Å	0·02 Å
O(15^{vii})–O(16^{vii})	2·75	2·75	0·02
N(12^{ii})—O(16^{vii})	2·75	2·75	0·02
N(12^{ii})—O(14^{i})	2·75	2·75	0·02
O(17^{viii})–O(16^{ii})	2·85	2·86	0·02
O(17^{viii})–N(12^{ii})	2·88	2·90	0·03
N(12^{ii})—O(15^{ii})	2·91	2·91	0·02
O(13^{iii})—O(17^{iii})	3·11	3·08	0·02
N(12^{i})—O(14^{i})	3·32	3·30	0·02
O(15^{vi})—O(16^{ii})	3·65	3·64	0·02
O(17^{iii})—O(16^{vi})	3·65	3·65	0·02
O(14^{i})—O(17^{viii})	3·93	3·93	0·02

Table 5. *Bond angles and standard deviations associated with the hydrogen bonding atoms*

Column *A* refers to those before anomalous dispersion corrections and column *B* to those after anomalous dispersion corrections.

For notation, see Table 4.

	A	*B*	
Water molecule O(16)			
O(15^{ii})—O(16^{ii})—O(17^{viii})	88·0°	87·8°	0·4°
O(15^{vii})–O(16^{vii})–N(12^{ii})	119·2	118·5	0·5
O(17^{iii})—O(16^{xii})–N(12^{ix})	133·8	133·8	0·4
Phenol oxygen O(13)			
C(3^{i})—O(13^{i})—O(17^{i})	146·2	146·0	0·9
Water molecule O(17)			
O(13^{iii})—O(17^{iii})—O(16^{xii})	101·2	101·5	0·2
O(16^{ii})—O(17^{viii})–O(15^{i})	137·2	136·2	0·4
N(12^{ii})—O(17^{viii})–O(16^{ii})	92·4	91·9	0·4
N(12^{ii})—O(17^{viii})–O(15^{i})	76·2	76·1	0·5
O(13^{viii})–O(17^{viii})–N(12^{ii})	134·2	134·1	0·4
O(13^{viii})–O(17^{viii})–O(15^{i})	116·7	117·2	0·4
Amino nitrogen N(12)			
O(15^{ii})—N(12^{ii})—O(16^{vii})	80·2	80·1	0·7
O(16^{vii})–N(12^{ii})—O(17^{viii})	116·1	115·6	0·8
O(15^{ii})—N(12^{ii})—O(17^{viii})	84·1	84·1	0·8
O(17^{viii})–N(12^{ii})—O(14^{i})	88·7	88·0	0·5
O(14^{i})—N(12^{ii})—O(15^{ii})	150·5	150·2	0·5
O(16^{vii})–N(12^{ii})—C(10^{ii})	103·7	104·1	1·2
O(17^{viii})–N(12^{ii})—C(10^{ii})	117·4	117·4	1·1
C(10^{ii})—N(12^{ii})—O(14^{i})	102·9	103·1	1·2
O(16^{vii})–N(12^{ii})—O(14^{i})	128·1	128·8	0·8
Carboxyl oxygen O(14)			
C(11^{i})—O(14^{i})—N(12^{ii})	116·2	117·3	1·4
Carboxyl oxygen O(15)			
O(17^{viii})–O(15^{i})—O(16^{i})	83·7	83·9	0·6
O(17^{viii})–O(15^{i})—N(12^{i})	148·4	148·0	0·8
O(16^{i})—O(15^{i})—N(12^{i})	93·9	94·0	0·4
C(11^{i})—O(15^{i})—O(16^{i})	141·7	140·6	1·4
C(11^{i})—O(15^{i})—O(17^{viii})	133·6	134·6	1·3

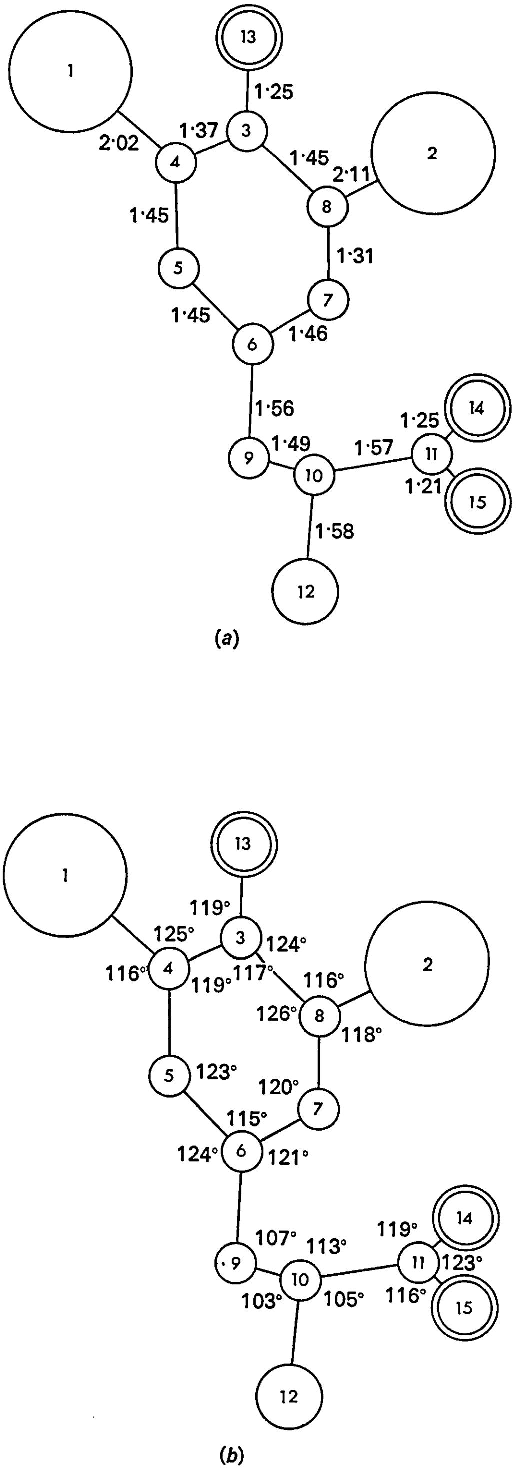

Fig. 1. Diagram of the molecule showing (*a*) bond lengths and (*b*) angles. The average deviations in bond lengths are: carbon–carbon 0·03, carbon–oxygen 0·02, carbon–nitrogen 0·02, carbon–iodine 0·02 Å. These bond lengths and angles were calculated with the atomic parameters after anomalous dispersion corrections. Large circles: I; medium circles: N; small circles: C: double circles: O.

from the full matrix least-squares totals with the use of the Busing & Levy (1962) function and error program. We would like to emphasize that we feel these deviations are underestimated by this method and should perhaps be 1·5 times the values quoted. The average carbon–carbon bond in the benzene ring is 1·42 ± 0·03 Å which compares with that of 1·39 ± 0·03 Å for tyrosine hydroiodide (Seely, 1966). Considering the *p*H at which the di-iodotyrosine was crystallized it seems reasonable to expect the carboxyl and amino groups to be charged. This is consistent with the carbon–oxygen bond lengths of 1·21 ± 0·02 Å and 1·25 ± 0·02 Å for di-iodotyrosine compared to 1·15 ± 0·03 Å in tyrosine hydroiodide which has an uncharged carboxyl group. The carbon–nitrogen bond length of 1·58 ± 0·02 Å compares reasonably with that of 1·55 ± 0·02 Å in tyrosine hydroiodide and is possibly significantly longer than the normal 1·48 Å quoted for a carbon–nitrogen bond (*Interatomic Distances and Configuration in Molecules and Ions*, 1958). The carbon–oxygen bond of the phenol group is 1·25 ± 0·02 Å, which is shorter than that of 1·41 ± 0·02 Å in tyrosine hydroiodide.

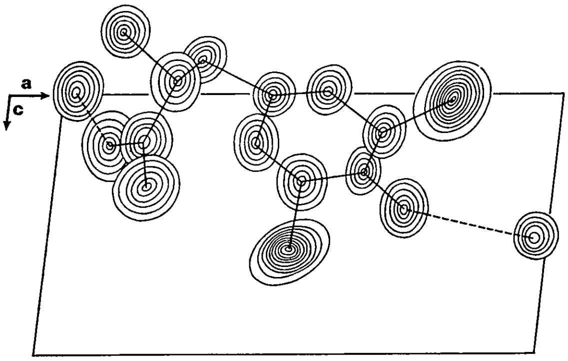

Fig. 2. A composite of the final three-dimensional electron density map viewed along the *b* axis. The contour levels are spaced every 1 e.Å^{-3} beginning with the 2 e.Å^{-3} level. The iodine contours are every 5 e.Å^{-3} beginning with the 2 e.Å^{-3} level. For the molecule to have the L configuration the *b* axis must be assumed to be into the page.

We believe the short bond length of the phenol group to be an artifact of our data rather than a consequence of any change in the benzene ring due to the iodine

Table 6. *Mean plane calculations through the atoms of the benzene ring*

The equation of the best plane through atoms C(3), C(4), C(5), C(6), C(7), C(8) is:

$$0{\cdot}1771X + 0{\cdot}7571Y - 0{\cdot}6289Z + 1{\cdot}5033 = 0$$

	Deviations from the plane
I(1)	−0·13 Å
I(2)	0·05
C(3)	0·02
C(4)	−0·01
C(5)	−0·01
C(6)	0·01
C(7)	−0·01
C(8)	−0·01
C(9)	0·11
O(13)	−0·04

These results were obtained with the atomic parameters after anomalous dispersion corrections.

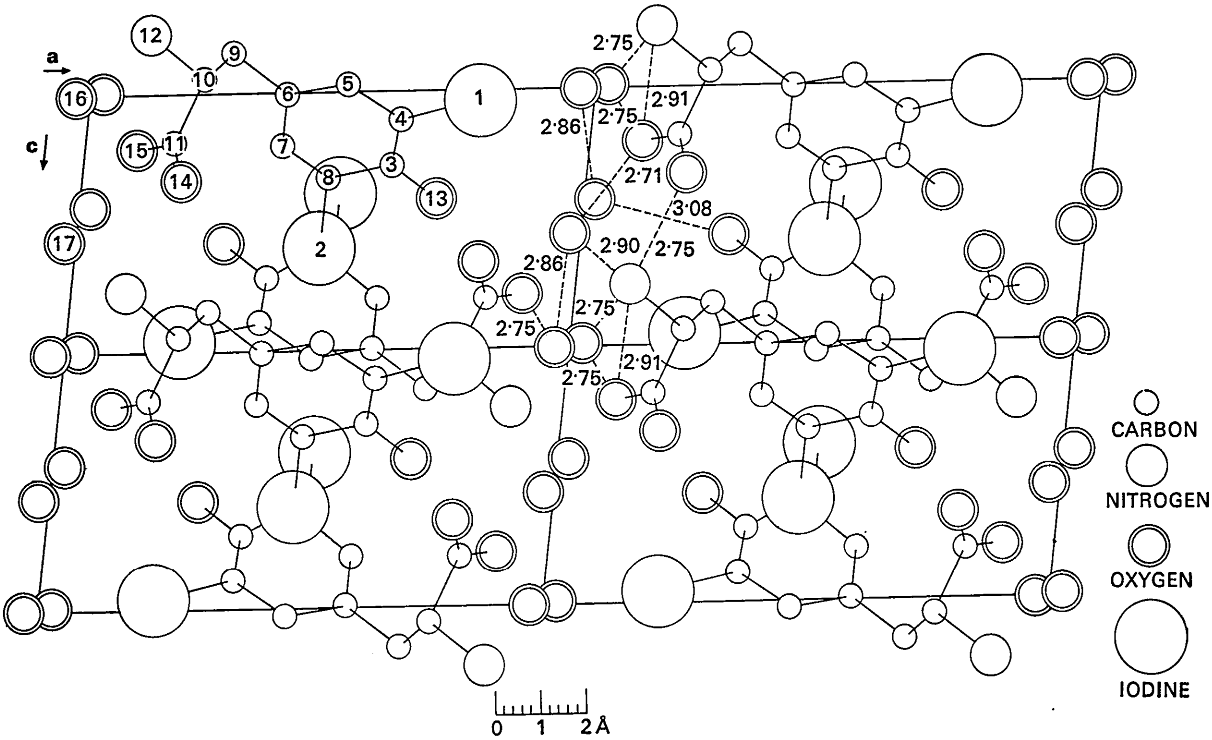

Fig. 3. Projection of the structure viewed down the *b* axis, showing those shortest intermolecular distances which are of interest. Four unit cells are shown. These distances were calculated with the atomic parameters after anomalous dispersion corrections.

Table 7. *The final observed and calculated structure factors obtained with the atomic parameters in Table* 2

F_o, F_c, and α(°) are multiplied by 10. < denotes unobserved reflections which were assigned an intensity value half of the minimum observed intensity and were included only in the calculation of the final cycle of structure factors.

* Denotes reflections cut off by the beam stop. Intensity values were assigned as in the case of the unobserved reflections.

h	K	F_o	F_c	α
Level ℓ = 0				
0	2	791	740	3355
0	4	567	528	735
0	6	916	879	96
0	8	325	319	2566
0	10	150	163	1140
0	12	254	249	25
1	0	914	838	1824
1	1	596	572	2889
1	2	811	782	352
1	3	490	461	2909
1	4	833	792	3117
1	5	794	738	2743
1	6	709	654	1991
1	7	448	438	2685
1	8	575	569	553
1	9	183	180	2716
1	10	249	245	3039
1	11	141	143	2995
1	12	197	211	1911
2	0	104	119	1
2	1	376	299	498
2	2	1595	1502	1997
2	3	339	337	3077
2	4	1181	1180	1434
2	5	197	202	2318
2	6	117	123	3406
2	7	128	112	2849
2	8	466	473	2217
2	9	194	172	2737
2	10	396	396	1396
2	11	< 72	17	2621
2	12	124	132	176
3	0	1181	1121	1327
3	1	747	702	1237
3	2	1051	1049	1077
3	3	838	909	890
3	4	449	450	2706
3	5	425	425	807
3	6	660	632	1042
3	7	224	206	1354
3	8	367	357	771
3	9	225	234	940
3	10	276	251	2886
3	11	< 73	105	805
3	12	127	139	1720
4	0	1325	1324	26
4	1	506	458	651
4	2	634	600	2846
4	3	301	292	1170
4	4	549	522	569
4	5	464	467	1417
4	6	745	734	37
4	7	280	256	631
4	8	275	276	2926
4	9	114	101	2454
4	10	219	237	1015
4	11	115	106	1423
4	12	106	111	91
5	0	< 45	51	3559
5	1	769	734	2493
5	2	718	735	360
5	3	141	112	2994
5	4	590	591	3157
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5	11	106	115	3234
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6	3	701	699	2913
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6	5	246	243	2778
6	6	128	122	3585
6	7	154	160	3412
6	8	216	237	2372
6	9	221	211	2965
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7	1	213	202	1772
7	2	462	459	1393
7	3	492	486	785
7	4	249	271	2318
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7	6	392	414	1879
7	7	132	130	1428
7	8	147	159	1085
7	9	213	214	910
7	10	120	110	2607
7	11	114	116	139
7	12	87	104	1936
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8	1	455	475	717
8	2	65	69	3291
8	3	185	194	1094
8	4	205	214	868
8	5	415	424	1287
8	6	192	221	67
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8	11	116	115	1195
8	12	82	94	138
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9	4	332	360	3449
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9	6	< 70	50	3198
9	7	108	139	2357
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10	8	95	89	2019
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11	7	87	110	2286
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12	2	< 76	67	2263
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12	4	< 79	33	1145
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12	6	20	21	3493
12	7	101	126	872
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13	2	142	158	223
13	3	105	122	889
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-10	-2	< 73	119	2284
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-9	-4	298	306	199
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-9	-2	229	229	85
-9	-1	166	172	85
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-3	-2	534	560	1485
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0	8	463	414	443
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1	8	351	351	2519
1	9	< 69	71	3268
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1	13	123	114	2453
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2	1	335	351	3096
2	2	729	693	1473
2	3	542	524	2568
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2	5	372	339	1947
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2	9	309	306	2645
2	10	204	208	2958
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2	12	171	177	1682
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3	4	876	836	1476
3	5	244	223	494
3	6	< 58	56	49
3	7	249	269	1265
3	8	331	334	2353
3	9	204	209	1065
3	10	217	219	1292
3	11	< 80	79	648
3	12	179	179	127
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4	2	779	766	219
4	3	355	313	373
4	4	894	868	3290
4	5	377	380	1094
4	6	214	211	1730
4	7	236	233	751
4	8	469	443	560
4	9	139	144	1125
4	10	247	248	3132
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4	12	117	132	1884
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5	2	456	481	2925
5	3	310	285	3057
5	4	411	400	578
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5	10	< 79	94	965
5	11	< 83	81	3419
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6	2	381	391	932
6	3	357	385	2903
6	4	274	283	3088
6	5	326	302	2829
6	6	390	394	1897
6	7	210	222	3039
6	8	205	195	802
6	9	116	120	2589
6	10	163	162	2778
6	11	< 85	58	2514
6	12	127	125	1876
7	0	230	227	1811
7	1	< 60	46	1377
7	2	733	717	2071
7	3	< 63	112	751
7	4	559	564	1428
7	5	< 67	98	937
7	6	< 70	76	659
7	7	162	148	703
7	8	230	264	2182
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8	3	281	288	1117
8	4	154	173	3351
8	5	257	264	944
8	6	260	265	1561
8	7	170	151	1100
8	8	166	182	762
9	0	529	556	37
9	1	< 69	82	3573
9	2	285	314	3101
9	3	< 72	14	1754
9	4	190	186	369
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10	3	185	205	2776
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11	1	< 79	21	3538
11	2	229	263	2026
11	3	< 81	108	2770
11	4	171	212	1619
12	0	142	179	1842
12	1	142	142	1215
12	2	< 84	90	1303
12	3	196	215	968
13	0	121	151	54
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-11	-4	215	218	2005
-11	-3	194	192	2788
-11	-2	308	347	1719
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-11	0	165	190	1825
-10	-6	213	246	1973
-10	-5	< 69	127	1434
-10	-4	65	83	1984
-10	-3	310	341	747
-10	-2	< 65	178	2444
-10	-1	146	174	500
-10	0	352	376	1835
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-9	-6	275	295	9
-9	-5	301	300	656
-9	-4	134	141	3369
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-9	-2	217	226	594
-9	-1	349	357	1181
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-8	-8	178	190	3579
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-8	-6	< 64	78	504
-8	-5	360	383	2264
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-8	-3	< 58	25	2434
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-8	-1	389	416	3021
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2	10	145	159	1690
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3	10	< 68	92	3197
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4	6	390	380	10
4	7	281	275	1291
4	8	< 62	43	2551
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5	2	716	681	277
5	3	424	393	2829
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5	9	211	215	2911
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6	4	289	301	1475
6	5	411	414	2961
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6	8	120	134	2361
6	9	85	62	2776
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7	2	437	416	1449
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7	6	325	329	1813
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9	2	299	332	427
9	3	< 65	46	3412
9	4	329	339	3310
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11	2	128	134	1518
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-16	-2	< 75	25	1238
-16	-1	116	93	2887
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-15	-5	57	56	3357
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-15	-2	79	66	2186
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-14	0	< 68	22	47
-13	-12	63	10	2362
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-13	-6	< 69	22	448
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-13	-3	72	66	871
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-12	-8	76	86	1007
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-12	-5	132	103	2618
-12	-4	69	43	3051
-12	-3	83	100	2811
-12	-2	67	97	751
-12	-1	133	130	2738
-12	0	270	213	46
-11	-12	59	51	1744
-11	-11	57	35	3009
-11	-10	55	43	904
-11	-9	76	81	3006
-11	-8	90	92	2883
-11	-7	112	102	3077
-11	-6	218	176	1701
-11	-5	48	65	2667
-11	-4	180	188	1265
-11	-3	91	127	2888
-11	-2	78	115	2169
-11	-1	141	145	2799
-11	0	326	357	1838
-10	-11	55	72	400
-10	-10	107	93	2456
-10	-9	73	84	840
-10	-8	< 64	124	1489
-10	-7	48	58	829
-10	-6	< 60	43	2808
-10	-5	78	101	587
-10	-4	288	303	2140
-10	-3	61	80	897
-10	-2	288	296	1611
-10	-1	156	145	1192
-10	0	55	44	3590
-9	-12	56	57	1891
-9	-11	76	65	1388
-9	-10	73	94	562
-9	-9	122	113	870
-9	-8	185	179	3144
-9	-7	92	112	575
-9	-6	44	49	2090
-9	-5	148	170	1259
-9	-4	246	264	390
-9	-3	252	242	1024
-9	-2	342	331	3243
-9	-1	211	214	974
-9	0	72	57	1857
-8	-12	134	85	101
-8	-11	< 68	13	2849
-8	-10	71	89	2619
-8	-9	83	98	2241
-8	-8	177	178	816
-8	-7	88	111	2002
-8	-6	396	392	147
-8	-5	< 52	37	2544
-8	-4	232	240	3143
-8	-3	181	195	2492
-8	-2	247	237	523
-8	-1	180	185	2015
-8	0	510	575	38
-7	-14	63	49	3114
-7	-13	79	71	3114
-7	-12	75	82	1803
-7	-11	88	86	2587
-7	-10	168	150	836
-7	-9	< 59	23	2201
-7	-8	98	111	3153
-7	-7	250	251	2810
-7	-6	264	274	1663
-7	-5	260	262	2445
-7	-4	213	202	795
-7	-3	100	114	2985
-7	-2	296	282	2855
-7	-1	443	434	2856
-7	0	396	448	1833
-6	-16	62	37	2547
-6	-15	59	38	2628
-6	-14	57	86	1262
-6	-13	< 70	43	897
-6	-12	< 67	40	402
-6	-11	99	106	175
-6	-10	200	184	2267
-6	-9	100	106	2760
-6	-8	275	268	1658
-6	-7	120	124	976
-6	-6	132	121	2811
-6	-5	238	280	592
-6	-4	487	536	2168
-6	-3	224	223	2814
-6	-2	558	652	1670
-6	-1	188	171	1755
-6	0	< 39	38	3543
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-5	-14	56	71	3025
-5	-13	< 69	28	111
-5	-12	72	79	1968
-5	-11	108	103	1705
-5	-10	80	119	671
-5	-9	258	275	905
-5	-8	244	283	2988
-5	-7	150	169	109
-5	-6	334	353	1972
-5	-5	118	112	1019
-5	-4	129	128	745
-5	-3	734	747	759
-5	-2	363	383	3037
-5	-1	282	284	626
-5	0	258	275	1848
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-4	-14	55	58	1164
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-4	-12	112	114	42
-4	-11	67	82	571
-4	-10	142	139	2820
-4	-9	127	149	2353
-4	-8	166	151	808
-4	-7	185	184	1401
-4	-6	516	465	87
-4	-5	289	302	420
-4	-4	319	359	3230
-4	-3	257	309	2623
-4	-2	466	466	562
-4	-1	468	455	1458
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-3	-16	60	51	894
-3	-15	57	29	875
-3	-14	55	61	3268
-3	-13	124	123	3044
-3	-12	49	63	2033
-3	-11	217	189	2247
-3	-10	182	176	787
-3	-9	42	46	1701
-3	-8	207	198	3409
-3	-7	382	470	3062
-3	-6	191	234	1239
-3	-5	461	481	2312
-3	-4	480	540	555
-3	-3	78	69	1671
-3	-2	390	361	3332
-3	-1	638	658	3003
-3	0	127	164	1836
-2	-16	60	37	2488
-2	-15	81	68	2793
-2	-14	94	76	1265
-2	-13	52	81	1359
-2	-12	< 63	47	150
-2	-11	180	177	75
-2	-10	169	134	2251
-2	-9	238	263	2674
-2	-8	321	347	1522
-2	-7	267	294	1871
-2	-6	57	69	1414
-2	-5	350	417	3593
-2	-4	403	422	2151
-2	-3	398	453	2866
-2	-2	416	697	1664
-2	-1	* 24	532	1792
-2	0	* 23	38	3506
-1	-15	99	75	1072
-1	-14	54	52	3048
-1	-13	89	100	3497
-1	-12	69	87	2272
-1	-11	139	136	1901
-1	-10	< 56	63	1129
-1	-9	255	257	807
-1	-8	242	256	2931
-1	-7	305	369	13
-1	-6	339	377	1892
-1	-5	344	389	1514
-1	-4	190	219	1419
-1	-3	824	854	903
-1	-2	206	257	1968
-1	-1	* 22	457	164
-1	0	* 20	808	1823
0	0	* 20	647	27
0	1	* 22	745	469
0	2	218	238	2884
0	3	255	252	3058
0	4	245	243	489
0	5	452	517	1340
0	6	271	284	123
0	7	374	410	452
0	8	66	75	0
0	9	100	121	3164
0	10	123	151	579
0	11	210	224	1373
0	12	69	87	170
0	13	155	143	535
0	14	< 70	30	1739
0	15	81	52	2904
1	0	* 24	269	26
1	1	* 25	678	2455
1	2	* 28	375	108
1	3	307	271	492
1	4	399	435	3386
1	5	535	604	3192
1	6	88	78	3510
1	7	346	385	2350
1	8	168	189	70
1	9	180	209	926
1	10	176	164	2969
1	11	192	168	3411
1	12	< 63	32	934
1	13	137	131	2236
1	14	< 70	37	180
1	15	57	55	1173
1	16	< 77	20	3254
1	17	62	44	3419
2	0	< 29	59	1859
2	1	548	563	3410
2	2	283	256	1889
2	3	792	803	2874
2	4	142	137	882
2	5	343	332	1963
2	6	90	87	2537
2	7	199	204	105
2	8	157	175	2158
2	9	348	346	2686
2	10	63	76	1947
2	11	156	143	1792
2	12	< 64	62	2562

Table 7 (*cont.*)

h	K	F_o	F_c	α
2	13	52	67	322
2	14	55	52	2302
2	15	100	84	2713
2	16	< 78	8	1368
2	17	63	49	1531
3	0	554	552	1826
3	1	484	461	1956
3	2	327	340	1748
3	3	596	641	1046
3	4	236	233	1546
3	5	188	168	3363
3	6	255	245	1819
3	7	260	267	2177
3	8	57	56	1896
3	9	233	246	915
3	10	120	129	2153
3	11	126	108	3403
3	12	< 65	40	1280
3	13	130	102	2140
3	14	< 72	5	2907
3	15	116	76	1074
3	16	< 78	34	2227
3	17	63	40	3198
4	0	160	165	12
4	1	499	512	647
4	2	30	52	1001
4	3	131	84	2869
4	4	160	176	276
4	5	505	571	1173
4	6	128	122	849
4	7	441	423	632
4	8	83	100	1191
4	9	116	124	2882
4	10	65	58	370
4	11	169	154	1306
4	12	< 66	10	1721
4	13	143	96	474
4	14	< 73	20	1788
4	15	83	64	2962
5	0	391	432	30
5	1	368	346	2211
5	2	327	332	3416
5	3	392	371	917
5	4	188	185	126
5	5	279	250	3275
5	6	160	134	233
5	7	173	148	2275
5	8	136	123	87
5	9	169	165	800
5	10	< 61	26	3203
5	11	187	147	3391
5	12	91	64	296
5	13	55	63	2338
5	14	< 74	25	45
5	15	60	50	903
6	0	< 44	27	3562
6	1	289	245	3491
6	2	70	67	3419
6	3	487	460	2722
6	4	118	122	3340
6	5	295	309	2137
6	6	58	90	1576
6	7	186	167	3349
6	8	< 59	42	1481
6	9	244	226	2750
6	10	< 63	52	3260
6	11	89	60	1785
6	12	< 69	30	2453
6	13	56	59	72
6	14	< 75	5	1352
6	15	61	45	2671
7	0	260	267	1825
7	1	316	283	2123
7	2	334	316	2076
7	3	145	101	699
7	4	217	201	1819
7	5	193	155	3191
7	6	74	81	2154
7	7	238	248	2058
7	8	66	83	2304
7	9	97	71	1407
7	10	88	72	1771
7	11	91	71	3193
7	12	< 71	38	572
7	13	81	72	2200
7	14	< 77	16	2643
7	15	62	36	944
8	0	144	146	1845
8	1	325	283	633
8	2	115	124	1319
8	3	41	63	1245
8	4	42	60	3082
8	5	247	223	1437
8	6	119	78	1749
8	7	210	185	474
8	8	68	64	1343
8	9	50	23	2090
8	10	< 68	27	3526
8	11	122	105	1355
8	12	80	28	1674
8	13	59	52	560
9	0	200	185	50
9	1	148	137	1761
9	2	167	165	3240
9	3	235	193	808
9	4	109	106	376
9	5	121	136	199
9	6	142	119	92
9	7	69	48	1433
9	8	50	60	3235
9	9	104	71	814
9	10	< 70	11	1051
9	11	79	49	3598
10	0	< 58	45	30
10	1	111	85	3338
10	2	129	110	147
10	3	210	164	2895
10	4	134	122	3298
10	5	48	41	2370
10	6	< 64	12	2623
10	7	114	87	3509
10	8	53	53	353
10	9	108	92	2758
10	10	< 72	54	3363
10	11	58	14	1853
11	0	48	70	1817
11	1	144	107	2453
11	2	145	138	2121
11	3	< 63	35	3208
11	4	122	102	1701
11	5	124	122	2868
11	6	< 67	8	887
11	7	75	73	2544
11	8	55	46	2372
11	9	< 73	21	850
11	10	58	21	1804
12	0	168	135	1846
12	1	72	49	629
12	2	114	92	1339
12	3	< 67	27	3025
12	4	< 68	25	1581
12	5	75	58	1193
12	6	94	79	1606
12	7	79	70	1006
12	8	57	44	1415
12	9	58	41	1941
13	0	53	51	65
13	1	54	55	1402
13	2	54	35	3433
13	3	122	85	1082
13	4	< 71	52	754
13	5	56	27	1253
13	6	57	48	103
13	7	58	24	1040
13	8	< 76	16	2723
13	9	86	47	837
14	0	56	53	33
14	1	< 73	7	2244
14	2	113	81	129
14	3	< 73	27	2450
14	4	100	78	3386
15	0	< 76	16	41
15	1	59	44	2595
15	2	< 76	34	2652
15	3	< 77	33	3036
15	4	< 78	26	1922
15	5	61	32	3133

Level ℓ = 4

h	K	F_o	F_c	α
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-17	-6	< 64	36	3513
-17	-5	< 63	22	2104
-17	-4	< 63	37	195
-17	-3	< 62	23	1189
-17	-2	49	55	147
-17	-1	< 62	9	3210
-17	0	109	75	47
-16	-3	106	83	2610
-16	-2	< 59	46	173
-16	-1	< 59	50	2412
-16	0	< 59	5	1742
-15	-8	49	52	1508
-15	-7	48	55	3146
-15	-6	< 60	11	1987
-15	-5	46	49	2816
-15	-4	79	66	1893
-15	-3	45	31	348
-15	-2	90	90	1906
-15	-1	45	36	3402
-15	0	< 56	45	1965
-14	-7	65	63	1088
-14	-6	45	47	2032
-14	-5	133	104	741
-14	-4	< 55	19	1997
-14	-3	< 55	39	773
-14	-2	< 54	47	2166
-14	-1	128	91	1179
-14	0	170	150	1831
-13	-9	47	54	1154
-13	-8	< 58	25	397
-13	-7	< 56	37	817
-13	-6	97	90	3459
-13	-5	42	69	1433
-13	-4	72	57	3154
-13	-3	116	115	899
-13	-2	41	66	426
-13	-1	40	46	435
-13	0	140	122	57
-12	-10	65	67	522
-12	-9	45	54	2429
-12	-8	62	54	3302
-12	-7	60	77	2206
-12	-6	< 52	15	990
-12	-5	< 51	36	3013
-12	-4	168	128	471
-12	-3	165	151	2718
-12	-2	164	150	3509
-12	-1	66	86	2355
-12	0	< 48	48	32
-11	-11	46	58	2623
-11	-10	< 56	44	2344
-11	-9	< 55	31	3392
-11	-8	119	106	1223
-11	-7	108	121	3235
-11	-6	< 50	23	452
-11	-5	122	132	2601
-11	-4	99	97	2002
-11	-3	37	61	2744
-11	-2	195	193	1341
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-11	0	< 45	14	1741
-10	-15	51	26	3295
-10	-14	< 63	14	2654
-10	-13	< 61	19	1044
-10	-12	47	40	1941
-10	-11	45	62	528
-10	-10	< 55	54	1035
-10	-9	< 53	40	462
-10	-8	121	95	2487
-10	-6	172	177	1837
-10	-5	103	92	523
-10	-4	101	106	1328
-10	-3	< 44	42	3197
-10	-2	158	162	2452
-10	-1	107	123	1326
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-9	-14	< 62	33	902
-9	-13	< 60	21	538
-9	-12	45	55	3468
-9	-11	< 55	21	1426
-9	-10	103	101	2490
-9	-9	107	111	1068
-9	-8	110	94	1315
-9	-7	64	106	712
-9	-6	139	154	3392
-9	-5	159	163	1252
-9	-4	175	197	2763
-9	-3	213	237	967
-9	-2	137	125	938
-9	-1	139	160	778
-9	0	209	227	43
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-8	-13	< 58	45	1699
-8	-12	< 56	42	1818
-8	-11	< 54	12	1875
-8	-10	115	121	471
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-8	-8	166	162	3204
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-7	-5	253	268	2673
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-7	-3	141	152	2994
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-6	-14	46	41	2913
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-6	-10	102	107	887
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-5	-15	< 60	14	1598
-5	-14	45	65	1355
-5	-13	< 55	54	892
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-5	-11	39	27	839
-5	-10	155	146	2386
-5	-9	71	92	946
-5	-8	239	205	1398
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-5	-5	137	142	592
-5	-4	359	365	2445
-5	-3	167	218	641
-5	-2	364	384	1552
-5	-1	375	352	900
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-4	-15	< 60	19	964
-4	-14	64	83	3091
-4	-13	< 54	40	1979
-4	-12	71	84	2039
-4	-11	67	68	1600
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-4	-9	60	90	1197
-4	-8	304	270	3241
-4	-7	30	60	1670
-4	-6	49	81	1682
-4	-5	166	161	1306
-4	-4	504	540	376
-4	-3	158	161	676
-4	-2	447	544	3269
-4	-1	112	118	792
-4	0	139	142	1829
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-3	-5	98	106	2542
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0	0	* 17	136	1856
0	1	* 18	216	1421
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0	3	398	338	964
0	4	363	364	3284
0	5	198	197	202
0	6	171	189	1749
0	7	92	97	1551
0	8	275	295	606
0	9	243	238	1057
0	10	113	113	3075
0	11	107	106	342
0	12	70	89	1450
0	13	< 53	5	2329
0	14	99	80	606
0	15	< 59	54	1253
0	16	49	48	2921
1	0	* 20	685	31
1	1	* 21	200	191
1	2	310	299	2965
1	3	428	339	2824
1	4	304	268	537
1	5	87	84	2142
1	6	439	394	3455
1	7	185	190	217
1	8	193	185	2659
1	9	165	152	2999
1	10	108	100	1135
1	11	76	76	1758
1	12	151	121	3589
1	13	60	54	602
1	14	63	55	2589
2	0	254	209	1858
2	1	467	452	2396
2	2	301	255	590
2	3	63	55	3370
2	4	254	223	3075
2	5	395	333	3301
2	6	288	283	1894
2	7	283	246	2523
2	8	102	92	490
2	9	< 44	57	1796
2	10	146	154	2729
2	11	116	105	3005
2	12	129	113	1927
2	13	105	82	2508
2	14	45	32	71
3	0	57	76	3585
3	1	336	333	623
3	2	472	467	1974
3	3	172	174	2290
3	4	389	361	1560
3	5	261	257	1819
3	6	59	41	315
3	7	138	149	416
3	8	162	163	1989
3	9	149	156	2606
3	10	105	86	1447
3	11	88	82	1801
3	12	< 52	56	3341
3	13	61	61	672
3	14	64	69	2157
4	0	303	256	1830
4	1	276	268	1527
4	2	175	143	761
4	3	394	379	935
4	4	67	38	11
4	5	177	153	360
4	6	165	191	1581
4	7	137	158	1761
4	8	171	192	521
4	9	208	199	1045
4	10	< 48	34	3099
4	11	57	86	152
4	12	73	69	1560
4	13	44	40	2572
5	0	342	313	41
5	1	218	225	450
5	2	102	80	3116
5	3	206	197	2847
5	4	160	150	416
5	5	171	151	1569
5	6	223	224	3482
5	7	181	190	569
5	8	71	65	3086
5	9	99	77	2853
5	10	68	77	656
5	11	41	52	1399
5	12	61	64	141
5	13	63	63	408
6	1	365	321	2329
6	2	146	142	501
6	3	30	27	1678
6	4	155	168	3128
6	5	209	224	3059
6	6	< 43	50	2837
6	7	160	183	2535
6	8	37	48	94
6	9	< 49	39	731
6	10	57	78	2603
6	11	84	75	3295
6	12	< 56	25	1543
6	13	65	61	2365
7	0	76	109	1831
7	1	131	127	3481
7	2	176	184	2123
7	3	176	168	2672
7	4	124	109	1659
7	5	137	134	1767
7	6	100	105	2405
7	7	104	103	377
7	8	77	79	2415
7	9	150	141	2593
7	10	< 53	28	1133
7	11	75	70	2020
8	0	81	63	1879
8	1	159	169	1714
8	2	48	82	1607
8	3	195	185	977
8	4	111	104	2088
8	5	102	95	306
8	6	74	61	1515
8	7	54	56	1719
8	8	56	65	785
8	9	131	105	922
8	10	< 54	20	2181
8	11	45	37	264
9	0	133	117	34
9	1	170	147	522
9	2	36	56	3527
9	3	36	53	3344
9	4	37	48	384
9	5	121	97	1268
9	6	39	30	320
9	7	156	132	578
9	8	< 53	29	697
9	9	< 54	16	3330
9	10	< 56	43	503
9	11	65	51	1146
10	0	75	105	33
10	1	113	99	2407
10	2	101	97	122
10	3	86	53	234
10	4	39	48	85
10	5	139	102	3162
10	6	< 52	19	147
10	7	59	59	2371
10	8	43	23	3460
10	9	< 56	30	1037
10	10	< 58	5	1820
10	11	48	44	3323
11	0	56	44	1842
11	1	56	43	3449
11	2	40	48	1630
11	3	122	102	2694
11	4	41	33	2359
11	5	126	77	2280
11	6	< 54	40	2731
11	7	44	44	3712
11	9	80	68	2680
12	0	73	53	1849
12	1	59	55	2076
12	2	< 54	39	1796
12	3	61	51	836
12	4	75	52	1442
12	5	< 56	22	403
12	6	< 57	37	1623
12	7	46	38	1962
13	0	63	22	0
13	1	88	59	770
13	2	< 56	26	728
13	3	< 57	26	1231
13	4	45	3	3075
13	5	80	47	1286
13	6	< 59	13	437
13	7	48	47	629
14	0	65	51	49
14	1	< 58	22	1656
14	2	47	42	3468

Level ℓ = 5

h	K	F_o	F_c	α
-19	0	47	24	1861
-17	0	77	45	55
-17	1	63	34	809
-17	2	45	40	16
-17	3	< 57	11	3013
-17	4	< 58	30	6
-17	5	46	16	1123
-16	0	< 55	16	75
-16	1	96	59	2760
-16	2	< 55	33	3544
-16	3	< 55	16	37
-16	4	< 56	36	3248
-16	5	77	52	3166
-15	0	71	47	1852
-15	1	< 52	13	641
-15	2	72	44	1947
-15	3	102	65	2629
-15	4	42	45	1871
-15	5	< 54	23	2025
-15	6	< 55	26	2065
-15	7	< 56	21	3038
-15	8	< 57	24	2264
-15	9	46	45	2749
-14	0	55	71	1840
-14	1	< 50	34	1658
-14	2	70	64	1475
-14	3	113	85	1042
-14	4	57	65	1742
-14	5	< 52	24	396
-14	6	< 53	39	1401
-14	7	< 54	39	1663
-14	8	< 55	20	1135
-14	9	63	48	1171
-13	0	< 47	31	63
-13	1	150	97	521
-13	2	38	41	301
-13	3	< 49	21	3194
-13	4	39	32	129
-13	5	119	77	1199
-13	6	< 51	33	417
-13	7	82	69	691
-12	0	112	117	45
-12	1	112	91	2415
-12	2	113	97	17
-12	3	< 45	11	328
-12	4	64	61	79
-12	5	160	124	3742
-12	6	86	74	3518
-12	7	39	44	2441
-12	8	57	42	3171
-12	9	72	39	630
-12	10	< 54	9	2560
-12	11	62	46	3526
-11	0	47	62	1851
-11	1	125	109	3308
-11	2	< 43	31	853
-11	3	168	159	2757
-11	4	35	37	2933
-11	5	101	73	1949
-11	6	37	61	2430
-11	7	53	60	3273
-11	8	39	28	2367
-11	9	106	82	2699
-11	10	41	28	2610
-11	11	61	57	2152
-10	0	< 39	62	1855
-10	1	135	123	1976
-10	2	155	142	1920
-10	3	129	134	1246
-10	4	153	142	1698
-10	5	89	74	3486
-10	6	35	32	1462
-10	7	102	83	1969
-10	8	84	58	2131
-10	9	87	48	989
-10	10	57	47	1648
-10	11	< 53	35	3321
-10	12	< 55	11	1063
-10	13	44	47	1873
-9	0	41	45	1880
-9	1	217	216	643
-9	2	111	96	731
-9	3	43	36	1650
-9	4	< 39	12	3199
-9	5	202	205	1223
-9	6	94	84	1510
-9	7	138	135	614
-9	8	102	107	877
-9	9	< 47	5	3421
-9	10	< 49	29	288
-9	11	70	65	1364
-9	12	< 53	24	2047
-9	13	43	49	381
-8	0	305	295	39
-8	1	101	91	2312
-8	2	151	160	3249
-8	3	156	161	836
-8	4	101	93	731
-8	5	152	142	3393
-8	6	144	134	166
-8	7	57	67	2042
-8	8	34	55	3240
-8	9	125	101	776
-8	10	38	50	960
-8	11	88	80	3374
-8	12	71	54	268
-7	0	70	64	1461
-7	1	107	106	279
-7	2	134	137	533
-7	3	319	323	2697
-7	4	235	205	3042
-7	5	120	115	2134
-7	6	95	113	2398
-7	7	131	111	2969
-7	8	47	57	492
-7	9	172	155	2835
-7	10	104	79	3167
-7	11	< 49	31	1439
-7	12	40	39	2193
-6	0	< 28	7	1706
-6	1	165	212	2599
-6	2	264	299	2135
-6	3	126	119	942
-6	4	241	219	1418
-6	5	170	167	3091
-6	6	76	76	3213
-6	7	163	152	2179
-6	8	163	151	2437
-6	9	76	79	1305
-6	10	113	104	1594
-6	11	106	77	2734
-6	12	< 50	37	253
-6	13	58	55	1989
-6	14	43	47	2580
-5	0	291	321	1841
-5	1	228	253	678
-5	2	221	212	1016
-5	3	63	58	1977
-5	4	124	112	2982
-5	5	252	248	1176
-5	6	223	232	1865
-5	7	136	115	601
-5	8	153	139	827
-5	9	33	42	2266
-5	10	70	64	3196
-5	11	128	106	1354
-5	12	77	80	1849
-5	13	41	44	520
-5	14	42	42	994
-4	0	403	425	36
-4	1	114	114	1565
-4	2	197	195	2816
-4	3	263	266	876
-4	4	214	229	999
-4	5	165	163	152
-4	6	253	257	146
-4	7	69	52	1420
-4	8	105	89	3002
-4	9	159	164	542
-4	10	108	103	1738
-4	11	73	67	32
-4	12	76	95	138
-4	13	< 51	41	1449
-4	14	42	37	2670
-4	15	< 55	36	445
-4	16	45	40	1231
-3	0	* 16	68	1870
-3	1	* 18	214	3394
-3	2	285	296	508
-3	3	228	208	2956
-3	4	350	342	3156
-3	5	33	35	1564
-3	6	141	122	2096
-3	7	153	129	3358
-3	8	229	177	427
-3	9	100	74	2775
-3	10	208	174	3241
-3	11	< 45	18	1954
-3	12	53	66	1920
-3	13	69	77	3394
-3	14	59	56	730
-3	15	< 55	28	2562
-3	16	45	36	3457
-2	0	* 8	169	44
-2	1	* 13	221	2599
-2	2	* 18	348	2264
-2	3	119	104	2791
-2	4	310	292	1380
-2	5	218	240	2918
-2	6	175	155	3495
-2	7	143	173	2220
-2	8	210	182	2460
-2	9	< 40	30	99
-2	10	154	137	1526
-2	11	50	51	3030
-2	12	65	70	62
-2	13	97	81	1827
-2	14	83	71	2537
-2	15	< 55	16	2365
-2	16	45	32	1443
-1	0	* 11	454	1842
-1	1	* 10	71	183
-1	2	* 17	335	1003
-1	3	57	52	1798
-1	4	131	118	2995
-1	5	197	195	1349
-1	6	301	333	1832
-1	7	< 34	21	1479
-1	8	204	168	966
-1	9	89	75	1735
-1	10	100	97	2752
-1	11	61	73	1497
-1	12	130	109	1942
-1	13	< 50	75	3269
-1	14	72	50	608
-1	15	< 55	29	1850
-1	16	45	37	3056
0	-16	45	33	2428
0	-15	< 55	38	1570
0	-14	72	67	1204
0	-13	40	52	705
0	-12	106	91	3458
0	-11	< 45	15	1493
0	-10	181	134	2445
0	-9	109	88	1393
0	-8	147	124	1296
0	-7	136	123	753
0	-6	211	229	3523
0	-5	145	115	859
0	-4	286	290	2624
0	-3	201	195	829
0	-2	164	198	1166
0	-1	* 14	109	681
0	0	* 11	429	31
1	0	< 18	7	2775
1	-14	59	60	3132
1	-13	40	34	2162
1	-12	54	66	1791
1	-11	36	61	1550
1	-10	157	148	517
1	-9	< 40	49	2234
1	-8	185	210	3209
1	-7	39	99	2244
1	-6	110	113	1645
1	-5	33	49	1199
1	-4	344	397	499
1	-3	< 74	47	139
1	-2	248	334	3345
1	-1	< 19	22	453
2	-14	59	60	1258
2	-13	40	9	2547
2	-12	77	85	143
2	-11	51	79	2874
2	-10	103	82	2318
2	-9	46	67	2486
2	-8	141	152	1203
2	-7	40	45	2462
2	-6	145	142	304
2	-5	112	132	2812
2	-4	179	206	2383
2	-3	113	119	2222
2	-2	233	288	1034
2	-1	155	193	2371
2	0	240	327	32
3	-16	46	31	686
3	-15	< 56	27	3572
3	-14	43	39	3162
3	-13	41	37	3012
3	-12	110	84	1948
3	-11	< 47	37	1240
3	-10	70	80	923
3	-9	< 42	30	3431
3	-8	109	122	2735
3	-7	114	141	3134
3	-6	220	262	1866
3	-5	< 33	22	2957
3	-4	170	175	998
3	-3	< 29	25	3289
3	-2	179	199	2444
3	-1	102	109	325
3	0	294	384	1840
4	-13	41	49	970
4	-12	56	71	3456
4	-11	< 48	42	360
4	-10	125	113	2459
4	-9	< 43	11	852
4	-8	97	83	1523
4	-7	97	89	900
4	-6	125	127	3566
4	-5	118	116	271
4	-4	194	221	2422
4	-3	78	88	976
4	-2	223	221	1520
4	-1	130	149	1258
4	0	91	95	64
5	-10	117	93	443
5	-9	50	56	727
5	-8	130	126	3395
5	-7	< 40	9	2521
5	-6	30	42	1641
5	-5	58	77	1460
5	-4	188	218	406
5	-3	93	113	1050
5	-2	184	182	3247
5	-1	51	52	3109
5	0	87	72	1842
6	-12	41	49	450
6	-11	< 50	23	154
6	-10	54	31	2102
6	-9	63	91	2405
6	-8	70	69	1070
6	-7	47	65	1802
6	-6	148	139	246
6	-5	31	37	2845
6	-4	94	83	2939
6	-3	123	149	2595
6	-2	135	153	835
6	-1	39	64	2458
6	0	195	200	41
7	-11	41	36	2461
7	-10	79	76	832
7	-9	38	29	374
7	-8	36	20	2989
7	-7	78	73	2942
7	-6	117	123	1929
7	-5	32	50	2479
7	-4	94	89	1047
7	-3	< 39	55	1229
7	-2	95	76	2564
7	-1	94	99	2915
7	0	143	109	1866
8	-10	57	48	2500
8	-9	< 50	16	2461
8	-8	38	46	1834
8	-7	63	70	1013
8	-6	35	26	3054
8	-5	34	66	344
8	-4	105	119	2392
8	-3	< 41	17	2522
8	-2	124	118	1602
8	-1	100	93	1179

Table 7 (*cont.*)

h	K	F_o	F_c	α
8	0	< 40	27	1785
9	−9	57	58	922
9	−8	< 50	45	3094
9	−7	< 48	25	367
9	−6	< 47	34	2397
9	−5	36	32	1361
9	−4	35	50	710
9	−3	104	84	974
9	−2	68	63	3071
9	−1	48	46	81
9	0	48	46	1822
10	−3	52	47	2645
10	−2	51	57	215
10	−1	36	48	2264
10	0	107	106	39
11	−4	55	56	835
11	−3	< 49	22	3393
11	−2	< 48	16	587
11	−1	53	70	3342
11	0	< 48	15	1902
12	−2	40	34	1471
12	−1	< 50	38	1560
12	0	< 50	16	1871
Level ℓ = 6				
−18	0	< 40	5	1837
−18	1	43	17	941
−17	0	< 38	2	294
−17	1	41	17	1312
−17	2	< 39	32	118
−17	3	< 39	28	1097
−17	4	42	25	3541
−16	0	69	46	49
−16	1	40	29	3116
−16	2	< 37	20	3085
−16	3	40	35	3120
−15	0	66	46	1851
−15	1	66	34	2560
−15	2	< 36	7	1872
−15	3	< 36	9	2516
−15	4	39	24	2209
−15	5	< 37	37	2925
−15	6	57	37	1853
−15	7	41	35	2481
−14	0	< 34	25	1845
−14	1	37	39	539
−14	2	52	53	1905
−14	3	< 35	18	2613
−14	4	53	51	1554
−14	5	< 36	43	1631
−14	6	39	17	1248
−14	7	40	23	1213
−13	0	35	13	1832
−13	1	35	26	1605
−13	2	61	50	480
−13	3	94	76	943
−13	4	36	25	3564
−13	5	36	37	489
−13	6	< 35	11	849
−13	7	38	33	1630
−13	8	< 36	26	636
−13	9	57	47	1028
−12	0	104	84	53
−12	1	57	48	267
−12	2	67	67	3379
−12	3	83	64	2858
−12	4	59	63	169
−12	5	< 33	17	1190
−12	6	62	56	3537
−12	7	64	60	122
−12	8	38	33	2856
−12	9	55	45	3199
−11	0	< 29	3	3159
−11	1	108	100	2535
−11	2	< 29	34	332
−11	3	32	36	906
−11	4	< 30	38	2671
−11	5	100	88	3206
−11	6	< 32	15	2101
−11	7	93	77	2506
−11	8	< 34	5	3142
−11	9	< 35	9	2201
−11	10	39	49	2626
−11	11	56	52	3040
−10	0	82	86	1933
−10	1	93	96	464
−10	2	111	103	2002
−10	3	95	87	2604
−10	4	87	76	1611
−10	5	110	96	1694
−10	6	< 30	40	1698
−10	7	< 31	21	287
−10	8	35	37	1994
−10	9	73	53	2769
−10	10	< 35	16	940
−10	11	55	42	1717
−9	0	77	89	1821
−9	1	102	93	1660
−9	2	28	34	1532
−9	3	169	164	1063
−9	4	< 27	34	1417
−9	5	43	50	182
−9	6	54	52	1149
−9	7	103	97	1807
−9	8	< 31	39	710
−9	9	99	99	1019
−9	10	< 34	15	1386
−9	11	39	45	90
−8	0	98	101	40
−8	1	115	141	232
−8	2	77	67	379
−8	3	106	106	2863
−8	4	87	88	28
−8	5	116	113	1320
−8	6	73	69	3599
−8	7	124	114	476
−8	8	< 30	7	1308
−8	9	34	37	3083
−8	10	35	38	241
−8	11	37	49	1429
−8	12	< 36	23	200
−8	13	69	48	357
−7	0	< 21	19	157
−7	1	161	189	2318
−7	2	92	122	3194
−7	3	85	88	1244
−7	4	68	62	3449
−7	5	154	165	3180
−7	6	63	50	3057
−7	7	129	130	2468
−7	8	77	74	3109
−7	9	33	39	551
−7	10	< 32	22	2312
−7	11	96	78	3231
−7	12	< 35	7	569
−7	13	68	43	2165
−6	0	78	67	1874
−6	1	130	141	41
−6	2	75	84	1831
−6	3	213	208	2731
−6	4	98	90	2490
−6	5	116	109	1566
−6	6	94	75	2190
−6	7	86	86	141
−6	8	52	51	1773
−6	9	121	118	2698
−6	10	34	38	3033
−6	11	79	76	1845
−6	12	37	32	2538
−5	0	26	37	1837
−5	1	115	130	1915
−5	2	121	123	2202
−5	3	222	219	945
−5	4	139	129	1597
−5	5	125	122	3591
−5	6	63	73	1219
−5	7	133	127	1880
−5	8	29	13	1843
−5	9	127	137	989
−5	10	33	41	1583
−5	11	< 32	39	3409
−5	12	36	33	752
−5	13	66	56	2046
−4	0	15	13	1854
−4	1	248	262	480
−4	2	101	103	479
−4	3	109	110	2822
−4	4	131	123	89
−4	5	160	141	1317
−4	6	66	64	1373
−4	7	155	166	465
−4	8	95	92	924
−4	9	30	26	2972
−4	10	32	52	246
−4	11	97	83	1176
−4	12	< 33	21	2070
−4	13	84	71	434
−3	0	* 11	206	42
−3	1	* 12	172	2287
−3	2	86	92	3240
−3	3	109	124	752
−3	4	94	81	706
−3	5	211	187	3287
−3	6	138	123	18
−3	7	132	140	2212
−3	8	80	67	2868
−3	9	74	75	824
−3	10	32	43	1909
−3	11	96	89	3196
−3	12	87	74	303
−2	0	* 3	169	1838
−2	1	* 9	126	17
−2	2	< 13	63	957
−2	3	277	269	2660
−2	4	165	147	3223
−2	5	109	98	1889
−2	6	112	117	2140
−2	7	112	116	3423
−2	8	< 26	15	1094
−2	9	138	139	2687
−2	10	63	59	3191
−2	11	82	78	1678
−2	12	79	67	2094
−2	13	< 35	28	315
−2	14	< 36	10	1193
−2	15	40	34	2501
−1	0	* 6	47	22
−1	1	* 8	156	2080
−1	2	* 12	184	2171
−1	3	194	179	1095
−1	4	215	183	1472
−1	5	106	93	3268
−1	6	48	53	509
−1	7	127	128	1961
−1	8	92	81	2340
−1	9	79	75	1168
−1	10	100	85	1872
−1	11	75	63	2895
−1	12	< 33	33	166
−1	13	53	55	1870
−1	14	39	41	2458
0	−15	40	18	2784
0	−14	39	34	2673
0	−13	37	56	1723
0	−12	62	59	1664
0	−11	68	69	737
0	−10	55	75	373
0	−9	< 28	25	3087
0	−8	109	117	2887
0	−7	115	124	1362
0	−6	103	130	2016
0	−5	156	178	627
0	−4	89	88	293
0	−3	54	50	3142
0	−2	121	145	2954
0	−1	165	200	1291
0	0	< 9	90	1870
1	−12	62	62	3383
1	−11	83	83	2100
1	−10	65	64	2512
1	−9	96	95	1363
1	−8	86	80	620
1	−7	65	66	3506
1	−6	140	155	60
1	−5	96	99	2272
1	−4	101	104	2879
1	−3	104	107	928
1	−2	181	198	894
1	−1	122	131	183
1	0	146	166	62
2	−13	38	41	2156
2	−12	36	29	2014
2	−11	34	14	293
2	−10	93	89	544
2	−9	76	76	2939
2	−8	58	52	2888
2	−7	77	76	1921
2	−6	77	90	1498
2	−5	< 22	21	7
2	−4	174	189	618
2	−3	163	165	2678
2	−2	144	140	2853
2	−1	109	113	2145
2	0	60	68	1874
3	−13	38	46	25
3	−12	37	37	3431
3	−11	35	48	2985
3	−10	100	85	2230
3	−9	< 29	19	870
3	−8	95	89	1234
3	−7	49	59	3456
3	−6	27	42	436
3	−5	87	77	2589
3	−4	163	155	2313
3	−3	44	31	515
3	−2	183	198	1352
3	−1	110	112	3218
3	0	< 18	8	1430
4	−12	37	45	1777
4	−11	36	45	701
4	−10	34	45	571
4	−9	33	43	3485
4	−8	103	97	2797
4	−7	< 27	50	1374
4	−6	141	123	1969
4	−5	88	82	709
4	−4	87	80	686
4	−3	< 22	44	3478
4	−2	92	104	2777
4	−1	63	51	1104
4	0	126	119	1863
5	−12	76	55	3518
5	−11	< 34	7	2416
5	−10	35	30	2660
5	−9	33	46	1232
5	−8	56	41	895
5	−7	31	45	680
5	−6	102	100	56
5	−5	< 26	30	1608
5	−4	71	68	2535
5	−3	73	66	1305
5	−2	94	95	932
5	−1	49	41	401
5	0	173	198	36
6	−12	39	45	1708
6	−11	38	25	1386
6	−10	36	41	542
6	−8	58	56	3265
6	−7	45	52	1977
6	−6	31	52	1691
6	−5	30	49	2251
6	−4	130	130	422
6	−3	48	51	2413
6	−2	108	100	3430
6	−1	< 25	34	3522
6	0	< 25	30	1876
7	−2	65	66	1614

atom. The positions of the phenol oxygen and the connected carbon atom, being between the two iodine atoms, are probably strongly affected by errors in the data due to the presence of the iodine atoms. To be sure there is some change in the benzene ring due to the iodine atoms as is demonstrated by the phenol *pK* of di-iodotyrosine being considerably lower than that of tyrosine. However, the *pK* of *p*-nitrophenol is quite similar to that of di-iodotyrosine, but the carbon to oxygen bonds were found to be normal (1·351 ± 0·006 and 1·361 ± 0·003, respectively) in two separate modifications (Coppens & Schmidt, 1965*a*,*b*).

The iodine–iodine distance of 6·05 ± 0·02 Å is shorter than that expected from consideration of van der Waals radii but agrees with values of 5·96 ± 0·02 Å and 6·03 ± 0·02 Å found in thyroxine (Hamilton, Pinkerton, Seely & Steinrauf, 1967).

Hydrogen bonding

The molecules of di-iodotyrosine are held together in the unit cell by an elaborate system of hydrogen bonds involving the carboxyl group, amino groups, and phenol group of the molecules and the two molecules of water of crystallization. The water molecules lie almost exactly along the *c* axis in the *ac* projection. The benzene rings are tilted slightly from the *ac* plane and packing in the **a** and **b** directions is governed by closest approach of the iodine atoms and the benzene rings.

A list of the shorter intermolecular distances with standard deviations is given in Table 4 and the corresponding angles in Table 5. The more interesting distances are also plotted on the packing diagram in Fig. 3.

Since the positions of the hydrogen atoms have not been determined, we can only postulate a hydrogen bonding scheme. A reasonable scheme would have the three hydrogen atoms from the charged amino group bonding to each of the water molecules and to O(14) of the carboxyl group in a tetrahedral arrangement. The hydrogen atoms of the water molecule of O(16) bond to O(15) of the carboxyl group and O(17) of the other water molecule; a hydrogen bond to O(16) is made by the amino group. The hydrogen atoms of the other water molecule, O(17), bond to O(13) of the phenol and to O(15) of the carboxyl group; bonds to O(17) are made by hydrogen atoms of the amino group and the other water molecule. The arrangement about each of the water molecules is only very approximately tetrahedral. In this scheme the hydrogen atom from the phenol group is not used. The hydrogen bond formed by the phenol group and the water molecule is 3·08 ± 0·2 Å, which is probably long owing to the short carbon–oxygen bond the phenol group makes with the benzene ring (mentioned above) and would be more satisfactory if the oxygen were moved farther from the benzene ring. However, when the oxygen was moved out, the least-squares refinement moved it back to the present position. The above water molecule is 0·56 Å from the plane of the benzene ring.

Mean plane calculation

The equation of the mean plane through the benzene ring is: $0{\cdot}1771X + 0{\cdot}7571Y - 0{\cdot}6289Z + 1{\cdot}5033 = 0$. The deviations of these atoms and of atoms O(13), C(9), I(1), and I(2) from this plane are shown in Table 6. None of these are considered significant. The closest approach of any other iodine atom to this plane is 3·3 Å. No π bonding is considered present between the iodine and the ring.

A three-dimensional Fourier map obtained from the copper data and the final phases for the molybdenum data proved to be almost impossible to interpret.

All computer programs used in this project were written by the crystallographic group here, except the Busing & Levy full-matrix least-squares program and

function and error program (Busing, Martin & Levy, 1962) and a much modified block-diagonal least-squares program (*World List of Crystallographic Computer Programs*, 1966).

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The Crystal Structure of Nitrilotriacetic Acid

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The crystal structure of nitrilotriacetic acid has been determined by the application of the symbolic addition technique. Three-dimensional data were collected photographically about two axes with copper $K\alpha$ radiation. The coordinates of the six methylene-hydrogen atoms and the coordinates and anisotropic temperature parameters for the thirteen heavier atoms were refined by the method of least squares. The final R index was 0·093. The molecule of nitrilotriacetic acid exists in the crystal as the zwitterion $HN^+(CH_2COOH)_2CH_2COO^-$. The hydrogen atom bonded to nitrogen appears to form a bifurcated hydrogen bond. The two remaining acidic hydrogen atoms form strong hydrogen bonds of only 2·51 Å length, which connect the molecules into two interpenetrating helical networks. The crystals are monoclinic, space group Cc with $a=7{\cdot}9936$, $b=18{\cdot}512$, $c=6{\cdot}493$ Å and $\beta=127{\cdot}73°$, but there exists a nearly orthogonal cell which provides a simple description of these networks. The bond distances and angles are normal within experimental error.

Introduction

Nitrilotriacetic acid is widely used in complexometric titrations, and its acid and complexometric properties have been thoroughly studied (Schwarzenbach, Kampitsch & Steiner, 1945; Schwarzenbach, Ackermann & Ruckstuhl, 1949; and Schwarzenbach & Freitag, 1951). The pK values for this molecule in $0{\cdot}1N$ KCl at 20°C, as determined by Schwarzenbach and his coworkers, are:

$$pK_1=1{\cdot}89$$
$$pK_2=2{\cdot}49$$
$$pK_3=9{\cdot}73\ .$$

The present investigation was initiated in order to determine whether the zwitterion, $HN^+(CH_2COOH)_2$-CH_2COO^-, is the stable form of the molecule in the crystal.

Experimental

Small needlelike crystals were obtained by slow evaporation of an aqueous solution of nitrilotriacetic acid. A pH of 2·3 was measured for this saturated solution. Unit-cell parameters were determined from Straumanis-type single-crystal rotation photographs about the b and c axes. The resulting values for the unit-cell dimensions are given in Table 1. The absence of hkl reflections with $h+k$ odd and the absence of $h0l$ reflections with l odd indicate that the space group is either Cc or $C2/c$.

The density of the crystals as measured by the flotation method is $1{\cdot}660 \pm 0{\cdot}003$ g.cm^{-3}. The density cal-

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