

## On a Family of OD-Structures of Uranyl Acetylacetonate

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Crystals of uranyl acetylacetonate obtained on crystallization show different diffraction diagrams. All of them can, however, be explained as originating from members of one and the same family of OD-structures. The OD-groupoid family is either

$$P \ 2 \ a \ (a) \qquad \qquad \qquad P \ m \ a \ (a)$$

$$\{2\frac{1}{2} \ n_{2,\frac{1}{2}} \ (a\frac{1}{2})\} \qquad \text{or} \qquad \qquad \qquad \{c_2 \ n_{2,\frac{1}{2}} \ (a\frac{1}{2})\}$$

with  $a = 16.6 \text{ \AA}$ ;  $b = 7.1 \text{ \AA}$ ;  $c_0 = 24.4 \text{ \AA}$ .

Approximate uranium coordinates were obtained from qualitative observations concerning the diffuse streaks. They are:

$$x_U = 0; y_U = 0.25; z_U = 0.052.$$

These are in accordance with the predominant maxima of the Patterson projections  $P(u,w)$  and  $P(u,v)$  for the monoclinic form.

Uranyl(IV) acetylacetonate was first prepared in 1904 by Biltz and Clinch<sup>1</sup> by mixing a solution of a uranyl salt with acetylacetonone and neutralizing the resulting solution with alkali or alkali carbonate. This compound has now been characterized as bis(acetylacetonato)dioxouranium(VI) hydrate  $\text{UO}_2\text{A}_2 \cdot \text{H}_2\text{O}$ , acetylacetonone being denoted by HA. A similar method has also been used by other authors.<sup>2-6</sup> From X-ray diffraction data for this compound Comyns *et al.*<sup>5</sup> concluded that it appears in several modifications, some of which are monoclinic and some orthorhombic. In a recent paper,<sup>7</sup> further X-ray diffraction data have been given for a monoclinic modification. Since several acetylacetonate complexes of tetravalent actinides and similar ions are under investigation at the Department of Inorganic Chemistry, University of Göteborg, a study of the crystal structure of uranyl acetylacetonate was considered to be of great interest.

## EXPERIMENTAL

*Preparation.* An aqueous solution of uranyl acetate was heated to 40°C and acetylacetone was added until the molar ratio U:HA was 1:4. The resulting solution was stirred rapidly for about an hour and was then allowed to stand and crystallize. After about 12 h a crystalline precipitate was collected on a filter, washed with cold water, and dried over silica gel in a desiccator.

Uranyl acetate was preferred to uranyl chloride or uranyl nitrate in the preparation, since the acetic acid formed during the reaction is a weak acid and does not appreciably alter the pH of the solution in contrast to HCl or HNO<sub>3</sub> which would have been formed had uranyl chloride or uranyl nitrate been used instead.

*Analysis.* The amount of uranium in the crystals was determined by heating a sample for about 30 min to approximately 800°C, the residue being weighed as U<sub>3</sub>O<sub>8</sub>. The amount of acetylacetone in the substance was determined spectrophotometrically using a method<sup>8</sup> described by Bonner and Thorne. A sample was dissolved in 0.05 M sulfuric acid and a solution containing iron(III) was added, whereupon the stable red compound FeA<sub>3</sub> was formed. The amount of FeA<sub>3</sub> was determined at 484 nm using a Zeiss PMQII photometer.

The amount of water was determined by drying a sample at 106°C in an oven. The results obtained are given in Table 1.

Table 1. Results of the chemical analysis.

	Calc. for UO <sub>2</sub> A <sub>2</sub> H <sub>2</sub> O	Found
% UO <sub>2</sub> <sup>2+</sup>	55.53	55.43
% C <sub>8</sub> H <sub>7</sub> O <sub>2</sub> <sup>-</sup> (=A)	40.76	41.02
% H <sub>2</sub> O	3.71	3.77
Sum	100.00	100.22

*X-Ray diffraction data.* Single crystals of about 0.015–0.050 mm in thickness were picked out, and rotation and Weissenberg photographs were taken of different crystals. Of some crystals extensive intensity data were collected using CuK $\alpha$  radiation and multiple film techniques. The sizes of these crystals were measured accurately so that a correction could be applied for absorption. The intensities of the reflections were estimated visually using a scale made from timed exposures of a strong spot in the Weissenberg photograph *h0l*.

## CRYSTAL DATA

The crystals investigated seem to belong to Form 1 described by Comyns, Gatehouse and Wait<sup>5</sup> since the lattice constants *a* and *b* for all crystals investigated agree within the limits of experimental error with those given by these authors. We have found some crystals which showed monoclinic symmetry and a geometry of the unit cell corresponding to form 1B,<sup>5</sup> and in some but not all of these crystals the reflections with *h* odd were joined by more or less continuous streaks parallel to  $\vec{c}^*$ . It would thus be more appropriate to call these "reflections" maxima on the streaks in contrast to the sharp reflections for *h* even. These continuous streaks are in disagreement with the

description given by the authors quoted <sup>5</sup> who stated that they observed streaks for  $hkl$  with  $l$  odd rather than  $h$  odd.

The positions and relative intensities of the sharp reflections with  $h$  even are the same for all crystals, as are the positions and the systematic absences of the diffuse streaks. The intensity distribution — including the positions of the maxima — along the diffuse streaks varies, however, from crystal to crystal; for all crystals investigated the positions of the maxima are such that they give integral indices referred to the large rectangular unit cell  $\vec{a} \vec{b} \vec{c}$  of the crystals named <sup>5</sup> 1A. For most crystals, even those with maxima which would give integral indices referred to the large rectangular cell but not to the small monoclinic cell, the intensity distribution of the maxima on the diffuse streaks shows monoclinic symmetry only. For some crystals, only the maxima corresponding to a  $B$ -face centred cell are present, but for other crystals this rule of systematic absences does not hold true.

Table 2. Lengths of reciprocal and real vectors characterizing the geometry of uranyl acetylacetonate.

$a^* = 0.0929$	$a = 16.6 \text{ \AA}$	$\alpha = \beta = \gamma = 90^\circ$
$b^* = 0.218$	$b = 7.1 \text{ \AA}$	
$c^* = 0.0316$	$c = 48.8 \text{ \AA}$	

NB: Whereas  $\vec{a}$  and  $\vec{b}$  correspond to translational vectors indicating periodicity, there is no periodicity corresponding to  $\vec{c}$ . This is the length of a vector corresponding to the reciprocal vector  $\vec{c}^*$  introduced to index the sharp reflections as well as the maxima on the streaks.

In spite of these variations from crystal to crystal, we may describe the distribution in reciprocal space for all these crystals with respect to reciprocal vectors  $\vec{a}^*$ ,  $\vec{b}^*$ ,  $\vec{c}^*$ , corresponding to the large rectangular cell (see Table 2 and Fig. 1) with intensities differing from zero for places in reciprocal space characterized by  $h$ ,  $k$ ,  $\xi'$  (where  $h$  and  $k$  are integral and  $\xi'$  may take any real value). The maxima on the streaks occur for integral values  $\xi' = l$ . The lengths of the vectors  $\vec{a}$ ,  $\vec{b}$ ,  $\vec{c}$  in direct space corresponding to these reciprocal vectors are also given in Table 2. The systematic absences valid for all crystals are listed in Table 3, together with the conditions for the maxima for three typical crystals denoted I, II, and III, and with observations concerning the symmetry of the observed intensities (see Fig. 1).

#### DETERMINATION OF SYMMETRY

In the following we proceed in a mode similar to that described earlier <sup>9</sup> in greater detail. The streaks parallel to  $\vec{c}^*$  show that the structures (at least of those crystals for which they have been observed) although periodic in the

Table 3. Summary of the observed distribution of intensity in reciprocal space.

- (i)  $(hk\xi')$  missing for  $h = 2\hat{h}$  with  $\xi' \neq l$  (no streaks visible)  
(ii)  $(\hat{h}kl)$  missing for  $l = 2n+1$  (only values  $l = 2\hat{l}$ )  
(iii)  $(\hat{h}k\hat{l})$  missing for  $\hat{h} + \hat{l} = 2n+1$   
(iv)  $(h0\xi')$  missing for  $h = 2n+1$  (no streaks for  $k = 0$ )  
(v)  $(hk0)$  missing for  $h = 2n+1$  (intensity of streaks vanishes when  $\xi'$  approaches 0)  
(vi)  $I(hk\xi') = I(\overline{h}\overline{k}\overline{\xi}')$   
(vii)  $I(\hat{h}k\hat{l}) = I(\overline{\hat{h}}\overline{k}\overline{\hat{l}})$

In general:

- (viii)  $I(hkl) \neq I(\overline{h}\overline{k}\overline{l})$  for  $h = 2n+1$   
(ix) Relative intensities  $I(\hat{h}k\hat{l})$  the same for different crystals.  
(x) Relative intensities  $I(hk\xi')$  for  $h$  odd vary from crystal to crystal.  
(xi) Maxima on diffuse streaks are found:  
for crystal I at  $h-l = 4n$   
for crystal II at  $h-l = 4n$  and at  $h+l = 4n$  } or at  $l = 2n+1$   
for crystal III at  $h+l = 4n$  and at  $l = 2n$  }

NB: The unit of  $\xi'$  has been chosen empirically as  $\vec{c}^*$ , so as to give integral indices for the sharp reflections and the maxima on the streaks. The  $\xi'$ -values deduced theoretically from the OD-groupoid family refer to a unit  $\vec{c}_m^* = 4\vec{c}^*$  which is reciprocal to  $\vec{c}_0$ .

$\vec{a}$  and  $\vec{b}$  directions are not periodic in the  $\vec{c}$  direction. The rule of absence (i) (cf. Table 3) may be interpreted in terms of a superposition structure with an electron density  $\hat{\rho}(xyz)$  related to the real structure (electron density  $\rho(xyz)$ ) in the following way:

$$\hat{\rho}(xyz) = \frac{1}{2}(\rho(x,y,z) + \rho(x + \frac{1}{2},y,z)) \quad (1)$$

Rule (ii) in Table 3 means that the superposition structure is periodic in three dimensions with lattice constants

$$\hat{a} = a/2; \quad \hat{b} = b; \quad \hat{c} = c/2$$

and rule (iii) shows that it is *B*-face centred.

The rules of absence (iv) and (v) indicate the presence of *a*-glide planes in the structure, perpendicular to  $\vec{b}$  and  $\vec{c}$ , respectively. A glide plane perpendicular to  $\vec{c}$ , which would hold good for the whole structure, would, however, result in a corresponding symmetry of the intensity distribution which we found to be violated for many crystals. These *a*-glide planes must therefore be at different *z*-positions, each holding true only for part of the structure.

Whether there exists a total *a*-glide plane perpendicular to  $\vec{b}$  or whether, even

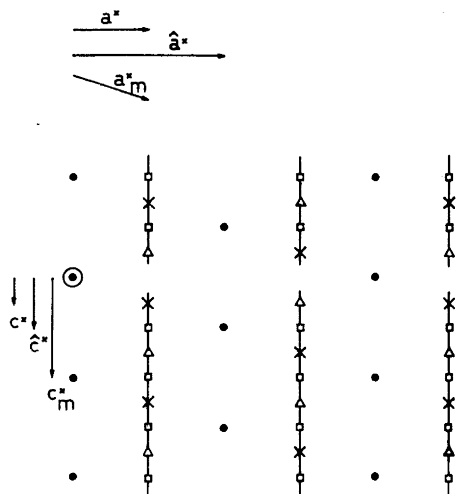


Fig. 1. Intensity distribution in reciprocal space. ● = sharp reflections. | = diffuse streaks present only for  $k \neq 0$ . Maxima on the diffuse streaks present in crystal I are denoted by ×, in crystal II by × Δ, in crystal III by Δ □.

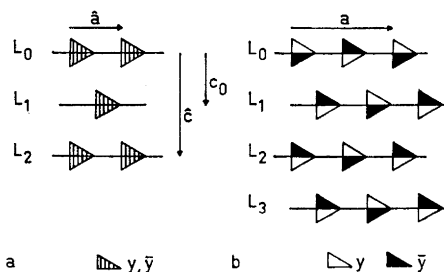


Fig. 2. Schematic drawing showing the symmetry of a) superposition structure and b) part of a real disordered structure. The sequence  $L_0 L_1 L_2$  corresponds to the sequence in crystal I of Table 2, the sequence  $L_1 L_2 L_3$  corresponds to the orthorhombic MDO-structure described in the text.  $\hat{c} = c/2$  and  $c_0 = c/4$ .

here, there exist partial glide planes, each holding for part of the structure only, cannot be decided at the present stage. The existence of a total  $a$ -glide plane would lead to monoclinic (or higher) symmetry of all crystals irrespective of the disorder or type of crystal; this has actually been observed in all crystals under investigation up to now.

Owing to the halving of the translation in the  $\vec{a}$  direction in the superposition structure, these glide planes (for any of the real structures) result in corresponding mirror planes in the superposition structure. Thus the minimum symmetry of the superposition structure is  $B2mm$ , which is in keeping with (vi) and (vii) (see Table 3 and Fig. 2); according to (ix), this superposition structure is the same for the different structures. We may consider the superposition structure to be built up of layers with periods  $\hat{a}$  and  $\hat{b}$ , each with the plane space group  $*P2m(m)$ , stacked in such a way as to produce the  $B$ -face centring. To each of these layers corresponds in a real structure a layer with plane space group  $P2a(a)$ . For each layer there exist two positions in keeping with the corresponding layer of the superposition structure, related by a shift of  $a/2 = \hat{a}$ . The crystals giving different intensity distributions for  $h$  odd (according to (x)) thus differ in the stacking of their layers. The lack of orthorhombic symmetry (viii) results from the partial

\* For nomenclature of plane space groups see Ref. 9–11 or 12.

character of the rotation diad parallel to  $\vec{a}$  and the  $a$ -glide perpendicular to  $\vec{c}$ . Thus a minimum symmetry corresponding to the schematic drawing in Fig. 2b results.

All pairs of successive layers are geometrically equivalent. These structures are therefore OD-structures and members of one and the same family of OD-structures.<sup>9,11,12</sup> The common features of their minimum symmetry are described by the OD-groupoid family

$$P \quad 2 \quad a \quad (a) \\ \{2_{\frac{1}{2}} \quad n_{2, \frac{1}{2}} \quad (a_{\frac{1}{2}})\}$$

It would not be possible to detect an additional mirror plane in the single layer perpendicular to  $\vec{a}$  and the resulting centre of symmetry by means of systematic absences or by a change in the symmetry of the intensities. There is, thus, still another OD-groupoid family in keeping with the observations listed in Table 3. Its symbol is

$$P \quad m \quad a \quad (a) \\ \{c_2 \quad n_{2, \frac{1}{2}} \quad (a_{\frac{1}{2}})\}$$

The volume occupied by a periodically repeating unit of one layer is  $abc_0$ , where  $c_0$  is the distance between the  $a$ -glide planes (perpendicular to  $\vec{c}$ ) of consecutive layers (see Fig. 2). As indicated by the density of the crystals, such a unit contains 4 formula units. The OD-groupoid family (of lower symmetry) contains 4 asymmetric units per repeat unit of the layer, whereas the family of higher symmetry contains 8 asymmetric units per repeat unit.

If every layer  $L_{p+1}$  of a structure may be obtained from  $L_p$  by a translation  $\vec{c}_m = \vec{c}_0 - \vec{a}/4$ , not only all pairs but also all triples of consecutive layers are geometrically equivalent (and the same holds for quadruples and higher  $n$ -tuples). Thus such a structure is a structure of maximum degree of order (MDO-structure, for short).<sup>9,11,12</sup> It is, of course, a periodic structure with reflections corresponding to the sharp reflections and the positions marked by crosses in Fig. 1. A twin to such a structure with translations  $\vec{c}_m' = \vec{c}_0 + \vec{a}/4$  would have reflections at the places marked by triangles instead of those marked by crosses. An essentially different MDO-structure results if the translations  $\vec{c}_m$  and  $\vec{c}_m'$  alternate. This structure is orthorhombic with periodicity  $2\vec{c}_0$ . Its reflections correspond to the places marked by squares in Fig. 1, as well as to the positions of the sharp reflections. Thus, we may conclude that all the maxima on the diffuse streaks so far observed are due to extended regions within the possibly disordered crystal, corresponding to one or the other of the MDO-structures described and that the crystals not showing any diffuse streaks are periodic MDO-structures or consist of extended MDO-regions.

## FOURIER TRANSFORM FOR THE OD-GROUPOID FAMILY

We shall start by considering the case of lower symmetry with one formula unit per asymmetric unit.

If not only the OD-groupoid family of the structure but also the exact stacking of the layers were known, then we could deduce the coordinates of the atoms related by symmetry to an atom at  $xyz$ . From these we could then — according to first principles — deduce a general formula for the Fourier transform in terms of the atomic scattering factors  $f_j$  and the coordinates  $x_j, y_j, z_j$  of the atoms within an asymmetric unit, similar to the structure factor formulae originally deduced by Lonsdale<sup>13</sup> and now in common use as part of *International Tables*, Vol. I.<sup>14</sup>

Because the exact stacking is not known and because we want the formula to be valid for crystals with different stacking sequences, we shall introduce parameters which characterize the stacking. The equipoints of a single layer with plane space group  $P2a(a)$  are

$$[L_0] = [xyz; x\bar{y}\bar{z}; \frac{1}{2} + x, \bar{y}, z; \frac{1}{2} + x, y, \bar{z}] \quad (2)$$

where  $x$  and  $y$  stand for  $x+m_1$  and  $y+m_2$ , respectively ( $m_i$  equal 0, 1, 2, ...), whereas  $z$  stands for  $z$  only, and is referred to the vector  $\vec{c}_0$  (see above). The geometrical part  $\varphi_0(h, k, \xi)$  of its Fourier transform is, accordingly

$$\varphi_0(h, k, \xi) = 4e^{2\pi i h x} \cos 2\pi k y \cos 2\pi \xi z \text{ for } h \text{ even} \quad (3a)$$

$$\varphi_0(h, k, \xi) = 4e^{2\pi i h x} \sin 2\pi k y \sin 2\pi \xi z \text{ for } h \text{ odd} \quad (3b)$$

where (as in the following) the unit  $\vec{c}_0^*$  of  $\xi$  is taken as the reciprocal of  $\vec{c}_0$ . The points of an even numbered layer  $L_{2q}$  related by symmetry to the point  $(xyz)$  of layer  $L_0$  have coordinates

$$[L_{2q}] = [L_0] + (\alpha_{2q}/2, 0, 2q) \quad (4)$$

those of the odd-numbered layers  $L_{2q+1}$

$$[L_{2q+1}] = [L_0] + (1/4 + \alpha_{2q+1}/2, 0, 2q + 1) \quad (5)$$

with  $\alpha_p = 0$  or 1. Thus the geometrical parts of the Fourier transforms are

$$\varphi_{2q}(hk\xi) = (-1)^{\alpha_{2q}h} e^{2\pi i 2q\xi} \varphi_0(hk\xi) \quad (6)$$

and

$$\varphi_{2q+1}(hk\xi) = i^h (-1)^{\alpha_{2q+1}h} e^{2\pi i (2q+1)\xi} \varphi_0(hk\xi) \quad (7)$$

respectively.

The Fourier transformation of the whole structure is thus

$$F(hk\xi) = S(h\xi)F_0(hk\xi) \quad (8)$$

with

$$F_0(hk\xi) = \sum_i f_i \varphi_0^{(i)}(hk\xi) \quad (9)$$

and

$$S(h\xi) = \sum_q (-1)^{\alpha_{2q}h} e^{2\pi i 2q\xi} + i^h (-1)^{\alpha_{2q+1}h} e^{2\pi i (2q+1)\xi} \quad (10)$$

$|S(h\xi)|^2$  is periodic:

$$|S(h, \xi)|^2 = |S(h + 4h' + 2v, \xi + l' + v/2)|^2 \quad (11)$$

where  $h'$ ,  $l'$ ,  $v$  may take any integral values.

#### DETERMINATION OF THE POSITION OF THE URANIUM ATOM

As a first approximation we may assume that the contributions from the other atoms to the structure factors may be neglected in comparison with the contribution from the uranium atoms. Thus from eqns. (9) and (3) it follows that

$$|F_0(hk\xi)|^2 \approx f_U^2 \cos^2 2\pi ky_U \cos^2 2\pi \xi z_U \quad \text{for } h \text{ even} \quad (12a)$$

and

$$|F_0(hk\xi)|^2 \approx f_U^2 \sin^2 2\pi ky_U \sin^2 2\pi \xi z_U \quad \text{for } h \text{ odd} \quad (12b)$$

Thus the intensities for  $h$  odd, *i.e.* on the diffuse streaks, ought to vanish or to be very small at  $\xi$ -values for which  $\xi \cdot z_U$  is an integral multiple of  $1/2$ . These points on the streaks may easily be detected. Confusion with points at which  $|S(h\xi)|^2$  is small or vanishing may be excluded because of the known periodicity of  $|S(h\xi)|^2$  along  $\xi$  [see eqn. (11)] and because they are the same for different crystals with different values of  $|S(h\xi)|^2$ . Actually, such small intensities occur for  $\xi$ -values of about 10, 19, or 20 or an integral multiple of about 9.7, so we may conclude that  $z_U = 0.052$  (referred to  $c$ ). If this is correct, then sharp reflections should be weak or missing when  $l$  is an odd multiple of 4.8 or  $l$  an odd multiple of about 2.4. Actually rather weak reflections occur for  $l = 2, 3, 7, \text{ and } 12$ . These values are near to odd multiples of 2.4.

An approximate value of  $y_U$  may be deduced from the fact that the sharp reflections ( $h$  even) are extremely weak on the ( $h1\xi$ )-Weissenberg diagrams and the diffuse maxima ( $h$  odd) are very strong, whereas the opposite is true for the ( $h2\xi$ ) diagrams. Thus  $y_U$  must be very near  $\frac{1}{4}$ . The origin of the single layer may be conveniently chosen so that  $x_U = 0$ . The approximate position of the uranium atom is thus (0, 0.25, 0.052).

According to eqn. (2), Patterson peaks due to the U—U-vectors are to be expected at the following coordinates:

$$0 \ 2y_U \ 2z_U; \quad \frac{1}{2} \ 0 \ 2z_U; \quad \frac{1}{2} \ 2z_U \ 0$$

$$0 \ 0.5 \ 0.104; \quad \frac{1}{2} \ 0 \ 0.104; \quad \frac{1}{2} \ 0.5 \ 0$$

(plus those related to them by orthorhombic symmetry). These positions may be compared with the Patterson projections  $P(u, w)$  and  $P(u, v)$  of the monoclinic crystal (Figs. 3 and 4) obtained before the OD-symmetry of these structures had been discussed. In comparing them with the Patterson positions just obtained, we have to keep in mind that the latter refer to the large rectangular cell with  $c = 4c_0$ , and that  $P(u, v)$  is a projection along the monoclinic  $c$ -axis. To facilitate an understanding of the projection  $P(u, v)$ , the monoclinic cell and half the large rectangular cell have been outlined in Fig. 3.



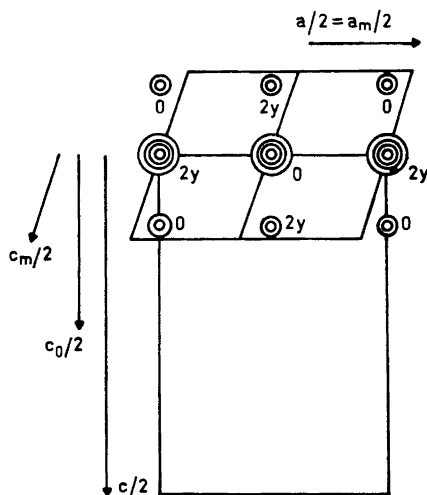


Fig. 3. Patterson projection  $P(u,w)$  of the monoclinic form with the monoclinic cell and half the rectangular "cell" outlined. The  $v$ -values at which the Patterson peaks are to be expected in space are indicated. (Origin at the centre.)

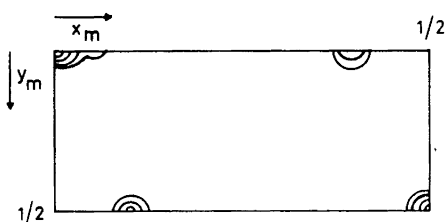


Fig. 4. Patterson projection  $P(u,v)$  of the monoclinic form.

So far we have discussed the uranium positions for the OD-groupoid family of lower symmetry only. The argument holds equally well for the family of higher symmetry, but these positions are now special positions on the mirror plane. It is not unlikely that the whole structure also complies with this higher symmetry and that the rest of the molecules occupy the general positions of this family.

The variation of the intensity with varying  $h$ , which is clearly visible for the weak reflections and maxima, is evidently due to the contributions from the light atoms. This makes us hope that we shall be able to obtain their positions as well. Work is being continued in this direction.

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The Patterson calculations were carried out on the SAAB D21 computer at the University of Göteborg Computer Centre.

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