THE CRYSTAL STRUCTURE OF DARAPSKITE

by

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Abstract

The space group of darapskite was determined to be $P2_1/m$, with the cell constants $a = 10.56$ Å, $b = 6.92$ Å $c = 5.19$ Å, and $\beta = 102.8°$ (monoclinic, 2nd setting). Each cell contains $2(Na_3NO_3SO_4 \cdot H_2O)$.

Integrated-intensity measurements were taken using a single-crystal diffractometer with Ni-filtered $CuK\alpha$ radiation. These data were corrected for absorption and Lorentz-polarization, and the resultant $|F_{hkl}|^2$ values were used to compute a three-dimensional Patterson synthesis. The sulfur inversion peak on the Harker section was identified using a superposition method and further confirmed by its occurrence with satellite peaks. Based on sulfur as a heavy atom, images in the Patterson synthesis revealed the locations of all other atoms except H_2O. A Fourier synthesis, computed with the known information, indicated the location of the H_2O molecule. The structure so determined was refined by the method of least-squares to $R = 8.8\%$.

The structure of darapskite is essentially an arrangement of atoms on two symmetry planes which are related to each other by the operation of a 2_1 screw axis. Located in the interstices between the planes and linking them together are Na^+ cations in octahedral co-ordination.

Table of Contents

List of Tables

List of Figures

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With special thanks, the author would also like to acknowledge her entire family, who helped her during vacation with the task of transferring numbers onto Patterson maps.

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I. INTRODUCTION

A. Previous work on darapskite

The mineral darapskite was discovered and first described by Dr. August Dietze, a chemist in Taltal, Chile. His chemical analysis, performed on material from the original locality in the Atacama Desert, showed that it is a hydrated double salt with the formula $Na_3NO_3SO_4 \cdot H_2O$. From that time to the present, darapskite is the only known, naturally-occurring combination of a sulfate with a nitrate.

Several years after Dietze's discovery, the new mineral was described in detail by A. Osann, a mineralogist in München, Germany. His investigation led to the conclusion that darapskite is monoclinic, belonging to crystal class 2/m. In physical appearance the crystals are colorless and transparent and have a habit which is usually tabular with the pinacoidal form (100) dominating. Because of the nearly square shape of this form, the crystals tend to be pseudo-tetragonal in appearance. A typical darapskite crystal showing these properties can be seen in Fig.1.

Perhaps the most interesting contribution to the understanding of this mineral which Osann made back in 1894 was his description of its twinning. According to him, "very often" crystals of darapskite are twinned, with (100) as the composition plane. Furthermore, the twinning repeats itself polysynthetically with the individual lamellae tending to paper-thinness. As can be expected, the crystals exhibit parting parallel to $\{100\}$. Fig.2

shows the twinning arrangement of two adjacent lamellae of darapskite in a cross-section perpendicular to the twin plane.

During the years that followed these preliminary investigations, relatively little work was done on naturally-occurring darapskite. A French chemist by the name of deSchulten discovered, however, that the spontaneous evaporation of a solution of sodium nitrate and sodium sulfate produces salt crystals which are identical with the mineral found in Chile. The chemical analysis of these crystals is shown in Table 1. Like the natural mineral, artificial darapskite crystallizes in tiny monoclinic tablets with $\{100\}$ as the dominant form.

Later, additional work done on the system Na_3NO_3 - Na_2SO_4 - H_2O at Yale University further established the equivalence of the double-salt crystals with darapskite as well as showing that an alleged mineral, nitroglauberite, does not exist but is a mixture of sodium nitrate and darapskite.

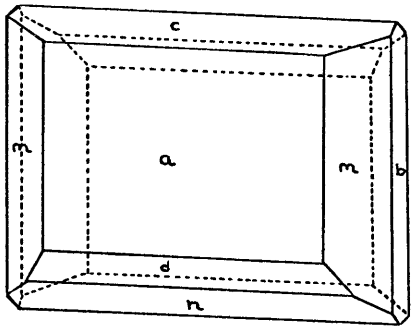


Fig. 1 [17]

Typical Darapskite Habit

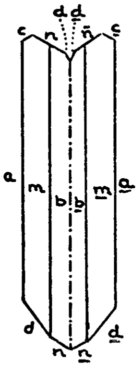


Fig. 2 [17]

Two adjacent twin lamellae.

Cross-section perpendicular to twin plane.

TABLE 1

Chemical analyses of darapskite

	Predicted for $Na_3NO_3SO_4 \cdot H_2O$	Material from Oficina Lautaro, Chile	Artificial material
		analyst: Dietze	analyst: de Schulten
Na_2O	37.94 %	38.27 %	37.96 %
N_2O_5	22.04	22.26	22.23
SO_3	32.67	32.88	32.45
H_2O	7.35	7.30	7.34
Total	100.00	100.71	99.98
Specific gravity		2.203	2.197

B. Occurrence

Although darapskite forms under a wide range of solubility conditions, its occurrence must obviously be greatly restricted by natural environment since it is a water-soluble mineral. The only place where darapskite can be found in considerable quantities is in the Atacama Desert in northern Chile, where it occurs in association with the famous Chilean nitrate deposits. The deposits are situated in interior dry valleys between the Coast Range and the Andes, at elevations ranging from 1,000 to 3,000 feet. Massive nitrate rock and superficial beds of caliche generally comprise the gentle slopes of the valleys, and the darapskite often occurs in cavities or crevices or as a surficial concentration here. Associated with darapskite are soda-niter, bloedite, halite, anhydrite, and kroehnkite.

In addition, darapskite associated with soda-niter and niter has been reported from Death Valley, San Bernadino County, California.

II. SELECTION OF SPECIMEN

The structure determination of darapskite began with the selection of a specimen suitable for analysis by x-ray techniques. For this purpose literally hundreds of crystals, both natural and artificial, were examined under the microscope, but all were proven to be unsatisfactory. The reason for this is that each little darapskite tablet, instead of being a single crystal, is in reality a composite of numerous twin individuals arranged in lamellar fashion. Thus a typical crystal, which is tabular parallel to (100) and also twinned on (100) resembles a little book with its pages analogous to polysynthetic twin lamellae.

That a twinned crystal is unsatisfactory for crystal-structure analysis is a well-established fact. The basic difficulty with twins is that their reciprocal lattices interpenetrate each other, and thus the symmetry and diffraction intensity information contained in an individual reciprocal lattice is likely to be masked. In the case of darapskite, which has point-group symmetry $2/\underline{m}$ and is twinned on (100), the two-fold axes of each twin have a common direction and their mirrors are parallel. However, the operation of the twin law augments the simple $2/\underline{m}$ symmetry of a twin individual by effectively introducing an extra mirror parallel to the two-fold axes. (See Fig.3) Thus, the twin composite of darapskite has orthorhombic symmetry $2/\underline{m}2/\underline{m}2/\underline{m}$.

Moreover, since the twinned crystal has its $\underline{b}$

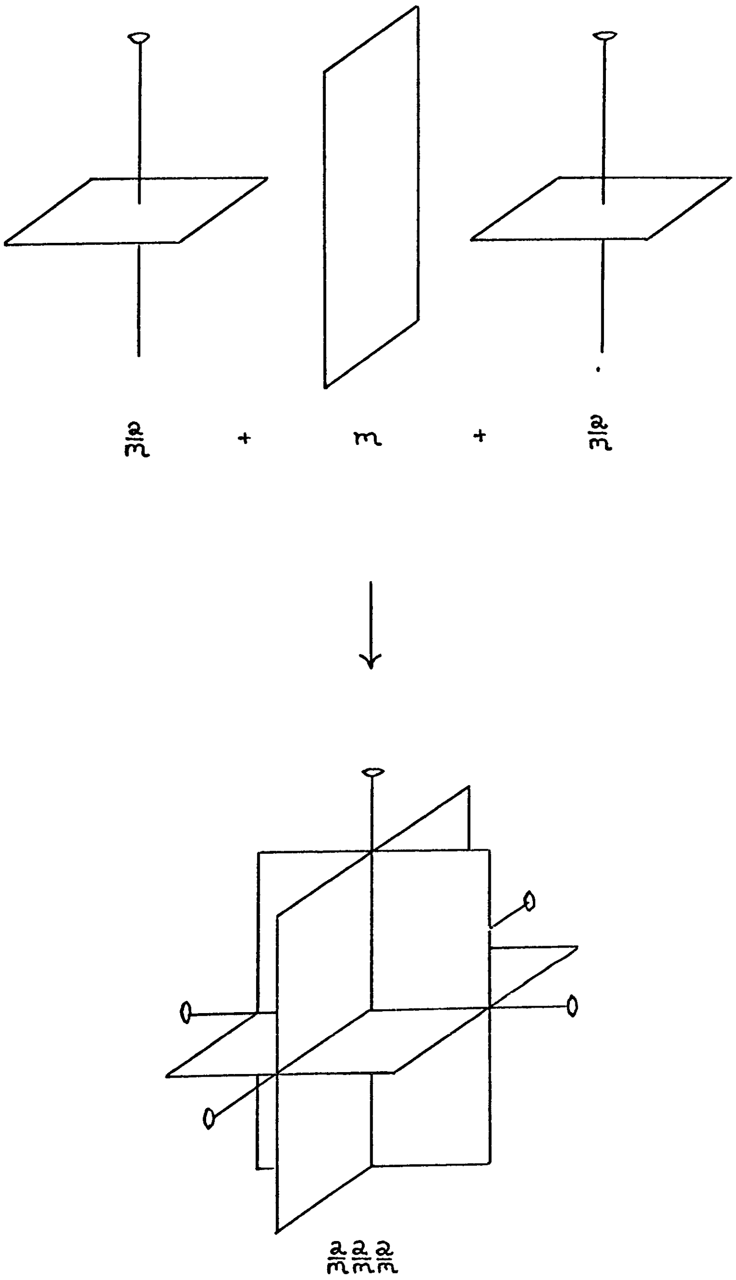


Fig. 3
SYMMETRY OF TWIN COMPOSITE

(2-fold) axes and (100) planes in common orientation, the $\underline{b}^*$ and $\underline{a}^*$ axes of the individual interpenetrating reciprocal lattices will lie in common directions. (See Fig.4.) Thus, the reciprocal-lattice points lying along these axes represent planes from both twins and the diffracted intensities corresponding to these points arise from simultaneous diffraction in both twins. The collection of $|F_{hkl}|$'s obtained from the x-ray investigation of a twinned crystal obviously conveys fictitious information about the structure of the single crystal, and therefore can not be tolerated for structure determination.

In consideration of these drawbacks and the lack of success in finding a single crystal of darapskite, it became necessary to use a non-conventional method to obtain an acceptable specimen. During the course of routine optical examinations, a fragment of an artificially-grown darapskite crystal was found which had several fairly wide twin lamellae. It seemed worthwhile to make use of the unusual width of these twins by endeavoring to separate them with a razor blade. Several smaller fragments were thus obtained, and one of them in particular was singled out as being a possible twin individual. After this fragment was mounted on a pyrex fiber with VYHH (polyvinyl chloride) glue and a precession orientation photograph was taken with $\underline{b}$ as the precessing axis, it seemed apparent that this crystal was indeed single.

In Fig.5 are shown two different zero-level precession photographs — one of the single crystal fragment and the other of a darapskite twin. For each, the unique axis (b) was the axis of precession, and thus the h0l reflections of the reciprocal lattice were recorded on the film. It is evident that for the single crystal, the pattern of these reflections shows typical monoclinic 2-fold symmetry; whereas for the twinned crystal the symmetry is 2mm, indicating orthorhombic. Therefore, by the straightforward comparison of these two zero-level reciprocal-lattice photographs, the authenticity of the single crystal was established.

The particular darapskite fragment which was selected for x-ray analysis was a rectangular prism with dimensions

.09mm X .015mm X .0075mm .

Its longest dimension was parellel to the c axis, and this was the direction along which the crystal was mounted to the goniometer head.

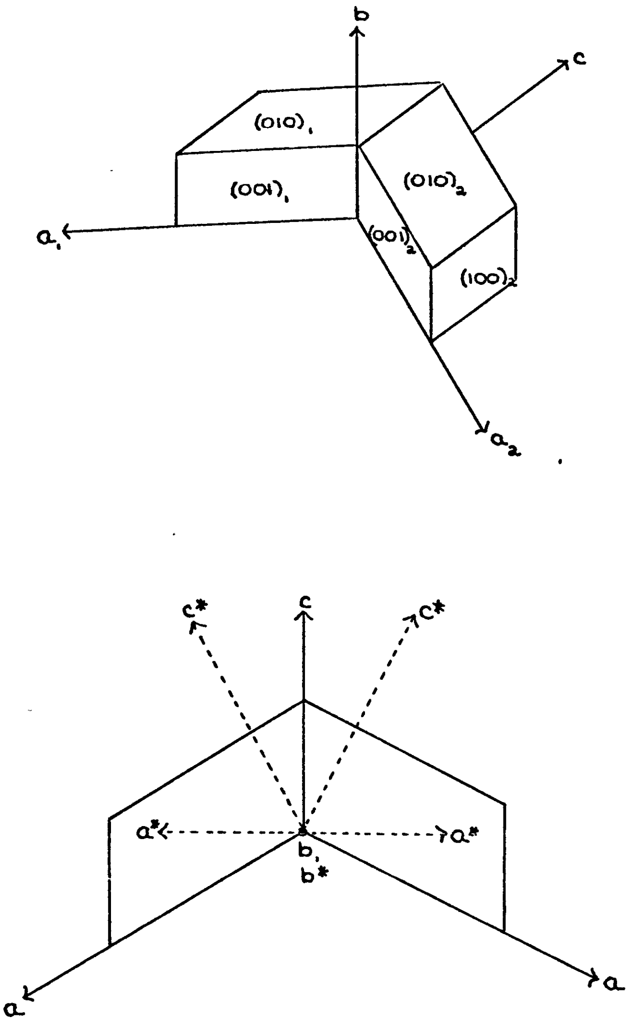
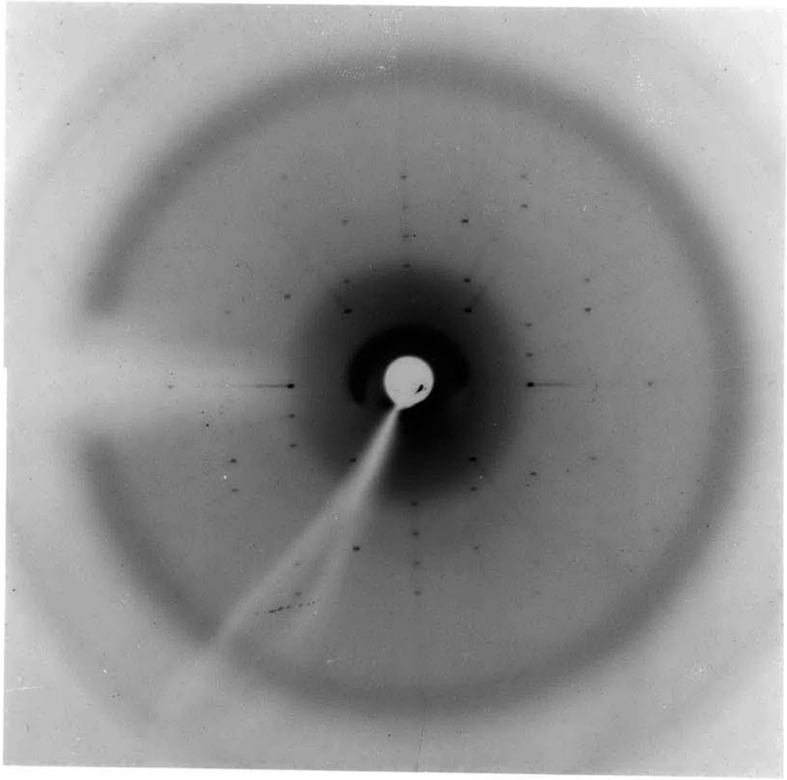


FIG. 4

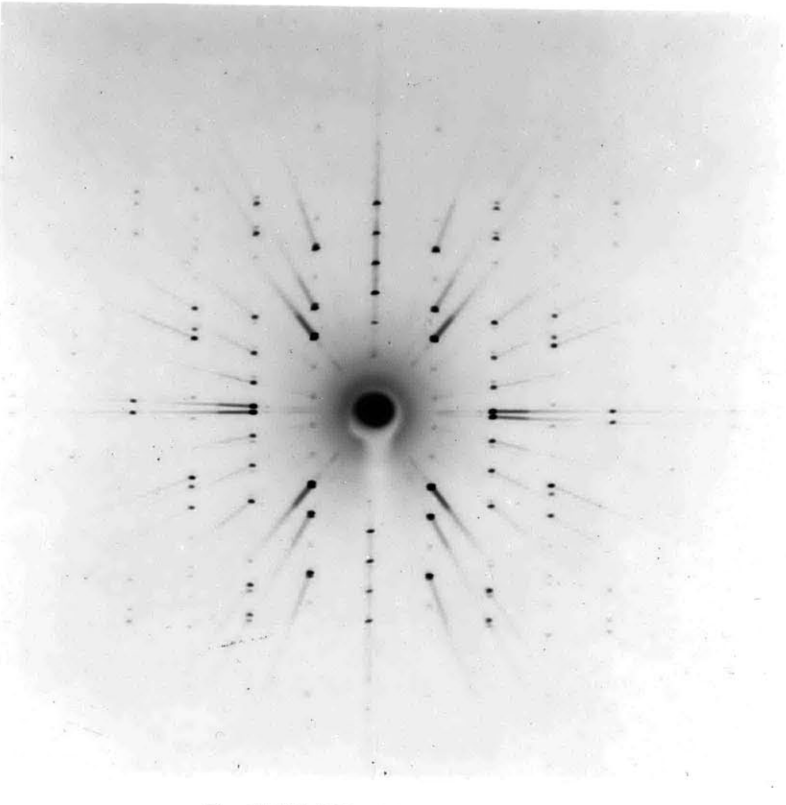
MONOCLINIC UNIT CELL TWINNED ON (100)

FIG. 5

DARAPSKITE ~ b axis ZERO-LEVEL PRECESSION PHOTOGRAPHS



A. SINGLE CRYSTAL



B. TWINNED CRYSTAL

III. SPACE - GROUP DETERMINATION

The initial steps of space-group interpretation necessarily include the determination of Friedel symmetry and lattice type, as well as the detection of glide planes and screw axes isogonal with the symmetry elements in the Friedel symmetry. To this end, the goniometer head holding the single crystal of darapskite was attached to a precession camera and x-ray photographs were taken using Mo$\underline{K}\alpha$ radiation. With the crystal in orientation such that its c axis was parallel to the dial axis of the camera and its b axis precessing around the direct x-ray beam, a series of photographs were taken representing the various reciprocal-lattice levels perpendicular to b. All of these photographs displayed 2-fold symmetry characteristic of the unique axis of monoclinic crystals. Furthermore, since x-ray diffraction effects are centrosymmetrical according to Friedel's Law, the Friedel symmetry of darapskite was derived by combining the 2-fold axis with an inversion center. Thus the Friedel symmetry was determined to be 2/m.

When the photographs of the different levels were superimposed with their origins and reciprocal axes (a^* and c^*) coinciding, the reflections for the levels fell right on top of one another. In this way it was shown that the reciprocal lattice of darapskite is primitive, and therefore the direct lattice is also primitive.

The remaining information which could be obtained from precession photographs, concerning the space group of darapskite, had to do with the detection of symmetry elements containing translation components. Since the zero-level b-axis photograph displayed no systematic extinctions in its 2-fold arrangement of reflections, it was inferred that there is no glide plane present which is isogonal with the mirror of the Friedel symmetry.

However, when the crystal was turned such that its a axis was the axis of precession, the resultant zero-level photograph exhibited a pattern of missing points along the b^* axis. Only the (0k0) reflections with $k = 2n$ were non-extinct. Therefore it was shown that there is definitely a 2_1 screw axis isogonal with the 2-fold axis of the Friedel symmetry.

In summarizing these results, it can be said that the diffraction symbol of darapskite is $2/mP2_1/-$. The possible space groups with which this is consistent are $P2_1$ and $P2_1/m$. However, dissolution experiments with etching solvents, which were performed by earlier investigators[17], have indicated that the point group is $2/m$. Thus it can be concluded that there is a mirror perpendicular to the screw axis, and the space group of darapskite is $P2_1/m$.

IV. UNIT-CELL DETERMINATION

A. Lattice constants

For the purpose of lattice constant refinement, two back-reflection Weissenberg photographs were taken using twinned darapskite crystals in b axis and c axis rotation. (The fact that the crystals were twinned would have no effect on the lattice constants.) Cu$K\alpha$ radiation was used and the resolved $\alpha_1 - \alpha_2$ doublets for some of the hkl reflections on each film were indexed. The vertical component of the distance between each indexed reflection and its equivalent on the other side of the center line of the photograph was then measured using a millimeter scale and vernier. From this film distance, F, the spacing between the planes in the crystal which gave rise to the reflection was easily calculated according to the following relationship[6] :

$$d_{hkl} = \frac{(n/2)\lambda}{\cos(F_{hkl}/4)} \qquad (1)$$

In addition, for a monoclinic crystal the interplanar spacing is expressed by

$$\frac{1}{d^2_{hkl}} = h^2 a^{*2} + k^2 b^{*2} + l^2 c^{*2} + 2hla^{*}c^{*}\cos\beta^{*} \quad (2)$$

where a*, b*, c*, and β^* are the lattice constants of the reciprocal cell. Thus, for each measured reflection an observational equation like (2) was set up in which the reciprocal-lattice constants were unknown quantities. The complete set of resultant simultaneous equations was then treated by a least-squares procedure in a computer program LCLSQ, written by Charles W. Burnham, which minimized the sums of the squares of the errors in the observable quantity d_{hkl}.

After several refinement cycles, the reciprocal and direct lattice constants of darapskite were determined to be the values shown in Table 2.

TABLE 2

Lattice constants of darapskite

Reciprocal

a^*	.09708	±	.00003 Å^{-1}
b^*	.14451	±	.00007 Å^{-1}
c^*	.19767	±	.00004 Å^{-1}
β^*	77.19	±	.02°

Direct

a	10.564	±	.004 Å
b	6.920	±	.003 Å
c	5.188	±	.001 Å
β	102.81	±	.02°

Unit cell volume = 369.8 ± .4 Å^3

B. Cell content

The mass contained in one unit cell of a crystal is given by $V\rho$, where V is the cell volume and ρ is the density of the substance. The mass can also be expressed by $(M/N)Z$ where M is the gram atomic weight of one formula unit, N is Avogadro's number, and Z is the number of formula units in one cell. Thus,

$$Z = \frac{V\rho N}{M} \qquad (3)$$

For darapskite,

$V = 369.8 \times 10^{-24}$ cm^3

$\rho = 2.20$ gm/cm^3

$N = 6.023 \times 10^{23}$

$$M\,(Na_3NO_3SO_4 \cdot H_2O) = 3(22.997) + 14.008 + 8(16.0) + 32.066 + 2(1.008) = 245.081 \text{ gm.}$$

If these values are substituted into (3), the result is $Z = 1.999 \simeq 2$. Therefore, there are two formula units of $Na_3NO_3SO_4 \cdot H_2O$ per unit cell of darapskite.

V. INTENSITY MEASUREMENTS

It is known that the intensity of the diffracted x-ray beam arising from a particular reciprocal lattice point hkl is proportional to the square of the structure factor, $|F_{hkl}|^2$, and that the structure factor is in turn a function of the various kinds of atoms and their positions in the unit cell. Because of this relationship, the collection of all diffracted intensities arising from a spherical region of the reciprocal lattice conveys valuable information pertaining to the structure of the crystal.

A. Experimental procedure

The intensities from darapskite were measured using an equi-inclination diffractometer with nickel-filtered, CuKα radiation. For each hkl reflection, the crystal setting ϕ and the scintillation counter setting Υ were computed by the program DFSET 4, written by Charles T. Prewitt. With the c axis of the darapskite single crystal as rotation axis, the intensity of each reflection with $l \geq 0$ lying within the CuKα sphere was measured. Thus, one hemisphere or two asymmetric Friedel units of reflection intensities were measured.

Throughout the data collection, the voltage on the x-ray tube was set at 35 KV and the current at 15 mA. The detector voltage and the pulse-height analyzer were adjusted to appropriate settings as described in the "Norelco Electronic Circuit Panel Instruction Manual" published by Philips Electronic Instruments.

B. Computation of intensities and data reduction

The integrated intensities were computed from the raw diffractometer measurements by means of the program FINTE 2 [14], which subtracted the average background reading for each peak and corrected for the counting interval used.

The integrated intensity I (which is the total diffracted energy received by the counter while the crystal is rotated through the Bragg position) is related to the structure factor F_{hkl} by

$$I_{hkl} = K\left|F_{hkl}\right|^2 (Lp)_{hkl}(T)_{hkl}. \qquad (4)$$

In this relationship Lp is the Lorentz-polarization factor, T is a transmission-correction term, and K is a constant for all reflections. The absolute value of each individual structure factor $\left|F_{hkl}\right|$ was thus computed by the program GAMP [15] and its subroutine GNABS [7], which essentially corrected the integrated intensity data for absorption and Lorentz-polarization.

The linear absorption coefficient μ_l for darapskite, used by GNABS to correct for absorption, was calculated to be 58.46 cm^{-1} from the known density of 2.20 gm/cm^3 [18] and from data tabulated in the *International Tables for X-ray Crystallography*.

VI. THREE-DIMENSIONAL PATTERSON SYNTHESIS

The next step in determining the crystal structure of darapskite was to compute a three-dimensional Patterson function $P(xyz)$ from the experimental diffraction data. The general form of this function is:

$$P(xyz) = \frac{1}{V} \sum_{h} \sum_{k} \sum_{l} \left| F_{hkl} \right|^{2} e^{2\pi i(hx+ky+lz)} \qquad (5)$$

and it gives the average value of the electron-density product at points at each end of a vector whose components are *xyz* as it ranges over a cell of volume *V*. The resulting three-dimensional Patterson synthesis has peaks at the ends of vectors which are interatomic vectors in the crystal structure.

A. Experimental procedure

The experimentally observed structure factors for one asymmetric unit of reciprocal space were used by the program GINPUT-GENFOR [21] to compute the Patterson function of darapskite in sections normal to the unique axis *b* (2nd setting, monoclinic). These sections were separated by intervals of 1/100 along *b*. Since the space group of darapskite is $P2_1/m$, the space group of its vector set is $2/m$. There are four operations in this space group so that its asymmetric unit is ¼ cell. Consequently, a block $a/2 \times b/2 \times c$ is an asymmetric unit in vector space for darapskite; and the Patterson function was computed at each grid point *xyz* for which $0 \le x \le .5$, $0 \le y \le .5$, $0 \le z \le 1.0$. The result was 51 Patterson sections normal to the $\underline{b}$ axis, from $\underline{P}(\underline{x}0\underline{z})$ to $\underline{P}(\underline{x}\tfrac{1}{2}\underline{z})$.

B. Preliminary considerations of the structure

As was previously determined, (p.23), the unit cell of darapskite contains two formula weights of composition $Na_3NO_3SO_4 \cdot H_2O$. The equipoints of space group $P2_1/m$ are:

2*a*	: on inversion centers
2*b*	
2*c*	
2*d*	
2*e*	: on mirrors
4*f*	: in the general position

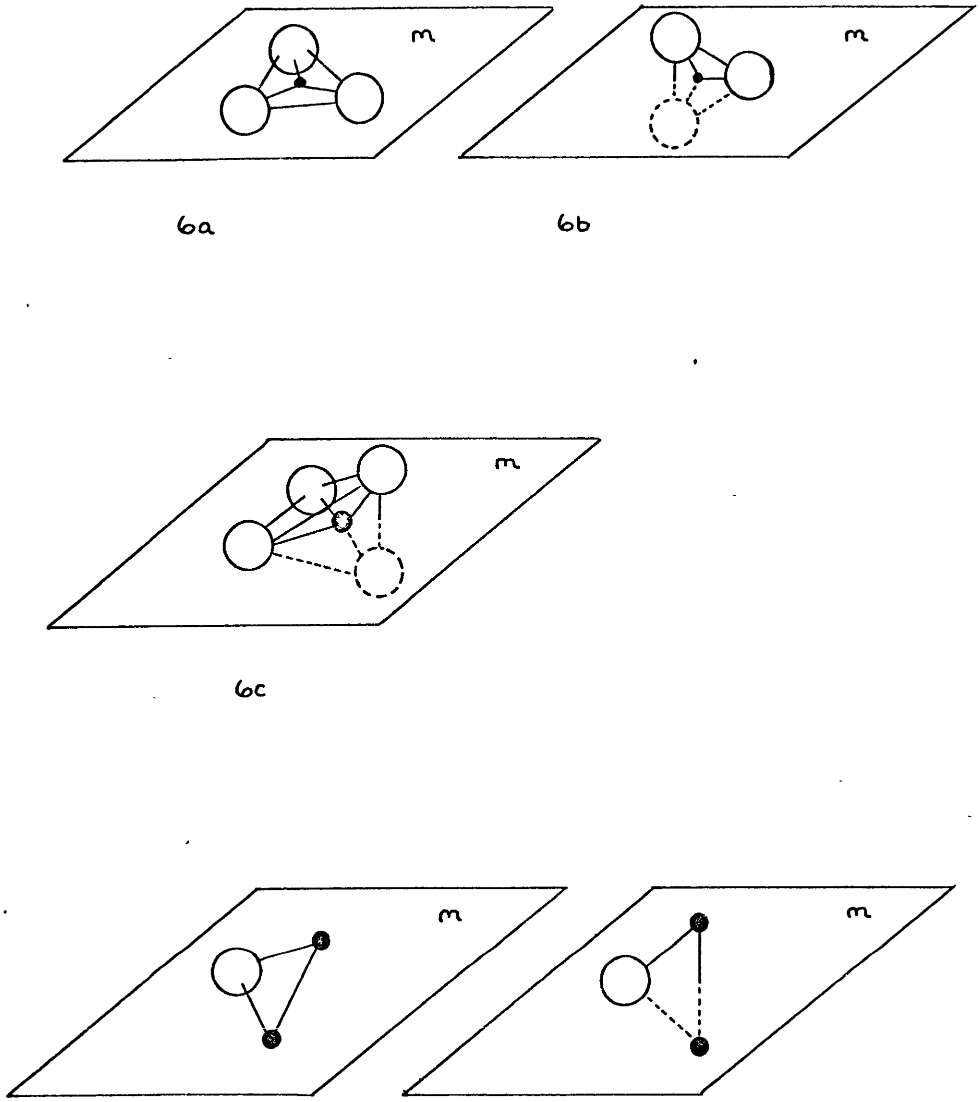
Since each cell has 2N, 2S, and $2H_2O$, these must all occupy special positions. Furthermore, in consideration of the symmetry requirements of the polyhedra formed by these atoms and their surrounding atoms (i.e. triangular NO_3, tetrahedral SO_4, angular H_2O), it is obvious that none of these could be situated on inversion centers. All, however, are consistent with mirror symmetry:

(i) The NO_3 equilateral triangle can either lie flat in the plane of a mirror (Fig.6a) or stand perpendicular to it with one O above the plane and another below (Fig.6b).

(ii) The SO_4 tetrahedron must be situated such that S and two O lie on a mirror, with the third and fourth oxygens above and below the plane respectively (Fig.6*c*).

(iii) The H_2O molecule can either lie flat in the plane of a mirror (Fig.6d) or stand perpendicular to it with one H above the plane and another below (Fig.6e).

The six Na atoms in the unit cell must either (1) all



6a 6b

6c

6d 6e

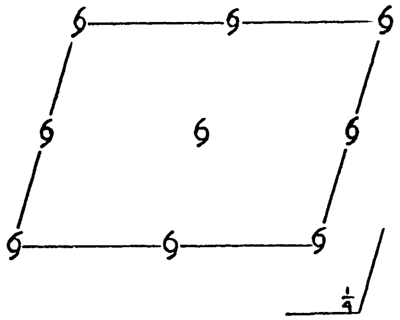
Fig. 6. Possible arrangements of NO_3, SO_4, and H_2O with respect to mirror

occupy three of the special positions (2*a*, 2*b*, 2*c*, 2*d*, or 2*e*) or (2) be situated such that four of them are in the general position (4*f*) and two are in one of the special positions. It seemed reasonable to assume at the outset that at least some of the Na must lie betweem the mirrors which are at 1/4 and 3/4 *b*, in order to utilize some of the otherwise empty space in the structure and to co-ordinate the two planes containing the H_2O, SO_4, and NO_3 groups.

In Table 3 are shown some characteristics of space group $P2_1/m$.

TABLE 3

Space group $P2_1/m$



2nd setting : unique axis b, origin at $\bar{1}$

Number of positions	Wyckoff notation	Point symmetry	Co-ordinates of equivalent positions
4	f	1	xyz, $\bar{x}\bar{y}\bar{z}$, $\bar{x}\ \tfrac{1}{2}+y\ \bar{z}$, $x\ \tfrac{1}{2}-y\ z$
2	e	m	$x\tfrac{1}{4}z$, $\bar{x}\tfrac{3}{4}\bar{z}$
2	d	$\bar{1}$	$\tfrac{1}{2}0\tfrac{1}{2}$, $\tfrac{1}{2}\tfrac{1}{2}\tfrac{1}{2}$
2	c	$\bar{1}$	$00\tfrac{1}{2}$, $0\tfrac{1}{2}\tfrac{1}{2}$
2	b	$\bar{1}$	$\tfrac{1}{2}00$, $\tfrac{1}{2}\tfrac{1}{2}0$
2	a	$\bar{1}$	000, $0\tfrac{1}{2}0$

C. Interpretation of the three-dimensional Patterson synthesis

The interpretation of the Patterson synthesis of darapskite began with a calibration of the expected peak heights for all pairs of atoms in the structure. The general basis for this calibration is that the height of a Patterson peak is proportional to the product of the number of electrons of the two atoms in the pair. "Half-ionized" states were assumed for the atoms involved. The results of this computation are shown in Table 4. The most prominent peaks to be expected are due to atom pairs S-S, S-Na, and S-O.

The general approach to solving the structure was to first determine the co-ordinates of the two symmetrically-equivalent sulfer atoms occurring on the mirrors. This required finding the location of the S-S inversion peak on the Harker section $P(x\frac{1}{2}z)$ of the Patterson synthesis. Since sulfer is a moderately heavy atom, it was expected that there would be an image of the complete structure from each of the sulfer atoms. In particular, the $P(x0z)$ section should show an image of the light atom arrangement within one mirror plane due to vectors originating from the sulfer atom in that plane; likewise, the $P(x\frac{1}{4}z)$ section should also show an image of the atom arrangement within the mirror due to vectors originating from the sulfer atom in the adjacent symmetry plane. A superposition of $P(x0z)$ and $P(x\frac{1}{4}z)$ should therefore indicate a coincidence of peaks when the corresponding images of the light atoms in the mirror plane are lined up.

This method, described elsewhere as the "vector-convergence method"*, was thus utilized to identify the S-S inversion peak. When the origin of the $P(x0z)$ map was displaced over the $P(x\frac{1}{2}z)$ map to a position such that exact coincidence of several peaks in both maps occurred, the Patterson peak in the Harker section coinciding with the origin of the zero-level section was accepted as the S-S inversion peak. This peak had co-ordinates typified by $2x, 2y, 2z$, where x, y, z are the co-ordinates of a sulfer atom in the actual crystal structure.

The identity of the S-S peak was further established by noting that it was accompanied by smaller peaks of the correct magnitude and at the proper tetrahedral distance and angular orientation to be the S-O satellite peaks.

With the co-ordinates of the SO_4 tetrahedra known, the co-ordinates of all the other lighter atoms on the mirror planes, except H_2O, were easily deduced. Furthermore, due to relatively heavy Na atoms located between the mirrors, section $P(x\ .25\ z)$ of the Patterson synthesis showed an image, uncluttered by background peaks, of the atomic arrangement on the symmetry planes (Fig.7). Therefore this section verified the positions of the atoms on equipoint 2*e*, as well as providing the co-ordinates of four Na atoms in the general position.

---

*M. J. Buerger, *Vector Space*, pp.252-253.

TABLE 4

Calibration of expected peak heights

(height of origin peak = 1302)

Atom	Z	Z^2	Z^2 • (no. atoms/cell)
S^0	16	256	256 • 2 = 512.00
Na^{+1}	10	100	100 • 6 = 160.00
$N^{+1/2}$	6½	42.25	42.25 • 2 = 84.50
$O^{-1/2}$	8½	72.25	72.25 • 16 = 1156.00
$H^{+1/4}$	¾	9/16	9/16 • 4 = 2.25
			ΣZ^2 = 2354.75

Atom pairs	Z_iZ_j	$H_{ij} = \frac{H_{origin}}{\Sigma Z^2} \cdot Z_iZ_j$
S-S	256	142
S-Na	160	88.5
S-N	104	57.5
S-O	136	75
Na-Na	100	55
Na-N	65	36
Na-O	85	47
N-N	42.25	23
N-O	55.25	31
O-O	72.25	40

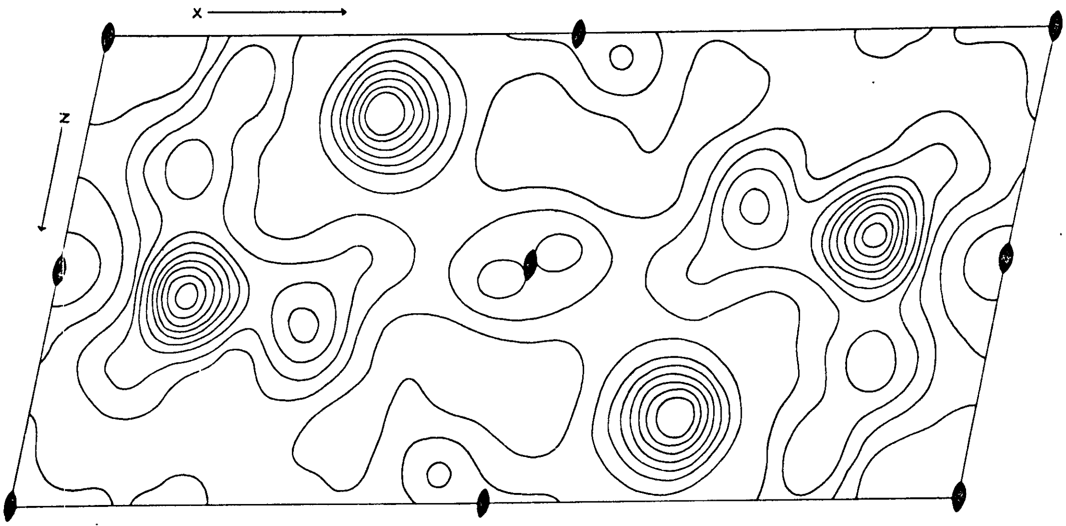


Fig. 7

Darapskite, Patterson section P(x .25 z)

VII. STRUCTURE REFINEMENT

Using atomic scattering factors obtained from the International Tables for X-ray Crystallography ~~and~~ and the atomic co-ordinates which were deduced from the Patterson synthesis, a preliminary set of structure factors $\underline{F}_{\underline{hkl}}$ for darapskite were computed by the program MIRA[23]. The phases of these calculated structure factors were then assigned to the absolute values of the corresponding observed structure factors. From the resultant set of $\underline{F}_{obs}$, the program GINPUT-GENFOR[21] computed a three-dimensional Fourier synthesis for one asymmetric unit of the crystal cell according to the formula:

$$\rho(\underline{x}\underline{y}\underline{z}) = \frac{1}{V}\sum_h\sum_k\sum_l \underline{F}_{\underline{hkl}} e^{-2\pi i(\underline{h}\underline{x}+\underline{k}\underline{y}+\underline{l}\underline{z})}. \qquad (6)$$

The contoured electron-density maps not only showed peaks at the previously determined atom locations, but also revealed the position of the water molecule on the symmetry plane.

Using the newly found co-ordinates of H_2O and the improved co-ordinates of the other atoms, which were suggested from the Fourier synthesis, a least-squares refinement of the structure was begun with computer program SFLSQ3[20]. At the start of this procedure, the residual factor $\underline{R}$ was 24%, where

$$\underline{R} = \frac{\sum\big||\underline{F}_{obs}| - |\underline{F}_{calc}|\big|}{\sum|\underline{F}_{obs}|} . \qquad (7)$$

After several refinement cycles in which the atom co-ordinates and isotropic thermal coefficients were allowed to vary, a weighting scheme was introduced which was based on a statistical comparison of the discrepancies between F_{obs} and F_{calc}* . In addition, corrections for anomalous scattering of the sulfur, sodium, and oxygen atoms were made (Table 5). With these changes, the least-squares refinement was continued until an unweighted R of 8.8% and a weighted R of 6.3% were reached. The final positional parameters for the various atoms in the unit cell are listed in Table 6. Final temperature factors are omitted because their values are not completely refined to date. An electron-density difference synthesis suggests, however, that O_5 (one of the nitrate group oxygens) has considerable anisotropic thermal motion and that perhaps an attempt should be made to represent it as two "half-atoms"[12]. Table 7 in the Appendix shows the agreement between the observed and computed structure factors.

Finally, a Fourier synthesis was computed using the observed structure factors and refined atom co-ordinates. A section through this synthesis, $P(x\frac{1}{4}z)$, showing the electron density on the symmetry plane at $\frac{1}{4}b$ can be seen in Fig.8. An interpretation of this electron density is shown in Fig.9.

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*B.J.Wuensch, personal communication.

TABLE 5

Dispersion corrections for atomic scattering factors

Cu*K*α radiation

at $(\sin\theta)/\lambda = 0$

Atom	Atomic number	$\Delta f'$	$\Delta f''$
N	7	0.0	0.0
O	8	0.0	0.1
Na	11	0.1	0.2
S	16	0.3	0.6

TABLE 6

Atomic positional parameters of darapskite

Atom	Equipoint in $P2_1/m$	Representative co-ordinates (2nd setting)	Parameters		
			x	y	z
S	2*e*	x $\frac{1}{4}$ z	.922		.176
Na(1)	2*e*	x $\frac{1}{4}$ z	.096		.767
Na(2)	4*f*	x y z	.226	.506	.365
N	2*e*	x $\frac{1}{4}$ z	.380		.928
O(1)	2*e*	x $\frac{1}{4}$ z	.065		.278
O(2)	2*e*	x $\frac{1}{4}$ z	.897		.884
O(3)	4*f*	x y z	.865	.422	.265
O(4)	2*e*	x $\frac{1}{4}$ z	.331		.687
O(5)	2*e*	x $\frac{1}{4}$ z	.502		.997
O(6)	2*e*	x $\frac{1}{4}$ z	.314		.095
O(7) = H_2O	2*e*	x $\frac{1}{4}$ z	.612		.541

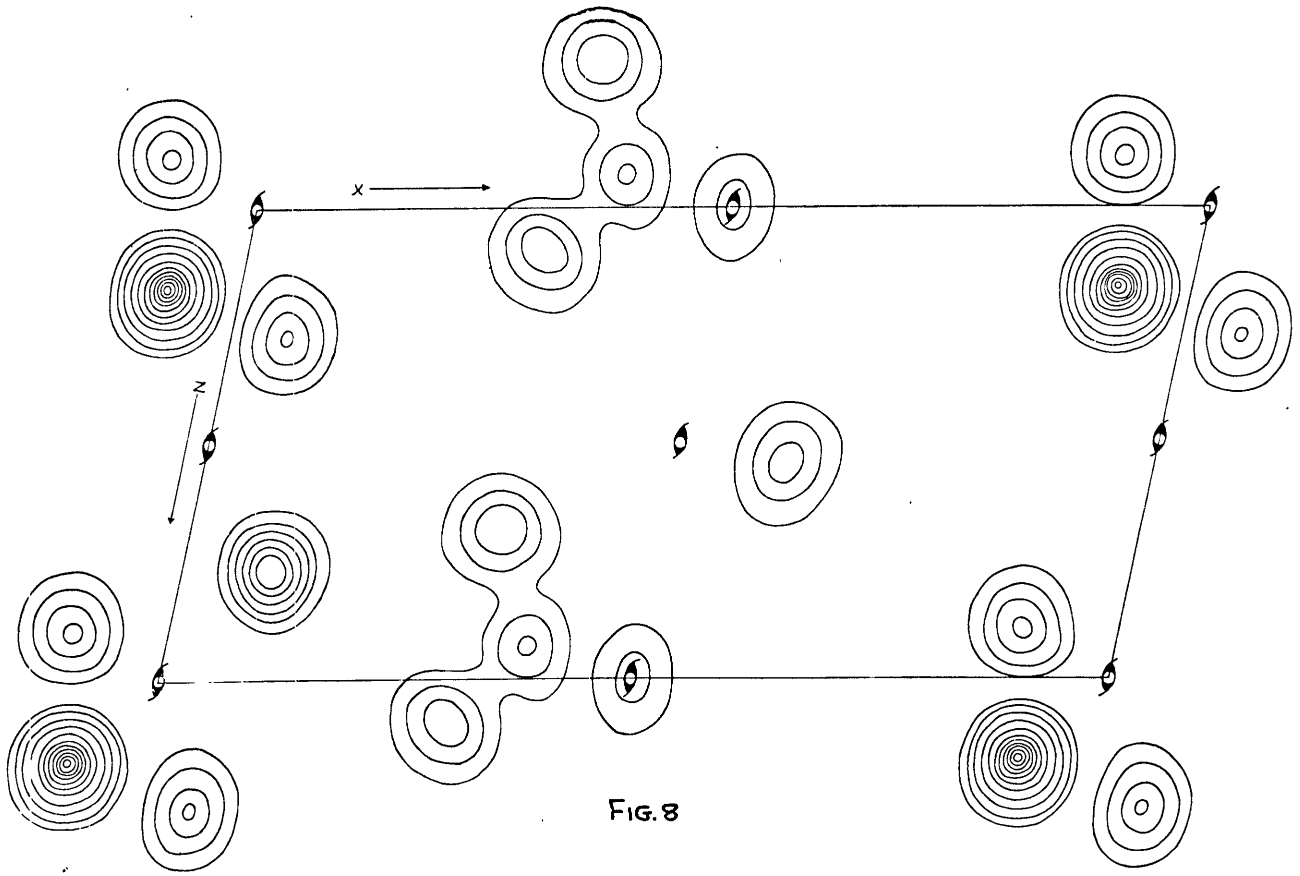

FIG. 8

DARAPSKITE, ELECTRON DENSITY $\rho(x\frac{1}{4}z)$, OMITTING ZERO CONTOUR

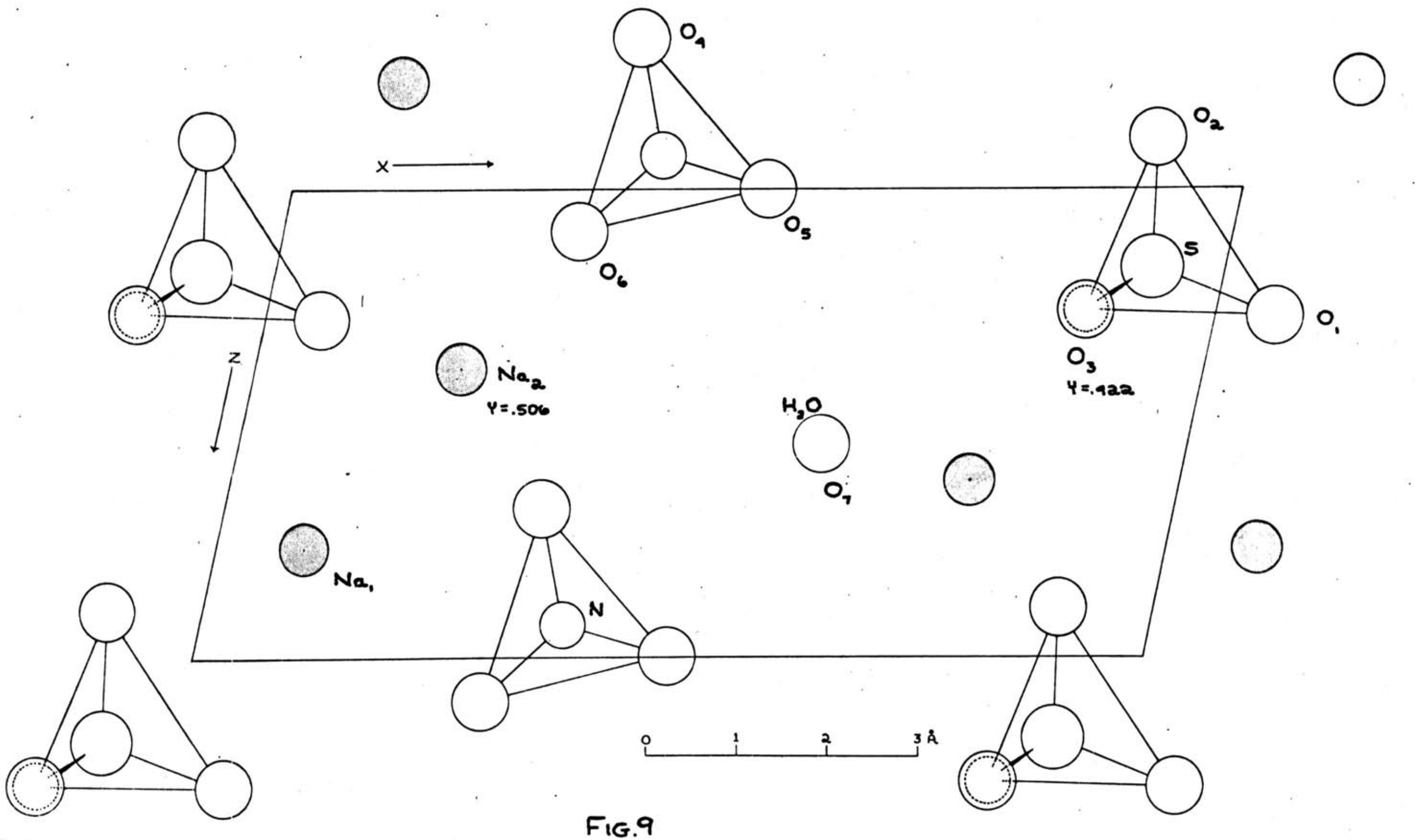

FIG. 9

INTERPRETATION OF FIG. 8; ALL ATOMS AT $Y = \frac{1}{4}$ EXCEPT Na_2 AND O_3

VIII. DESCRIPTION OF THE STRUCTURE

The structure of darapskite consists essentially of an arrangement of atoms on mirror planes which are related to each other by the operation of a 2_1 screw axis. Sandwiched between the planes and linking them together are octahedrally co-ordinated Na atoms, each of which is bonded to three oxygen atoms from the plane above and three from the plane below.

Within the mirror planes themselves, the atomic arrangement is such that the H_2O molecules and Na atoms provide linkages between the SO_4 and NO_3 groups. Each Na in the plane of the mirror is co-ordinated by seven oxygen atoms in all. Five of these oxygens belong to NO_3 and SO_4 groups in the same plane as the Na, while two oxygens belong to SO_4 groups in the adjacent planes. On the other hand, each tetrahedrally co-ordinated H_2O molecule is bonded (by hydrogen bonds) to two oxygen atoms of two different NO_3 groups in the same plane and to two Na atoms in the general position above and below the mirror. These relationships concerning the locations of atoms on the symmetry planes are shown in Fig. 9.

In summation, the structure of darapskite can be regarded as consisting of alternating layers of atoms perpendicular to the b axis. These layers are of two types: the first (at 0, $\frac{1}{2}\underline{b}$, $\frac{2}{2}\underline{b}$, ...) is composed exclusively of Na atoms, while the second (at $\frac{1}{4}\underline{b}$, $\frac{3}{4}\underline{b}$, $\frac{5}{4}\underline{b}$, ...) is composed of SO_4, NO_3, H_2O, and Na atoms. Cohesion of the layers is effected by bonding between

Na and O such that Na in the general position is in 6-fold co-ordination and Na on the mirrors is in 7-fold co-ordination. Perfect cleavage on {010} arises from the rupture of Na-O bonds between the layers.

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Appendix

TABLE 7

Structure Factors

h	K	L	F_{OBS}	F_{CALC}
1	0	0	54.31	55.47
-1	0	0	56.55	55.47
2	0	0	3.22	3.73
-2	0	0	3.97	3.73
3	0	0	18.25	15.63
-3	0	0	18.79	15.63
4	0	0	46.92	45.93
-4	0	0	47.93	45.93
5	0	0	47.33	45.10
-5	0	0	43.22	45.10
6	0	0	43.53	42.28
-6	0	0	43.52	42.28
7	0	0	52.73	48.96
-7	0	0	52.90	48.96
8	0	0	11.38	6.05
-8	0	0	11.71	6.05
9	0	0	13.30	15.99
-9	0	0	13.38	15.99
10	0	0	15.65	12.34
-10	0	0	15.25	12.34
11	0	0	7.91	6.32
-11	0	0	8.34	6.32
12	0	0	8.90	8.84
-12	0	0	9.36	8.84
1	-1	0	8.35	7.67
-1	-1	0	7.39	7.67
2	-1	0	47.33	47.39
-2	-1	0	46.04	47.39
3	-1	0	26.64	25.42
-3	-1	0	25.40	25.42
4	-1	0	26.60	24.16
-4	-1	0	25.98	24.16
5	-1	0	6.91	7.40
-5	-1	0	6.83	7.40
6	-1	0	3.04	.67
-6	-1	0	2.57	.67
7	-1	0	20.00	19.12
-7	-1	0	20.17	19.12
8	-1	0	0.	2.31
-8	-1	0	0.	2.31
9	-1	0	5.42	.51
-9	-1	0	0.	.51
10	-1	0	7.82	9.27
-10	-1	0	9.14	9.27
11	-1	0	5.52	3.58
-11	-1	0	0.	3.58

h	k	l	F_{obs}	F_{calc}
12	-1	0	2.62	3.13
-12	-1	0	2.62	3.13
0	-2	0	96.63	95.26
1	-2	0	3.65	5.56
-1	-2	0	3.50	5.56
2	-2	0	76.32	74.10
-2	-2	0	76.21	74.10
3	-2	0	52.56	49.24
-3	-2	0	52.72	49.24
4	-2	0	60.21	60.68
-4	-2	0	63.26	60.68
5	-2	0	68.06	65.93
-5	-2	0	66.94	65.93
6	-2	0	16.62	16.18
-6	-2	0	17.53	16.18
7	-2	0	25.18	24.38
-7	-2	0	25.36	24.38
8	-2	0	19.72	21.48
-8	-2	0	19.77	21.48
9	-2	0	20.76	17.62
-9	-2	0	21.56	17.62
10	-2	0	19.74	18.78
-10	-2	0	19.64	18.78
11	-2	0	21.04	22.84
-11	-2	0	21.19	22.84
12	-2	0	16.67	17.19
-12	-2	0	16.73	17.19
1	-3	0	38.55	35.40
-1	-3	0	38.34	35.40
2	-3	0	6.54	2.40
-2	-3	0	2.16	2.40
3	-3	0	8.00	4.97
-3	-3	0	7.08	4.97
4	-3	0	6.98	7.12
-4	-3	0	8.37	7.12
5	-3	0	24.80	22.84
-5	-3	0	24.47	22.84
6	-3	0	15.26	14.24
-6	-3	0	14.50	14.24
7	-3	0	5.61	8.24
-7	-3	0	0.	8.24
8	-3	0	2.05	3.01
-8	-3	0	5.24	3.01
9	-3	0	12.43	11.80
-9	-3	0	11.21	11.80
10	-3	0	16.67	17.04
-10	-3	0	16.93	17.04
11	-3	0	3.58	4.40

h	K	L	FOBS	FCALC
-11	-3	0	3.92	4.40
0	-4	0	112.46	112.43
1	-4	0	25.32	20.57
-1	-4	0	25.05	20.57
2	-4	0	5.99	2.85
-2	-4	0	4.89	2.85
3	-4	0	9.87	7.21
-3	-4	0	10.23	7.21
4	-4	0	9.28	7.56
-4	-4	0	7.43	7.56
5	-4	0	18.14	19.95
-5	-4	0	17.11	19.95
6	-4	0	40.36	36.91
-6	-4	0	39.43	36.91
7	-4	0	46.11	45.34
-7	-4	0	45.81	45.34
8	-4	0	8.42	6.92
-8	-4	0	3.82	6.92
9	-4	0	9.01	8.79
-9	-4	0	8.91	8.79
10	-4	0	13.34	13.07
-10	-4	0	13.91	13.07
11	-4	0	.94	2.70
-11	-4	0	0.	2.70
1	-5	0	0.	2.92
-1	-5	0	0.	2.92
2	-5	0	21.51	21.00
-2	-5	0	21.68	21.00
3	-5	0	11.47	9.97
-3	-5	0	9.69	9.97
4	-5	0	8.23	7.97
-4	-5	0	5.28	7.97
5	-5	0	7.38	6.32
-5	-5	0	6.47	6.32
6	-5	0	0.	.93
-6	-5	0	3.86	.93
7	-5	0	9.27	11.36
-7	-5	0	10.01	11.36
8	-5	0	2.32	1.93
-8	-5	0	3.79	1.93
9	-5	0	5.69	1.53
-9	-5	0	6.16	1.53
10	-5	0	6.76	5.63
-10	-5	0	5.57	5.63
0	-6	0	41.50	44.44
1	-6	0	23.89	20.40
-1	-6	0	24.06	20.40
2	-6	0	28.84	28.17

h	K	L	F_{obs}	F_{calc}
-2	-6	0	28.80	28.17
3	-6	0	9.64	8.25
-3	-6	0	9.97	8.25
4	-6	0	42.19	38.91
-4	-6	0	41.00	38.91
5	-6	0	35.63	35.28
-5	-6	0	34.74	35.28
6	-6	0	10.51	9.82
-6	-6	0	9.58	9.82
7	-6	0	2.53	6.96
-7	-6	0	3.34	6.96
8	-6	0	8.35	6.90
-8	-6	0	8.42	6.90
9	-6	0	8.14	7.78
-9	-6	0	8.38	7.78
1	-7	0	5.24	3.74
-1	-7	0	3.43	3.74
2	-7	0	9.49	10.09
-2	-7	0	10.08	10.09
3	-7	0	12.17	11.21
-3	-7	0	11.79	11.21
4	-7	0	4.11	1.33
-4	-7	0	0.	1.33
5	-7	0	0.	3.82
-5	-7	0	1.72	3.82
6	-7	0	2.91	.69
-6	-7	0	2.20	.69
7	-7	0	4.83	5.43
-7	-7	0	3.60	5.43
0	-8	0	33.82	36.06
1	-8	0	13.54	11.90
-1	-8	0	13.31	11.90
2	-8	0	1.54	1.80
-2	-8	0	3.76	1.80
3	-8	0	4.47	4.27
-3	-8	0	4.58	4.27
4	-8	0	2.46	4.05
-4	-8	0	3.22	4.05
1	0	1	13.16	12.93
-2	0	1	7.72	8.30
2	0	1	74.53	76.34
-3	0	1	70.44	69.07
3	0	1	62.35	66.18
-4	0	1	44.61	46.87
4	0	1	13.46	13.21
-5	0	1	13.96	12.17
5	0	1	73.04	72.30
-6	0	1	46.49	42.91

h	k	l	Fobs	Fcalc
6	0	1	21.01	23.08
-7	0	1	8.97	8.36
7	0	1	9.23	5.44
-8	0	1	1.88	6.21
8	0	1	6.90	9.37
-9	0	1	1.41	2.06
9	0	1	19.29	16.35
-10	0	1	25.88	24.31
10	0	1	0.	6.22
-11	0	1	15.91	19.89
11	0	1	1.67	4.43
-12	0	1	4.02	.86
0	-1	1	9.93	10.87
-1	-1	1	20.49	22.08
1	-1	1	47.02	45.24
-2	-1	1	66.01	64.75
2	-1	1	33.41	31.85
-3	-1	1	3.23	2.64
3	-1	1	8.88	11.59
-4	-1	1	27.19	25.63
4	-1	1	4.31	4.68
-5	-1	1	7.93	8.15
5	-1	1	18.80	18.99
-6	-1	1	16.18	16.02
6	-1	1	3.22	1.36
-7	-1	1	2.75	2.02
7	-1	1	8.55	5.95
-8	-1	1	32.23	30.99
8	-1	1	3.17	3.79
-9	-1	1	21.80	23.09
9	-1	1	8.59	7.97
-10	-1	1	12.44	13.06
10	-1	1	7.13	5.61
-11	-1	1	12.80	12.37
11	-1	1	6.67	5.68
-12	-1	1	8.36	7.89
0	-2	1	62.21	64.18
-1	-2	1	15.91	21.80
1	-2	1	48.84	46.20
-2	-2	1	19.27	18.36
2	-2	1	3.47	1.75
-3	-2	1	11.60	13.44
3	-2	1	26.35	25.49
-4	-2	1	10.04	9.25
4	-2	1	23.17	22.95
-5	-2	1	42.60	42.30
5	-2	1	9.48	9.77
-6	-2	1	35.64	35.07

h	K	L	Fobs	Fcalc
6	-2	1	5.73	5.81
-7	-2	1	4.66	5.85
7	-2	1	35.97	34.52
-8	-2	1	39.83	38.58
8	-2	1	26.75	29.40
-9	-2	1	26.13	26.94
9	-2	1	4.25	4.58
-10	-2	1	12.60	10.11
10	-2	1	3.44	1.55
-11	-2	1	0.	3.55
11	-2	1	8.09	7.73
-12	-2	1	6.11	5.41
0	-3	1	22.63	21.02
-1	-3	1	1.85	.86
1	-3	1	2.13	4.43
-2	-3	1	49.03	47.75
2	-3	1	23.96	24.50
-3	-3	1	24.85	24.04
3	-3	1	13.67	11.50
-4	-3	1	44.81	43.08
4	-3	1	20.24	20.52
-5	-3	1	17.32	17.35
5	-3	1	6.23	5.55
-6	-3	1	21.91	21.32
6	-3	1	8.19	7.89
-7	-3	1	20.82	20.88
7	-3	1	20.53	18.59
-8	-3	1	38.59	38.01
8	-3	1	11.92	11.91
-9	-3	1	18.73	19.71
9	-3	1	8.08	8.28
-10	-3	1	0.	4.42
10	-3	1	0.	1.57
-11	-3	1	3.35	3.89
11	-3	1	10.74	10.75
0	-4	1	0.	5.00
-1	-4	1	20.93	16.68
1	-4	1	4.72	6.43
-2	-4	1	20.06	22.96
2	-4	1	24.19	25.06
-3	-4	1	30.17	31.15
3	-4	1	37.43	33.31
-4	-4	1	45.96	41.08
4	-4	1	10.28	11.52
-5	-4	1	22.79	23.92
5	-4	1	29.36	29.74
-6	-4	1	11.67	11.50
6	-4	1	7.03	5.12

h	K	L	F_{OBS}	F_{CALC}
-7	-4	1	7.19	3.21
7	-4	1	0.	.99
-8	-4	1	2.49	2.44
8	-4	1	12.23	12.40
-9	-4	1	10.72	9.42
9	-4	1	19.96	20.01
-10	-4	1	23.30	24.06
10	-4	1	8.49	10.90
-11	-4	1	13.68	15.81
0	-5	1	6.29	5.47
-1	-5	1	11.59	9.75
1	-5	1	20.11	19.83
-2	-5	1	27.68	26.11
2	-5	1	7.29	8.93
-3	-5	1	6.37	6.04
3	-5	1	9.43	8.02
-4	-5	1	10.34	10.25
4	-5	1	10.07	8.84
-5	-5	1	0.	.26
5	-5	1	13.53	13.25
-6	-5	1	11.08	9.95
6	-5	1	2.86	.90
-7	-5	1	2.02	2.12
7	-5	1	7.58	4.32
-8	-5	1	17.82	19.07
8	-5	1	5.08	3.76
-9	-5	1	15.35	16.97
9	-5	1	3.60	4.93
-10	-5	1	9.19	9.97
0	-6	1	21.49	22.83
-1	-6	1	20.92	19.48
1	-6	1	39.34	33.81
-2	-6	1	32.13	26.98
2	-6	1	18.34	16.92
-3	-6	1	14.37	14.65
3	-6	1	7.99	4.14
-4	-6	1	5.17	.99
4	-6	1	10.97	9.72
-5	-6	1	8.36	10.31
5	-6	1	0.	1.21
-6	-6	1	12.35	10.46
6	-6	1	9.96	8.08
-7	-6	1	1.80	.09
7	-6	1	20.98	21.96
-8	-6	1	14.61	15.26
8	-6	1	14.43	15.03
0	-7	1	10.07	7.33
-1	-7	1	12.09	11.18

h	K	L	F_{OBS}	F_{CALC}
1	-7	1	8.38	8.20
-2	-7	1	14.11	16.32
2	-7	1	9.24	8.82
-3	-7	1	0.	2.90
3	-7	1	1.83	2.81
-4	-7	1	9.36	9.44
4	-7	1	1.73	1.22
-5	-7	1	4.20	3.82
5	-7	1	9.64	9.19
-6	-7	1	8.07	8.53
6	-7	1	7.01	6.26
-7	-7	1	8.38	8.39
0	-8	1	0.	3.13
-1	-8	1	8.06	6.86
1	-8	1	0.	2.03
-2	-8	1	6.89	5.14
2	-8	1	10.56	10.53
-3	-8	1	12.93	12.45
0	0	2	50.77	50.42
1	0	2	46.81	43.78
-2	0	2	20.11	21.24
2	0	2	17.38	17.28
-3	0	2	16.12	16.70
3	0	2	5.22	8.55
-4	0	2	51.87	50.26
4	0	2	12.02	11.89
-5	0	2	8.04	6.24
5	0	2	59.97	64.18
-6	0	2	19.77	20.47
6	0	2	53.44	52.47
-7	0	2	31.01	33.51
7	0	2	10.80	10.39
-8	0	2	32.19	31.65
8	0	2	3.10	7.97
-9	0	2	11.05	7.72
9	0	2	14.71	13.43
-10	0	2	14.55	12.55
10	0	2	4.35	4.97
-11	0	2	0.	2.94
-12	0	2	5.25	4.63
0	-1	2	15.58	16.76
-1	-1	2	1.95	1.02
1	-1	2	15.08	12.33
-2	-1	2	15.53	14.92
2	-1	2	4.68	2.86
-3	-1	2	37.80	36.08
3	-1	2	8.56	7.04
-4	-1	2	16.93	15.86

h	K	L	Fobs	Fcalc
4	-1	2	5.81	4.67
-5	-1	2	4.70	3.75
5	-1	2	9.01	10.64
-6	-1	2	10.00	8.12
6	-1	2	4.66	3.08
-7	-1	2	14.53	13.39
7	-1	2	4.33	5.18
-8	-1	2	25.50	25.22
8	-1	2	9.67	9.69
-9	-1	2	0.	5.94
9	-1	2	0.	.49
-10	-1	2	0.	1.13
10	-1	2	0.	1.79
-11	-1	2	4.06	3.06
-12	-1	2	12.84	14.95
0	-2	2	5.36	4.47
-1	-2	2	8.33	10.81
1	-2	2	75.02	71.41
-2	-2	2	21.04	19.47
2	-2	2	8.81	8.00
-3	-2	2	72.25	71.77
3	-2	2	35.56	35.71
-4	-2	2	10.02	8.82
4	-2	2	23.24	22.39
-5	-2	2	28.95	31.19
5	-2	2	20.98	20.35
-6	-2	2	25.72	26.49
6	-2	2	20.53	19.15
-7	-2	2	22.62	23.50
7	-2	2	10.91	13.72
-8	-2	2	12.07	9.49
8	-2	2	28.79	27.62
-9	-2	2	5.38	4.48
9	-2	2	11.66	10.83
-10	-2	2	29.31	28.57
10	-2	2	22.57	25.68
-11	-2	2	8.19	6.57
-12	-2	2	14.41	16.79
0	-3	2	14.78	13.74
-1	-3	2	22.89	22.28
1	-3	2	22.32	22.27
-2	-3	2	19.17	18.13
2	-3	2	18.08	19.18
-3	-3	2	34.88	35.53
3	-3	2	4.78	6.19
-4	-3	2	1.55	1.68
4	-3	2	5.85	4.61
-5	-3	2	26.41	24.45

h	K	L	F_{OBS}	F_{CALC}
5	-3	2	0.	.55
-6	-3	2	8.29	7.77
6	-3	2	8.84	10.10
-7	-3	2	12.23	12.02
7	-3	2	0.	1.87
-8	-3	2	13.01	14.26
8	-3	2	11.50	12.17
-9	-3	2	4.36	3.79
9	-3	2	7.78	8.83
-10	-3	2	0.	7.77
-11	-3	2	5.56	3.91
0	-4	2	19.69	15.82
-1	-4	2	54.31	55.66
1	-4	2	4.47	7.31
-2	-4	2	28.98	24.12
2	-4	2	11.64	11.32
-3	-4	2	2.69	6.00
3	-4	2	3.67	2.67
-4	-4	2	10.98	12.24
4	-4	2	5.57	2.56
-5	-4	2	6.92	6.89
5	-4	2	40.36	38.27
-6	-4	2	3.60	4.29
6	-4	2	37.25	38.61
-7	-4	2	37.30	36.13
7	-4	2	0.	1.80
-8	-4	2	29.81	30.86
8	-4	2	2.95	1.08
-9	-4	2	2.62	2.45
9	-4	2	7.13	7.91
-10	-4	2	3.52	4.88
-11	-4	2	3.72	3.19
0	-5	2	13.97	12.38
-1	-5	2	0.	1.34
1	-5	2	0.	.68
-2	-5	2	9.10	8.10
2	-5	2	3.34	1.07
-3	-5	2	10.33	12.54
3	-5	2	6.19	4.67
-4	-5	2	6.27	7.56
4	-5	2	3.93	.23
-5	-5	2	2.40	2.92
5	-5	2	5.12	6.05
-6	-5	2	5.01	.72
6	-5	2	6.37	5.48
-7	-5	2	11.38	10.79
7	-5	2	7.91	7.19
-8	-5	2	13.86	14.82

h	K	L	Fobs	Fcalc
8	-5	2	6.69	7.47
-9	-5	2	2.57	2.58
-10	-5	2	0.	1.37
0	-6	2	22.34	19.17
-1	-6	2	11.41	13.54
1	-6	2	37.16	37.16
-2	-6	2	12.75	10.90
2	-6	2	4.74	4.30
-3	-6	2	21.49	21.61
3	-6	2	29.54	26.83
-4	-6	2	0.	1.98
4	-6	2	24.53	23.49
-5	-6	2	17.98	17.76
5	-6	2	19.96	19.35
-6	-6	2	9.42	9.13
6	-6	2	9.07	10.74
-7	-6	2	5.06	6.94
-8	-6	2	0.	2.48
0	-7	2	6.26	4.97
-1	-7	2	3.77	3.81
1	-7	2	2.75	.69
-2	-7	2	0.	2.10
2	-7	2	5.58	4.57
-3	-7	2	8.12	7.91
3	-7	2	3.37	4.98
-4	-7	2	0.	1.88
4	-7	2	0.	2.20
-5	-7	2	8.36	7.79
-6	-7	2	3.76	1.21
0	0	3	11.52	10.59
1	0	3	65.85	63.85
2	0	3	50.21	54.72
-12	0	3	10.83	12.94
-3	0	3	5.66	5.76
3	0	3	15.80	17.79
-4	0	3	41.50	39.36
4	0	3	21.01	21.99
-5	0	3	11.74	13.95
5	0	3	19.53	22.90
-6	0	3	4.31	3.12
6	0	3	26.94	27.05
-7	0	3	17.00	19.14
7	0	3	11.40	13.89
-8	0	3	6.25	5.43
8	0	3	21.69	22.08
-9	0	3	7.90	10.08
9	0	3	8.74	11.54
-10	0	3	2.83	4.20

h	K	L	Fobs	Fcalc
-11	0	3	16.27	17.03
0	-1	3	12.86	12.75
-1	-1	3	23.10	22.67
1	-1	3	12.03	12.62
-2	-1	3	16.47	16.71
2	-1	3	4.39	3.32
-3	-1	3	18.62	21.60
3	-1	3	27.68	28.50
-4	-1	3	11.45	12.16
4	-1	3	27.93	27.75
-5	-1	3	21.38	20.49
5	-1	3	2.78	1.84
-6	-1	3	2.40	1.82
6	-1	3	12.38	12.89
-7	-1	3	7.09	7.67
7	-1	3	0.	1.17
-8	-1	3	3.09	2.58
8	-1	3	13.27	14.01
-9	-1	3	24.89	26.26
9	-1	3	3.21	3.15
-10	-1	3	17.80	18.84
-11	-1	3	8.87	11.52
-12	-1	3	9.29	10.41
0	-2	3	37.36	39.65
-1	-2	3	30.18	31.25
1	-2	3	27.82	25.77
-2	-2	3	7.82	9.38
2	-2	3	9.67	11.58
-3	-2	3	29.97	31.19
3	-2	3	8.89	8.15
-4	-2	3	28.85	28.34
4	-2	3	14.44	15.73
-5	-2	3	9.78	9.58
5	-2	3	8.06	6.73
-6	-2	3	18.79	19.40
6	-2	3	38.82	39.92
-7	-2	3	19.69	19.05
7	-2	3	15.20	18.21
-8	-2	3	8.24	6.95
8	-2	3	6.23	6.36
-9	-2	3	25.19	25.70
9	-2	3	4.11	5.57
-10	-2	3	12.40	13.05
-11	-2	3	1.09	.27
0	-3	3	7.96	8.88
-1	-3	3	13.72	12.90
1	-3	3	26.50	27.26
-2	-3	3	24.30	24.14

h	K	L	F_{OBS}	F_{CALC}
2	-3	3	1.63	1.20
-3	-3	3	32.44	33.78
3	-3	3	13.83	12.65
-4	-3	3	22.17	22.73
4	-3	3	13.96	11.59
-5	-3	3	20.36	20.67
5	-3	3	5.81	5.80
-6	-3	3	3.27	4.04
6	-3	3	11.69	12.37
-7	-3	3	18.82	18.66
7	-3	3	4.61	4.18
-8	-3	3	11.09	12.56
8	-3	3	6.65	6.28
-9	-3	3	21.41	23.05
-10	-3	3	8.71	9.48
-11	-3	3	2.19	3.08
0	-4	3	3.39	1.37
-1	-4	3	3.92	2.63
1	-4	3	35.80	35.51
-2	-4	3	16.04	15.08
2	-4	3	28.60	27.05
-3	-4	3	4.51	6.45
3	-4	3	0.	3.41
-4	-4	3	31.67	32.50
4	-4	3	15.47	15.19
-5	-4	3	26.29	25.18
5	-4	3	12.60	11.37
-6	-4	3	10.35	12.35
6	-4	3	13.09	13.20
-7	-4	3	9.07	9.87
7	-4	3	9.14	8.16
-8	-4	3	10.35	9.02
-9	-4	3	3.49	.20
-10	-4	3	3.36	1.72
0	-5	3	8.54	9.03
-1	-5	3	13.03	12.06
1	-5	3	4.83	4.97
-2	-5	3	8.62	9.77
2	-5	3	2.24	2.15
-3	-5	3	14.69	15.28
3	-5	3	20.43	20.69
-4	-5	3	11.56	10.89
4	-5	3	19.05	19.73
-5	-5	3	10.06	12.46
5	-5	3	1.15	.74
-6	-5	3	5.37	.34
6	-5	3	8.23	8.16
-7	-5	3	2.97	4.89

h	k	L	F_{OBS}	F_{CALC}
-8	-5	3	1.12	2.14
-9	-5	3	15.33	17.95
0	-6	3	21.26	20.19
-1	-6	3	7.38	8.05
1	-6	3	18.43	19.34
-2	-6	3	10.20	11.26
2	-6	3	9.33	7.41
-3	-6	3	19.38	18.30
3	-6	3	9.15	10.39
-4	-6	3	5.00	9.59
4	-6	3	7.18	7.31
-5	-6	3	7.02	7.94
-6	-6	3	6.43	5.71
-7	-6	3	13.29	11.66
0	-7	3	2.03	2.47
-1	-7	3	9.74	10.89
1	-7	3	7.80	8.23
-2	-7	3	10.09	11.86
-3	-7	3	11.24	11.63
-4	-7	3	7.61	6.63
0	0	4	3.02	3.80
1	0	4	5.61	3.65
2	0	4	5.56	3.75
-3	0	4	1.79	5.16
3	0	4	13.30	13.31
-4	0	4	.90	3.29
4	0	4	28.02	34.08
-5	0	4	9.16	8.58
5	0	4	26.64	29.33
-6	0	4	8.66	10.51
6	0	4	7.25	5.09
-7	0	4	9.96	10.35
7	0	4	8.35	12.38
-8	0	4	10.75	13.73
-9	0	4	31.04	32.65
-10	0	4	8.53	11.50
-11	0	4	6.45	5.61
0	-1	4	10.68	8.93
-1	-1	4	9.56	9.90
1	-1	4	0.	2.07
-2	-1	4	27.23	27.47
2	-1	4	13.85	15.22
-3	-1	4	21.27	21.66
3	-1	4	10.97	11.17
-4	-1	4	8.81	11.76
4	-1	4	14.89	15.37
-5	-1	4	6.71	6.23
5	-1	4	14.88	15.99

h	K	L	Fobs	Fcalc
-6	-1	4	8.89	6.88
6	-1	4	3.90	6.55
-7	-1	4	23.07	23.40
7	-1	4	3.98	4.20
-8	-1	4	19.76	22.07
-9	-1	4	10.37	12.09
-10	-1	4	5.91	7.34
-11	-1	4	2.70	3.92
0	-2	4	13.90	16.75
-1	-2	4	2.70	2.21
1	-2	4	2.40	2.59
-2	-2	4	22.05	21.41
2	-2	4	33.25	33.92
-3	-2	4	12.45	13.64
3	-2	4	23.65	24.82
-4	-2	4	41.34	41.91
4	-2	4	4.84	4.97
-5	-2	4	24.66	24.99
5	-2	4	4.88	1.22
-6	-2	4	21.82	23.05
6	-2	4	3.46	2.96
-7	-2	4	19.81	21.10
7	-2	4	7.73	9.62
-8	-2	4	4.83	5.06
-9	-2	4	9.87	10.15
-10	-2	4	7.47	10.52
0	-3	4	14.46	12.63
-1	-3	4	18.59	19.12
1	-3	4	2.66	.94
-2	-3	4	32.85	34.48
2	-3	4	19.22	21.48
-3	-3	4	22.62	23.41
3	-3	4	17.24	18.93
-4	-3	4	1.59	3.33
4	-3	4	16.23	16.15
-5	-3	4	8.17	8.48
5	-3	4	8.49	8.49
-6	-3	4	15.45	15.50
6	-3	4	0.	1.77
-7	-3	4	18.21	18.99
-8	-3	4	10.30	11.74
-9	-3	4	4.26	4.07
-10	-3	4	4.59	3.94
0	-4	4	12.67	12.27
-1	-4	4	7.65	6.23
1	-4	4	11.31	11.71
-2	-4	4	41.95	43.42
2	-4	4	9.14	7.51

h	K	L	Fobs	Fcalc
-3	-4	4	17.33	16.36
3	-4	4	8.01	6.84
-4	-4	4	6.30	7.56
4	-4	4	17.77	18.77
-5	-4	4	8.43	8.72
5	-4	4	15.35	16.77
-6	-4	4	2.98	1.98
-7	-4	4	0.	.42
-8	-4	4	12.71	15.44
-9	-4	4	20.91	23.69
0	-5	4	9.31	8.01
-1	-5	4	9.89	9.50
1	-5	4	0.	1.79
-2	-5	4	15.97	17.26
2	-5	4	6.71	8.33
-3	-5	4	11.81	12.28
3	-5	4	5.03	5.75
-4	-5	4	5.71	6.77
-5	-5	4	3.72	4.57
-6	-5	4	4.48	5.08
-7	-5	4	15.94	17.29
0	-6	4	11.41	13.41
-1	-6	4	5.02	5.21
-2	-6	4	6.66	9.17
-3	-6	4	3.18	3.77
-4	-6	4	16.03	17.70
0	0	5	3.51	3.18
-1	0	5	1.39	5.43
1	0	5	16.53	20.32
2	0	5	10.21	11.66
3	0	5	17.09	16.62
-4	0	5	23.80	25.03
4	0	5	2.44	5.36
-5	0	5	2.69	1.98
-6	0	5	10.95	11.37
-7	0	5	4.14	3.21
-8	0	5	2.50	3.85
-9	0	5	10.40	10.70
0	-1	5	10.56	11.00
-1	-1	5	3.04	2.63
1	-1	5	11.72	10.85
-2	-1	5	11.56	10.89
2	-1	5	3.82	4.86
-3	-1	5	1.17	1.79
3	-1	5	5.90	6.76
-4	-1	5	21.25	21.91
4	-1	5	5.73	4.76
-5	-1	5	9.14	9.69

h	K	L	Fobs	Fcalc
−6	−1	5	20.06	21.02
−7	−1	5	18.71	18.88
−8	−1	5	2.33	2.77
−9	−1	5	2.72	4.59
0	−2	5	2.87	2.82
−1	−2	5	23.51	25.49
1	−2	5	4.93	5.78
−2	−2	5	20.11	20.08
2	−2	5	7.89	5.71
−3	−2	5	10.40	10.72
3	−2	5	3.51	3.45
−4	−2	5	1.97	4.69
4	−2	5	4.18	4.54
−5	−2	5	0.	.58
−6	−2	5	2.17	4.67
−7	−2	5	4.79	3.28
−8	−2	5	7.74	8.65
0	−3	5	15.92	18.59
−1	−3	5	3.60	1.02
1	−3	5	15.92	16.87
−2	−3	5	4.24	2.79
2	−3	5	6.78	6.14
−3	−3	5	11.12	11.97
3	−3	5	4.89	3.25
−4	−3	5	23.46	24.94
−5	−3	5	6.47	7.67
−6	−3	5	12.23	13.19
−7	−3	5	11.23	10.65
0	−4	5	6.50	6.13
−1	−4	5	6.16	4.77
1	−4	5	12.66	12.84
−2	−4	5	3.61	2.74
−3	−4	5	0.	4.36
−4	−4	5	12.45	13.55
−5	−4	5	3.51	4.89
−6	−4	5	12.57	12.95