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The Structure of (+)-Demethanolaconinone Hydriodide Trihydrate

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The space group of (+)-demethanolaconinone hydriodide trihydrate,

$$C_{19}H_{17}N.C_2H_5(OH)_4.O.(OCH_3)_3.HI.3\,H_2O$$

is $P2_12_12_1$, the unit-cell dimensions are

$$a=14\cdot69,\ b=22\cdot08,\ c=8\cdot33\ \text{Å};\ Z=4\ .$$

The coordinates of the iodine atom were found by means of two-dimensional Patterson syntheses and ϱ_0 maps were obtained for the three projections using the heavy-atom technique. The structure was solved only on the basis of the above empirical formula from a three-dimensional Patterson synthesis interpreted by vector convergence method. The coordinates were refined by two-dimensional methods and one three-dimensional differential synthesis. The carbon–nitrogen skeleton was found to be the same as in the case of des-(oxymethylene)-lycoctonine. The absolute configuration of the (+)-isomer has been determined.

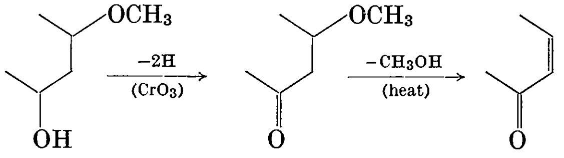
Introduction

Aconitine is the most readily accessible and important alkaloid of the Aconitum class. The chemical studies on aconitine reported before 1900 have been fully reviewed by Schulze (1906), and those covering the first half of the 20th century by Stern (1954).

When this investigation was undertaken the ring structure of aconitine was unknown. Although Schneider (1956) suggested that aconitine possesses the same carbon–nitrogen skeleton as that derived for lycoctonine from an X-ray analysis of des-(oxymethylene) lycoctonine hydriodide (Przybylska & Marion, 1956), no chemical evidence of this postulate was forthcoming until 1959. It was certain, however, that it contains three hydroxyls, four methoxyl groups, an acetate, a benzoate and an ethyl group attached to the nitrogen atom.

The examination of crystals and determination of space groups of aconitine hydriodide, aconine hydriodide, benzoylaconine hydriodide and demethanolaconinone hydriodide showed that the last compound is the most suitable for a detailed analysis.

Aconitine is oxidised by chromic acid to the ketone aconitinone, which very readily loses one methoxyl group which is eliminated as methanol and gives rise to aconitoline.



Aconitine Aconitinone Aconitoline

The presence of an $\alpha\beta$-unsaturated carbonyl group in aconitoline has been proved by Mayer & Marion (1959) and indirectly by the results of this X-ray investigation.

Demethanolaconinone was obtained by saponification of aconitoline which removes the acetyl and benzoyl groups. This compound was then converted to hydriodide, which was found to contain three molecules of water of crystallization.

The chemical reactions which lead to the formation of demethanolaconinone hydriodide from aconitine are carried out under mild conditions and it can safely be assumed that the ring skeleton of these two compounds is the same.

The empirical formula of demethanolaconinone hydriodide trihydrate could be expanded to

$$C_{19}H_{17}N\,.\,C_2H_5(OH)_4\,.\,O\,.\,(OCH_3)_3\,.\,HI\,.\,3\,H_2O\ .$$

In this X-ray analysis the molecular structure of demethanolaconinone has been solved on the basis of this formula alone and no other chemical evidence was taken into account.

Soon after the results of this work were submitted for publication (Przybylska & Marion, 1959*a*), Wiesner *et al.* (1959) proposed the complete structure of aconitine, in which the only ambiguity was that one hydroxyl could exchange position with a methoxyl group. That structure, the result of an independent chemical study, was found to be in full agreement with this X-ray investigation.

Experimental

Pure, colourless crystals of the trihydrate were prepared by Mayer (1959). They were crystallized from methanol-ether, m.p. was 240–241 °C., with decomposition, when immersed at 220 °C. The hydrate dried in air was used for chemical analysis, and the optical rotation was found by Dr Mayer to be

$$[\alpha]_D^{25} = +59{\cdot}2 \pm 1{\cdot}5° \ (c,\ 1{\cdot}04 \text{ in water}).$$

The crystals were found to belong to the orthorhombic bisphenoidal class, and the systematic extinctions were only $h00$ when h is odd, $0k0$ when k is odd, and $00l$ when l is odd. The space group is therefore $P2_12_12_1$. The unit cell, whose dimensions measured from precession photographs are

$$a = 14{\cdot}69 \pm 0{\cdot}03,\ b = 22{\cdot}08 \pm 0{\cdot}03,\ c = 8{\cdot}33 \pm 0{\cdot}02\ \text{Å}\ ,$$

contains four molecules. The crystals were elongated along the c axis. The density measured at 23·2 °C. by flotation was found to be 1·582 g.cm.$^{-3}$ and the calculated value for the trihydrate is 1·592 g.cm.$^{-3}$. The number of electrons per unit cell, $F(000)$, is 1336; the absorption coefficient for Cu $K\alpha$ radiation is 107 cm.$^{-1}$.

Three-dimensional data were collected from multiple-film, equi-inclination Weissenberg photographs of $hk0 \ldots hk6$ and of $0kl \ldots 9kl$ reflections. All the $hk0$–$hk6$ reflections were measured whereas the $0kl$–$9kl$ reflections were used only to supplement the data. The $h0l$ reflections were also collected. Out of a possible 3477 reflections which lie within the range of Cu $K\alpha$ radiation, a total of 2773 were measured. All crystals used for this investigation were cut so that they were cylindrical in shape with diameters ranging from 0·12 to 0·20 mm. and therefore absorption corrections were not applied.

Some intensities were roughly corrected for the variation in spot size, and all of them were corrected for the Lorentz and polarization factors and correlated. Overall scale and temperature factors were evaluated using the $hk0$ data. Later, when the structure was solved, the absolute scale of F_o values was readjusted by direct comparison with F_c values.

Determination of the structure

The three Fourier projections of the molecule on (001), (010) and (100) planes were calculated by the heavy-atom technique, using the coordinates of the iodine atom of

$$x = 0{\cdot}167,\ y = 0{\cdot}209,\ z = 0{\cdot}170\ ,$$

which were derived from two-dimensional Patterson syntheses. A three-dimensional Patterson synthesis was then computed for 30,000 points by Dr F. R. Ahmed using the generalised programme as described by Ahmed & Barnes (1958). The intensities were sharpened and modified as described by Donohue & Trueblood (1952). The $F^2(000)$ term was omitted.

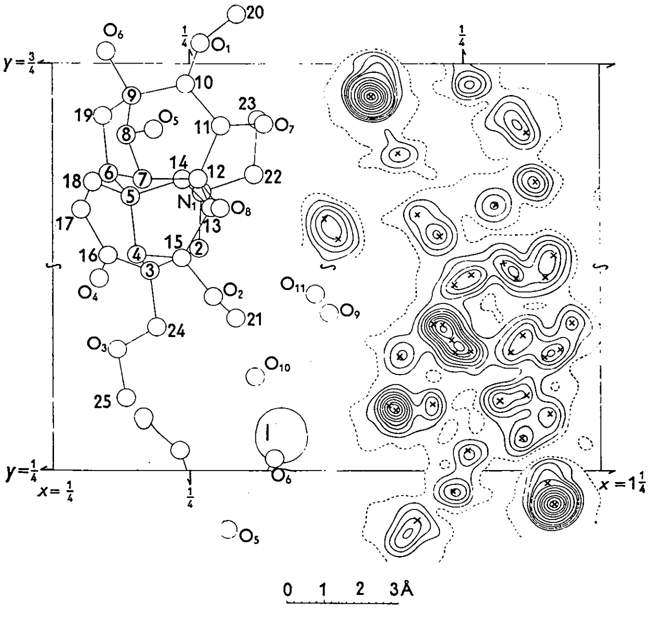


Fig. 1. The structure of (+)-demethanolaconinone hydriodide trihydrate projected down the c axis. The first broken contour is at 2 e.Å^{-2}. All other intervals are at 2 e.Å^{-2} with the exception of those above 10 e.Å^{-2} for the iodine atom which are at 10 e.Å^{-2}.

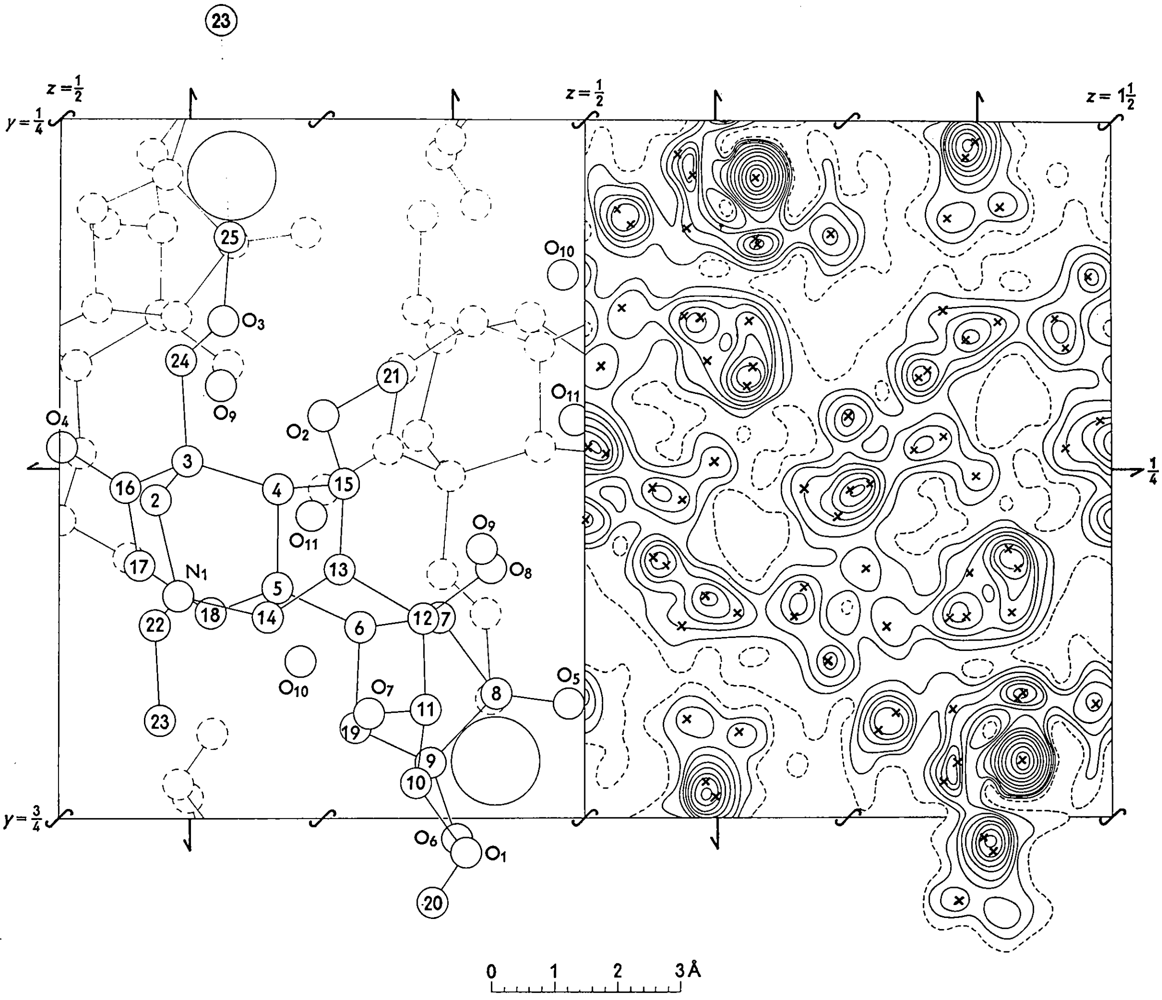


Fig. 2. A projection along the *a* axis. The contours of the light atoms are drawn at 2 $e.\text{Å}^{-2}$ starting with the broken line at 4 $e.\text{Å}^{-2}$. The iodine atom contours are at intervals of 10 $e.\text{Å}^{-2}$, starting at 10 $e.\text{Å}^{-2}$.

The results of this synthesis were then interpreted by a vector convergence method which was first described by Beevers & Robertson (1950). A new three-dimensional electron-density synthesis, called vector-convergence distribution was constructed by four transcriptions of the Patterson synthesis with its origin transferred to the four equivalent positions of the iodine atom. The positive peaks only of the Patterson sections perpendicular to the *c* axis were plotted on sheets of perspex, which were then assembled together to give a three-dimensional model. Those peaks of the vector-convergence distribution which showed four-fold coincidences were correlated with two-dimensional Fourier projections and in spite of the presence of some spurious peaks, it was possible to locate 32 of the light atoms of the molecule.

All these atoms were treated as C atoms for the first structure-factor calculations for the *hk*0 and 0*kl* zones. Only one cycle of refinement using F_o-F_c syntheses for these two zones established the location of all the remaining atoms of the molecule with the exception of one oxygen of the molecule of water of crystallization. It indicated also which atoms are oxygen atoms.

The ring skeleton was found to be identical with that in des-(oxymethylene)-lycoctonine (Przybylska, 1961). It would have been impossible at this stage of the work to recognize the nitrogen atom. The ethyl group was, however, identified and using the chemical evidence that the ethyl group is attached to the nitrogen atom (Jacobs & Elderfield, 1936; Majima & Tamura, 1936), it was possible to distinguish the nitrogen atom from the other light atoms of the molecule.

It is interesting that even at this early stage of the work, the calculation of the bond lengths revealed the position of the carbonyl group and the C=C double bond, as these interatomic distances were found to be considerably shorter. This result was found in agreement with the chemical evidence. (Mayer & Marion, 1959).

By one more two-dimensional cycle of ϱ_o and $\varrho_o-\varrho_c$ syntheses for all three zones it was possible to locate unambiguously the last oxygen atom of the water of crystallization. One additional refinement cycle brought the R factor for the $hk0$, $0kl$ and $h0l$ zones to 0·15, 0·20 and 0·22 respectively. At this stage the structure factors were evaluated for the three-dimensional data and the overall factor was found to be 0·19. A differential synthesis which was then carried out gave average atomic shifts of 0·037, 0·026 and 0·040 Å in the x, y and z directions respectively, and the corresponding maximum shifts were 0·093, 0·077 and 0·166 Å. These are the final values, already corrected for the finite-summation errors given by the F_c differential synthesis and multiplied by a factor of 1·7 to allow for the fact that the structure has no centre of symmetry. The bond lengths calculated with the new set of coordinates gave considerably better agreement with theoretical values. The structure-factors for all three-dimensional data were evaluated again and the overall discrepancy factor was as before $R=0·19$, whereas the discrepancy factors for the three zones became slightly lower. Including the unobserved reflections, they are:

$$R(hk0)=0·13,\ R(h0l)=0·20,\ R(0kl)=0·21\ .$$

The R values for the $h0l$ and $0kl$ spectra are considerably higher than for the $hk0$ reflections, because the z coordinates are not refined to the same degree of accuracy as the x and y coordinates. The c axis is the shortest and the [010] and [100] projections do not offer as good a resolution of atomic peaks as the projection along the c axis.

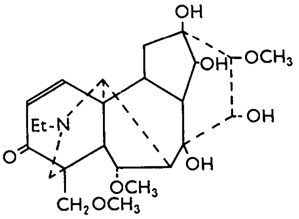


Fig. 3. The diterpenoid representation of (+)-demethanolaconinone.

The scattering curves of McWeeny (1951) were used for carbon, nitrogen and oxygen and that of Thomas & Umeda (1957) for iodine. The temperature factor $B=4·0$ Å^2 was employed in structure-factor calculations. The F_o-F_c electron-density maps indicated an anisotropic thermal motion of the iodine ion in a direction almost parallel to the a axis. No allowance, however, was made for this motion in the calculation of F_c values as the molecular structure of (+)-demethanolaconinone has been solved beyond any doubt and further refinement seemed unnecessary. For the same reason the contribution made by hydrogen atoms was not taken into account.

Table 1. *Fractional atomic coordinates*

Atom	x	y	z
N_1	0·522	0·592	−0·271
C_2	0·516	0·523	−0·314
C_3	0·427	0·495	−0·254
C_4	0·403	0·515	−0·082
C_5	0·389	0·586	−0·084
C_6	0·351	0·614	0·074
C_7	0·413	0·607	0·224
C_8	0·384	0·661	0·331
C_9	0·394	0·710	0·206
C_{10}	0·492	0·725	0·179
C_{11}	0·556	0·672	0·197
C_{12}	0·515	0·608	0·193
C_{13}	0·538	0·573	0·034
C_{14}	0·486	0·607	−0·102
C_{15}	0·485	0·511	0·041
C_{16}	0·350	0·514	−0·372
C_{17}	0·301	0·570	−0·345
C_{18}	0·323	0·604	−0·211
C_{19}	0·341	0·685	0·065
C_{20}	0·586	0·811	0·208
C_{21}	0·583	0·436	0·134
C_{22}	0·617	0·613	−0·316
C_{23}	0·624	0·680	−0·306
C_{24}	0·439	0·424	−0·266
C_{25}	0·383	0·335	−0·175
O_1	0·519	0·775	0·273
O_2	0·542	0·463	0·002
O_3	0·367	0·396	−0·187
O_4	0·334	0·485	−0·494
O_5	0·434	0·668	0·469
O_6	0·347	0·765	0·255
O_7	0·635	0·675	0·089
O_8	0·555	0·572	0·321
O_9	0·753	0·442	−0·192
O_{10}	0·618	0·362	0·459
O_{11}	0·728	0·466	0·480
I	0·166	0·209	0·171

The projections of the structure along the c and a axes are given in Fig. 1 and 2 respectively. Their explanatory diagrams, as well as the structural formula in Fig. 3 show the absolute configuration of (+)-demethanolaconinone as determined having made use of the anomalous dispersion of the Cu $K\alpha$ radiation by the iodine atom. (Przybylska & Marion, 1959*b*). The final atomic coordinates given in Table 1, in a right-handed system of axes, belong to the same molecular model.

Discussion

The molecule of (+)-demethanolaconinone has been found to have the same carbon–nitrogen skeleton as des-(oxymethylene)-lycoctonine (Przybylska, 1961). The functional groups of both compounds that are attached to the same carbon atoms of the skeleton are directed the same way with the exception of one methoxyl group attached to C_{15}. This group is equatorial in des-(oxymethylene)-lycoctonine and axial in demethanolaconinone.

The bond lengths and angles are given in Fig. 4. The average of C–C bonds excluding the $C_{17}=C_{18}$ double bond is 1·54 Å and the average of C–O bonds omitting the value for the $C_{16}=O_4$ bond is 1·41 Å. The standard deviations of the coordinates were cal-

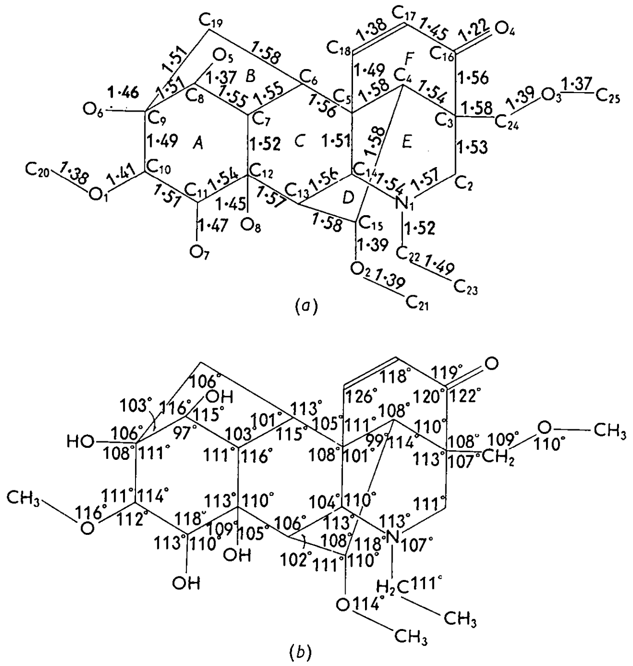


Fig. 4. (*a*) Intramolecular bond lengths, (*b*) angles.

culated using Cruickshank's formula (1949). The r.m.s. value for the light atoms other than hydrogen, multiplied by 1·66 to allow for the lack of a centre of symmetry came to 0·031 Å and for the iodine atom 0·002 Å. The e.s.d. for the light atom bonds is about 0·044 Å ($= \sqrt{(2)} \times 0{\cdot}031$). Using Cruickshank & Robertson's (1953) criterion for the 5% significance level, the deviations as high as ±0·09 Å ($1{\cdot}96 \times 0{\cdot}044$) in these bonds from their nominal values should not be taken as significant. None of the bonds exceed this limit. Only N_1–C_2 and C_{16}–C_{17} bonds deviate by this amount from the theoretical N–C and C–C values, whereas all others were found to lie within ±0·06 Å.

The standard deviation $\sigma(\theta)$ for several angles was calculated using the equation of Ahmed & Cruickshank (1953) and they averaged to about 2·2°. That means that differences as high as 6° ($\sqrt{(2)} \times 1{\cdot}96 \times 2{\cdot}2$) between a pair of observed angles should not be regarded as significant.

The only angles that are significantly smaller than the tetrahedral value can be found in five-membered rings. All the angles for these two rings vary from 97° to 108°, with the average of 102·4°. Significantly larger angles are at C_{16}, C_{17} and C_{18} of the ring *F*. These results are in good agreement with the position of the C=C bond and the C=O group. The $C_{17}=C_{18}$ bond is 1·38 Å long and $C_{16}=O_4$ is 1·22 Å and as the atoms forming these bonds were identified as carbon and oxygen, these lengths leave no doubt that they represent double bonds. Also in agreement with this conclusion, the C_{17} atom was found to be planar with C_3, C_{16}, C_{18} and C_5 atoms.

The angle C_{14}–N_1–C_{22}, found to be 118°, is considerably larger than the corresponding angle in des-(oxymethylene)-lycoctonine. It is possible that the position of the nitrogen atom is less accurately determined than most atoms and this is actually indicated by the length of N_1–C_2 bond (1·57 Å). On the other hand some increase in that angle from the tetrahedral value is not unexpected as the hydroxyl group attached to C_{11}, which is not present in des-(oxymethylene)-lycoctonine, is directed towards the ethyl group.

The C_{10}–C_{11}–C_{12} angle is also large (118°). This is in agreement with the distortion found in the ring *A*, which was found to be of a flattened boat form. The equation of the plane best fitting the C_7, C_9, C_{10} and C_{12} atoms is

$$0{\cdot}153x + 0{\cdot}072y + 0{\cdot}986z - 3{\cdot}723 = 0$$

and the distances of C_8 and C_{11} from that plane are +0·90 and +0·21 Å respectively. A similar distortion was found in the same ring in the skeleton of des-(oxymethylene)-lycoctonine. This was an expected result as the oxygen atoms of the functional groups attached to C_8, C_{10} and C_{12}, which are responsible for this flattening of the ring, are directed the same way in both compounds.

The rings *C* and *E* are in 'chair' form. The most interesting intramolecular distances are listed in Table 2.

Table 2. *Intramolecular distances*

	Intramolecular distances		Intramolecular distances
C_{23}–C_{14}	3·10 Å	O_1–O_5	3·13 Å
C_{23}–O_7	3·30	O_1–O_8	4·53
C_3–C_5	2·52	O_5–O_8	3·03
C_7–C_9	2·30	O_5–C_{11}	2·89
C_5–C_{13}	2·42	O_7–C_{14}	3·10
O_3–O_4	3·26		

The closest C · · · C approach is shown in Fig. 2. It takes place between the atoms C_{23} and C_{25} and is equal to 3·43 Å. This distance can be considered as normal, since the bonds C_{22}–C_{23} and O_3–C_{25} are almost parallel to each other and therefore the hydrogen atoms, which form tetrahedral angles with these bonds, do not lie along the line joining the C_{23} and C_{25} atoms. Six other approaches are lower than 4·0 Å and they range from 3·92 to 3·96 Å.

The shortest C · · · O intermolecular distances were those between O_{10} of the molecule of water of crystallization, C_{20} and C_{21} of neighbouring molecules. They were found to be 3·49 and 3·20 Å respectively. All other C · · · O approaches are greater than 3·5 Å.

The distances between $O_{10} \cdots O_6$, $O_{10} \cdots O_{11}$ and $O_9 \cdots O_{11}$ are about 2·82 Å, whereas the angles C_9–O_6–O_{10}, O_6–O_{10}–O_{11} and O_{10}–O_{11}–O_9 are 112, 124 and 89° respectively. These results indicate that the hydrogen bonds probably link all these oxygen atoms together.

Although close contact between the nitrogen atom and the iodine ion might be expected, the distance between these two atoms is over 5 Å. The iodine atom, however, is closely associated with five oxygen atoms, two of which belong to the water molecules. They were all found at distances varying from 3·52 to 3·62 Å, and are shown by dotted lines in Fig. 1.

I am very grateful to Dr Léo Marion for suggesting this problem for X-ray analysis, for his unfailing support and many discussions. I should like to thank Dr F. R. Ahmed for his invaluable assistance in computation and helpful advice and Dr W. H. Barnes for his continued interest in this investigation. Thanks are also due to Miss C. Saunderson and Miss C. Philo for their help with computing and preparing of the data. I am also indebted to the staff of the Computation Centre at the University of Ottawa and of Structures Laboratory, the Division of Mechanical Engineering, National Research Council for the use of the IBM 650 and FERUT computers.

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Remarks on a Non-Statistical Approach to the *F* Phase Problem

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The non-negativity criterion is that the volume $A(\varrho).d\varrho$, for which the expected ϱ lies in the interval $(\varrho, \varrho+d\varrho)$, shall vanish for $\varrho<0$. However, providing the overlap is negligible, the expected volume $A(\varrho)$ within one cell is known for all ϱ values. It is possible to set up general A criteria, since A can be studied for arbitrary phase angles. Some preliminary examples of the use of the A concept are given and the results compared with known results.

The first attempts to solve the phase problem—among which the Harker & Kasper (1948) and Sayre (1952) methods are outstanding—were non-statistical in nature. Since then, starting with Zachariasen (1952), interest has been focused more and more upon statistical methods (e.g., Klug (1958)).

A disadvantage of the statistical approach is that it often lacks physical perspicuity, and it is often difficult to visualize how much of the experimental and general information has really been used to advantage. The hypotheses are often difficult to assess and sometimes even to accept. For instance, of fundamental interest in most statistical attempts to solve the sign problem is the probability distribution $\mathscr{P}(E)$ (and analogous joint probability distributions). The functions $\mathscr{P}$ are usually derived on the assumption that the atomic parameters are random variables subject only to symmetry relations. However, the *a priori* probability of a certain value $\mathbf{r}_{(n)}$ may range from zero to unity; in fact, the probability of any one atomic-coordinate value is a function of all other coordinates (cf. e.g., Bertaut (1955)). However elaborate the assumptions made about interatomic vectors, *some* sort of *a priori* assumption must be made. In the following paper (Löfgren, 1961) it will be shown that these assumptions critically influence certain expressions that are therefore unpredictable.

Instead of *statistically* deriving the properties of