A Thesis

CRYSTAL STRUCTURE ANALYSES
of
ANDROSTANE DERIVATIVES
and
ORGANO-ARSENIC COMPOUNDS.

Submitted to the University of Glasgow
for the Degree of Doctor of Philosophy
in the Faculty of Science.

by

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Summary.

The thesis is divided into three parts: the first part is a brief review of the theory and methods of X-ray crystallography and the second and third parts contain examples of the application of the heavy atom method to structural problems in organic and organo-metallic chemistry respectively.

The widespread occurrence of 4,4-dimethyl substituted 3-keto-5α-steroids and triterpenoids in nature and the high degree of substitution in ring A of these compounds has aroused interest in the conformation of ring A. In order to obtain quantitative information about the effect of the 1-3 interaction of the axial methyl groups on the conformation of ring A, the crystal structure analyses of 3-keto-4,4-dimethyl-5α-androstane-17β-iodoacetate and its 19-nor analogue have been completed and are contained in part II of this thesis. The analyses establish that, in the solid state, ring A adopts a partially flattened chair conformation in the former case and an undistorted chair conformation in the latter.

Part III contains the crystal structure analyses of three compounds, viz. triphenyl arsenic dihydroxide, triphenyl arsenic hydroxychloride and triphenyl arsenic hydroxybromide. These compounds are produced by either complete or partial hydrolysis of the corresponding

triphenylarsine dihalides and have been formulated as $Ph_3As(OH)_2$, $Ph_3As(OH)Cl$ and $Ph_3As(OH)Br$ respectively. By analogy with other compounds containing penta-coordinate group VB elements, these compounds have been assumed to adopt a trigonal bipyramidal structure. The crystal structure analysis of the dihydroxide establishes that, in the solid state, the compound is, in fact, triphenylarsine oxide monohydrate. The analyses of the hydroxyhalides establish that the halogen is not bonded to the arsenic atom in either case and that the compounds are more correctly formulated as 1:1 adducts of triphenylarsine oxide and the corresponding halogen acid. The composition of the adducts is maintained by a very short O...Halogen hydrogen bond.

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PART I

METHODS OF X-RAY ANALYSIS

1.1 Introduction.

By the first decade of this century, classical crystallography had been developed to such a stage that it was possible to derive the crystal class and the three-dimensional symmetry of a crystal by examination of its external morphology. Further development of the subject was made possible by von Laue's realisation in 1912 that X-radiation has a wavelength of the same order of magnitude as the distance between adjacent atoms in solids. A crystal will therefore diffract X-rays in much the same way as a diffraction grating diffracts light. The science of crystallography has now become almost totally concerned with interpretation of the diffraction patterns produced when a crystal is irradiated with monochromatic X-rays.

Since the angles of diffraction are dependent on the lattice translations of the crystal and the X-rays are diffracted by the electrons present in the crystal, it is now possible to determine, from the diffraction pattern, not only the symmetry of the crystals, but also the electron-density distribution and the atomic positions.

The method of X-ray diffraction found immediate success in the structures of ionic crystals but molecular structures proved to be a much more complex problem. The major advance in the study of molecular crystals came with the realisation that an atom, or group of atoms, which would dominate the scattering would reduce the complexity of the problem. This method, known as the heavy atom method (section 1.6.1), has been used to great effect in solving complex structures of many classes of compounds, e.g. steroids, terpenes and proteins.

The advent of the electronic computer has eased the burden of calculation which was placed on the crystallographer by the size and complexity of the calculations involved in the interpretation of the diffraction pattern. In some cases, with suitable programming, it has become possible to collect the diffraction data and solve the structure automatically.

Crystallography, in its present state of development, has become one of the most powerful methods available for detailed study of structure and bonding in molecules and has been the catalyst for the development of many different fields of study.

1.2 Geometry of X-ray Diffraction.

Consider a beam of monochromatic X-rays of wavelength falling on a crystal, the direction of the incident beam being defined by the unit vector $\lambda \underline{s}_o$ (fig. 1.1). Let the beam be diffracted in the direction of the unit vector $\lambda \underline{s}$. The path difference between the waves scattered at a point A and those scattered in the same direction at the origin O, is then,

$$OB - AC = \lambda \underline{r}.\underline{s} - \lambda \underline{r}.\underline{s}_o = \lambda \underline{r}.\underline{S} \qquad (1)$$

where $\underline{r}$ is the vector position of A relative to the origin and $\underline{S} = \underline{s} - \underline{s}_o$. The vector $\underline{S}$ defines the spatial relationship of the incident and diffracted beams and it can be seen (fig. 1.2) that, if the angle between the incident and diffracted beam is 2θ,

$$|\underline{S}| = 2\sin\theta/\lambda . \qquad (2)$$

The path difference $\lambda \underline{r}.\underline{S}$ corresponds to a phase difference of $2\pi \underline{r}.\underline{S}$.

If the electron content of the volume element around A is $\rho(\underline{r})dV$, the scattered wave, relative to a wave scattered by a single electron at the origin, is defined, both in amplitude and phase, by

$$G_A(\underline{S}) = \rho(\underline{r})dV.\exp 2\pi i\underline{r}.\underline{S}. \qquad (3)$$

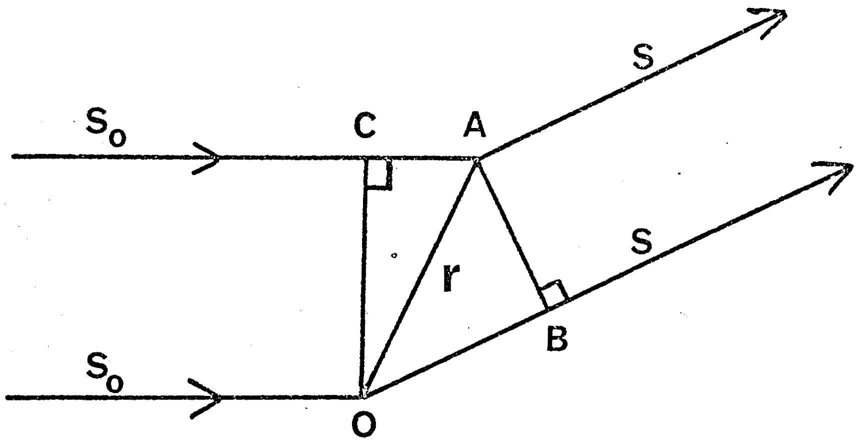


Fig. 1.1

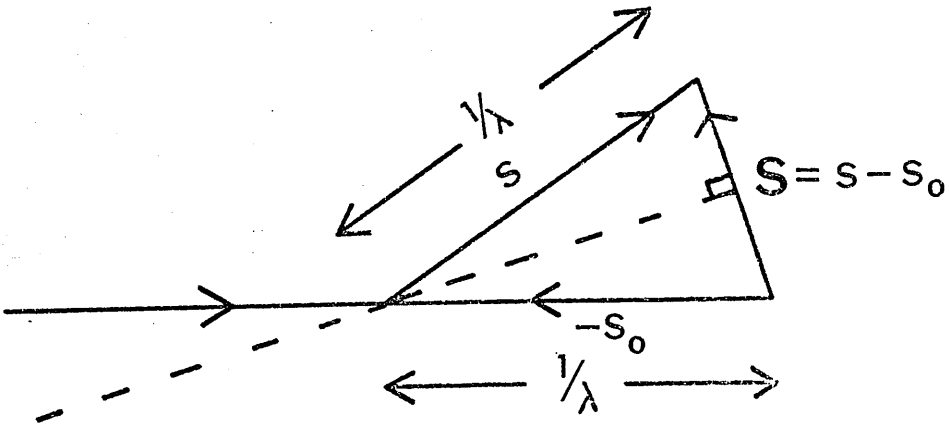


Fig. 1.2

In a crystal with lattice translations $\underline{a}$, $\underline{b}$ and $\underline{c}$, the electron-densities at all points B, defined by the vector equation

$$\underline{R} = \underline{r} + u\underline{a} + v\underline{b} + w\underline{c} \tag{4}$$

where u, v and w are integers, will be identical because of the periodicity of the electron density. The wave scattered in the direction $\lambda\underline{s}$ from any one of these points is therefore

$$\begin{aligned} G_B(\underline{S}) &= \rho(\underline{r})dV.\exp 2\pi i\underline{R}.\underline{S}. \\ &= G_A(\underline{S}).\exp 2\pi i(u\underline{a} + v\underline{b} + w\underline{c}).\underline{S}. \end{aligned} \tag{5}$$

The path difference between the waves is therefore $\lambda(u\underline{a} + v\underline{b} + w\underline{c}).\underline{S}$. For the scattered wave to have maximum intensity, the path difference must be an integral number of wavelengths. That is

$$\lambda(u\underline{a} + v\underline{b} + w\underline{c}).\underline{S} = n\lambda \tag{6}$$

where n is an integer, for all values of u, v and w. Hence

$$\begin{aligned} \underline{a}.\underline{S} &= h \\ \underline{b}.\underline{S} &= k \\ \underline{c}.\underline{S} &= l \end{aligned} \tag{7}$$

where h, k and l are integers must also hold. The equations

(7) are known as the Laue Equations.

$\underline{a}.\underline{S} = h$ represents a set of planes perpendicular to $\underline{a}$, with mutual separation $1/|\underline{a}|$. Similarly $\underline{b}.\underline{S} = k$ and $\underline{c}.\underline{S} = l$ represent sets of planes normal to $\underline{b}$ and $\underline{c}$ respectively. Since $\underline{a}$, $\underline{b}$ and $\underline{c}$ are never parallel, the Laue equations (7), represent three sets of planes which intersect to form a lattice, which is known as the reciprocal lattice. If the primitive translations of the reciprocal lattice are $\underline{a}^*$, $\underline{b}^*$ and $\underline{c}^*$, the reciprocal lattice is then defined by

$$\underline{S} = h\underline{a}^* + k\underline{b}^* + l\underline{c}^* \qquad (8)$$

From equations (7) and (8), it is apparent that $\underline{a}^*$ is normal to $\underline{b}$ and $\underline{c}$, and that the projection of $\underline{a}^*$ upon $\underline{a}$ is $1/|\underline{a}|$. Similar results follow for $\underline{b}^*$ and $\underline{c}^*$. The primitive translations of the reciprocal lattice are related to those of the real space lattice by

$$\begin{aligned} \underline{a}^* &= \underline{b} \times \underline{c}\,/V \\ \underline{b}^* &= \underline{c} \times \underline{a}\,/V \\ \underline{c}^* &= \underline{a} \times \underline{b}\,/V \end{aligned} \qquad (9)$$

where V is the volume of the unit cell in real space.

The Laue equations (7) may be rewritten in the form

$$\underline{a}/h.\underline{S} = \underline{b}/k.\underline{S} = \underline{c}/l.\underline{S} = 1 \qquad (10)$$

By definition, $\underline{a}/h$, $\underline{b}/k$ and $\underline{c}/l$ are the intercepts on the crystal axes of a plane with Miller indices (hkl). Since the projections of three points in the plane on $\underline{S}$ are equal, $\underline{S}$ must be normal to the plane and $|\underline{S}| = 1/d_{hkl}$ where d_{hkl} is the origin to plane distance. Substituting in (2) for $|\underline{S}|$ yields

$$\lambda = 2d_{hkl} \sin\theta . \tag{11}$$

Since $\underline{S}$ is normal to the plane with Miller indices (hkl), $\underline{s}_o$ and $\underline{s}$ make equal angles with the plane (fig. 1.2). In this manner, diffraction may be regarded as reflection from the crystal plane with Miller indices (hkl). This treatment was first derived by W.L.Bragg (1913) and allowed interpretation of the Laue equations (7) by reducing their complexity.

1.3 The Structure Factor.

The wave, $G(\underline{S})$, scattered by the entire contents of one unit cell is completely defined by

$$G(\underline{S}) = \int \rho(\underline{r}) \exp 2\pi i\, \underline{r}.\underline{S}\, dV \qquad (12)$$

where the integral is taken over the volume of one unit cell. $G(\underline{S})$ is the Fourier transform of the electron-density distribution.

If the unit cell contains N atoms, the vector position of the n^{th} atom is defined by

$$\underline{r}_n = \underline{x}_n + \underline{y}_n + \underline{z}_n \qquad (13)$$

where $\underline{x}_n$, $\underline{y}_n$ and $\underline{z}_n$ are the atomic coordinates. The total wave scattered from the n^{th} atom with respect to the origin of the unit cell is then

$$\int \rho_n(\underline{r}) \exp 2\pi i(\underline{r} + \underline{r}_n).\underline{S}\, dV$$

$$= \int \rho_n(\underline{r}) \exp 2\pi i\, \underline{r}.\underline{S}\, dV \exp 2\pi i\, \underline{r}_n.\underline{S}. \qquad (14)$$

The total wave scattered by the unit cell may be regarded as the sum of the waves scattered by the N individual atoms and is therefore

$$G(\underline{S}) = \sum_{1}^{N} \int \rho_n(\underline{r}) \exp 2\pi i\, \underline{r}.\underline{S}\, dV \exp 2\pi i\, \underline{r}_n.\underline{S}. \qquad (15)$$

The function

$$f_n(\underline{S}) = \int\!\!\int \rho_n(\underline{r}) \exp 2\pi i\, \underline{r}.\underline{S}\, dV \qquad (16)$$

is the Fourier transform of the atomic electron density and is known as the <u>atomic scattering factor</u>. If the atom is assumed to be centrosymmetric, the function $f_n(\underline{S})$ is real, if the origin of $\underline{r}$ has been taken at the centre of symmetry; if spherical symmetry is also assumed for the atom, $\rho_n(\underline{r})$ is a function only of $\underline{r}$. Therefore $f_n(\underline{S})$ is a function only of $|\underline{S}| = 2 \sin \theta/\lambda$ (from (2)).

The function $f_n(\underline{S})$ defines the wave scattered from a point $\underline{r}$ in the atom, relative to that scattered by a single electron. The phase differences between the waves arise from the finite volume occupied by the electronic distribution of the atom and are a function only of the scattering angle. Hence as $|\underline{S}|$ tends to zero, $f_n(\underline{S})$ tends to Z, the atomic number. As the angle of diffraction increases, the phase differences become larger and the scattered beam becomes weaker; that is the scattering factor becomes less than Z.

Since $\rho_n(\underline{r})$ is known accurately only for the hydrogen atom, the scattering factors for other atoms are, in general, only approximate. Values for atomic scattering factors, calculated on the assumption of spherical symmetry for the atoms, may be found, for example, in International Tables for X-ray Crystallography, Volume III, (1962).

Substitution of (16) into (15) yields

$$G(\underline{S}) = \sum_{1}^{N} f_n(\underline{S}) \exp 2\pi i \, \underline{r}_n \cdot \underline{S}. \tag{17}$$

The total wave scattered by the crystal has an appreciable amplitude only when the Laue equations (7) are simultaneously satisfied; that is, when

$$\underline{S} = h\underline{a}^* + k\underline{b}^* + l\underline{c}^*. \tag{8}$$

Equation (13) may be rewritten as

$$\underline{r}_n = x_n\underline{a} + y_n\underline{b} + z_n\underline{c} \tag{18}$$

where x_n, y_n and z_n are the fractional coordinates, with respect to the crystal axes, of the n^{th} atom. Substitution of (18) and (8) into (17) leads to

$$\begin{aligned} G(\underline{S}) = \sum_{1}^{N} f_n(\underline{S}) \exp 2\pi i(&hx_n\underline{a}.\underline{a}^* + kx_n\underline{a}.\underline{b}^* + lx_n\underline{a}.\underline{c}^* \\ &+ hy_n\underline{b}.\underline{a}^* + ky_n\underline{b}.\underline{b}^* + ly_n\underline{b}.\underline{c}^* \\ &+ hz_n\underline{c}.\underline{a}^* + kz_n\underline{c}.\underline{b}^* + lz_n\underline{c}.\underline{c}^*). \end{aligned} \tag{19}$$

From the definition of the reciprocal lattice, it can be shown that $\underline{a}.\underline{a}^* = 1$ and $\underline{a}.\underline{b}^* = \underline{a}.\underline{c}^* = 0$. Similar results hold for $\underline{b}$ and $\underline{c}$. Hence (19) becomes

$$G(\underline{S}) = F(hkl) = \sum_{1}^{N} f_n(\underline{S}) \exp 2\pi i(hx_n + ky_n + lz_n) \tag{20}$$

Thus the structure factor, F(hkl), is a special value of the Fourier transform of the electron-density distribution which can only be observed when the Laue equations (7) are satisfied, that is, when the total wave scattered by one unit cell has a non-zero amplitude. The structure factor then describes the amplitude and phase of this wave and therefore, since all unit cells must scatter in phase, of the total wave scattered by the crystal. F(hkl) is a complex quantity and may be written

$$F(hkl) = G(\underline{S}) = A + iB \tag{21}$$

where

$$A = \sum_{1}^{N} f_n \cos 2\pi(hx_n + ky_n + lz_n) \tag{22}$$

and

$$B = \sum_{1}^{N} f_n \sin 2\pi(hx_n + ky_n + lz_n). \tag{23}$$

The modulus of the structure factor, $|F(hkl)|$, which is known as the structure amplitude, and the phase angle $\alpha(hkl)$ are then defined by

$$|F(hkl)| = (A^2 + B^2)^{1/2} \tag{24}$$

$$\alpha(hkl) = \tan^{-1} B / A. \tag{25}$$

If the origin of the unit cell is taken at a centre of symmetry, the Fourier transform of the electron density, and therefore the structure factor, is real; that is, B = 0 and the phase angle α(hkl) is restricted to 0° or 180°.

The atomic scattering factor (16) is normally calculated for the isolated atom at rest, and thermal vibration therefore has no effect. In the crystal, however, the effect of thermal vibration of the atoms becomes important, since it causes atoms which should scatter in phase to scatter slightly out of phase reducing the amplitude of the scattered wave. This corresponds to the electron density of the atom in the crystal appearing to be more diffuse than that of an isolated atom. Waller (1927) has shown that the scattering factor for an atom vibrating isotropically is related to that of the atom at rest by

$$f_T = f_o \exp(-B \sin^2\theta / \lambda^2) \qquad (26)$$

where f_o is the scattering factor of the atom at rest. B, the temperature coefficient, may be evaluated from heat capacity data (Debye, 1914; Waller, 1923) and has a value

$$B = 8\pi^2 U \qquad (27)$$

where U = $\overline{u}^2$, the mean square amplitude of vibration of the atoms.

The Debye-Waller correction for thermal vibration (26) assumes that the atoms are vibrating equally in all directions, that is, that the spherical symmetry of the atom is maintained. In general, however, atoms vibrate with different amplitudes in different directions and the electron density is smeared over a small anisotropic volume which may be regarded as a triaxial ellipsoid. Each non-equivalent atom may then be described by a different ellipsoid and the ellipsoids will, in general, be differently orientated. The vibration of the atom may then be described by a symmetric tensor $\underline{U}$, with six independent components, such that the mean square amplitude of vibration in the direction of a unit vector $\underline{l} = \underline{l}_1 + \underline{l}_2 + \underline{l}_3$ is

$$\bar{u}^2 = \sum_{i=1}^{3} \sum_{j=1}^{3} \underline{U}_{ij} \cdot \underline{l}_i \cdot \underline{l}_j \tag{28}$$

where $\underline{U}$ and $\underline{l}$ are defined with respect to the reciprocal axes $\underline{a}^*$, $\underline{b}^*$ and $\underline{c}^*$. The modified scattering factor for an atom vibrating with anisotropic motion is then given by

$$f_T = f_o \exp[-2\pi^2 (U_{11} h^2 \underline{a}^{*2} + U_{22} k^2 \underline{b}^{*2} + U_{33} l^2 \underline{c}^{*2} + 2U_{23} kl\underline{b}^*.\underline{c}^* + 2U_{31} lh\underline{c}^*.\underline{a}^* + 2U_{12} hk\underline{a}^*.\underline{b}^*)] \tag{29}$$

where f_o is the scattering factor for the atom at rest (Cruickshank, 1965).

1.4 Fourier Series Representation of the Electron Density.

Since the electron density function $\rho(\underline{r})$ is periodic in three dimensions, it can be represented by a three-dimensional Fourier series. If u,v and w are integers and x, y and z fractional coordinates,

$$\rho(\underline{r}) = \rho(xyz) = \sum_{u}\sum_{v}\sum_{w}{}_{-\infty}^{+\infty} A(uvw)\ \exp[-2\pi i(ux + vy + wz)]. \tag{30}$$

The structure factor F(hkl) is the Fourier transform of the electron density $\rho(xyz)$. Therefore

$$F(hkl) = V\int_0^1\int_0^1\int_0^1 \rho(xyz)\exp\ 2\pi i(hx + ky + lz)dx.dy.dz. \tag{31}$$

Substituting for $\rho(xyz)$ in (31) and rearranging yields

$$F(hkl) = V\sum_{u}\sum_{v}\sum_{w}{}_{-\infty}^{+\infty}\int_0^1\int_0^1\int_0^1 A(uvw)\exp 2\pi i(h - u)x.\exp 2\pi i(k - v)y \times \exp 2\pi i(l - w)z.dx.dy.dz. \tag{32}$$

The triple integral in (32) has a value of zero unless $h = u$, $k = v$ and $l = w$. When the conditions $h = u$ etc. are satisfied, the integral has the value

$$F(hkl) = V.A(uvw) \tag{33}$$

which gives

$$\rho(xyz) = \frac{1}{V}\sum_{h}\sum_{k}\sum_{l}{}_{-\infty}^{+\infty} F(hkl)\ \exp[-2\pi i(hx + ky + lz)] \qquad (34)$$

$$\rho(xyz)$$

on substitution in (30). Thus the electron-density distribution may be represented by a three-dimensional Fourier series in which the coefficients are the structure factors scaled by the reciprocal of the unit cell volume.

1.5 Factors Affecting Intensity.

The total energy $E(hkl)$ which is reflected from the crystal plane with indices hkl when a crystal rotates with uniform angular velocity about an axis normal to the incident X-ray beam is given by

$$E(hkl) = K.L(hkl).p(hkl).\left|F(hkl)\right|^2 \qquad (35)$$

where K is a constant for the experiment. The value of K is given by

$$K = \frac{I \,.\, N^2 \,.\, \lambda^3 .\, dV}{\omega} \,.\, \frac{e^4}{m^2 .\, c^4} \qquad (36)$$

where I is the intensity of the incident beam,

N is the number of unit cells per unit volume,

λ is the wavelength of the X-radiation,

dV is the volume of the crystal,

e is the electronic charge,

ω is the angular velocity,

m is the mass of the electron,

c is the velocity of light.

The Lorentz Factor, $L(hkl)$, allows correction for the varying angular velocities with which the reciprocal lattice points pass through the surface of the sphere of reflection and is therefore dependent on the experimental conditions. Since the angular velocity of the planes is dependent on the angle between the incident beam and the plane normal to the rotation axis of the crystal, a rotation factor is normally incorporated in the Lorentz factor. The correction to be applied in the case of equi-inclination Weissenberg geometry is given by Tunell (1939) as

$$L(hkl) = \sin\theta.(\cos^2\mu - \cos^2\theta)^{-1/2}.(\sin 2\theta)^{-1} \qquad (37)$$

where μ is the angle between the incident beam and the plane normal to the rotation axis of the crystal.

If the X-ray beam incident on the crystal is unpolarised, the electric vector of the beam will be randomly orientated. Consideration of the geometrical conditions necessary for reflection from a plane shows that the component of the electric vector normal to the reflecting plane will be reduced by an amount which is proportional to the reflecting angle θ. This corresponds to a partial polarisation of the diffracted beam and results in a reduction of its intensity. The correction for this effect, which is independent of experimental conditions, is

$$p(hkl) = (1 + \cos 2\theta) / 2. \qquad (38)$$

Expression (35) relates the energy of the diffracted beam and the modulus of the structure factor $|F(hkl)|$. Since the intensity of the beam is proportional to the energy, (35) may be rewritten

$$I(hkl) \propto K.L(hkl).p(hkl).|F(hkl)|^2. \qquad (39)$$

The intensity of the diffracted beam may be measured either from the blackening produced on a photographic plate or by monitoring the diffracted beam with a quantum counter. The set of structure amplitudes $|F(hkl)|$ may, therefore, be measured experimentally but only on a relative scale. Several methods have been proposed for placing the structure amplitudes on absolute scale; for example, by comparison with a standard reflection from a standard crystal (Robertson, 1934), from the decrease in the average structure amplitude with increasing scattering angle (Wilson, 1942) or by comparison with the calculated structure amplitudes for the correct structural model.

In the above discussion some factors of geometric origin which cause a reduction in the intensity of the diffracted beam have been considered. The intensity of the diffracted beam, however, is also affected by physical factors which depend on the nature of the crystalline material under examination. The most important of these factors are absorption, primary extinction and secondary extinction.

Since X-radiation is absorbed by matter, there will be a reduction in the intensity of the emergent beam in comparison with that of the incident beam when X-rays travel through a crystal. The relationship between the intensities of the incident and emergent beams is given by

$$I = I_o e^{-\mu t} \tag{40}$$

where I_o is the intensity of the incident beam, μ is the linear absorption coefficient of the material and t is the distance the beam travels through the crystal. Since the path length through an irregularly shaped crystal will, in general, be different for each plane which satisfies the Bragg reflecting conditions, correction for absorption becomes extremely difficult. The effects of absorption, however, can be greatly reduced by selection of extremely small crystals and a suitable wavelength of X-rays.

Consider a beam of X-rays incident on a crystal plane which is in such an orientation as to satisfy the conditions of Bragg reflection. The reflected beam from this plane will be directed at such an angle as to satisfy the reflecting conditions for other members of the stack of planes and may therefore be reflected back into the incident direction. Since the incident and the multiply reflected beam will be exactly out of phase, the intensity of both the incident and the reflected beams will be reduced. This effect is known as *primary extinction*.

Secondary extinction arises from attenuation of the incident beam, as it penetrates the crystal, by planes which satisfy the reflecting conditions. The intensity of the beam incident on a crystal plane is therefore the intensity of the incident beam less that which has been reflected by any plane preceding the plane under consideration.

1.6 The Phase Problem.

It has been shown (section 1.4) that the structure factors F(hkl) are related to the coefficients of the Fourier series which represents the electron-density distribution in the crystal. The structure factors are complex quantities which are characterised by an amplitude $|F(hkl)|$ and a phase angle $\alpha(hkl)$. At the present time, methods of observing the relative intensities of the diffracted beams, which are related to the structure amplitudes by (39), are well established but no experimental method of observing the relative phase angles has been discovered. The science of X-ray crystallography is therefore concerned primarily with attempts to determine the phase angles of the diffracted beams. This constitutes what is known as the phase problem in X-ray crystallography and although many ways of overcoming this problem have been developed, none of them guarantees success.

The earliest structure solutions were done on a trial-or-error basis in which a structural model was proposed and the corresponding structure amplitudes compared with the observed values. This approach proved valuable for a number of structures which contained a small number of atoms but the number of possible structures for a molecule containing a large number of atoms would be astronomical. The number of possibilities may be reduced by constraints imposed by space group symmetry and consideration of atomic radii, but the trial-and-error method would obviously not be practicable for complex structures. For this reason a number of methods of overcoming the phase problem have been developed and some of the more important are reviewed below.

1.6.1 The Heavy-Atom Method.

This method of determining the unknown phase angles associated with the known structure amplitudes depends on the presence of an atom or small group of atoms of high atomic number compared with the major component of the structure. If such a grouping is not present naturally in the molecule, it must be incorporated by chemical means. Since the diffraction of X-radiation is dependent on the electron density of the atoms, the heavy atom will dominate the diffracted spectra. Thus the contribution of the heavy atom to the structure factors will outweigh that of the light atoms. If the position of the heavy atom can then be determined, a phase angle, which will approximate to the phase angle of the complete structure, may then be calculated for each structure amplitude. The electron-density distribution may then be evaluated by summation of the appropriate Fourier series, using the calculated phase angles and the observed structure amplitudes, and further features of the molecule may be revealed. It is then possible to derive the complete molecular structure by an iterative process of improving the phase angles by including the light atoms in the structure factor calculation and then evaluating the resultant electron-density distribution.

The resolution of the electron-density distribution derived from a partial knowledge of the structure may be improved by weighting the Fourier coefficients in a manner proportional to the probability of the phase angle being approximately correct. Methods of weighting the Fourier coefficients according to these criteria have been devised for centrosymmetric structures (Woolfson, 1956) and for non-centrosymmetric structures (Sim, 1960).

The first direct application of the heavy-atom method was in the crystal structure analysis of platinum phthalocyanine (Robertson and Woodward, 1940). The position of the heavy atom was fixed by the space group symmetry and the first electron-density distribution, based on the phase angles derived from the heavy atom position, revealed the entire structure of this very complex molecule.

The major disadvantage of the heavy-atom method is contained in the basic assumption that the heavy atom contribution to the structure factors will be greater than that of the rest of the molecule. The positions of the lighter atoms are therefore determined from the minor component of the structure factors and for this reason are less accurate. Despite the lessened accuracy of the positional parameters, the heavy-atom method is one of the most powerful approaches in structural chemistry.

1.6.2 The Isomorphous Replacement Method.

For this method of phase angle determination to be applicable, at least one heavy-atom derivative isomorphous with the parent compound must be available. Since the replacement of an atom in the parent compound by a heavier atom will give rise to differences in the intensities of the diffracted beams, and therefore in the structure amplitudes, of the two isomorphs, the heavy atom need not dominate the scattering as in the case of the heavy-atom method. If the position of the heavy atom is known, its contribution, $\underline{f}_H$, to the structure factors can be calculated and the phase angles associated with the diffracted spectra may be derived, for a centrosymmetric structure, by solution of the vector equation

$$\underline{F}_T - \underline{F}_O = \underline{f}_H \tag{41}$$

where $\underline{F}_T$ is the structure factor of the heavy-atom isomorph and F_o is that of the parent compound. The structure factors must be on absolute scale to permit solution of the phase problem by this method.

The above relationship was used to great effect in the elucidation of the crystal structures of the phthalocyanines. Two of the many possible isomorphs viz. metal-free phthalocyanine and nickel phthalocyanine were used (Robertson, 1935; 1936; Robertson and Woodward, 1937).

Non-centrosymmetric structures may also be solved by use of the isomorphous replacement method but, because of the continuously variable nature of the phase angles, three isomorphs are required, two containing heavy atoms and the parent compound. The phase angles may then be derived by graphical combination of the three structure factor vectors (Harker, 1956).

Since the addition of a heavy atom to an organic molecule may result in marked changes in the crystal structure the main limitation of the isomorphous replacement method is the condition of isomorphism. Crystals containing molecules of solvent have been used with great effect in structure solution by the isomorphous replacement method (Cheung and Sim, 1966) but in some cases this leads to very unsatisfactory results (Petersen et al., 1967). Nevertheless, the isomorphous replacement method is finding a wide application in the field of protein crystallography where the addition of heavy atoms or groups of atoms makes little difference to the crystal or molecular structure.

1.6.3 The Patterson Function.

The Patterson function (Patterson, 1934; 1935) arose from a consideration of the self-convolution of the electron density and is defined by

$$P(uvw) = V\int_0^1\int_0^1\int_0^1 \rho(xyz)\,\rho(x + u, y + v, z + w)\,dxdydz. \qquad (42)$$

Substitution of the values for the electron density derived from equation (34) yields

$$P(uvw) = \frac{1}{V}\int_0^1\int_0^1\int_0^1 \sum_h \sum_k \sum_{l=-\infty}^{+\infty} \sum_{h'} \sum_{k'} \sum_{l'} F(hkl)\exp[-2\pi i(hx+ky+lz)]\times$$

$$F(h'k'l')\exp[-2\pi i(h'x+k'y+l'z)]\exp[-2\pi i(h'u+k'v+l'w)].$$

$$dxdydz. \qquad (43)$$

The triple integral in (43) has non-zero values only when $h = -h'$, $k = -k'$ and $l = -l'$; therefore

$$P(uvw) = \frac{1}{V}\sum_h \sum_k \sum_{l=-\infty}^{+\infty} F(hkl).F(\bar{h}\bar{k}\bar{l})\exp[-2\pi i(h'u+k'v+l'w)]. \qquad (44)$$

Since $F(hkl)$ and $F(\bar{h}\bar{k}\bar{l})$ are complex conjugates,

$$F(hkl).F(\bar{h}\bar{k}\bar{l}) = |F(hkl)|^2 = |F(\bar{h}\bar{k}\bar{l})|^2 \qquad (45)$$

and equation (44) becomes

$$P(uvw) = \frac{1}{V}\sum_{h}\sum_{k}\sum_{l}{}_{-\infty}^{+\infty} |F(hkl)|^{2} \exp 2\pi i(hu + kv + lw). \quad (46)$$

The Patterson function is therefore a Fourier series in which the coefficients are the squares of the structure amplitudes. Since these quantities can always be derived from the intensities of the diffraction spectra, the series can be summed without ambiguity. From (42) it can be seen that the function P(uvw) will attain large values only when the vector (uvw) corresponds to a vector between two peaks in the electron-density distribution; the value of P(uvw) is then approximately equal to the product of the electron densities of the two peaks.

The Patterson function therefore yields a vector map which will have peaks at distances from the origin which correspond to interatomic vectors. This would appear to be a general solution to the phase problem but, for a structure containing N atoms, it can be shown that N(N - 1) distinct peaks should be present in the Patterson function. If these N(N - 1) peaks were fully resolved , complete interpretation of the Patterson function would be possible but this is not normally the case even for reasonably simple molecules. Overlap of peaks arising from interatomic vectors which are almost parallel obscures detail and complicates interpretation of the Patterson function.

The problem is somewhat simplified by the use of space group symmetry. Harker (1936) pointed out that certain symmetry elements gave rise to a concentration of peaks on certain plane and line sections of the Patterson function and that examination of these sections might ease interpretation.

Since the height of a peak in the Patterson function is proportional to the product of the electron densities at the ends of the vector (uvw), the presence of a heavy atom which dominates the scattering simplifies the interpretation. After the coordinates of the heavy atom have been derived, the structure solution may proceed by the heavy atom method (section 1.6.1) or, if a non-heavy atom isomorph is available, by the isomorphous replacement method (section 1.6.2).

1.6.4 Direct Methods.

Although the solution of an unknown structure by methods involving use of heavy atoms or the Patterson function may be regarded as a direct solution of the phase problem, the term direct method has become restricted, by usage, to those methods which attempt to derive the phase angles of the structure factors without postulating atomic positions.

The first direct method of structure solution was based on inequality relationships which arise between the structure amplitudes when the electron density is constrained to being positive (Harker and Kasper, 1948). Further development of direct methods was due to Sayre (1952) who derived equality relationships between the structure amplitudes. Using these equality relationships, Karle and Hauptman (1954) formulated a statistical treatment of the structure amplitudes which can yield information about the phase angles. This method is known as the symbolic additon method and has proved to be very powerful for structure solution.

Direct methods have been widely applied in the case of centrosymmetric structures where the phase angles are constrained by symmetry but more recently non-centrosymmetric structures have been solved, notably by the symbolic addition method (for example, the alkaloid reserpine, Karle and Karle, 1968).

1.7 Methods of Refinement.

In the initial stages of a crystal structure analysis, the correctness or incorrectness of the proposed model is normally decided by comparison of Fo and Fc, the observed and calculated structure amplitudes. The agreement between these quantities is expressed as a residual, R, defined by

$$R = \frac{\sum \big| |Fo| - |Fc| \big|}{\sum |Fo|} . \qquad (47)$$

Refinement of the structural model by systematically varying the atomic parameters will therefore lead to a minimum value of R.

Since the refinement of a structure consists of improving the atomic positions, and therefore the calculated phase angles, solution of the structure by use of consecutive Fourier syntheses will merge with refinement by Fourier methods. Fourier refinement is therefore important as it can proceed when the structure is only partially known.

1.7.1 Fourier Refinement.

The process of Fourier refinement may be regarded as beginning when either a heavy atom or a group of light atoms has been located in an unknown structure. The atomic coordinates may be used to calculate approximate phase angles which are used in conjunction with the observed amplitudes to evaluate the electron-density distribution by summation of the appropriate Fourier series. It may be possible to identify further molecular features in the electron-density distribution and inclusion of these in the calculation of the phase angles will yield improved values. The process is repeated until the phase angles calculated from the coordinates derived from the electron-density distribution show no difference from those used in its calculation.

In the preliminary stages of refinement by this method only those reflections for which the phasing is reliable should be included in the calculations. These reflections may be selected by the crystallographer or, alternatively, the reflections may be weighted according to the reliability of their calculated phase angles (Woolfson, 1956; Sim, 1960).

The electron-density distribution is subject to termination of series errors because the Fourier series(34) is summed over a finite number of terms. Truncation of the series in this manner results in the peaks in the electron-density maps being surrounded by diffraction ripples. Overlap of these ripples will result in slight displacements of the derived atomic positions. Booth (1946) proposed a simple correction, known as the backshift correction, for this effect. It involves the calculation of two separate Fourier syntheses using Fo and Fc as coefficients. Both syntheses will be subject to the same termination of series errors and the atomic coordinates will therefore be similarly affected in both cases. In the case of the Fc synthesis, however, specific coordinates were used in the calculation and the positions derived from the syntheses would be unaltered but for the presence of termination of series errors. The corrected atomic coordinates may then be derived by subtracting the differences between the coordinates used to calculate the Fc synthesis and those derived from it from the coordinates derived from the Fo synthesis.

An important development of Fourier refinement is the use of the difference synthesis (Booth, 1948b; Cochran, 1951) which arose from the Bunn's error synthesis (Bunn, 1949). The difference synthesis consists of a Fourier synthesis in which the coefficients are (Fo - Fc). It has the important property that the topography of the map is characteristically flat when the proposed and actual structures match. Errors in atomic coordinates and thermal parameters may therefore be detected and corrected by use of the difference synthesis but its most common use is for locating hydrogen atoms and for checking that the refinement of a structure is complete.

1.7.2 The Method of Least-squares.

Since the refinement of a structure consists of varying the atomic parameters in some systematic manner so as to minimise some function of the differences between the observed and calculated structure amplitudes, the problem is amenable to solution by the least-squares method. The first application of this method to crystal structure analysis was in the case of melamine (Hughes, 1941). The function normally minimised is

$$M = \sum_{hkl} w(|Fo| - |Fc|)^2 = \sum w\Delta^2 \qquad (48)$$

where the summation is carried out over the independent structure amplitudes and w is a weight chosen to reflect the accuracy of the observations. Ideally, the appropriate weights are given by

$$w = 1/\sigma^2(hkl) \qquad (49)$$

where $\sigma^2(hkl)$ is the variance of the observed structure amplitude. Normally only relative weights which depend on some function of $|Fo|$ are used. The validity of the weighting scheme may be tested by examining the average $w\Delta^2$ when the observations are grouped in any systematic manner; the values in each group should be approximately the same.

If the parameters $p_1, p_2 \ldots p_n$ are those whose values are to be determined from Fc, then the condition that $dM/dp_j = 0$ must be satisfied for the function M to have a minimum value. That is

$$\sum w \Delta \frac{\partial |Fc|}{\partial p_j} = 0 . \qquad (50)$$

For values of p_j which approximate closely to the correct values, Δ may be expanded as a first-order Taylor series

$$\Delta(\underline{p} + \underline{e}) = \Delta(\underline{p}) - \sum_{i=1}^{n} e_i \frac{\partial |Fc|}{\partial p_i} \qquad (51)$$

where e_i is the required change in p_i and $\underline{p}$ and $\underline{e}$ represent the complete set of parameters and changes.

Substituting (51) in (50) yields the <u>normal equations</u>.

$$\sum_{i=1}^{n} \left\{ \sum_{hkl} w \frac{\partial |Fc|}{\partial p_i} . \frac{\partial |Fc|}{\partial p_j} \right\} e_i = \sum_{hkl} w \Delta \frac{\partial |Fc|}{\partial p_j} \qquad (52)$$

There are, therefore, n equations in n unknowns which must be solved for the corrections e_i to the individual parameters.

The normal equations are more conveniently represented, in matrix form, by

$$\sum_i a_{ij} . e_i = b_j \qquad (53)$$

where

$$a_{ij} = \sum_{hkl} w \frac{\partial |Fc|}{\partial p_i} . \frac{\partial |Fc|}{\partial p_j} \qquad (54)$$

and

$$b_j = \sum_{hkl} w \Delta \frac{\partial |Fc|}{\partial p_j}. \qquad (55)$$

The solution of (53) is given by

$$e_i = \sum (a^{-1})_{ij} . b_j \qquad (56)$$

where $(a^{-1})_{ij}$ is the inverse matrix to a_{ij}.

Limitation of the expansion of Δ as a Taylor series (51) to only two terms enforces the necessity of calculating several cycles of least-squares refinement before convergence is reached.

The course of the refinement may be followed by examining the value of $\sum w \Delta^2$ after each cycle, by checking that the residual has decreased or by using a modified residual defined by

$$R = \sum w \Delta^2 / \sum w |Fo|^2 . \qquad (57)$$

The main disadvantage of the modified residual is that it may be affected by changes in the weighting scheme.

In general the size of the matrix of the normal equations (53) which must be evaluated to give the changes in the parameters exceeds the available high speed store in all but the largest computers. For this reason the block-diagonal approximation, in which the elements of the matrix arising from correlations between the atoms are ignored, is normally employed. In some extreme cases, all off-diagonal elements of the matrix are ignored. Both approximations involve an increase in the number of cycles required to achieve convergence.

In the structure analyses described in the following chapters of this thesis, either the full matrix of the normal equations was used or a chain of 9 × 9 matrices corresponding to three positional and six anisotropic thermal parameters was employed.

1.8 Accuracy of Results.

At the conclusion of the refinement of a crystal structure, the agreement between the observed and calculated structure amplitudes becomes of minor importance. Interest now turns to the structure of the molecule and detailed consideration of its geometry. It is therefore necessary to have some assessment of the accuracy with which the atomic positions have been determined so that useful comparisons with theoretical predictions and other experimental observations may be made. The most convenient way of expressing the accuracy of the atomic positions is by deriving their estimated standard deviations (σ). When weights appropriate to the structure factor data have been applied in the least-squares refinement, the variance $\sigma^2(p_i)$ of a parameter p_i is given by

$$\sigma^2(p_i) = (a^{-1})_{ii} \sum w\Delta^2 / (m - n) \qquad (58)$$

where m is the number of observations, n is the number of parameters defining the structure and $(a^{-1})_{ii}$ is the diagonal element of the inverse matrix of the normal equations. Since $(a^{-1})_{ii}$ is evaluated in the course of the least-squares refinement, the standard deviations are easily derived. The covariance of the parameters p_i and p_j may also be estimated as

$$\mathrm{cov}(p_i, p_j) = (a^{-1})_{ij} \sum w \Delta^2 / (m - n)$$

$$= \sigma^2(p_i) . \sigma^2(p_j) . r_{ij} \qquad (59)$$

where r_{ij} is the correlation coefficient between the two parameters. By application of statistical methods and use of equations (58) and (59) it is possible to derive the estimated standard deviations of the individual measurements.

In order to decide whether an observed value $\underline{x}$ which has a standard deviation s, based on n degrees of freedom, differs significantly from a theoretical value p, it is necessary to apply a statistical significance test defined by

$$t_o = (\underline{x} - p) / s. \qquad (60)$$

This is a random variable having a Student distribution with n degrees of freedom. When n is large, as is normally the case in X-ray crystallography, the distribution is normal and $t_o = 2.6$ or 3.3 respectively for the 1% or 0.1% significance levels (Fisher and Yates, 1953). If $2.6 < t_o < 3.3$, the difference probably does not arise from random errors. The relevant equation for two experimentally observed values is

$$t = (l_1 - l_2)/(s_1^2 + s_2^2)^{1/2} \qquad (61)$$

where l_1 and l_2 are the individual measurements and s_1 and s_2

are their respective standard deviations (Cruickshank and Robertson, 1953).

If the agreement between a set of measurements and their expected values is under consideration, (for example, the planarity or non-planarity of a molecular feature), the χ^2 distribution may be used. If d_i is the deviation of the i^{th} member of a set of n atoms, then

$$\chi^2 = \sum_{i=1}^{n} d_i^2/\sigma^2 \qquad (62)$$

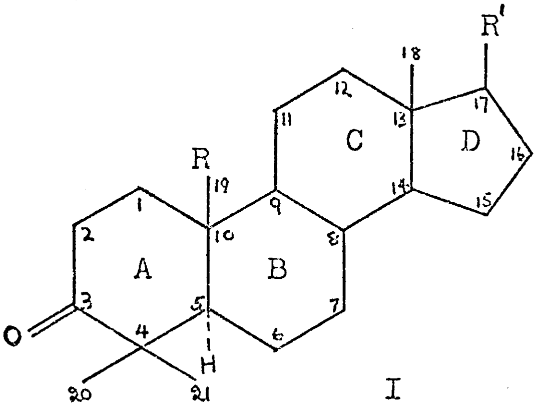
where σ is the average standard deviation in the positional parameters of the n atoms. The probability that the deviation is due to random errors may then be found from tables of the χ^2 distribution for n - 3 degrees of freedom (Fisher and Yates, 1953).

PART II

CRYSTAL STRUCTURE ANALYSES
OF
ANDROSTANE DERIVATIVES

Introduction.

The conformation of ring A in 3-keto-4,4-dimethyl-5α-steroids (I), which has been the subject of some controversy in the literature (Allinger et al., 1962, Lehn et al., 1962), is of interest because of the high degree of substitution in ring A which leads to a large number of steric interactions, and the widespread occurrence of such compounds in nature.

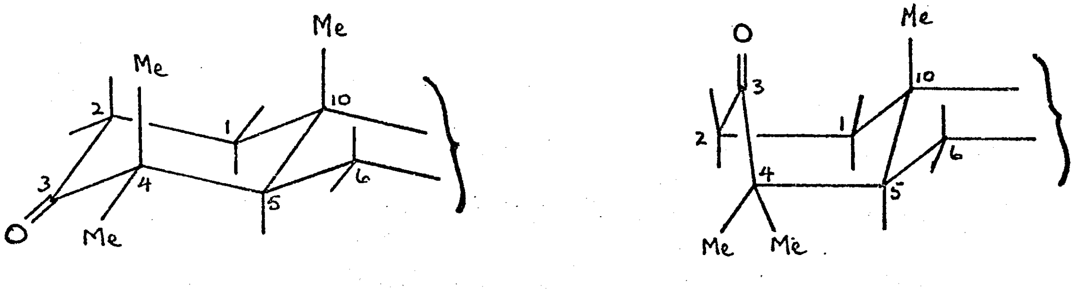


I

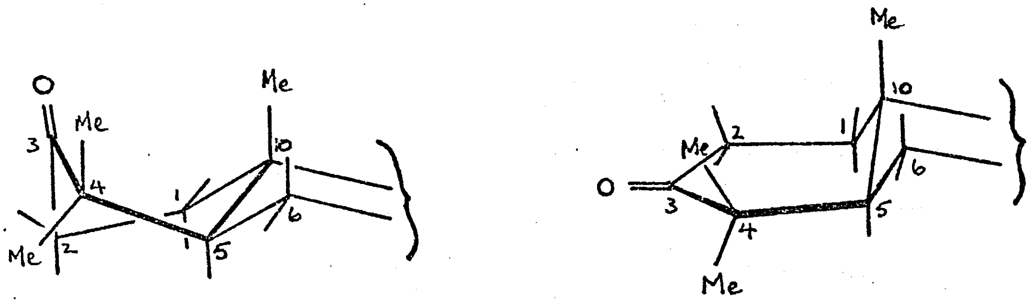
II R = Me R' = $OCOCH_2I$.

III R = H R' = $OCOCH_2I$.

In the chair conformation (fig. 2a) it is evident from an examination of molecular models that the most important conformation determining interaction is that between the axial methyl groups at C(4) and C(10). The enthalpy for such an interaction, 3.7 kcal./mole (Allinger and Miller, 1961), is greater than that required for chair ⟶ boat transformation of cyclohexanone (2.8 kcal./mole, Allinger and Blatter, 1961). The classical boat conformation (fig. 2b), on the other hand, suffers from a bowsprit interaction between the carbonyl group and the C(10) methyl group in addition to the increased interaction between one of the C(4) methyl groups and the hydrogen atoms of the C(6) methylene group.



(a) chair (b) classical boat



(c) skewed boat (d) flattened chair

Figure 2.

The lack of agreement between the predicted and observed Cotton effect in the optical rotatory dispersion curves for 3-keto-4,4-dimethyl-5α-steroids (Djerassi, 1960) has been rationalised by the assumption of a skewed boat conformation (fig. 2c) for ring A (Holker and Whalley, 1961).

From dipole moment studies of 3-keto-4,4-dimethyl-5α-steroids and triterpenoids, Lehn, Levisalles and Ourisson (1962) concluded that, in solution, a mixture of conformers, comprising 70 % chair and 30 % boat conformations, is present. They could not, however, exclude the possibility of a single conformation with any certainty. A systematic study of dipole moments has also been carried out by Allinger and Da Rooge (1962) and their interpretation of the results is in favour of a flattened chair conformation (fig. 2d) in which the interaction between the axial methyl groups at C(4) and C(10) has been reduced by rotation about the C(4)-C(5) bond. The net effect of such a rotation is to move C(3) towards the plane of the atoms C(1),C(2),C(4) and C(5), but the rotation would be insufficient to cause chair → boat transformation. Such a flattened chair conformation for ring A can also explain the apparently anomalous optical rotatory dispersion curves (Allinger and Da Rooge, 1962).

Comparison of the solid state and solution infra-red spectra of 3-keto-4,4-dimethyl-5α-steroids and triterpenoids has also led Allinger and Da Rooge (1962) to propose that the same (flattened chair) conformation is adopted by ring A in both phases.

To obtain quantitative information about the conformation of ring A in 3-keto-4,4-dimethyl-5α-steroids, the crystal structure analyses of 3-keto-4,4-dimethyl-5α-androstane-17β-iodoacetate and its 19-nor analogue have been completed. Molecular geometry calculations establish that , in the solid state, ring A adopts a partially flattened chair conformation in the former case and an undistorted chair conformation in the latter.

CHAPTER 1

The crystal and molecular structure of 3-keto-4,4-dimethyl-5α-androstane-17β-iodoacetate.

1.1 Experimental.

Crystal Data

3-keto-4,4-dimethyl-5α-androstane-17β-iodoacetate.

$C_{23}H_{35}O_3I$, F.W. = 486.4.

Orthorhombic, a = 14.96 ± 2 Å, b = 19.53 ± 2 Å, c = 7.47 ± 3 Å.

U = 2183 Å^3, F(000) = 1000 electrons,

D_m = 1.50 gm.cm.$^{-3}$, Z = 4, D_x = 1.48 gm.cm.$^{-3}$,

Space group, $P2_12_12_1$ (D_2^4, No. 19).

Linear absorption coefficient (Mo Kα, λ = 0.7107 Å) = 16 cm.$^{-1}$

The crystals used in this analysis were supplied by Professor W.B. Whalley. They consisted of colourless needles elongated along the c axis. The cell dimensions were found from Weissenberg photographs of the hk0 reciprocal lattice net using Cu Kα radiation (λ = 1.5418 Å) and from precession photographs of the h0l and hk0 reciprocal lattice nets taken with Mo Kα radiation (λ = 0.7107 Å). The space group was uniquely determined as $P2_12_12_1$ (D_2^4) from the systematically absent spectra.

Three-dimensional intensity data were collected using a Hilger and Watts Y-190 linear diffractometer (see appendix I). The crystal was rotated about its c-axis and the intensities from the reciprocal lattice nets hk0-hk7 obtained using Mo Kα radiation; allowance was made for background radiation using balanced strontium oxide / zirconium oxide Ross filters (Ross, 1926). A one minute oscillation of 3° was employed and each reflection measured a minimum of twice with each filter. The maximum reflecting angle permitted was $\theta = 30^\circ$.

The measurements made on the diffractometer were processed using a suite of programs (Sime, 1965) which corrected the intensities for background radiation before reducing them to structure amplitudes by application of the appropriate Lorentz, polarisation and rotation factors (Tunell, 1939). No absorption correction was applied as the crystal was almost transparent to Mo Kα radiation ($\mu = 16\mathrm{cm.}^{-1}$). A total of 2089 independent structure amplitudes were obtained, which represents 85 % of the data which would be available to Cu Kα radiation.

The structure amplitudes were placed on approximately absolute scale by ensuring that $k\sum|Fo| = \sum|Fc|$ in the first structure factor calculation. The final scale was determined by the least-squares refinement.

1.2 Solution of the Structure.

The Harker sections of the three-dimensional Patterson function were calculated and examination showed the presence of only one major peak on each section (figure 1.1), corresponding to the iodine-iodine vectors arising from iodine atoms related by the two-fold screw axes. The coordinates of these peaks were derived using the interpolation formulae of Booth (1948a) and led to the fractional coordinates of the iodine atom (0.291, -0.050, 0.299).

The phase angles calculated with these coordinates were used in conjunction with the observed structure amplitudes to evaluate the first electron-density distribution which revealed the entire structure. The coordinates of the twenty-seven non-hydrogen atoms (i.e. 1 iodine, 23 carbon and 3 oxygen atoms) were used to calculate a second set of structure factors for which the residual R $(= \sum||Fo| - |Fc||/\sum|Fc|)$ was 0.265 over all independent structure amplitudes. The different chemical species present in the structure were differentiated on the basis of peak height and knowledge of the chemical structure. The atomic scattering factors used in the second structure factor calculation (and throughout the remainder of the analysis) were those of Hoerni and Ibers (1954) for carbon and oxygen and those of Thomas and Umeda

(1957) for iodine. A second electron-density distribution was evaluated using the improved phases and the observed structure amplitudes. The improved coordinates derived from this distribution reduced the residual to 0.257 in the next structure factor calculation.

1.3 Least-squares Refinement.

The atomic parameters were refined in eight cycles of least-squares minimisation of the function $M = \sum w(|Fo| - |Fc|)^2$. The block-diagonal approximation to the normal matrix was employed throughout the refinement. As no significant deterioration of the crystal was observed in the course of the data collection, only one overall scale factor was refined.

In the first two cycles, three positional and an isotropic thermal parameter were varied for each atom; all observations were given unit weight. These cycles reduced the residual R from 0.257 to 0.209. A weighting scheme of the form

$$w = 1 / [p_1 + |Fo| + p_2 |Fo|^2]$$

where $p_1 = 2|Fo|_{min}$ and $p_2 = 2/|Fo|_{max}$, which has been recommended by Cruickshank (1961), was then applied and a further two cycles of least-squares refinement calculated.

The residual was reduced to 0.190 but examination of a bivariate analysis of $\langle w\Delta^2\rangle$ by magnitude of $|Fo|$ and $\sin^2\theta/\lambda^2$ indicated that the weighting scheme was unsuitable; there was a significant increase in $\langle w\Delta^2\rangle$ with increasing $\sin^2\theta/\lambda^2$. The weighting scheme was altered to one of the form

$$w = k[\exp-(A + B|Fo| + C|Fo|^2 + D/s + E/s^2 + Fs)]$$

where k is an arbitrary scale factor and $s = \sin^2\theta/\lambda^2$. The coefficients in the expression were determined by the least-squares method using a program written by Dr. D.R. McGregor (1967). The final values were $A = 2.55$, $B = 3.85 \times 10^{-2}$, $C = -4.34 \times 10^{-4}$, $D = 8.35 \times 10^{-3}$, $E = -7.15 \times 10^{-6}$ and $F = -3.72 \times 10^{-1}$ which gave rise to a weighting scheme in which different batches of $\langle w\Delta^2\rangle$ by $|Fo|$ and $\sin^2\theta/\lambda^2$ did not differ by more than 15 %.

The refinement converged in a further four cycles of calculation in which three positional and six anisotropic thermal parameters were refined for each atom. The residual was reduced to 0.142 and no shifts of greater than $0.2\,\sigma$ in coordinates or thermal parameters were observed in the final cycle. The course of the refinement is summarised in table 1.1.

The structure factors calculated using the atomic parameters from the final cycle of refinement were used to phase electron-density and difference syntheses. The electron-density synthesis is shown by means of a superimposed contour diagram in figure 1.2 which also explains the numbering system adopted for the molecule. The estimated standard deviation of the electron density, calculated using the approximate formula of Cruickshank (1949), was 0.24 e/Å^3 and no peaks of greater than three times this value were found in the difference synthesis so the refinement was regarded as being complete. A low order difference map, calculated using those planes for which $\sin \theta / \lambda \leq 0.35$, showed diffuse positive regions in some of the expected hydrogen positions but it was not possible to determine the coordinates with any accuracy. No allowance was therefore made for the hydrogen contribution to the scattering in the analysis.

The atomic coordinates (fractional and in Ångstroms referred to the crystal axes) and the anisotropic thermal parameters of the structural model are given in tables 1.2, 1.6 and 1.3 respectively. The observed and final calculated structure amplitudes are given in table 1.4; an analysis of the structure factor data as a function of layer line index and magnitude of $|Fo|$ is contained in table 1.5. Bond lengths and angles are given in tables 1.7 and 1.8; some intramolecular and all intermolecular distances less than

4.0 Ångstroms are contained in tables 1.9 and 1.10 respectively. Deviations of atoms from selected mean planes through portions of the molecular framework are given in table 1.11.

Estimated standard deviations, where quoted, are in units of the last decimal place of the quantity to which they refer.

TABLES AND DIAGRAMS

TABLES AND DIAGRAMS

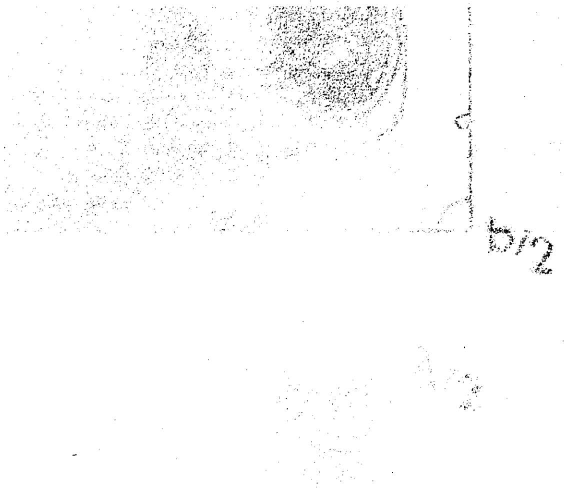
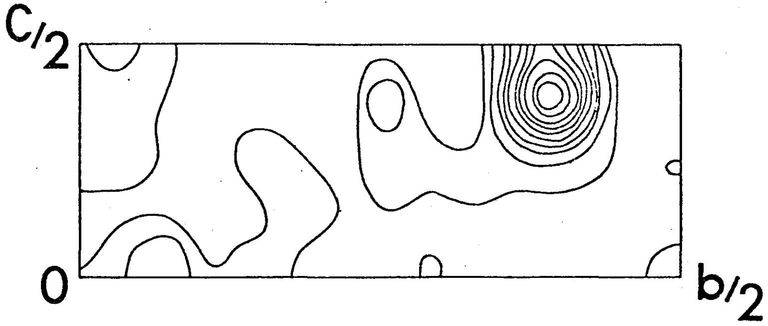


Figure 1.1

Harker sections at u =1/2, v = 1/2 and w = 1/2.

Contours are at arbitrary levels.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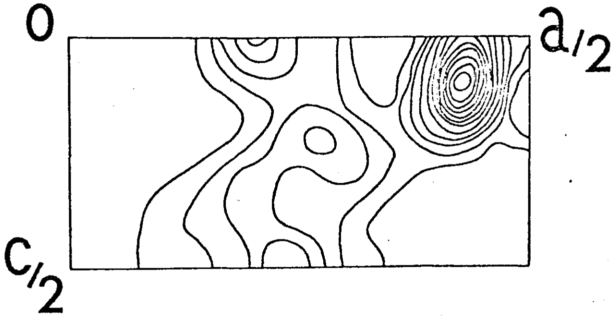
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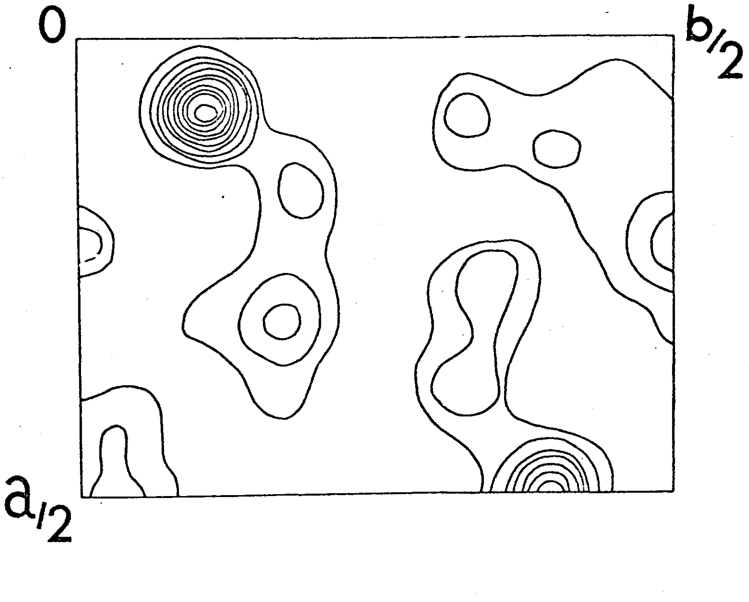
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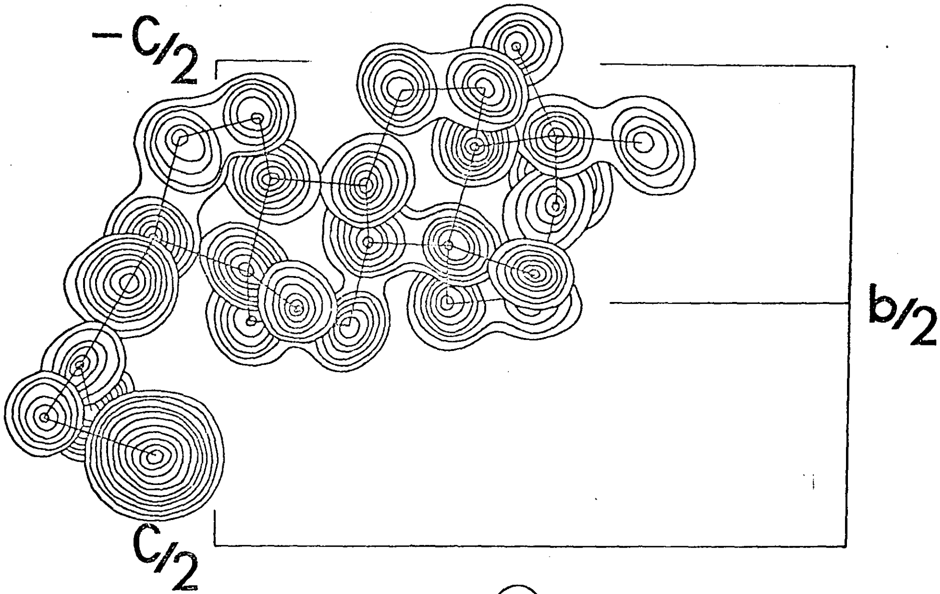
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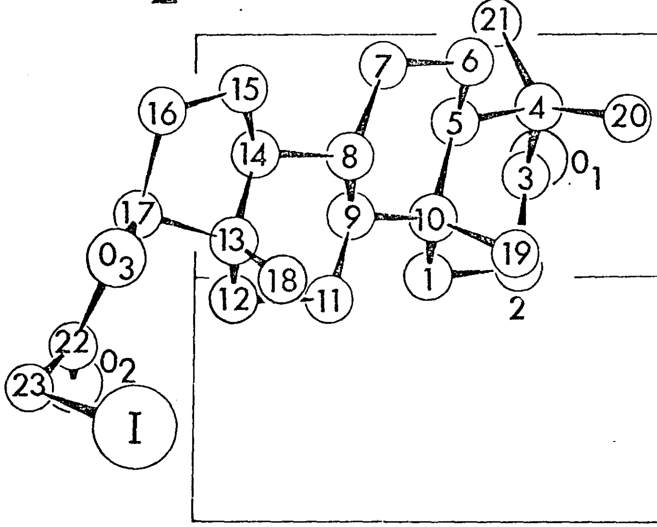
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8
0₁
3
17
9
10
13
19
0₃
18
12
11
1
2
22
0₂
23
I

Figure 1.3

The molecular packing as viewed down c.

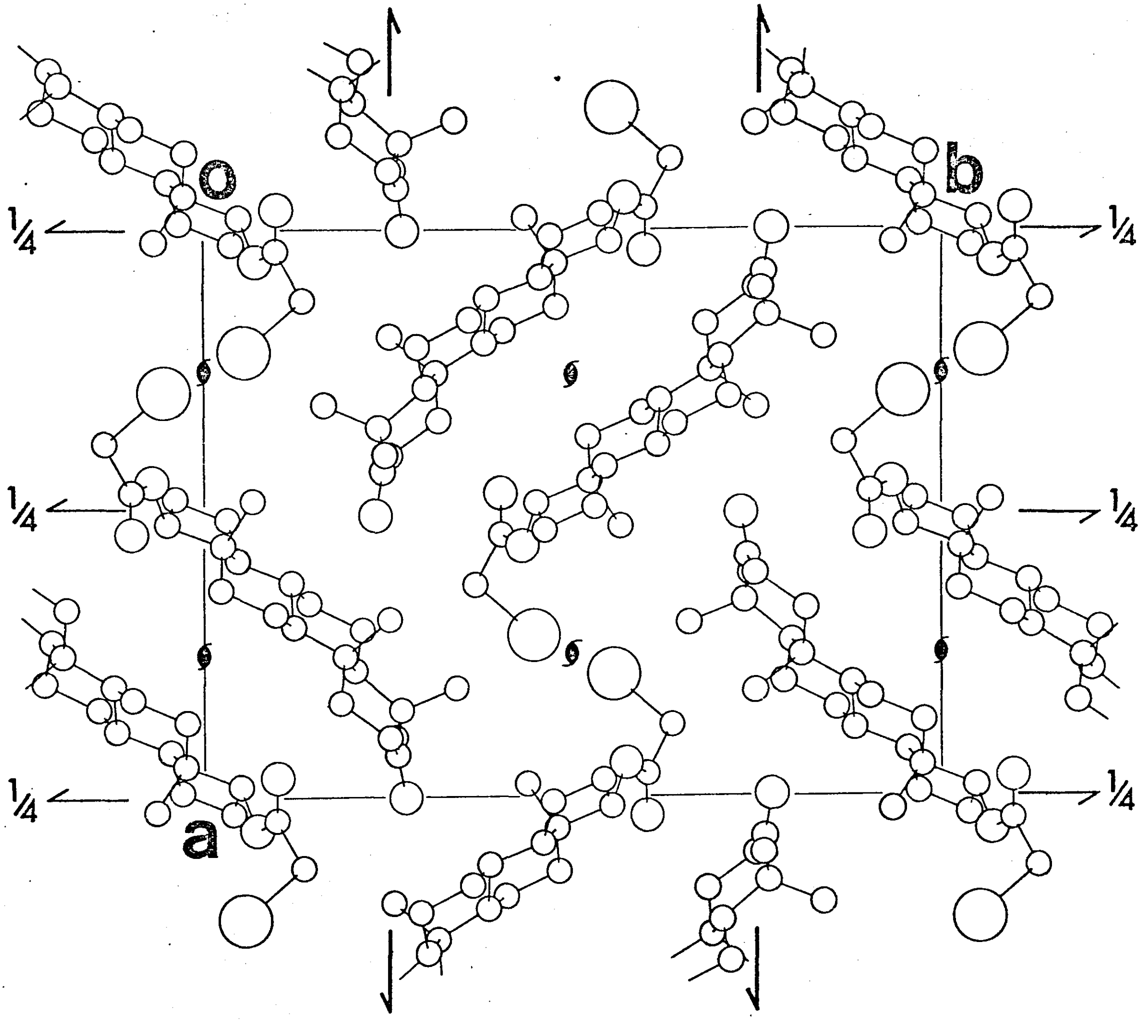
o
b
a
1/4
1/4
1/4
1/4
1/4
1/4

TABLE 1.1

COURSE OF ANALYSIS

(I) Fourier refinement.

	Atoms included in structure factor calculation	R
1	Iodine only	0.453
2	I + 23 C + 3 O	0.265
3	I + 23 C + 3 O	0.259

(II) Least-squares refinement.

Cycles	Final R	Final R$'$	*
1 - 2	0.209	0.050	(a)
3 - 5	0.178	0.031	(b)
6 - 9	0.142	0.028	(c)

*(a) Block-diagonal, U_{iso}, unit weights.

(b) As above, weighting scheme 1 applied.

(c) All atoms anisotropic, weighting scheme 2 applied.

TABLE 1.2

FRACTIONAL COORDINATES AND E.S.D.S.

ATOM	X/a	Y/b	Z/c
C(1)	0.8372 ± 14	0.1861 ± 10	-0.0151 ± 31
C(2)	0.9025 ± 17	0.2462 ± 13	-0.0245 ± 41
C(3)	0.9253 ± 16	0.2615 ± 15	-0.2085 ± 44
C(4)	0.8513 ± 14	0.2689 ± 8	-0.3532 ± 28
C(5)	0.7840 ± 12	0.2055 ± 8	-0.3324 ± 28
C(6)	0.7063 ± 16	0.2082 ± 11	-0.4533 ± 33
C(7)	0.6576 ± 13	0.1402 ± 10	-0.4561 ± 31
C(8)	0.6281 ± 12	0.1194 ± 9	-0.2598 ± 29
C(9)	0.7059 ± 13	0.1195 ± 10	-0.1360 ± 26
C(10)	0.7537 ± 14	0.1905 ± 10	-0.1338 ± 31
C(11)	0.6789 ± 15	0.0933 ± 9	0.0529 ± 27
C(12)	0.6345 ± 14	0.0220 ± 11	0.0458 ± 28
C(13)	0.5547 ± 13	0.0235 ± 9	-0.0802 ± 33
C(14)	0.5879 ± 11	0.0439 ± 9	-0.2656 ± 29
C(15)	0.5096 ± 17	0.0280 ± 11	-0.3951 ± 40
C(16)	0.4763 ± 21	-0.0450 ± 16	-0.3163 ± 39
C(17)	0.5182 ± 11	-0.0495 ± 9	-0.1306 ± 27
C(18)	0.4776 ± 14	0.0657 ± 9	0.0002 ± 37
C(19)	0.6897 ± 17	0.2476 ± 11	-0.0561 ± 31
C(20)	0.8107 ± 17	0.3408 ± 13	-0.3323 ± 37

C(21)	0.8888 ± 19	0.2640 ± 13	-0.5393 ± 41
C(22)	0.4629 ± 18	-0.0975 ± 14	0.1464 ± 52
C(23)	0.3795 ± 15	-0.1322 ± 12	0.2317 ± 35
I(1)	0.2875 ± 1	-0.0533 ± 1	0.3043 ± 3
O(1)	1.0035 ± 13	0.2696 ± 9	-0.2577 ± 32
O(2)	0.5364 ± 10	-0.0962 ± 9	0.2112 ± 30
O(3)	0.4446 ± 12	-0.0732 ± 10	-0.0115 ± 28

TABLE 1.3

ANISOTROPIC TEMPERATURE FACTORS AND E.S.D.S.

ATOM	U11	U22	U33	2U23	2U31	2U12
C(1)	0.0486	0.0488	0.0520	0.0182	0.0075	-0.0063
	106	95	128	188	228	166
C(2)	0.0651	0.0652	0.0719	0.0050	-0.0221	-0.0188
	142	125	174	250	308	228
C(3)	0.0532	0.0976	0.0762	0.0172	0.0289	-0.0377
	124	175	183	335	317	246
C(4)	0.0616	0.0312	0.0406	0.0052	0.0320	-0.0228
	114	71	112	145	213	148
C(5)	0.0325	0.0434	0.0488	0.0259	-0.0002	-0.0128
	76	78	114	160	210	138
C(6)	0.0525	0.0555	0.0649	0.0117	-0.0283	0.0174
	113	104	146	206	268	195
C(7)	0.0467	0.0462	0.0517	0.0151	-0.0030	0.0146
	102	91	127	180	216	160
C(8)	0.0373	0.0487	0.0491	-0.0080	0.0058	-0.0275
	84	87	125	166	194	151
C(9)	0.0402	0.0619	0.0331	0.0107	-0.0090	-0.0380
	90	102	100	163	196	177
C(10)	0.0563	0.0504	0.0479	0.0231	0.0440	0.0009
	112	96	125	182	221	174
C(11)	0.0627	0.0421	0.0327	0.0077	-0.0053	-0.0286
	119	85	102	152	206	169
C(12)	0.0498	0.0709	0.0299	0.0316	0.0107	0.0227
	107	119	109	186	207	197
C(13)	0.0448	0.0384	0.0680	0.0134	0.0320	0.0067
	97	82	147	188	233	154

C(14)	0.0340	0.0469	0.0536	0.0082	0.0025	-0.0074
	78	87	121	176	190	142
C(15)	0.0671	0.0541	0.0839	0.0093	-0.0380	-0.0551
	138	110	178	240	306	212
C(16)	0.0989	0.0958	0.0583	-0.0115	-0.0346	-0.0138
	200	184	157	333	339	330
C(17)	0.0301	0.0383	0.0532	-0.0107	0.0379	-0.0026
	75	74	114	168	174	135
C(18)	0.0555	0.0368	0.0822	0.0109	0.0372	0.0426
	113	85	164	199	262	165
C(19)	0.0724	0.0543	0.0476	-0.0109	0.0469	-0.0005
	149	104	134	195	257	203
C(20)	0.0652	0.0796	0.0584	0.0084	0.0089	-0.0220
	138	144	150	252	290	233
C(21)	0.0779	0.0716	0.0745	-0.0270	0.0444	-0.0578
	166	146	189	274	330	261
C(22)	0.0666	0.0729	0.1167	0.0639	0.0371	0.0310
	146	146	261	349	370	246
C(23)	0.0533	0.0669	0.0622	0.0174	0.0313	-0.0244
	110	121	152	226	251	201
I(1)	0.0688	0.0898	0.1014	0.0139	0.0226	0.0006
	9	10	14	23	23	18
O(1)	0.0669	0.0810	0.0927	0.0198	0.0230	-0.0201
	95	106	141	214	219	177
O(2)	0.0537	0.0796	0.0850	0.0069	-0.0044	0.0260
	82	100	127	214	211	153
O(3)	0.0614	0.0914	0.0780	0.0132	0.0102	-0.0219
	96	120	125	209	211	180

TABLE 1.4

FINAL OBSERVED AND CALCULATED STRUCTURE AMPLITUDES.

H	K	L	Fo	Fc
0	24	0	6.9	4.5
0	22	0	14.4	5.1
0	20	0	17.1	17.7
0	18	0	14.4	13.4
0	16	0	22.5	25.6
0	14	0	17.4	20.4
0	12	0	61.7	59.3
0	10	0	117.7	116.1
0	6	0	175.1	167.0
0	4	0	110.1	101.6
0	2	0	102.6	117.1
1	24	0	8.4	8.0
1	23	0	4.7	1.0
1	22	0	4.6	3.7
1	21	0	14.0	9.0
1	20	0	7.4	0.5
1	19	0	24.5	20.3
1	18	0	22.2	15.6
1	17	0	31.3	38.3
1	16	0	5.2	5.9
1	15	0	7.1	2.0
1	14	0	31.7	33.3
1	13	0	6.6	15.7
1	12	0	9.4	0.6
1	11	0	91.1	89.5
1	10	0	72.2	65.8
1	9	0	144.1	129.6
1	7	0	86.7	84.4
1	6	0	17.7	13.1
1	5	0	48.5	45.6
1	4	0	71.3	80.7
1	3	0	142.1	139.8
1	2	0	31.8	39.1
1	1	0	91.5	103.5
2	22	0	4.6	3.2
2	21	0	8.9	3.1
2	20	0	19.2	18.3
2	19	0	7.2	1.9
2	18	0	15.5	17.5
2	17	0	5.5	2.1
2	16	0	27.7	28.2
2	14	0	3.4	7.7
2	13	0	34.0	36.7
2	12	0	50.7	49.8
2	11	0	19.6	15.6
2	10	0	41.2	47.2
2	9	0	44.9	31.2
2	8	0	94.3	92.3
2	7	0	50.7	52.6
2	6	0	30.0	21.7
2	5	0	50.9	48.0
2	4	0	25.3	13.1
2	3	0	248.1	221.7
2	2	0	26.9	4.9
2	1	0	21.0	16.0
2	0	0	97.1	95.3
3	23	0	4.8	2.9
3	22	0	4.6	3.0
3	21	0	7.7	4.9
3	20	0	6.1	7.4
3	19	0	20.9	16.3
3	18	0	5.7	7.5
3	17	0	10.3	14.6
3	16	0	10.0	7.1
3	15	0	3.6	4.4
3	14	0	41.6	41.6
3	13	0	23.7	25.7
3	12	0	29.6	30.7
3	11	0	55.1	52.6
3	10	0	28.1	20.7
3	9	0	98.5	94.0
3	8	0	61.0	52.3
3	7	0	8.3	12.4
3	6	0	123.1	121.9
3	5	0	13.8	17.7
3	4	0	54.9	51.3
3	3	0	139.4	142.3
3	2	0	32.5	25.3
3	1	0	128.6	132.8
4	24	0	7.0	0.6
4	23	0	4.8	5.7
4	22	0	4.7	2.6
4	21	0	10.1	11.7
4	20	0	7.6	13.7
4	19	0	4.2	4.5
4	18	0	13.0	16.5
4	17	0	11.9	9.8
4	16	0	8.5	4.5
4	15	0	44.1	40.9
4	14	0	3.6	2.7
4	13	0	42.9	40.3
4	12	0	30.2	30.4
4	11	0	37.5	41.4
4	10	0	21.9	28.6
4	9	0	21.6	23.5
4	8	0	130.2	114.6
4	6	0	59.1	51.2
4	5	0	144.7	122.2
4	4	0	26.7	31.3
4	3	0	78.8	84.0
4	2	0	126.9	114.5
4	1	0	83.3	74.3
4	0	0	129.5	129.3
5	22	0	8.2	10.8
5	21	0	7.9	3.1
5	20	0	4.4	2.6
5	19	0	6.1	7.2
5	18	0	10.2	10.8
5	17	0	7.0	9.9
5	16	0	31.8	33.8
5	15	0	12.4	15.2
5	14	0	52.2	52.9
5	13	0	4.9	4.0
5	12	0	29.3	30.5
5	11	0	11.2	14.1
5	10	0	12.3	17.3
5	9	0	52.1	50.9
5	8	0	56.2	49.0
5	7	0	20.6	25.7
5	6	0	139.5	126.5
5	5	0	51.3	46.9
5	4	0	145.6	145.7
5	3	0	32.4	33.6
5	2	0	62.2	67.5
5	1	0	90.1	76.2
6	23	0	8.5	6.1
6	22	0	8.3	3.8
6	21	0	11.3	4.7
6	20	0	11.9	4.5
6	19	0	4.4	1.1
6	18	0	4.2	0.6
6	17	0	8.2	11.3
6	16	0	6.9	10.1
6	15	0	31.6	29.5
6	14	0	8.3	1.7
6	13	0	27.8	26.7
6	12	0	3.5	5.5
6	11	0	37.7	38.5
6	10	0	3.2	8.7
6	9	0	10.3	12.2
6	8	0	22.3	15.3
6	7	0	43.0	51.7
6	6	0	5.6	6.2
6	5	0	159.7	157.4
6	3	0	74.8	74.9
6	2	0	11.3	9.5
6	1	0	57.7	57.0
6	0	0	35.5	31.2
7	22	0	6.3	7.6
7	21	0	6.6	4.5
7	20	0	6.5	4.6
7	19	0	6.3	6.4
7	18	0	8.6	2.6
7	17	0	8.4	7.5
7	16	0	9.9	14.7
7	15	0	9.6	6.0
7	14	0	24.1	24.0
7	13	0	3.7	2.1
7	12	0	45.5	48.3
7	11	0	7.7	3.3
7	10	0	3.4	5.7
7	9	0	30.9	27.1
7	8	0	9.4	15.0
7	7	0	19.3	15.5
7	6	0	98.3	94.7
7	5	0	2.9	3.0
7	4	0	104.7	104.5
7	3	0	12.0	15.3
7	2	0	82.3	79.9
7	1	0	2.7	7.0
8	22	0	9.8	1.6
8	21	0	11.7	2.4
8	20	0	4.7	9.3
8	19	0	6.4	7.4
8	18	0	4.4	1.1
8	17	0	14.2	13.6
8	16	0	5.9	1.6
8	15	0	35.8	37.3
8	14	0	3.9	0.2
8	13	0	47.3	47.1
8	12	0	3.7	3.2
8	11	0	25.3	26.1
8	10	0	13.9	11.6
8	9	0	6.8	7.9
8	8	0	27.2	28.3
8	7	0	64.2	63.7
8	6	0	18.3	19.4
8	5	0	79.0	84.5
8	4	0	3.0	6.8
8	3	0	106.7	98.8
8	2	0	25.5	27.6
8	1	0	35.1	27.4
8	0	0	28.3	31.5
9	20	0	4.7	3.0
9	19	0	11.3	5.0
9	18	0	11.9	10.1
9	17	0	15.2	6.7
9	16	0	10.4	8.5
9	15	0	8.3	1.0
9	14	0	27.9	26.9
9	13	0	3.9	9.9
9	12	0	29.3	29.0
9	11	0	9.1	10.7
9	10	0	8.1	3.9
9	9	0	32.4	30.8
9	8	0	35.5	41.2
9	7	0	3.4	4.8
9	6	0	53.7	50.0
9	5	0	8.6	3.1
9	4	0	64.5	58.4
9	3	0	25.3	20.7
9	2	0	33.8	36.4
9	1	0	50.6	47.3
10	20	0	6.9	9.7
10	19	0	9.4	7.2
10	18	0	8.0	10.1
10	17	0	6.3	0.7
10	16	0	12.4	8.9
10	15	0	9.5	15.0
10	14	0	7.2	1.0
10	13	0	12.8	11.9
10	12	0	23.7	22.1
10	11	0	21.5	3.4
10	10	0	50.0	46.1
10	9	0	12.8	17.6
10	8	0	24.7	24.9
10	7	0	30.6	24.7
10	6	0	18.7	27.0
10	5	0	31.7	28.0
10	4	0	24.3	30.3
10	3	0	37.4	33.3
10	2	0	21.4	20.3
10	1	0	25.0	20.0
10	0	0	64.2	61.6
11	19	0	4.8	2.5
11	18	0	11.6	9.8
11	17	0	10.3	7.0
11	16	0	4.5	5.5
11	15	0	4.4	7.0
11	14	0	12.9	7.6
11	13	0	10.3	5.6
11	12	0	13.0	15.0
11	11	0	22.3	18.8
11	10	0	14.7	12.9
11	9	0	51.0	50.2
11	8	0	3.8	2.9
11	7	0	18.5	18.7
11	6	0	28.7	29.0
11	5	0	13.4	3.1
11	4	0	34.2	35.0
11	3	0	24.6	23.4
11	2	0	39.8	41.1
11	1	0	55.4	51.2
12	18	0	10.8	11.1
12	17	0	6.7	2.3
12	16	0	4.6	8.1
12	15	0	16.3	6.4
12	14	0	13.3	5.0
12	13	0	12.3	8.3
12	12	0	25.5	20.3
12	11	0	5.9	9.7
12	10	0	12.2	13.8
12	9	0	5.7	4.1
12	8	0	23.6	21.3
12	7	0	12.8	15.1
12	6	0	20.2	22.1
12	5	0	22.6	16.8
12	4	0	3.7	6.7
12	3	0	32.3	32.5
12	2	0	56.0	49.8
12	1	0	13.7	3.4
12	0	0	61.6	61.2
13	15	0	4.7	0.6
13	14	0	6.5	7.3
13	13	0	9.0	5.0
13	12	0	4.4	2.6
13	11	0	19.8	19.7
13	10	0	6.0	7.0
13	9	0	39.5	38.4
13	8	0	10.1	4.9
13	7	0	35.0	27.8
13	6	0	12.6	4.7
13	5	0	4.0	7.2
13	4	0	7.8	7.4
13	3	0	24.0	23.4
13	2	0	6.7	3.6
13	1	0	48.0	42.3
14	15	0	4.8	6.5
14	14	0	9.4	2.6
14	13	0	11.4	4.0
14	12	0	17.6	21.3
14	11	0	9.0	3.1
14	10	0	27.1	20.5
14	9	0	4.3	1.4
14	8	0	19.1	19.2
14	7	0	4.2	0.5
14	6	0	13.8	10.6
14	5	0	10.1	11.2
14	4	0	16.9	18.5
14	3	0	14.1	8.5
14	2	0	23.3	22.5
14	1	0	15.6	16.8
14	0	0	43.3	38.9
15	14	0	15.4	6.2
15	13	0	4.8	1.6
15	12	0	13.3	11.2
15	11	0	4.6	4.2
15	10	0	11.2	8.4
15	9	0	15.6	14.4
15	8	0	7.7	8.2
15	7	0	9.8	14.2
15	6	0	13.8	5.8
15	5	0	10.6	4.5
15	4	0	9.6	12.3
15	3	0	15.9	17.4
15	2	0	4.2	4.4
15	1	0	22.0	17.2
16	10	0	14.2	9.6
16	9	0	4.7	2.1
16	8	0	22.2	17.9
16	7	0	7.9	7.7
16	6	0	16.4	9.6
16	5	0	14.2	9.5
16	4	0	4.5	4.1
16	3	0	13.3	7.8
16	2	0	12.5	13.3
16	1	0	13.3	6.1
16	0	0	20.7	20.3
17	8	0	10.8	2.8
17	7	0	4.8	5.3
17	6	0	4.7	10.3
17	5	0	4.7	3.0
17	4	0	9.3	12.8
17	3	0	12.3	12.6
17	2	0	4.6	6.0
17	1	0	11.3	13.4
18	0	0	6.8	4.0
17	9	1	8.4	3.1
17	8	1	12.7	7.2
17	7	1	4.8	4.2
17	6	1	4.7	5.3
17	5	1	4.7	6.3
17	4	1	8.1	4.0
17	3	1	8.0	8.4
17	2	1	14.6	7.3
17	1	1	8.0	6.9
17	0	1	6.5	10.0
16	12	1	9.8	7.0
16	11	1	4.3	9.7
16	10	1	10.6	7.4
16	9	1	4.7	7.1
16	8	1	6.6	4.6
16	7	1	22.5	10.9
16	6	1	20.3	16.6
16	5	1	4.5	5.1
16	4	1	22.3	16.4
16	3	1	10.0	7.7
16	2	1	7.7	13.6
16	1	1	13.3	16.8
16	0	1	9.9	8.7
15	12	1	8.2	4.2
15	11	1	10.4	10.7
15	10	1	16.5	13.1
15	9	1	4.5	3.6
15	8	1	8.9	8.1
15	7	1	15.9	18.0
15	6	1	15.1	13.8
15	5	1	22.0	19.4
15	4	1	16.6	16.6
15	3	1	20.9	22.0
15	2	1	17.0	16.2
15	1	1	12.7	3.5
15	0	1	11.2	6.6
14	14	1	17.7	17.0
14	13	1	4.6	2.9
14	12	1	11.2	8.8
14	11	1	4.5	2.6
14	10	1	4.4	5.4
14	9	1	4.3	5.9
14	8	1	18.2	14.8
14	7	1	4.2	7.2
14	6	1	24.4	24.2
14	5	1	4.1	8.1
14	4	1	33.6	28.9
14	3	1	10.0	16.4
14	2	1	15.2	18.2
14	1	1	10.7	5.3
14	0	1	15.7	15.1
13	17	1	8.4	4.1
13	16	1	13.5	6.0
13	15	1	12.4	10.5
13	14	1	4.6	7.1
13	13	1	14.2	12.6
13	12	1	6.2	2.7
13	11	1	10.6	13.7
13	10	1	9.5	6.7
13	9	1	4.2	4.2
13	8	1	10.9	7.7
13	7	1	31.9	30.2
13	6	1	8.9	10.1
13	4	1	13.6	17.7
13	3	1	35.5	32.0
13	2	1	16.4	13.1
13	1	1	3.9	11.5
13	0	1	3.9	4.3
12	18	1	6.9	0.7
12	17	1	4.7	6.8
12	16	1	9.3	9.8
12	15	1	4.5	5.6
12	14	1	16.0	14.5
12	13	1	13.0	7.0
12	12	1	13.4	16.5
12	11	1	4.2	6.8
12	10	1	5.8	6.3
12	9	1	23.0	21.2
12	8	1	11.8	4.9
12	7	1	12.9	16.9
12	6	1	22.9	24.6
12	5	1	11.3	16.0
12	4	1	39.0	39.9
12	3	1	22.2	23.1
12	2	1	45.9	46.4
12	1	1	16.4	12.9
12	0	1	5.2	3.4
11	18	1	6.7	2.2
11	17	1	6.5	6.2
11	16	1	9.0	3.7
11	15	1	17.0	15.0
11	14	1	11.4	12.2
11	13	1	16.3	17.9
11	12	1	14.8	14.0
11	11	1	21.2	23.0
11	10	1	19.6	20.0
11	9	1	13.9	9.2
11	8	1	31.1	27.6
11	7	1	26.0	29.9
11	6	1	41.3	43.0
11	5	1	51.1	51.9
11	4	1	18.8	23.3
11	3	1	31.1	31.7
11	2	1	41.7	40.9
11	1	1	22.6	21.6
11	0	1	39.5	38.5
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10	18	1	4.6	4.1
10	17	1	11.9	12.5
10	16	1	7.6	10.0
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10	14	1	20.4	21.6
10	13	1	15.7	13.8
10	12	1	20.2	22.3
10	11	1	24.1	29.0
10	10	1	11.9	12.2
10	9	1	27.9	27.5
10	8	1	16.6	22.6
10	7	1	24.3	24.6
10	6	1	37.1	38.1
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10	4	1	45.8	44.9
10	3	1	35.9	35.3
10	2	1	25.4	24.0
10	1	1	45.3	48.7
10	0	1	19.4	22.4
9	20	1	14.3	6.9
9	19	1	12.2	4.0
9	18	1	11.0	6.9
9	17	1	4.4	1.7
9	16	1	12.1	11.2
9	15	1	7.2	11.3
9	14	1	12.8	10.4
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9	12	1	19.5	20.6
9	10	1	38.7	38.1
9	9	1	7.9	11.9
9	8	1	31.9	42.0
9	7	1	27.4	28.1
9	6	1	30.1	28.7
9	5	1	27.5	27.8
9	4	1	24.9	25.3
9	3	1	47.6	47.9
9	2	1	40.4	38.0
9	1	1	36.5	34.3
9	0	1	71.6	65.0
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8	19	1	16.9	12.3
8	18	1	7.6	3.5
8	17	1	16.0	16.1
8	16	1	16.1	14.6
8	15	1	9.9	9.2
8	14	1	5.5	7.1
8	13	1	17.9	16.3
8	12	1	10.5	15.5
8	11	1	38.0	38.6
8	10	1	13.1	14.5
8	9	1	62.2	61.9
8	8	1	25.3	23.0
8	7	1	42.5	45.1
8	6	1	41.5	41.5
8	5	1	33.0	31.0
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8	3	1	48.9	50.0
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8	1	1	99.9	98.9
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7	19	1	17.8	12.3
7	18	1	14.9	14.3
7	17	1	5.9	6.4
7	16	1	13.5	15.8
7	15	1	10.4	12.2
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7	12	1	29.1	32.4
7	11	1	14.3	12.2
7	10	1	56.0	55.7
7	9	1	29.3	30.0
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7	7	1	19.1	21.2
7	6	1	34.0	34.9
7	5	1	17.6	16.0
7	4	1	13.3	14.7
7	3	1	32.2	30.3
7	2	1	36.8	36.5
7	1	1	13.0	20.3
7	0	1	103.5	104.8
6	22	1	12.6	4.2
6	21	1	11.4	5.0
6	20	1	6.4	8.3
6	19	1	10.7	16.3
6	18	1	4.2	3.6
6	17	1	11.6	18.1
6	16	1	11.9	13.3
6	15	1	6.7	5.5
6	14	1	8.3	14.8
6	13	1	14.8	12.2
6	12	1	25.0	22.1
6	11	1	47.6	44.6
6	10	1	32.3	30.6
6	9	1	87.3	87.1
6	8	1	10.4	12.4
6	7	1	51.1	52.7
6	6	1	49.6	48.3
6	5	1	8.6	5.8
6	4	1	11.8	15.5
6	3	1	83.2	80.4
6	2	1	47.4	43.0
6	1	1	105.0	101.8
6	0	1	43.1	39.0
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5	21	1	4.6	2.1
5	20	1	8.9	12.6
5	19	1	4.3	7.5
5	18	1	22.4	21.1
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5	16	1	16.0	15.1
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5	14	1	3.6	2.3
5	13	1	20.1	25.9
5	12	1	24.3	27.3
5	11	1	32.8	30.6
5	10	1	50.7	50.8
5	9	1	33.2	35.7
5	8	1	86.2	83.4
5	7	1	13.8	14.8
5	6	1	62.9	62.3
5	5	1	54.5	44.7
5	4	1	18.5	21.3
5	3	1	42.1	39.6
5	2	1	75.7	76.2
5	1	1	40.7	39.5
5	0	1	153.7	143.5
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4	21	1	9.0	4.9
4	20	1	8.8	6.7
4	19	1	10.4	14.6
4	18	1	8.2	10.0
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4	16	1	20.2	9.9
4	15	1	12.8	12.1
4	14	1	11.2	12.9
4	13	1	23.9	23.3
4	12	1	12.3	9.8
4	11	1	45.2	49.5
4	10	1	41.9	39.4
4	9	1	68.2	66.1
4	8	1	13.2	16.2
4	7	1	42.2	50.3
4	6	1	103.7	100.4
4	5	1	35.0	33.9
4	4	1	69.4	64.1
4	3	1	41.6	36.0
4	2	1	58.3	56.9
4	1	1	121.7	114.3
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3	22	1	13.1	4.5
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3	20	1	4.3	8.9
3	19	1	11.9	4.0
3	18	1	4.1	12.5
3	17	1	21.1	20.7
3	16	1	13.1	8.6
3	15	1	34.5	32.8
3	14	1	3.5	2.0
3	13	1	24.2	29.7
3	12	1	35.8	37.2
3	11	1	40.5	41.8
3	10	1	71.1	72.5
3	9	1	51.7	45.7
3	8	1	93.7	90.9
3	7	1	71.6	71.2
3	6	1	62.4	58.1
3	5	1	153.1	146.8
3	4	1	52.9	63.6
3	3	1	127.5	121.2
3	2	1	133.2	122.6
3	1	1	74.6	70.9
3	0	1	133.3	110.3
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2	23	1	4.7	5.2
2	22	1	8.0	8.9
2	21	1	4.5	4.1
2	20	1	10.6	7.0
2	19	1	15.0	8.4
2	18	1	9.0	6.4
2	17	1	22.9	20.2
2	16	1	23.6	28.8
2	15	1	23.6	25.0
2	14	1	59.0	64.1
2	13	1	8.1	12.3
2	12	1	59.2	63.2
2	11	1	58.1	60.9
2	10	1	12.2	12.4
2	9	1	33.3	34.5
2	8	1	27.6	32.0
2	7	1	17.6	19.1
2	6	1	162.4	151.9
2	5	1	63.1	57.9
2	4	1	115.6	98.4
2	3	1	87.4	84.6
2	2	1	108.3	102.9
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1	24	1	4.9	5.3
1	23	1	8.2	10.5
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1	21	1	4.4	8.8
1	20	1	16.6	9.3
1	19	1	4.1	3.0
1	18	1	5.7	14.6
1	17	1	14.9	15.0
1	16	1	5.3	1.3
1	15	1	28.3	32.5
1	14	1	28.7	32.6
1	13	1	47.3	50.2
1	12	1	32.6	37.3
1	11	1	50.1	54.2
1	10	1	8.0	3.9
1	9	1	8.9	9.5
1	8	1	24.3	26.0
1	7	1	80.7	84.8
1	6	1	70.3	69.6
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0	21	1	4.4	6.2
0	20	1	8.6	7.8
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0	18	1	10.6	9.3
0	17	1	19.6	9.9
0	16	1	13.9	13.4
0	15	1	33.6	30.6
0	14	1	76.4	71.3
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1	16	2	7.5	9.9
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2	18	2	11.4	16.9
2	17	2	16.5	17.3
2	16	2	18.0	22.1
2	15	2	30.0	36.7
2	14	2	20.5	21.8
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2	2	2	127.1	113.4
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3	19	2	14.0	16.0
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8	1	2	40.1	39.8
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9	20	2	10.7	10.5
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9	12	2	25.2	24.7
9	11	2	3.7	9.2
9	10	2	20.3	20.7
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10	12	2	19.5	19.8
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10	0	2	44.3	45.7
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11	15	2	4.4	7.5
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11	13	2	9.4	8.7
11	12	2	9.2	12.0
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11	10	2	8.8	8.6
11	9	2	26.5	26.7
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16	0	2	9.9	11.3
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17	3	2	10.4	5.7
17	2	2	4.6	7.0
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17	0	2	16.7	9.4
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0	19	3	4.2	3.0
0	18	3	4.0	7.3
0	17	3	13.5	5.6
0	16	3	15.0	15.1
0	15	3	25.5	27.6
0	14	3	25.7	26.9
0	13	3	43.4	48.5
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0	11	3	21.5	22.2

H	K	L	Fo	Fc
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0	4	3	108.0	105.9
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1	23	3	4.8	3.5
1	22	3	10.4	5.5
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1	19	3	8.4	0.4
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1	17	3	5.5	7.9
1	16	3	14.6	16.1
1	15	3	10.2	16.6
1	14	3	24.3	28.5
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1	12	3	25.0	22.2
1	11	3	24.4	25.4
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2	20	3	15.7	5.1
2	19	3	4.2	1.6
2	18	3	11.5	3.8
2	17	3	11.1	13.5
2	16	3	9.3	14.9
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4	19	3	7.4	4.6
4	18	3	4.2	11.9
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7	20	3	4.6	4.5
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4	7	4	22.2	19.3
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7	1	6	15.9	16.4
7	2	6	5.0	4.1
7	3	6	15.3	21.7
7	4	6	11.6	13.6
7	5	6	22.5	21.9
7	6	6	15.7	16.6
7	7	6	7.2	10.9
7	8	6	6.7	4.6
7	9	6	4.9	3.8
7	10	6	11.0	3.9
0	1	7	2.9	3.7
0	2	7	3.0	0.9
0	3	7	5.7	5.0
0	4	7	1.9	0.6
0	5	7	29.6	28.4
0	6	7	17.3	14.6
0	7	7	2.5	6.3
0	8	7	7.2	2.2
0	9	7	5.0	4.0
0	10	7	13.3	4.0
0	11	7	14.1	9.2
0	12	7	11.1	11.9
0	13	7	3.5	0.3
1	0	7	3.9	9.5
1	1	7	2.1	6.3
1	2	7	9.4	15.3
1	3	7	15.3	20.5
1	4	7	3.4	6.1
1	5	7	20.6	16.2
1	6	7	12.2	10.6
1	7	7	12.5	3.4
1	8	7	2.7	2.9
1	9	7	4.1	3.0
1	10	7	3.1	1.4
1	11	7	5.6	4.3
1	12	7	4.5	6.5
1	13	7	3.5	1.9
2	0	7	1.5	4.7
2	1	7	7.5	5.5
2	2	7	10.2	12.5
2	3	7	5.0	7.5
2	4	7	11.2	11.5
2	5	7	12.6	13.0
2	6	7	22.2	16.7
2	7	7	2.6	2.9
2	8	7	13.0	11.4
2	9	7	14.4	9.2
2	10	7	3.1	2.7
2	11	7	14.2	9.6
2	12	7	10.2	5.2
2	13	7	3.6	4.3
3	0	7	16.1	17.2
3	1	7	1.9	2.0
3	2	7	11.9	12.3
3	3	7	9.7	10.1
3	4	7	5.7	6.0
3	5	7	3.4	7.1
3	6	7	10.2	7.5
3	7	7	3.3	3.4
3	8	7	19.0	10.3
3	9	7	3.0	1.1
3	10	7	3.2	1.3
3	11	7	3.3	6.0
3	12	7	4.9	2.5
3	13	7	8.1	5.3
4	0	7	2.2	3.5
4	1	7	15.5	15.3
4	2	7	14.4	3.7
4	3	7	16.0	12.5
4	4	7	12.7	9.5
4	5	7	6.3	4.7
4	6	7	15.6	7.5
4	7	7	7.5	3.6
4	8	7	15.2	6.6
4	9	7	19.5	12.2
4	10	7	13.8	3.8
4	11	7	25.0	11.9
4	12	7	3.5	1.6
4	13	7	11.5	7.6
5	0	7	10.3	11.5
5	1	7	16.1	12.5
5	2	7	13.1	13.3
5	3	7	2.6	3.9
5	4	7	15.4	11.6
5	5	7	10.3	4.1
5	6	7	2.9	2.4
5	7	7	3.0	3.2
5	8	7	16.1	10.0
5	9	7	3.2	5.1
5	10	7	15.3	5.4
5	11	7	14.3	9.1
5	12	7	3.6	3.6
5	13	7	10.6	3.3

TABLE 1.5

(A) ANALYSIS OF STRUCTURE FACTOR DATA AS A FUNCTION OF LAYER LINE INDEX. ALL FIGURES ARE ON ABSOLUTE SCALE.

l	$\Sigma\lvert F_o\rvert$	$\Sigma\lvert F_c\rvert$	$\Sigma\Delta$	N	R	$\Sigma\Delta/N$
0	9564	8991	1322	337	0.1382	3.92
1	9952	9685	1150	352	0.1156	3.27
2	8566	8744	974	349	0.1137	2.79
3	7059	7201	1027	347	0.1455	2.96
4	4536	4125	687	234	0.1514	2.93
5	3355	3229	565	243	0.1684	2.32
6	1466	1448	402	144	0.2745	2.79
7	794	645	285	83	0.3584	3.43
ALL	45292	44068	6412	2089	0.1416	3.07

(B) ANALYSIS OF STRUCTURE FACTOR DATA BY MAGNITUDE OF Fo.

0 - 13	7634	7172	2793	1021	0.3659	2.74
13 - 25	9409	8952	1681	523	0.1786	3.21
25 - 50	11611	11689	987	337	0.0850	2.92
50 - 75	7061	6974	439	120	0.0622	3.66
75 - 100	3730	3659	157	43	0.0422	3.66
100 - 250	5846	5620	353	45	0.0605	7.86

TABLE 1.6

ORTHOGONAL COORDINATES AND E.S.D.S.

ATOM	X	Y	Z
C(1)	12.525 ± 21	3.635 ± 19	-0.112 ± 24
C(2)	13.501 ± 26	4.808 ± 26	-0.183 ± 31
C(3)	13.843 ± 25	5.108 ± 29	-1.558 ± 33
C(4)	12.735 ± 21	5.252 ± 16	-2.638 ± 21
C(5)	11.728 ± 18	4.014 ± 16	-2.483 ± 21
C(6)	10.567 ± 23	4.067 ± 21	-3.387 ± 25
C(7)	9.838 ± 20	2.738 ± 19	-3.407 ± 23
C(8)	9.396 ± 18	2.333 ± 18	-1.941 ± 22
C(9)	10.560 ± 19	2.334 ± 19	-1.016 ± 20
C(10)	11.275 ± 22	3.720 ± 20	-0.999 ± 23
C(11)	10.156 ± 22	1.822 ± 18	0.395 ± 20
C(12)	9.492 ± 21	0.430 ± 22	0.342 ± 21
C(13)	8.299 ± 20	0.458 ± 18	-0.599 ± 25
C(14)	8.795 ± 17	0.858 ± 18	-1.984 ± 22
C(15)	7.623 ± 25	0.547 ± 22	-2.951 ± 30
C(16)	7.126 ± 32	-0.878 ± 31	-2.362 ± 29
C(17)	7.752 ± 16	-0.968 ± 17	-0.975 ± 20
C(18)	7.145 ± 21	1.282 ± 18	0.001 ± 27
C(19)	10.318 ± 25	4.835 ± 21	-0.419 ± 23
C(20)	12.128 ± 25	6.655 ± 26	-2.483 ± 27

C(21)	13.297 ± 29	5.157 ± 26	-4.029 ± 31
C(22)	6.925 ± 27	-1.903 ± 27	1.093 ± 39
C(23)	5.678 ± 22	-2.582 ± 23	1.731 ± 26
I(1)	4.300 ± 2	-1.042 ± 2	2.273 ± 2
O(1)	15.013 ± 19	5.265 ± 18	-1.925 ± 24
O(2)	8.024 ± 16	-1.879 ± 17	1.578 ± 22
O(3)	6.652 ± 18	-1.430 ± 20	-0.086 ± 21

TABLE 1.7

BOND LENGTHS AND E.S.D.S.

C(1) - C(2)	1.528 ± 33 A	C(10) - C(19)	1.579 ± 31 A
C(1) - C(10)	1.534 ± 31	C(11) - C(12)	1.543 ± 29
C(2) - C(3)	1.448 ± 44	C(12) - C(13)	1.520 ± 30
C(3) - C(4)	1.554 ± 36	C(13) - C(14)	1.524 ± 32
C(4) - C(5)	1.603 ± 32	C(13) - C(17)	1.572 ± 25
C(4) - C(20)	1.537 ± 31	C(14) - C(15)	1.551 ± 33
C(4) - C(21)	1.503 ± 37	C(15) - C(16)	1.620 ± 39
C(5) - C(6)	1.472 ± 30	C(16) - C(17)	1.524 ± 36
C(5) - C(10)	1.579 ± 30	C(18) - C(13)	1.540 ± 29
C(6) - C(7)	1.515 ± 29	C(22) - C(23)	1.556 ± 37
C(7) - C(8)	1.585 ± 31	O(1) - C(3)	1.237 ± 32
C(8) - C(9)	1.487 ± 28	O(2) - C(22)	1.201 ± 34
C(8) - C(14)	1.593 ± 25	O(3) - C(17)	1.488 ± 26
C(9) - C(10)	1.560 ± 28	O(3) - C(22)	1.300 ± 42
C(9) - C(11)	1.555 ± 28	I(1) - C(23)	2.136 ± 23

TABLE 1.8

INTERBOND ANGLES AND E.S.D.S.

C(2) -C(1)-C(10)	116.8±19	C(7) -C(8) -C(14)	108.5±16
C(1) -C(2)-C(3)	110.7±23	C(9) -C(8) -C(14)	108.2±15
C(2) -C(3)-C(4)	120.7±21	C(8) -C(9) -C(10)	111.5±16
C(2) -C(3)-O(1)	122.1±26	C(8) -C(9) -C(11)	111.1±16
C(4) -C(3)-O(1)	117.1±27	C(10)-C(9) -C(11)	113.7±16
C(3) -C(4)-C(5)	108.0±16	C(1) -C(10)-C(5)	108.7±17
C(3) -C(4)-C(20)	107.2±18	C(1) -C(10)-C(9)	109.3±16
C(3) -C(4)-C(21)	111.8±20	C(1) -C(10)-C(19)	108.7±17
C(5) -C(4)-C(20)	116.5±17	C(5) -C(10)-C(9)	106.7±16
C(5) -C(4)-C(21)	106.0±17	C(5) -C(10)-C(19)	112.8±16
C(20)-C(4)-C(21)	107.4±18	C(9) -C(10)-C(19)	110.7±17
C(4) -C(5)-C(6)	114.1±16	C(9) -C(11)-C(12)	112.2±16
C(4) -C(5)-C(10)	114.5±16	C(11)-C(12)-C(13)	110.0±17
C(6) -C(5)-C(10)	110.9±16	C(12)-C(13)-C(14)	108.2±16
C(5) -C(6)-C(7)	110.9±17	C(12)-C(13)-C(17)	113.8±15
C(6) -C(7)-C(8)	110.2±18	C(12)-C(13)-C(18)	110.9±19
C(7) -C(8)-C(9)	110.9±15	C(14)-C(13)-C(17)	97.7±16

C(14)-C(13)-C(18)	117.3±17	C(13)-C(17)-O(3)	113.3±16
C(17)-C(13)-C(18)	108.5±15	C(16)-C(17)-O(3)	105.0±17
C(8) -C(14)-C(13)	109.9±16	C(23)-C(22)-O(2)	125.2±30
C(8) -C(14)-C(15)	119.2±16	C(23)-C(22)-O(3)	111.2±23
C(13)-C(14)-C(15)	105.5±16	O(2) -C(22)-O(3)	123.4±28
C(14)-C(15)-C(16)	100.5±20	C(22)-C(23)-I(1)	107.8±15
C(15)-C(16)-C(17)	104.8±21	C(17)-O(3) -C(22)	120.0±19
C(13)-C(17)-C(16)	107.9±18		

TABLE 1.9

INTRAMOLECULAR NON-BONDED DISTANCES

(A) 1-4 TRANSANNULAR DISTANCES.

C(1)...C(4)	3.01 Å	C(7)...C(10)	2.97 Å
C(1)...C(11)	3.03	C(7)...C(15)	3.15
C(2)...C(5)	3.01	C(8)...C(12)	2.97
C(3)...C(10)	2.97	C(9)...C(13)	2.97
C(5)...C(8)	2.93	C(11)...C(14)	2.91
C(6)...C(9)	2.94	C(15)...C(17)	2.49

(B) 1-4 DISTANCES INVOLVING METHYL GROUPS.

C(18)...C(8)	3.15 Å	C(19)...C(8)	3.07 Å
C(18)...C(11)	3.08	C(19)...C(11)	3.13
C(18)...C(15)	3.08	C(20)...C(2)	3.25
C(18)...C(16)	3.20	C(20)...C(6)	3.29
C(19)...C(2)	3.19	C(20)...C(10)	3.40
C(19)...C(4)	3.31	C(21)...C(6)	3.01
C(19)...C(6)	3.08		

(C) 1-3 METHYL - METHYL DISTANCE.

C(19)...C(20)	3.29 Å

TABLE 1.10

INTERMOLECULAR DISTANCES LESS THAN 4.0 Å.

O(1)....C(23)	ii	3.20 Å	C(3)....C(22)	ii	3.77 Å
O(2)....C(1)	iii	3.29	O(2)....C(16)	i	3.78
O(2)....C(3)	iii	3.33	C(11)...C(7)	i	3.79
O(1)....O(2)	ii	3.45	O(2)....C(4)	iii	3.80
O(2)....C(5)	iii	3.45	C(3)....C(18)	iv	3.80
O(1)....C(22)	ii	3.47	O(2)....C(10)	iii	3.82
C(19)...C(23)	vii	3.53	O(2)....C(15)	i	3.83
O(1)....C(7)	v	3.60	C(2)....C(18)	iv	3.85
O(2)....C(2)	iii	3.65	C(3)....C(23)	ii	3.89
C(2)....C(21)	i	3.65	O(2)....C(21)	iii	3.94
O(1)....C(19)	iv	3.66	C(1)....C(21)	i	3.94
O(1)....C(6)	v	3.75	C(6)....C(23)	vi	3.96
O(1)....C(18)	iv	3.77	C(20)...C(18)	iv	3.97

The Roman numerals indicate the transformations which must be applied to the atomic coordinates given in Table 1.2.

i	x,	y,	1 - z.
ii	3/2 - x,	-y,	1/2 + z.
iii	3/2 - x,	-y,	-1/2 + z.
iv	1/2 + x,	1/2 - y,	- z.
v	1/2 + x,	1/2 - y,	1 - z.
vi	1 - x,	1/2 + y,	1/2 - z.
vii	1 - x,	1/2 + y,	-1/2 - z.

The transformations are applied to the coordinates of the second atom.

TABLE 1.11

MEAN MOLECULAR PLANES

(A) DEVIATIONS FROM MEAN PLANES (IN ÅNGSTROMS)

Plane 1 defined by atoms C(1),C(2),C(3),C(4),C(5),C(10).

C(1)	0.215	C(4)	-0.189
C(2)	-0.185	C(5)	0.220
C(3)	0.176	C(10)	-0.237

Plane 2 defined by atoms C(1),C(2),C(4),C(5).

C(3)	0.517	C(10)	-0.642

Plane 3 defined by atoms C(5),C(6),C(7),C(8),C(9),C(10).

C(5)	0.270	C(8)	-0.226
C(6)	-0.251	C(9)	0.253
C(7)	0.214	C(10)	-0.260

Plane 4 defined by atoms C(6),C(7),C(9),C(10).

C(5)	0.716	C(8)	-0.671

Plane 5 defined by atoms C(8),C(9),C(11),C(12),C(13),C(14).

C(8)	-0.258	C(12)	0.237
C(9)	0.226	C(13)	-0.279
C(11)	-0.212	C(14)	0.284

Plane 6 defined by atoms C(8),C(11),C(12),C(14).

C(9)	0.667	C(13)	-0.737

Plane 7 defined by atoms C(15),C(16),C(17).

C(13)	-0.339	C(14)	0.427

Plane 8 defined by atoms C(2),C(4),C(6),C(8),C(10),C(11),C(13).

C(2)	0.060	C(10)	-0.224
C(4)	0.152	C(11)	-0.046
C(6)	-0.053	C(13)	0.153
C(8)	-0.041		

Plane 9 defined by atoms C(1),C(3),C(5),C(7),C(9),C(12),C(14).

C(1)	-0.115	C(9)	-0.132
C(3)	0.182	C(12)	0.058
C(5)	-0.067	C(14)	0.120
C(7)	-0.047		

(B) PLANE EQUATIONS

Plane No.	P	Q	R	S	RMS D
1	0.5240	-0.7950	-0.3056	3.4922	0.205
2	0.7118	-0.6170	-0.3356	6.7070	0.003
3	0.6484	-0.7006	-0.2980	5.2621	0.246
4	0.8384	-0.4410	-0.3203	8.1415	0.009
5	0.6889	-0.6381	-0.3439	5.9091	0.251
6	0.8634	-0.3688	-0.3441	7.9401	0.020
7	0.8059	-0.4433	-0.3924	7.0591	
8	0.6059	-0.7120	-0.3550	4.7615	0.124
9	0.6288	-0.6977	-0.3433	5.4927	0.112

P,Qand R are direction cosines of the plane normal, S is the plane to origin distance and RMS D is the root mean square deviation in Angstroms, of the atoms defining the plane from the plane.

The plane equation is then

$$PX + QY + RZ = S$$

where X,Y and Z are the atomic coordinates in Angstroms referred to the crystal axes.

1.4 Discussion.

The most interesting feature in the structure of 3-keto-4,4-dimethyl-5α-androstane-17β-iodoacetate (II) is the effect of the 1-3 diaxial transannular interaction of the methyl groups C(19) and C(20) on the conformation of ring A. The deviations of the ring A atoms (C(1),C(2), C(3),C(4),C(5) and C(10)) from the mean plane through them (plane 1, table 1.11) are alternately positive and negative, characteristic of the chair form of cyclohexanone, permitting rejection, at least in the solid state, of a boat conformation for this type of compound (Holker and Whalley, 1961). The individual deviations of the atoms from the plane (table 1.11), which range from 0.18 Å to 0.24 Å, are smaller than the value of 0.26 Å expected for an undistorted cyclohexanone system and indicate a flattening of the ring.

Klyne and Prelog (1960) have shown that the conformation of a six-membered ring is characterised by six valency angles and six torsional (or Newmann) angles. [The torsional angle ϕ(1-2-3-4) denotes the angle between the plane defined by C(1),C(2) and C(3) and the plane defined by C(2),C(3) and C(4) (fig. 3), and is represented by 2-3 in the following tables.]

The observed values of the torsional angles (table A)

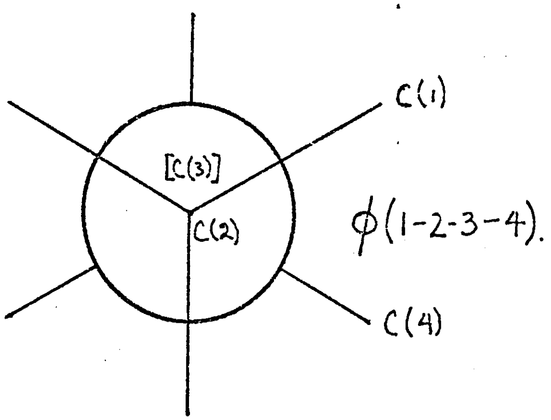


Figure 3.

are all smaller than the standard values, confirming that ring A has been flattened. The maximum difference between the observed and standard values of the torsional angles is 9.9° in the case of ϕ(3-4-5-10) and must be due to the steric repulsion between the axial methyl groups (C(19) and C(20)) causing a rotation about the C(4)-C(5) bond. This rotation forces C(3) towards the plane of C(1),C(2),C(4) and C(5) in addition to reducing the torsional angle ϕ(3-4-5-10).(fig. 4).

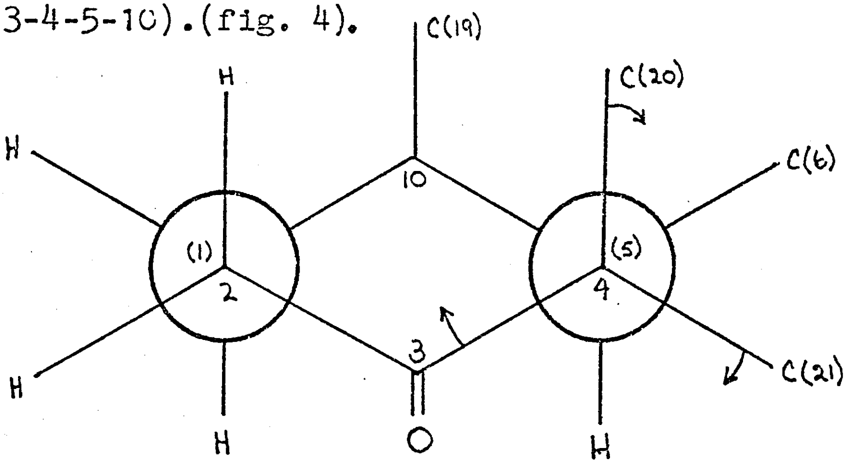


Figure 4.

Table A.

Torsional angles in ring A			Valence angles in ring A		
Angle	Observed	Standard*	Angle	Observed	Standard*
1-2	-50.8°	-55.8°	2-1-10	116.8°	113.3°
2-3	+48.5	+52.3	1-2-3	110.7	112.7
3-4	-47.0	-52.6	2-3-4	120.7	120.0
4-5	+46.6	+56.5	3-4-5	108.0	112.7
10-5	-50.8	-55.1	4-5-10	111.4	113.1
10-1	+52.6	+54.9	5-10-1	108.7	106.4

* The standard values are weighted average values of the torsional angles and valence angles of ring A in substituted cyclopentano-perhydrophenanthrene molecules in which there is no apparent deformation of ring A (Geise, Altona and Romers, 1967).

Displacement of C(20) by such a mechanism is also apparent in (i) the reduction of the distance of C(3) from the plane C(1),C(2),C(4) and C(5) (plane 2, table 1.11) to 0.51 Å compared with the value of 0.73 Å in an undistorted system (Sim, 1965), (ii) the increase in the C(19)...C(20) non-bonded distance from the normal transannular distance of 2.52 Å to 3.29 Å, (iii) the increase in the intramolecular distances C(20)...C(2) and C(20)...C(10) (3.25 Å and 3.40 Å respectively compared with 3.0 Å in an undistorted cyclohexanone system) and (iv) the increase in the torsional angle ϕ(O(1)-3-4-21) from 0° in an undistorted system involving a trigonal carbon atom (Geise, Altona and Romers, 1967) to 17.1° in this system. The value of 3.29 Å for the separation of the axial methyl groups compares with the values of 3.28 Å in 8β-methyltesterone-17β - monobromoacetate (Koyama et al., 1967) and 3.31 Å in the p-bromophenacyl ester of labdanolic acid (Bjamer, Ferguson and Melville, 1968).

Increased interaction between the C(20) methyl group and the C(6) methylene group of ring B is relieved by distortion of the C(5)-C(4)-C(20) bond angle to 116.5° so that the non-bonded distance C(20)...C(6) is increased to 3.29 Å.

The bond angles at C(10) show no significant deviations from tetrahedral but the increase in the angle C(5)-C(10)-C(19) (112.7°) is possibly caused by slight displacement of C(19) which could arise because of the mutual repulsion between the axial methyl groups C(19) and C(20). Such a displacement, however, would be expected to be small because of the increased interaction between the C(19) methyl group and the C(11) methylene group in ring C.

Ring B adopts the normal chair conformation with the atoms alternately above and below the mean plane through them (plane 3, table 1.11). The root mean square deviation of the atoms comprising ring B (i.e. C(5) to C(10) inclusive) from the least-squares plane through the ring (0.25 Å) compares favourably with the value of 0.26 Å for the idealised chair conformation of cyclohexane. The torsional angles and valence angles in ring B are contained in table B.

Table B.

Torsional angles in ring B			Valence angles in ring B		
Angle	Observed	Standard	Angle	Observed	Standard
9-10	-58.6°	-56.9°	10-9-8	111.5°	112.5°
10-5	+60.6	+58.0	9-10-5	106.7	107.2
5-6	-61.6	-57.2	10-5-6	110.9	112.2
6-7	+56.8	+53.4	5-6-7	110.9	111.3
7-8	-54.5	-51.9	6-7-8	110.2	113.1
8-9	+56.8	+54.6	7-8-9	110.9	110.8

No significant deviations from the standard values of these angles (Geise, Altona and Romers, 1967) are found suggesting that conformational transmission effects arising from the distortion of ring A are minimal.

The observed and standard values for the torsional and valence angles present in ring C are contained in table C. The most marked difference between the observed and standard valence angles is in the angle C(8)-C(14)-C(13) (observed 109.9°, standard 113.8°). The valence angles used to derive

Table C.

Torsional angles in ring C			Valence angles in ring C		
Angle	Observed	Standard	Angle	Observed	Standard
11-12	-55.5°	-55.0°	11-12-13	110.0°	110.2°
11-9	+55.0	+53.7	9-11-12	112.2	111.1
9-8	-56.4	-52.8	8-9-11	111.1	111.6
8-14	+62.2	+56.7	9-8-14	108.2	108.9
14-13	-63.2	-59.2	8-14-13	109.9	113.8
13-12	+59.6	+55.6	12-13-14	108.2	109.5

the standard angle, range from 116.8° to 113.4° and the deviations of these angles from tetrahedral have been attributed to the strain associated with the trans-coupling of a five- and six-membered ring (Geise and Romers, 1966; Geise, Romers and Rutten, 1966; High and Kraut, 1966). In the case of 3-keto-4,4-dimethyl-5α-androstane-17β-iodoacetate, however, the strain of trans-coupling the rings (C and D) is manifested in distortions of the external angles at the ring junction,(C(12)-C(13)-C(17) 113.8°, C(8)-C(14)-C(15) 119.2°) rather than the internal angles of ring C (C(12)-C(13)-C(14) 108.2°, C(8)-C(14)-C(13) 109.9°). Since a small increase in valency angle in a cyclic system leads to a relatively large decrease in torsional angle, the observed and standard values for the torsional angles at the ring junction are in poor agreement (see table C).

Ring D adopts a half-chair conformation (Hendrickson, 1961) with C(13) and C(14) displaced by -0.34 Å and +0.43 Å respectively from the plane defined by C(15),C(16) and C(17) (plane 7, table 1.11). The valence angles in ring D (table 1.8) are all smaller than tetrahedral and reflect the strain inherent in five-membered rings. The greatest deviation is found in the angle C(14)-C(13)-C(17) (97.7°). Similar values for this angle are found in 4-bromo-estradiol (98.5°) (Norton, Kartha and Lu, 1964), 4-bromo-9β,10α-pregna-4,6-diene-3,20-dione (98.3°) (Romers, Heykoop, Hesper and Geise, 1966), and androsterone (99.2°) (High and Kraut, 1966). The contraction of this angle may be attributed to the strain of trans-coupling rings C and D, the full substitution at C(13) and steric repulsion between the β-methyl group at C(13) and the β-substituent at C(17).

The perhydrophenanthrene skeleton of the molecule is slightly convex towards the β-side. This effect is noticed in the deviations of the atoms from the mean plane through the β-atoms (C(2),C(4),C(6),C(8),C(10),C(11) and C(13)) (plane 8, table 1.11) and the corresponding plane through the α-atoms (C(1),C(3),C(5),C(7),C(9),C(12) and C(14)), (plane 9, table 1.11). The deviations are summarised in figure 5; the atoms at both ends of the molecule are below their respective planes (denoted by positive distances), whilst the atoms in the middle of the molecule are above their respective planes (denoted by negative distances).

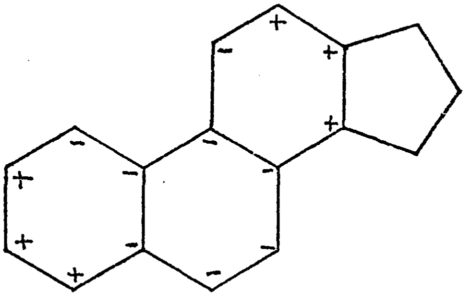


Figure 5.

The bending is caused by steric interactions between the β-methyl groups at C(10) and C(13) and the axial hydrogen atoms at C(8) and C(11) and leads to compression of the axial hydrogen atoms on the α-side of the molecule. The separation of the hydrogen atoms on the α-side of the molecule (approximately 2.9 Å), however, is such that there will be little resistance to their compression.

Table D contains the average values for the different types of bonds found in the substituted cyclopentanoperhydrophenanthrene system present in the molecule. No significant deviations from these average values are found in the individual bond lengths (cf. table 1.8) supporting the conclusion that the steric overcrowding in the molecule has been relieved by bond angle deformation and not by bond length deformation.

Table D.

Average bond lengths (in Ångstroms)

type	No.	Mean Value	Literature*
$C_{sp^3}-C_{sp^3}$	22	1.546 ± 0.030	1.537 ± 0.005
$C_{sp^3}-C_{sp^2}$	2	1.501 ± 0.040	1.510 ± 0.005
$C_{sp^2}=O$	1	1.237 ± 0.032	1.215 ± 0.005

* Sutton (1965).

The bond lengths and valency angles in the iodoacetoxy group (tables 1.7 and 1.8) compare favourably with the accepted mean values (Sutton, 1965).

The packing of the molecules in the unit cell, as projected down the c-axis is shown in figure 1.3. The intermolecular distances (table 1.10) all fall within the range regarded as normal for van der Waals contacts and no short intermolecular distances are observed. This result indicates that the conformational distortions discussed above arise from intramolecular forces and not from crystal-packing forces.

CHAPTER 2.

The Crystal and Molecular Structure

of

3-keto-4,4-dimethyl-19-nor-5α-androstane-17β-iodoacetate.

2.1 Experimental.

The crystals used in this analysis were supplied by Professor W.B.Whalley. They consisted of colourless needles elongated along the c-axis. Unit cell dimensions were obtained from rotation and Weissenberg photographs (Cu Kα radiation, $\lambda = 1.5418$ Å) taken about the c-axis, and from precession photographs of the h0l and 0kl reciprocal lattice nets (Mo Kα radiation, $\lambda = 0.7107$ Å). The space group was determined uniquely from the systematically absent spectra.

Crystal Data.

3-keto-4,4-dimethyl-19-nor-5α-androstane-17β-iodoacetate.

$C_{22}H_{33}O_3I$, F.W. = 472.4.

Orthorhombic, a = 11.32 ± 2 Å, b = 25.42 ± 3 Å, c = 7.55 ± 2 Å.

U = 2173 $Å^3$, F(000) = 968 electrons, Dm = 1.44 gm.cm.$^{-3}$,

Z = 4, Dx = 1.444 gm.cm.$^{-3}$, Space Group $P2_12_12_1$ (D_2^4, No, 19).

Linear absorption coefficient (Cu Kα) = 119.5 cm.$^{-1}$

(Mo Kα) = 15.6 cm.$^{-1}$

2.2 Data Collection (by diffractometer).

The intensity data were collected using a Hilger and Watts Y-190 linear diffractometer (see Appendix I). The crystal was rotated about its c-axis and the intensities from the reciprocal lattice nets hk0-5 obtained using Mo Kα radiation. Because of marked deterioration of the crystal on prolonged exposure to X-radiation, two crystals were used for the data collection. The reciprocal lattice nets hk0,1,2 were collected from the first crystal using a one minute oscillation cycle of 3°. Technical difficulties enforced the use of a half-minute scanning cycle in the collection of subsequent layers from the second crystal. Balanced strontium oxide / zirconium oxide Ross filters (Ross, 1926) were used to monochromate the molybdenum radiation, and each reflection was measured twice with each filter. The maximum reflecting angle was $\theta = 30^\circ$.

The intensity data were reduced to structure amplitudes by applying Lorentz, polarisation and rotation factors (Tunell, 1939) appropriate to a small mosaic crystal. No absorption correction was applied. A total of 1776 independent structure amplitudes were obtained, which represents 89 % of the data which would be available to Cu Kα radiation.

The structure amplitudes were placed on approximately absolute scale by ensuring that $k\sum|Fo| = \sum|Fc|$ for each reciprocal lattice net in the first structure factor calculations. The final scale was determined by the least-squares refinement.

2.3 Structure Solution.

The Harker sections at u = 1/2, v = 1/2 and w = 1/2 were calculated and are shown in figure 2.1. The coordinates of the peaks marked A, B and C which correspond to the vectors arising from the iodine atoms related by the two-fold screw axes were derived by the method of Booth (1948a) and gave the fractional coordinates of the iodine atom (-0.250, -0.099, 0.192). The apparent discrepancies in the heights of peaks B and C are explained by the proximity of the x-coordinate to 0.25 which causes the peaks related by the mirror planes at u = 1/2 and v = 1/2 to coalesce.

Approximate phase angles, based on the scattering of the heavy atom only, were used in conjunction with the observed structure amplitudes (modified by the weighting function proposed by Sim (1960)) to calculate the electron-density distribution. Interpretation of the resultant distribution was somewhat complicated by the presence of pseudo-symmetry in the form of mirror planes parallel to (100) at x = 1/4 and 3/4.

Consideration of known chemical features of the molecule permitted selection of three probable atomic sites in the side chain of ring D, but the bulk of the molecule was obscured by the pseudo-symmetry. Gradually, by successive rounds of structure factor and electron-density calculations, it was possible to derive coordinates for the twenty-six non-hydrogen atoms in the structure. The solution of the structure is summarised in the following table:

	Atoms included in the structure factor calculation	R
1	Iodine only	0.484
2	I + 2 O + 1 C	0.444
3	I + 2 O + 11 C	0.400
4	I + 2 O + 14 C	0.396
5	I + 3 O + 19 C	0.370
6	I + 3 O + 22 C	0.331
7	Backshift correction*	0.303

* Booth, (1946).

Individual chemical types were identified by consideration of the known chemical structure and assigned the corresponding atomic scattering factors (carbon and oxygen, Hoerni and Ibers, 1954; iodine, Thomas and Umeda, 1957). During the structure solution an overall U_{iso} of $0.05\ \mathring{A}^2$ was assumed.

2.4 Least-squares Refinement.

The atomic parameters of the structural model were refined by the method of least-squares. Three cycles of minimisation of the function $M = \sum w(|Fo| - |Fc|)^2$, in which three positional and an isotropic thermal parameter for each atom were varied, reduced the residual R to 0.242. The full matrix of the normal equations was evaluated in each cycle; unit weights were applied to the observations and individual layer scale factors were refined. The parameter shifts calculated in the third cycle were not significant in comparison with the estimated standard deviations but the poor agreement between the observed and calculated structure amplitudes suggested the presence of serious errors. Bond lengths and angles were calculated and showed marked differences from the expected values; as much as 0.3 Å in some cases. These differences were attributed to systematic errors in the data.

Two main sources of error were revealed by a detailed analysis of the structure factor data, viz. the extremely low counting rates obtained on the diffractometer (approximately 60 % of the reflections had count rates of less than 15 counts per minute) and the marked decomposition of the crystal on prolonged exposure to X-radiation.

In an attempt to overcome these errors the data were recollected using photographic techniques. This overcomes the problem of decomposition by giving a time-averaged value for each intensity in a particular reciprocal lattice net. The problem of weak reflecting power of the crystal may be overcome by increasing the duration of the exposure.

2.5 Collection of Photographic Data.

The crystals were rotated about their needle (c) axis and the hk0-5 reciprocal lattice nets recorded by means of equatorial and equi-inclination Weissenberg photographs using Cu Kα radiation. The multiple film technique (Robertson, 1943) and the interfilm scale factors of Rossman (1956) were used to achieve correlation of the strong and weak reflections. The intensities were estimated visually by comparison with a calibrated step-wedge. A total of five crystals were used for the collection of the intensity data.

The intensities were reduced to structure amplitudes by applying Lorentz, polarisation and rotation factors (Tunell, 1939) appropriate to a small mosaic crystal. No absorption correction was applied. A total of 852 independent structure amplitudes (table 2.2) were obtained which represents 43 % of the data available to Cu Kα

radiation.

The observed structure amplitudes were placed on approximately absolute scale by ensuring that $k \sum |Fo| = \sum |Fc|$ in the first structure factor calculation. The final scale factor was determined by the least-squares refinement.

2.6 Refinement of the Photographic Data.

The structure was refined in four cycles of least-squares minimisation of the function $M = \sum w(|Fo| - |Fc|)^2$. Three positional and an isotropic thermal parameter were refined for each atom; individual layer scale factors were also refined. The full matrix of the normal equations was evaluated in each cycle. The weighting scheme was of the form

$$w = 1 / [p_1 + |Fo| + p_2 |Fo|^2]$$

where $p_1 = 1.40$ and $p_2 = 0.037$ (Cruickshank, 1961). No shifts in coordinates or thermal parameters were greater than 0.25σ in the fourth cycle of least-squares refinement. The agreement index was reduced from 0.176 to 0.122 in the four cycles of isotropic refinement.

The structure factors calculated with the final molecular parameters were used to phase electron-density and difference syntheses. The electron-density synthesis is shown by means of a superimposed contour diagram on (010) in figure 2.2 which also explains the numbering system adopted.

The difference map showed a complex region of positive and negative density in the region of the iodine atom and this was tentatively ascribed to uncorrected anisotropic thermal motion. As the standard deviation of the electron density, calculated using the approximate formula of Cruickshank (1949), was 0.3 e/Å^3, no attempt was made to locate the hydrogen atoms in the difference synthesis.

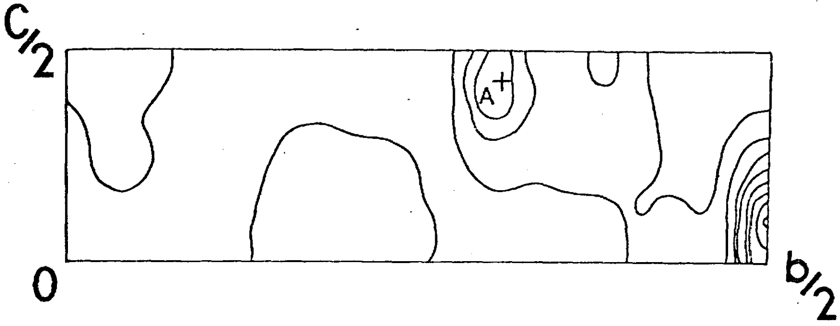
The atomic parameters are contained in table 2.1. Table 2.2 contains the observed and calculated structure amplitudes and table 2.3 an analysis of the structure factor data by layer line index and magnitude of |Fo| . Atomic coordinates (in Ångstroms) with respect to the crystal axes are given in table 2.4; tables 2.5 and 2.6 contain bond lengths and interbond angles respectively. The more important interatomic non-bonded distances are given in tables 2.7 (intramolecular) and 2.8 (intermolecular). Deviations of atoms from selected mean planes through portions of the molecule are given in table 2.9. Estimated standard deviations, where quoted, are in units of the last decimal place of the quantity to which they refer.

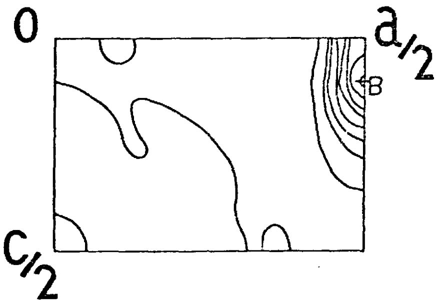
TABLES AND DIAGRAMS

Figure 2.1

Harker sections at u =1/2, v = 1/2 and w = 1/2.

Contours are at arbitrary levels.


c/2
A
0
b/2


0
a/2
B
c/2

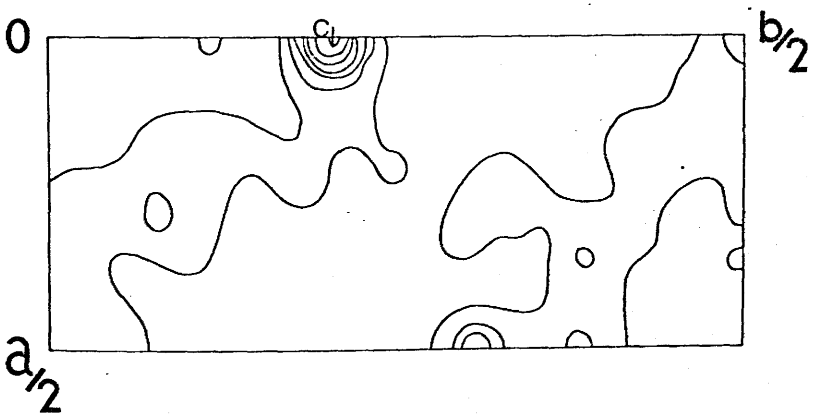
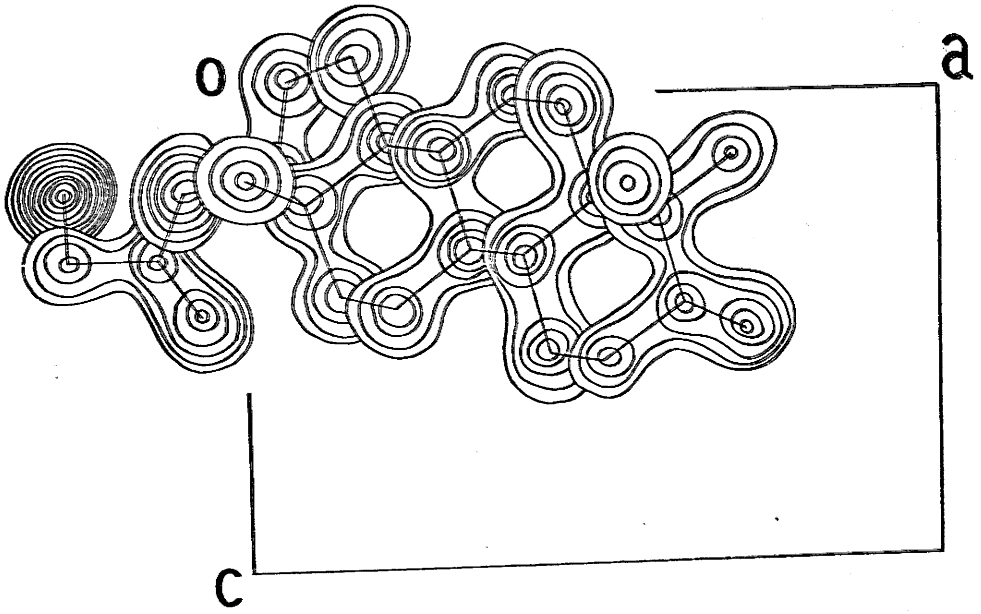

0
C
b/2
a/2

Figure 2.2

Final electron-density distribution shown by means of superimposed contour sections parallel to (010). Contours are at intervals of 1 e/Å^{3} except around the iodine atom where they are at intervals of 7 e/Å^{3}.


o
a
c

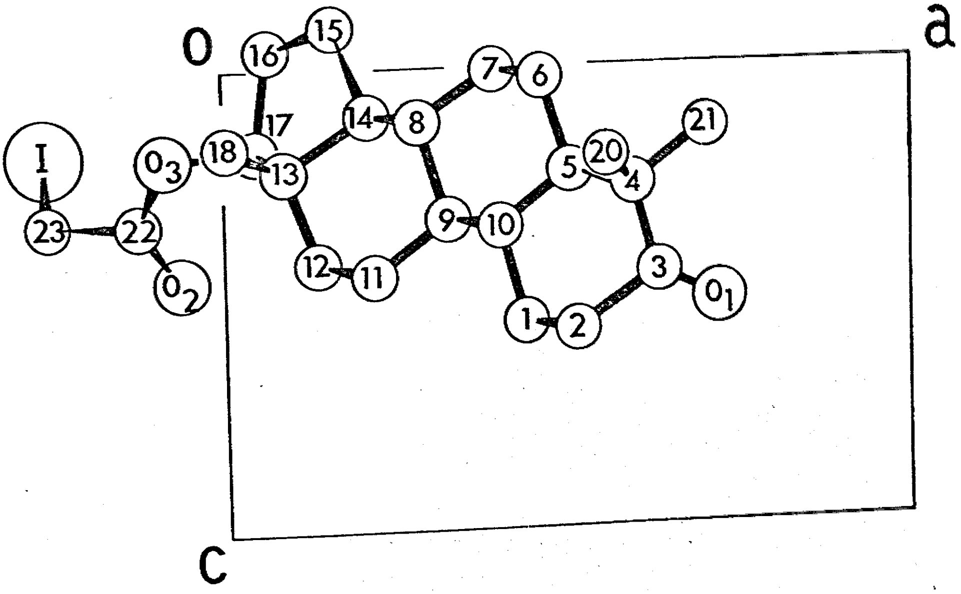
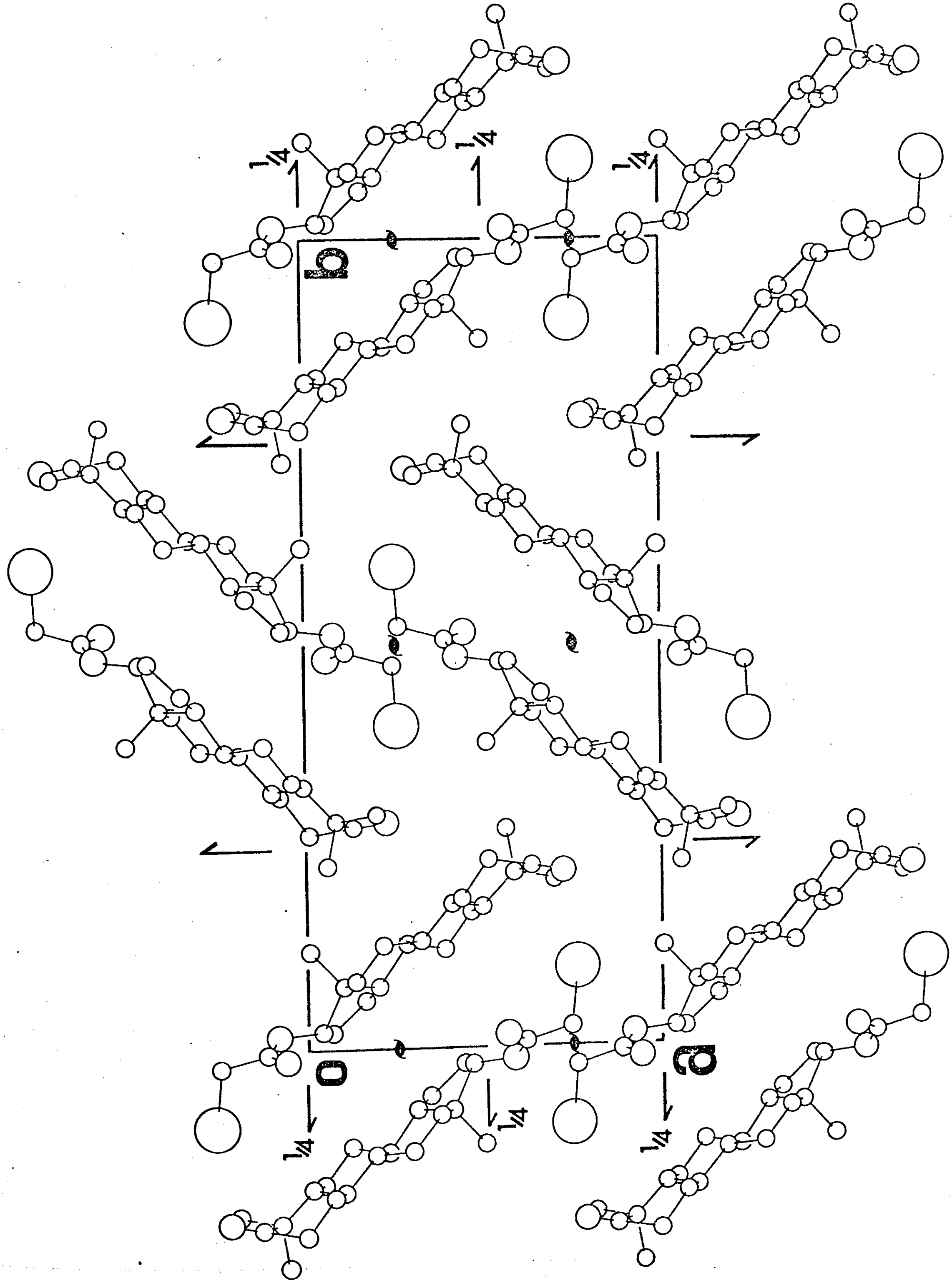

o
a
c
I
23
22
O3
O2
18
17
16
15
14
13
12
11
8
7
6
9
10
5
20
4
21
3
O1
1
2

Figure 2.3

The molecular packing as projected on (001).



0
a
b
1/4
1/4
1/4
1/4
1/4
1/4

TABLE 2.1

FRACTIONAL COORDINATES AND ISOTROPIC TEMPERATURE FACTORS WITH E.S.D.S.

ATOM	X/a	Y/b	Z/c	Uiso
I(1)	-0.2567	-0.0987	0.1952	0.0934
	3	1	5	8
O(1)	0.7218	0.2178	0.5070	0.1069
	24	12	48	100
O(2)	-0.0595	-0.0145	0.4625	0.0874
	23	11	42	88
O(3)	-0.0818	0.0144	0.1921	0.0579
	18	8	35	61
C(1)	0.4305	0.1857	0.5473	0.0444
	25	11	49	81
C(2)	0.5088	0.2333	0.5693	0.0744
	32	15	63	114
C(3)	0.6270	0.2251	0.4519	0.0612
	30	13	56	94
C(4)	0.5822	0.2171	0.2629	0.0582
	27	13	54	97
C(5)	0.4931	0.1746	0.2281	0.0552
	28	13	53	88
C(6)	0.4603	0.1693	0.0269	0.0571
	28	13	52	92
C(7)	0.3824	0.1218	0.0007	0.0494
	25	12	52	80
C(8)	0.2782	0.1297	0.1191	0.0543
	26	12	50	92
C(9)	0.3165	0.1328	0.3251	0.0465
	26	11	48	78

C(10)	0.4006 26	0.1796 12	0.3524 49	0.0497 86
C(11)	0.2128 30	0.1286 14	0.4431 58	0.0695 109
C(12)	0.1322 26	0.0827 11	0.4146 51	0.0474 83
C(13)	0.0950 26	0.0769 12	0.2168 51	0.0475 82
C(14)	0.2008 28	0.0770 13	0.0974 51	0.0546 91
C(15)	0.1583 30	0.0606 13	-0.0870 57	0.0631 101
C(16)	0.0657 30	0.0193 16	-0.0398 58	0.0701 108
C(17)	0.0406 36	0.0199 17	0.1689 68	0.0868 124
C(18)	0.0068 32	0.1207 14	0.1683 60	0.0733 108
C(20)	0.5555 31	0.2722 14	0.1974 63	0.0748 118
C(21)	0.6950 34	0.2085 16	0.1553 66	0.0819 112
C(22)	-0.1148 34	-0.0077 13	0.3340 65	0.0724 116
C(23)	-0.2426 33	-0.0246 14	0.3287 56	0.0748 101

TABLE 2.2

FINAL OBSERVED AND CALCULATED STRUCTURE AMPLITUDES.

H	K	L	Fo	Fc
0	4	0	81.8	75.7
0	6	0	105.0	93.6
0	8	0	96.7	92.3
0	10	0	196.8	163.8
0	12	0	39.2	43.0
0	14	0	32.8	28.6
0	16	0	57.3	45.7
0	18	0	14.7	10.9
0	20	0	15.8	15.9
0	22	0	20.1	21.8
0	24	0	13.5	13.4
0	26	0	15.9	22.1
1	2	0	32.9	26.3
1	3	0	36.9	20.6
1	4	0	187.5	165.6
1	5	0	30.5	29.6
1	6	0	26.8	28.9
1	7	0	92.1	79.9
1	8	0	12.9	9.0
1	9	0	187.8	164.4
1	10	0	12.2	8.2
1	11	0	95.7	95.1
1	13	0	9.6	7.8
1	15	0	104.9	104.8
1	16	0	31.9	31.5
1	17	0	39.8	42.9
1	19	0	46.4	64.6
1	21	0	33.7	31.5
1	25	0	22.1	24.9
2	0	0	111.8	125.8
2	1	0	46.2	34.6
2	2	0	69.8	59.3
2	3	0	9.7	9.7
2	4	0	185.0	203.6
2	5	0	155.5	133.8
2	6	0	195.8	194.0
2	7	0	10.7	10.0
2	8	0	25.9	28.2
2	10	0	94.2	99.9
2	11	0	21.4	8.8
2	12	0	14.1	9.6
2	13	0	25.2	22.7
2	14	0	31.1	26.2
2	15	0	47.7	53.2
2	16	0	52.3	57.9
2	19	0	12.7	10.5
2	20	0	39.1	46.1
2	22	0	15.1	13.2
2	25	0	9.3	3.7
2	26	0	12.8	15.5
3	1	0	140.2	145.1
3	2	0	12.9	21.3
3	3	0	92.2	96.8
3	4	0	52.1	44.8
3	5	0	122.9	132.4
3	5	0	11.2	8.0
3	7	0	108.7	102.8
3	8	0	17.4	6.8
3	9	0	49.1	46.9
3	10	0	118.3	81.8
3	11	0	147.3	103.4
3	12	0	7.0	4.5
3	13	0	31.0	34.4
3	14	0	25.9	23.1
3	15	0	85.2	87.0
3	17	0	32.1	23.5
3	19	0	19.4	16.5
3	20	0	11.5	5.1
3	21	0	23.3	21.5
3	25	0	15.9	15.6
4	0	0	144.6	142.7
4	1	0	14.5	10.7
4	2	0	6.6	10.6
4	3	0	20.4	24.9
4	4	0	109.5	110.0
4	5	0	23.3	17.9
4	6	0	72.3	81.9
4	7	0	37.8	45.7
4	8	0	44.8	44.2
4	9	0	33.7	37.3
4	10	0	73.8	74.9
4	11	0	17.5	24.5
4	12	0	15.8	12.7
4	13	0	13.4	8.3
4	14	0	64.7	63.3
4	15	0	25.0	19.2
4	16	0	53.1	51.0
4	20	0	26.0	26.9
4	24	0	11.4	10.8
4	26	0	13.5	16.5
5	1	0	95.9	107.7
5	2	0	25.0	23.6
5	3	0	17.6	3.3
5	4	0	19.0	16.6
5	5	0	98.8	115.6
5	6	0	14.0	28.4
5	7	0	44.3	41.7
5	8	0	18.5	20.3
5	9	0	77.4	70.4
5	10	0	10.5	3.7
5	11	0	56.6	57.4
5	13	0	10.1	7.3
5	15	0	40.5	38.1
5	17	0	21.6	26.0
5	19	0	23.3	26.0
5	21	0	23.4	18.3
5	25	0	13.6	11.3
6	0	0	124.0	127.0
6	1	0	13.8	3.9
6	2	0	51.0	50.4
6	3	0	21.0	28.5
6	4	0	21.3	27.3
6	5	0	10.2	16.5
6	6	0	37.1	37.1
6	7	0	21.2	16.2
6	8	0	31.2	26.1
6	9	0	33.4	24.8
6	10	0	86.7	86.1
6	11	0	21.1	17.8
6	12	0	28.9	26.0
6	14	0	30.3	29.0
6	16	0	35.5	36.1
6	19	0	15.2	9.8
6	20	0	20.3	14.6
6	24	0	13.5	13.1
7	1	0	64.3	74.8
7	2	0	9.5	10.3
7	3	0	9.6	8.4
7	4	0	21.6	22.4
7	5	0	33.9	41.6
7	7	0	13.0	10.8
7	9	0	44.4	45.4
7	10	0	13.8	3.8
7	11	0	28.8	30.7
7	14	0	25.6	24.6
7	15	0	36.0	34.9
7	16	0	11.7	6.0
7	17	0	15.6	9.7
7	19	0	16.4	15.5
7	21	0	12.9	11.2
8	0	0	41.1	45.2
8	3	0	20.1	22.0
8	4	0	41.8	50.0
8	6	0	40.8	50.6
8	10	0	39.2	32.7
8	12	0	11.6	14.5
8	13	0	15.1	15.0
8	14	0	24.4	26.3
8	16	0	20.3	18.7
8	18	0	11.4	8.4
8	20	0	14.0	10.7
9	1	0	25.9	29.2
9	3	0	15.9	14.7
9	4	0	19.6	15.6
9	5	0	35.4	40.8
9	6	0	14.8	19.5
9	7	0	13.3	13.2
9	8	0	11.6	11.4
9	9	0	21.3	17.5
9	10	0	15.1	13.1
9	11	0	21.4	18.9
9	15	0	16.3	15.3
9	17	0	12.8	4.9
10	0	0	13.5	14.4
10	2	0	15.1	15.8
10	3	0	11.7	12.5
10	4	0	11.7	8.7
10	6	0	13.6	16.6
10	9	0	16.5	6.1
10	14	0	15.6	13.6
10	16	0	18.1	16.1
10	21	0	9.0	3.2
11	1	0	15.0	22.4
11	5	0	17.6	24.6
11	8	0	13.0	9.2
11	10	0	12.6	2.9
0	2	1	37.5	45.7
0	3	1	54.8	51.6
0	4	1	132.8	130.3
0	5	1	20.4	11.5
0	6	1	56.1	64.2
0	7	1	85.6	98.1
0	8	1	116.2	120.2
0	9	1	79.4	76.8
0	12	1	85.0	81.1
0	14	1	69.4	59.7
0	16	1	44.5	50.9
0	17	1	39.1	38.9
0	18	1	72.4	64.4
0	19	1	34.4	31.0
0	22	1	43.3	46.2
0	23	1	22.7	18.6
1	0	1	13.2	11.0
1	1	1	94.2	120.6
1	2	1	121.4	139.0
1	3	1	168.3	242.0
1	4	1	54.6	46.5
1	5	1	50.5	43.2
1	6	1	14.8	13.5
1	7	1	124.4	154.4
1	8	1	94.4	91.1
1	9	1	68.1	63.5
1	10	1	13.8	18.7
1	11	1	60.8	51.9
1	12	1	19.9	16.9
1	13	1	90.7	92.7
1	14	1	24.3	22.2
1	15	1	14.7	9.8
1	16	1	26.6	23.8
1	17	1	35.9	27.8
1	18	1	31.2	22.6
1	19	1	34.6	33.5
1	23	1	29.4	20.6
2	0	1	25.9	24.4
2	1	1	52.5	52.9
2	2	1	147.9	170.3
2	3	1	70.9	70.5
2	4	1	106.5	114.9
2	5	1	44.7	51.3
2	6	1	68.5	64.6
2	7	1	63.8	78.3
2	8	1	91.3	89.9
2	10	1	27.5	22.2
2	11	1	54.0	44.6
2	12	1	105.9	114.4
2	13	1	42.2	37.8
2	14	1	68.9	63.7
2	15	1	23.8	18.9
2	16	1	31.4	33.8
2	17	1	28.3	27.0
2	18	1	47.9	41.5
2	20	1	18.0	9.1
2	22	1	32.1	21.4
3	0	1	44.3	40.1
3	1	1	80.5	107.1
3	2	1	48.8	48.8
3	3	1	124.6	119.0
3	4	1	37.0	31.3
3	5	1	27.3	29.5
3	6	1	22.1	13.7
3	7	1	123.6	119.1
3	8	1	49.7	54.1
3	9	1	35.8	46.6
3	10	1	34.3	27.2
3	11	1	71.1	69.3
3	12	1	42.3	48.9
3	13	1	64.9	61.9
3	14	1	25.9	19.5
3	15	1	19.1	17.4
3	17	1	46.0	43.5
3	19	1	30.8	20.9
3	22	1	26.3	13.0
3	23	1	29.2	25.6
4	0	1	29.3	27.2
4	1	1	19.5	29.8
4	2	1	91.4	107.5
4	3	1	30.3	33.8
4	4	1	83.8	85.5
4	5	1	19.5	17.8
4	6	1	59.3	44.1
4	7	1	58.4	49.6
4	8	1	105.2	110.1
4	9	1	22.5	21.5
4	11	1	34.3	35.3
4	12	1	56.4	60.7
4	14	1	60.8	57.1
4	16	1	23.8	16.4
4	18	1	41.7	39.0
4	21	1	22.7	13.7
4	22	1	22.7	18.5
5	0	1	46.2	43.3
5	1	1	52.1	54.7
5	2	1	46.7	50.1
5	3	1	102.8	117.6
5	5	1	22.0	14.6
5	6	1	22.5	22.7
5	7	1	70.6	68.3
5	8	1	40.0	34.8
5	9	1	43.6	42.4
5	10	1	23.1	21.7
5	11	1	44.0	34.8
5	12	1	27.0	21.6
5	13	1	55.7	55.6
5	14	1	23.4	22.9
5	17	1	38.0	30.3
5	18	1	22.3	16.9
5	19	1	29.2	18.6
6	0	1	21.3	25.6
6	1	1	31.7	30.5
6	2	1	73.9	83.7
6	3	1	21.7	21.1
6	4	1	41.7	38.8
6	6	1	52.9	54.3
6	7	1	25.5	15.7
6	8	1	58.3	53.4
6	9	1	36.1	41.6
6	11	1	28.1	22.2
6	12	1	51.3	50.2
6	14	1	30.2	22.0
6	16	1	25.6	20.9
6	17	1	22.5	15.4
6	18	1	32.1	26.7
7	1	1	35.4	38.7
7	2	1	26.3	23.3
7	3	1	66.6	59.0
7	4	1	21.9	20.5
7	5	1	15.6	16.3
7	6	1	38.8	34.1
7	7	1	46.9	45.5
7	8	1	34.7	30.4
7	9	1	40.9	42.5
7	10	1	17.0	20.4
7	13	1	40.1	35.9
7	17	1	26.3	20.2
8	1	1	20.3	17.5
8	2	1	40.8	42.1
8	3	1	20.5	17.7
8	4	1	37.7	32.9
8	5	1	29.4	33.0
8	6	1	29.7	19.2
8	7	1	30.1	27.5
8	8	1	39.3	34.9
8	12	1	41.0	30.9
8	13	1	22.6	17.5
8	14	1	18.5	19.0
9	3	1	31.1	25.0
9	7	1	22.4	20.7
9	11	1	26.3	23.3
9	13	1	32.0	26.4
0	1	2	58.1	71.3
0	2	2	63.6	61.5
0	3	2	85.8	58.0
0	4	2	87.7	84.5
0	5	2	114.9	103.5
0	6	2	76.4	78.3
0	7	2	38.0	38.3
0	8	2	26.2	20.1
0	9	2	61.2	60.4
0	10	2	82.3	86.8
0	11	2	58.1	58.6
0	12	2	33.6	34.6
0	13	2	14.1	18.2
0	14	2	38.7	38.2
0	15	2	39.1	39.3
0	16	2	36.3	45.2
0	17	2	16.9	17.1
0	19	2	18.1	17.7
0	20	2	24.0	29.6
0	21	2	10.9	12.0
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1	1	2	136.6	133.0
1	2	2	34.4	34.0
1	3	2	45.7	38.7
1	4	2	83.9	89.0
1	5	2	127.4	121.7
1	6	2	89.1	78.7
1	7	2	7.0	14.0
1	8	2	13.7	9.3
1	9	2	62.4	63.6
1	10	2	63.5	63.0
1	11	2	83.8	81.9
1	12	2	17.4	15.5
1	13	2	25.3	29.9
1	14	2	33.4	35.5
1	15	2	40.9	43.1
1	16	2	34.7	29.1
1	17	2	17.0	19.5
1	18	2	12.5	17.4
1	19	2	22.3	21.1
1	20	2	24.1	26.3
1	21	2	24.5	17.5
2	0	2	93.0	89.2
2	1	2	53.5	51.6
2	2	2	68.4	62.0
2	3	2	40.8	36.1
2	4	2	76.0	80.4
2	5	2	134.5	88.7
2	6	2	51.1	57.3
2	7	2	57.0	52.2
2	8	2	27.6	25.0
2	9	2	52.7	55.7
2	10	2	82.6	73.2
2	11	2	52.2	54.4
2	12	2	38.6	36.0
2	13	2	12.0	13.9
2	14	2	57.0	55.4
2	15	2	38.7	35.2
2	16	2	35.4	35.8
2	17	2	18.7	21.2
2	19	2	18.4	23.2
2	20	2	30.7	32.2
2	21	2	17.4	17.8
3	0	2	65.4	72.8
3	1	2	76.9	77.1
3	2	2	29.0	15.3
3	3	2	29.8	22.0
3	4	2	54.1	60.0
3	5	2	118.7	113.3
3	6	2	51.4	55.1
3	7	2	30.5	36.4
3	8	2	27.3	25.3
3	9	2	50.4	46.6
3	10	2	59.1	62.7
3	11	2	52.1	51.2
3	12	2	14.6	14.8
3	13	2	10.8	17.4
3	14	2	26.0	23.6
3	15	2	41.7	45.4
3	16	2	25.3	28.0
3	17	2	17.7	13.4
3	18	2	10.5	14.5
3	19	2	13.2	11.6
3	20	2	24.5	26.2
3	21	2	15.6	21.4
4	0	2	86.7	87.7
4	1	2	108.0	105.4
4	2	2	33.9	34.1
4	3	2	52.9	45.0
4	4	2	81.5	87.2
4	5	2	66.6	66.0
4	6	2	66.8	68.3
4	7	2	37.3	43.8
4	8	2	20.4	17.7
4	9	2	54.6	55.0
4	10	2	53.6	57.8
4	11	2	46.9	40.7
4	12	2	11.0	12.8
4	13	2	14.7	15.5
4	14	2	46.2	45.0
4	15	2	28.2	28.9
4	16	2	25.1	28.9
4	20	2	19.1	16.4
4	21	2	19.1	18.0
5	0	2	27.1	32.6
5	1	2	50.5	57.7
5	2	2	13.3	8.9
5	3	2	44.0	45.9
5	4	2	21.7	29.3
5	5	2	62.4	55.7
5	6	2	51.7	48.1
5	7	2	25.4	25.1
5	8	2	32.8	31.5
5	9	2	28.0	33.7
5	10	2	48.4	43.1
5	11	2	45.9	43.3
5	12	2	20.3	15.4
5	13	2	17.0	15.1
5	14	2	25.8	23.0
5	15	2	40.2	35.5
5	16	2	26.0	27.6
5	18	2	19.0	20.3
5	19	2	15.6	15.6
5	20	2	19.1	18.3
5	21	2	13.4	13.0
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6	1	2	45.3	40.7
6	2	2	27.8	28.3
6	3	2	24.0	31.4
6	4	2	33.2	27.0
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6	6	2	60.5	63.4
6	7	2	23.1	24.6
6	8	2	16.0	10.7
6	9	2	21.2	20.1
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6	12	2	22.7	24.4
6	14	2	18.3	10.1
6	15	2	18.7	10.0
6	16	2	27.9	30.5
6	17	2	15.6	11.0
6	19	2	15.6	9.6
6	20	2	15.4	14.1
7	0	2	28.6	30.8
7	1	2	40.5	46.7
7	2	2	22.8	21.8
7	3	2	19.9	21.4
7	4	2	34.8	32.9
7	5	2	40.1	42.3
7	6	2	28.3	22.2
7	8	2	12.3	4.6
7	9	2	48.8	44.5
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7	11	2	18.2	21.1
7	12	2	23.9	25.1
7	13	2	18.8	19.4
7	14	2	25.7	25.7
7	15	2	23.4	20.0
7	16	2	15.6	13.8
8	0	2	39.2	33.9
8	1	2	16.0	16.8
8	2	2	17.6	24.2
8	3	2	17.7	16.4
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8	6	2	22.2	15.2
8	9	2	15.2	19.6
8	10	2	36.0	33.9
8	12	2	17.4	21.1
8	13	2	19.2	13.7
8	14	2	24.7	22.4
8	15	2	19.1	12.9
8	16	2	13.4	8.3
9	0	2	15.3	13.0
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9	2	2	13.2	12.0
9	3	2	10.8	9.6
9	4	2	26.6	24.4
9	5	2	29.9	36.5
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9	8	2	15.6	18.0
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9	10	2	13.5	15.4
9	11	2	13.5	16.3
9	14	2	13.2	4.8
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10	4	2	17.5	13.1
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10	6	2	19.1	22.3
10	8	2	15.5	12.4
10	9	2	17.2	15.7
10	10	2	13.2	9.7
10	11	2	10.7	10.8
0	2	3	53.7	67.2
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0	4	3	48.8	40.5
0	5	3	58.8	44.6
0	6	3	84.3	74.5
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0	8	3	75.6	66.9
0	9	3	68.3	54.5
0	10	3	10.4	7.6
0	11	3	12.7	11.4
0	12	3	25.1	19.9
0	13	3	51.8	60.3
0	14	3	12.8	14.1
0	15	3	10.9	16.0
0	16	3	11.4	11.4
0	17	3	32.3	37.5
0	18	3	21.1	25.7
0	19	3	27.9	27.7
1	0	3	31.4	26.0
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1	2	3	82.0	85.9
1	3	3	40.7	43.2
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2	6	3	32.7	28.9
2	7	3	96.0	91.5
2	8	3	45.1	45.5
2	9	3	49.1	44.3
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2	12	3	38.3	38.5
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2	14	3	22.9	25.7
2	17	3	26.9	25.7
2	19	3	17.8	21.2
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3	9	3	35.2	33.4
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3	12	3	49.1	57.1
3	13	3	33.2	37.1
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7	13	3	20.2	20.6
7	18	3	19.6	21.0
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0	15	4	36.7	31.6
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1	8	4	18.4	18.1
1	9	4	17.6	12.6
1	10	4	60.3	64.5
1	12	4	23.1	19.3
1	13	4	15.3	14.7
1	14	4	33.7	31.7
1	16	4	36.1	30.4
1	20	4	25.6	21.1
2	0	4	32.4	16.2
2	1	4	36.9	40.1
2	2	4	34.0	25.2
2	3	4	64.2	58.6
2	5	4	76.2	76.2
2	6	4	23.4	23.4
2	7	4	53.5	50.7
2	8	4	17.5	9.6
2	9	4	41.2	41.6
2	10	4	16.8	14.6
2	11	4	43.1	37.7
2	15	4	35.6	41.8
3	0	4	51.9	59.6
3	1	4	26.6	21.0
3	2	4	31.9	36.9
3	3	4	29.9	30.9
3	4	4	63.8	73.3
3	5	4	31.0	29.4
3	6	4	69.1	76.8
3	8	4	21.1	16.9
3	10	4	45.8	49.8
3	12	4	27.1	23.7
3	14	4	20.4	16.4
3	16	4	30.4	29.1
4	1	4	47.4	45.9
4	2	4	12.4	9.2
4	3	4	32.0	28.7
4	5	4	62.5	63.2
4	9	4	40.9	38.3
4	10	4	26.7	21.7
4	11	4	45.0	41.7
4	12	4	28.4	28.7
4	13	4	26.6	23.3
4	14	4	21.1	14.4
4	15	4	30.5	25.6
5	0	4	56.4	63.2
5	1	4	19.7	12.3
5	2	4	22.2	20.9
5	4	4	36.7	40.1
5	6	4	33.4	40.7
5	7	4	26.5	28.0
5	9	4	27.8	25.1
5	10	4	41.8	47.2
5	12	4	29.7	25.7
5	14	4	21.7	21.6
6	0	4	26.8	22.5
6	1	4	34.6	40.1
6	5	4	40.8	44.1
6	7	4	20.2	10.0
6	8	4	29.0	27.2
6	9	4	38.1	36.2
6	10	4	21.2	14.4
6	11	4	30.4	25.0
7	0	4	29.1	26.9
7	4	4	24.1	22.8
7	6	4	30.0	26.5
7	10	4	25.4	23.7
8	1	4	25.1	22.4
8	3	4	28.2	26.7
8	5	4	31.1	34.8
0	1	5	16.2	26.9
0	3	5	53.5	51.3
0	4	5	7.9	8.8
0	6	5	12.6	7.6
0	7	5	56.2	56.3
0	8	5	21.7	17.5
0	9	5	41.2	38.6
0	10	5	12.8	9.8
0	11	5	13.4	11.8
0	12	5	11.5	7.8
0	13	5	16.8	9.1
0	14	5	15.1	12.4
0	17	5	22.7	25.4
0	19	5	20.7	21.7
1	1	5	11.8	13.2
1	2	5	47.3	54.9
1	3	5	10.8	11.9
1	4	5	28.9	26.5
1	6	5	24.0	28.7
1	8	5	56.3	61.6
1	9	5	14.2	11.2
1	12	5	23.3	32.7
1	14	5	23.1	24.2
1	16	5	20.5	19.4
1	18	5	22.8	26.1
2	0	5	5.4	4.0
2	1	5	68.8	59.1
2	2	5	8.8	8.8
2	3	5	63.9	62.0
2	4	5	14.9	17.1
2	7	5	45.3	46.4
2	9	5	25.7	23.9
2	10	5	13.4	12.2
2	11	5	24.2	18.6
2	12	5	16.7	17.7
2	13	5	29.9	31.8
3	0	5	19.8	15.4
3	1	5	13.4	12.3
3	2	5	28.7	30.7
3	3	5	25.8	23.8
3	4	5	35.4	32.8
3	6	5	20.9	20.2
3	7	5	16.2	12.8
3	8	5	28.2	33.2
3	12	5	31.1	35.6
3	14	5	23.9	19.2
3	16	5	16.1	11.5
3	18	5	20.7	21.4
4	1	5	30.4	29.9
4	3	5	55.5	54.4
4	4	5	16.3	14.2
4	7	5	46.7	49.9
4	8	5	11.5	15.6
4	9	5	20.4	26.7
4	10	5	17.1	14.8
4	11	5	17.5	19.4
4	13	5	20.3	23.0
5	1	5	13.6	13.5
5	2	5	36.2	37.3
5	3	5	15.9	9.9
5	4	5	19.8	18.1
5	6	5	16.7	14.0
5	7	5	19.0	17.3
5	8	5	35.6	36.2
5	11	5	20.3	18.7
5	12	5	20.5	18.6
5	13	5	18.5	12.8
5	14	5	16.1	17.9
6	1	5	21.0	21.5
6	3	5	17.3	19.5
6	4	5	17.4	12.2
6	5	5	19.7	14.0
6	7	5	25.4	29.2
6	8	5	20.3	20.0
6	9	5	22.4	19.9
6	12	5	13.2	14.1
6	13	5	13.1	17.7
7	0	5	15.7	13.0
7	2	5	30.2	30.9
7	4	5	22.4	22.2
7	5	5	15.9	14.9
7	6	5	25.1	16.6
7	8	5	26.3	27.5

TABLE 2.3

(A) ANALYSIS OF STRUCTURE FACTOR DATA AS A FUNCTION OF LAYER LINE INDEX. ALL FIGURES ARE ON ABSOLUTE SCALE.

l	$\Sigma\|Fo\|$	$\Sigma\|Fc\|$	$\Sigma\Delta$	N	R	$\Sigma\Delta/N$
0	7094	6834	929	176	0.1310	5.28
1	7231	7080	894	153	0.1236	5.84
2	7210	7071	759	197	0.1052	3.85
3	5266	5133	675	157	0.1282	4.30
4	2942	2851	384	86	0.1307	4.47
5	1986	1941	241	83	0.1212	2.90
ALL	31729	30910	3882	852	0.1224	4.56

(B) ANALYSIS OF STRUCTURE FACTOR DATA BY MAGNITUDE OF Fo.

0 - 13	891	886	294	80	0.3305	3.68
13 - 25	5382	5041	977	290	0.1815	3.37
25 - 50	10144	9875	1169	294	0.1152	3.97
50 - 75	6200	6147	570	104	0.0919	5.48
75 - 150	7816	7674	718	77	0.0919	9.33
150 - 300	1297	1287	154	7	0.1189	22.02

TABLE 2.4

ORTHOGONAL COORDINATES AND E.S.D.S.

ATOM	X	Y	Z
I(1)	-2.906 ± 3	-2.508 ± 3	1.474 ± 3
O(1)	8.171 ± 27	5.536 ± 31	3.828 ± 36
O(2)	-0.674 ± 26	-0.369 ± 27	3.492 ± 32
O(3)	-0.925 ± 20	0.367 ± 20	1.451 ± 27
C(1)	4.873 ± 28	4.721 ± 28	4.132 ± 37
C(2)	5.760 ± 36	5.930 ± 39	4.298 ± 48
C(3)	7.098 ± 34	5.723 ± 34	3.412 ± 43
C(4)	6.591 ± 31	5.520 ± 33	1.985 ± 41
C(5)	5.582 ± 32	4.438 ± 32	1.722 ± 40
C(6)	5.210 ± 31	4.303 ± 33	0.203 ± 39
C(7)	4.329 ± 29	3.097 ± 32	0.006 ± 39
C(8)	3.149 ± 29	3.297 ± 31	0.899 ± 38
C(9)	3.582 ± 29	3.376 ± 27	2.454 ± 36
C(10)	4.535 ± 30	4.565 ± 30	2.661 ± 37
C(11)	2.409 ± 34	3.269 ± 37	3.346 ± 44
C(12)	1.496 ± 30	2.102 ± 29	3.130 ± 39
C(13)	1.075 ± 29	1.955 ± 30	1.637 ± 39
C(14)	2.273 ± 32	1.958 ± 32	0.736 ± 38
C(15)	1.792 ± 34	1.541 ± 33	-0.657 ± 43
C(16)	0.744 ± 35	0.492 ± 40	-0.301 ± 44

C(17)	0.460 ± 41	0.505 ± 42	1.275 ± 51
C(18)	0.077 ± 36	3.067 ± 37	1.271 ± 45
C(20)	6.288 ± 35	6.919 ± 36	1.490 ± 48
C(21)	7.868 ± 38	5.301 ± 40	1.173 ± 50
C(22)	-1.299 ± 38	-0.196 ± 34	2.522 ± 49
C(23)	-2.746 ± 37	-0.626 ± 36	2.482 ± 42

TABLE 2.5

BOND LENGTHS AND E.S.D.S.

C(1) - C(2)	1.51 ± 5 A	C(9) - C(11)	1.48 ± 5 A
C(1) - C(10)	1.52 ± 5	C(11)- C(12)	1.50 ± 5
C(2) - C(3)	1.62 ± 5	C(12)- C(13)	1.56 ± 5
C(3) - C(4)	1.53 ± 6	C(13)- C(14)	1.50 ± 5
C(3) - O(1)	1.17 ± 5	C(13)- C(17)	1.62 ± 5
C(4) - C(21)	1.51 ± 5	C(13)- C(18)	1.54 ± 5
C(4) - C(20)	1.53 ± 5	C(14)- C(15)	1.53 ± 5
C(4) - C(5)	1.50 ± 5	C(15)- C(16)	1.53 ± 5
C(5) - C(6)	1.50 ± 5	C(16)- C(17)	1.60 ± 7
C(5) - C(10)	1.50 ± 5	C(22)- C(23)	1.51 ± 5
C(6) - C(7)	1.51 ± 5	O(3) - C(22)	1.27 ± 5
C(7) - C(8)	1.49 ± 5	O(3) - C(17)	1.40 ± 5
C(8) - C(9)	1.62 ± 5	O(2) - C(22)	1.17 ± 5
C(8) - C(14)	1.61 ± 4	I(1) - C(23)	2.14 ± 4
C(9) - C(10)	1.54 ± 4		

TABLE 2.6

INTERBOND ANGLES AND E.S.D.S.

C(2) -C(1)-C(10)	109 ± 3	C(7) -C(8) -C(14)	105 ± 3
C(1) -C(2)-C(3)	109 ± 3	C(9) -C(8) -C(14)	107 ± 3
C(2) -C(3)-C(4)	115 ± 3	C(8) -C(9) -C(10)	109 ± 3
C(2) -C(3)-O(1)	123 ± 4	C(8) -C(9) -C(11)	111 ± 3
C(4) -C(3)-O(1)	125 ± 4	C(10)-C(9) -C(11)	118 ± 3
C(3) -C(4)-C(5)	118 ± 3	C(1) -C(10)-C(5)	119 ± 3
C(3) -C(4)-C(21)	104 ± 3	C(1) -C(10)-C(9)	110 ± 3
C(3) -C(4)-C(20)	104 ± 3	C(5) -C(10)-C(9)	107 ± 3
C(5) -C(4)-C(21)	118 ± 3	C(9) -C(11)-C(12)	117 ± 3
C(5) -C(4)-C(20)	111 ± 3	C(11)-C(12)-C(13)	112 ± 3
C(20)-C(4)-C(21)	98 ± 3	C(12)-C(13)-C(14)	111 ± 3
C(4) -C(5)-C(6)	113 ± 3	C(12)-C(13)-C(17)	114 ± 3
C(4) -C(5)-C(10)	108 ± 3	C(12)-C(13)-C(18)	110 ± 3
C(6) -C(5)-C(10)	118 ± 3	C(14)-C(13)-C(17)	100 ± 3
C(5) -C(6)-C(7)	110 ± 3	C(14)-C(13)-C(18)	112 ± 3
C(6) -C(7)-C(8)	106 ± 3	C(17)-C(13)-C(18)	110 ± 3
C(7) -C(8)-C(9)	112 ± 3	C(8) -C(14)-C(13)	112 ± 3

C(8) -C(14)-C(15)	119 ± 3	C(16)-C(17)-O(3)	107 ± 3
C(13)-C(14)-C(15)	107 ± 3	C(23)-C(22)-O(2)	120 ± 4
C(14)-C(15)-C(16)	101 ± 3	C(23)-C(22)-O(3)	113 ± 4
C(15)-C(16)-C(17)	110 ± 3	O(2) -C(22)-O(3)	128 ± 4
C(13)-C(17)-C(16)	99 ± 3	C(22)-C(23)-I(1)	110 ± 2
C(13)-C(17)-O(3)	116 ± 3	C(17)-O(3) -C(22)	116 ± 3

TABLE 2.7

INTRAMOLECULAR NON-BONDED DISTANCES

(A) 1-4 TRANSANNULAR DISTANCES.

C(1)...C(4)	2.86 Å	C(7)...C(10)	3.04 Å
C(1)...C(11)	2.97	C(7)...C(15)	3.05
C(2)...C(5)	2.98	C(8)...C(12)	3.02
C(3)...C(10)	2.91	C(9)...C(13)	3.00
C(5)...C(8)	2.81	C(11)...C(14)	2.92
C(6)...C(9)	2.93	C(15)...C(17)	2.57

(B) 1-4 DISTANCES INVOLVING METHYL GROUPS.

C(18)...C(8)	3.10 Å	C(20)...C(2)	3.02 Å
C(18)...C(11)	3.11	C(20)...C(6)	3.11
C(18)...C(15)	3.00	C(20)...C(10)	3.16
C(18)...C(16)	3.09	C(21)...C(6)	3.00

TABLE 2.8

INTERMOLECULAR DISTANCES LESS THAN 4.0 Å

O(1)...I(1)	ii	3.37 Å	C(1)...C(7)	i	3.83 Å
O(2)...C(7)	ii	3.40	C(2)...C(6)	i	3.86
C(23)...O(3)	iii	3.40	C(3)...C(11)	v	3.92
O(1)...C(1)	v	3.43	C(21)...C(18)	iv	3.92
O(1)...C(10)	v	3.47	O(1)...C(11)	v	3.93
O(1)...C(2)	v	3.53	O(2)...I(1)	iii	3.96
C(1)...C(6)	i	3.66	C(3)...I(1)	ii	3.98
O(2)...C(23)	iii	3.69	C(23)...C(23)	iii	3.98
C(23)...C(16)	iii	3.79	C(11)...C(15)	i	3.99
C(12)...C(15)	i	3.82			

The Roman numerals indicate the transformations which must be applied to the atomic coordinates given in Table 2.1.

i	x,	y,	1 + z.
ii	1/2 - x,	- y,	1/2 + z.
iii	-1/2 - x,	- y,	1/2 + z.
iv	1/2 + x,	1/2 - y,	- z.
v	1/2 + x,	1/2 - y,	1 - z.

The transformations are applied to the second atom in each case.

TABLE 2.9

MEAN MOLECULAR PLANES

(A) DEVIATIONS FROM MEAN PLANES (IN ÅNGSTROMS).

Plane 1 defined by atoms C(1),C(2),C(3),C(4),C(5),C(10).

C(1)	-0.257	C(4)	0.203
C(2)	0.292	C(5)	-0.155
C(3)	-0.264	C(10)	0.181

Plane 2 defined by atoms C(1),C(2),C(4),C(5).

C(3)	-0.734	C(10)	0.560

Plane 3 defined by atoms C(5),C(6),C(7),C(8),C(9),C(10).

C(5)	-0.210	C(8)	0.285
C(6)	0.225	C(9)	-0.237
C(7)	-0.274	C(10)	0.210

Plane 4 defined by atoms C(6),C(7),C(9),C(10).

C(5)	-0.602	C(8)	0.725

Plane 5 defined by atoms C(8),C(9),C(11),C(12),C(13),C(14).

C(8)	0.258	C(12)	-0.178
C(9)	-0.224	C(13)	0.232
C(10)	0.191	C(14)	-0.279

Plane 6 defined by atoms C(8),C(11),C(12),C(14).

C(9)	-0.649	C(13)	0.656

Plane 7 defined by atoms C(14),C(15),C(16),C(17).

C(14)	-0.037	C(17)	0.037
C(15)	0.058	C(13)	0.693
C(16)	-0.058		

(B) PLANE EQUATIONS

Plane No.	P	Q	R	S	RMS D
1	-0.4701	0.8374	-0.2788	0.7681	0.231
2	-0.6993	0.6385	-0.3214	-1.6722	0.050
3	-0.5116	0.8265	-0.2349	0.6179	0.242
4	0.0011	0.9987	-0.0510	3.1734	0.077
5	-0.5715	0.7980	-0.1910	0.4017	0.230
6	-0.7694	0.5857	-0.2549	-0.7553	0.036
7	-0.6598	0.7486	-0.0646	-0.0448	0.049

P,Q and R are the direction cosines of the plane normal, S is the plane to origin distance and RMS D is the root mean square deviation (in Angstroms) of the atoms defining the plane from the plane.

The plane equation is then

$$PX + QY + RZ = S$$

where X,Y and Z are the atomic coordinates in Angstroms referred to the crystal axes.

atoms and selected groups of them. The root mean square deviation of the six atoms from the plane defined by C(1), C(2), C(3), C(4), C(5) and C(10) (plane 1, table 1.9) is 0.23 Å which is in good agreement with the value of 0.227 Å for ring A in androsterone (Norton and Ohrt, 1964). The atoms are alternately above and below the plane which is characteristic of the chair conformation. Further evidence for the chair conformation is found in the large displacements of C(3) and C(10) (0.73 Å above and 0.56 Å below respectively) from the plane defined by C(1), C(2), C(4) and C(5) (plane 2, table 2.9).

The adoption of a chair conformation for ring A in this compound permits rejection of the proposal of a skewed boat conformation for ring A in 3-keto-4,4-dimethyl-19-nor-5α-steroids, made by Holker and Whalley (1961). [It should be noted that Holker and Whalley (1961) carried out their investigations in the liquid phase and this investigation has been in the solid state. The conformations in the two states may be different, but evidence from infra-red spectroscopy (Allinger and Da Rooge, 1962) suggests the same conformation for this type of molecule in both solid phase and solution.]

Rings B and C display the normal chair conformation of cyclohexane. The deviations of the atoms from the mean planes through the respective rings (table 2.9) are alternately positive and negative which is characteristic of the chair conformation. The root mean square deviations of the atoms from these planes are: ring B, 0.24 Å and ring C, 0.23 Å, which compare favourably with the values of 0.233 Å found in androsterone (Norton and Ohrt, 1964), 0.25 Å in 4-bromoestradiol (Norton, Kartha and Lu, 1964) and 0.25 Å in 3-keto-4,4-dimethyl-5α-androstane-17β-iodoacetate (Part II, Chapter 1 of this thesis).

Ring D adopts an envelope conformation with C(13) displaced 0.69 Å from the plane of C(14),C(15),C(16) and C(17) (plane 7, table 2.9). The envelope conformation with C(13) displaced is in agreement with the conformation of minimum energy calculated by Brutcher and Bauer (1962) for the cyclopentane ring in 17β-substituted steroids.

The packing of the molecules in the unit cell as projected on (001) is shown in figure 2.3. No short intermolecular distances are found (table 2.8) and the crystals appear to be held together only by van der Waals forces.

PART III

CRYSTAL STRUCTURE ANALYSES
OF
ORGANO-ARSENIC COMPOUNDS

Introduction.

Like most compounds of group VB elements with halogens, triaryl arsenic dihalides R_3AsX_2 are hydrolytically unstable. The hydrolysis has generally been regarded as occurring in two stages, corresponding to the replacement of one or both halogens by a hydroxyl group (Goddard, 1930). Corresponding to the series of arsines and arsenic dihalides, there is therefore a series of hydrolysis products generally referred to in the literature as the hydroxyhalides $R_3As(OH)Hal$ and the dihydroxides $R_3As(OH)_2$.

By analogy with the structures of other compounds containing penta-coordinate group VB elements, the hydrolysis products have been assumed to have trigonal bipyramidal structures in which the most electronegative substituents occupy the apical positions. Recent examination of the infra-red spectra and conductance properties of these compounds, however, suggests that they do not form a regular series in the structural sense (Harris and Inglis, 1967). It has been suggested by several authors (Jensen, 1943 ; Harris and Inglis, 1967) that the compound usually recognised as the dihydroxide is, in fact, the monohydrated triarylarsine oxide ($R_3AsO.H_2O$) whereas the conductance properties of the triaryl arsenic hydroxyhalides are more in keeping with a penta-coordinate arsenic atom (assumed to be a trigonal bipyramid). Failure to locate an absorption which could be attributed to the arsenic-halogen stretching frequency in the infra-red spectra of the hydroxyhalides, however, has cast grave doubts on the validity of the trigonal bipyramidal structure (Harris and Inglis, 1967). In order to obtain conclusive information about the structures of these compounds and to examine the bonding systems present, the crystal structure analyses of triphenylarsenic dihydroxide , triphenyl arsenic hydroxychloride and triphenyl arsenic hydroxybromide have been completed. The analysis of the dihydroxide confirms the suggestion that these compounds are indeed the triarylarsine oxide monohydrates in the solid state whilst the analyses of the hydroxychloride and hydroxybromide establish that the arsenic atom is tetrahedrally coordinated and that the compounds are more correctly regarded as 1:1 adducts of triphenylarsine oxide and the corresponding halogen acid.

Chapter 1.

The Crystal and Molecular Structure of Triphenylarsine Oxide Monohydrate.

1.1 Experimental.

The crystals used in this analysis were supplied by Dr. G.S. Harris and consisted of large colourless plates. Unit cell dimensions were derived from oscillation and zero-layer Weissenberg photographs (Cu Kα radiation, $\lambda = 1.5418$ Å) with the crystal rotating about its a-axis, and from precession photographs of the h0l reciprocal lattice net (Mo Kα radiation, $\lambda = 0.7107$ Å). The space group was determined uniquely from the systematic absences as $P2_1/c$ (C_{2h}^5, No. 14).

Crystal Data.

Triphenylarsine oxide monohydrate, $[C_{18}H_{15}AsO.H_2O]$.

F.W. = 340.1,

Monoclinic, a = 11.14 ± 2 Å, b = 16.65 ± 3 Å, c = 11.41 ± 2 Å,

$\beta = 130.7 \pm 0.5^\circ$.

U = 1608 Å^3, F(000) = 696 electrons

Dm = 1.39 gm.cm.$^{-3}$, Z = 4, Dx = 1.40 gm.cm.$^{-3}$,

Space Group $P2_1/c$ (C_{2h}^5, No. 14).

Linear absorption coefficient = 31.4 cm.$^{-1}$ (Cu Kα radiation).

The intensity data were estimated visually from equi-inclination Weissenberg photographs of the 0kl-9kl reciprocal lattice nets taken with Cu Kα radiation. The multiple film technique (Robertson, 1943) and interfilm scale factors of Rossman (1956) were used. The observed intensities were reduced to structure amplitudes by applying the appropriate Lorentz, polarisation and rotation factors (Tunell, 1939), yielding a total of 2187 independent structure amplitudes (table 1.5) which represents 60 % of the data available to Cu Kα radiation. No absorption correction was applied .

The data were initially placed on approximately absolute scale by comparison with the calculated structure factors. The final scale factor was determined by the least-squares refinement.

1.2 Solution and Refinement of the Structure.

In the space group $P2_1/c$, the equivalent positions are

$$\pm(x,y,z), \quad \pm(x, 1/2 - y, 1/2 + z).$$

A heavy atom situated in a general position will therefore give rise to the following vectors:

2x, 2y, 2z,	(single weight vector).
0, 1/2 + 2y, 1/2,	(double weight vector).
2x, 1/2, 1/2 + 2z,	(double weight vector).

Thus, in the three-dimensional Patterson function, three dominant peaks would be expected; viz. one peak in a general position, one peak on the Harker section at $v = 1/2$ and one peak on the Harker line at $u = 0$, $w = 1/2$.

The asymmetric unit of the Harker section (shown in figure 1.1) and the Harker line were calculated and interpretation, using the analytical expressions given above, led to the following coordinates for the heavy atom; 0.250, 0.144, 0.000.

When these coordinates are substituted in the structure factor expression for $P2_1/c$, it becomes apparent that the heavy atom will have no contribution to those planes for which $(h + k + l)$ is odd. This indicates that the heavy atom positions approximate to a higher symmetry than that of the true space group.

A set of structure factors, based on the heavy atom position, were calculated and the calculated phase angles used in conjunction with the observed amplitudes to evaluate an electron-density synthesis. The observed amplitudes were weighted using the method proposed by Woolfson (1956) which will give zero weight to those planes for which the heavy atom makes no contribution. The higher symmetry of the heavy atom arrangement was manifested in the presence of a diad-axis at (1/4, 0 , 0) and an inversion centre at (1/4, 1/4, 1/4) superimposed on the symmetry elements of the true space group. It was possible, however, to recognise the phenyl groups which were known to be present in the structure. Arbitrary selection of one set of phenyl groups over the corresponding pseudo-set was shown, by consideration of the structure factor expression, to result only in a consistent inversion of the phase angles of the structure factors. After selection of the first benzene ring, it was possible to eliminate the pseudo-rings on the basis of inordinate steric overcrowding of the molecule and impossibly short intermolecular contacts.

Coordinates were derived from the electron-density synthesis for two of the three phenyl groups present in the molecule using a peak searching program written by Dr. D.R. McGregor (1967). Inclusion of the thirteen atoms (1 arsenic and 12 carbon) in the structure factor calculation reduced the agreement index R from 0.64 with the heavy atom only to 0.36. A second electron-density distribution was calculated using the improved phase angles and the modified values of |Fo| (Woolfson, 1956), and showed no pseudo-symmetry. It was possible to derive the coordinates of nineteen of the twenty-one atoms present in the structure from this map using the peak-searching program (McGregor, 1967). The process was repeated until all the atoms were located and is summarised in table 1.1 part A.

The structure was refined in eight cycles of minimisation of the function $\sum w(|Fo| - |Fc|)^2$ by the method of least-squares. D.R. Pollard's least-squares program (1968) was used in the initial stages. Each reciprocal lattice net was given an individual scale factor and each atom a single isotropic temperature factor. The observations were all given unit weight. After four cycles of full-matrix refinement, the R-factor was reduced to 0.150.

The Glasgow SFLS program (Cruickshank and Smith, 1965) was then used to continue the refinement. Lingafelter and Donohue (1966) have shown that, if the observations are collected up only one axis, there is insufficient information to permit the refinement of individual layer scale factors and anisotropic thermal parameters. On the assumption that the data were on the same relative scale at the conclusion of the isotropic refinement, it was therefore decided to refine anisotropic thermal parameters and a single overall scale factor. Consequently least-squares totals were accumulated in the fifth cycle to permit calculation of the anisotropic thermal parameters for all atoms. As a result of allowing for the anisotropic vibrations of the atoms, R fell from 0.145 in cycle five to 0.127 in cycle six (the block-diagonal approximation to the matrix of the normal equations was used in these cycles).

Using the structure factors calculated in cycle six a low order (Fo - Fc) map was calculated using only those planes for which $\sin \theta / \lambda < 0.36$. In this map it was possible to locate the hydrogen atoms bonded to the phenyl groups but it was not possible to locate the hydrogen atoms of the molecule of water of crystallisation present in the structure. The aromatic hydrogen atoms were therefore included in the structure factor calculation in calculated positions but were not refined.

A further two cycles of refinement in which the block-diagonal approximation to the normal matrix was used produced convergence. The R-factor over the 2187 independent reflections was reduced to 0.114.

The weighting scheme applied in the last three cycles of refinement was of the form

$$w = 1 / [5.16 + |Fo| + 0.0131 |Fo|^2 + 0.0002 |Fo|^3]$$

Cruickshank (1961). Examination of a bivariate analysis of $\sum w\Delta^2$ by magnitude of $|Fo|$ and $\sin^2\theta/\lambda^2$ showed a reasonably flat distribution for the average values of $w\Delta^2$.

The structure factors calculated in the final cycle of least-squares refinement were used to phase electron-density and difference syntheses. The electron-density synthesis is shown by means of a composite contour diagram in figure 1.2 which also explains the numbering system adopted for the molecule. The standard deviation of the electron density, calculated using the approximate formula of Cruickshank (1949), was 0.2 $e/\mathring{A}^3$ and in terms of this no significant features appeared in the difference map indicating that the refinement was complete.

The atomic scattering factors used throughout the analysis were those of Hoerni and Ibers (1954) for carbon and oxygen; Freeman and Watson (1961) for arsenic ; Stewart, Davidson and Simpson (1965) for hydrogen.

The atomic parameters are presented in tables 1.2 (fractional coordinates), 1.3(anisotropic thermal parameters) and 1.4 (assumed hydrogen parameters). The observed and calculated structure factors, at the conclusion of the refinement, are contained in table 1.5 and an analysis of the structure factor data by layer line index and magnitude of $|Fo|$ in table 1.6. Atomic coordinates in Ångstroms referred to orthogonal axes defined by X' parallel to a^*, Y parallel to b and Z' parallel to c are given in table 1.7 for the heavier atoms present in the structure. Tables 1.8, 1.9, 1.10, 1.11 and 1.12 contain bond lengths, interbond angles, intramolecular distances, intermolecular distances and mean planes through portions of the molecule respectively. Estimated standard deviations, where quoted, are in units of the last decimal place of the quantity to which they refer.

The packing of the molecules in the unit cell as viewed down the b-axis is shown in figure 1.3.

TABLES AND DIAGRAMS

Figure 1.1

Harker section at v = 1/2, contours at arbitrary levels.

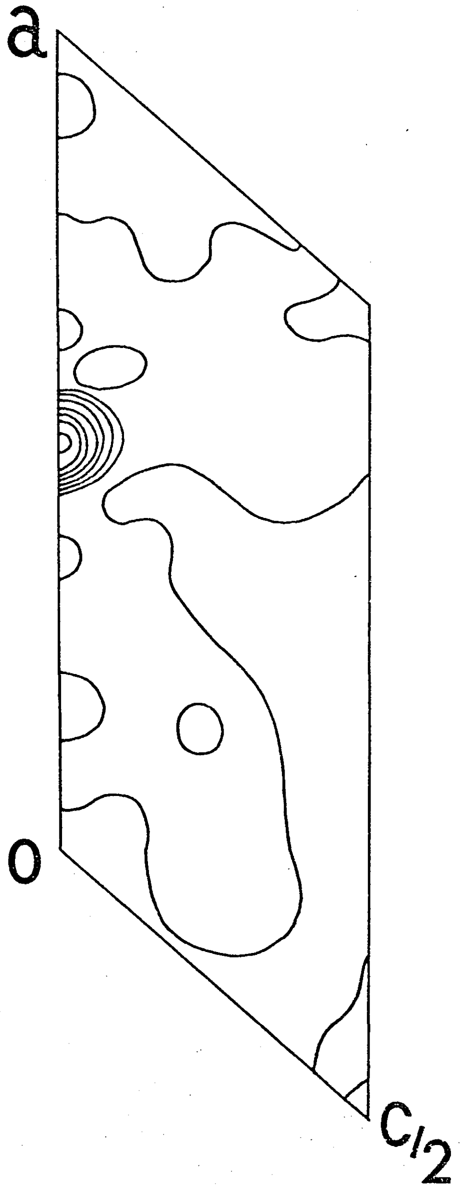
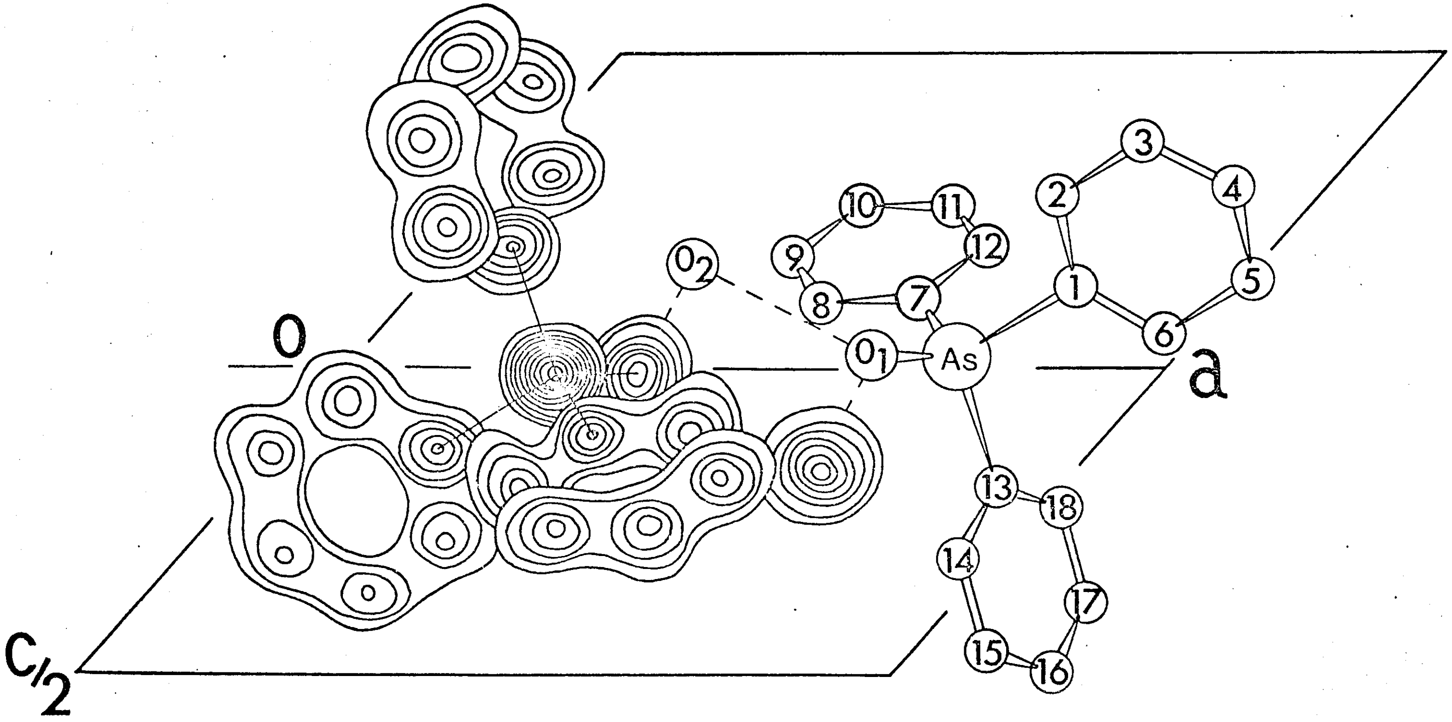

a
0
c/2

Figure 1.2

Composite electron-density diagram, projected on (010). Contours are at intervals of 1 $e/\mathring{A}^{3}$ except around the arsenic atom which is contoured at intervals of 5 $e/\mathring{A}^{3}$.



0
a
c/2
As
O_1
O_2
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18

Figure 1.3

The packing of the molecules as projected down b. Molecules in the upper half of the cell are drawn with bolder lines; hydrogen-bonding is shown by means of broken lines.

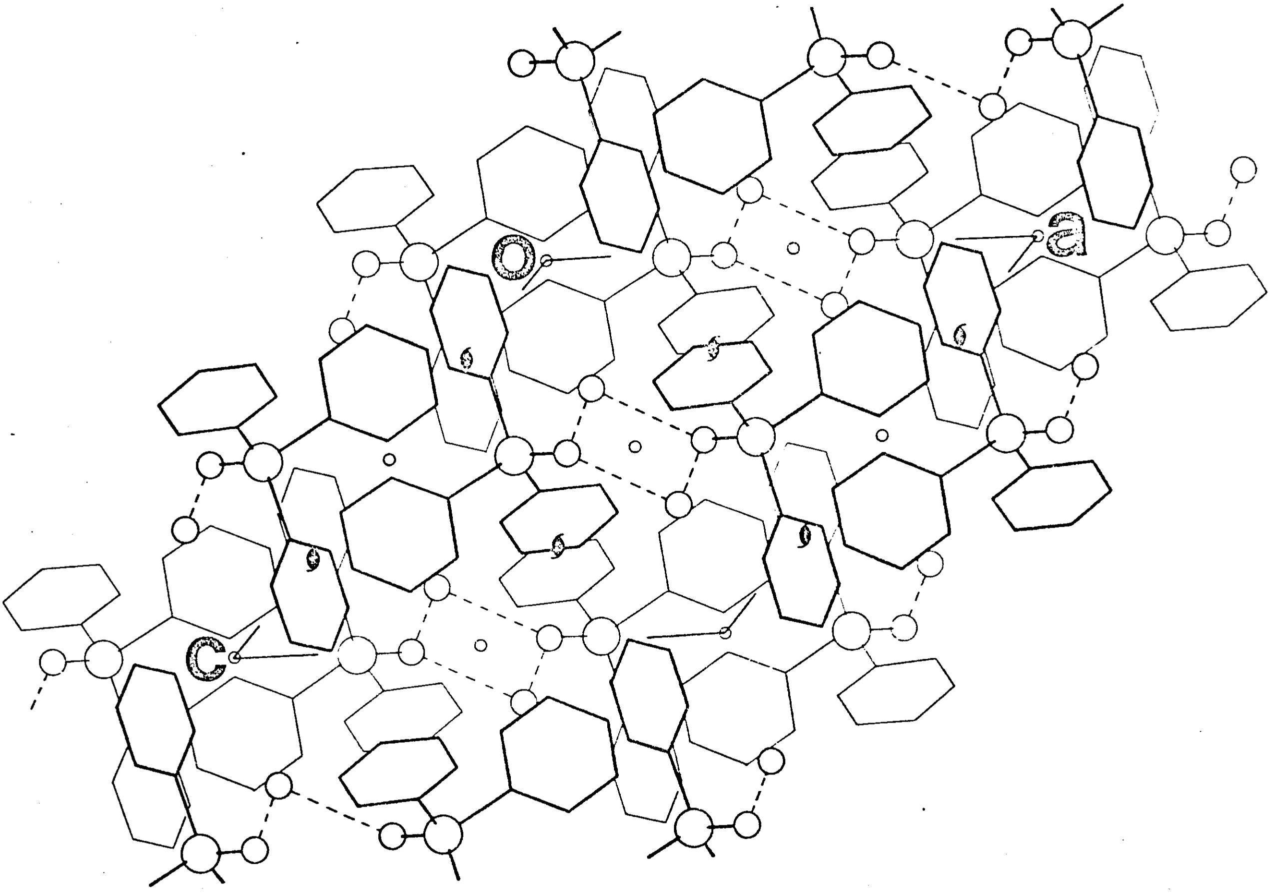

O
a
c

TABLE 1.1

COURSE OF ANALYSIS

(A) Fourier Refinement

	Atoms included in structure factor calculation.	R
1	As only	0.64
2	As + 12 C	0.364
3	As + 16 C + 2 O	0.320
4	As + 18 C + 2 O	0.299

(B) Least-squares refinement.

Cycles	Final R	Final R'	*
1 - 4	0.150	-	(a)
5	0.145	-	(b)
6	0.127	0.030	(c)
7 - 9	0.114	0.021	(d)

* (a) Full-matrix, isotropic, layer scales, unit weights.

(b) As above, weighting scheme applied.

(c) Block-diagonal, anisotropic, one scale factor.

(d) As above, hydrogen atoms included but not refined.

TABLE 1.2

FRACTIONAL COORDINATES AND E.S.D.S.

ATOM	X/a	Y/b	Z/c
As(1)	0.2588 ± 1	0.1448 ± 1	0.0114 ± 1
O(1)	0.3678 ± 8	0.0770 ± 4	0.0139 ± 8
O(2)	0.6898 ± 9	0.0583 ± 5	0.1595 ± 9
C(1)	0.1927 ± 12	0.1081 ± 5	0.1213 ± 11
C(2)	0.3070 ± 14	0.0840 ± 8	0.2684 ± 14
C(3)	0.2649 ± 20	0.0557 ± 9	0.3533 ± 15
C(4)	0.1053 ± 21	0.0550 ± 9	0.2823 ± 17
C(5)	-0.0067 ± 22	0.0782 ± 9	0.1366 ± 21
C(6)	0.0369 ± 16	0.1059 ± 8	0.0522 ± 17
C(7)	0.3740 ± 10	0.2424 ± 5	0.1090 ± 9
C(8)	0.4855 ± 12	0.2632 ± 6	0.1001 ± 11
C(9)	0.5733 ± 14	0.3343 ± 7	0.1745 ± 13
C(10)	0.5421 ± 14	0.3793 ± 7	0.2531 ± 12
C(11)	0.4308 ± 15	0.3601 ± 6	0.2597 ± 13
C(12)	0.3426 ± 15	0.2898 ± 6	0.1879 ± 13
C(13)	0.0772 ± 11	0.1677 ± 7	-0.1944 ± 11
C(14)	0.0427 ± 13	0.1175 ± 7	-0.3118 ± 11
C(15)	-0.0917 ± 20	0.1356 ± 9	-0.4626 ± 15
C(16)	-0.1942 ± 16	0.1964 ± 10	-0.4943 ± 14
C(17)	-0.1554 ± 17	0.2446 ± 9	-0.3784 ± 20
C(18)	-0.0258 ± 13	0.2321 ± 7	-0.2292 ± 14

TABLE 1.3

ANISOTROPIC TEMPERATURE FACTORS AND E.S.D.S.

ATOM	U11	U22	U33	2U23	2U31	2U12
As(1)	0.0511	0.0588	0.0488	0.0010	0.0595	0.0197
	6	5	5	8	9	8
O(1)	0.0584	0.0741	0.0841	-0.0160	0.0999	0.0357
	38	46	47	71	75	67
O(2)	0.0671	0.0671	0.0867	-0.0314	0.0774	-0.0018
	41	48	51	77	80	70
C(1)	0.0711	0.0457	0.0667	-0.0060	0.0993	-0.0011
	58	48	52	78	99	82
C(2)	0.0803	0.0938	0.0701	0.0043	0.0894	-0.0356
	73	88	66	118	123	129
C(3)	0.1374	0.1012	0.0715	-0.0052	0.1100	-0.0838
	123	105	75	141	167	185
C(4)	0.1636	0.0865	0.1166	-0.0512	0.2218	-0.0902
	145	94	110	155	233	184
C(5)	0.1494	0.0918	0.1517	-0.0001	0.2483	-0.0084
	137	99	142	182	261	184
C(6)	0.0853	0.0919	0.1078	0.0550	0.1461	0.0370
	80	88	93	144	156	134
C(7)	0.0493	0.0490	0.0434	-0.0033	0.0537	0.0038
	45	46	39	64	73	69
C(8)	0.0565	0.0673	0.0663	0.0031	0.0839	0.0110
	52	61	56	88	97	88
C(9)	0.0706	0.0749	0.0764	0.0195	0.0972	-0.0091
	65	69	66	103	117	104
C(10)	0.0775	0.0684	0.0606	0.0015	0.0740	-0.0069
	70	65	57	92	110	105

C(11)	0.0991	0.0554	0.0749	-0.0046	0.1228	-0.0022
	81	59	62	95	128	104
C(12)	0.0958	0.0634	0.0876	-0.0329	0.1483	-0.0127
	77	61	71	102	135	109
C(13)	0.0483	0.0757	0.0631	0.0273	0.0692	0.0010
	49	64	54	90	90	86
C(14)	0.0729	0.0747	0.0540	0.0023	0.0655	-0.0355
	65	69	52	90	102	104
C(15)	0.1201	0.1190	0.0688	-0.0194	0.1089	-0.1107
	107	112	70	138	155	177
C(16)	0.0734	0.1260	0.0652	0.0713	0.0317	-0.0628
	72	120	69	138	114	151
C(17)	0.0899	0.1086	0.1361	0.1418	0.1417	0.0751
	89	112	125	196	188	163
C(18)	0.0688	0.0756	0.0894	0.0446	0.1039	0.0442
	65	74	76	117	123	108

TABLE 1.4

ASSUMED FRACTIONAL COORDINATES AND TEMPERATURE FACTORS OF HYDROGEN ATOMS

ATOM	X/a	Y/b	Z/c	Uiso
H(2)	0.430	0.084	0.319	0.0500
H(3)	0.355	0.037	0.473	0.0500
H(4)	0.077	0.036	0.352	0.0500
H(5)	-0.135	0.074	0.084	0.0500
H(6)	-0.055	0.125	-0.070	0.0500
H(8)	0.508	0.225	0.037	0.0500
H(9)	0.663	0.353	0.167	0.0500
H(10)	0.613	0.433	0.314	0.0500
H(11)	0.409	0.400	0.320	0.0500
H(12)	0.255	0.271	0.197	0.0500
H(14)	0.120	0.067	-0.284	0.0500
H(15)	-0.117	0.101	-0.559	0.0500
H(16)	-0.306	0.204	-0.610	0.0500
H(17)	-0.231	0.297	-0.409	0.0500
H(18)	-0.001	0.271	-0.138	0.0500

The hydrogen atoms are numbered to correspond to the carbon atom to which they are bonded.

TABLE 1.5

FINAL OBSERVED AND CALCULATED STRUCTURE AMPLITUDES.

H	K	L	Fo	Fc
0	20	0	9.1	8.9
0	18	0	9.4	-9.3
0	14	0	34.2	33.0
0	10	0	31.5	-75.1
0	8	0	46.1	40.5
0	6	0	51.9	-43.1
0	4	0	142.4	-149.6
0	2	0	68.2	-74.2
0	17	1	15.6	-17.3
0	15	1	14.9	12.6
0	13	1	38.3	33.3
0	11	1	30.8	-29.2
0	10	1	31.5	26.4
0	9	1	29.9	-23.6
0	8	1	8.1	6.9
0	7	1	62.6	61.2
0	6	1	47.5	-37.9
0	5	1	79.9	-77.9
0	4	1	114.0	-126.7
0	3	1	71.5	-75.4
0	2	1	16.6	14.8
0	1	1	95.3	105.4
0	20	2	7.4	8.4
0	18	2	12.2	-16.0
0	16	2	11.0	-9.5
0	14	2	28.5	28.3
0	12	2	6.2	3.1
0	11	2	8.3	8.2
0	10	2	39.7	-36.0
0	9	2	15.9	-13.9
0	8	2	41.3	35.5
0	7	2	47.4	40.2
0	6	2	50.3	52.6
0	5	2	22.6	-12.8
0	4	2	65.8	-69.4
0	2	2	32.6	-29.5
0	1	2	94.2	-109.4
0	0	2	34.2	40.6
0	17	3	17.0	-19.0
0	15	3	13.8	11.8
0	13	3	28.2	25.5
0	12	3	7.4	7.1
0	11	3	37.0	-34.1
0	10	3	14.4	-12.6
0	9	3	16.1	-13.4
0	7	3	55.3	56.6
0	6	3	31.8	26.9
0	5	3	23.8	-20.6
0	4	3	12.0	11.9
0	3	3	66.3	-75.7
0	2	3	63.8	-74.2
0	1	3	57.2	69.7
0	18	4	10.2	-11.4
0	16	4	8.6	-10.1
0	14	4	28.5	34.0
0	12	4	9.3	-8.2
0	11	4	14.1	13.3
0	10	4	32.4	-31.3
0	9	4	15.7	-16.9
0	8	4	21.6	24.3
0	7	4	10.8	9.2
0	6	4	55.7	56.3
0	5	4	7.0	8.3
0	4	4	80.3	-94.2
0	3	4	11.0	-11.1
0	2	4	8.9	-9.8
0	1	4	31.4	-36.0
0	0	4	71.6	86.1
0	17	5	11.6	-11.2
0	16	5	7.2	-5.9
0	15	5	7.0	6.0
0	14	5	6.4	-5.2
0	13	5	26.6	25.2
0	11	5	26.2	-26.7
0	10	5	9.2	8.6
0	9	5	22.0	-19.6
0	8	5	11.0	-11.4
0	7	5	42.6	44.3
0	6	5	12.7	13.3
0	4	5	15.8	15.5
0	3	5	38.9	-42.8
0	2	5	6.7	-8.7
0	1	5	26.8	28.0
0	16	6	6.8	-5.0
0	15	6	10.8	-9.2
0	14	6	17.9	16.1
0	13	6	14.9	13.2
0	10	6	22.2	-24.0
0	9	6	8.6	-8.6
0	7	6	9.1	-7.9
0	6	6	34.2	33.4
0	5	6	29.6	27.6
0	4	6	42.1	-40.9
0	3	6	17.4	-14.1
0	1	6	7.6	-9.0
0	0	6	44.1	58.2
0	16	7	4.9	-4.6
0	13	7	13.0	12.3
0	12	7	14.6	13.0
0	11	7	14.4	-14.9
0	10	7	7.5	-5.8
0	8	7	11.9	-12.3
0	7	7	22.2	20.4
0	6	7	9.4	9.0
0	5	7	15.1	-15.0
0	3	7	27.3	-26.0
0	2	7	12.4	-12.8
0	1	7	32.4	27.2
0	14	8	8.6	9.0
0	13	8	7.8	6.5
0	11	8	8.3	5.8
0	10	8	15.4	-15.0
0	9	8	7.8	-7.2
0	8	8	14.2	12.6
0	6	8	8.4	6.5
0	5	8	19.2	18.6
0	4	8	20.5	-18.8
0	3	8	16.0	-15.1
0	2	8	9.0	-7.5
0	1	8	16.5	-16.9
0	0	8	33.5	33.0
0	12	9	5.6	6.3
0	11	9	8.5	-8.9
0	7	9	16.2	17.3
0	6	9	6.7	5.5
0	5	9	6.0	-4.7
0	3	9	21.4	-18.9
0	2	9	18.2	-17.3
0	1	9	14.2	12.5
0	9	10	4.4	-6.4
0	6	10	7.6	6.3
0	5	10	8.3	8.3
0	4	10	6.8	-6.4
0	2	10	8.3	-8.2
0	1	10	11.1	-8.4
0	0	10	17.6	14.8
0	2	11	5.8	-3.0
0	1	11	5.0	4.9
1	0	8	21.6	-17.9
1	0	6	10.2	-11.8
1	0	4	41.3	-42.2
1	0	2	20.8	15.7
1	0	-6	24.7	22.7
1	0	-10	13.6	13.7
1	1	9	13.2	-10.7
1	1	8	21.6	-19.8
1	1	7	10.3	-9.3
1	1	6	30.5	-29.6
1	1	5	36.7	-32.2
1	1	4	51.9	-59.0
1	1	3	31.0	-27.6
1	1	2	21.1	-26.3
1	1	1	32.5	-36.3
1	1	-2	61.0	-78.5
1	1	-4	52.3	-51.3
1	1	-5	7.5	5.5
1	1	-6	37.0	-37.3
1	1	-7	9.4	8.6
1	1	-8	22.0	-19.9
1	1	-9	14.5	13.7
1	1	-10	13.0	-14.1
1	1	-11	8.0	6.4
1	2	9	18.4	-18.1
1	2	7	23.5	-22.0
1	2	5	51.7	-53.3
1	2	4	30.4	-27.3
1	2	3	77.9	-103.5
1	2	2	35.6	34.0
1	2	1	116.5	-171.2
1	2	0	19.0	19.4
1	2	-1	93.1	-115.1
1	2	-2	36.9	-33.6
1	2	-3	118.6	-151.8
1	2	-4	9.2	-8.1
1	2	-5	53.8	-62.9
1	2	-6	10.5	-10.5
1	2	-7	55.7	-60.0
1	2	-9	22.0	-20.8
1	2	-11	15.6	-14.2
1	3	9	12.6	12.9
1	3	8	9.4	-7.9
1	3	7	15.9	16.8
1	3	6	21.3	-18.5
1	3	5	16.3	17.7
1	3	4	39.8	-38.1
1	3	3	52.1	55.9
1	3	2	84.3	-90.9
1	3	1	7.1	-4.0
1	3	0	84.2	-104.2
1	3	-2	19.1	16.8
1	3	-3	7.2	5.2
1	3	-4	51.4	-56.2
1	3	-5	23.7	-23.5
1	3	-6	19.5	-19.1
1	3	-7	14.2	-14.7
1	3	-8	27.3	-26.7
1	3	-9	14.4	-13.8
1	3	-11	10.2	-9.6
1	4	9	7.0	6.5
1	4	8	21.1	19.7
1	4	7	17.3	16.5
1	4	6	10.4	10.0
1	4	5	28.0	24.2
1	4	4	52.2	46.5
1	4	3	9.3	7.0
1	4	2	22.7	-22.0
1	4	1	70.2	67.6
1	4	0	23.0	-17.8
1	4	-1	73.1	86.8
1	4	-2	59.8	-73.0
1	4	-4	15.3	8.7
1	4	-5	49.9	47.6
1	4	-6	15.6	-15.0
1	4	-7	16.7	17.9
1	4	-8	10.9	-10.1
1	4	-9	12.5	10.1
1	4	-10	14.4	-15.4
1	4	-11	6.2	5.3
1	5	8	14.2	12.6
1	5	6	39.8	35.5
1	5	5	23.8	17.3
1	5	4	53.9	54.8
1	5	3	15.9	-12.8
1	5	2	72.6	85.2
1	5	1	37.2	29.2
1	5	0	73.4	72.0
1	5	-1	28.4	-26.9
1	5	-2	52.4	62.7
1	5	-3	18.2	-15.7
1	5	-4	38.3	47.1
1	5	-5	17.2	14.7
1	5	-6	39.9	43.4
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7	10	-5	16.4	14.6
7	12	-5	15.2	-15.9
7	14	-5	4.4	5.6
7	16	-5	11.3	10.5
7	1	-6	51.7	41.3
7	2	-6	13.1	-10.9
7	3	-6	29.4	24.2
7	4	-6	16.6	13.5
7	5	-6	55.4	-47.6
7	7	-6	11.6	11.7
7	9	-6	31.5	30.0
7	10	-6	12.6	-12.0
7	11	-6	20.5	-22.2
7	12	-6	7.9	6.9
7	13	-6	8.8	-7.8
7	14	-6	5.4	-6.0
7	15	-6	12.4	12.0
7	1	-7	15.4	-17.2
7	2	-7	66.8	55.1
7	3	-7	11.9	9.0
7	4	-7	23.5	-18.7
7	5	-7	13.4	12.1
7	6	-7	32.4	-30.9
7	8	-7	27.9	29.3
7	9	-7	8.5	-9.2
7	10	-7	13.5	13.0
7	11	-7	8.9	9.4
7	12	-7	20.0	-22.9
7	15	-7	6.3	-6.1
7	16	-7	9.9	11.3
7	1	-8	56.5	54.3
7	2	-8	5.1	-4.7
7	3	-8	13.8	10.0
7	5	-8	34.8	-32.7
7	6	-8	9.5	6.4
7	7	-8	7.4	-5.7
7	8	-8	6.2	-3.6
7	9	-8	24.3	29.0
7	10	-8	5.6	4.5
7	11	-8	11.8	-12.3
7	13	-8	7.8	-8.5
7	15	-8	10.9	10.0
7	1	-9	7.9	-8.4
7	2	-9	23.7	25.7
7	4	-9	11.7	-12.5
7	5	-9	10.4	9.2
7	6	-9	23.3	-24.1
7	8	-9	17.1	17.9
7	10	-9	5.6	5.6
7	12	-9	14.2	-17.8
7	14	-9	5.9	7.5
7	15	-9	3.3	-5.0
7	16	-9	7.6	5.5
7	0	-10	29.0	-27.1
7	1	-10	13.4	16.2
7	2	-10	10.2	9.3
7	3	-10	8.7	7.4
7	4	-10	8.2	8.1
7	5	-10	24.5	-25.2
7	7	-10	5.6	4.9
7	8	-10	6.4	-6.0
7	9	-10	13.7	15.2
7	10	-10	4.6	4.2
7	11	-10	7.6	-9.1
7	13	-10	7.2	-5.2
7	14	-10	5.5	-5.2
7	15	-10	7.5	9.3
7	1	-11	8.9	-10.4
7	2	-11	26.0	26.7
7	3	-11	10.5	12.3
7	4	-11	12.6	-14.8
7	6	-11	13.6	-15.5
7	7	-11	8.6	-10.8
7	8	-11	11.5	15.2
7	10	-11	5.6	6.1
7	11	-11	6.8	7.0
7	12	-11	10.3	-9.9
7	13	-11	4.1	-4.3
7	1	-12	11.8	13.0
7	3	-12	10.4	12.1
7	5	-12	19.2	-23.6
7	7	-12	3.9	3.6
7	9	-12	9.7	11.1
7	10	-12	3.9	4.7
7	11	-12	3.5	-3.9
7	1	-13	4.7	-4.7
7	2	-13	11.8	9.4
7	3	-13	4.5	5.9
7	4	-13	4.4	-4.2
7	6	-13	7.4	-7.1
7	7	-13	3.7	-3.4
7	8	-13	3.9	5.0
7	0	-14	5.8	-6.4
7	1	-14	7.5	7.1
7	3	-14	3.9	4.2
7	4	-14	7.2	9.6
7	5	-14	5.2	-7.5
8	0	0	19.0	20.3
8	1	0	3.7	-5.0
8	2	0	8.9	-9.1
8	3	0	10.4	-13.8
8	4	0	11.1	-11.0
8	5	0	14.5	17.4
8	6	0	6.4	5.2
8	8	0	11.2	14.3
8	9	0	6.9	-6.7
8	10	0	19.7	-17.5
8	14	0	8.4	9.9
8	1	1	14.4	14.1
8	2	1	10.3	-11.4
8	3	1	16.7	-17.0
8	4	1	10.1	12.4
8	5	1	7.1	-7.7
8	6	1	6.5	7.2
8	7	1	16.3	15.8
8	8	1	9.7	-9.5
8	11	1	9.8	-8.8
8	12	1	3.8	4.9
8	0	2	16.2	21.5
8	1	2	9.9	-11.7
8	2	2	3.3	-2.9
8	3	2	3.3	4.9
8	4	2	13.8	-13.0
8	5	2	8.3	7.1
8	6	2	5.7	4.6
8	8	2	6.5	6.7
8	9	2	7.0	-6.9
8	10	2	8.5	-13.0
8	2	3	5.2	-7.7
8	3	3	10.0	-11.1
8	7	3	5.1	6.5
8	1	-1	19.1	20.3
8	2	-1	6.9	-8.3
8	3	-1	27.2	-31.5
8	6	-1	3.7	2.3
8	7	-1	17.8	16.4
8	8	-1	3.7	-4.2
8	9	-1	4.5	-5.5
8	11	-1	13.9	-11.7
8	12	-1	4.0	4.2
8	13	-1	12.7	13.1
8	14	-1	4.6	-4.8
8	15	-1	3.0	3.3
8	0	-2	40.5	48.5
8	1	-2	12.0	-11.2
8	3	-2	8.1	-7.1
8	4	-2	40.0	-44.3
8	5	-2	13.3	14.0
8	6	-2	19.4	20.3
8	7	-2	8.7	-9.9
8	8	-2	13.4	13.3
8	10	-2	17.9	-20.1
8	14	-2	15.1	15.7
8	15	-2	5.6	-4.8
8	16	-2	3.3	-4.1
8	1	-3	34.3	36.9
8	2	-3	16.3	-14.7
8	3	-3	33.7	-35.0
8	4	-3	10.1	11.5
8	5	-3	15.1	-14.6
8	6	-3	13.6	14.5
8	7	-3	26.5	26.5
8	8	-3	12.2	-14.2
8	9	-3	9.9	-8.8
8	11	-3	15.5	-15.5
8	13	-3	11.9	8.8
8	14	-3	5.6	-4.1
8	15	-3	8.9	9.3
8	16	-3	2.5	3.3
8	17	-3	5.1	-6.8
8	0	-4	39.1	43.4
8	1	-4	9.1	-7.7
8	2	-4	3.0	2.6
8	4	-4	44.9	-44.1
8	5	-4	17.4	13.9
8	6	-4	27.8	24.2
8	8	-4	13.5	12.7
8	9	-4	4.4	-3.0
8	10	-4	16.9	-15.8
8	12	-4	5.2	-5.5
8	13	-4	5.0	-4.7
8	14	-4	13.2	15.7
8	15	-4	3.8	-4.5
8	16	-4	7.1	-7.7
8	17	-4	3.4	2.4
8	1	-5	28.9	35.2
8	2	-5	5.6	5.7
8	3	-5	53.3	-43.0
8	4	-5	13.3	9.7
8	5	-5	25.5	-20.6
8	6	-5	5.1	-3.1
8	7	-5	31.3	27.0
8	8	-5	5.8	-5.0
8	10	-5	10.8	10.5
8	11	-5	14.5	-12.1
8	13	-5	14.1	14.3
8	14	-5	8.3	-7.5
8	15	-5	4.4	3.3
8	17	-5	8.2	-10.0
8	2	-6	6.3	-4.0
8	3	-6	22.8	20.9
8	4	-6	67.8	-50.6
8	5	-6	10.7	-11.1
8	6	-6	20.0	14.4
8	8	-6	26.2	25.2
8	9	-6	8.6	-9.6
8	10	-6	20.4	-24.2
8	11	-6	5.2	4.5
8	13	-6	3.5	-4.2
8	14	-6	17.7	18.0
8	16	-6	3.4	-4.5
8	1	-7	44.3	34.8
8	2	-7	17.9	15.3
8	3	-7	59.2	-47.7
8	4	-7	10.6	7.2
8	5	-7	6.8	-3.8
8	7	-7	35.1	33.1
8	8	-7	9.4	-9.8
8	9	-7	12.3	-13.0
8	11	-7	13.6	-12.4
8	13	-7	10.9	12.0
8	14	-7	4.7	-4.0
8	15	-7	4.3	5.6
8	17	-7	8.1	-11.8
8	1	-8	7.4	-6.1
8	2	-8	17.3	-14.5
8	3	-8	16.2	12.4
8	4	-8	33.4	-29.9
8	6	-8	18.6	17.1
8	7	-8	9.5	-10.5
8	8	-8	16.5	18.7
8	10	-8	18.5	-22.9
8	11	-8	4.5	3.7
8	12	-8	3.6	4.8
8	13	-8	3.4	-2.7
8	14	-8	11.7	12.3
8	16	-8	5.4	-6.1
8	1	-9	21.5	17.6
8	2	-9	11.0	10.2
8	3	-9	22.3	-19.5
8	5	-9	9.5	-8.5
8	6	-9	7.1	-5.5
8	7	-9	20.2	19.5
8	8	-9	4.5	-4.5
8	9	-9	9.0	-9.8
8	10	-9	5.8	5.6
8	11	-9	10.5	-10.3
8	13	-9	11.9	11.2
8	15	-9	5.6	5.0
8	16	-9	4.0	5.9
8	0	-10	40.4	37.5
8	1	-10	17.7	19.1
8	2	-10	13.9	-11.3
8	3	-10	8.7	9.1
8	4	-10	17.4	-16.3
8	6	-10	12.6	11.3
8	8	-10	11.4	11.8
8	9	-10	6.7	7.0
8	10	-10	14.7	-19.5
8	11	-10	3.4	-5.5
8	13	-10	5.7	-5.1
8	14	-10	10.6	11.9
8	1	-11	17.0	15.4
8	2	-11	7.3	7.6
8	3	-11	22.8	-24.4
8	4	-11	4.5	4.9
8	5	-11	3.7	-2.8
8	6	-11	4.5	-5.2
8	7	-11	17.3	19.5
8	8	-11	5.0	5.0
8	9	-11	4.1	-4.7
8	11	-11	9.6	-9.8
8	12	-11	5.4	-4.1
8	13	-11	6.4	8.3
8	0	-12	14.2	12.8
8	1	-12	7.6	7.9
8	4	-12	19.6	-19.3
8	6	-12	9.5	11.4
8	8	-12	9.0	8.0
8	9	-12	5.5	5.2
8	10	-12	8.0	-7.4
8	1	-13	9.3	7.7
8	2	-13	7.5	7.7
8	3	-13	13.2	-12.2
8	6	-13	4.9	-3.9
8	7	-13	8.9	7.9
8	9	-13	3.6	-4.2
8	0	-14	10.4	9.7
8	1	-14	6.0	5.5
8	3	-14	7.1	7.0
8	4	-14	8.0	-8.9
8	5	-14	5.0	-5.9
8	6	-14	4.1	6.0
9	0	0	11.5	-12.4
9	1	0	15.1	-16.4
9	2	0	4.0	5.5
9	3	0	7.9	-7.5
9	4	0	4.5	5.0
9	5	0	12.7	10.9
9	9	0	10.3	-11.2
9	10	0	5.9	5.8
9	11	0	5.3	5.5
9	1	1	7.2	-6.5
9	2	1	12.9	-12.4
9	3	1	8.7	9.3
9	6	1	6.9	7.6
9	7	1	4.9	-6.7
9	8	1	6.6	-6.5
9	9	1	4.6	5.1
9	1	-1	5.7	-6.0
9	2	-1	19.6	-21.2
9	3	-1	10.1	10.9
9	4	-1	4.7	3.9
9	6	-1	12.0	15.4
9	7	-1	7.2	-8.3
9	8	-1	11.9	-13.0
9	10	-1	4.1	-4.0
9	12	-1	11.5	11.9
9	0	-2	5.4	-5.1
9	1	-2	29.3	-33.5
9	3	-2	9.0	-10.2
9	4	-2	19.9	10.0
9	5	-2	15.2	17.3
9	6	-2	5.7	-6.8
9	7	-2	7.4	9.6
9	9	-2	15.0	-15.3
9	11	-2	16.5	8.9
9	13	-2	6.0	5.5
9	1	-3	5.1	-7.4
9	2	-3	35.3	-37.5
9	3	-3	10.7	11.1
9	4	-3	13.6	13.0
9	6	-3	22.5	22.3
9	7	-3	7.3	-8.7
9	8	-3	18.6	-17.0
9	12	-3	14.0	13.0
9	0	-4	10.3	-10.4
9	1	-4	28.9	-29.4
9	3	-4	19.6	-19.3
9	4	-4	12.7	12.4
9	5	-4	30.4	30.0
9	6	-4	13.7	-15.1
9	8	-4	10.5	11.4
9	9	-4	15.4	-14.4
9	11	-4	6.5	5.7
9	12	-4	5.4	-5.9
9	13	-4	10.4	9.0
9	15	-4	9.0	-10.1
9	1	-5	4.3	-3.0
9	2	-5	35.8	-40.4
9	3	-5	3.8	2.4
9	4	-5	4.3	-2.4
9	5	-5	10.2	-8.8
9	6	-5	30.4	27.8
9	7	-5	5.3	4.8
9	8	-5	16.2	-13.5
9	9	-5	11.6	11.7
9	10	-5	5.8	-4.6
9	12	-5	16.3	17.2
9	13	-5	8.0	-5.7
9	16	-5	8.4	-11.2
9	3	-6	40.4	-39.3
9	4	-6	5.7	-4.0
9	5	-6	34.0	30.5
9	6	-6	10.3	-9.6
9	7	-6	6.0	-3.9
9	8	-6	4.5	2.7
9	9	-6	18.3	-14.4
9	10	-6	6.6	5.3
9	11	-6	10.1	9.7
9	12	-6	4.6	-4.4
9	13	-6	16.9	15.6
9	15	-6	11.1	-10.4
9	1	-7	8.8	11.5
9	2	-7	42.5	-36.9
9	3	-7	10.2	9.2
9	4	-7	23.5	20.0
9	5	-7	13.6	-10.9
9	6	-7	17.2	11.6
9	7	-7	7.5	-7.9
9	8	-7	23.3	-20.8
9	10	-7	7.4	-6.1
9	12	-7	20.3	24.4
9	14	-7	5.6	-6.0
9	16	-7	8.4	-10.5
9	1	-8	22.3	-15.1
9	1	-8	19.0	-15.1
9	3	-8	13.0	-8.7
9	4	-8	20.7	-17.6
9	5	-8	30.4	29.0
9	7	-8	8.2	-7.9
9	9	-8	15.4	-13.0
9	11	-8	11.9	12.6
9	12	-8	7.7	-9.2
9	13	-8	6.6	6.5
9	15	-8	10.8	-11.8
9	1	-9	8.5	6.2
9	2	-9	35.2	-32.3
9	3	-9	8.9	-8.9
9	4	-9	15.3	12.6
9	6	-9	19.1	20.7
9	8	-9	19.8	-20.8
9	9	-9	5.7	4.8
9	10	-9	4.6	-1.8
9	11	-9	6.3	-7.3
9	12	-9	13.5	15.1
9	15	-9	6.0	5.1
9	1	-10	33.0	-31.3
9	3	-10	4.5	-3.1
9	5	-10	19.6	15.2
9	9	-10	16.5	-15.1
9	11	-10	7.3	9.5
9	12	-10	4.8	-5.1
9	13	-10	7.4	7.2
9	14	-10	5.0	7.0
9	2	-11	16.6	-16.1
9	4	-11	8.1	6.6
9	6	-11	12.0	12.8
9	7	-11	5.6	4.5
9	8	-11	9.6	-11.6
9	12	-11	10.2	11.6
9	0	-12	11.1	11.4
9	1	-12	15.2	-14.6
9	2	-12	6.5	-5.8
9	3	-12	7.9	-7.2
9	4	-12	5.5	-5.1
9	5	-12	11.1	11.4
9	8	-12	4.9	5.3
9	9	-12	9.3	-9.0
9	12	-12	5.1	-1.4
9	1	-13	6.6	6.2
9	2	-13	16.3	-16.5
9	3	-13	7.1	-5.7
9	4	-13	5.7	5.3
9	6	-13	7.8	9.0
9	8	-13	7.7	-6.4
9	0	-14	8.2	7.9
9	1	-14	8.1	-7.0
9	3	-14	5.4	-4.9
9	5	-14	8.2	9.5

TABLE 1.6

(A) ANALYSIS OF STRUCTURE FACTOR DATA AS A FUNCTION OF LAYER LINE INDEX. ALL FIGURES ARE ON ABSOLUTE SCALE.

h	Σ|Fo|	Σ|Fc|	ΣΔ	N	R	ΣΔ/N
0	3557	3565	380	138	0.1067	2.75
1	6202	6173	792	267	0.1278	2.97
2	6151	6150	670	282	0.1089	2.37
3	5620	5464	650	253	0.1156	2.57
4	4910	4807	553	250	0.1127	2.21
5	4163	4142	412	224	0.0991	1.84
6	3674	3609	393	226	0.1069	1.74
7	2978	2926	345	204	0.1157	1.69
8	2514	2463	316	196	0.1256	1.61
9	1793	1737	216	147	0.1205	1.47
ALL	41561	41035	4726	2187	0.1137	2.16

(B) ANALYSIS OF STRUCTURE FACTOR DATA BY MAGNITUDE OF Fo.

0 - 13	8964	8493	1331	1072	0.1484	1.24
13 - 25	10692	10241	1219	612	0.1141	1.99
25 - 50	12363	12236	1091	359	0.0883	3.04
50 - 100	8614	9046	959	136	0.1113	7.05
100 - 150	928	1025	126	8	0.1359	15.77

TABLE 1.7

ORTHOGONAL COORDINATES AND E.S.D.S.

ATOM	X'	Y	Z'
As(1)	2.185 ± 1	2.410 ± 1	-1.752 ± 1
O(1)	3.105 ± 7	1.282 ± 7	-2.515 ± 7
O(2)	5.823 ± 8	0.971 ± 8	-3.194 ± 9
C(1)	1.627 ± 10	1.799 ± 9	-0.017 ± 9
C(2)	2.591 ± 12	1.399 ± 13	0.831 ± 13
C(3)	2.236 ± 17	0.928 ± 15	2.105 ± 16
C(4)	0.889 ± 18	0.915 ± 14	2.456 ± 13
C(5)	-0.056 ± 18	1.301 ± 15	1.607 ± 16
C(6)	0.312 ± 13	1.763 ± 14	0.327 ± 13
C(7)	3.157 ± 8	4.036 ± 8	-1.475 ± 9
C(8)	4.099 ± 10	4.382 ± 10	-2.387 ± 10
C(9)	4.840 ± 12	5.566 ± 12	-2.176 ± 12
C(10)	4.576 ± 11	6.316 ± 12	-1.052 ± 12
C(11)	3.637 ± 13	5.995 ± 10	-0.169 ± 11
C(12)	2.893 ± 12	4.825 ± 11	-0.346 ± 10
C(13)	0.652 ± 9	2.792 ± 11	-2.780 ± 11
C(14)	0.361 ± 11	1.956 ± 12	-3.868 ± 11
C(15)	-0.774 ± 17	2.257 ± 16	-4.612 ± 14
C(16)	-1.639 ± 13	3.271 ± 17	-4.228 ± 17
C(17)	-1.312 ± 14	4.072 ± 16	-3.188 ± 18
C(18)	-0.218 ± 11	3.865 ± 12	-2.427 ± 13

TABLE 1.8

BOND LENGTHS AND E.S.D.S.

As(1) - C(1)	1.922± 9	C(8) - C(9)	1.412±15
As(1) - C(7)	1.914± 8	C(9) - C(10)	1.377±17
As(1) - C(13)	1.885±10	C(10) - C(11)	1.329±17
As(1) - O(1)	1.644± 7	C(11) - C(12)	1.398±16
C(1) - C(2)	1.345±16	C(12) - C(7)	1.402±13
C(2) - C(3)	1.404±20	C(13) - C(14)	1.403±16
C(3) - C(4)	1.393±24	C(14) - C(15)	1.390±20
C(4) - C(5)	1.328±23	C(15) - C(16)	1.387±22
C(5) - C(6)	1.410±21	C(16) - C(17)	1.353±24
C(6) - C(1)	1.360±17	C(17) - C(18)	1.348±19
C(7) - C(8)	1.356±13	C(18) - C(13)	1.425±16

TABLE 1.9

INTERBOND ANGLES AND E.S.D.S.

O(1)-As(1)-C(1)	111.3± 4	As(1) - C(7) -C(12)	119.9± 7
O(1)-As(1)-C(7)	111.5± 4	C(8) - C(7) -C(12)	121.9± 9
O(1)-As(1)-C(13)	109.9± 4	C(7) - C(8) -C(9)	118.5± 9
C(1)-As(1)-C(7)	106.7± 4	C(8) - C(9) -C(10)	118.6±10
C(7)-As(1)-C(13)	108.6± 4	C(9) - C(10)-C(11)	123.1±11
C(1)-As(1)-C(13)	108.7± 4	C(10)- C(11)-C(12)	119.6±10
As(1)- C(1)-C(2)	117.1± 8	C(7) - C(12)-C(11)	118.2±10
As(1)- C(1)-C(6)	121.2± 8	As(1) - C(13)-C(14)	118.1± 8
C(2)- C(1)-C(6)	121.7±10	As(1) - C(13)-C(18)	120.9± 8
C(1)- C(2)-C(3)	119.4±12	C(14)- C(13)-C(18)	120.9±10
C(2)- C(3)-C(4)	118.4±14	C(13)- C(14)-C(15)	117.1±11
C(3)- C(4)-C(5)	121.7±14	C(14)- C(15)-C(16)	121.3±13
C(4)- C(5)-C(6)	119.3±16	C(15)- C(16)-C(17)	119.6±14
C(1)- C(6)-C(5)	119.4±13	C(16)- C(17)-C(18)	122.6±14
As(1)- C(7)-C(8)	118.1± 7	C(13)- C(18)-C(17)	118.1±12

TABLE 1.10

INTRAMOLECULAR DISTANCES (IN ANGSTROMS).

As...C(2)	2.80	O(1)...C(6)	4.01
As...C(6)	2.87	O(1)...C(12)	4.16
As...C(8)	2.82	O(1)...C(18)	4.21
As...C(14)	2.88	C(1)...C(7)	3.08
As...C(12)	2.88	C(1)...C(13)	3.09
As...C(18)	2.89	C(7)...C(13)	3.09
O(1)...C(1)	2.94	C(6)...C(12)	4.06
O(1)...C(7)	2.95	C(6)...C(18)	3.50
O(1)...C(13)	2.89	C(12)...C(18)	3.87

HYDROGEN BOND DISTANCES

O(1)...O(2)	2.819 ± 10
O(1)...O(2)*	2.784 ± 10

O(2)* refers to the coordinates given in table 1.2 transformed through an inversion centre.

TABLE 1.11

INTERMOLECULAR DISTANCES LESS THAN 3.8 Å.

C(5).....C(17)	iii	3.33	C(9).....C(16)	i	3.67
C(4).....C(14)	v	3.44	C(12)....C(14)	iii	3.68
C(3).....C(9)	iii	3.49	C(9).....C(17)	iv	3.69
O(2).....C(15)	i	3.53	C(6).....C(17)	iii	3.69
C(11)....O(1)	iii	3.56	C(14)....O(2)	vi	3.70
C(5).....O(2)	ii	3.56	C(11)....O(2)	iii	3.71
C(5).....C(18)	iii	3.57	C(8).....C(16)	i	3.72
C(3).....C(10)	iii	3.62	C(3).....C(8)	iii	3.75
C(5).....C(6)	v	3.63	C(2).....C(9)	iii	3.77
C(6).....C(6)	v	3.64	C(4).....C(18)	iii	3.80

Roman numerals refer to the following transformations of the fractional coordinates given in table 1.2.

i	1 + x,	y,	1 + z.
ii	-1 + x,	y,	z.
iii	x,	1/2 - y,	1/2 + z.
iv	1 + x,	1/2 - y,	1/2 + z.
v	-x,	-y,	-z.
vi	1 - x,	-y,	-z.

The transformations are applied to the coordinates of the second atom in each case.

TABLE 1.12

MEAN MOLECULAR PLANES

(A) Atoms defining planes.

PLANE 1 C(1) TO C(6).

PLANE 2 C(7) TO C(12).

PLANE 3 C(13) TO C(18).

PLANE 4 O(1), As(1), C(1).

PLANE 5 O(1), As(1), C(7).

PLANE 6 O(1), As(1), C(13).

(B) Plane equations.

Plane No.	P	Q	R	S	RMS D
1	-0.0724	-0.9318	-0.3557	-1.7841	0.006
2	-0.6816	0.5182	-0.5167	0.7067	0.006
3	0.5179	0.6136	-0.5960	3.7057	0.018
4	-0.8239	-0.3974	-0.4041	-2.0506	-
5	-0.3180	0.3404	-0.8849	1.6753	-
6	0.4977	0.7265	-0.4739	3.6685	-

P,Q and R are direction cosines of the plane normal, S is the origin to plane distance and RMS D the root mean square deviation of the atoms defining the plane, from the plane (in Angstroms). The plane equation is then

$$PX' + QY + RZ' = S$$

where X', Y and Z' are the atomic coordinates in Angstroms referred to orthogonal axes defined by X' parallel to $\underline{a}^*$, Y parallel to $\underline{b}$, and Z' parallel to $\underline{c}$.

(C) Deviations from mean molecular planes (in Angstroms).

	Plane 1 i = 0	Plane 2 i = 6	Plane 3 i = 12
C(1 + i)	-0.005	-0.004	0.000
C(2 + i)	-0.001	0.003	-0.013
C(3 + i)	0.008	0.003	0.028
C(4 + i)	-0.009	-0.009	-0.029
C(5 + i)	0.003	0.008	0.015
C(6 + i)	0.004	-0.002	-0.001

(D) Dihedral angles.

Plane A	Plane B	$\angle$AB
1	4	55.0
2	5	31.8
3	6	9.6

1.3 Discussion.

The crystal structure analysis establishes that the compound produced by complete hydrolysis of triphenyl arsenic dihalides, which has been generally regarded as triphenyl arsenic dihydroxide [$Ph_3As(OH)_2$] is more correctly formulated as triphenylarsine oxide monohydrate [$Ph_3AsO.H_2O$].

The arsenic is tetrahedrally coordinated but the expected C_{3v} symmetry of the molecule is destroyed by the unequal rotations of the phenyl groups. The phenyl groups are planar within experimental error (table 1.12) and are inclined at 55°, 32° and 10° respectively to the planes defined by O(1),As,C(1 + i) where i = 0, 6 and 12 (table 1.12). Since the intermolecular distances (table 1.11) involving carbon atoms are in the range regarded as being normal for van der Waals contacts, the orientations of the phenyl groups may be ascribed to crystal packing forces.

The average C_{sp^2} - C_{sp^2} (aromatic) bond length of 1.38 ± 0.02 Å and the average bond angle at a trigonal carbon atom of 120.0 ± 1.2° are in accord with the standard values quoted by Sutton (1965). No significant deviations from the means are found in the individual values (tables 1.8 and 1.9).

The arsenic-oxygen bond length found in this analysis is 1.644 ± 0.007 Å which compares favourably with the values of 1.646 ± 0.006 Å found in haidingerite [$CaH.(AsO_4)H_2O$] (Calleri and Ferraris, 1967) and 1.649 ± 0.016 Å in the 1:1 adduct of N-methyl quinolone and arsenic acid (Currie and Speakman, 1968).

The bonding may be considered as arising from the formation of four σ bonds using the sp^3 hybrid orbitals of the arsenic atom. The arsenic-oxygen bond length is then shortened by overlap between the filled oxygen $p\pi$ orbitals and the vacant arsenic $d\pi$ orbitals. A similar explanation of the bonding in analogous phosphorus and sulphur compounds has been proposed by Cruickshank (1961).

The average arsenic-carbon bond length is 1.907 ± 0.009 Å which does not compare with the value of 1.966 ± 0.021 Å found in arsenobenzene (Hedberg, Hughes and Waser, 1961) and the mean value of 1.96 ± 0.01 Å reported by Sutton (1965). Using the criteria of Cruickshank and Robertson (1953), these bond distances are significantly different at the 0.1 % level. The arsenic-carbon distance, however, compares favourably with the values of 1.897 ± 0.009 Å in tetraphenylarsonium 3-fluoro-1,1,4,5,5-pentacyano-2-azapentadienide (Palenik, 1966), 1.90 ± 0.01 Å in $[(C_6H_5)_4As]^+[BrIBr]^-$ (McEwan, 1965) and 1.91 ± 0.01 Å in $[(C_6H_5)_4As]^+[RuCl_4(H_2O)_2]^-.H_2O$ (Hopkins, Zalkin, Templeton and Adamson, 1966). The shortening of the arsenic-carbon bond lengths in these ionic compounds is attributed to a formal charge effect arising from the positive charge on the arsenic atom. A similar shortening mechanism may be envisaged in triphenylarsine oxide monohydrate. The polar nature of the arsenic-oxygen bond will leave the arsenic with a partial positive charge which might lead to slight bond shortening. Evidence for the polar nature of the arsenic-oxygen bond is found in the relatively high dipole moments in benzene solution of both the anhydrous and monohydrated triphenylarsine oxides (5.51 D. and 5.81 D. respectively ; Jensen, 1943).

The molecules in the unit cell are held together by a hydrogen-bonding system around the crystallographic centres of symmetry. Each triphenylarsine oxide molecule forms a hydrogen bond with two water molecules, forming what may be regarded as centrosymmetric dimers (see figure 1.3). The O...H...O distances are 2.81 Å and 2.78 Å which agree with values found for O...H...O distances in various crystalline hydrates (e.g. Baur, 1965). The hydrogen bonding may therefore be regarded as arising from interaction between the positive ends of the water dipole and the negative end of the arsenic-oxygen dipole.

Chapter 2.

The Crystal and Molecular Structure
of
Triphenyl Arsenic Hydroxychloride.

2.1 Introduction.

The crystal structure analyses of triphenyl arsenic hydroxychloride and triphenyl arsenic hydroxybromide will be described in this chapter and in the succeeding chapter respectively. These compounds are produced by partial hydrolysis of triphenylarsine dichloride and dibromide respectively and have generally been regarded as having trigonal bipyramidal structures in which the arsenic atom is penta-coordinate. The solution of their crystal structures establishes conclusively that, in the solid state, the structures consist of tetrahedrally coordinated arsenic atoms which are hydrogen bonded through oxygen to the halogen.

Although the compounds do not form an isomorphous series the environments of the arsenic atoms are so similar that the crystal and molecular structures of the two compounds will be discussed together at the end of the succeeding chapter (i.e. Part III, Chapter 3, section 3.4).

2.2 Experimental.

Crystal Data.

Triphenyl arsenic hydroxychloride

$C_{18}H_{16}AsOCl$, F.W = 358.7.

Monoclinic, $a = 17.38 \pm 3$ Å, $b = 11.44 \pm 2$ Å, $c = 17.21 \pm 3$ Å,

$\beta = 100^{\circ}\ 28' \pm 30'$

$U = 3365$ Å^3, $F(000) = 1456$ electrons.

$Dm = 1.44$ gm.cm.$^{-3}$, $Z = 8$, $Dx = 1.416$ gm.cm.$^{-3}$,

Space Group $P2_1/c$ (C^5_{2h}, No. 14).

Linear absorption coefficient (Mo K_α) $= 23.3$ cm.$^{-1}$

The crystals used in this analysis consisted of thin colourless plates and were supplied by Dr. G.S. Harris. Unit cell dimensions were derived from oscillation and zero-layer Weissenberg photographs taken about the b-axis with Cu $K\alpha$ radiation ($\lambda = 1.5418$ Å) and from precession photographs of the 0kl reciprocal lattice net (Mo $K\alpha$ radiation, $\lambda = 0.7107$ Å).

Intensity data were collected using a Hilger and Watts Y-190 linear diffractometer (Appendix I). The crystal was rotated about its b-axis and the h0l-h10l reciprocal lattice nets explored using Mo $K\alpha$ radiation. A one minute oscillation cycle of 3° was employed for each reflection; compensation for background radiation was achieved by means of balanced strontium oxide / zirconium oxide filters (Ross, 1926). Each reflection was measured twice with each filter. The maximum reflecting angle allowed was $\theta = 30^{\circ}$.

Since the random background count was of the order of 15 counts per minute, all reflections with count rates of less than 20 counts/minute were excluded from the analysis. The remaining 2420 reflections (table 2.4), which represents 33 % of the copper sphere, were reduced to structure amplitudes by application of the appropriate Lorentz, polarisation and rotation factors (Tunell, 1939). No correction for absorption was applied.

The data were initially placed on approximately absolute scale by comparison with the calculated structure factors. The final scale was determined by the least-squares refinement.

2.3 Solution of the Structure.

Triphenyl arsenic hydroxychloride crystallises in the monoclinic system, space group $P2_1/c$, with eight molecules per unit cell, i.e. two molecules per asymmetric unit. In the preliminary examination of the vector set which may be derived for the heavy atoms present in the structure, only those vectors which would arise between the arsenic atoms were considered. This reduced the complexity of the problem by reducing the number of heavy atoms from four to two per asymmetric unit with a consequent reduction in the number of possible vectors.

Two types of arsenic-arsenic vectors may be recognised; those due to atoms in symmetry related positions and those due to non-symmetry related atoms arising from the presence of two arsenic atoms per asymmetric unit. The former vectors may be derived by linear combinations of the coordinates of the equivalent positions and will have the following coordinates:

$2x_i, 2y_i, 2z_i$ (single weight)

$0, 1/2 + 2y_i, 1/2$ (double weight)

$2x_i, 1/2, 1/2 + 2z_i$ (double weight)

where the subscript i refers to either As(1) or As(2).

The non-symmetry related vectors were derived by graphical construction of the Patterson diagram for two atoms per asymmetric unit in a monoclinic cell, space group $P2_1/c$. Reduction of the resultant vector set by the reduced symmetry of the Patterson space group indicated that four double weight vectors should be present in the asymmetric unit of the Patterson function. The coordinates of these vectors are:

$$(x_1 - x_2),\ (y_1 - y_2),\ (z_1 - z_2)$$

$$(x_1 + x_2),\ (y_1 + y_2),\ (z_1 + z_2)$$

$$(x_1 - x_2),\ (1/2 + y_1 + y_2),\ (1/2 + z_1 - z_2)$$

$$(x_1 + x_2),\ (1/2 + y_1 - y_2),\ (1/2 + z_1 + z_2)$$

The asymmetric unit of the three-dimensional Patterson function was calculated. An empirical sharpening function which increased the magnitudes of the F^2 coefficients as $\sin \Theta/\lambda$ increased was applied.

Examination of the Harker line at u = 0, w = 1/2 showed the presence of only one major peak at v = 0 instead of the expected two peaks of comparable height. The peak height corresponded to the estimated value for an eight-fold arsenic-arsenic vector and was interpreted in terms of both arsenic atoms in the asymmetric unit having a fractional y-coordinate of 0.25.

Substitution of the value $y_i = 0.25$ in the analytical expressions given above for both the symmetry and non-symmetry related vectors indicates that all peaks due to arsenic-arsenic vectors will be concentrated on the sections of the three-dimensional Patterson function at v = 1/2 and v = 0.

The Harker section at v = 1/2 (figure 2.1) contains a total of six major peaks, four of which correspond to double weight arsenic-arsenic vectors (peaks A, B, C and D) and two which correspond to four-fold arsenic-arsenic vectors (peaks E and F). The section at v = 0 contained three major non-origin peaks: the eight-fold peak on the Harker line and two four-fold arsenic-arsenic vectors. The increase in weights of the rotation peak and the vectors from non-symmetry related atoms is due to coalescence of two peaks across the mirror planes at v = 0, 1/2.

Derivation of the atomic coordinates for both arsenic atoms by application of the above analytical expressions led to the possibility of two solutions both of which satisfied the vector set, viz. peaks A and D as rotation peaks, peaks B and C as corresponding Harker peaks and vice versa.

In order to differentiate between these solutions an attempt was made to locate the vector set arising from the chlorine atoms present in the structure. However, no solution could be found. It was therefore decided to calculate electron-density distributions based on the two solutions for the coordinates of the arsenic atoms.

<u>Position 1</u> Peaks A and D as rotation peaks.

As(1)	0.0749	0.250	0.0935
As(2)	0.4108	0.250	0.0596

Examination of the structure factor expression for $P2_1/c$ shows that atoms having a y-coordinate of 0.25 have no contribution to those planes with $l \neq 2n$. For this reason all planes for which $l \neq 2n$ were removed from the analysis in the initial stages. Structure factors were then calculated over the reduced data and were used to phase an electron-density synthesis. The resultant map was somewhat complicated by a pseudo-mirror plane at $y = 1/4$ but one possible chlorine could be located. Inclusion of the suspected chlorine atom in the structure factor calculation over all the data reduced the R-factor from 0.68 to 0.52. The improved phasing of the second electron-density map allowed location of a second possible chlorine site and showed areas of incompletely resolved electron density which could be interpreted in terms of benzene rings attached to the arsenic atoms. A third cycle of Fourier refinement, however, yielded no improved resolution. Further attempts to improve the phasing using this solution of the Patterson function were therefore suspended in order to explore the possibilities of the second solution.

<u>Position 2</u> Peaks B and C as rotation peaks.

As(1)	0.0875	0.250	0.1900
As(2)	0.4243	0.250	0.1515

An electron-density synthesis was calculated based on the phases of the arsenic and the observed structure amplitudes which were modified by the method of Woolfson (1956). The distribution showed regions of positive electron density which could be interpreted in terms of phenyl groups bonded to the arsenic atoms. The poor phasing of the map, however, obscured the positions of the chlorine atoms. A second cycle of Fourier refinement was calculated based on the phases derived from the arsenic atoms and one phenyl group. Examination of the resultant map showed tentative positions for the chlorine atoms and poorly resolved phenyl groups. It was then realised that the x and z coordinates of the proposed chlorine atoms were very similar to those of the arsenic atoms. For this reason, the chlorine-chlorine vectors arising from atoms related by the two-fold screw axes would superpose on the arsenic-arsenic vector arising in the same manner, but the vectors between atoms related by the inversion centres would not superpose.

This rationalised the differences in peak heights between the double-weight vectors A and D, and B and C on the Harker section. Inclusion of the chlorine atoms in the structure factor calculation reduced the R-factor from 0.68 (As only) to 0.47. The resultant improvement in the phase angles permitted the location of eighteen carbon atoms in the subsequent electron-density synthesis. A further two cycles of structure factors and Fourier syntheses yielded coordinates for all non-hydrogen atoms in the structure (i.e. 2 arsenic, 2 chlorine, 2 oxygen and 36 carbon atoms), and reduced the R-factor to 0.226.

2.4 Least-squares Refinement.

The atomic parameters were refined by least-squares minimisation of the function $M = \sum w(|Fo| - |Fc|)^2$. In the initial stages of the refinement, positional and isotropic thermal parameters were refined for each atom. Although the same counting times had been used for each reflection during the data collection, inspection of the intensities of several low-order reflections selected as standards revealed the necessity for refining individual layer scale factors. Four cycles of calculation, in which each observation was given unit weight and the block-diagonal approximation to the matrix of the normal equations was used, reduced the R-factor to 0.126. Inspection of the layer scale factors at this stage showed that their inclusion was justified; there was an approximately 33 % difference between the scale factor for h0l and h10l which compared favourably with the trend in the standard reflections.

The unweighted differences between the observed and calculated structure factors were analysed as functions of $|Fo|$, $\sin\theta/\lambda$ and layer line index. From this analysis it was apparent that equal weights were appropriate to approximately 80 % of the data. Planes with large $|Fo|$ at low $\sin\theta/\lambda$ values, however, required some down weighting. Similar conclusions have been reached about data collected using linear diffractometers in a number of analyses carried out in this laboratory and elsewhere (e.g. Manojlović, 1968; Wheatley, 1964). The weighting scheme which was adopted and applied throughout the remainder of the refinement was of the form

$$\sqrt{w} = 1 \text{ if } |Fo| \leq 100, \text{ else } \sqrt{w} = 100/|Fo|.$$

The refinement was continued by replacing the isotropic thermal parameters with six anisotropic thermal parameters. Lingafelter and Donohue (1966) have shown that, if the intensity data have been collected up only one axis and the individual reciprocal lattice nets require separate scale factors, then the thermal parameter, U_{ii}, corresponding to the rotation axis is indeterminate. For this reason, the U_{22} thermal parameter was held constant at the isotropic value and the refinement continued.

The refinement converged in a further three cycles of calculation in which the atomic coordinates, the determined anisotropic thermal parameters and individual layer scale factors were adjusted using the block-diagonal approximation to the matrix of the normal equations. The complete refinement (including Fourier refinement) is summarised in table 2.1.

In the final cycle of refinement, the shifts in both positional and thermal parameters were not significant being less than one-quarter of the corresponding standard deviations.

The structure factors calculated in the final cycle of refinement were used to phase electron-density and difference syntheses. The electron-density distribution is shown by means of a superimposed contour diagram in figure 2.2 which also explains the numbering system adopted for the molecules. Diffuse regions of positive electron density were found in the difference map in the expected positions for some of the hydrogen atoms bonded to the aromatic rings but the hydrogen contribution to the scattering was ignored. The standard deviation of the electron density, calculated using the approximate formula of Cruickshank (1949), was 0.17 $e/\AA^3$ and in terms of this no significant features appeared in the difference map so the refinement was regarded as being complete.

The atomic scattering factors used in this analysis were: for carbon and oxygen, Hoerni and Ibers (1954); for arsenic, Freeman and Watson (1961); for chlorine, Dawson (1960).

The atomic parameters are given in tables 2.2 (fractional coordinates) and 2.3 (anisotropic temperature factors). The final observed and calculated structure amplitudes and an analysis of them as functions of Fo and layer line index are contained in tables 2.4 and 2.5 respectively. Atomic coordinates, in Ångstoms, referred to orthogonal axes defined by X' parallel to a*, Y parallel to b and Z' parallel to c, are presented in table 2.6. Bond lengths, interbond angles, intermolecular distances and mean molecular planes are contained in tables 2.7 to 2.10. Estimated standard deviations, where quoted, are in units of the last decimal place of the quantity to which they refer.

The general packing of the molecules in the unit cell is shown in figure 2.3 and the coordination of the arsenic atom is shown in figure 2.4.

TABLES AND DIAGRAMS

Figure 2.1

Harker section at v = 1/2. Contours are at arbitrary levels. Peaks A and D are arsenic-arsenic vectors arising from the 2_1-axes, B and C are rotation peaks and E and F are vectors between non-symmetry related arsenic atoms.

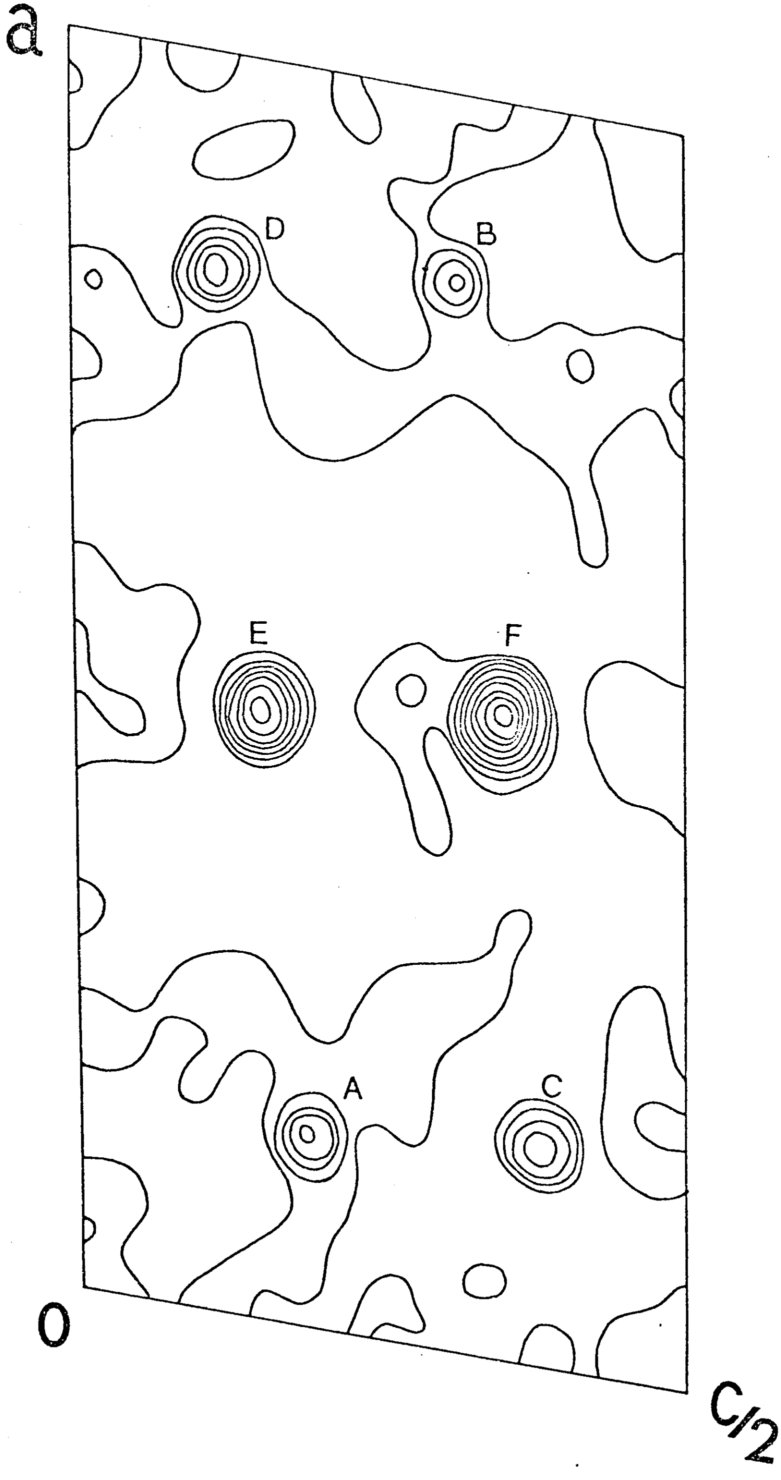
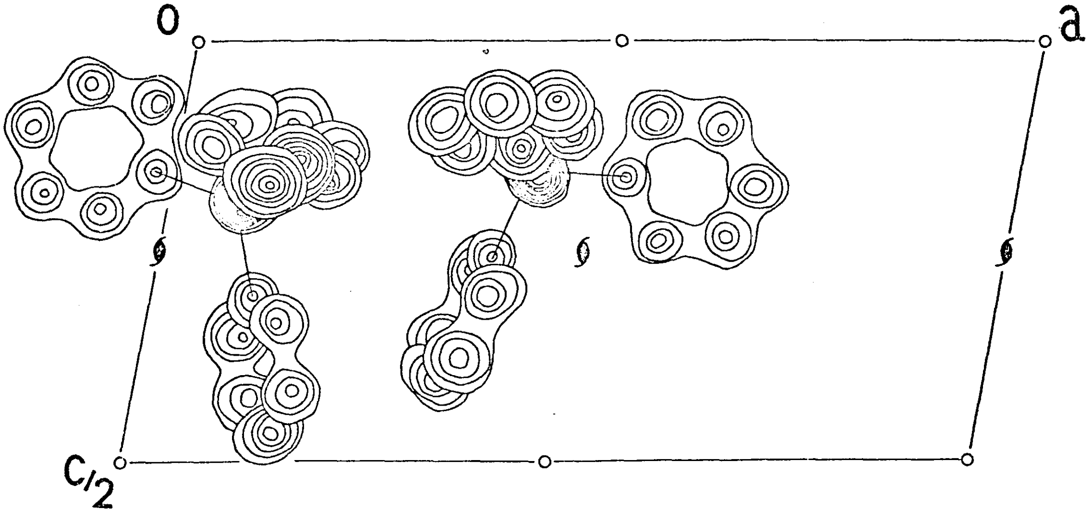

a
D
B
E
F
A
C
0
c/2

Figure 2.2

Superimposed contour diagram of the electron-density viewed down *b*. Contours are at intervals of 1 e/Å^3 except around the arsenic and chlorine atoms where they are at intervals of 5 e/Å^3 and 3 e/Å^3 respectively.


0
a
c/2

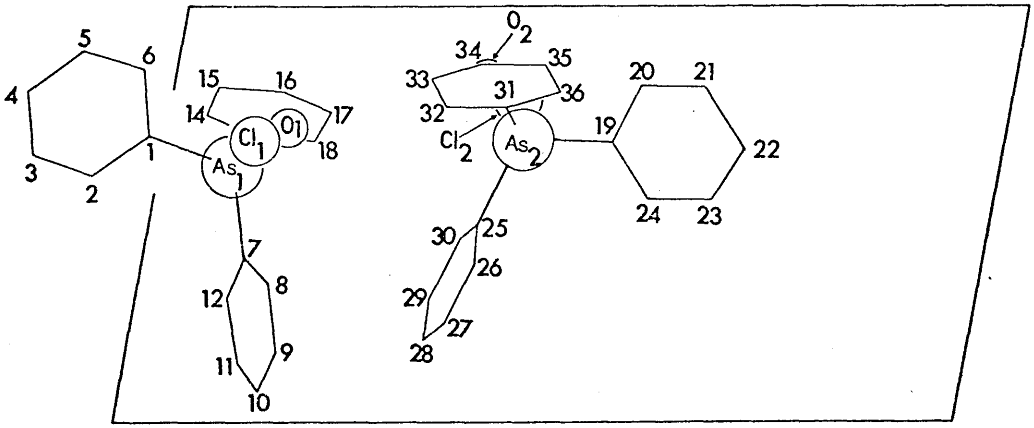

1
2
3
4
5
6
As1
Cl1
O1
7
8
9
10
11
12
14
15
16
17
18
O2
19
20
21
22
23
24
As2
Cl2
25
26
27
28
29
30
31
32
33
34
35
36

Figure 2.3

Molecular packing as viewed down *b*.

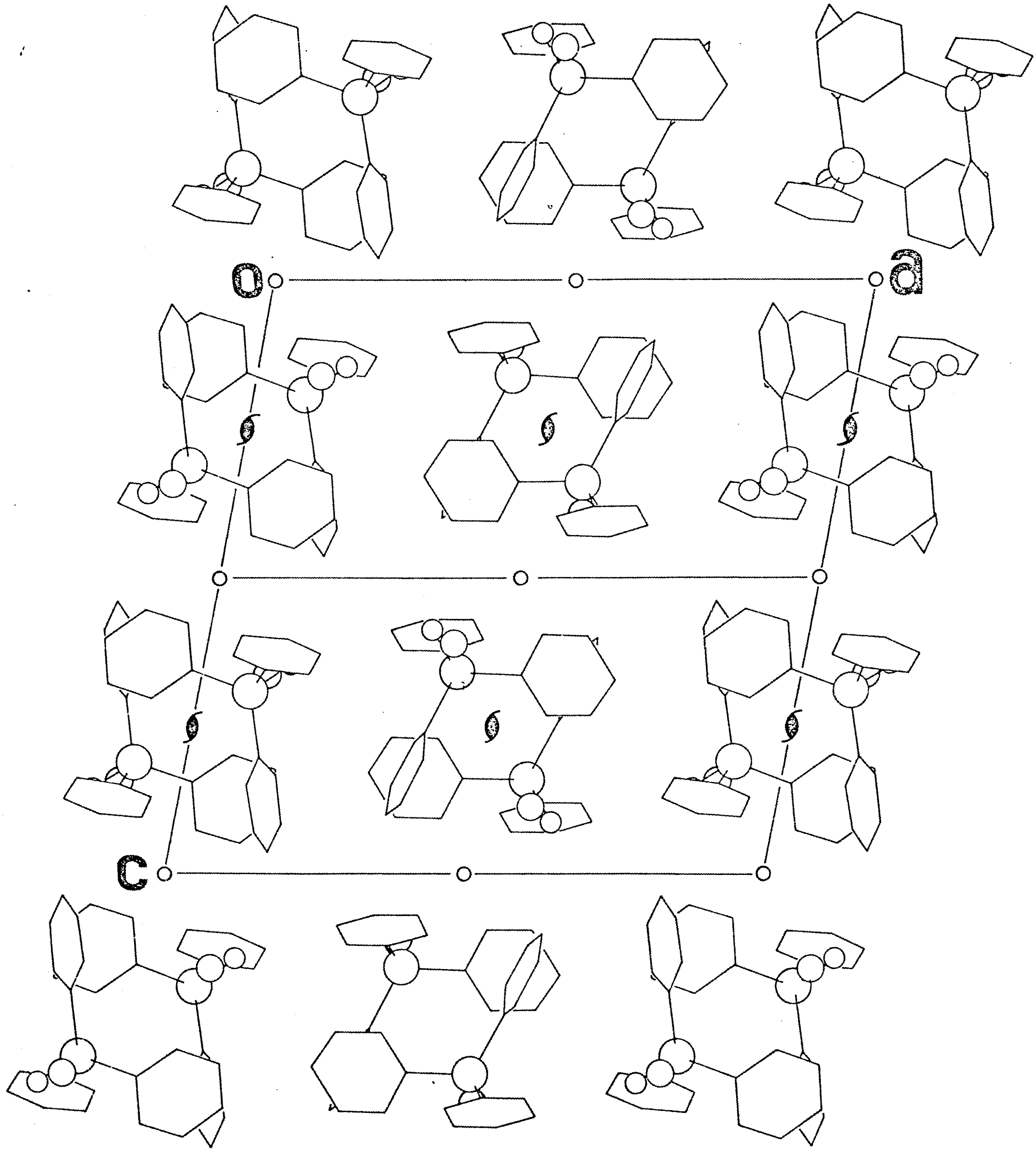

o
a
c

Figure 2.4

Molecular diagram.

(Hydrogen-bonding is indicated by means of a broken line.)

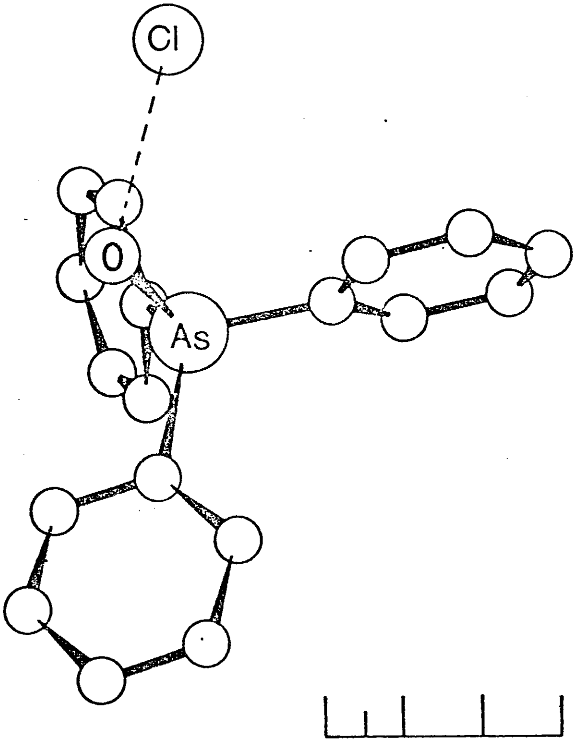

Cl
O
As
0 1 2 3 Å

TABLE 2.1

COURSE OF ANALYSIS

(A) Fourier Refinement

I Position 1

	Atoms included in calculation of the structure factors	R
1	2 As	0.68
2	2 As + Cl	0.52
3	2 As + 2 Cl	0.47

Lack of positive electron density which could be interpreted in terms of three phenyl groups bonded to each arsenic atom led to this solution of the Patterson function being abandoned.

II Position 2

1	2 As	0.68
2	2 As + 6 C	0.59
3	2 As + 2 Cl	0.49
4	2 As + 2 Cl + 18 C	0.398
5	2 As + 2 Cl + 2 O + 33 C	0.253
6	2 As + 2 Cl + 2 O + 36 C	0.226

(B) Least-squares refinement

Cycles	Final R	Final R'	*
1 - 4	0.127	-	(a)
5 - 7	0.103	0.015	(b)

*(a) Block-diagonal, isotropic, layer scales, unit weights.

(b) Block-diagonal, anisotropic (U_{22} held constant) layer scales, weighting scheme applied.

TABLE 2.2

FRACTIONAL COORDINATES AND E.S.D.S.

ATOM	X/a	Y/b	Z/c
As(1)	0.0868 ± 1	0.2609 ± 2	0.1896 ± 1
Cl(1)	0.1121 ± 5	0.5857 ± 7	0.1627 ± 4
O(1)	0.1461 ± 8	0.3445 ± 13	0.1436 ± 7
C(1)	-0.0161 ± 13	0.3094 ± 20	0.1538 ± 12
C(2)	-0.0725 ± 13	0.3045 ± 20	0.2049 ± 12
C(3)	-0.1504 ± 11	0.3386 ± 21	0.1771 ± 13
C(4)	-0.1732 ± 14	0.3709 ± 24	0.0976 ± 13
C(5)	-0.1147 ± 13	0.3727 ± 21	0.0508 ± 11
C(6)	-0.0397 ± 11	0.3471 ± 19	0.0744 ± 11
C(7)	0.1189 ± 11	0.2806 ± 20	0.3005 ± 10
C(8)	0.1519 ± 16	0.3832 ± 26	0.3305 ± 14
C(9)	0.1761 ± 15	0.3911 ± 24	0.4146 ± 14
C(10)	0.1651 ± 12	0.2953 ± 20	0.4607 ± 11
C(11)	0.1334 ± 13	0.1954 ± 21	0.4327 ± 12
C(12)	0.1075 ± 16	0.1813 ± 24	0.3463 ± 14
C(13)	0.1020 ± 11	0.1058 ± 19	0.1548 ± 11
C(14)	0.0370 ± 13	0.0381 ± 22	0.1256 ± 13
C(15)	0.0541 ± 16	-0.0747 ± 26	0.0967 ± 15
C(16)	0.1301 ± 18	-0.1126 ± 28	0.0981 ± 15
C(17)	0.1944 ± 15	-0.0386 ± 25	0.1295 ± 15
C(18)	0.1790 ± 12	0.0699 ± 21	0.1595 ± 12

As(2)	0.4251 ± 1	0.2302 ± 2	0.1561 ± 1
Cl(2)	0.4105 ± 5	-0.0896 ± 8	0.1120 ± 4
O(2)	0.3685 ± 8	0.1470 ± 13	0.0865 ± 7
C(19)	0.5314 ± 11	0.1962 ± 19	0.1597 ± 11
C(20)	0.5564 ± 13	0.1510 ± 22	0.0918 ± 11
C(21)	0.6345 ± 13	0.1224 ± 23	0.0967 ± 13
C(22)	0.6922 ± 12	0.1437 ± 22	0.1651 ± 13
C(23)	0.6630 ± 13	0.1874 ± 22	0.2318 ± 14
C(24)	0.5850 ± 14	0.2153 ± 22	0.2292 ± 11
C(25)	0.3920 ± 11	0.2002 ± 18	0.2564 ± 10
C(26)	0.3943 ± 14	0.3026 ± 21	0.3056 ± 12
C(27)	0.3682 ± 15	0.2743 ± 26	0.3809 ± 14
C(28)	0.3455 ± 14	0.1704 ± 24	0.3978 ± 15
C(29)	0.3465 ± 13	0.0762 ± 23	0.3505 ± 14
C(30)	0.3726 ± 15	0.0900 ± 23	0.2746 ± 13
C(31)	0.4023 ± 11	0.3870 ± 19	0.1228 ± 10
C(32)	0.3253 ± 11	0.4259 ± 20	0.1173 ± 11
C(33)	0.3048 ± 15	0.5337 ± 25	0.0898 ± 14
C(34)	0.3605 ± 14	0.6105 ± 24	0.0698 ± 14
C(35)	0.4377 ± 15	0.5692 ± 25	0.0734 ± 15
C(36)	0.4605 ± 12	0.4585 ± 21	0.1004 ± 12

TABLE 2.3

ANISOTROPIC TEMPERATURE FACTORS AND E.S.D.S.

ATOM	U11	U22	U33	2U23	2U31	2U12
As(1)	0.0519	0.0464	0.0416	-0.0014	0.0177	-0.0042
	10	0	9	22	16	24
Cl(1)	0.1224	0.0948	0.0837	-0.0044	0.0605	0.0062
	57	0	45	84	84	96
O(1)	0.0676	0.0572	0.0505	-0.0180	0.0257	-0.0146
	86	0	74	146	132	154
C(1)	0.0800	0.0635	0.0510	0.0072	0.0241	-0.0169
	146	0	118	225	214	251
C(2)	0.0769	0.0616	0.0620	-0.0199	0.0657	-0.0022
	142	0	129	225	226	239
C(3)	0.0493	0.0681	0.0816	-0.0543	0.0319	0.0051
	116	0	148	259	220	231
C(4)	0.0843	0.0777	0.0714	-0.0288	0.0213	0.0215
	161	0	146	278	252	295
C(5)	0.0789	0.0605	0.0457	0.0227	-0.0098	0.0292
	144	0	110	221	203	249
C(6)	0.0594	0.0563	0.0531	0.0077	0.0249	0.0563
	123	0	115	218	195	220
C(7)	0.0663	0.0587	0.0435	-0.0216	0.0136	0.0054
	121	0	101	208	179	232
C(8)	0.1149	0.0897	0.0698	-0.0315	0.0465	-0.0413
	205	0	152	302	295	341
C(9)	0.1045	0.0830	0.0727	-0.0317	0.0603	-0.0736
	190	0	154	289	284	310
C(10)	0.0748	0.0635	0.0444	0.0003	0.0062	-0.0081
	136	0	108	214	197	242

C(11)	0.0790	0.0716	0.0578	0.0166	0.0206	0.0296
	150	0	128	239	226	261
C(12)	0.1140	0.0815	0.0653	0.0255	0.0511	0.0083
	200	0	146	276	284	318
C(13)	0.0535	0.0498	0.0579	-0.0001	0.0259	-0.0132
	116	0	117	213	189	211
C(14)	0.0812	0.0693	0.0622	-0.0071	0.0187	-0.0396
	153	0	136	258	235	268
C(15)	0.1190	0.0894	0.0808	-0.0114	0.0633	0.0324
	214	0	173	311	319	351
C(16)	0.1417	0.0984	0.0758	0.0438	0.0638	0.0118
	249	0	168	328	337	392
C(17)	0.0951	0.0840	0.0856	-0.0144	0.0346	0.0034
	183	0	173	315	293	326
C(18)	0.0583	0.0620	0.0642	0.0182	-0.0012	-0.0078
	125	0	132	239	208	238
As(2)	0.0557	0.0478	0.0408	0.0045	0.0188	-0.0034
	11	0	9	22	16	23
Cl(2)	0.1199	0.0961	0.0841	-0.0049	0.0498	0.0002
	57	0	46	85	84	95
O(2)	0.0782	0.0591	0.0447	0.0228	0.0071	-0.0129
	94	0	73	146	135	163
C(19)	0.0618	0.0541	0.0422	0.0124	-0.0006	0.0091
	120	0	102	193	178	214
C(20)	0.0870	0.0672	0.0422	0.0102	0.0155	0.0278
	155	0	110	225	215	265
C(21)	0.0772	0.0740	0.0781	-0.0278	0.0658	0.0067
	153	0	150	274	253	273
C(22)	0.0655	0.0718	0.0683	0.0298	0.0188	0.0384
	136	0	137	259	223	254
C(23)	0.0603	0.0716	0.0832	0.0007	0.0103	0.0141
	133	0	155	268	233	248

C(24)	0.0966	0.0722	0.0469	0.0314	0.0156	0.0559
	163	0	115	234	219	279
C(25)	0.0670	0.0486	0.0382	0.0060	0.0359	-0.0033
	124	0	97	187	181	212
C(26)	0.0949	0.0696	0.0457	-0.0386	0.0446	0.0130
	163	0	113	221	221	260
C(27)	0.1007	0.0925	0.0709	0.0039	0.0336	0.0476
	178	0	146	310	260	336
C(28)	0.0778	0.0816	0.0965	0.0228	0.0559	-0.0074
	159	0	178	312	281	293
C(29)	0.0673	0.0752	0.0821	0.0482	-0.0139	-0.0150
	141	0	160	278	241	270
C(30)	0.0996	0.0749	0.0592	0.0118	0.0128	-0.0195
	178	0	135	264	253	302
C(31)	0.0701	0.0523	0.0417	0.0027	0.0563	0.0119
	128	0	100	204	190	225
C(32)	0.0579	0.0620	0.0571	0.0558	0.0211	-0.0061
	122	0	124	222	198	232
C(33)	0.1014	0.0834	0.0663	0.0286	0.0010	0.0188
	186	0	150	290	271	324
C(34)	0.0898	0.0783	0.0717	0.0058	0.0233	-0.0225
	168	0	150	286	261	300
C(35)	0.0861	0.0845	0.0875	-0.0030	0.0300	-0.0146
	168	0	174	306	283	311
C(36)	0.0679	0.0633	0.0621	0.0053	0.0337	-0.0151
	133	0	130	245	217	248

TABLE 2.4

FINAL OBSERVED AND CALCULATED STRUCTURE AMPLITUDES.

H	K	L	Fo	Fc
22	0	0	15.1	3.5
20	0	0	20.5	-12.2
18	0	0	38.9	-30.7
17	0	0	21.3	-13.0
16	0	0	19.6	-19.6
15	0	0	41.3	-37.9
14	0	0	25.1	23.9
13	0	0	20.2	-22.5
12	0	0	78.8	76.3
11	0	0	25.4	25.4
10	0	0	24.1	32.8
9	0	0	53.7	52.0
8	0	0	60.6	-65.7
7	0	0	56.9	61.7
6	0	0	117.9	-138.1
4	0	0	437.5	-476.5
17	0	-2	43.1	40.2
15	0	-2	40.5	42.0
14	0	-2	11.0	-11.2
12	0	-2	27.8	-29.3
11	0	-2	61.1	-61.1
9	0	-2	177.9	-134.5
8	0	-2	32.4	28.4
7	0	-2	35.9	-34.5
6	0	-2	88.3	73.8
5	0	-2	208.0	212.7
4	0	-2	59.9	57.6
3	0	-2	241.6	251.8
2	0	-2	105.6	-105.4
2	0	2	51.9	-50.1
3	0	2	234.7	-241.7
4	0	2	166.1	166.1
5	0	2	87.0	-80.1
6	0	2	178.6	164.7
7	0	2	106.0	110.9
8	0	2	18.5	15.1
9	0	2	119.7	118.0
10	0	2	92.7	-98.9
11	0	2	22.3	19.2
12	0	2	82.0	-86.6
14	0	2	9.8	-18.7
15	0	2	25.8	-23.6
16	0	2	24.6	25.8
18	0	2	41.0	37.2
19	0	2	21.3	8.7
20	0	4	11.6	-9.6
19	0	4	15.8	-16.4
16	0	4	11.0	-9.7
15	0	4	51.3	47.7
13	0	4	58.8	63.7
12	0	4	27.4	35.1
11	0	4	32.1	-26.5
10	0	4	9.8	10.1
9	0	4	135.6	-131.5
8	0	4	11.3	9.2
7	0	4	133.6	-132.5
6	0	4	33.3	-25.2
4	0	4	79.3	83.7
3	0	4	145.0	152.3
2	0	4	145.5	135.5
1	0	4	119.3	118.9
0	0	4	442.3	-420.7
1	0	-4	9.9	9.4
2	0	-4	292.1	-238.6
3	0	-4	99.3	-93.3
4	0	-4	126.4	125.0
5	0	-4	124.6	-126.3
6	0	-4	74.1	71.6
7	0	-4	26.3	-27.2
8	0	-4	92.4	95.1
9	0	-4	75.4	77.3
11	0	-4	65.0	67.9
12	0	-4	77.8	-83.7
13	0	-4	8.3	7.2
14	0	-4	44.4	-41.9
15	0	-4	17.7	-15.4
16	0	-4	8.5	10.2
17	0	-4	20.2	-22.6
18	0	-4	33.9	36.5
20	0	-4	31.4	26.0
21	0	-4	17.6	10.6
20	0	-6	22.6	-22.3
18	0	-6	23.0	-24.6
14	0	-6	86.0	81.2
12	0	-6	104.3	104.4
10	0	-6	20.9	-22.5
9	0	-6	71.0	66.2
8	0	-6	167.2	-165.8
7	0	-6	64.8	58.7
6	0	-6	99.6	-101.3
5	0	-6	85.3	-82.2
4	0	-6	84.2	83.6
3	0	-6	209.7	-197.4
2	0	-6	111.2	114.8
1	0	-6	15.0	14.8
0	0	6	77.4	90.4
1	0	6	216.4	200.1
2	0	6	14.9	12.5
3	0	6	30.1	-33.0
4	0	6	148.8	-146.9
5	0	6	84.0	-78.3
6	0	6	113.7	-108.0
7	0	6	61.3	56.5
8	0	6	55.6	54.8
9	0	6	55.5	54.0
10	0	6	105.9	105.3
12	0	6	40.7	42.7
13	0	6	15.0	-19.5
15	0	6	34.6	-34.6
16	0	6	21.7	-25.9
18	0	6	8.6	-17.5
19	0	6	18.5	16.7
19	0	8	19.2	-3.0
18	0	8	16.3	20.7
16	0	8	30.5	26.4
15	0	8	13.2	7.3
12	0	8	55.6	-49.6
10	0	8	79.5	-62.4
9	0	8	22.3	27.7
8	0	8	26.0	-24.5
7	0	8	63.5	61.1
6	0	8	55.4	62.4
5	0	8	78.4	80.7
4	0	8	135.2	123.1
3	0	8	34.4	-34.8
2	0	8	13.9	-11.2
1	0	8	131.1	-175.1
0	0	8	81.5	-72.6
1	0	-8	86.8	-85.3
2	0	-8	54.9	-54.5
3	0	-8	143.0	143.6
4	0	-8	47.0	-41.7
5	0	-8	150.9	143.7
6	0	-8	58.2	54.2
8	0	-8	35.6	36.3
9	0	-8	82.2	-75.8
10	0	-8	31.2	32.1
11	0	-8	107.0	-108.3
12	0	-8	23.0	23.3
13	0	-8	42.0	-40.6
14	0	-8	36.8	-33.2
15	0	-8	36.5	32.1
16	0	-8	24.3	-21.1
17	0	-8	39.9	41.5
19	0	-8	27.4	21.4
20	0	-8	13.2	-1.3
21	0	-10	23.5	-2.1
20	0	-10	5.1	13.6
19	0	-10	25.4	-25.1
17	0	-10	30.1	-29.7
16	0	-10	12.7	-6.8
14	0	-10	36.8	-35.6
13	0	-10	50.4	53.9
12	0	-10	38.2	-38.5
11	0	-10	69.6	71.8
10	0	-10	25.1	32.5
9	0	-10	18.4	7.6
8	0	-10	61.1	65.5
7	0	-10	50.7	-51.5
5	0	-10	72.3	-71.6
4	0	-10	42.6	-43.2
3	0	-10	32.5	-31.4
2	0	-10	11.1	3.2
0	0	10	14.8	-8.1
1	0	10	85.6	81.2
3	0	10	73.8	96.4
4	0	10	9.9	19.5
5	0	10	41.9	-43.9
6	0	10	18.8	-12.4
7	0	10	124.7	-120.4
8	0	10	35.5	-33.6
9	0	10	53.8	-54.9
10	0	10	4.2	-2.4
11	0	10	20.2	16.6
12	0	10	26.3	22.0
13	0	10	40.5	40.7
15	0	10	10.9	19.3
17	0	10	22.1	-5.4
17	0	12	19.6	8.3
15	0	12	13.6	-16.8
13	0	12	35.3	-29.1
12	0	12	12.5	8.7
11	0	12	22.4	-15.3
10	0	12	21.3	22.2
8	0	12	33.2	29.2
7	0	12	57.7	51.3
6	0	12	21.5	-15.4
5	0	12	15.1	5.8
4	0	12	91.6	-90.2
3	0	12	37.8	-40.2
2	0	12	29.9	-28.1
1	0	12	48.2	-50.4
0	0	12	66.8	67.9
1	0	-12	26.2	-25.3
2	0	-12	73.1	72.1
3	0	-12	19.8	17.0
4	0	-12	76.5	75.9
5	0	-12	33.1	32.6
7	0	-12	19.3	20.2
8	0	-12	72.5	-71.6
9	0	-12	3.9	5.8
10	0	-12	58.1	-55.2
11	0	-12	17.7	24.5
13	0	-12	7.4	10.6
14	0	-12	39.4	32.8
16	0	-12	42.4	41.5
17	0	-12	11.7	-6.0
19	0	-12	15.2	-3.3
19	0	-14	28.7	24.1
17	0	-14	30.3	21.4
15	0	-14	16.4	-15.9
13	0	-14	55.8	-56.3
11	0	-14	18.3	-25.3
10	0	-14	36.1	29.4
9	0	-14	12.4	16.3
8	0	-14	36.5	33.3
7	0	-14	22.4	13.4
6	0	-14	15.9	-14.7
5	0	-14	26.4	26.3
4	0	-14	35.7	-33.6
2	0	-14	26.5	-23.5
1	0	-14	13.0	-18.3
0	0	14	18.1	-14.5
2	0	14	34.9	31.5
4	0	14	61.8	57.6
7	0	14	23.4	26.6
8	0	14	26.4	-25.9
9	0	14	12.3	5.3
10	0	14	37.1	-41.7
12	0	14	16.6	-17.1
13	0	16	17.1	15.2
12	0	16	19.1	1.7
11	0	16	13.7	12.4
10	0	16	13.4	20.1
8	0	16	16.1	15.8
7	0	16	41.4	-37.7
5	0	16	25.1	-24.3
2	0	16	10.7	-5.6
1	0	16	42.0	41.9
0	0	16	25.4	-23.7
1	0	-16	64.7	63.7
5	0	-16	59.3	-54.2
7	0	-16	52.4	-54.0
9	0	-16	16.6	-15.5
10	0	-16	19.1	12.0
13	0	-16	36.2	32.4
15	0	-16	31.4	25.0
16	0	-16	24.3	-17.1
16	0	-18	21.9	21.6
14	0	-18	25.1	12.9
12	0	-18	18.5	-8.9
11	0	-18	16.9	-11.9
10	0	-18	40.5	-37.0
8	0	-18	23.0	-27.7
7	0	-18	30.6	31.7
6	0	-18	13.9	19.9
4	0	-18	42.4	40.1
2	0	-18	30.5	25.6
1	0	-18	32.7	-33.0
1	0	18	22.5	-25.3
2	0	18	33.4	-28.5
3	0	18	10.7	0.6
4	0	18	12.5	-19.3
5	0	18	18.4	22.0
6	0	18	21.9	16.7
8	0	20	21.3	-21.1
2	0	20	39.4	38.3
0	0	20	25.5	25.1
4	0	-20	45.9	-43.9
5	0	-20	5.0	-0.3
6	0	-20	16.5	-20.2
7	0	-20	14.1	12.5
8	0	-20	11.2	20.0
9	0	-20	13.3	3.1
9	0	-22	20.2	-19.5
7	0	-22	5.3	-17.0
4	0	-22	14.0	5.5
1	0	-22	11.9	20.5
18	1	0	20.5	23.2
17	1	0	22.5	-21.2
16	1	0	11.2	3.4
15	1	0	45.7	-45.5
14	1	0	28.1	-30.8
13	1	0	25.8	-24.3
12	1	0	36.4	-37.0
11	1	0	51.7	59.6
10	1	0	11.5	12.8
9	1	0	105.5	102.1
8	1	0	21.5	23.8
7	1	0	117.9	112.1
5	1	0	30.4	20.4
4	1	0	39.7	39.5
3	1	0	192.9	-208.4
10	1	3	14.3	-25.7
9	1	3	12.4	20.5
6	1	3	17.9	-21.5
5	1	3	68.5	-65.7
4	1	3	49.3	-47.6
3	1	3	24.3	-26.0
2	1	3	92.8	-83.1
1	1	3	37.3	34.1
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1	1	-3	47.2	48.9
2	1	-3	83.8	72.4
3	1	-3	52.4	50.0
4	1	-3	21.5	-19.7
5	1	-3	67.6	-57.1
6	1	-3	56.4	56.4
7	1	-3	41.2	-37.1
8	1	-3	61.4	56.5
9	1	-3	12.4	13.9
10	1	-3	14.9	15.0
11	1	-3	5.8	8.8
12	1	-3	10.6	7.3
14	1	-3	17.2	-18.2
15	1	-3	14.5	13.7
16	1	-3	11.9	-13.3
19	1	-3	17.6	-4.6
21	1	-4	27.2	-17.6
18	1	-4	31.4	23.2
17	1	-4	43.7	37.5
16	1	-4	11.9	5.4
15	1	-4	44.6	42.4
14	1	-4	64.0	-64.7
13	1	-4	17.4	-25.0
12	1	-4	75.3	-73.8
11	1	-4	83.9	-87.5
10	1	-4	17.9	22.5
9	1	-4	55.8	-53.5
8	1	-4	116.3	114.2
7	1	-4	19.2	-17.7
6	1	-4	90.4	93.5
5	1	-4	32.1	34.4
4	1	-4	26.0	20.2
3	1	-4	104.7	95.4
2	1	-4	156.0	-154.7
1	1	-4	22.4	17.9
0	1	4	229.2	219.7
1	1	4	88.8	78.5
2	1	4	9.2	6.3
3	1	4	27.0	20.7
4	1	4	79.6	-79.5
5	1	4	34.3	-36.8
6	1	4	114.8	-109.7
8	1	4	82.9	-83.8
9	1	4	5.7	-6.2
10	1	4	66.2	68.2
12	1	4	104.2	104.4
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16	1	4	25.5	-33.4
18	1	4	41.5	-36.7
19	1	4	14.0	3.8
14	1	5	13.9	-8.5
12	1	5	13.5	22.5
11	1	5	21.9	-24.5
9	1	5	8.3	-6.1
8	1	5	11.2	-12.4
7	1	5	40.6	49.1
6	1	5	67.2	69.2
5	1	5	11.1	-5.4
3	1	5	10.9	-9.5
2	1	5	85.1	-76.4
1	1	5	40.6	-30.5
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2	1	-5	9.7	9.3
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5	1	-5	76.0	-67.8
7	1	-5	22.5	-21.9
8	1	-5	26.4	-29.5
9	1	-5	44.3	44.1
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11	1	-5	16.4	21.3
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16	1	-5	14.6	4.9
21	1	-6	21.5	13.9
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17	1	-6	31.2	-35.3
16	1	-6	12.0	-2.3
15	1	-6	36.1	-38.6
13	1	-6	20.0	12.7
12	1	-6	15.4	-13.1
11	1	-6	122.1	122.6
10	1	-6	7.4	-6.5
9	1	-6	123.2	120.0
8	1	-6	24.5	-25.4
7	1	-6	70.4	-72.1
6	1	-6	32.3	-34.7
5	1	-6	192.1	-191.0
4	1	-6	24.2	22.4
3	1	-6	142.0	-138.1
2	1	-6	9.4	21.3
0	1	6	23.1	-22.6
1	1	6	151.5	-170.2
2	1	6	22.4	-20.2
3	1	6	138.6	-136.6
4	1	6	65.7	64.0
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6	1	6	82.7	79.0
7	1	6	121.2	117.5
9	1	6	30.2	32.7
10	1	6	56.3	-59.1
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12	1	6	59.7	-57.3
13	1	6	24.7	-31.4
15	1	6	31.7	-31.3
16	1	6	29.9	29.4
17	1	6	13.5	0.7
18	1	6	23.7	23.2
19	1	6	15.4	11.0
10	1	7	16.4	3.5
7	1	7	16.0	-15.6
4	1	7	25.1	23.0
3	1	7	31.9	32.0
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0	1	7	10.2	22.5
1	1	-7	37.3	-37.7
2	1	-7	13.9	-17.4
3	1	-7	49.6	-43.1
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5	1	-7	26.1	22.1
6	1	-7	32.4	-32.3
7	1	-7	32.9	33.1
8	1	-7	54.3	-50.3
16	1	-8	22.5	22.5
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12	1	-8	39.9	42.6
11	1	-8	50.9	-53.5
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9	1	-8	46.0	-49.0
8	1	-8	87.7	-86.8
7	1	-8	49.9	46.1
6	1	-8	62.4	-63.8
5	1	-8	94.8	93.5
4	1	-8	33.4	26.9
3	1	-8	51.4	52.3
2	1	-8	132.1	125.4
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3	1	8	79.3	77.3
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6	1	8	28.7	30.5
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13	1	8	47.8	45.6
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12	1	9	22.7	-17.3
11	1	9	13.1	-7.5
9	1	9	21.6	20.6
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5	1	9	17.0	-15.9
2	1	9	34.2	42.0
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4	1	-9	49.7	-50.0
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7	1	-9	8.0	13.2
8	1	-9	22.1	15.5
11	1	-9	14.4	-5.1
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9	1	1	29.6	-30.5
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6	1	-1	72.9	66.8
7	1	-1	15.4	-4.3
9	1	-1	40.6	-44.0
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12	1	-1	18.6	18.5
20	1	-2	29.7	-25.8
19	1	-2	11.6	2.2
18	1	-2	43.1	-42.2
15	1	-2	11.4	-11.6
14	1	-2	61.9	60.3
13	1	-2	25.5	21.4
12	1	-2	85.6	95.7
8	1	-2	130.8	-129.4
7	1	-2	50.0	-48.1
6	1	-2	106.9	-112.5
5	1	-2	66.6	68.6
4	1	-2	38.7	29.0
3	1	-2	159.0	154.6
3	1	2	153.3	160.8
5	1	2	42.9	53.0
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7	1	2	115.6	-114.4
8	1	2	63.6	68.9
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12	1	2	41.3	-39.3
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9	1	-10	10.8	-13.3
8	1	-10	113.6	124.0
7	1	-10	33.7	38.1
6	1	-10	37.1	39.9
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4	1	-10	103.7	-104.5
2	1	-10	112.8	-111.6
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2	1	10	35.4	-33.5
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4	1	10	117.7	-122.4
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17	1	-12	32.3	-37.5
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7	1	12	73.5	75.5
8	1	12	24.4	-23.6
9	1	12	20.6	24.8
10	1	12	30.6	-31.0
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2	1	-13	17.4	-15.7
16	1	-14	35.4	32.9
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7	1	-14	62.2	63.9
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7	2	0	22.0	-20.5
8	2	0	39.9	40.0
9	2	0	25.7	-30.1
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14	2	3	22.5	-18.3
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13	2	-7	13.5	-22.8
14	2	-7	15.1	-20.5

H	K	L	Fo	Fc
17	2	-8	46.0	-41.0
16	2	-8	14.4	-2.2
15	2	-8	27.1	-23.9
14	2	-8	25.1	25.6
13	2	-8	42.2	42.4
12	2	-8	25.6	26.3
11	2	-8	91.9	89.4
10	2	-8	20.1	-23.8
9	2	-8	57.4	56.6
8	2	-8	70.8	-76.5
7	2	-8	44.7	-45.1
6	2	-8	56.8	-52.6
5	2	-8	127.2	-125.3
4	2	-8	25.2	26.0
3	2	-8	103.6	-103.4
2	2	-8	170.7	161.7
1	2	-8	46.8	45.9
0	2	8	112.0	108.9
1	2	8	131.4	126.5
2	2	8	71.3	-68.7
3	2	8	64.0	61.5
4	2	8	97.4	-98.5
6	2	8	71.8	-71.9
7	2	8	44.8	-41.3
8	2	8	8.5	13.9
9	2	8	33.9	-32.9
10	2	8	75.3	77.0
12	2	8	40.7	35.6
10	2	9	23.9	10.0
9	2	9	14.1	-6.4
8	2	9	27.3	26.8
7	2	9	15.2	-13.6
6	2	9	11.7	17.9
5	2	9	13.4	6.7
4	2	9	25.5	-28.5
3	2	9	91.4	84.1
2	2	9	36.5	-37.7
1	2	9	20.9	27.6
0	2	9	31.5	-24.8
1	2	-9	107.7	107.1
3	2	-9	66.6	71.8
4	2	-9	40.8	-36.0
5	2	-9	35.4	-30.6
6	2	-9	30.9	-30.7
7	2	-9	42.2	-34.5
10	2	-9	47.3	47.2
12	2	-9	28.5	28.4
19	2	-10	24.7	15.6
17	2	-10	23.2	25.7
16	2	-10	10.5	18.1
15	2	-10	15.8	19.4
14	2	-10	29.5	26.3
12	2	-10	17.5	12.4
11	2	-10	84.0	-86.2
10	2	-10	32.3	-31.0
9	2	-10	48.9	-50.7
8	2	-10	46.8	-43.9
7	2	-10	88.5	86.9
6	2	-10	16.4	20.1
5	2	-10	118.6	123.1
4	2	-10	35.2	32.7
3	2	-10	53.3	55.9
2	2	-10	15.3	15.3
1	2	-10	69.4	-70.8
0	2	10	19.7	20.8
1	2	10	138.9	-140.8
2	2	10	25.1	23.9
3	2	10	66.6	-66.7
4	2	10	20.9	17.5
5	2	10	52.8	51.3
6	2	10	15.9	-7.5
7	2	10	101.3	98.2
8	2	10	16.9	-12.3
9	2	10	37.3	42.9
10	2	10	21.3	-16.6
13	2	10	34.9	-35.7
14	2	10	15.7	4.0
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6	2	11	39.9	-42.2
5	2	11	26.5	27.9
4	2	11	33.3	-32.7
2	2	11	19.4	22.6
1	2	11	55.0	-53.7
0	2	11	81.9	89.7
1	2	-11	15.8	12.6
2	2	-11	16.9	-10.6
4	2	-11	67.9	76.2
5	2	-11	42.2	40.8
6	2	-11	30.9	34.8
9	2	-11	31.5	-25.4
10	2	-11	29.2	-25.4
11	2	-11	14.3	4.1
16	2	-12	29.6	-27.0
14	2	-12	42.1	-43.6
12	2	-12	18.5	-14.1
10	2	-12	49.8	54.4
8	2	-12	83.4	86.0
7	2	-12	10.3	-13.7
6	2	-12	21.6	17.4
5	2	-12	47.4	-48.1
4	2	-12	84.6	-86.6
3	2	-12	17.5	-19.8
2	2	-12	113.5	-113.6
1	2	-12	46.5	49.2
0	2	12	33.6	-25.7
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2	2	12	57.6	54.2
4	2	12	52.4	59.2
5	2	12	35.8	-37.1
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7	2	12	42.0	-44.7
8	2	12	33.2	-26.4
10	2	12	27.5	-31.2
11	2.	12	24.4	24.0
12	2	12	11.0	-5.9
13	2	12	17.5	29.6
3	2	13	25.9	-30.7
0	2	13	18.0	-18.4
1	2	-13	31.5	-26.9
2	2	-13	19.9	12.7
3	2	-13	19.5	-22.3
5	2	-13	22.3	-23.1
7	2	-13	16.2	6.5
9	2	-13	29.9	33.8
17	2	-14	24.9	-25.2
13	2	-14	45.4	42.6
11	2	-14	45.0	48.2
10	2	-14	31.5	-32.9
8	2	-14	41.5	-49.7
7	2	-14	46.0	-47.6
5	2	-14	32.9	-26.8
4	2	-14	50.9	53.6
2	2	-14	50.6	55.1
1	2	-14	18.3	25.8
1	2	14	26.3	26.7
2	2	14	60.4	-63.0
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7	2	14	22.6	-20.6
8	2	14	35.7	37.6
10	2	14	38.7	36.3
4	2	15	26.4	15.9
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4	2	-15	42.2	-41.7
6	2	-15	30.9	-37.9
13	2	-16	37.5	-43.5
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10	2	-16	23.1	-16.6
8	2	-16	18.9	-18.3
7	2	-16	50.8	57.0
5	2	-16	51.4	57.8
4	2	-16	23.9	21.7
1	2	-16	63.0	-66.7
1	2	16	44.6	-43.8
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9	2	-17	27.6	-24.9
10	2	-18	45.2	45.3
8	2	-18	32.9	35.5
4	2	-18	53.7	-53.9
2	2	-18	37.2	-33.3
1	2	-18	27.7	32.6
0	2	18	23.8	19.6
1	2	18	20.2	22.2
2	2	18	40.7	39.6
2	3	0	40.7	44.1
3	3	0	264.2	279.9
4	3	0	20.2	-23.0
5	3	0	85.3	84.5
6	3	0	67.7	-66.7
7	3	0	128.2	-127.0
8	3	0	12.2	-10.2
9	3	0	117.3	-124.9
10	3	0	35.2	35.6
11	3	0	38.1	-36.1
12	3	0	41.6	41.6
14	3	0	24.9	20.4
15	3	0	33.1	40.0
17	3	0	29.0	26.5
18	3	0	26.0	-21.6
10	3	1	17.6	18.7
8	3	1	40.3	40.0
7	3	1	27.7	-30.1
6	3	1	18.3	13.9
4	3	1	12.9	-8.9
3	3	1	29.9	17.8
3	3	-1	29.8	-34.1
5	3	-1	44.7	-40.8
6	3	-1	12.8	18.7
7	3	-1	14.2	-2.6
9	3	-1	14.5	-7.3
10	3	-1	23.7	-24.1
18	3	-2	37.5	33.5
17	3	-2	14.6	-2.0
15	3	-2	16.8	-2.2
14	3	-2	59.6	-66.4
12	3	-2	83.6	-85.1
11	3	-2	11.8	4.1
10	3	-2	23.4	-29.3
9	3	-2	30.5	26.9
8	3	-2	114.3	114.3
7	3	-2	23.7	18.1
6	3	-2	193.9	197.6
5	3	-2	58.6	-59.2
4	3	-2	44.4	41.6
3	3	-2	39.8	-41.0
3	3	2	106.0	-105.2
4	3	2	108.2	-105.4
5	3	2	46.0	-46.9
6	3	2	113.9	-122.7
7	3	2	93.1	94.8
8	3	2	34.8	-29.6
9	3	2	99.6	105.3
10	3	2	27.2	36.1
11	3	2	20.1	14.2
12	3	2	49.6	51.2
13	3	2	39.1	-41.4
14	3	2	19.8	25.6
15	3	2	41.6	-37.6
10	3	3	22.5	24.1
9	3	3	21.9	24.8
8	3	3	23.1	-27.4
7	3	3	19.0	22.9
6	3	3	48.7	-51.6
4	3	3	32.9	36.0
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2	3	3	6.9	2.6
1	3	3	98.4	-90.0
1	3	-3	195.9	-175.2
2	3	-3	19.4	27.4
4	3	-3	38.6	-32.9
5	3	-3	23.9	21.0
7	3	-3	32.2	30.6
9	3	-3	29.0	23.1
10	3	-3	20.9	-19.9
11	3	-3	13.9	-17.8
12	3	-3	18.8	-18.8
13	3	-3	17.9	-26.1
16	3	-3	16.8	15.5
17	3	-3	21.7	11.1
17	3	-4	41.2	-43.2
15	3	-4	32.7	-37.1
14	3	-4	41.1	43.6
12	3	-4	33.0	41.4
11	3	-4	68.5	71.3
9	3	-4	94.2	96.6
8	3	-4	70.4	-70.5
7	3	-4	24.5	26.8
6	3	-4	108.5	-104.1
5	3	-4	115.0	-106.3
4	3	-4	23.0	-20.2
3	3	-4	138.4	-131.7
2	3	-4	190.1	171.3
1	3	-4	19.4	-12.5
0	3	4	189.6	-187.0
1	3	4	107.7	-101.3
2	3	4	13.2	-9.6
3	3	4	76.6	-73.7
4	3	4	148.1	146.0
6	3	4	109.2	110.7
7	3	4	52.8	54.3
10	3	4	58.1	-63.9
12	3	4	74.3	-68.1
16	3	4	31.3	34.7
18	3	4	28.2	29.4
12	3	5	20.2	-21.6
11	3	5	44.4	50.6
10	3	5	23.0	-20.4
9	3	5	25.9	26.6
6	3	5	16.3	-17.9
5	3	5	49.3	-43.4
2	3	5	55.3	51.4
1	3	5	23.1	13.0
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1	3	-5	41.1	-31.1
2	3	-5	23.9	-21.6
4	3	-5	41.7	-33.1
6	3	-5	28.5	-31.6
7	3	-5	22.2	-22.8
9	3	-5	21.8	-26.0
10	3	-5	31.8	29.9
12	3	-5	31.3	28.1
13	3	-5	26.9	32.3
15	3	-5	26.7	8.1
17	3	-6	41.0	43.7
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12	3	-6	13.4	11.4
11	3	-6	84.3	-87.8
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9	3	-6	85.5	-86.9
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7	3	-6	30.3	32.8
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5	3	-6	141.7	138.7
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1	3	6	101.8	99.5
2	3	6	16.5	16.0
3	3	6	136.6	132.6
4	3	6	29.4	-23.6
5	3	6	11.7	-12.0
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7	3	6	122.7	-124.5
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10	3	6	22.6	32.6
12	3	6	36.1	34.8
13	3	6	42.0	42.9
15	3	6	24.7	20.2
16	3	6	21.1	-17.8
11	3	7	17.4	-4.8
10	3	7	25.5	24.6
8	3	7	27.9	29.8
6	3	7	30.9	30.2
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3	3	-7	14.8	-10.7
4	3	-7	10.5	7.3
6	3	-7	34.9	35.6
7	3	-7	15.4	10.5
10	3	-7	30.5	-29.3
12	3	-7	22.4	-25.6
16	3	-8	30.9	-24.6
14	3	-8	52.2	-53.2
13	3	-8	13.0	8.5
12	3	-8	17.7	-24.5
10	3	-8	23.3	25.7
9	3	-8	13.0	-9.8
8	3	-8	80.5	76.3
6	3	-8	74.0	67.9
5	3	-8	47.0	-47.7
4	3	-8	69.4	-69.8
3	3	-8	75.9	-76.7
2	3	-8	118.4	-117.6
1	3	-8	18.3	15.1
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1	3	8	112.8	-117.7
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3	3	8	86.1	-87.6
4	3	8	76.3	-79.9
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8	3	8	37.0	36.5
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3	3	9	17.9	21.4
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0	3	9	13.9	-4.2
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7	3	-9	35.7	40.8
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6	3	-10	23.5	-17.0
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1	3	-10	35.5	40.5
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2	3	10	38.1	36.1
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6	3	10	49.9	49.2
8	3	10	22.9	-24.4
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7	3	-11	36.2	-37.0
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12	3	-11	39.6	38.4
17	3	-12	24.3	28.2
16	3	-12	20.6	11.0
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11	3	-12	43.5	-42.4
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6	4	0	141.1	-150.2
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3	4	0	8.9	-4.3
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7	4	-4	25.4	26.7
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1	4	4	100.4	103.5
2	4	4	50.4	-47.2
3	4	4	100.7	96.7
4	4	4	44.5	-38.9
5	4	4	18.7	17.9
6	4	4	23.6	31.1
7	4	4	58.0	-57.4
9	4	4	90.4	-89.2
11	4	4	36.7	-41.4
13	4	4	31.2	32.0
15	4	4	49.6	48.7
9	4	5	17.7	14.1
8	4	5	54.6	62.5
7	4	5	18.9	-14.7
6	4	5	19.2	25.2
5	4	5	36.6	-39.3
4	4	5	39.4	-43.7
3	4	5	20.3	-15.3
1	4	5	69.9	63.9
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1	4	-5	110.5	-102.7
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3	4	-5	46.3	-39.1
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6	4	-5	48.1	-48.5
7	4	-5	39.0	38.0
8	4	-5	22.3	-31.6
9	4	-5	27.7	25.5
13	4	-5	22.1	-30.1
17	4	-6	23.6	-13.3
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11	4	-6	32.9	35.4
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8	4	-6	91.4	-89.4
7	4	-6	64.6	-64.7
6	4	-6	97.4	-95.7
5	4	-6	49.1	48.0
4	4	-6	23.2	15.5
3	4	-6	82.0	74.3
2	4	-6	140.4	140.1
0	4	6	132.9	131.2
1	4	6	84.4	-72.3
2	4	6	13.8	5.7
3	4	6	41.5	-43.7
4	4	6	101.9	-104.4
5	4	6	41.9	41.3
6	4	6	72.3	-73.0
7	4	6	38.6	41.3
9	4	6	33.9	35.1
10	4	6	30.4	35.1
12	4	6	40.7	38.8
13	4	6	30.8	-35.8
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11	4	7	22.8	-33.7
10	4	7	25.1	16.6
7	4	7	18.3	18.2
5	4	7	21.7	21.1
4	4	7	18.8	24.2
3	4	7	22.4	17.7
2	4	7	79.6	-75.9
0	4	7	103.4	-99.6
1	4	-7	31.8	33.2
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12	4	-7	37.1	34.2
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16	4	-7	23.5	-11.9
13	4	-8	30.2	-35.0
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11	4	-8	60.2	-60.5
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6	4	-8	65.6	66.7
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6	4	8	59.1	62.4
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10	4	8	65.4	-67.7
13	4	1	19.7	-19.3
11	4	1	41.2	-43.0
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6	4	1	32.7	28.9
5	4	1	65.6	74.6
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11	4	-1	24.8	17.1
13	4	-1	20.2	21.9
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17	4	-2	33.1	29.4
15	4	-2	33.8	42.2
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7	4	-10	50.0	-51.9
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12	4	-12	23.6	15.9
11	4	-12	26.5	-23.0
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6	4	-12	36.4	-40.5
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2	4	-12	87.5	89.9
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8	4	14	37.3	-38.1
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14	4	-16	31.1	-12.3
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7	4	-16	54.1	-50.0
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7	5	0	16.4	-0.4
6	5	0	22.2	25.7
5	5	0	72.6	-77.3
4	5	0	14.6	-20.0
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4	5	1	21.2	23.1
3	5	1	13.3	-9.3
4	5	-1	24.5	-31.2
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6	5	-1	19.7	-15.7
7	5	-1	15.6	17.9
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14	5	-2	36.8	35.6
12	5	-2	64.0	70.6
9	5	-2	23.4	-21.8
8	5	-2	85.5	-87.8
7	5	-2	28.4	-23.1
6	5	-2	89.5	-95.0
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4	5	-2	32.4	33.5
3	5	-2	58.7	52.9
3	5	2	84.2	79.3
4	5	2	26.9	26.1
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6	5	2	76.6	80.9
7	5	2	26.3	-24.0
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11	5	2	38.4	-37.0
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13	5	2	27.9	23.3
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8	5	3	20.2	26.3
7	5	3	39.2	-37.3
5	5	3	16.9	-13.3
3	5	3	16.0	-14.2
1	5	3	75.2	75.3
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H	K	L	Fo	Fc
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4	6	2	57.8	-51.9
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7	6	2	50.4	-51.4
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12	6	2	42.9	41.4
13	6	2	22.7	21.6
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10	6	3	28.8	34.5
8	6	3	61.0	60.5
7	6	3	21.3	-28.5
6	6	3	24.1	23.3
5	6	3	35.8	39.6
4	6	3	27.7	-33.5
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2	6	3	83.5	-79.9
1	6	3	34.8	-38.3
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2	6	-3	67.2	-61.5
4	6	-3	52.5	-49.5
6	6	-3	18.2	-16.7
7	6	-3	16.6	20.2
8	6	-3	38.0	40.2
10	6	-3	64.7	62.4
11	6	-3	31.2	-23.4
12	6	-4	49.2	47.6
11	6	-4	33.2	-33.8
9	6	-4	52.5	-56.1
8	6	-4	68.4	-65.2
6	6	-4	84.8	-76.0
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3	6	-4	91.0	92.7
2	6	-4	88.8	79.1
1	6	-4	15.1	14.3
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1	6	4	95.5	-101.9
2	6	4	31.3	-33.0
3	6	4	65.9	-24.6
4	6	4	54.4	-53.2
7	6	4	70.5	66.7
9	6	4	70.1	73.1
13	6	4	37.0	-45.9
8	6	5	30.1	-33.1
6	6	5	20.5	-22.4
5	6	5	40.6	49.0
4	6	5	19.7	21.1
2	6	5	30.0	25.6
1	6	5	44.5	-49.3
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1	6	-5	53.0	53.6
2	6	-5	24.4	-21.9
3	6	-5	48.2	43.4
4	6	-5	35.3	33.0
7	6	-5	61.1	-57.6
10	6	-5	20.8	20.6
14	6	-6	43.1	-38.0
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9	6	-6	30.7	-34.2
8	6	-6	63.4	68.3
6	6	-6	73.0	67.5
5	6	-6	22.2	26.8
2	6	-6	109.5	-104.7
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2	6	6	32.3	30.8
4	6	6	101.9	93.6
6	6	6	60.0	60.2
9	6	6	37.9	-32.3
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12	6	6	28.7	-24.7
13	6	-7	27.3	-19.3
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11	6	-7	22.6	15.2
10	6	-7	47.5	-49.9
9	6	-7	36.1	37.0
8	6	-7	34.9	-29.1
7	6	-7	21.5	15.7
4	6	-7	44.3	42.3
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1	6	-7	74.6	-74.1
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7	6	8	37.4	-41.4
6	6	8	52.5	-48.8
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3	6	-8	56.5	-54.0
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9	6	-8	33.9	38.0
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11	6	-9	22.1	-16.6
9	6	-9	30.4	-29.1
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2	6	9	64.8	-71.2
6	6	9	41.4	42.5
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2	6	10	34.8	-31.2
1	6	10	78.3	-84.6
1	6	-10	23.1	-32.8
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3	6	-11	40.0	40.0
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4	6	-12	29.7	-34.4
8	6	-12	43.3	33.7
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9	6	-13	31.1	26.0
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4	7	2	33.3	-52.4
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9	7	2	40.4	42.9
10	7	2	48.7	42.4
11	7	2	25.1	30.8
11	7	3	30.5	-34.8
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3	7	-3	48.3	-52.0
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11	7	-3	21.0	-17.8
13	7	-3	23.6	-28.3
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6	7	4	73.6	73.8
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11	7	5	18.9	32.2
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7	7	5	35.2	-35.5
5	7	5	42.0	-35.6
4	7	5	43.8	44.5
2	7	5	57.8	50.3
1	7	5	23.5	18.0
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1	7	-5	70.2	74.5
2	7	-5	30.4	-23.2
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14	7	-6	25.1	16.8
11	7	-6	37.3	-37.1
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7	7	-6	20.2	-6.5
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1	7	-14	25.5	26.6
2	7	14	25.0	-23.0
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7	7	14	28.0	23.6
1	7	-15	29.3	-21.2
15	7	0	33.3	29.1
9	7	0	61.6	-72.0
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6	9	-2	67.2	-64.5
5	9	-2	15.9	9.2
4	9	-2	31.6	-30.0
3	9	-2	17.2	26.3
1	9	-3	49.0	50.9
4	9	-1	43.7	-54.1
3	9	11	26.5	29.8
1	9	-11	25.2	-20.0
6	9	-11	31.2	27.4
0	9	13	32.4	31.5
6	9	-13	34.4	-36.5
3	10	2	14.6	14.6
10	10	3	33.1	27.7
8	10	3	29.8	30.5
5	10	3	18.6	18.5
4	10	3	29.4	-33.7
2	10	3	50.6	-55.3
0	10	3	13.6	-14.8
2	10	-3	36.7	-42.8
4	10	-3	43.7	-45.5
8	10	-3	25.2	18.8
10	10	-3	27.7	32.9
5	10	-4	26.7	33.5
3	10	-4	35.0	42.7
1	10	4	16.8	-26.2
2	10	4	18.7	-10.4
3	10	4	30.3	-37.1
5	10	4	20.9	-13.8
7	10	4	22.5	15.9
5	10	5	27.4	23.6
4	10	5	25.6	22.3
2	10	5	28.2	34.9
1	10	5	17.0	-14.4
1	10	-5	47.1	45.8
4	10	-5	21.3	24.2
5	10	-5	21.6	-22.2
7	10	-5	36.9	-38.4
9	10	-5	32.6	-27.5
13	10	-5	33.0	26.6
13	10	-6	31.0	-13.5
9	10	-6	25.2	23.6
8	10	-6	25.6	23.9
7	10	-6	28.6	18.6
6	10	-6	25.4	18.4
3	10	-6	26.1	-25.4
3	10	6	27.5	27.4
4	10	6	23.1	30.3
5	10	7	41.2	-47.9
3	10	7	25.0	-31.1
1	10	-7	40.9	-43.1
3	10	-7	35.7	-35.4
4	10	-7	25.0	25.3
6	10	-7	24.9	14.9
7	10	-7	25.3	23.5
9	10	-7	24.2	21.7
10	10	-7	27.5	-21.7
8	10	-8	31.5	-19.7
6	10	-8	23.3	-26.2
2	10	-8	21.4	22.1
4	10	8	37.3	-28.6
4	10	9	24.5	-14.9
3	10	9	27.0	21.2
2	10	9	41.2	-34.6
4	10	-9	27.6	-36.9
6	10	-9	30.9	-33.4
10	10	-9	34.8	21.4
11	10	-10	29.5	-23.6
5	10	-10	29.0	28.8
1	10	0	9.4	-16.1
11	10	1	28.7	33.0
7	10	1	34.6	-40.6
5	10	1	35.1	-39.9
0	10	1	9.0	3.1
4	10	-1	26.1	30.8
5	10	-1	32.0	30.5
7	10	-1	20.8	34.6
10	10	-1	34.1	-26.1
6	10	11	35.4	-24.2
3	10	11	29.8	16.7
1	10	-11	35.7	23.4
8	10	-12	32.1	25.3
3	10	13	34.4	-24.6
1	10	-13	31.8	-19.1
5	10	-2	23.2	-23.2
4	10	-2	21.0	-9.1
3	10	-2	21.3	-25.1

TABLE 2.5

(A) ANALYSIS OF STRUCTURE FACTOR DATA AS A FUNCTION OF LAYER LINE INDEX. ALL FIGURES ARE ON ABSOLUTE SCALE.

k	$\Sigma\lvert F_o\rvert$	$\Sigma\lvert F_c\rvert$	$\Sigma\Delta$	N	R	$\Sigma\Delta/N$
0	14491	14139	1241	278	0.0856	4.46
1	14803	14484	1446	336	0.0977	4.30
2	15906	15622	1513	364	0.0951	4.16
3	12632	12291	1220	296	0.0966	4.12
4	12164	11965	1138	268	0.0936	4.25
5	9966	9776	1010	245	0.1014	4.12
6	7552	7535	646	161	0.0855	4.01
7	6089	5957	775	158	0.1273	4.91
8	4749	4637	663	132	0.1396	5.02
9	3412	3216	607	107	0.1779	5.67
10	2124	2025	436	75	0.2051	5.81
ALL	103889	101648	10696	2420	0.1030	4.42

(B) ANALYSIS OF STRUCTURE FACTOR DATA BY MAGNITUDE OF Fo.

0 - 25	13213	12379	3659	711	0.2770	5.15
25 - 50	37328	36289	4494	1078	0.1204	4.17
50 - 75	20624	20613	1140	340	0.0553	3.35
75 - 150	25796	25529	1095	257	0.0425	4.26
150 - 300	6050	5942	247	32	0.0408	7.72
300 - 600	880	897	61	2	0.0692	30.44

TABLE 2.6

ORTHOGONAL COORDINATES AND E.S.D.S.

ATOM	X′	Y	Z′
As(1)	1.484 ± 2	2.985 ± 2	2.989 ± 2
Cl(1)	1.916 ± 8	6.701 ± 8	2.447 ± 7
O(1)	2.496 ± 13	3.941 ± 15	2.011 ± 12
C(1)	-0.275 ± 22	3.539 ± 23	2.697 ± 20
C(2)	-1.239 ± 22	3.483 ± 23	3.756 ± 20
C(3)	-2.570 ± 19	3.874 ± 24	3.523 ± 22
C(4)	-2.960 ± 24	4.243 ± 27	2.226 ± 23
C(5)	-1.960 ± 22	4.264 ± 24	1.236 ± 20
C(6)	-0.679 ± 19	3.971 ± 22	1.406 ± 19
C(7)	2.032 ± 19	3.210 ± 23	4.796 ± 18
C(8)	2.596 ± 27	4.384 ± 30	5.208 ± 23
C(9)	3.010 ± 26	4.475 ± 28	6.579 ± 23
C(10)	2.821 ± 21	3.378 ± 23	7.407 ± 19
C(11)	2.280 ± 22	2.236 ± 24	7.026 ± 21
C(12)	1.837 ± 27	2.074 ± 28	5.620 ± 23
C(13)	1.743 ± 19	1.211 ± 22	2.342 ± 19
C(14)	0.632 ± 23	0.436 ± 25	2.044 ± 21
C(15)	0.924 ± 28	-0.855 ± 30	1.494 ± 25
C(16)	2.223 ± 31	-1.288 ± 32	1.277 ± 25
C(17)	3.322 ± 25	-0.442 ± 29	1.616 ± 25
C(18)	3.060 ± 20	0.800 ± 24	2.179 ± 21

As(2)	7.265 ± 2	2.633 ± 2	1.345 ± 2
Cl(2)	7.016 ± 8	-1.025 ± 9	0.630 ± 7
O(2)	6.297 ± 14	1.682 ± 15	0.325 ± 12
C(19)	9.082 ± 19	2.244 ± 21	1.070 ± 18
C(20)	9.510 ± 22	1.727 ± 25	-0.178 ± 19
C(21)	10.844 ± 22	1.400 ± 26	-0.339 ± 22
C(22)	11.831 ± 21	1.644 ± 25	0.654 ± 21
C(23)	11.331 ± 22	2.143 ± 25	1.895 ± 24
C(24)	9.998 ± 23	2.463 ± 25	2.097 ± 19
C(25)	6.700 ± 19	2.290 ± 20	3.174 ± 17
C(26)	6.739 ± 23	3.462 ± 24	4.014 ± 19
C(27)	6.293 ± 25	3.138 ± 30	5.393 ± 23
C(28)	5.904 ± 24	1.950 ± 28	5.755 ± 25
C(29)	5.922 ± 22	0.872 ± 27	4.937 ± 24
C(30)	6.367 ± 25	1.030 ± 27	3.550 ± 22
C(31)	6.876 ± 20	4.427 ± 22	0.843 ± 17
C(32)	5.559 ± 19	4.873 ± 23	0.992 ± 19
C(33)	5.210 ± 26	6.105 ± 28	0.582 ± 23
C(34)	6.160 ± 25	6.984 ± 27	0.063 ± 23
C(35)	7.481 ± 25	6.512 ± 29	-0.118 ± 25
C(36)	7.870 ± 21	5.245 ± 24	0.274 ± 20

TABLE 2.7

BOND LENGTHS AND E.S.D.S.

As(1) - O(1)	1.702 ± 14 A	As(2) - O(2)	1.697 ± 14 A
As(1) - C(1)	1.868 ± 22	As(2) - C(19)	1.879 ± 20
As(1) - C(7)	1.901 ± 18	As(2) - C(25)	1.946 ± 17
As(1) - C(13)	1.906 ± 21	As(2) - C(31)	1.902 ± 21
C(1) - C(2)	1.43 ± 3	C(19)- C(20)	1.42 ± 3
C(2) - C(3)	1.41 ± 3	C(20)- C(21)	1.38 ± 3
C(3) - C(4)	1.40 ± 3	C(21)- C(22)	1.42 ± 3
C(4) - C(5)	1.41 ± 3	C(22)- C(23)	1.43 ± 3
C(5) - C(6)	1.33 ± 3	C(23)- C(24)	1.39 ± 3
C(6) - C(1)	1.42 ± 3	C(24)- C(19)	1.39 ± 3
C(7) - C(8)	1.37 ± 4	C(25)- C(26)	1.44 ± 3
C(8) - C(9)	1.43 ± 3	C(26)- C(27)	1.48 ± 3
C(9) - C(10)	1.39 ± 3	C(27)- C(28)	1.30 ± 4
C(10)- C(11)	1.32 ± 3	C(28)- C(29)	1.35 ± 4
C(11)- C(12)	1.48 ± 3	C(29)- C(30)	1.47 ± 3
C(12)- C(7)	1.42 ± 3	C(30)- C(25)	1.36 ± 3
C(13)- C(14)	1.39 ± 3	C(31)- C(32)	1.40 ± 3
C(14)- C(15)	1.43 ± 4	C(32)- C(33)	1.35 ± 4
C(15)- C(16)	1.39 ± 4	C(33)- C(34)	1.39 ± 4
C(16)- C(17)	1.43 ± 4	C(34)- C(35)	1.41 ± 4
C(17)- C(18)	1.39 ± 4	C(35)- C(36)	1.38 ± 4
C(18)- C(13)	1.39 ± 3	C(36)- C(31)	1.41 ± 3

TABLE 2.8

INTERBOND ANGLES AND E.S.D.S.

O(1) -As(1) -C(1)	107.7 ± 8	O(2) -As(2) -C(19)	110.3 ± 8
O(1) -As(1) -C(7)	108.0 ± 8	O(2) -As(2) -C(25)	107.5 ± 7
O(1) -As(1) -C(13)	104.3 ± 8	O(2) -As(2) -C(31)	104.7 ± 8
C(1) -As(1) -C(13)	110.6 ± 9	C(19)-As(2) -C(31)	110.7 ± 9
C(1) -As(1) -C(7)	112.6 ± 9	C(19)-As(2) -C(25)	112.4 ± 8
C(7) -As(1) -C(13)	113.2 ± 9	C(25)-As(2) -C(31)	110.8 ± 8
As(1) - C(1) -C(2)	120.4 ±15	As(2) -C(19) -C(20)	119.8 ±15
As(1) - C(1) -C(6)	120.0 ±15	As(2) - C(19)-C(24)	119.7 ±15
C(2) - C(1) -C(6)	119.5 ±19	C(20)- C(19)-C(24)	120.5 ±19
C(1) - C(2) -C(3)	120.2 ±19	C(19)- C(20)-C(21)	118.7 ±19
C(2) - C(3) -C(4)	119.3 ±19	C(20)- C(21)-C(22)	123.2 ±20
C(3) - C(4) -C(5)	117.2 ±20	C(21)- C(22)-C(23)	115.1 ±19
C(4) - C(5) -C(6)	126.4 ±20	C(22)- C(23)-C(24)	122.9 ±20
C(5) - C(6) -C(1)	117.3 ±18	C(23)- C(24)-C(19)	119.3 ±19
As(1) - C(7) -C(8)	120.5 ±16	As(2) - C(25)-C(26)	113.3 ±14
As(1) - C(7) -C(12)	114.7 ±16	As(2) - C(25)-C(30)	119.7 ±15
C(8) - C(7) -C(12)	124.8 ±19	C(26)- C(25)-C(30)	126.9 ±18
C(7) - C(8) -C(9)	117.5 ±23	C(25)- C(26)-C(27)	110.8 ±20
C(8) - C(9) -C(10)	118.8 ±24	C(26)- C(27)-C(28)	123.2 ±24

C(9) - C(10) -C(11)	124.6 ±20	C(27)- C(28)-C(29)	123.7 ±24
C(10)- C(11) -C(12)	119.3 ±21	C(28)- C(29)-C(30)	119.3 ±24
C(11)- C(12) -C(13)	115.0 ±22	C(29)- C(30)-C(25)	115.9 ±21
As(1) - C(13) -C(14)	118.9 ±15	As(2) - C(31)-C(32)	117.7 ±15
As(1) - C(13) -C(18)	116.4 ±16	As(2) - C(31)-C(36)	120.7 ±15
C(14)- C(13) -C(18)	124.7 ±21	C(32)- C(31)-C(36)	121.5 ±20
C(13)- C(14) -C(15)	115.0 ±21	C(31)- C(32)-C(33)	120.3 ±20
C(14)- C(15) -C(16)	122.2 ±26	C(32)- C(33)-C(34)	120.9 ±23
C(15)- C(16) -C(17)	119.9 ±27	C(33)- C(34)-C(35)	118.3 ±25
C(16)- C(17) -C(18)	118.8 ±23	C(34)- C(35)-C(36)	122.2 ±23
C(17)- C(18) -C(13)	119.4 ±20	C(35)- C(36)-C(31)	116.7 ±20

TABLE 2.9

INTERMOLECULAR DISTANCES LESS THAN 3.80 Å.

C(23)....C(33)	vii	3.49 Å	C(2).....C(15)	viii	3.64 Å
C(22)....C(3)	i	3.51	C(27)....O(2)	iii	3.65
Cl(2)....C(24)	vii	3.52	Cl(2)....C(26)	vii	3.65
Cl(1)....C(2)	viii	3.53	Cl(1)....C(16)	ii	3.66
Cl(2)....C(21)	iv	3.55	Cl(1)....C(5)	v	3.71
C(29)....C(36)	vii	3.57	Cl(2)....C(20)	iv	3.72
C(34)...Cl(2)	ii	3.58	C(10)....C(18)	iii	3.72
C(10)....O(1)	iii	3.60	C(5).....C(11)	viii	3.72
C(30)....C(36)	vii	3.61	C(29)....C(36)	vii	3.74
C(23)....C(34)	vii	3.62	C(36)....C(35)	vi	3.76
C(24)....C(34)	vii	3.62	C(4).....C(33)	v	3.76
C(11)....O(1)	iii	3.63	C(4).....C(12)	viii	3.80

Roman numerals refer to the following transformations of the coordinates given in table 2.2.

i	1 + x,	y,	z.
ii	x,	1 + y,	z.
iii	x,	1/2 - y,	1/2 + z.
iv	1 - x,	- y,	- z.
v	- x,	1 - y,	- z.
vi	1 - x,	1 - y,	- z.
vii	1 - x,	1/2 + y,	3/2 - z.
viii	- x,	3/2 + y,	3/2 - z.

The transformations are applied to the coordinates of the second atom in each case.

TABLE 2.10

MEAN MOLECULAR PLANES

(A) Atoms defining planes.

Plane 1	C(1) TO C(6)	Plane 7	O(1),As(1),C(1).
Plane 2	C(7) TO C(12)	Plane 8	O(1),As(1),C(7).
Plane 3	C(13) TO C(18)	Plane 9	O(1),As(1),C(13).
Plane 4	C(19) TO C(24)	Plane 10	O(2),As(2),C(19).
Plane 5	C(25) TO C(30)	Plane 11	O(2),As(2),C(25).
Plane 6	C(31) TO C(36)	Plane 12	O(2),As(2),C(31).

(B) Plane equations.

Plane No.	P	Q	R	S	RMS D *
1	-0.2096	-0.9497	-0.2328	-3.9373	0.015
2	0.9052	-0.3434	-0.2503	-0.4601	0.002
3	-0.0145	0.3932	-0.9193	-1.7158	0.010
4	0.1599	0.9272	-0.3388	3.1709	0.016
5	-0.9428	0.2010	-0.2658	-6.7211	0.015
6	-0.2233	-0.3463	-0.9111	-3.8285	0.014
7	-0.0869	-0.6663	-0.7406	-4.3319	-
8	0.6326	-0.7685	-0.0960	-1.6423	-
9	-0.7490	0.1277	-0.6502	-2.6738	-
10	0.0453	0.7083	-0.7044	1.2470	-
11	-0.6641	0.7448	-0.0653	-2.9508	-
12	0.7383	-0.0286	-0.6739	4.3816	-

* These quantities are defined in table 1.12. The plane equation is

$$PX' + QY + RZ' = S$$

where X', Y and Z' are the atomic coordinates in Angstroms referred to orthogonal axes defined by X' parallel to $\underline{a}^*$, Y parallel to $\underline{b}$, and Z' parallel to $\underline{c}$.

(C) Deviations from mean molecular planes(in Angstroms).

	Plane 1	Plane 2	Plane 3
	i = 0	i = 6	i = 12
C(1 + i)	0.006	-0.004	0.014
C(2 + i)	0.015	0.001	-0.001
C(3 + i)	-0.023	0.002	-0.007
C(4 + i)	0.010	-0.001	0.003
C(5 + i)	0.011	-0.002	0.009
C(6 + i)	-0.019	0.004	-0.017
	Plane 4	Plane 5	Plane 6
C(19 + i)	0.000	0.021	-0.008
C(20 + i)	0.012	-0.004	-0.005
C(21 + i)	-0.024	-0.015	0.020
C(22 + i)	0.024	0.016	-0.023
C(23 + i)	-0.014	0.001	0.011
C(24 + i)	0.002	-0.019	0.005

(D) Dihedral angles (in degrees).

PLANE A	PLANE B	∠AB
1	7	34.6
2	8	30.6
3	9	48.8
4	10	25.5
5	11	37.5
6	12	62.7

Chapter 3.

The Crystal and Molecular Structure of Triphenyl Arsenic Hydroxybromide.

3.1 Crystal Data.

The crystals used in this analysis were provided by Dr. G.S. Harris. They consisted of colourless needles elongated along the b-axis. Unit cell dimensions were derived from oscillation and zero-layer Weissenberg photographs (Cu $K\alpha$ radiation, $\lambda = 1.5418$ Å), taken with the crystal rotating about its needle axis, and from precession photographs (Mo $K\alpha$ radiation, $\lambda = 0.7107$ Å) of the 0kl reciprocal lattice net. The space group was determined uniquely from the systematically absent spectra.

Triphenyl arsenic hydroxybromide, $C_{18}H_{16}AsOBr$.

F.W. = 403.1.

Monoclinic, a = 10.39 ± 2 Å, b = 11.86 ± 2 Å, c = 17.48 ± 3 Å,

$\beta = 127^{\circ}\,25' \pm 30'$.

U = 1711 Å^3, F(000) = 800 electrons,

Dm = 1.55 gm.cm.$^{-3}$, Z = 4, Dx = 1.564 gm.cm.$^{-3}$,

Space Group $P2_1/c$ (C^5_{2h}, No. 14).

Linear absorption coefficient = 62.2 cm.$^{-1}$ (Cu $K\alpha$ radiation).

3.2 Collection of the Intensity Data.

The intensity data were derived from zero-layer and equi-inclination Weissenberg photographs of the h0l-h9l reciprocal lattice nets (Cu $K\alpha$ radiation) by visual comparison with a calibrated step-wedge. The multiple film technique (Robertson, 1943) and interfilm scale factors of Rossman (1956) were used. The intensities were reduced to structure amplitudes by applying the Lorentz, polarisation and rotation factors (Tunell, 1939) appropriate to a small mosaic crystal. No allowance was made for absorption. A total of 2245 independent structure amplitudes (table 3.4) were obtained which represents some 63 % of the data available to Cu Kα radiation.

The data were placed on approximately absolute scale by comparison with the first set of calculated structure factors. The final scale was determined by the least-squares refinement.

3.3 Solution and Refinement of the Structure.

The solution and refinement of the structure of triphenyl arsenic hydroxybromide are summarised in table 3.1.

The coordinates of the two heavy atoms were derived from the three-dimensional Patterson function by application of the analytical expressions given in the preceding chapter. The Harker section is shown in figure 3.1. Peaks A and C are due to the vectors arising between one heavy atom set related by the two-fold screw axes and the inversion centres respectively; peak B is due to the vector arising between the other heavy atoms related by the two-fold screw axes. The coordinates derived gave an unambiguous solution to the Patterson function and were:

heavy atom 1	0.2845	0.2500	0.2145
heavy atom 2	0.2178	-0.0846	0.2298.

It was impossible to differentiate between the bromine and arsenic atoms at this stage and both atoms were assigned scattering curves for arsenic in the structure factor calculation. An electron-density synthesis calculated using the phase angles derived from the heavy atoms and the observed structure amplitudes clearly revealed the positions of all non-hydrogen atoms present in the structure. Inclusion of these atoms (18 carbon and 1 oxygen) in the the structure factor calculation reduced the residual R from 0.478 with the heavy atoms only to 0.213.

The structure was then refined in eight cycles of least-squares minimisation of the function $M = \sum w(|Fo| - |Fc|)^2$. D.R. Pollard's least-squares program (1968) was used in the initial stages of the refinement. The parameters refined were three positional and one isotropic thermal parameter for each atom and an individual scale factor for each reciprocal lattice net. Three consecutive cycles of refinement, in which the observations were given unit weights and the full-matrix of the normal equations was evaluated, reduced R, the agreement index, to 0.133.

On the assumption that the data were now on the same relative scale, the isotropic thermal parameters were replaced by six anisotropic temperature factors and the individual scale factors by one overall scale. The refinement was continued using the Glasgow SFLS program (Cruickshank and Smith, 1965). Convergence was achieved in a further five cycles in which the block-diagonal approximation to the matrix of the normal equations was used. The R-factor at the conclusion of the refinement was 0.098.

The weighting scheme which was applied in the last three cycles of refinement was of the form

$$w = 1 / [p_1 + |Fo| + p_2 |Fo|^2]$$

(Cruickshank, 1961). The final values of the parameters p_1 and p_2 were 4.0 and 0.0106 respectively.

The structure factors calculated in the final cycle of refinement were used to phase electron-density and difference syntheses. The electron-density synthesis is shown by means of a superimposed contour diagram in figure 3.2 which also explains the numbering system adopted for the molecule. The difference synthesis showed diffuse regions of positive density in positions which corresponded to several of the aromatic hydrogen atoms but, because of lack of resolution of the hydrogen positions, their contribution to the scattering was ignored. The standard deviation of the electron density (Cruickshank, 1949) was 0.20 $e/\overset{o}{A}{}^3$ and in terms of this no significant features appeared in the difference synthesis.

The atomic scattering factors used throughout this analysis were those of Hoerni and Ibers (1954) for carbon and oxygen; those of Freeman and Watson (1961) for arsenic and bromine.

The fractional coordinates and anisotropic thermal parameters are contained in tables 3.2 and 3.3 respectively. The observed and calculated structure factors, at the conclusion of the refinement, and an analysis of the data by magnitude of |Fo| and layer line index are presented in tables 3.4 and 3.5. Atomic coordinates, in Ångstroms, referred to orthogonal axes defined by X' parallel to a*, Y parallel to b and Z' parallel to c are given in table 3.6. Bond lengths, valence angles, intermolecular distances and mean molecular planes are presented in tables 3.7, 3.8, 3.9 and 3.10 respectively. Estimated standard deviations, where quoted, are in units of the last decimal place of the quantity to which they refer.

The general packing of the molecules in the unit cell, as projected on (100), is shown in figure 3.3.

TABLES AND DIAGRAMS

Figure 3.1

Harker section at $v = 1/2$. Peaks A and C are due to arsenic-arsenic vectors and peak B is due to a bromine -bromine vector. Contours are at arbitrary levels.

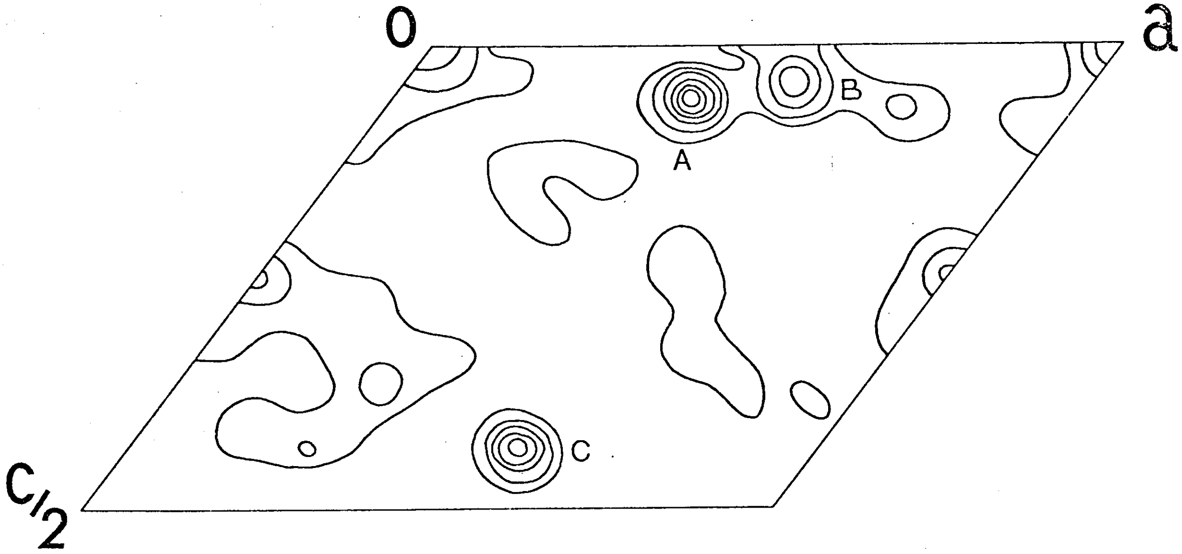

0
a
c/2
A
B
C

Figure 3.2

Superimposed contour diagram of the electron-density projected on (010). Contours are at intervals of 1 $e/\AA^3$ except around the arsenic and bromine atoms where they are at intervals of 5 $e/\AA^3$.

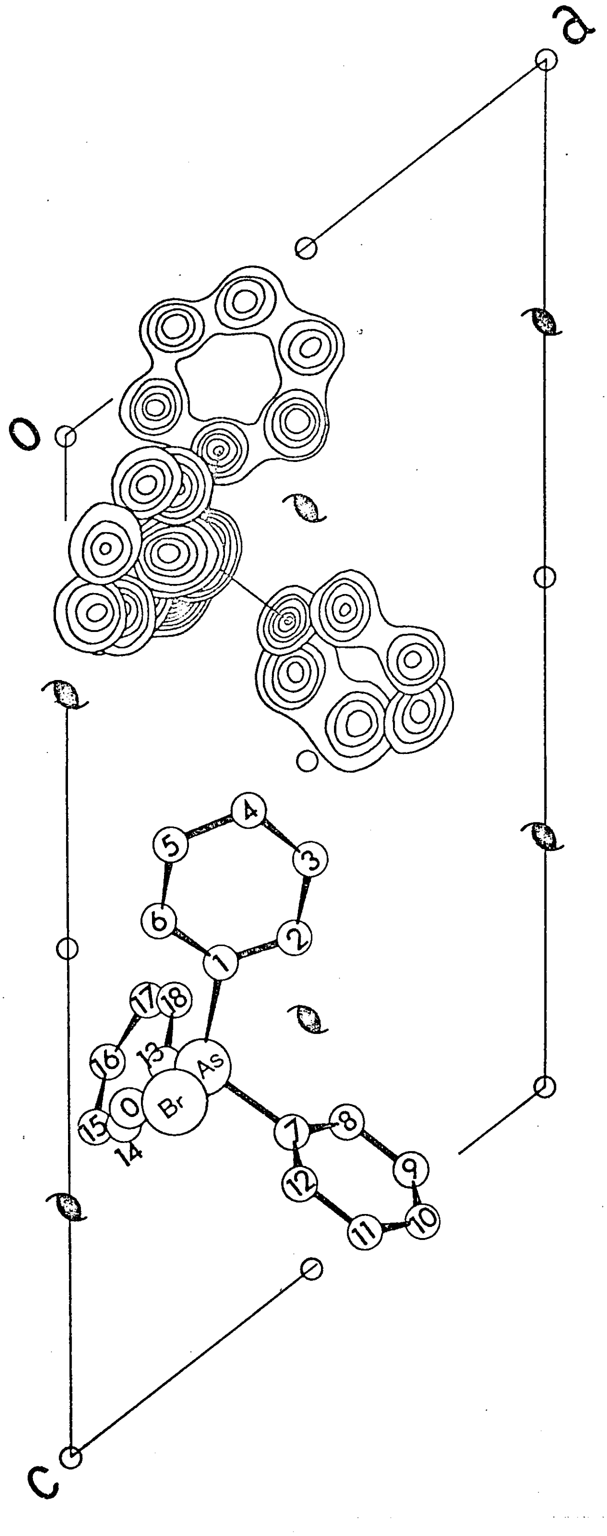

a
0
c
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
As
Br
O

Figure 3.3

The molecular packing as projected on (100). Hydrogen-bonding is shown by means of broken lines. (For clarity the 2_1-axes and c-glide plane symbols have been omitted.)

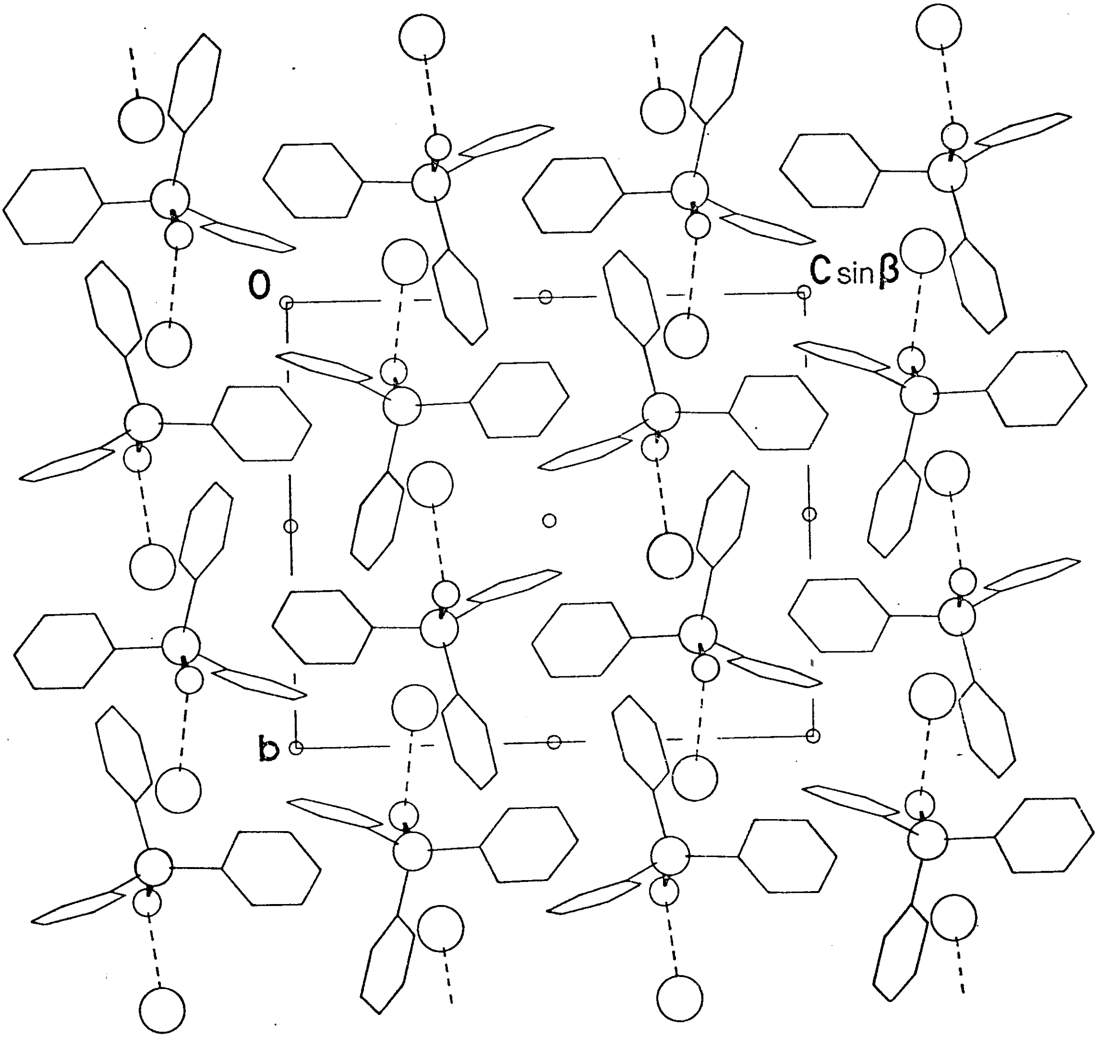
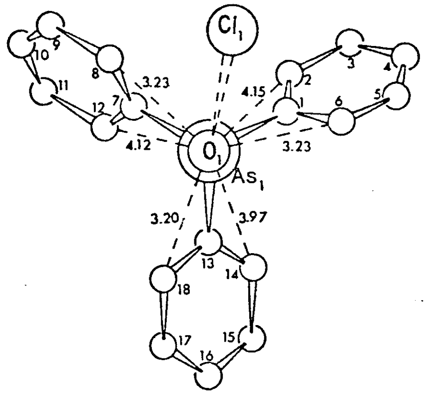

0
C sin β
b

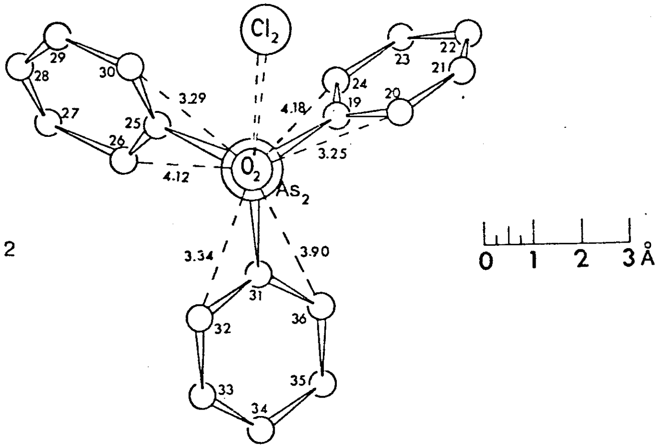
Figure 3.4

Projections of triphenylarsenic hydroxychloride and triphenylarsenic hydroxybromide down the oxygen-arsenic bond. Some intramolecular distances are shown.

CLAS
molecule 1



CLAS
molecule 2



BRASO

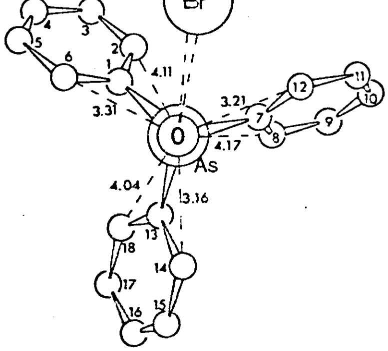


TABLE 3.1

COURSE OF ANALYSIS

(A) Fourier refinement.

	Atoms included in structure factor calculation.	R
1	2 Arsenic	0.478
2.	As + Br + O + 18 C	0.213

(B) Least-squares refinement.

Cycles	Final R	Final R′	*
1 - 3	0.133	-	(a)
4 - 5	0.110	0.022	(b)
6 - 8	0.098	0.017	(c)

*(a) Full-matrix, isotropic, layer scales, unit weights.

(b) Block-diagonal, anisotropic, one scale factor,unit weights.

(c) As above, weighting scheme applied.

TABLE 3.2

FRACTIONAL COORDINATES AND E.S.D.S.

ATOM	X/a	Y/b	Z/c
As(1)	0.2818 ± 1	0.2370 ± 1	0.2186 ± 1
Br(1)	0.2192 ± 2	-0.0870 ± 1	0.2297 ± 1
O(1)	0.1290 ± 8	0.1608 ± 6	0.2048 ± 5
C(1)	0.3201 ± 11	0.1839 ± 8	0.1320 ± 6
C(2)	0.4787 ± 12	0.1763 ± 11	0.1639 ± 8
C(3)	0.5042 ± 15	0.1393 ± 12	0.0977 ± 9
C(4)	0.3764 ± 17	0.1125 ± 12	0.0056 ± 8
C(5)	0.2229 ± 15	0.1201 ± 11	-0.0235 ± 7
C(6)	0.1903 ± 12	0.1541 ± 10	0.0401 ± 7
C(7)	0.4681 ± 12	0.2322 ± 10	0.3487 ± 6
C(8)	0.5833 ± 11	0.3144 ± 10	0.3827 ± 7
C(9)	0.7228 ± 13	0.3112 ± 11	0.4810 ± 8
C(10)	0.7387 ± 15	0.2249 ± 14	0.5395 ± 8
C(11)	0.6241 ± 17	0.1451 ± 13	0.5037 ± 9
C(12)	0.4844 ± 15	0.1443 ± 11	0.4072 ± 8
C(13)	0.2011 ± 13	0.3840 ± 9	0.1871 ± 7
C(14)	0.1149 ± 15	0.4258 ± 10	0.2173 ± 8
C(15)	0.0565 ± 17	0.5372 ± 13	0.1938 ± 10
C(16)	0.0847 ± 16	0.6033 ± 11	0.1452 ± 10
C(17)	0.1709 ± 17	0.5641 ± 11	0.1124 ± 10
C(18)	0.2281 ± 16	0.4551 ± 11	0.1337 ± 9

TABLE 3.3

ANISOTROPIC TEMPERATURE FACTORS AND E.S.D.S.

ATOM	U11	U22	U33	2U23	2U31	2U12
As(1)	0.0535	0.0597	0.0465	0.0091	0.0597	0.0065
	5	8	5	8	8	8
Br(1)	0.0919	0.0682	0.0784	-0.0002	0.0943	-0.0103
	8	10	7	11	13	12
O(1)	0.0628	0.0474	0.0658	0.0066	0.0796	-0.0070
	37	47	38	60	64	59
C(1)	0.0585	0.0547	0.0477	0.0081	0.0671	0.0030
	48	67	43	75	78	81
C(2)	0.0580	0.1006	0.0641	-0.0129	0.0795	0.0001
	53	97	59	113	99	109
C(3)	0.0777	0.1029	0.0833	0.0021	0.1149	0.0075
	71	108	76	135	131	132
C(4)	0.1008	0.0902	0.0621	0.0001	0.1095	0.0206
	87	97	61	116	129	139
C(5)	0.0823	0.0844	0.0495	-0.0039	0.0729	-0.0031
	70	89	51	101	104	121
C(6)	0.0634	0.0682	0.0487	-0.0035	0.0549	0.0106
	54	80	48	86	86	94
C(7)	0.0551	0.0856	0.0410	0.0148	0.0547	0.0251
	48	83	42	84	79	92
C(8)	0.0547	0.0713	0.0604	0.0007	0.0598	0.0001
	51	84	55	95	90	92
C(9)	0.0577	0.0854	0.0716	-0.0204	0.0648	0.0130
	56	96	65	116	103	108
C(10)	0.0672	0.1192	0.0619	-0.0213	0.0503	0.0183
	69	119	68	132	115	135

C(11)	0.0908	0.1058	0.0583	0.0239	0.0672	-0.0165
	84	117	64	125	120	151
C(12)	0.0846	0.0893	0.0512	0.0216	0.0705	-0.0091
	74	98	53	105	107	125
C(13)	0.0794	0.0550	0.0541	-0.0002	0.0933	0.0032
	62	67	48	85	95	97
C(14)	0.0782	0.0705	0.0703	-0.0207	0.0954	0.0031
	67	90	65	101	116	107
C(15)	0.0942	0.0806	0.0887	-0.0036	0.1117	0.0310
	86	99	86	136	150	141
C(16)	0.0813	0.0479	0.1011	0.0039	0.0899	0.0074
	76	81	91	125	140	112
C(17)	0.0924	0.0715	0.0880	0.0394	0.1050	-0.0017
	84	94	84	127	145	129
C(18)	0.0900	0.0636	0.0778	0.0223	0.1093	0.0021
	76	81	71	113	128	120

TABLE 3.4

FINAL OBSERVED AND CALCULATED STRUCTURE AMPLITUDES.

H	K	L	Fo	Fc
12	0	-6	12.8	8.5
12	0	-8	7.3	-4.9
12	0	-12	13.1	5.7
12	0	-14	15.8	-10.6
12	0	-16	20.3	13.7
11	0	-6	28.1	18.0
11	0	-8	23.8	-16.2
11	0	-10	21.9	16.5
11	0	-12	22.0	-18.5
11	0	-14	24.5	19.7
11	0	-16	20.3	-16.3
11	0	-18	12.8	10.1
10	0	0	15.6	9.1
10	0	-2	12.2	-8.1
10	0	-4	18.3	12.4
10	0	-6	29.6	-23.0
10	0	-5	13.6	9.0
10	0	-12	9.6	-6.6
10	0	-15	23.2	-14.5
10	0	-18	24.3	16.5
10	0	-20	23.0	-20.8
9	0	2	14.6	9.4
9	0	0	17.2	-11.4
9	0	-2	8.6	-6.1
9	0	-4	8.9	-8.1
9	0	-8	18.9	13.6
9	0	-10	46.7	-39.3
9	0	-12	24.8	18.9
9	0	-14	24.8	-17.4
9	0	-16	33.4	24.4
9	0	-18	21.6	-16.3
8	0	4	7.9	8.8
8	0	-2	13.7	11.0
8	0	-6	26.2	18.8
8	0	-8	32.1	-23.2
8	0	-10	42.2	32.7
8	0	-12	16.8	14.7
8	0	-14	12.5	9.3
8	0	-16	11.9	7.2
8	0	-18	20.1	-13.7
8	0	-20	35.3	23.5
7	0	2	15.9	-9.7
7	0	0	7.8	6.2
7	0	-2	26.6	-19.8
7	0	-4	38.3	28.9
7	0	-6	20.5	-13.3
7	0	-8	13.7	10.6
7	0	-10	14.5	10.8
7	0	-12	36.2	-28.6
7	0	-14	25.8	19.0
7	0	-16	40.4	-28.2
7	0	-18	29.0	22.6
7	0	-20	22.9	-14.5
6	0	8	17.9	-12.9
6	0	6	23.2	18.7
6	0	4	40.2	-34.0
6	0	2	31.2	24.6
6	0	0	36.5	-29.1
6	0	-2	32.2	26.0
6	0	-4	27.9	-21.8
6	0	-6	32.1	-26.4
6	0	-8	20.8	14.5
6	0	-10	30.8	-24.5
6	0	-12	28.7	25.7
6	0	-14	20.7	-16.7
6	0	-16	12.6	-8.6
6	0	-18	8.5	5.2
6	0	-20	20.8	-12.1
5	0	10	20.7	-14.2
5	0	8	38.1	23.7
5	0	6	23.9	-17.7
5	0	4	40.2	29.4
5	0	2	17.9	14.2
5	0	0	29.6	-24.9
5	0	-2	55.9	44.3
5	0	-4	40.0	-36.4
5	0	-6	52.5	44.9
5	0	-8	30.7	-24.1
5	0	-10	21.4	16.1
5	0	-12	18.7	-11.3
5	0	-14	23.8	-17.3
5	0	-16	32.1	24.1
5	0	-18	9.1	-5.4
5	0	-20	12.4	8.1
4	0	10	18.1	-9.9
4	0	8	15.3	11.5
4	0	6	42.9	-34.6
4	0	4	69.5	56.1
4	0	2	79.2	-70.1
4	0	0	108.7	113.7
4	0	-2	79.8	-74.9
4	0	-4	33.3	30.4
4	0	-6	53.2	-49.6
4	0	-8	6.0	5.0
4	0	-10	9.6	-6.6
4	0	-12	32.4	-24.8
4	0	-14	24.3	15.9
4	0	-16	21.8	-15.9
3	0	14	24.0	17.1
3	0	12	38.6	-27.2
3	0	10	48.2	38.2
3	0	8	50.5	-44.6
3	0	6	73.6	60.8
3	0	4	37.4	-33.2
3	0	2	54.2	44.7
3	0	0	71.2	-66.5
3	0	-4	113.0	120.4
3	0	-6	57.1	-52.9
3	0	-8	135.4	115.4
3	0	-10	78.5	-65.2
3	0	-12	67.5	49.8
3	0	-14	7.3	-6.9
3	0	-16	16.7	12.5
3	0	-18	10.1	8.2
2	0	14	15.4	-12.2
2	0	12	27.0	24.9
2	0	8	25.1	-20.3
2	0	6	35.7	32.7
2	0	4	133.5	-128.1
2	0	2	77.4	83.7
2	0	-2	170.8	191.9
2	0	-6	112.9	110.2
2	0	-8	68.0	-56.6
2	0	-10	9.4	-5.7
2	0	-12	68.5	53.0
2	0	-14	41.8	-30.7
2	0	-16	22.8	19.1
2	0	-18	20.2	-15.8
1	0	16	11.4	7.5
1	0	14	22.9	-16.6
1	0	12	40.3	31.2
1	0	10	84.5	-81.8
1	0	8	56.4	87.2
1	0	6	131.8	-116.7
1	0	4	180.6	184.6

H	K	L	Fo	Fc
1	0	-4	138.0	-140.5
1	0	-6	129.2	128.8
1	0	-8	174.2	-167.6
1	0	-10	109.9	104.4
1	0	-12	87.4	-71.9
1	0	-14	21.4	13.6
0	0	16	32.9	-24.2
0	0	14	31.1	28.4
0	0	12	45.3	-35.0
0	0	10	33.5	24.2
0	0	8	62.3	51.4
0	0	6	42.2	-35.3
0	0	4	138.3	147.7
12	1	-5	1.6	-2.0
12	1	-8	13.5	11.0
12	1	-10	13.5	-12.7
12	1	-11	5.9	5.7
12	1	-12	13.6	11.9
12	1	-14	7.9	-6.9
12	1	-16	7.8	6.9
12	1	-18	6.3	-5.4
11	1	-2	7.8	7.8
11	1	-4	11.7	-8.7
11	1	-6	15.7	14.6
11	1	-8	15.9	-13.4
11	1	-10	11.4	12.3
11	1	-15	9.1	7.4
11	1	-16	6.5	5.2
11	1	-17	5.6	-5.4
11	1	-18	11.3	-9.6
11	1	-20	10.8	9.1
10	1	0	15.5	11.8
10	1	-1	5.1	4.8
10	1	-2	16.5	-13.7
10	1	-4	6.3	5.0
10	1	-7	12.7	-10.4
10	1	-8	6.8	-6.3
10	1	-9	6.8	5.1
10	1	-10	10.8	8.1
10	1	-11	5.9	-5.6
10	1	-12	13.7	-12.7
10	1	-14	22.3	20.7
10	1	-15	8.1	-7.3
10	1	-16	24.9	-22.2
10	1	-18	18.7	16.6
10	1	-20	5.4	-5.1
9	1	2	7.4	5.8
9	1	-2	7.5	-7.2
9	1	-4	30.8	27.6
9	1	-6	36.1	-36.6
9	1	-7	6.0	-4.0
9	1	-8	34.4	32.7
9	1	-9	6.0	5.3
9	1	-10	30.7	-32.1
9	1	-11	11.9	-11.2
9	1	-12	21.6	20.1
9	1	-13	10.9	10.5
9	1	-14	23.7	-22.7
9	1	-15	8.4	-8.5
9	1	-16	8.9	7.9
9	1	-17	9.8	8.9
9	1	-18	7.6	7.3
9	1	-19	8.7	-7.4
9	1	-20	19.7	-15.3
8	1	4	14.9	-13.5
8	1	3	17.7	-7.1
8	1	2	26.4	23.6
8	1	1	8.1	6.3
8	1	0	24.9	-19.9
8	1	-1	10.4	-8.2
8	1	-2	45.9	44.0
8	1	-3	9.7	9.0
8	1	-4	47.5	-42.6
8	1	-5	10.5	-8.6
8	1	-6	41.2	35.3
8	1	-7	12.6	12.9
8	1	-8	9.8	-7.5
8	1	-10	19.1	-14.6
8	1	-11	23.7	22.3
8	1	-12	31.9	29.9
8	1	-13	6.8	-6.5
8	1	-14	35.9	-33.9
8	1	-16	32.6	28.7
8	1	-17	8.9	8.4
8	1	-18	28.9	-28.0
8	1	-19	3.0	-2.8
8	1	-20	23.7	19.0
8	1	-21	8.4	4.0
7	1	6	24.2	-21.1
7	1	4	26.9	24.2
7	1	2	20.1	-17.3
7	1	0	10.8	-9.8
7	1	-2	7.3	5.9
7	1	-4	35.1	-33.0
7	1	-5	18.9	-16.8
7	1	-6	52.9	56.0
7	1	-7	21.6	23.7
7	1	-8	70.1	-77.5
7	1	-9	16.5	-14.1
7	1	-10	76.2	82.0
7	1	-11	17.8	15.9
7	1	-12	53.1	-59.2
7	1	-13	29.6	-28.4
7	1	-14	35.4	32.2
7	1	-15	28.3	27.5
7	1	-16	6.9	-4.0
7	1	-17	13.6	-12.1
7	1	-20	7.2	7.5
6	1	9	6.8	-5.7
6	1	7	12.2	9.2
6	1	5	17.9	-13.0
6	1	4	20.2	16.0
6	1	3	16.1	13.2
6	1	2	60.5	-60.4
6	1	1	11.7	-10.5
6	1	0	61.8	65.2
6	1	-1	16.5	14.2
6	1	-2	77.4	-78.6
6	1	-3	35.8	-33.7
6	1	-4	43.1	52.0
6	1	-5	32.7	32.9
6	1	-6	56.6	-61.8
6	1	-7	20.7	-16.7
6	1	-8	28.3	29.4
6	1	-10	13.5	11.6
6	1	-11	10.3	-8.5
6	1	-12	23.8	-20.4
6	1	-13	10.2	-8.2
6	1	-14	49.3	48.1
6	1	-15	20.0	16.3
6	1	-16	37.3	-41.7
6	1	-17	18.7	-16.6
6	1	-18	36.2	35.2
6	1	-19	10.1	9.0
6	1	-20	18.7	-15.9

H	K	L	Fc	Fc
5	1	10	14.9	12.9
5	1	8	23.5	-23.9
5	1	6	23.9	25.5
5	1	4	38.1	-34.9
5	1	3	16.7	13.5
5	1	2	43.1	42.0
5	1	1	15.9	-14.0
5	1	0	9.5	-7.3
5	1	-1	25.9	26.2
5	1	-2	26.1	-18.0
5	1	-3	22.9	-24.2
5	1	-4	61.5	67.4
5	1	-5	58.0	59.2
5	1	-6	65.6	-69.6
5	1	-7	59.6	-65.9
5	1	-8	85.6	105.2
5	1	-9	36.5	38.8
5	1	-10	74.2	-87.0
5	1	-11	45.3	-44.4
5	1	-12	52.6	51.7
5	1	-13	31.8	28.2
5	1	-14	44.2	-40.2
5	1	-15	21.4	-15.2
5	1	-16	37.3	28.2
5	1	-18	7.9	-6.9
5	1	-20	8.0	-7.8
4	1	12	15.0	13.0
4	1	11	11.4	-8.2
4	1	10	15.9	-14.6
4	1	9	12.8	12.1
4	1	7	21.3	-16.9
4	1	6	16.4	14.3
4	1	5	35.6	31.9
4	1	4	19.5	-15.7
4	1	3	37.3	-32.9
4	1	2	52.8	53.1
4	1	1	41.2	44.3
4	1	0	55.2	-53.5
4	1	-1	35.0	-32.7
4	1	-2	94.1	111.8
4	1	-3	40.4	38.8
4	1	-4	56.4	-64.6
4	1	-6	74.3	83.0
4	1	-7	22.0	-21.2
4	1	-8	57.7	-60.0
4	1	-9	34.4	30.2
4	1	-10	16.5	-9.9
4	1	-11	25.3	-19.4
4	1	-12	23.4	20.4
4	1	-13	31.9	27.0
4	1	-14	42.8	-39.1
4	1	-15	25.5	-25.2
4	1	-16	13.8	12.0
4	1	-17	14.6	14.4
4	1	-18	29.6	-28.5
4	1	-20	19.0	18.9
3	1	13	4.8	-5.2
3	1	10	10.2	-8.2
3	1	8	26.4	24.6
3	1	6	27.7	-22.8
3	1	5	17.6	12.4
3	1	4	28.9	26.8
3	1	3	52.8	-53.0
3	1	2	48.7	-50.3
3	1	1	55.0	62.2
3	1	0	32.0	31.3
3	1	-1	69.5	-74.1
3	1	-2	11.9	-5.9
3	1	-3	88.6	87.6
3	1	-4	42.9	-42.8
3	1	-5	112.4	-112.6
3	1	-6	15.0	18.5
3	1	-7	66.1	69.1
3	1	-8	56.1	-56.8
3	1	-9	27.8	-22.8
3	1	-10	84.2	88.7
3	1	-11	30.7	27.4
3	1	-12	76.2	-74.5
3	1	-14	42.8	42.3
3	1	-15	8.5	4.9
3	1	-16	14.9	-13.0
3	1	-17	6.5	7.2
3	1	-18	15.4	10.2
2	1	15	5.8	5.4
2	1	14	5.7	6.6
2	1	13	12.3	-8.6
2	1	12	17.7	-15.1
2	1	11	21.4	21.0
2	1	10	18.9	17.1
2	1	9	28.1	-24.4
2	1	8	19.9	-17.7
2	1	7	36.0	38.2
2	1	6	20.9	16.8
2	1	5	70.9	-72.7
2	1	4	36.7	-28.5
2	1	3	41.5	40.7
2	1	2	12.2	0.9
2	1	0	24.4	32.2
2	1	-1	43.4	-41.7
2	1	-2	130.7	-135.5
2	1	-3	49.5	-45.9
2	1	-4	38.0	49.4
2	1	-5	36.1	-34.6
2	1	-6	54.3	-66.6
2	1	-7	64.1	52.5
2	1	-8	60.0	59.8
2	1	-9	49.8	-48.6
2	1	-10	47.7	-42.3
2	1	-11	31.8	27.1
2	1	-12	21.1	19.0
2	1	-13	28.0	-26.2
2	1	-14	26.7	24.9
2	1	-15	15.0	14.7
2	1	-16	18.1	-16.7
2	1	-17	6.7	-5.9
2	1	-18	22.7	18.9
1	1	16	13.6	11.3
1	1	15	5.3	-5.7
1	1	14	22.3	-16.1
1	1	13	6.7	6.7
1	1	12	21.5	20.1
1	1	10	17.3	-12.9
1	1	9	13.7	-11.0
1	1	8	16.0	12.7
1	1	7	14.7	14.8
1	1	6	11.5	8.3
1	1	5	63.4	-67.1
1	1	4	35.7	-33.9
1	1	3	127.5	120.1
1	1	2	86.9	81.8
1	1	1	93.8	-102.9
1	1	0	86.8	-86.4
1	1	-2	94.3	90.4
1	1	-3	48.9	-55.6
1	1	-4	12.7	-6.5

H	K	L	Fo	Fc
1	1	-5	90.9	100.5
1	1	-6	20.6	-18.8
1	1	-7	51.8	-52.0
1	1	-8	38.5	36.0
1	1	-9	25.4	21.6
1	1	-10	42.9	-42.0
1	1	-11	12.1	-10.0
1	1	-12	46.6	45.1
1	1	-14	39.2	-34.6
1	1	-16	25.3	20.3
1	1	-17	7.6	-7.4
1	1	-18	11.1	-10.6
0	1	15	14.5	-11.2
0	1	14	7.5	5.3
0	1	13	18.9	16.4
0	1	12	15.0	13.9
0	1	11	33.1	-30.8
0	1	10	36.2	-23.0
0	1	9	40.7	39.5
0	1	8	43.6	43.5
0	1	7	47.6	-45.0
0	1	6	96.8	-89.4
0	1	5	67.2	61.0
0	1	4	75.7	69.6
0	1	3	40.4	28.4
0	1	2	40.5	-46.7
12	2	-6	6.7	-8.2
12	2	-8	6.7	7.5
12	2	-10	5.2	-6.2
12	2	-14	4.3	4.7
12	2	-16	11.4	-10.5
12	2	-18	11.4	9.4
11	2	-2	4.4	5.7
11	2	-3	3.6	-4.1
11	2	-4	5.8	-6.3
11	2	-5	4.4	3.2
11	2	-6	5.8	-4.9
11	2	-7	6.0	-6.2
11	2	-8	6.2	7.3
11	2	-9	7.8	8.3
11	2	-10	9.1	-10.2
11	2	-12	12.1	13.4
11	2	-14	14.8	-16.0
11	2	-16	13.9	15.7
11	2	-18	8.0	-6.7
10	2	0	3.6	-4.6
10	2	-2	8.7	7.7
10	2	-4	13.9	-16.4
10	2	-5	4.7	5.4
10	2	-6	24.3	26.0
10	2	-7	6.9	-6.6
10	2	-8	20.6	-24.1
10	2	-9	9.9	11.4
10	2	-10	15.2	16.9
10	2	-11	11.5	-13.2
10	2	-12	10.3	-12.7
10	2	-13	12.0	13.3
10	2	-14	6.9	8.0
10	2	-15	6.7	-6.1
10	2	-17	6.3	7.0
10	2	-18	7.2	-8.2
10	2	-20	12.4	12.2
9	2	2	16.1	-15.7
9	2	1	9.8	9.1
9	2	0	22.2	21.2
9	2	-1	11.4	-12.7
9	2	-2	15.5	-15.6
9	2	-3	15.4	13.4
9	2	-4	25.2	25.0
9	2	-5	21.0	-20.9
9	2	-6	12.0	-12.6
9	2	-7	15.3	15.5
9	2	-9	7.1	-5.2
9	2	-10	17.4	19.2
9	2	-11	9.2	-9.2
9	2	-12	19.9	-25.6
9	2	-14	25.3	27.3
9	2	-15	10.4	-9.9
9	2	-16	26.0	-27.0
9	2	-17	6.1	4.7
9	2	-18	15.5	17.9
9	2	-19	7.6	-7.2
9	2	-20	13.4	-12.5
9	2	-21	3.6	4.3
8	2	4	13.5	-12.7
8	2	3	7.2	6.4
8	2	2	8.7	9.6
8	2	-1	7.1	2.8
8	2	-2	17.4	-18.3
8	2	-3	10.0	9.6
8	2	-4	29.6	29.0
8	2	-5	16.9	-17.1
8	2	-6	44.6	-42.2
8	2	-7	23.7	31.5
8	2	-8	54.5	56.2
8	2	-9	44.1	-45.5
8	2	-10	50.0	-56.6
8	2	-11	19.6	23.7
8	2	-12	32.2	34.9
8	2	-13	10.0	-10.0
8	2	-14	15.9	-16.9
8	2	-15	10.1	11.3
8	2	-17	6.9	-6.9
8	2	-18	7.1	6.9
8	2	-20	12.0	-13.2
7	2	6	5.7	6.5
7	2	5	4.3	-5.4
7	2	4	13.6	-15.0
7	2	3	19.3	20.7
7	2	2	24.5	25.1
7	2	1	19.9	-19.2
7	2	0	51.8	-45.9
7	2	-1	24.6	23.3
7	2	-2	49.5	49.6
7	2	-3	36.9	-35.1
7	2	-4	49.1	-52.2
7	2	-5	35.4	33.8
7	2	-6	26.4	25.6
7	2	-7	10.4	-11.5
7	2	-8	18.3	-21.7
7	2	-9	4.4	4.2
7	2	-10	16.3	-19.3
7	2	-11	18.6	23.7
7	2	-12	41.3	40.9
7	2	-13	24.4	-13.5
7	2	-14	32.6	-35.1
7	2	-15	14.5	14.9
7	2	-16	30.2	30.5
7	2	-17	7.0	-6.8
7	2	-18	25.1	-25.7
7	2	-19	10.1	11.2
7	2	-20	19.1	13.3
6	2	8	15.9	16.2
6	2	7	8.4	-7.4
6	2	6	19.9	-24.1

H	K	L	Fo	Fc
6	2	5	5.7	4.0
6	2	4	30.1	31.4
6	2	3	28.8	-27.5
6	2	2	19.9	-18.4
6	2	1	19.9	-21.3
6	2	0	6.5	6.8
6	2	-1	13.5	14.4
6	2	-2	29.2	27.1
6	2	-3	38.1	-36.7
6	2	-4	37.1	-40.5
6	2	-5	36.5	40.3
6	2	-6	53.9	58.5
6	2	-7	66.2	-70.3
6	2	-8	67.2	-75.2
6	2	-9	37.8	39.3
6	2	-10	76.0	81.1
6	2	-11	26.3	-29.1
6	2	-12	52.0	-51.0
6	2	-13	20.1	23.0
6	2	-14	20.4	22.6
6	2	-15	11.6	-10.0
6	2	-16	8.7	-10.7
6	2	-17	5.7	3.7
6	2	-18	8.2	9.3
6	2	-19	5.6	-4.7
6	2	-20	5.4	6.1
5	2	10	7.1	7.7
5	2	9	6.2	5.8
5	2	8	8.3	-8.4
5	2	7	8.9	-10.2
5	2	5	5.8	7.5
5	2	4	19.8	19.0
5	2	3	25.1	-22.9
5	2	2	53.5	-49.8
5	2	1	68.4	70.5
5	2	0	76.0	79.1
5	2	-1	58.4	-54.5
5	2	-2	66.4	-65.9
5	2	-3	24.9	27.9
5	2	-4	60.0	60.3
5	2	-5	4.5	-1.4
5	2	-6	41.2	-46.7
5	2	-7	17.8	25.5
5	2	-8	43.5	47.5
5	2	-9	35.3	40.0
5	2	-10	8.0	-5.6
5	2	-11	17.1	-16.4
5	2	-12	26.7	-24.9
5	2	-13	14.3	14.3
5	2	-14	30.4	29.2
5	2	-15	12.2	-11.8
5	2	-16	30.2	-29.9
5	2	-17	11.7	12.6
5	2	-18	19.9	21.0
5	2	-19	5.4	-6.1
5	2	-20	22.8	-21.1
5	2	-21	7.3	8.6
4	2	11	5.4	5.9
4	2	10	10.5	13.2
4	2	8	17.3	-18.2
4	2	7	11.6	11.7
4	2	6	26.2	24.7
4	2	4	43.5	-45.3
4	2	3	12.9	8.9
4	2	2	38.3	39.4
4	2	1	22.2	23.3
4	2	0	44.5	-43.4
4	2	-1	33.9	-43.8
4	2	-2	27.3	-18.6
4	2	-3	108.9	113.8
4	2	-4	27.6	25.5
4	2	-5	54.2	-60.5
4	2	-5	47.0	-48.6
4	2	-7	23.3	33.9
4	2	-8	76.7	79.4
4	2	-9	74.7	-75.6
4	2	-10	77.2	-77.3
4	2	-11	23.9	22.7
4	2	-12	67.2	62.8
4	2	-13	19.0	-16.8
4	2	-14	42.8	-43.4
4	2	-15	5.8	3.7
4	2	-16	25.2	26.3
4	2	-17	16.8	-15.1
4	2	-18	9.3	-9.9
3	2	12	13.2	16.7
3	2	10	17.0	-19.2
3	2	9	12.0	-15.2
3	2	8	12.2	11.5
3	2	7	13.9	15.3
3	2	6	13.9	-9.5
3	2	5	39.3	-43.2
3	2	4	4.6	-3.1
3	2	3	68.5	69.5
3	2	2	32.8	32.6
3	2	1	47.1	-41.9
3	2	0	64.4	-65.3
3	2	-1	11.5	15.4
3	2	-2	65.3	71.7
3	2	-3	64.1	-47.8
3	2	-4	100.2	-108.2
3	2	-5	43.1	-40.6
3	2	-6	96.1	102.7
3	2	-8	73.6	-68.5
3	2	-9	8.9	-11.0
3	2	-10	13.6	11.2
3	2	-11	12.4	10.1
3	2	-12	13.3	13.0
3	2	-13	35.4	-38.2
3	2	-14	12.2	-12.5
3	2	-15	5.7	7.4
3	2	-16	21.2	21.7
3	2	-17	6.6	-7.4
3	2	-18	20.5	-20.4
3	2	-19	10.7	10.3
3	2	-20	16.6	17.7
2	2	14	8.7	8.7
2	2	12	12.2	-12.0
2	2	8	11.2	12.6
2	2	7	7.6	6.9
2	2	6	21.7	-22.7
2	2	5	5.7	-3.0
2	2	4	97.5	83.2
2	2	3	47.1	42.3
2	2	2	71.6	-62.2
2	2	1	106.8	-103.4
2	2	0	51.0	43.1
2	2	-1	119.1	118.3
2	2	-2	51.3	33.5
2	2	-3	14.2	-27.8
2	2	-4	42.4	35.4
2	2	-5	155.9	100.7
2	2	-6	19.5	18.6
2	2	-7	32.1	-27.3
2	2	-8	75.9	-74.3

H	K	L	Fo	Fc
2	2	-9	45.1	46.1
2	2	-10	72.9	69.6
2	2	-11	15.8	-15.1
2	2	-12	56.6	-56.8
2	2	-13	42.3	41.6
2	2	-14	30.2	28.7
2	2	-16	20.4	-20.6
2	2	-17	7.9	7.0
2	2	-18	9.8	11.3
1	2	15	4.9	-3.3
1	2	14	7.2	7.5
1	2	13	14.7	15.5
1	2	12	14.5	-13.5
1	2	11	15.3	-16.5
1	2	10	24.5	25.7
1	2	9	13.3	15.6
1	2	8	30.1	-29.5
1	2	7	49.4	-48.5
1	2	6	46.4	46.4
1	2	5	22.9	24.9
1	2	4	52.8	-51.1
1	2	3	58.0	-52.1
1	2	2	40.6	-31.1
1	2	1	23.4	-15.8
1	2	0	112.1	118.9
1	2	-2	66.2	-57.9
1	2	-3	74.4	-61.7
1	2	-4	28.8	29.7
1	2	-6	52.5	-50.8
1	2	-7	30.1	-35.4
1	2	-8	57.6	55.2
1	2	-9	34.4	29.5
1	2	-10	34.0	-30.9
1	2	-11	14.0	-14.5
1	2	-12	9.7	8.8
1	2	-13	11.6	11.5
1	2	-15	17.4	-18.5
1	2	-16	12.5	-12.0
1	2	-17	11.8	11.1
1	2	-18	7.4	8.8
0	2	16	18.2	13.1
0	2	14	20.5	-21.2
0	2	13	8.2	5.2
0	2	12	26.7	26.4
0	2	11	6.2	-4.2
0	2	10	14.0	-14.8
0	2	9	31.3	23.5
0	2	8	9.0	10.0
0	2	7	23.4	-23.5
0	2	6	11.1	-8.2
0	2	5	89.3	77.7
0	2	4	30.6	-29.2
0	2	3	121.9	-112.8
0	2	2	12.8	9.7
12	3	-6	4.7	5.6
12	3	-7	3.5	-4.0
12	3	-8	13.6	-12.1
12	3	-10	17.8	14.7
12	3	-11	5.4	-5.6
12	3	-12	16.8	-14.5
12	3	-14	16.6	12.9
12	3	-16	10.6	-9.2
11	3	-4	12.3	10.2
11	3	-5	3.9	-4.4
11	3	-6	12.7	-10.5
11	3	-8	13.0	12.3
11	3	-10	9.0	-8.5
11	3	-14	9.5	11.9
11	3	-16	15.4	-14.7
11	3	-18	18.8	16.4
11	3	-20	13.5	-12.6
10	3	0	7.6	-8.3
10	3	-2	7.1	6.7
10	3	-4	8.1	5.0
10	3	-5	8.4	5.9
10	3	-6	7.1	-7.4
10	3	-7	5.9	5.8
10	3	-8	16.0	13.8
10	3	-10	20.3	-21.9
10	3	-11	5.2	4.2
10	3	-12	27.4	31.3
10	3	-14	25.4	-25.6
10	3	-15	10.6	11.3
10	3	-16	16.5	16.9
10	3	-17	7.0	-6.7
10	3	-18	7.8	-7.5
9	3	-2	12.2	11.8
9	3	-4	20.5	-20.9
9	3	-5	9.6	8.4
9	3	-6	26.6	20.6
9	3	-7	12.2	-13.2
9	3	-8	22.4	-21.5
9	3	-9	11.0	10.8
9	3	-10	9.1	8.9
9	3	-11	6.8	5.3
9	3	-13	4.3	-0.6
9	3	-14	15.1	-15.1
9	3	-15	7.9	-8.8
9	3	-16	12.4	13.1
9	3	-18	18.0	-18.8
9	3	-19	8.5	7.0
9	3	-20	14.7	15.9
8	3	4	7.4	7.3
8	3	2	9.3	-9.5
8	3	1	6.4	6.4
8	3	0	8.9	8.3
8	3	-2	10.1	-8.1
8	3	-3	10.5	9.0
8	3	-5	15.8	-15.5
8	3	-6	5.9	-4.3
8	3	-7	5.9	3.7
8	3	-8	19.8	-20.5
8	3	-10	34.7	34.1
8	3	-11	25.9	-24.9
8	3	-12	27.7	-30.8
8	3	-14	20.7	21.2
8	3	-15	6.1	-5.2
8	3	-16	11.6	-12.8
8	3	-17	6.5	4.8
8	3	-18	10.7	11.5
8	3	-19	6.7	-7.4
8	3	-20	5.5	-5.9
7	3	6	9.2	11.0
7	3	5	6.1	-5.9
7	3	4	12.0	-13.2
7	3	3	7.7	7.8
7	3	2	11.7	12.3
7	3	-5	6.7	-5.4
7	3	-6	7.2	-5.1
7	3	-7	15.0	11.3
7	3	-8	25.0	23.7
7	3	-9	23.4	-25.9
7	3	-10	10.8	-10.4
7	3	-11	12.0	12.5
7	3	-14	9.6	8.8

H	K	L	Fo	Fc
7	3	-15	6.1	-5.2
7	3	-16	8.6	-8.0
7	3	-18	14.1	11.2
7	3	-20	8.5	-9.0
6	3	8	7.3	6.7
6	3	7	5.6	-3.6
6	3	6	12.6	-15.5
6	3	4	9.1	8.5
6	3	1	17.6	-16.1
6	3	0	17.7	-15.4
6	3	-1	20.1	17.9
6	3	-2	19.4	17.4
6	3	-3	9.9	-9.0
6	3	-4	15.5	-15.4
6	3	-5	6.0	7.1
6	3	-6	21.2	20.3
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3	9	8	7.1	6.4
3	9	7	9.3	-10.6
3	9	6	14.0	-15.0
3	9	5	19.2	18.9
3	9	4	32.0	33.4
3	9	3	23.7	-20.9
3	9	2	37.7	-33.4
3	9	1	29.5	25.3
3	9	0	46.8	45.4
3	9	-1	27.7	-23.8
3	9	-2	52.0	-47.9
3	9	-3	9.7	9.0
3	9	-4	47.3	46.6
3	9	-5	15.3	12.7
3	9	-6	19.0	-16.3
3	9	-8	10.6	8.5
3	9	-9	6.0	4.0
3	9	-11	10.6	-8.2
3	9	-12	7.9	-7.5
3	9	-13	14.4	14.8
3	9	-14	15.9	15.2
3	9	-15	7.2	-6.7
3	9	-16	8.0	-7.7
2	9	12	10.4	-11.7
2	9	11	10.2	9.3
2	9	10	19.9	19.8
2	9	9	19.3	-14.8
2	9	8	20.8	-24.9
2	9	7	15.0	16.6
2	9	6	25.5	23.7
2	9	5	12.9	-10.9
2	9	4	31.5	-27.0
2	9	3	13.0	10.8
2	9	2	20.6	17.5
2	9	1	5.1	2.8
2	9	0	6.6	-5.4
2	9	-1	21.0	-17.1
2	9	-2	24.6	-23.5
2	9	-3	30.4	31.2
2	9	-4	37.9	35.6
2	9	-5	26.0	-21.0
2	9	-6	55.8	-47.6
2	9	-7	6.9	4.9
2	9	-8	36.6	31.6
2	9	-9	19.9	-18.0
2	9	-10	25.5	-25.6
2	9	-11	15.0	11.3
2	9	-12	13.8	18.8
2	9	-13	7.4	-6.2
2	9	-14	13.3	-11.2
1	9	13	7.3	6.8
1	9	12	8.1	8.1
1	9	11	6.0	-4.4
1	9	8	15.0	-13.7
1	9	7	17.5	15.2
1	9	6	27.7	27.4
1	9	5	22.2	-20.8
1	9	4	41.1	-43.0
1	9	3	21.9	20.8
1	9	2	53.5	51.4
1	9	1	27.0	-27.6
1	9	0	53.9	-73.6
1	9	-1	18.8	24.9
1	9	-2	44.4	61.0
1	9	-3	24.3	-26.9
1	9	-4	49.9	-48.5
1	9	-5	5.2	7.3
1	9	-6	28.2	24.2
1	9	-7	9.5	-8.3
1	9	-8	10.1	-7.4
1	9	-9	8.4	-6.5
1	9	-11	6.2	5.1
1	9	-12	6.5	7.3
1	9	-13	13.4	-10.6
1	9	-14	8.2	-7.0
1	9	-15	6.7	6.1
0	9	14	7.7	-8.0
0	9	13	8.5	7.7
0	9	12	17.0	15.5
0	9	11	10.0	-7.9
0	9	10	28.6	-30.4
0	9	9	24.7	25.4
0	9	8	33.5	34.7
0	9	7	24.3	-22.0
0	9	6	37.9	-39.2
0	9	5	22.2	18.2
0	9	4	32.6	30.0
0	9	3	6.2	-3.3
0	9	2	24.5	-23.2

TABLE 3.5

(A) ANALYSIS OF STRUCTURE FACTOR DATA AS A FUNCTION OF LAYER LINE INDEX. ALL FIGURES ARE ON ABSOLUTE SCALE.

k	$\Sigma\|F_o\|$	$\Sigma\|F_c\|$	$\Sigma\Delta$	N	R	$\Sigma\Delta/N$
0	5889	5046	967	148	0.1642	6.53
1	8411	8080	830	283	0.0987	2.93
2	7957	7923	678	298	0.0852	2.27
3	6430	6025	599	250	0.0932	2.40
4	6658	6641	586	278	0.0881	2.11
5	5902	5787	436	270	0.0738	1.61
6	4851	4793	448	254	0.0923	1.76
7	4202	4169	381	228	0.0907	1.67
8	3665	3501	344	224	0.0940	1.54
9	2895	2705	326	192	0.1141	1.70
ALL	56822	54670	5596	2425	0.0985	2.31

(B) ANALYSIS OF STRUCTURE FACTOR DATA BY MAGNITUDE OF Fo.

0 - 13	8404	7966	1190	1030	0.1417	1.16
13 - 25	11745	11090	1399	641	0.1191	2.18
25 - 50	17399	16690	1580	498	0.0908	3.17
50 - 75	9397	9121	797	157	0.0848	5.08
75 - 100	5280	5225	366	62	0.0693	5.90
100 - 200	4597	4579	263	37	0.0573	7.12

TABLE 3.6

ORTHOGONAL COORDINATES AND E.S.D.S.

ATOM	X'	Y	Z'
As(1)	2.325 ± 1	2.811 ± 1	2.042 ± 1
Br(1)	1.809 ± 1	-1.032 ± 1	2.631 ± 1
O(1)	1.065 ± 6	1.907 ± 7	2.766 ± 6
C(1)	2.641 ± 9	2.181 ± 10	0.287 ± 8
C(2)	3.950 ± 10	2.091 ± 14	-0.156 ± 10
C(3)	4.162 ± 12	1.652 ± 15	-1.475 ± 11
C(4)	3.107 ± 14	1.335 ± 14	-2.278 ± 11
C(5)	1.839 ± 12	1.424 ± 13	-1.818 ± 10
C(6)	1.571 ± 10	1.827 ± 12	-0.501 ± 10
C(7)	3.863 ± 10	2.754 ± 12	3.141 ± 9
C(8)	4.814 ± 9	3.729 ± 12	3.008 ± 10
C(9)	5.965 ± 11	3.691 ± 13	3.846 ± 12
C(10)	6.096 ± 13	2.667 ± 16	4.769 ± 13
C(11)	5.151 ± 14	1.721 ± 16	4.865 ± 13
C(12)	3.998 ± 12	1.712 ± 14	4.059 ± 11
C(13)	1.660 ± 11	4.554 ± 11	2.001 ± 8
C(14)	0.948 ± 12	5.050 ± 12	3.073 ± 11
C(15)	0.466 ± 14	6.371 ± 15	3.031 ± 13
C(16)	0.699 ± 13	7.155 ± 13	2.004 ± 15
C(17)	1.411 ± 14	6.690 ± 14	0.886 ± 14
C(18)	1.882 ± 13	5.397 ± 13	0.897 ± 11

TABLE 3.7

BOND LENGTHS AND E.S.D.S.

As(1) - O(1)	1.712 ± 7 Å	C(8) - C(9)	1.425 ± 15 Å
As(1) - C(1)	1.891 ± 9	C(9) - C(10)	1.385 ± 20
As(1) - C(7)	1.891 ± 10	C(10) - C(11)	1.341 ± 21
As(1) - C(13)	1.867 ± 11	C(11) - C(12)	1.406 ± 18
C(1) - C(2)	1.385 ± 14	C(12) - C(7)	1.396 ± 16
C(2) - C(3)	1.406 ± 16	C(13) - C(14)	1.379 ± 15
C(3) - C(4)	1.363 ± 18	C(14) - C(15)	1.407 ± 19
C(4) - C(5)	1.351 ± 18	C(15) - C(16)	1.312 ± 20
C(5) - C(6)	1.403 ± 15	C(16) - C(17)	1.404 ± 20
C(6) - C(1)	1.376 ± 13	C(17) - C(18)	1.376 ± 19
C(7) - C(8)	1.368 ± 15	C(18) - C(13)	1.407 ± 15

INTRAMOLECULAR DISTANCES

Br(1)...O(1)	3.034 ± 12	As(1)...Br(1)	3.922 ± 3

TABLE 3.8

INTERBOND ANGLES AND E.S.D.S.

O(1)-As(1)-C(1)	109.8± 4	As(1) -C(7) -C(12)	118.8± 8
O(1)-As(1)-C(7)	109.7± 4	C(8) -C(7) -C(12)	121.9±10
O(1)-As(1)-C(13)	103.9± 4	C(7) -C(8) -C(9)	119.1±10
C(1)-As(1)-C(7)	113.2± 4	C(8) -C(9) -C(10)	119.2±11
C(1)-As(1)-C(13)	110.5± 4	C(9) -C(10)-C(11)	120.2±12
C(7)-As(1)-C(13)	109.3± 4	C(10)-C(11)-C(12)	122.8±13
As(1)- C(1)-C(2)	118.5± 7	C(7) -C(12)-C(11)	116.8±12
As(1)- C(1)-C(6)	119.2± 7	As(1) -C(13)-C(14)	120.2± 8
C(2)- C(1)-C(6)	122.4± 9	As(1) -C(13)-C(18)	121.4± 8
C(1)- C(2)-C(3)	117.5±10	C(14)-C(13)-C(18)	118.4±10
C(2)- C(3)-C(4)	120.6±11	C(13)-C(14)-C(15)	119.4±11
C(3)- C(4)-C(5)	120.6±11	C(14)-C(15)-C(16)	121.6±13
C(4)- C(5)-C(6)	121.2±11	C(15)-C(16)-C(17)	121.0±13
C(1)- C(6)-C(5)	117.6± 9	C(16)-C(17)-C(18)	118.6±12
As(1)- C(7)-C(8)	119.2± 8	C(13)-C(18)-C(17)	121.0±11

TABLE 3.9

INTERMOLECULAR DISTANCES < 3.80 ANGSTROMS

C(10)...O(1)	i	3.52	C(5)...C(9)	vi	3.71
C(10)...C(3)	ii	3.55	C(2)...C(16)	vi	3.75
C(2)...C(17)	vi	3.61	C(17)...C(18)	iv	3.75
O(1)...C(15)	v	3.63	Br(1)...C(5)	i	3.76
C(9)...O(1)	i	3.63	C(4)...C(9)	vi	3.77
C(11)...C(3)	ii	3.64	C(4)...C(4)	iii	3.79
C(17)...C(17)	iv	3.66	C(12)...Br(1)	vi	3.79
C(10)...C(14)	i	3.65			

Roman numerals indicate the following transformations which must be applied to the atomic coordinates given in table 3.2.

i	1 + x,	1/2 - y,	1/2 + z.
ii	x,	1/2 - y,	1/2 + z.
iii	1 - x,	- y,	- z.
iv	- x,	1 - y,	- z.
v	- x,	-1/2 + y,	1/2 - z.
vi	1 - x,	-1/2 + y,	1/2 - z.

The transformations are applied to the coordinates of the second atom in each case.

TABLE 3.10

MEAN MOLECULAR PLANES

(A) Atoms defining planes.

Plane 1	C(1) TO C(6)	Plane 4	O(1),As(1),C(1).
Plane 2	C(7) TO C(12)	Plane 5	O(1),As(1),C(7).
Plane 3	C(13)TO C(18)	Plane 6	O(1),As(1),C(13).

(B) Plane Equations.

Plane No.	P	Q	R	S	RMSD *
1	-0.0523	0.9465	-0.3183	1.8225	0.009
2	0.4784	-0.5508	-0.6839	-1.8110	0.004
3	-0.8514	-0.3159	-0.4187	-3.6868	0.005
4	-0.6709	0.6512	-0.3547	-0.4537	-
5	0.3126	-0.8198	-0.4798	-2.5574	-
6	-0.3950	-0.1719	-0.9025	-3.2445	-

*These quantities are defined in table 1.12. The plane is given by

$$PX' + QY + RZ' = S$$

where X', Y and Z' are the atomic coordinates in Angstroms referred to orthogonal axes defined by X' parallel to a*, Y parallel to b and Z' parallel to c.

(C) Deviations from mean planes(in Angstroms).

	Plane 1	Plane 2	Plane 3
	i = 0	i = 6	i = 12
C(1 + i)	0.012	-0.006	-0.003
C(2 + i)	-0.001	0.003	-0.003
C(3 + i)	-0.007	0.002	0.008
C(4 + i)	0.003	-0.003	-0.008
C(5 + i)	0.008	0.000	0.002
C(6 + i)	-0.016	0.005	0.004

(D) Dihedral angles (in degrees).

Plane A	Plane B	∠AB
1	4	40.1
2	5	21.7
3	6	39.8

3.4 Discussion.

The crystal structure analyses of triphenyl arsenic hydroxychloride (CLAS) and triphenyl arsenic hydroxybromide (BRASO) establish that the partial hydrolysis of triphenyl-arsine dihalides does not give rise to compounds in which the arsenic atom is penta-coordinate, as was generally considered to be the case. The compounds are more correctly formulated as 1:1 adducts of triphenylarsine oxide and the corresponding halogen acid.

The arrangement around the arsenic atoms in both cases is approximately tetrahedral. One O-As-C bond angle in each molecule, however, is found to differ significantly from the tetrahedral value. The observed values are 104.3 ± 0.8° for O(1)-As(1)-C(13) and 104.7 ± 0.8° for O(2)-As(2)-C(31) in CLAS (there are two molecules in the asymmetric unit) and 103.9 ± 0.4° for O(1)-As(1)-C(13) in BRASO. It can be seen from figure 3.4 that this corresponds to the angle on the opposite side of the arsenic from the halogen atom in each molecule. A mechanism involving attraction between the oxygen atom and the hydrogen atom on the upper carbon of the benzene ring (that is C(18) and C(32) in CLAS and C(14) in BRASO) may be the cause of this contraction in the angle. The remaining O-As-C and C-As-C bond angles approximate closely to the tetrahedral value (tables 2.8

and 3.8).

The halogen atom is not bonded directly to the arsenic atom in either CLAS or BRASO, the arsenic-halogen distances being 3.78 Å and 3.74 Å in CLAS and 3.92 Å in BRASO. The composition of the adduct is maintained by hydrogen bonding between the halogen acid and the triphenylarsine oxide moiety. The O...H...Cl distances in CLAS are 2.854 Å and 2.817 Å (both ± 0.017 Å) which do not agree with the value 3.05 ± 0.02 Å in D(-)-isoleucine hydrochloride (Trommel and Bijvoet, 1954) or the average value of 3.08 ± 0.01 Å reported by Pimentel and McClellan (1960); in BRASO the O...H...Br distance is 3.034 ± 0.012 Å which is considerably shorter than the value found in D(-)-isoleucine hydrobromide (3.29 ± 0.02 Å, Trommel and Bijvoet, 1954) and the mean value of 3.38 ± 0.01 Å given by Pimentel and McClellan (1960). The significant decrease in the oxygen-halogen distance is comparable with the shortening of the oxygen-oxygen distance in short, possibly symmetric, hydrogen bonds (Speakman, 1967). The separation of the oxygen and halogen atoms in CLAS and BRASO makes it improbable that the hydrogen atom (in the hydrogen bond) is symmetrically disposed as this would involve an oxygen-hydrogen distance of the order of 1.4-1.5 Å. Unfortunately, the position of the hydrogen bonded proton could not be determined with any degree of accuracy in a difference map; only a diffuse region of positive density (≤ 0.4 e/Å^3) was observed in either case. Evidence for the position of this proton may be derived, however, from an examination of the arsenic-oxygen bond lengths and the spectral properties of both compounds.

The arsenic-oxygen bond lengths observed in CLAS and BRASO are 1.702 Å and 1.697 Å (both ± 0.014 Å) and 1.712 ± 0.007 Å respectively, which compare favourably with the value of 1.69 ± 0.01 Å found in o-phenylenediarsine oxychloride (Trotter, 1962) where the arsenic-oxygen bond is formally regarded as being single. The value for the arsenic-oxygen double bond has been determined as 1.644 ± 0.007 Å (Part III, Chapter 1 of this thesis). The significant (Cruickshank and Robertson, 1953) increase in the arsenic-oxygen distance may be rationalised by assuming that the proton in the hydrogen bond is closely associated with the oxygen of the triphenylarsine oxide moiety. Such an association would lead to a reduction of the double-bond character of the arsenic-oxygen bond by reducing the overlap between the filled oxygen $p\pi$ orbitals and the vacant arsenic $d\pi$ orbitals.

The most prominent feature in the infra-red spectra of CLAS and BRASO is a complex absorption in the region 2750-2000 cm.$^{-1}$. This absorption has been assigned to an (O-H) stretching frequency under the influence of strong hydrogen bonding. Similar absorptions are observed in the infra-red spectra of the analogous amine oxide hydrohalides (Giguere and Chin, 1961; Tsoucaris, 1961) and phosphine oxide adducts (Hadzi, 1962). In these cases, strong hydrogen bonding with possible proton transfer has been proposed.

The absorption frequency corresponding to the As=O stretching mode which is observed at <u>ca</u>. 890 cm.$^{-1}$ in triphenylarsine oxide monohydrate is not present in the spectra of either CLAS or BRASO. Both hydroxyhalides have transmission windows in this region. The As-O stretching frequency in the hydroxyhalides has been assigned as a strong absorption <u>ca</u>. 790 cm.$^{-1}$. The decrease (100 cm.$^{-1}$) in the absorption frequency of the arsenic-oxygen stretching mode is compatible with an increase in bond length and therefore with reduced double-bond character; it is also in agreement with the assumption of protonation of the oxygen of the triphenylarsine oxide.

Phillips and Tyree (1961) have observed smaller decreases in ν(As=O) in the infra-red spectra of complexes of triphylarsine oxide and various Lewis acids. These shifts have been attributed to a decrease in the back-donation from oxygen to arsenic caused by the oxygen acting as the donor atom.

The nuclear magnetic resonance spectra of the series of compounds Ph_3AsO, $Ph_3AsOHCl$ and $Ph_3AsOHBr$ yield further evidence for the association of the proton in the hydrogen bond with the oxygen of the triphenylarsine oxide moiety. The signal corresponding to the aromatic protons of triphenylarsine oxide occurs at 2.4τ when the spectrum is recorded under anhydrous conditions in deuterochloroform. Introduction of a strongly protonating agent (trifluoroacetic acid) to the solvent results in a shift of the signal to 2.14τ . The signal for the aromatic protons in both CLAS and BRASO appears at $\sim 2.15\tau$ irrespective of the presence of strongly protonating or deprotonating agents in the solvent (trifluoroacetic acid or hexamethylphosphoramide). The chemical shift, in the signal corresponding to the aromatic protons, has also been observed in the analogous triarylphosphine oxides under similar conditions (Keat, 1968), in which case it has been interpreted as arising from a reduction of the back-donation from the oxygen to phosphorus by protonation of the oxygen. This could result in either an increase in overlap between the vacant arsenic $d\pi$ orbitals and the aromatic π electron system or an increased inductive effect, both of which would result in a downfield shift in the nuclear magnetic resonance signal. Both mechanisms, however, are compatible with the protonation of the oxygen atom of the phosphine oxide. A similar argument may be adopted in the case of triarylarsine oxides which are stronger bases (Hadzi, Klofutar and Oblak, 1968) and therefore more susceptible to protonation.

Increase in overlap between the vacant arsenic $d\pi$ orbitals and the aromatic π electron system, or an increased inductive effect might lead to a shortening of the arsenic-carbon bond lengths. The mean observed values of the arsenic-carbon bond lengths are 1.897±0.020 Å in CLAS and 1.883 ± 0.011 Å in BRASO. These values are not significantly different and compare favourably with the values of 1.899 ± 0.009 Å in tetraphenylarsonium 3-fluoro-1,1,4,5,5-pentacyano-2-azapentadieneide (Palenik, 1966), 1.907 ± 0.009 Å in triphenylarsine oxide monohydrate (Part III, Chapter 1 of this thesis) and 1.90 ± 0.01 Å in $[(C_6H_5)_4As]^+ [BrIBr]^-$ (McEwan, 1965). The values in all these compounds differ significantly (Cruickshank and Robertson, 1953) from the value of 1.966 ± 0.021 Å in arsenobenzene (Hedberg, Hughes and Waser, 1961) and 1.96 ± 0.01 Å (mean value, Sutton, 1965). The decrease in the arsenic-carbon bond length may arise from the change in coordination of the arsenic atom (three-coordinate in arsenobenzene and four-coordinate in the other compounds) but has been ascribed, in the case of the tetraphenylarsonium compounds, to a formal charge effect.

Since the arsenic atom in CLAS and BRASO has, at least, a partial positive charge (the dipole moment of CLAS in benzene solution is 9.2 D.; Jensen, 1943), a formal charge effect may also be responsible for the shortening of the carbon-arsenic bonds. Shortening of the bonds by a mechanism involving increased overlap of the $d\pi$ and $p\pi$ orbitals or by an increased inductive effect, however, cannot be dismissed and may contribute to some extent in the case of CLAS and BRASO.

The mean carbon-carbon (aromatic) bond lengths found in these analyses are 1.40 ± 0.04 Å in CLAS and 1.39±0.03 Å in BRASO; the average values of the corresponding valence angles are 119.6 ± 2.1° and 119.9 ± 1.6° respectively. (Individual values of bond lengths are in tables 2.8 and 3.8; angles in tables 2.9 and 3.9). These values are in accord with the accepted values (Sutton, 1965).

The phenyl groups are planar within experimental error (tables 2.10 and 3.10) and adopt a conformation in which the triphenylarsine oxide moiety has no symmetry. The phenyl groups in each molecule are rotated in the same sense (figure 3.4) forming a "propellor type" arrangement, but the rings are rotated about the arsenic-carbon bonds by differing amounts. The rotations are given by the dihedral angles between the plane of the phenyl group and the plane defined by the atoms O-As-C(i) where C(i) is the carbon atom bonded to arsenic. The dihedral angles angles are given in table A (a positive dihedral angle represents a clockwise rotation of the phenyl group from the plane O-As-C(i)).

Table A.

Molecule	Plane 1	Plane 2	Angle
CLAS(1)	O-As-C(1)	C(1)-C(6)	-34.6°
	O-As-C(7)	C(7)-C(12)	-30.6
	O-As-C(13)	C(13)-C(18)	-48.8
CLAS(2)	O-As-C(19)	C(19)-C(24)	-25.5
	O-As-C(25)	C(25)-C(30)	-37.5
	O-As-C(31)	C(31)-C(36)	-62.7
BRASO	O-As-C(1)	C(1)-C(6)	+40.1
	O-As-C(7)	C(7)-C(12)	+21.7
	O-As-C(13)	C(13)-C(18)	+39.8

The relevant plane equations are given in tables 2.10 (CLAS) and 3.10 (BRASO). As the phenyl groups of the two crystallographically independent molecules of CLAS in the asymmetric unit do not have their phenyl groups in the same orientation, the distortion may be ascribed to crystal packing forces in both compounds.

The packing of the molecules of CLAS in the unit cell, projected down the b-axis is shown in figure 2.3. There are two molecules in the asymmetric unit and they pack with the arsenic-chlorine vector approximately parallel to the b-axis but directed in opposite senses. The molecules form infinite columns up the two-fold screw axes, the halogen atom occupying the space beneath the phenyl groups of the next molecule. The intermolecular distances (table 2.9) are all in the range corresponding to normal van der Waals contacts.

The molecular packing of BRASO as projected on (100) is shown in figure 3.3. No short intermolecular contacts are found (table 3.9).

The structures of triphenyl arsenic hydroxychloride and triphenyl arsenic hydroxybromide therefore correspond to the 1:1 adducts of triphenylarsine oxide and either hydrogen chloride or hydrogen bromide. The composition of the adduct is maintained by hydrogen bonding arising from competition for the proton between the arsenic base and the conjugate base of the halogen acid. The proton in the hydrogen bond has not been located but the crystallographic and spectral evidence supports the contention that the proton is more closely associated with the oxygen atom. Since the infrared spectra of these compounds resemble the spectra of compounds which contain an oxygen-oxygen hydrogen bond which is symmetrical, the hydrogen bonds in CLAS and BRASO may be regarded as the hetero-atom analogue of the short, possibly symmetric, oxygen-oxygen hydrogen bonds.

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Appendix.

The Linear Diffractometer

Appendix.

The Linear Diffractometer.

The Hilger and Watts Y-190 linear diffractometer (Arndt and Phillips, 1961) is a device for measuring the integrated intensities of diffracted X-ray beams automatically. It consists of a stabilised X-ray generator, a mechanical analogue of the reciprocal lattice for positioning both crystal and counter simultaneously, a radiation counter and punch / printer output for recording the measured intensities. Molybdenum radiation is normally employed and is monochromated by use of balanced strontium oxide / zirconium oxide Ross filters. A scintillation counter / pulse height analyser system is used to measure the intensities.

The mechanical analogue of the reciprocal lattice is composed of three slides, the movements of which are linked with those of the crystal and counter. The crystal is set with each of its reciprocal axes parallel to one of the slides and each crystal plane is brought into the reflecting condition by movement of the slides. As movement of the crystal and counter are linked, when a crystal plane satisfies the reflecting conditions the counter is in a position to receive the diffracted beam. In the normal mode of operation, the intensity of each reflection in a reciprocal lattice net is measured by automatic scanning on two slides; the reflections in upper layers are brought into the reflecting condition by adjusting the third slide manually as in the equi-inclination Weissenberg photographic method.

The integrated intensity is measured by rotating the crystal plane out of the reflecting condition and counting the background for t seconds. The crystal plane is then turned at constant speed for 2t seconds through the reflecting condition, counting continuously . Finally the background is measured for a further t seconds. The intensity of the reflection is then given by the second count minus the sum of the first and third. The complete scanning cycle is repeated for each filter and the integrated intensity is the difference between the counts with the zirconium filter and those with the strontium filter.