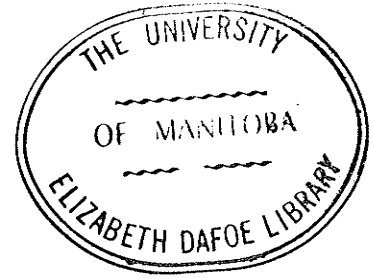


MINERALOGICAL STUDY
AND CRYSTAL STRUCTURE
REFINEMENT OF CANADIAN
SODALITE

by
VERNON A. BARRETT



A Thesis
Submitted to
The Faculty of Graduate Studies and Research
University of Manitoba

In Partial Fulfillment
of the Requirements for the Degree
Master of Science
1968

c Vernon A. Barrett 1968

TO

ELLEN

ABSTRACT

Sodalite specimens from Bancroft, Ontario and Ice River, B.C. have been examined mineralogically, and the crystal structure of the Ice River sodalite has been refined. Both sodalites are shown to have a chemical composition close to $\text{Na}_8\text{Al}_6\text{Si}_6\text{O}_{24}\text{Cl}_2$. The refractive indices, densities, and cell edges of both minerals were found to be the same, namely 1.488 ± 0.001 , 2.31 ± 0.02 g/cc., and $8.878 \pm 0.001 \text{ \AA}$ respectively. Complete indexed powder data are given. The extinction conditions confirmed the previously derived non-centric space group $\overline{P4}3n$.

The crystal structure of Ice River, B.C. sodalite has been refined by Structure Factor Least Squares (SFLS) and Fourier three-dimensional syntheses. Three-dimensional intensity data was collected from Weissenberg photographs using a triple film pack and Mo X-radiation. The intensities were measured by visual comparison with a standard intensity scale, and structure amplitudes were derived by applying the Lorentz-polarization, elongation, and scale factors to each reflection. The observed structure factors were placed on an absolute scale by comparison with calculated structure factor data.

The computer programs used in the refinement were those of Dr. F.R. Ahmed, Division of Pure Physics, National Research Council, modified by the author to take account of the non-centric cubic character of the structure. A comparison is made with the structural results recently published by Löns & Schultz (1967). Using

the atomic parameters and isotropic temperature coefficients of Lons & Schultz the unweighted R-index obtained with the author's intensity data was 0.196. This high value is assumed to be due to the fact that the original intensities were fairly inaccurate. A Fourier and least-squares refinement is used here whereas Lons & Schultz employed only the latter.

An attempt to interpret the Si-Al distribution and the Na coordination in terms of electrostatic charge distribution produced no satisfactory conclusions.

TABLE OF CONTENTS

Chapter	Page
ABSTRACT.....	i
TABLE OF CONTENTS.....	iii
LIST OF TABLES.....	vi
LIST OF ILLUSTRATIONS.....	vii
ACKNOWLEDGEMENTS.....	viii
I. INTRODUCTION.....	1
II. LITERATURE SURVEY.....	4
III. PHYSICAL AND CHEMICAL PROPERTIES.....	6
A. Source of Material.....	6
B. Refractive Index.....	6
C. Density.....	6
D. Chemical Analyses and Cell Content.....	6
IV. X-RAY POWDER DATA.....	11
A. Preparation of the Sample.....	11
B. Lattice Spacings.....	11
C. Refined Cell Edge.....	13
V. SINGLE CRYSTAL PHOTOGRAPHS.....	19
VI. COMPUTATIONAL ASPECTS OF THE STRUCTURE REFINEMENT.....	24
A. Introduction.....	24
B. Program NRC-2: Data Reduction and Tape Generation....	24
(a) General Description.....	24
(b) Application to Sodalite.....	26
C. Program EXPAND.....	27

TABLE OF CONTENTS(continued-2)

Chapter	Page
D. Program NRC-10:Structure Factor Least Squares.....	28
(a) General Description.....	28
(b) Application to Sodalite.....	31
E. Program NRC-8:Fourier.....	32
(a) General Description.....	32
(b) Application to Sodalite.....	35
VII. DATA REDUCTION RESULTS.....	36
VIII. RESULTS AND DISCUSSION OF THE REFINEMENT.....	46
IX. CHARGE DISTRIBUTION CONSIDERATIONS.....	65
A. Introduction.....	65
B. Calculations.....	67
X. DISCUSSION OF THE RESULTS.....	70
APPENDIX I: UPPER LEVEL WEISSENBERG INTENSITY CORRECTION....	71
SUBROUTINE ELONGD.....	71
A. General Description.....	71
B. Listing.....	73
APPENDIX II: SORTING SUBROUTINES.....	74
SUBROUTINES SORTA, SORTB, AND SORTC.....	74
A. Introduction.....	74
B. Subroutine SORTA.....	74
(a) General Description.....	74
(b) Listing.....	75
C. Subroutine SORTB.....	74
(a) General Description.....	74

TABLE OF CONTENTS(continued-3)

Chapter	Page
(b) Listing.....	77
D. Subroutine SORTC.....	76
(a) General Description.....	76
(b) Listing.....	79
APPENDIX III: LISTING OF PROGRAM EXPAND.....	84
A. Main Program.....	84
B. Subroutine SETUP.....	87
C. Subroutine SORGEN.....	88
REFERENCES.....	93

LIST OF TABLES

Table	Page
I. Chemical Analysis of Ice River, B.C. sodalite.....	8
II. Chemical Analysis of Bancroft, Ontario sodalite.....	9
III. d-spacings from Powder Photographs of Ice River, B.C. sodalite.....	14
IV. Results of the Data Reduction and Tape Generation Program	37
V. Positional and Thermal Parameters.....	48
VI. Refined Structure Factors.....	49
VII. Interatomic Distances and Bond Angles.....	70

LIST OF ILLUSTRATIONS

Fig.	Page
1. Powder Photograph of Ice River, B.C. Sodalite.....	12
2. Plot to Minimize Absorption Errors in Determination of Accurate Cell Edge of Ice River, B.C. Sodalite.....	17
3. Zero Level Weissenberg Photograph and Intensity Scale (both in triple film packs) for Ice River, B.C. Sodalite.	20
4. 6th Level $\psi_c[001]$ Weissenberg Photographs of Ice River, B.C. Sodalite.....	21
5. Plot of Undistorted Reciprocal Lattice Resulting from the Reading of Ice River, B.C. Weissenberg Photographs.....	22
6. $\rho_o : (001)$ section at $z = 0.0 \text{ \AA}^o$	58
7. $\rho_o : (001)$ section at $z = 1.211 \text{ \AA}^o$	59
8. $\rho_o : (001)$ section at $z = 1.614 \text{ \AA}^o$	60
9. $\rho_o - \rho_c : (001)$ section at $z = 0.00 \text{ \AA}^o$	61
10. $\rho_o - \rho_c : (001)$ section at $z = 1.211 \text{ \AA}^o$	62
11. $\rho_o - \rho_c : (001)$ section at $z = 1.614 \text{ \AA}^o$	63

ACKNOWLEDGEMENTS

This project was undertaken at the suggestion of Dr. R.B. Ferguson of the Geology Department of the University of Manitoba, and the author was supported in part from Dr. Ferguson's National Research Council grant. The author wishes to express his sincere appreciation to Dr. Ferguson for his patience, understanding, and guidance during the course of this research project.

Grateful acknowledgement is made to Dr. F.R. Ahmed and staff of the National Research Council for providing the programs with which this research was carried out; and for his help in many of the computing difficulties which occurred.

In addition, the author would like to thank the following: Mr. K. Ramlal and Mr. G. Irwin for assistance with the chemical analyses; Dr. D. McBride and Mr. L. Delbaere for their assistance in applying the computer programs to sodalite; K. Schmidt and C. Abraham for their assistance with several of the programs written by the author.

CHAPTER I. INTRODUCTION

This project was undertaken for the purposes of providing additional physical and chemical data on two well known Canadian sodalites, of refining the crystal structure of sodalite, and of attempting to interpret the refined structure in terms of electrostatic charge distribution.

The experimental work was done in the X-Ray Crystallography Laboratory of the Geology Department of the University of Manitoba at the suggestion, and under the direction of Professor R.B. Ferguson. The mineral sodalite, $\text{Na}_4\text{Al}_3\text{Si}_3\text{O}_{12}\text{Cl}$, was chosen because of Ferguson's interest in the silicates, in particular the triclinic feldspars to which cubic sodalite is closely related. Two of the main points of interest in the feldspar and related aluminosilicate structures are:

- (a) The Si-Al distribution or "order-disorder", and
- (b) The coordination of the alkali or alkali-earth cation.

Ferguson (Ferguson, Traill, & Taylor, 1958; Ferguson, 1960) concluded through an ionic interpretation of the structure of the feldspar albite $\text{NaAlSi}_3\text{O}_8$, that:

- (a) Low albite is not fully ordered, but only largely ordered with respect to Si and Al, and
- (b) The Na atom is 6 coordinated.

These conclusions seem to be strongly confirmed by recent results of Gait & Ferguson (1968). Ferguson felt that a refinement of the related feldspathoid mineral sodalite might produce

results which could be interpreted in a similar manner.

The determination of a completely unknown structure involves solving the structure with no previous knowledge of the atomic positions. For a structure refinement, approximate atomic positions from previous work are available, and the refinement involves locating the atoms more accurately in the cell unit. Therefore, a structure refinement is generally less difficult and less time-consuming than a structure determination. We concluded that this might leave time for charge considerations and calculations.

The original solution of the sodalite structure was that of Pauling (1930), in which he stated that Na was 4 coordinated. However, in view of the interpretation of the albite structure by Ferguson and Gait & Ferguson, in which Na was found to be 6 coordinated, this seemed to be an abnormally small coordination for Na. It was with the idea of re-examining this 4 coordination of Na, and of looking more closely at the Si-Al distribution that the refinement of this mineral was undertaken. At the beginning of this project, a search of the literature to September 1967 revealed that no refinement had been published, and so work was begun on the structure at that time. However, in late October 1967 when this work was nicely begun, a refinement of sodalite was published by Löns & Schultz (1967).

At this time a decision was made to proceed with the refinement because:

(a) Löns & Schultz had completed their structure refinement using the least squares method only; they had not done a Fourier analysis.

(b) They had again interpreted the Na as having a coordination of 4.

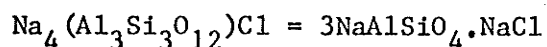
The computer programs used in the present refinement were written by Dr. F.R. Ahmed and his staff of the Division of Pure Physics, National Research Council, Ottawa and kindly provided by him. Considerable difficulty was experienced in applying the programs to sodalite, primarily because they were written for low-symmetry crystal systems whereas the sodalite structure is cubic. This factor, combined with the fact that sodalite is non-centric, created certain difficulties which could be surmounted only by the author writing several subroutines that enabled the programs to cope with a non-centric cubic structure.

The computations were done on the I.B.M. 360/65 computer at the Institute for Computer Studies of the University of Manitoba.

CHAPTER II. LITERATURE SURVEY

Sodalite, named in allusion to its sodium content, was first described by Thomson (1810) in a paper entitled "Chemical Analysis of Sodalite, a New Mineral from Greenland". The first mineralogical paper on sodalite from the Bancroft, Ontario region was by Walker & Parsons (1925), who described, not blue sodalite, but an evanescent pink form later to be called hackmanite. The presence of pink sodalite (hackmanite) in the Ice River, B.C. region was first mentioned by Miser & Glass (1941) who acknowledged the source as a personal communication from A.L. Parsons.

The only chemical analysis of a Canadian sodalite is that of a hackmanite from Bancroft, Ontario published by Walker & Parsons (1925). Analyses of blue sodalite found in the literature were those of specimens from Magnet Cove, Arkansas (Miser & Glass, 1941), Greenland (Thomson, 1810), and Bolivia (Löns & Schultz, 1967). The ideal chemical formula of sodalite is



The X-ray powder diffraction pattern of sodalite was first described by Barth (1926), who stated that the cell edge obtained was $8.85\overset{\circ}{\text{Å}} (=kX)$. The first structure determination of sodalite was that of Pauling (1930). In this publication he stated that sodalite has space group symmetry $T_d^4 (=P\bar{4}3n)$, a cell edge of $8.87\overset{\circ}{\text{Å}} [=kX]$, and a structure in which each unit

cell contains two "molecules" of $\text{Na}_4\text{Si}_3\text{Al}_3\text{O}_{12}\text{Cl}$. Gruner (1931) discussed the structure of sodalite, which he concluded " consists of a framework of $\text{Al}_6\text{Si}_6\text{O}_{24}$ in which tetrahedra of AlO_4 alternate with SiO_4 groups". According to Jaeger (1929) and Pauling (1930), the Na and Cl are inside the framework with each Na linked to three O and one Cl and each Cl to four Na. Barth (1932) reported on the structures of the sodalite family, and stated that sodalite has the space group T_d^1 ($=P\bar{4}3m$), and a cell edge of 8.89\AA ($=kX$). Taylor (1934) discusses the nature and properties of aluminosilicate structures, including the sodalite structure. Kotaczkowska (1935) in a paper on sodalite, gave the following statement: "Crystal of Sodalite from Mt. Vesuvius, Italy, gives by rotation method cubic cell edge 8.89\AA [$=kX$] containing $3\text{Na}_2\text{Al}_2\text{Si}_2\text{O}_8 \cdot 2\text{NaCl}$, space group= T_d^3 ($I\bar{4}3m$)". In the same paper a crystal structure based on Goldschmidt values of interatomic distances is illustrated and described. Lons & Schultz (1967) found sodalite to have space group symmetry $P\bar{4}3n$ with cell edge $8.870\text{\AA} \pm .004\text{\AA}$; and they refined the structure by three-dimensional methods. The most recent report on Canadian sodalite was published by James (1968) where "An Occurrence of Sodalite at the Princess Quarry, Bancroft, Ontario" is described.

CHAPTER III. PHYSICAL AND CHEMICAL PROPERTIES

A. Source of Material

The mineral specimens came from the Mineralogy Museum of the Department of Geology. The Ice River, B.C. blue specimen was macroscopically pure whereas the Bancroft, Ontario specimen contained some nepheline and cancrinite in the blue sodalite.

B. Refractive Index

Using Na-light and the oil immersion method, the refractive index of the Ice River, B.C. and Bancroft, Ontario sodalites was found to be the same, namely:

$$n = 1.488 \pm 0.001$$

C. Density

The densities were determined with the Berman density balance, and two measurements were made on two different specimens of each mineral. The resulting densities of Ice River, B.C. and Bancroft, Ontario sodalites were found to be the same within the limit of error:

$$D = 2.31 \pm 0.02 \text{ grams/cc.}$$

D. Chemical Analyses and Cell Content

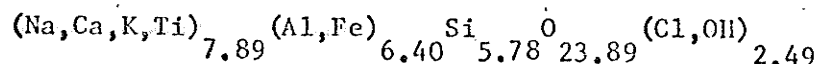
Chemical analyses were performed on both the Ice River, B.C. and Bancroft, Ontario sodalites. Specimens were prepared by crushing about 0.5 grams of sodalite to a maximum particle size

of 4 mm. and removing visible impurities under a binocular microscope. Analyses for Al_2O_3 , Fe (total as Fe_2O_3), MgO, CaO, K_2O , and TiO_2 were done on the X-ray fluorescence spectrometer in the Geology Department of the University of Manitoba by Mr. K. Ramlal. Sodium was determined by Mr. G. Irwin on the atomic absorption unit in the same department. Chlorine and S were determined by wet chemical methods as AgCl and $BaSO_4$ respectively by the author. The total water content was derived by subtraction of the Cl and S contents from the total loss in weight on heating for two hours at $1200^{\circ}C$.

The results of the chemical analyses of the Ice River, B.C. and Bancroft, Ontario sodalites and the corresponding cell contents are listed in the first column of Tables I and II respectively. Both analyses, particularly that of the Ice River specimen, are low in total weight percent. Presumably this is due to small amounts of components such as CO_2 , MgO, F, etc. which were not determined in the chemical analysis.

For Ice River, B.C. sodalite

From Table I the number of atoms in the unit cell is $(Na_{7.84} Ca_{0.04} K_{0.008} Ti_{0.003}) (Al_{6.36} Si_{5.78} Fe_{0.04}) O_{23.89} (Cl_{1.75} OH_{0.74})$ which may be written



This confirms the simplest ideal chemical formula as $Na_4Al_3Si_3O_{12}Cl$, and the cell content as $Z = 2$. The ideal density of the mineral using the ideal cell content (2) and the actual molec-

TABLE I

Sodalite, Ice River, B.C.: Chemical Analysis and Cell Content

	<u>Analysis, Wt. %</u>	<u>"molecules"</u> <u>per unit cell</u>	<u>cations</u> <u>per unit cell</u>		<u>anions</u> <u>per unit cell</u>	
					<u>O</u>	<u>Cl</u>
SiO ₂	35.30	5.78	Si	5.78	11.56	
Al ₂ O ₃	32.92	3.18	Al	6.36	9.54	
Fe (total as Fe ₂ O ₃)	0.34	0.021	Fe	0.04	0.06	
CaO	0.24	0.04	Ca	0.04	0.04	
Na ₂ O	24.70	3.92	Na	7.84	3.92	
K ₂ O	0.04	0.004	K	0.008	0.004	
TiO ₂	0.021	0.003	Ti	0.003	0.006	
Cl	6.30	1.75				1.75
H ₂ O (±)	0.68	0.37			-0.37	0.74 OH
S	0.00					
Less O for Cl	-1.42	-0.87			-.087	
Totals	99.12				0 23.89	Cl, OH 2.49

TABLE II

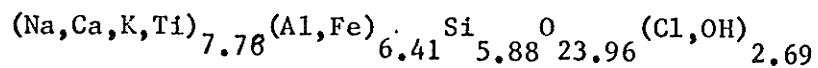
Sodalite, Bancroft, Ontario: Chemical Analysis and Cell Content

	<u>Analysis, Wt. %</u>	<u>"molecules"</u> <u>per unit cell</u>	<u>cations</u> <u>per unit cell</u>		<u>anions</u> <u>per unit cell</u>	
					<u>O</u>	<u>Cl</u>
SiO ₂	36.10	5.88	Si	5.88	11.76	
Al ₂ O ₃	33.20	3.19	Al	6.38	} 6.41	9.57
Fe (total as Fe ₂ O ₃)	0.26	0.016	Fe	0.032		0.048
CaO	0.23	0.04	Ca	0.04	} 7.76	0.04
Na ₂ O	24.45	3.86	Na	7.72		3.86
K ₂ O	0.085	0.009	K	0.018		0.009
TiO ₂	0.024	0.003	Ti	0.003		0.006
Cl	5.31	1.47				1.47
H ₂ O (±)	1.12	0.61			-0.61	1.22 OH
S	0.00					
Less O for Cl	-1.20	-0.73			-0.73	
Totals	<u>99.58</u>				0	<u>23.96</u> Cl, OH 2.69

ular weight of the unit cell as calculated from the results of the chemical analysis is 2.310 grams/cc..

For Bancroft, Ontario sodalite

As above, from Table II the composition is $(\text{Na}_{7.72}\text{Ca}_{0.04}\text{K}_{0.018}\text{Ti}_{0.003})(\text{Al}_{6.38}\text{Si}_{5.88}\text{Fe}_{0.032})\text{O}_{23.96}(\text{Cl}_{1.47}\text{OH}_{1.22})$ which may be written as



The density, as calculated by the previously described method was 2.316 grams/cc..

It is of interest that these two sodalites from different localities have very similar physical and chemical properties.

CHAPTER IV. X-RAY POWDER DATA

A. Preparation of the Sample

A sample of the Ice River, British Columbia sodalite was rolled into a fine collodion rod and mounted in a large (114.6 mm. diameter) Straumanis type Philips powder camera equipped with a fine-focus collimator. The powder photograph which was obtained with Ni-filtered Cu radiation, is shown in Fig. 1.

B. Lattice Spacings

The position of each reflection of the film in Fig. 1 was read to approximately 0.02 mm., and the intensity estimated visually. In the 114.6 mm. diameter camera used, the ideal semi-circumference is 180 mm.. Film shrinkage was corrected for by using the midpoints of the front-and back-reflection regions. If s_1 (mm) is the distance from the center of the front-reflection region to a given reflection, and if X (mm) is the measured distance from the center of the back-reflection region to the center of the front-reflection region, i.e. the effective half-circumference of the camera, then the value of s_2 corrected for film shrinkage, is

$$s_2 = \frac{180}{X} \times s_1 \text{ (mm)}$$

In this way, the lattice spacings (d 's) were corrected for film shrinkage. The d values of the reflections were also calculated by a computer program (NRC-22) which was given the cell edge as

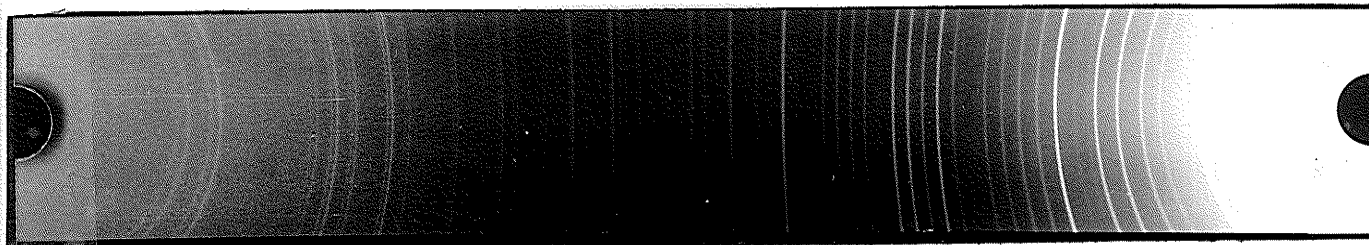


Fig. 1. Powder photograph of the Ice River, B.C. sodalite (Cu/Ni radiation)
Camera diameter 114.6 mm.; contact print.

8.878^oÅ, the refined best value as derived below. Table III contains the d_{obs} , d_{calc} , θ , and intensity of each reflection. The data given in Table III are considerably more complete than those given in the A.S.T.M.X-Ray Powder Data File (1967), Card Number 3-0338.

C. Refined Cell Edge

A refined cell edge was arrived at using eight reflections of low d value which were measured on the powder photograph. Of these eight, six were α_1 and two were α_2 . Their positions in relation to a mm. scale were read by two different people, and a weighted average was taken. Reasonably accurate a_o values were calculated using the cubic spacing formula, and these were then plotted against their corresponding values of the function $1/2(\cos^2\theta/\sin\theta + \cos^2\theta/\theta)$, (Nelson & Riley, (1945) in Nuffield (1966), p.162). This graph, which is drawn in order to minimize absorption errors, is shown in Fig. 2.

The best straight line through these points was calculated by computer, using a least squares curve fitting program of K.L. Schmidt of the Institute for Computer Studies, University of Manitoba. As shown in Fig. 2, the intercept derived from the computations is 8.878^oÅ, and therefore the best cell edge for this specimen of sodalite is taken to be 8.878^oÅ.

E.W. Nuffield (1966) estimates that the accuracy of a cell edge derived in this way is about one part in 10,000; the cell

TABLE III

Sodalite, Ice River, B.C.; X-Ray Powder Data, Cu/Ni Radiation

$a_0 = 8.878\text{\AA}$; space group $P\bar{4}3n$

<u>$d_{\text{obs}}\text{\AA}$</u>	<u>$d_{\text{calc}}\text{\AA}$</u>	<u>$\theta_{\text{obs}} (\text{CuK}\alpha)_1^\circ$</u>	<u>I</u>	<u>hkl</u>
6.3	6.3	7.025	8	110
4.45	4.44	9.97	4	200
3.63	3.62	12.46	10	211
2.81	2.81	15.91	5	310
2.57	2.56	17.47	7	222
2.38	2.37	18.94	7	321
2.22	2,20	20.49	1	400
2.10	2.09	21.57	8	330,411
1.99	1.99	22.80	3	420
1.896	1.892	23.99	3	332
1.814	1.812	25.15	3	422
1.743	1.741	26.25	3	431
1.621	1.621	28.40	3	521
1.569	1.569	29.42	6	440
1.523	1.523	30.41	4	433,530
1.480	1.480	31.40	6	442,600
1.441	1.440	32.35	6	532,611
1.404	1.404	33.30	1/2	620
1.371	1.370	34.22	3	541
1.339	1.338	35.15	3	622

TABLE III (continued-2)

$d_{\text{obs}}^{\text{\AA}}$	$d_{\text{calc}}^{\text{\AA}}$	$\theta(\text{CuK}\alpha)^{\circ}$	I	hkl
1.310	1.309	36.05	3	631
1.281	1.281	36.98	3	444
1.231	1.231	38.78	1/2	640
1.209	1.208	39.61	6	552,633,721
1.188	1.186	40.46	1/2	642
1.167	1.166	41.36	2	730
1.129	1.128	43.08	4	651,732
1.110	1.110	44.01	1/2	800
1.093	1.093	44.80	2	554,741,811
1.078	1.077	45.63	2	644,820
1.019	1.018	49.08	2	662
1.007	1.005	49.93	2	752
0.994	0.993	50.83	3	840
0.982	0.980	51.65	1	833,910
0.969	0.969	52.68	1	842
0.958	0.957	53.53	3	655,761,921
0.936	0.936	55.35	1	754,851,930
0.917	0.916	57.18	2	763,932
0.906	0.906	58.23	2	844
0.8972	0.8968	59.23	1	770,853,941
0.8792	0.8791	61.18	2	772,10.2.2
0.8546	0.8543	64.33	2	666,10.2.2

TABLE III (continued-3)

$d_{\text{obs}} \text{ \AA}$	$d_{\text{calc}} \text{ \AA}$	$\theta (\text{CuK}_{\alpha})^{\circ}$	I	hkl
0.8466	0.8465	65.48	4	765,952,10.3.1
0.8316	0.8315	67.85	2	774,855,871
0.8243	0.8243	69.13	3	864,10.4.0
0.8172	0.8173	70.48	2	961,10.3.3
0.7910	0.7909	76.85	4	963,10.5.1,11.2.1
0.7847	0.7847	78.98	3	880

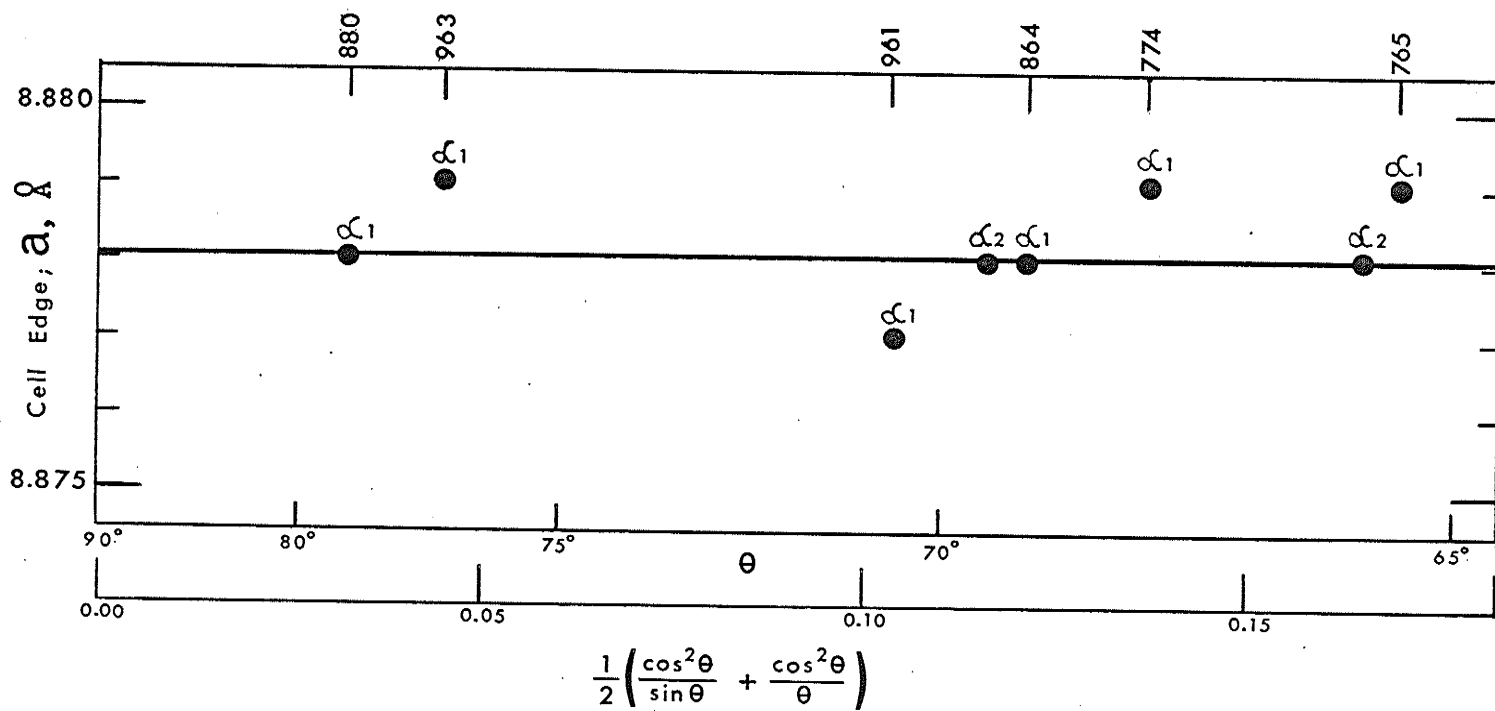


Fig. 2. Sodalite. Plot of apparent a_0 against $\frac{1}{2} \left[\frac{\cos^2 \theta}{\sin \theta} + \frac{\cos^2 \theta}{\theta} \right]$

edge given here is therefore regarded as accurate to $\pm 0.001 \text{ \AA}$.

This compares with $a_0 = 8.870 \pm 0.004 \text{ \AA}$ given by Löns & Schultz (1967) for a specimen from Bolivia.

CHAPTER V. SINGLE-CRYSTAL PHOTOGRAPHS

A crystal of Ice River, B.C. sodalite was first ground to a sphere of approximately 0.4 mm. diameter by using the NONIUS crystal grinder, and this crystal was used for all single-crystal photographs. All photographs were taken with unfiltered Mo X-radiation..

The Buerger Precession instrument was used to orient the crystal about a principal axis, taken as $c[001]$. Several oscillation setting photographs were taken to assure alignment of the crystal prior to taking the Weissenberg equi-inclination photographs, and to provide the settings for the upper level Weissenberg photographs. Zero to 10th level Weissenberg photographs were taken around the $[001]$ axis using the triple film pack method with brass foil separating the films. The 0- and 6- level Weissenberg photographs are shown in Figs. 3 and 4 respectively. The positions of the reflections on the 0- and 1- level Weissenberg photographs were measured and plotted in the usual manner to give the undistorted reciprocal lattice, and to establish the simplest unit cell (Fig. 5). It is important to note that some of the reflections, shown as open circles in Fig. 5, violate the body-centering condition of $h+k+l = 2n$. Therefore, we conclude that the lattice type is "P". The only other systematic condition shown in any of the reflections is hhl present only with $l = 2n$. Therefore, we confirm that for a non-centric structure the space group must be $P\bar{4}3n$.

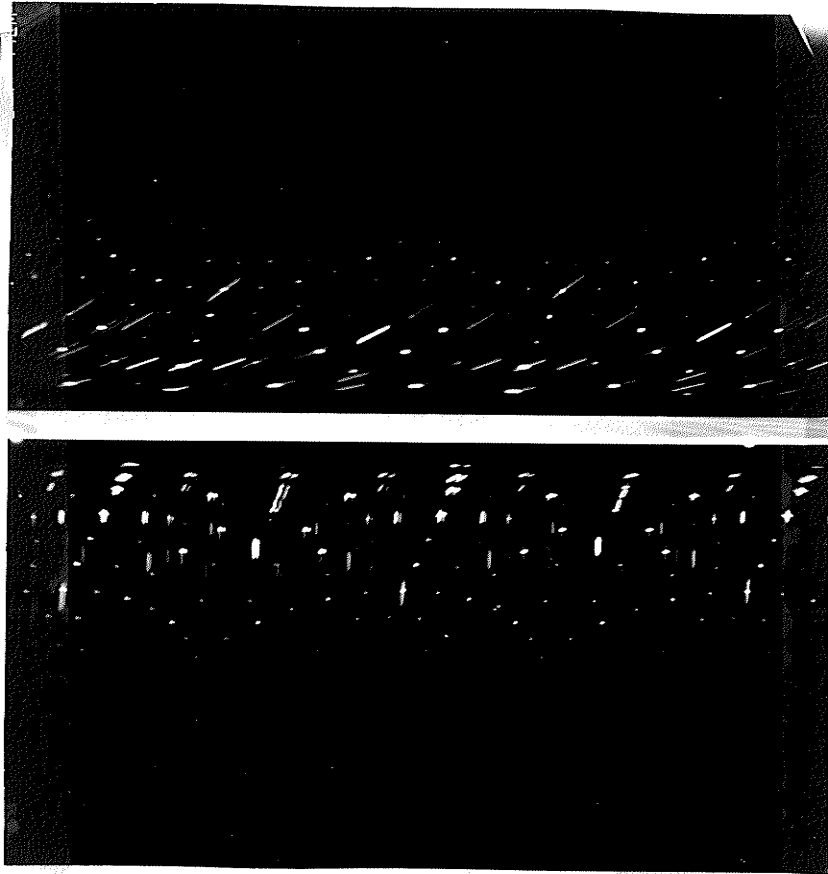


Fig. 4. 6th level Weissenberg photograph of Ice River, B.C. sodalite σ_c [001], Mo/ -, showing the elongation of reflections on the upper half of the film and the contraction of them on the lower half of the film.

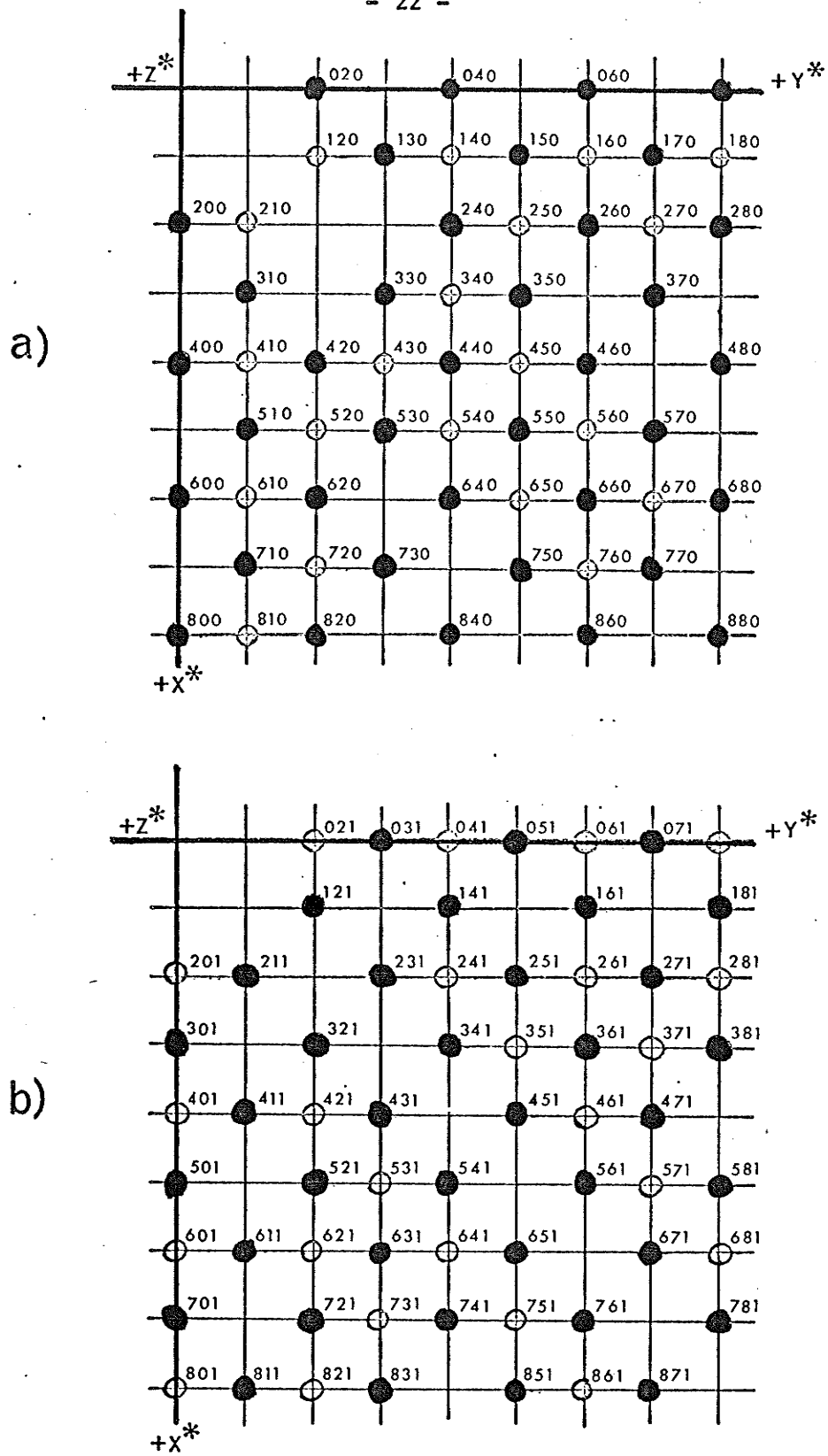


Fig. 5. Plot of position of reflections from zero-(a) and first-(b) level Weissberg photographs to establish the undistorted reciprocal lattice and the simplest unit cell. The open circles represent the reflections which violate the body-centering condition.

The intensities were measured by means of an intensity scale which was constructed using a triple film pack and an isolated reflection (440) photographed for different known periods by the oscillation method. The intensity scale is shown in Fig. 3.

Splitting of some of the reflections was found to occur on lower halves of upper-level photographs; the effect is shown for the 6-level photograph in Fig.4. This is attributed to a slight misalignment in two portions of the crystal. To counteract this effect in reading the intensities of the reflections, they were read on only the top halves of the photographs, and the effect of the elongation on the intensities was taken account of in the computer programs as described in Appendix I.

Seven hundred and thirty-one (731) reflections were observed in all, but many of these are structurally equivalent due to the high symmetry. A scale factor was established for each level of reflections by comparison of exposure times from level to level. The intensities of equivalent reflections were combined into 372 non-equivalent reflections by a computer program which is described in Appendix II.

CHAPTER VI. COMPUTATIONAL ASPECTS OF THE STRUCTURE REFINEMENT

A. Introduction

The programs used by the author for the structure refinement were obtained from Dr. F.R. Ahmed, Division of Pure Physics, National Research Council, Ottawa. Of the series of programs which were available from NRC the ones used were:

1. NRC-2...Data Reduction and Tape Generation
2. NRC-8...Fourier
3. NRC-10...Structure Factor Least Squares (block diagonal)
4. NRC-12...Scan of Interatomic Distances and Angles
5. NRC-22...Calculation of d-Spacings

An additional program (EXPAND) had to be written by the author for reasons which are given below. Because of modifications to the original programs of Ahmed required to adapt them to the present cubic non-centric structure, fairly detailed descriptions of these programs, mainly from Ahmed's reports are given here.

B. Program NRC-2: Data Reduction and Tape Generation (1966) by F.R. Ahmed and C.P. Saunderson.

(a) General Description

This program was written for low symmetry systems and therefore required some alteration by the author. Four subroutines were written to adapt the program to the cubic system, and they are described later. The purpose of this program is to generate, from the lists read from cards, a tape which may be used as input for the Structure Factor Least Squares, Fourier, and several

other programs. Program NRC-2, after modification by the author, carries out the following steps for each reflection hkl :

1. Reads the indices h , k , l , and the observed related intensity.

2. Tests signs of h , k , l , and reverses signs if two (2) or three (3) indices are negative. For this cubic structure, only positive Miller indices were used as input.

3. Determines the "parity" and the "multiplicity". Parity signifies the even/odd combination of the three (3) indices h , k , l , and is for use later in Fourier analyses. The parity is redetermined in subroutine SORTC (written by the author and referred to later) due to the combining and rearranging of the equivalent reflections into a systematic listing of the non-equivalent reflections as necessitated by the cubic symmetry. The multiplicity consists of all possible positive and negative combinations of h , k , l : thus the multiplicity is 2 for $h00$, 4 for $hk0$ and 8 for hkl . The Fourier programs require the multiplicity of each reflection.

4. Calculates $\sin^2\theta$.

5. Interpolates the atomic scattering factor (f) curves to give the f -values for each chemically different atom.

6. Corrects the intensity for the effect of elongation on the top-half of upper level Weissenberg photographs. The author found it necessary to write a subroutine (ELONGD) to carry this out because none was included in the original program. The sub-

routine is described in Appendix I.

7. Applies the Lorentz-polarization and scale corrections to the observed relative intensity to give relative F_o^2 and relative F_o .

8. Stores all data except the h, k, l values on magnetic disk by the direct access method. Calls subroutine SORTA (written by the author) which finds equivalent reflections as they come from NRC-2 and links them together by an indicator which is stored with their h, k, and l values in the computer memory. Subroutine SORTA is described in Appendix II.

9. Returns to Step 1 to read in the next reflection. When all the reflections have been dealt with, the computer applies subroutines SORTB and SORTC, written by the author and described in Appendix II. SORTB averages F_o and F_o^2 of equivalent reflections and stores them under a resultant reflection with $h>k>l$. Subroutine SORTC (referred to previously) sorts all the non-equivalent reflections in increasing order of l, k, and h respectively. It then calculates the proper parity for each reflection and outputs the hkl and associated data on the line printer and the output tape.

(b) Application to Sodalite

A cell edge of 8.878\AA was used by the program. The wavelength of $\text{MoK}\alpha$ X-radiation was taken to be 0.7107\AA and the program was instructed to apply the data to a non-centric cubic primitive lattice. The equivalent positions used were those given in

the International Tables, (1952), Volume I, space group $P\bar{4}3n$, number 218, with O, Al, Si, Na, and Cl having the (i), (c), (d), (e), and (a) positions respectively. The atomic scattering factor (f) curves for Si^{+4} , Al^{+3} , Cl^{-1} , and Na^{+1} were obtained from International Tables, (1959), Volume II, p.202, whereas that for O^{-2} was from Internationale Tabellen, (1935), Volume II, p.57. The program interpolated all of the curves, none of which was corrected for anomalous scattering. The initial parameters inserted were those of Pauling (1930), and the isotropic temperature factors those of Löns & Schultz (1967).

C. Program EXPAND

This program was written by the author to expand the non-equivalent reflections which are on the output tape of NRC-2 (after SORTA, SORTB, and SORTC), into all possible equivalent reflections with positive values of h, k, and l; e.g. the reflection 321 becomes 321, 312, 123, 132, 213, 231. This was necessary because in the space group $P\bar{4}3n$, three of these reflections will differ in phase from the other three when $h+k+l = 2n+1$, thus introducing an error into the Fourier program (NRC-8) if they were all given the same phase. For this reason, the expansion process had to be performed before the SFLS program (NRC-10) which calculates the phase of each reflection. The logic of this program is very similar to that of subroutine SORTA and SORTC which have been described above. Therefore, only a listing of the program is given in Appendix III and the reader is referred to the above-mentioned

subroutines for the description of the logic involved.

D. Program NRC-10: Structure Factor Least Squares (SFLS) by

F.R. Ahmed

(a) General Description

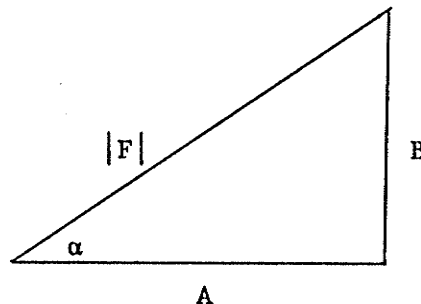
The purpose of this program is three-fold:

1. To calculate the structure factors from a given set of temperature factors, atomic scattering factor (f) values (used in NRC-2), and positional parameters for each atom.
2. To refine, by block-diagonal least squares approximation, the positional parameters, the isotropic or anisotropic temperature factor, and the occupational factor for each atom (if desired). The program also calculates an overall scale factor which converts the calculated structure factors (F_c) to the scale of the observed structure amplitudes $|F_o|$.
3. To calculate the estimated standard deviations of the refined parameters (if desired).

The structure factor equation may best be described as the sum of a real and an imaginary term:

$$F(hkl) = A(hkl) + iB(hkl)$$

or, vectorially



Where:

A = the sum over all atoms of $f \cos \alpha$ for a given reflection,

B = the sum over all atoms of $f \sin \alpha$ for the same reflection,

f being the atomic scattering factor corrected for atomic vibration, and

α = the phase angle = $\tan^{-1} \left(\frac{B}{A} \right)$.

Therefore $|F|$ may also be written as

$$|F| = (A^2 + B^2)^{1/2} .$$

The least squares portion of the program minimizes the quantity R, where

$$\begin{aligned} R &= \Sigma (|F_o| - |F_c|)^2 \\ &= \Sigma (\Delta F)^2, \end{aligned}$$

Σ = sums over all non-equivalent (orthorhombic) reflections,

and

$|F_c|$ = calculated structure amplitude on the scale of $|F_o|$.

If p_j ($j=1, \dots, N$) are the N parameters in $|F_c|$, then R is a minimum when these parameters satisfy the N equations such that

$$\frac{\delta R}{\delta p_j} = 0 \quad j=1, \dots, N$$

The purpose of the least squares refinement is therefore to arrive at a set of parameters p_j , $j=1, N$ such that R is a minimum. Weighting factors may be applied to the $|F_o|$ values to take account of the differences in accuracy of the original intensities; this might accelerate the minimizing of R.

The block-diagonal approximation form of matrix calculation is used to reduce the amount of calculation, and to allow for the refinement of a large number of parameters simultaneously by ignoring the interactions between the parameters of different atoms.

The program executes the following steps:

1. Calculates the structure factor for a given reflection by computing the A and B contributions of each prototype atom and all its equivalent atoms in the cell, yielding A_c , B_c , F_c , and

$$\Delta F = |(|F_c| - |F_o|)|.$$

2. Calculates (relative) A observed (A_o) and B observed (B_o) by using F_o and the calculated phase angle, α .

3. Prints out hkl , $\sin^2\theta$, F_o , F_c , A_o , B_o , A calculated (A_c), B calculated (B_c), F , and $100 \times \frac{\Delta F}{|F_o|}$ for a given reflection on the line printer.

4. Calculates for the least squares refinement, the contributions to the derivatives for a given reflection considering in turn all the parameters of each non-equivalent atom in the cell.

5. Accumulates the contributions of the reflection to the overall sums.

6. Repeats steps 1 to 5 for the remaining non-equivalent (orthorhombic) reflections.

7. Calculates shifts to be applied to the present parameters in order to minimize the R value, and computes the estimated standard deviation of these shifts. The new positional and thermal parameters are then arrived at by summing the shifts

and adding this sum to the original parameter to give a refined parameter.

8. Calculates scale factor to bring $|F_c|$ to the scale of $|F_o|$ by summing all of the $|F_o|$'s and all of the $|F_c|$'s and comparing them.

9. Calculates the R value using the old parameters and prints this out on the printer. The R value using the new scale and the overall temperature factor correction is also calculated and printed at this point. As well, the new parameters, old scale, shifts, and estimated standard deviations are printed.

10. Repeats, on order, steps 1 to 8 with the new parameters and the new scale.

11. Recycles a programmed number of times and then terminates.

(b) Application to Sodalite

The program was instructed to apply the data given to a non-centric structure. The output tape of the program NRC-2 was the main source of data input to this program. No weighting schemes or "fudge" factors, provision for which exists in the program, were used in the refinement. The program was instructed to refine the temperature factors (isotropic) for all of the atoms, the positional parameters x, y, z for O^{-2} , and x, x, x for Na^{+1} . A slight alteration was made to the program so that, for Na, the shifts which are calculated by the program are equal for all three parameters.

An attempt was made to refine the structure by treating the thermal motion of the atoms as anisotropic but the program produced unreasonable results.

E. Program NRC-8: Fourier for Distorted and Undistorted Nets

by F.R. Ahmed

(a) General Description

The purpose of this program is to compute Fourier summations along lines parallel to the unit-cell axes at specified fractions of the cell edge.

The general form of the expression for computing the electron density (ρ) at any point XYZ using a Fourier summation is:

$$\rho(XYZ) = \frac{1}{V} \sum_h \sum_k \sum_l F(hkl) e^{-2\pi i (hX+kY+lZ)}$$

where $h, k,$ and l are Miller indices,

$F(hkl)$ is the structure factor of the plane hkl ,

and V is the volume of the unit cell in \AA^3 .

The particular form of this equation which must be used for input to Program NRC-8 is:

$$\rho(XYZ) = \frac{1}{V} \left\{ F(000) + \sum_h \sum_k \sum_l \left[\sum_{ij} R_{ij}(hkl) \cdot \text{trg} 2\pi hX \cdot \text{trg} 2\pi kY \cdot \text{trg} 2\pi lZ + \sum_{ij} S_{ij}(hkl) \cdot \text{trg} 2\pi hX \cdot \text{trg} 2\pi kY \cdot \text{trg} 2\pi lZ \right] \right\}$$

where $F(000)$ is the total number of electrons in the unit-cell,

trg is a cosine or sine,

$$R_{ij}(hkl) = m \cdot \left[\pm A(hkl) \pm A(\bar{h}kl) \pm A(h\bar{k}l) \pm A(hk\bar{l}) \right], \text{ or}$$

$$m. [\pm A(hkl) \pm A(\bar{h}kl)], \text{ or}$$

$$m. [\pm A(hkl) \pm A(hk\bar{l})], \text{ or}$$

$$m. [\pm A(hkl)],$$

$S_{ij}(hkl)$ = similar expressions to R_{ij} for the imaginary parts
 $B(hkl)$,

m = "multiplicity" (1, 2, 4, or 8), and

\sum_{ij} = summation over the number of terms ($i=1, 2, \text{ or } 4$) in each
 group (j).

Some clarification is desired at this point. The "multiplicity" as explained in Chapter VI, Section B is the total number of positive and negative combinations of the three Miller indices h , k , and l . The terms A and B in the above expression are those of the structure factor expression

$$F(hkl) = A(hkl) + iB(hkl).$$

For a centric structure, the B 's will all be zero and the A terms may be equally well represented as F terms. In the expression $R_{ij}(hkl) = m. \pm A(hkl) \dots$, the programmer puts in the terms $A(hkl)$, $A(\bar{h}kl)$, and so on only if a reflection such as hkl or $\bar{h}kl$ exists in the data which the program will use. Only in a low-symmetry system such as the monoclinic does a reflection such as $\bar{h}kl$ have no equivalent such as hkl ; in contrast, in a high symmetry system such as the cubic there is always a reflection of the type hkl which is equivalent in amplitude to the reflection $\bar{h}kl$. Thus in

the case of cubic sodalite all reflections can be expressed in positive indices and hence the form of the expression $R_{ij}(hkl)$ was $m \pm A(hkl)$. Each A or B is called a "term". However, the electron density expression is sometimes able to be simplified if it is known that of h, k, and l, h and l are even, or that some similar condition holds. For this reason, each reflection has been given a "parity" in NRC-2 which tells whether hkl, the indices of a given reflection, are even, even, even, or odd, even, even, or something similar. The "parity" is used in this program, NRC-8, to save time in the Fourier summations. It is for this reason that the programmer may, depending on the space group, have to assign a certain "group" to each "parity" and give a special form of the Fourier expression for each group in this program.

The program is capable of performing Fourier summations using three different kinds of input data as terms in the series: F_o , F_o^2 , or $|F_o - F_c| = \Delta F$. An F_o synthesis produces an electron density map, an F_o^2 synthesis, a Patterson or vector map, and an $(F_o - F_c)$ synthesis an electron difference map.

The program executes the following steps:

1. Reads a reflection and associated data from the input tape which is the output tape of the SFLS (NRC-10) program.

2. Tests the "parity" of that reflection and calculates the pertinent A (or F) and B terms which apply to the group to which the parity refers.

3. Applies the proper multiplicity to these coefficients.
4. Stores the results in a core memory.
5. Repeats steps 1 to 4 for all remaining reflections.
6. Performs the Fourier summations for one level (Z) of the unit cell over a given range of (X) and (Y) at a given interval along the axes (say $\frac{a}{4}$).
7. Outputs the $\rho(XYZ)$ values for this level on tape or on printer or on both.
8. Repeats steps 6 and 7 as many times as needed to cover the levels desired.

(b) Application to Sodalite

For sodalite, the electron density expression for space group $P\bar{4}3n$ in volume I of the International Tables for X-Ray Crystallography (1952) is

$$\rho(XYZ) = \frac{8}{V_c} \sum_0^{\infty} \sum_0^{\infty} \sum_0^{\infty} \left[|F(hkl)| \left[\cos 2\pi hX \cos 2\pi kY \cos 2\pi lZ \cos \alpha_{hkl} - \sin 2\pi hX \sin 2\pi kY \sin 2\pi lZ \sin \alpha_{hkl} \right] \right]$$

which, on substitution of $\cos \alpha_{hkl} = \frac{A(hkl)}{|F(hkl)|}$

$$\text{and } \sin \alpha_{hkl} = \frac{B(hkl)}{|F(hkl)|}$$

$$\rho(XYZ) = \frac{8}{V_c} \sum_0^{\infty} \sum_0^{\infty} \sum_0^{\infty} \left[A(hkl) \cos 2\pi hX \cos 2\pi kY \cos 2\pi lZ - B(hkl) \sin 2\pi hX \sin 2\pi kY \sin 2\pi lZ \right]$$

This expression was derived from, and applies to, the ortho-

rhombic system. Therefore, in order to apply it to the cubic system, the non-equivalent reflections must be expanded to the equivalent reflections thus in effect treating the data as orthorhombic. A more detailed description was previously given in the discussion of Program EXPAND, written by the author.

All reflections were given the same parity because no simplification of this expression occurs if h, k, and l are all even; or if h and k are even; and so on.

The form of this expression as it is used in the program is

$$\rho(XYZ) = \frac{1}{V} \left\{ F(000) + \sum_{h=0}^{\infty} \sum_{k=0}^{\infty} \sum_{l=0}^{\infty} \left[m.A(hkl) \cos^{\frac{2\pi}{\lambda}} hX \cos^{\frac{2\pi}{\lambda}} kY \cos^{\frac{2\pi}{\lambda}} lZ - m.B(hkl) \sin^{\frac{2\pi}{\lambda}} hX \sin^{\frac{2\pi}{\lambda}} kY \sin^{\frac{2\pi}{\lambda}} lZ \right] \right\}.$$

CHAPTER VII. DATA REDUCTION RESULTS

Program NRC-2 (Data Reduction and Tape Generation) produced the results shown in Table IV. In this table the values listed for each reflection are:

(a) H, K, L: the three Miller indices h, k, and l.

(b) SINSQ: the value of $\sin^2\theta$ where θ is the angle of diffraction of the X-rays from the plane hkl.

(c) IOBS: the original relative intensity after it has been corrected for elongation of the reflections read on the upper half of the Weissenberg photographs.

(d) FOBS-SQ: the resultant amplitude (F_o^2) after the original intensity has been corrected for elongation, Lorentz-polarization, and scale effects.

(e) FOBS: $|F_o|$, the square root of (d).

These data are then used as tape input to program NRC-10 (Structure Factor Least Squares).

TABLE IV

RESULTS OF THE DATA REDUCTION AND TAPE GENERATION PROGRAM

H	K	L	SINSQ	IOBS	FUBS-SQ	FUBS
1	1	0	0.0032	0.0	0.0	0.0
2	0	0	0.0064	175.00	28.40	5.33
2	1	0	0.0080	5.83	1.02	1.01
2	1	1	0.0096	639.68	73.18	8.55
2	2	0	0.0128	0.0	0.0	0.0
2	2	2	0.0192	596.91	138.54	11.77
3	1	0	0.0160	109.80	28.10	5.30
3	2	0	0.0208	1.41	0.23	0.48
3	2	1	0.0224	158.36	41.29	6.43
3	3	0	0.0288	440.00	156.63	12.52
3	3	2	0.0352	84.48	27.23	5.18
4	0	0	0.0256	90.00	30.06	5.48
4	1	0	0.0272	1.03	0.35	0.60
4	1	1	0.0288	574.11	202.88	14.24
4	2	0	0.0320	111.56	30.09	5.48
4	2	1	0.0336	1.22	0.36	0.60
4	2	2	0.0384	107.61	31.66	5.63
4	3	0	0.0401	1.19	0.39	0.63
4	3	1	0.0417	35.85	11.87	3.43
4	3	2	0.0465	3.15	1.19	1.08
4	3	3	0.0545	100.13	40.11	6.33
4	4	0	0.0513	537.17	219.81	14.82
4	4	2	0.0577	188.10	81.28	9.01
4	4	4	0.0769	303.77	154.65	12.44
5	1	0	0.0417	3.58	1.56	1.25
5	2	0	0.0465	1.05	0.47	0.68
5	2	1	0.0481	30.01	11.03	3.26
5	3	0	0.0545	75.65	31.32	5.49
5	3	1	0.0561	1.20	0.48	0.69
5	3	2	0.0609	131.41	59.54	7.52
5	4	0	0.0657	1.18	0.52	0.72
5	4	1	0.0673	51.30	24.41	4.83
5	4	2	0.0721	1.19	0.57	0.75
5	4	3	0.0801	2.77	1.43	1.19
5	5	0	0.0801	1.00	0.64	0.80

TABLE IV (continued-2)

H	K	L	SINSQ	IOBS	FOBS-SQ	FOBS
5	5	2	0.0865	35.28	20.31	4.46
5	5	4	0.1057	6.99	4.47	2.08
6	0	0	0.0577	299.00	157.04	12.53
6	1	0	0.0593	1.02	0.55	0.74
6	1	1	0.0609	16.67	9.11	3.02
6	2	0	0.0641	18.17	9.88	3.14
6	2	1	0.0657	1.06	0.59	0.77
6	2	2	0.0705	160.20	61.04	7.75
6	3	1	0.0737	85.31	36.89	6.07
6	3	2	0.0785	1.21	0.58	0.76
6	3	3	0.0865	139.97	66.10	8.13
6	4	0	0.0833	26.94	13.21	3.63
6	4	1	0.0849	1.19	0.62	0.78
6	4	2	0.0897	9.20	4.46	2.02
6	4	3	0.0977	1.19	0.69	0.83
6	4	4	0.1089	16.90	10.13	3.18
6	5	0	0.0977	1.61	0.87	0.92
6	5	1	0.0993	42.96	24.80	4.94
6	5	2	0.1041	1.18	0.71	0.84
6	5	3	0.1121	5.91	3.75	1.91
6	5	4	0.1234	1.18	0.80	0.90
6	5	5	0.1378	24.58	17.34	4.06
6	6	0	0.1153	1.00	0.81	0.90
6	6	2	0.1218	92.71	61.40	7.76
6	6	4	0.1410	7.59	5.78	2.40
6	6	6	0.1730	25.84	22.27	4.72
7	1	0	0.0801	2.04	1.31	1.14
7	2	0	0.0849	1.04	0.67	0.82
7	2	1	0.0865	64.94	42.72	6.53
7	3	0	0.0929	90.21	45.70	6.73
7	3	1	0.0945	1.74	0.81	0.90
7	3	2	0.0993	22.93	12.49	3.53
7	4	1	0.1057	20.99	12.28	3.49
7	4	2	0.1105	1.20	0.74	0.86
7	4	3	0.1186	2.66	1.90	1.26
7	5	0	0.1186	2.69	1.80	1.33
7	5	1	0.1202	1.18	0.78	0.88
7	5	2	0.1250	14.87	9.78	3.10
7	5	3	0.1330	1.18	0.85	0.92
7	5	4	0.1442	6.69	5.14	2.25
7	6	0	0.1362	1.17	0.84	0.92
7	6	1	0.1378	6.71	4.87	2.21
7	6	2	0.1426	1.18	0.88	0.94
7	6	3	0.1506	10.97	8.65	2.92
7	6	4	0.1618	1.18	0.98	0.99
7	6	5	0.1762	9.83	8.67	2.93
7	7	0	0.1570	9.61	8.16	2.86

TABLE IV (continued-3)

H	K	L	SINSQ	IOBS	FOBS-SQ	FOBS
7	7	2	0.1634	11.87	10.61	3.24
7	7	4	0.1826	14.86	14.40	3.76
7	7	6	0.2147	8.14	8.58	2.93
8	0	0	0.1025	70.00	52.26	7.23
8	1	0	0.1041	1.02	0.77	0.88
8	1	1	0.1057	22.68	17.51	4.18
8	2	0	0.1089	63.28	35.59	5.75
8	2	2	0.1153	16.49	8.22	2.80
8	3	1	0.1186	3.89	2.58	1.55
8	3	2	0.1234	1.23	0.75	0.86
8	3	3	0.1314	23.60	15.60	3.81
8	4	0	0.1282	129.56	106.32	10.30
8	4	2	0.1346	9.90	6.91	2.60
8	4	3	0.1426	1.20	0.86	0.92
8	4	4	0.1538	42.79	33.01	5.59
8	5	1	0.1442	2.37	1.72	1.31
8	5	3	0.1570	1.63	1.19	1.08
8	5	4	0.1682	1.18	0.98	0.99
8	5	5	0.1826	6.07	4.99	2.23
8	6	0	0.1602	1.17	0.92	0.96
8	6	1	0.1618	1.17	0.94	0.97
8	6	2	0.1666	1.18	0.96	0.98
8	6	3	0.1746	1.18	1.00	1.00
8	6	4	0.1858	15.44	14.36	3.73
8	6	6	0.2179	8.81	8.87	2.96
8	7	1	0.1826	5.74	5.38	2.28
8	7	3	0.1955	1.57	1.49	1.20
8	7	5	0.2211	1.93	2.08	1.42
8	7	6	0.2387	1.54	1.74	1.30
8	7	7	0.2595	1.76	2.05	1.42
8	8	0	0.2051	26.03	28.51	5.14
8	8	2	0.2115	3.93	4.14	2.02
8	8	4	0.2307	24.61	28.58	5.16
8	8	6	0.2627	2.87	3.47	1.85
8	8	8	0.3076	3.47	4.59	2.14
9	1	0	0.1314	2.03	1.80	1.34
9	2	0	0.1362	1.36	0.74	0.85
9	2	1	0.1378	22.60	11.55	3.38
9	3	1	0.1458	1.26	0.82	0.90
9	3	2	0.1506	10.67	6.84	2.61
9	4	1	0.1570	7.47	5.22	2.28
9	4	2	0.1618	1.22	0.91	0.95
9	4	3	0.1698	3.26	2.51	1.58
9	5	0	0.1698	7.77	5.66	2.34
9	5	2	0.1762	19.60	15.10	3.83
9	5	4	0.1955	1.19	1.08	1.04
9	6	1	0.1890	10.73	9.26	3.04

TABLE IV (continued-4)

H	K	L	SINSQ	IOBS	FOBS-SQ	FOBS
9	6	3	0.2019	9.88	9.11	3.02
9	6	5	0.2275	1.98	1.99	1.40
9	7	0	0.2083	1.16	1.13	1.06
9	7	1	0.2099	1.17	1.15	1.07
9	7	2	0.2147	3.83	4.02	1.97
9	7	4	0.2339	2.76	2.90	1.70
9	7	6	0.2659	5.09	5.93	2.42
9	8	0	0.2323	1.16	1.20	1.10
9	8	3	0.2467	5.80	6.43	2.50
9	8	5	0.2724	1.57	1.82	1.33
9	9	2	0.2659	3.39	4.12	2.03
9	9	4	0.2852	1.74	2.14	1.45
9	9	6	0.3172	2.29	3.13	1.77
9	9	8	0.3621	2.29	3.38	1.84
10	1	0	0.1618	1.01	1.04	1.02
10	1	1	0.1634	14.34	14.89	3.86
10	2	0	0.1666	6.98	3.65	1.90
10	2	1	0.1682	1.54	0.76	0.85
10	2	2	0.1730	35.27	18.80	4.29
10	3	1	0.1762	9.78	6.52	2.54
10	3	2	0.1810	1.26	0.95	0.97
10	3	3	0.1890	3.41	2.19	1.48
10	4	0	0.1858	23.54	17.65	4.19
10	4	2	0.1922	1.22	1.02	1.00
10	4	4	0.2115	4.47	3.60	1.90
10	5	1	0.2019	4.77	4.29	2.05
10	5	3	0.2147	5.70	5.12	2.25
10	5	4	0.2259	1.92	2.06	1.40
10	5	5	0.2403	4.32	4.09	2.02
10	6	0	0.2179	3.15	2.97	1.72
10	6	2	0.2243	22.17	21.65	4.64
10	6	4	0.2435	2.80	2.87	1.69
10	6	6	0.2756	4.70	5.52	2.31
10	7	1	0.2403	4.29	4.62	2.14
10	7	3	0.2531	9.78	10.96	3.30
10	7	5	0.2788	2.76	3.28	1.81
10	7	7	0.3172	1.79	2.27	1.49
10	8	0	0.2627	3.07	3.49	1.86
10	8	2	0.2691	1.59	1.74	1.31
10	8	4	0.2884	2.33	2.88	1.69
10	8	6	0.3204	1.57	2.03	1.41
10	8	8	0.3653	2.32	3.34	1.83
10	9	0	0.2900	1.00	1.55	1.24
10	9	1	0.2916	2.31	2.86	1.69
10	9	3	0.3044	3.48	4.43	2.10
10	9	5	0.3300	2.36	3.06	1.72
10	9	7	0.3685	2.70	3.99	1.99

TABLE IV (continued-5)

H	K	L	SINSQ	IOBS	FOBS-SQ	FOBS
10	9	9	0.4197	2.30	3.57	1.89
10	10	0	0.3204	1.72	2.26	1.49
10	10	2	0.3268	4.49	6.32	2.51
10	10	4	0.3460	2.86	4.08	2.02
10	10	6	0.3781	3.41	5.21	2.28
10	10	8	0.4229	1.14	1.83	1.35
10	10	10	0.4806	4.55	7.41	2.72
11	1	0	0.1955	1.00	1.16	1.08
11	2	0	0.2003	1.04	1.21	1.10
11	2	1	0.2019	3.10	3.67	1.92
11	3	0	0.2083	3.60	4.30	2.07
11	3	2	0.2147	13.71	16.53	4.07
11	4	1	0.2211	4.73	5.90	2.42
11	4	3	0.2339	1.07	1.36	1.16
11	5	2	0.2403	2.15	2.73	1.65
11	5	4	0.2595	1.09	1.45	1.20
11	6	0	0.2515	1.06	1.39	1.18
11	6	1	0.2531	8.58	11.34	3.37
11	6	3	0.2659	1.09	1.46	1.21
11	6	5	0.2916	2.76	3.87	1.96
11	7	0	0.2724	2.15	3.00	1.73
11	7	2	0.2788	1.66	2.31	1.50
11	7	4	0.2980	2.21	3.25	1.80
11	7	6	0.3300	2.23	3.44	1.86
11	8	1	0.2980	1.09	1.56	1.25
11	8	3	0.3108	1.10	1.60	1.27
11	8	5	0.3364	2.22	3.37	1.83
11	8	7	0.3749	1.67	2.80	1.64
11	9	0	0.3236	3.27	4.78	2.19
11	9	2	0.3300	3.31	4.88	2.21
11	9	4	0.3493	1.11	1.70	1.30
11	9	6	0.3813	1.12	1.78	1.33
11	9	8	0.4262	1.12	1.86	1.36
11	10	1	0.3557	1.10	1.70	1.30
11	10	3	0.3685	1.12	1.73	1.31
11	10	5	0.3941	2.24	3.58	1.89
11	10	7	0.4326	1.70	2.81	1.66
11	10	9	0.4838	1.68	2.83	1.66
11	11	0	0.3877	1.00	1.86	1.36
11	11	2	0.3941	1.03	1.90	1.38
11	11	4	0.4133	1.05	1.96	1.40
11	11	6	0.4454	1.08	1.97	1.40
11	11	8	0.4902	1.10	1.97	1.40
11	11	10	0.5479	1.12	1.85	1.36
12	0	0	0.2307	26.00	34.10	5.84
12	1	0	0.2323	1.01	1.34	1.16
12	1	1	0.2339	2.04	2.75	1.66

TABLE IV (continued-6)

H	K	L	SINSQ	IOBS	FOBS-SQ	FOBS
12	2	0	0.2371	2.56	3.38	1.83
12	2	2	0.2435	8.32	11.11	3.33
12	3	1	0.2467	2.08	2.84	1.69
12	4	0	0.2563	17.55	24.32	4.91
12	4	2	0.2627	3.69	5.15	2.26
12	4	4	0.2820	11.81	17.10	4.13
12	5	1	0.2724	2.11	3.02	1.74
12	5	3	0.2852	1.62	2.32	1.50
12	5	5	0.3108	2.17	3.27	1.81
12	6	0	0.2884	4.73	6.91	2.62
12	6	2	0.2948	1.63	2.35	1.51
12	6	4	0.3140	2.17	3.30	1.82
12	7	1	0.3108	1.07	1.68	1.30
12	7	3	0.3236	1.09	1.72	1.31
12	7	7	0.3877	1.11	1.95	1.40
12	8	0	0.3332	4.72	7.56	2.70
12	8	2	0.3396	2.17	3.40	1.84
12	8	4	0.3589	4.89	8.05	2.79
12	8	6	0.3909	1.11	1.84	1.36
12	8	8	0.4358	3.33	5.73	2.39
12	9	1	0.3621	1.09	1.75	1.32
12	9	5	0.4005	1.68	2.72	1.63
12	9	7	0.4390	1.11	1.96	1.40
12	9	9	0.4902	1.12	1.90	1.38
12	10	0	0.3909	1.67	2.62	1.60
12	10	2	0.3973	1.03	1.91	1.38
12	10	4	0.4165	1.69	2.73	1.64
12	10	8	0.4934	1.11	1.91	1.38
12	11	1	0.4262	1.01	2.01	1.42
12	11	3	0.4390	1.04	2.00	1.41
12	11	7	0.5031	1.08	2.12	1.46
12	11	9	0.5543	2.20	3.77	1.94
12	12	0	0.4614	4.00	7.96	2.82
12	12	2	0.4678	1.02	2.01	1.42
12	12	4	0.4870	2.09	4.09	2.02
12	12	6	0.5191	1.07	2.00	1.41
12	12	8	0.5639	1.08	1.93	1.39
13	1	0	0.2724	2.02	3.02	1.74
13	2	1	0.2788	3.60	5.40	2.32
13	3	0	0.2852	1.05	1.57	1.25
13	3	2	0.2916	1.57	2.38	1.52
13	4	1	0.2980	1.02	1.63	1.28
13	4	3	0.3108	1.06	1.66	1.29
13	5	0	0.3108	1.09	1.64	1.28
13	5	2	0.3172	2.12	3.33	1.83
13	5	4	0.3364	1.07	1.74	1.32
13	6	1	0.3300	2.12	3.43	1.85

TABLE IV (continued-8)

H	K	L	SINSQ	IOBS	FOBS-SQ	FOBS
14	9	7	0.5223	1.65	2.96	1.70
14	10	0	0.4742	2.21	3.77	1.88
14	10	2	0.4806	2.10	3.96	1.91
14	10	6	0.5319	2.13	3.98	2.00
14	10	10	0.6344	1.09	1.70	1.30
14	11	1	0.5095	1.01	2.07	1.44
14	11	3	0.5223	1.03	2.03	1.42
14	11	5	0.5479	1.05	1.99	1.41
14	11	9	0.6376	1.08	1.72	1.31
14	12	4	0.5703	1.04	1.98	1.41
14	12	6	0.6024	1.06	1.88	1.37
15	1	0	0.3621	2.01	3.65	1.91
15	2	1	0.3685	3.06	5.56	2.36
15	3	2	0.3813	1.04	1.87	1.37
15	4	3	0.4005	1.04	1.93	1.39
15	5	2	0.4069	2.05	3.85	1.96
15	6	3	0.4326	1.60	2.94	1.69
15	7	2	0.4454	2.05	3.98	2.00
15	8	3	0.4774	1.04	2.03	1.43
15	9	2	0.4966	1.02	2.02	1.42
15	9	6	0.5479	1.06	1.98	1.41
15	10	1	0.5223	1.01	2.06	1.44
15	10	3	0.5351	1.03	2.02	1.42
15	11	6	0.6120	1.05	1.86	1.36
15	12	5	0.6312	1.04	1.81	1.35
16	0	0	0.4101	6.00	11.48	3.39
16	2	2	0.4229	1.03	1.96	1.40
16	3	1	0.4262	1.53	2.99	1.70
16	4	0	0.4358	2.55	4.94	2.21
16	4	2	0.4422	1.03	1.99	1.41
16	4	4	0.4614	3.15	6.09	2.47
16	5	1	0.4518	1.01	2.04	1.43
16	6	0	0.4678	2.07	3.99	2.00
16	6	2	0.4742	1.02	2.02	1.42
16	7	0	0.4886	1.00	2.01	1.42
16	8	0	0.5127	5.00	10.03	3.17
16	8	2	0.5191	1.02	2.02	1.42
16	10	0	0.5703	1.00	1.95	1.40
16	10	2	0.5767	1.02	1.95	1.40
16	11	1	0.6056	1.01	1.93	1.39
16	12	0	0.6408	2.00	3.57	1.89
17	2	1	0.4710	1.02	2.01	1.42
17	3	2	0.4838	1.03	2.03	1.42
17	4	0	0.4886	1.00	2.01	1.42
17	4	3	0.5031	1.03	2.04	1.43
17	5	2	0.5095	1.02	2.02	1.42
17	7	2	0.5479	2.04	3.99	2.00

TABLE IV (continued-9)

H	K	L	SINSQ	IOBS	FOBS-SQ	FOBS
17	8	0	0.5655	1.00	1.96	1.40
17	9	0	0.5928	2.00	3.81	1.95
17	9	2	0.5992	1.02	1.90	1.38
18	2	2	0.5319	1.02	2.01	1.42
18	6	2	0.5832	1.02	1.94	1.39
18	10	0	0.6793	2.00	3.32	1.82
19	2	1	0.5864	1.02	1.93	1.39
20	0	0	0.6408	2.00	3.57	1.89

CHAPTER VIII. RESULTS AND DISCUSSION OF THE REFINEMENT

Isotropic temperature factors were used in all stages of the refinement. A least squares refinement was attempted starting with Pauling's positional parameters and Löns & Schultz's isotropic temperature factors. Refinement by this method caused the positional parameters to approach those of Löns & Schultz; however, repeated refinements showed that it was impossible to lower the R-index below 0.20. This suggested that the intensity data were fairly inaccurate. The R-index obtained with the author's intensity data and the positional and thermal parameters of Löns & Schultz was 0.196. Further refinements resulted in only small shifts in the parameters that gave somewhat higher R-indices. This confirmed that the author's intensity data were inaccurate. (Their intensities were measured photometrically while the author's were done visually.)

Electron density (ρ_o) and difference ($\rho_o - \rho_c$) maps were computed using the data^{of} Löns & Schultz (1967). Sections through one atom of each type in the structure are shown in Fig. 6, 7, 8, 9, 10, and 11.

Within the limits of this work the results appear to confirm Löns & Schultz's positional and temperature factors (Table V).

In Table VI are given the structure factor data using the author's intensities and Löns & Schultz's parameters. This table contains the FO, FC, AO, BO, AC, BC, and ALPHAC (calculated phase

angle) for each reflection HKL.

The Program NRC-12, Scan of Interatomic Distances and Bond Angles by M.E. Pippy and F.R. Ahmed, given the parameters of Löns & Schultz (Table V) and the author's cell edge, produced the results shown in Table VII.

TABLE V
Positional and Thermal Parameters

<u>Atom</u>	<u>Position</u>	<u>Pauling's Positional Parameters (1930)</u>	<u>Löns & Schultz (1967) Positional Parameters</u>	<u>Isotropic Temperature Factors, Å²</u>
Cl	0,0,0,			1.90
Na	x,x,x,	x=0.175	x=0.1777	1.49
Si	1/4,0,1/2			0.35
Al	1/4,1/2,0			0.80
O	x,y,z,	x=0.135 y=0.440 z=0.150	x=0.1401 y=0.4385 z=0.1487	0.89

TABLE VI

Refined Structure Factor After Execution of Structure Factor

Least Squares (SFLS) Program

H	K	L	FO	FC	AO	BO	AC	BC	ALPHAC
1	1	0	0.0	4.94	0.0	0.0	-4.94	0.0	180.00
2	0	0	5.33	4.34	5.33	-0.00	4.34	-0.00	360.00
2	1	0	1.01	0.75	-1.01	-0.00	-0.75	-0.00	180.00
2	1	1	8.55	11.89	7.32	-4.43	10.17	-6.16	328.78
2	2	0	0.0	0.04	0.0	0.0	0.04	-0.00	360.00
2	2	2	11.77	12.52	-10.21	5.86	-10.86	6.24	150.12
3	1	0	5.30	4.47	-5.30	0.00	-4.47	0.00	180.00
3	2	0	0.48	0.19	0.48	0.00	0.19	0.00	0.00
3	2	1	6.43	6.19	6.12	-1.94	5.90	-1.87	342.38
3	3	0	12.52	11.23	12.52	0.00	11.23	0.00	0.00
3	3	2	5.18	4.57	4.30	-2.89	3.80	-2.55	326.17
4	0	0	5.48	5.02	5.48	0.00	5.02	0.00	0.00
4	1	0	0.60	0.25	0.60	0.00	0.25	0.00	0.00
4	1	1	14.24	9.24	3.50	13.81	2.27	8.96	75.78
4	2	0	5.48	5.27	5.48	0.00	5.27	0.00	0.00
4	2	1	0.60	0.24	-0.17	0.57	-0.07	0.23	106.33
4	2	2	5.63	4.88	-1.34	5.47	-1.16	4.74	103.73
4	3	0	0.63	0.20	-0.63	0.00	-0.20	0.00	180.00
4	3	1	3.43	3.35	-3.08	1.52	-3.01	1.48	153.74
4	3	2	1.08	0.66	0.09	-1.08	0.05	-0.66	274.66
4	3	3	6.33	5.02	-5.95	2.15	-4.72	1.71	160.12
4	4	0	14.82	14.61	14.82	-0.00	14.61	-0.00	360.00
4	4	2	9.01	8.61	6.66	-6.07	6.36	-5.80	317.67
4	4	4	12.44	11.80	11.16	5.48	10.59	5.20	26.17
5	1	0	1.25	1.16	1.25	0.0	1.16	0.0	0.0
5	2	0	0.68	0.19	-0.68	0.00	-0.19	0.00	180.00
5	2	1	3.26	3.66	3.25	-0.27	3.65	-0.30	355.30
5	3	0	5.49	5.82	-5.49	-0.00	-5.82	-0.00	180.00
5	3	1	0.69	0.46	0.58	0.38	0.38	0.25	33.67
5	3	2	7.52	7.57	7.52	0.10	7.57	0.10	0.72
5	4	0	0.72	0.24	-0.72	0.0	-0.24	0.0	180.00
5	4	1	4.83	4.98	-2.80	-3.94	-2.89	-4.06	234.57
5	4	2	0.75	0.65	-0.73	0.20	-0.63	0.17	164.97
5	4	3	1.19	0.93	-0.49	-1.09	-0.38	-0.85	245.88
5	5	0	0.80	0.50	0.80	0.00	0.50	0.00	0.00

TABLE VI (continued-2)

H	K	L	FD	FC	AO	BO	AC	BC	ALPHAC
5	5	2	4.46	4.76	2.75	3.51	2.94	3.74	51.89
5	5	4	2.08	2.29	-1.58	1.35	-1.74	1.49	139.46
6	0	0	12.53	11.30	12.53	0.00	11.30	0.00	0.00
6	1	0	0.74	0.23	0.74	-0.00	0.23	-0.00	360.00
6	1	1	3.02	3.10	2.90	0.83	2.98	0.85	15.88
6	2	0	3.14	3.05	-3.14	0.00	-3.05	0.00	180.00
6	2	1	0.77	0.34	-0.25	-0.72	-0.11	-0.32	250.81
6	2	2	7.75	7.29	-7.50	-1.97	-7.05	-1.85	194.70
6	3	1	6.07	5.25	5.40	2.77	4.67	2.39	27.15
6	3	2	0.76	0.10	-0.56	0.52	-0.07	0.07	137.20
6	3	3	8.13	7.49	8.01	1.38	7.38	1.27	9.74
6	4	0	3.63	3.48	3.63	0.0	3.48	0.0	0.0
6	4	1	0.78	0.71	0.75	-0.23	0.68	-0.21	342.71
6	4	2	2.02	2.23	-1.98	-0.40	-2.18	-0.44	191.45
6	4	3	0.83	0.67	0.81	0.15	0.66	0.13	10.75
6	4	4	3.18	2.99	3.17	0.30	2.97	0.28	5.35
6	5	0	0.92	0.91	-0.92	0.0	-0.91	0.0	180.00
6	5	1	4.94	5.62	4.93	0.23	5.61	0.26	2.67
6	5	2	0.84	0.10	-0.77	-0.34	-0.09	-0.04	203.87
6	5	3	1.91	1.73	1.25	-1.45	1.13	-1.31	310.82
6	5	4	0.90	0.56	-0.49	-0.75	-0.31	-0.47	236.73
6	5	5	4.06	4.67	3.87	-1.20	4.46	-1.38	342.75
6	6	0	0.90	0.31	0.90	0.00	0.31	0.00	0.00
6	6	2	7.76	7.08	-7.60	-1.58	-6.93	-1.44	191.73
6	6	4	2.40	2.01	-0.72	2.29	-0.60	1.92	107.46
6	6	6	4.72	4.82	-4.53	1.30	-4.64	1.33	163.95
7	1	0	1.14	1.00	-1.14	-0.00	-1.00	-0.00	180.00
7	2	0	0.82	0.46	0.82	0.0	0.46	0.0	0.0
7	2	1	6.53	6.21	5.51	-3.52	5.24	-3.34	327.44
7	3	0	6.73	5.77	-6.73	-0.00	-5.77	-0.00	180.00
7	3	1	0.90	0.29	-0.43	0.79	-0.14	0.25	118.55
7	3	2	3.53	3.39	3.52	0.20	3.38	0.19	3.20
7	4	1	3.49	3.32	-0.96	3.36	-0.91	3.19	105.90
7	4	2	0.86	0.70	-0.75	-0.41	-0.62	-0.34	208.63
7	4	3	1.26	0.49	0.85	-0.93	0.33	-0.36	312.36
7	5	0	1.33	1.27	-1.33	0.00	-1.27	0.00	180.00
7	5	1	0.88	0.44	0.59	0.66	0.30	0.33	48.25
7	5	2	3.10	3.68	2.67	1.58	3.17	1.87	30.62
7	5	3	0.92	0.44	-0.25	0.89	-0.12	0.42	105.93
7	5	4	2.25	2.40	-1.44	-1.73	-1.54	-1.85	230.25
7	6	0	0.92	0.47	0.92	0.0	0.47	0.0	0.0
7	6	1	2.21	2.39	2.17	-0.42	2.35	-0.46	348.95
7	6	2	0.94	0.41	-0.16	-0.93	-0.07	-0.40	260.06
7	6	3	2.92	2.76	2.88	0.52	2.72	0.49	10.31
7	6	4	0.99	0.32	0.98	-0.14	0.31	-0.05	351.66
7	6	5	2.93	3.06	2.93	0.14	3.06	0.15	2.76
7	7	0	2.86	2.52	2.86	-0.00	2.52	-0.00	360.00

TABLE VI (continued-3)

H	K	L	FO	FC	AO	BO	AC	BC	ALPHAC
7	7	2	3.24	3.68	2.85	-1.55	3.23	-1.75	331.50
7	7	4	3.76	3.44	-3.23	1.93	-2.95	1.76	149.22
7	7	6	2.93	2.88	2.87	-0.58	2.83	-0.57	348.62
8	0	0	7.23	6.48	7.23	-0.00	6.48	-0.00	360.00
8	1	0	0.88	0.43	-0.88	-0.00	-0.43	-0.00	180.00
8	1	1	4.18	4.28	-3.69	-1.98	-3.77	-2.02	208.25
8	2	0	5.75	6.31	5.75	0.00	6.31	0.00	0.00
8	2	2	2.80	3.01	-2.64	0.94	-2.84	1.01	160.40
8	3	1	1.55	1.45	1.25	-0.93	1.16	-0.87	323.28
8	3	2	0.86	0.86	0.81	0.30	0.80	0.30	20.34
8	3	3	3.81	4.32	-3.71	-0.87	-4.20	-0.98	193.15
8	4	0	10.30	9.40	10.30	0.00	9.40	0.00	0.00
8	4	2	2.60	2.73	2.04	1.62	2.13	1.70	38.51
8	4	3	0.92	0.19	0.13	-0.92	0.03	-0.19	278.26
8	4	4	5.59	6.38	5.31	-1.75	6.06	-1.99	341.79
8	5	1	1.31	1.57	-1.30	0.14	-1.56	0.17	173.65
8	5	3	1.08	0.43	-0.48	0.97	-0.19	0.39	116.05
8	5	4	0.99	0.13	0.59	-0.79	0.08	-0.11	306.49
8	5	5	2.23	2.58	-1.84	1.26	-2.13	1.46	145.61
8	6	0	0.96	0.34	0.96	0.00	0.34	0.00	0.00
8	6	1	0.97	0.50	-0.97	0.03	-0.50	0.02	178.26
8	6	2	0.98	0.55	0.94	-0.28	0.53	-0.16	343.29
8	6	3	1.00	0.20	-0.86	-0.51	-0.17	0.10	149.46
8	6	4	3.73	4.10	3.68	-0.61	4.04	-0.67	350.56
8	6	6	2.96	3.16	-2.81	-0.90	-3.01	-0.97	197.81
8	7	1	2.28	2.37	-2.01	-1.07	-2.09	-1.11	208.05
8	7	3	1.20	0.77	-1.20	-0.03	-0.77	-0.02	181.63
8	7	5	1.42	1.04	-1.01	1.00	-0.74	0.73	135.31
8	7	6	1.30	0.91	1.28	-0.25	0.89	-0.17	349.09
8	7	7	1.42	1.05	-1.19	-0.77	-0.88	-0.57	212.78
8	8	0	5.14	6.20	5.14	0.00	6.20	0.00	0.00
8	8	2	2.02	2.10	2.01	0.25	2.09	0.26	7.20
8	8	4	5.16	5.96	5.11	0.75	5.90	0.86	8.31
8	8	6	1.85	1.87	1.83	0.25	1.85	0.26	7.90
8	8	8	2.14	2.69	2.14	-0.01	2.69	-0.02	359.66
9	1	0	1.34	1.25	-1.34	0.0	-1.25	0.0	180.00
9	2	0	0.85	0.74	0.85	0.00	0.74	0.00	0.00
9	2	1	3.38	3.72	3.02	1.52	3.32	1.67	26.74
9	3	1	0.90	0.43	-0.84	-0.31	-0.40	-0.15	200.20
9	3	2	2.61	2.60	2.48	-0.83	2.47	-0.82	341.52
9	4	1	2.28	2.05	-2.28	-0.07	-2.06	-0.06	181.78
9	4	2	0.95	0.42	-0.89	0.34	-0.39	0.15	158.98
9	4	3	1.58	1.34	-0.83	1.34	-0.71	1.14	121.92
9	5	0	2.34	2.64	-2.34	0.00	-2.64	0.00	180.00
9	5	2	3.83	4.56	3.04	-2.33	3.62	-2.78	322.48
9	5	4	1.04	0.60	-0.95	0.41	-0.55	0.24	156.61
9	6	1	3.04	2.97	2.84	1.09	2.77	1.06	20.91

TABLE VI (continued-4)

H	K	L	FD	FC	AO	BO	AC	BC	ALPHAC
9	6	3	3.02	2.99	2.92	0.77	2.90	0.76	14.75
9	6	5	1.40	1.17	1.40	-0.02	1.17	-0.02	359.09
9	7	0	1.06	0.70	-1.06	-0.00	-0.70	-0.00	180.00
9	7	1	1.07	0.38	1.04	0.25	0.37	0.09	13.29
9	7	2	1.97	1.99	1.93	0.38	1.95	0.39	11.20
9	7	4	1.70	1.56	-1.61	-0.56	-1.47	-0.52	199.33
9	7	6	2.42	2.47	2.36	0.53	2.41	0.54	12.64
9	8	0	1.10	0.54	-1.10	-0.00	-0.54	-0.00	180.00
9	8	3	2.50	2.45	-2.43	-0.59	-2.38	-0.58	193.61
9	8	5	1.33	1.17	-0.54	-1.22	-0.47	-1.07	246.35
9	9	2	2.03	1.70	1.83	0.88	1.53	0.74	25.73
9	9	4	1.45	0.98	-0.82	1.19	-0.56	0.81	124.69
9	9	6	1.77	1.76	1.77	0.03	1.76	0.03	0.83
9	9	8	1.84	1.42	-1.81	0.35	-1.40	0.27	169.06
10	1	0	1.02	0.71	-1.02	0.0	-0.71	0.0	180.00
10	1	1	3.86	4.05	3.77	0.82	3.95	0.86	12.31
10	2	0	1.90	1.71	-1.90	0.00	-1.71	0.00	180.00
10	2	1	0.85	0.35	0.33	0.79	0.14	0.32	66.98
10	2	2	4.29	4.70	-4.20	0.90	-4.60	0.98	167.96
10	3	1	2.54	2.63	2.36	-0.94	2.45	-0.97	338.34
10	3	2	0.97	0.29	0.24	-0.94	0.07	-0.28	284.30
10	3	3	1.48	1.42	1.45	-0.30	1.39	-0.29	348.16
10	4	0	4.19	4.22	4.19	-0.00	4.22	-0.00	360.00
10	4	2	1.00	0.94	-0.60	-0.80	-0.56	-0.75	232.98
10	4	4	1.90	2.02	1.69	0.86	1.80	0.91	26.88
10	5	1	2.05	2.29	2.00	-0.46	2.23	-0.52	346.92
10	5	3	2.25	2.31	2.16	0.64	2.21	0.65	16.45
10	5	4	1.40	0.64	-1.05	0.93	-0.48	0.43	138.58
10	5	5	2.02	2.35	1.97	0.44	2.29	0.52	12.67
10	6	0	1.72	1.23	-1.72	-0.00	-1.23	-0.00	180.00
10	6	2	4.64	4.83	-4.55	0.94	-4.73	0.98	168.36
10	6	4	1.69	1.48	-1.24	-1.16	-1.08	-1.01	223.07
10	6	6	2.31	2.96	-2.29	-0.32	-2.94	-0.41	187.89
10	7	1	2.14	2.18	1.91	0.97	1.94	0.98	26.78
10	7	3	3.30	3.55	3.29	-0.34	3.53	-0.37	354.06
10	7	5	1.81	1.83	1.72	-0.55	1.74	-0.56	342.20
10	7	7	1.49	0.87	-0.57	1.38	-0.33	0.80	112.21
10	8	0	1.86	1.70	1.86	0.00	1.70	0.00	0.00
10	8	2	1.31	1.19	-1.30	0.16	-1.18	0.14	173.05
10	8	4	1.69	1.35	1.69	0.00	1.35	0.00	0.16
10	8	6	1.41	0.74	-0.72	1.22	-0.38	0.64	120.50
10	8	8	1.83	1.94	1.82	-0.12	1.94	-0.13	356.30
10	9	0	1.24	0.43	-1.24	-0.00	-0.43	-0.00	180.00
10	9	1	1.69	1.44	1.30	-1.08	1.10	-0.92	320.08
10	9	3	2.10	2.13	2.09	-0.27	2.11	-0.28	352.52
10	9	5	1.72	1.58	1.64	0.50	1.51	0.46	17.06
10	9	7	1.99	1.10	1.79	-0.88	0.99	-0.48	333.86

TABLE VI (continued-5)

H	K	L	FO	FC	AO	BO	AC	BC	ALPHAC
10	9	9	1.89	1.39	1.89	-0.07	1.39	-0.05	357.90
10	10	0	1.49	0.96	1.49	0.00	0.96	0.00	0.00
10	10	2	2.51	2.95	-2.39	-0.78	-2.80	-0.92	198.19
10	10	4	2.02	1.84	-1.76	0.99	-1.60	0.90	150.56
10	10	6	2.28	2.35	-2.28	-0.03	-2.35	-0.03	180.73
10	10	8	1.35	0.82	-1.09	-0.79	-0.66	-0.48	215.89
10	10	10	2.72	3.01	-2.70	0.31	-2.99	0.34	173.49
11	1	0	1.08	0.23	1.08	0.00	0.23	0.00	0.00
11	2	0	1.10	0.30	1.10	-0.00	0.30	-0.00	360.00
11	2	1	1.92	1.66	1.57	1.10	1.36	0.96	35.06
11	3	0	2.07	1.80	-2.07	-0.00	-1.80	-0.00	180.00
11	3	2	4.07	3.76	4.02	0.63	3.72	0.58	8.85
11	4	1	2.42	2.31	-2.11	-1.19	-2.02	-1.13	209.27
11	4	3	1.16	0.74	-0.89	-0.75	-0.56	-0.48	220.39
11	5	2	1.65	1.67	1.64	-0.22	1.65	-0.23	352.18
11	5	4	1.20	1.15	-0.85	0.85	-0.82	0.82	135.02
11	6	0	1.18	0.83	1.18	-0.00	0.83	-0.00	360.00
11	6	1	3.37	3.42	3.29	-0.72	3.34	-0.73	347.65
11	6	3	1.21	0.83	0.61	-1.04	0.42	-0.72	300.48
11	6	5	1.96	2.10	1.91	0.43	2.05	0.46	12.59
11	7	0	1.73	1.77	-1.73	0.00	-1.77	0.00	180.00
11	7	2	1.50	1.40	1.41	0.50	1.32	0.47	19.48
11	7	4	1.80	0.70	-1.60	-0.84	-0.62	-0.33	207.69
11	7	6	1.86	1.73	1.85	-0.08	1.73	-0.08	357.39
11	8	1	1.25	0.75	-0.51	1.14	-0.30	0.68	113.92
11	8	3	1.27	0.86	-1.04	0.72	-0.71	0.49	145.38
11	8	5	1.83	1.50	-1.79	-0.38	-1.47	-0.31	191.97
11	8	7	1.64	0.34	-1.28	1.04	-0.26	0.21	140.92
11	9	0	2.19	1.89	-2.19	0.00	-1.89	0.00	180.00
11	9	2	2.21	2.16	2.21	0.13	2.15	0.13	3.39
11	9	4	1.30	0.47	-0.02	-1.30	-0.01	-0.47	268.97
11	9	6	1.33	0.64	0.49	-1.24	0.23	-0.59	291.52
11	9	8	1.36	0.79	-1.27	0.49	-0.73	0.28	158.99
11	10	1	1.30	0.99	1.26	0.32	0.96	0.24	14.13
11	10	3	1.31	0.89	1.20	0.53	0.82	0.36	23.66
11	10	5	1.89	1.40	1.88	-0.21	1.39	-0.16	353.60
11	10	7	1.66	1.55	1.66	-0.05	1.55	-0.05	358.24
11	10	9	1.66	1.19	1.59	0.47	1.14	0.34	16.38
11	11	0	1.36	0.17	-1.36	-0.00	-0.17	-0.00	180.00
11	11	2	1.38	1.34	1.27	-0.54	1.24	-0.52	337.09
11	11	4	1.40	0.53	-0.93	1.04	-0.36	0.40	131.87
11	11	6	1.40	0.96	1.21	0.71	0.83	0.48	30.19
11	11	8	1.40	1.45	-1.36	-0.36	-1.41	-0.37	194.66
11	11	10	1.36	0.89	1.32	-0.35	0.86	-0.23	345.26
12	0	0	5.84	5.91	5.84	0.0	5.91	0.0	0.0
12	1	0	1.16	0.46	1.16	0.00	0.46	0.00	0.00
12	1	1	1.66	1.11	-1.06	-1.27	-0.71	-0.85	230.23

TABLE VI (continued-6)

H	K	L	FO	FC	AO	BO	AC	BC	ALPHAC
12	2	0	1.83	1.54	1.83	0.00	1.54	0.00	0.00
12	2	2	3.33	2.64	-0.70	-3.26	-0.56	-2.58	257.82
12	3	1	1.69	1.45	-1.52	0.73	-1.31	0.62	154.51
12	4	0	4.91	4.69	4.91	0.00	4.69	0.00	0.00
12	4	2	2.26	2.09	2.20	0.54	2.03	0.50	13.84
12	4	4	4.13	4.33	4.13	-0.13	4.33	-0.14	358.13
12	5	1	1.74	1.55	-0.68	1.60	-0.61	1.43	113.20
12	5	3	1.50	1.32	-1.49	-0.19	-1.31	-0.17	187.40
12	5	5	1.81	2.46	-0.34	-1.78	-0.46	-2.42	259.29
12	6	0	2.62	2.23	2.62	-0.00	2.23	-0.00	360.00
12	6	2	1.51	1.27	-1.49	0.25	-1.26	0.21	170.36
12	6	4	1.82	1.61	1.59	0.88	1.40	0.78	29.09
12	7	1	1.30	0.86	-1.18	-0.53	-0.78	-0.35	204.13
12	7	3	1.31	0.68	-1.21	0.50	-0.63	0.26	157.49
12	7	7	1.40	0.92	-1.26	-0.60	-0.83	-0.40	205.52
12	8	0	2.70	3.20	2.70	-0.00	3.20	-0.00	360.00
12	8	2	1.84	2.06	1.48	-1.10	1.66	-1.22	323.52
12	8	4	2.79	3.40	2.79	-0.02	3.40	-0.03	359.52
12	8	6	1.36	0.89	1.36	-0.04	0.89	-0.03	358.12
12	8	8	2.39	2.91	2.36	-0.42	2.87	-0.51	349.96
12	9	1	1.32	0.98	-1.30	-0.25	-0.96	-0.19	190.95
12	9	5	1.63	1.44	-1.00	1.28	-0.89	1.14	128.03
12	9	7	1.40	0.71	-1.19	0.74	-0.60	0.37	148.19
12	9	9	1.38	0.90	-0.43	-1.31	-0.28	-0.85	252.06
12	10	0	1.60	1.41	1.60	-0.00	1.41	-0.00	360.00
12	10	2	1.38	0.77	-1.21	0.67	-0.67	0.37	150.85
12	10	4	1.64	1.18	1.37	-0.90	0.99	-0.65	326.81
12	10	8	1.38	0.80	1.32	0.40	0.76	0.23	17.00
12	11	1	1.42	0.52	-1.33	-0.48	-0.49	-0.18	199.97
12	11	3	1.41	0.99	-1.32	-0.50	-0.93	-0.35	200.53
12	11	7	1.46	0.51	-1.45	-0.16	-0.51	-0.06	186.36
12	11	9	1.94	0.64	-1.91	-0.32	-0.63	-0.10	189.34
12	12	0	2.82	2.60	2.82	0.00	2.60	0.00	0.00
12	12	2	1.42	1.40	0.76	1.20	0.75	1.18	57.46
12	12	4	2.02	2.23	2.01	0.20	2.22	0.22	5.68
12	12	6	1.41	1.06	1.28	-0.59	0.97	-0.44	335.34
12	12	8	1.39	1.90	1.31	0.45	1.79	0.62	19.09
13	1	0	1.74	1.38	-1.74	0.00	-1.38	0.00	180.00
13	2	1	2.32	2.29	2.31	-0.17	2.29	-0.17	355.87
13	3	0	1.25	0.98	-1.25	-0.00	-0.98	-0.00	180.00
13	3	2	1.52	1.36	1.23	0.90	1.10	0.80	36.18
13	4	1	1.28	0.18	-1.19	-0.46	-0.17	-0.06	200.88
13	4	3	1.29	1.16	-0.98	-0.84	-0.88	-0.76	220.65
13	5	0	1.28	0.92	-1.28	-0.00	-0.92	-0.00	180.00
13	5	2	1.83	1.61	1.73	0.58	1.53	0.51	18.51
13	5	4	1.32	0.90	-1.31	0.19	-0.89	0.13	171.68
13	6	1	1.85	1.33	1.01	-1.56	0.72	-1.12	302.91

TABLE VI (continued-7)

H	K	L	FO	FC	AO	BO	AC	BC	ALPHAC
13	6	3	1.87	2.03	1.79	-0.54	1.95	-0.59	343.17
13	6	5	1.63	1.45	1.44	0.75	1.29	0.67	27.34
13	7	2	1.91	1.65	1.87	-0.37	1.62	-0.32	348.69
13	7	4	1.38	0.96	-1.24	-0.60	-0.87	0.42	154.35
13	7	6	1.68	1.02	1.54	-0.69	0.93	-0.41	335.94
13	8	0	1.34	0.37	1.34	-0.00	0.37	-0.00	360.00
13	8	1	1.35	1.24	-1.32	0.31	-1.21	0.28	166.84
13	8	3	1.36	0.76	-0.84	1.07	-0.47	0.60	128.23
13	8	7	1.42	0.85	-1.42	-0.09	-0.85	-0.05	183.45
13	9	0	1.38	0.11	-1.38	-0.00	-0.11	-0.00	180.00
13	9	2	1.36	0.92	1.24	0.57	0.84	0.39	24.77
13	9	4	1.95	1.34	-1.55	-1.19	-1.06	-0.82	217.59
13	9	6	1.96	1.43	1.88	-0.58	1.37	-0.43	342.75
13	9	8	1.39	0.49	-0.95	1.02	-0.33	0.36	133.03
13	10	1	1.95	1.48	1.64	1.06	1.24	0.81	32.92
13	10	3	1.95	1.27	1.94	0.24	1.26	0.16	7.00
13	10	5	1.39	0.99	1.12	-0.82	0.80	-0.58	323.99
13	10	7	1.41	0.60	0.98	1.01	0.42	0.43	45.77
13	10	9	1.36	0.49	1.32	0.32	0.48	0.12	13.57
13	11	2	1.42	0.77	1.28	-0.62	0.69	-0.33	334.24
13	11	4	1.43	0.87	-1.29	0.61	-0.78	0.37	154.71
13	11	6	2.00	1.18	1.75	0.96	1.03	0.57	28.67
13	11	10	1.31	0.63	1.16	-0.62	0.56	-0.30	331.84
13	12	1	1.44	0.69	-1.09	-0.94	-0.52	-0.45	220.83
13	12	7	1.43	0.61	-1.26	-0.67	-0.54	-0.29	208.13
14	0	0	2.86	2.67	2.86	-0.00	2.67	-0.00	360.00
14	1	1	1.85	0.87	1.53	-1.03	0.72	-0.49	325.97
14	2	2	3.69	3.55	-3.66	-0.48	-3.52	-0.46	187.44
14	3	1	1.87	1.96	1.85	-0.25	1.94	-0.27	352.20
14	3	3	1.88	1.57	1.87	-0.24	1.56	-0.20	352.73
14	4	2	1.33	1.23	-1.27	0.41	-1.17	0.38	162.03
14	4	4	1.92	1.57	1.91	-0.24	1.55	-0.19	352.93
14	5	1	1.90	1.35	1.81	0.59	1.28	0.42	18.16
14	5	3	1.92	1.68	1.90	0.26	1.67	0.23	7.72
14	5	5	1.94	0.83	1.76	-0.83	0.75	-0.35	334.60
14	6	0	1.90	1.01	-1.90	0.00	-1.01	0.00	180.00
14	6	2	1.92	1.93	-1.89	0.35	-1.90	0.35	169.46
14	6	4	1.37	0.48	-1.16	-0.73	-0.41	-0.25	211.95
14	6	6	2.42	3.05	-2.40	-0.29	-3.03	-0.37	186.94
14	7	1	1.40	1.19	1.40	-0.09	1.19	-0.08	356.34
14	7	5	1.42	1.05	1.41	0.16	1.05	0.12	6.30
14	7	7	2.05	2.15	2.05	0.01	2.15	0.01	0.23
14	8	0	1.95	1.50	1.95	-0.00	1.50	-0.00	360.00
14	8	4	1.40	1.21	1.38	0.25	1.19	0.22	10.28
14	8	6	1.41	0.67	-1.29	0.56	-0.62	0.27	156.45
14	9	1	1.97	1.58	1.93	-0.40	1.54	-0.32	348.20
14	9	5	1.40	1.05	1.35	0.36	1.01	0.27	14.83

TABLE VI (continued-8)

H	K	L	FO	FC	AO	BO	AC	BC	ALPHAC
14	9	7	1.70	0.99	1.70	-0.09	0.99	-0.05	356.94
14	10	0	1.88	1.80	-1.88	-0.00	-1.80	-0.00	180.00
14	10	2	1.91	2.12	-1.91	-0.09	-2.12	-0.10	182.78
14	10	6	2.00	1.97	-1.99	0.18	-1.96	0.18	174.77
14	10	10	1.30	0.79	-1.29	-0.16	-0.78	-0.10	187.05
14	11	1	1.44	0.98	1.41	0.29	0.96	0.20	11.52
14	11	3	1.42	1.30	1.42	0.11	1.30	0.10	4.61
14	11	5	1.41	0.44	1.19	-0.75	0.37	-0.24	327.64
14	11	9	1.31	0.54	1.23	0.45	0.51	0.19	20.27
14	12	4	1.41	0.79	1.38	-0.30	0.77	-0.17	347.78
14	12	6	1.37	0.67	-1.31	-0.41	-0.64	-0.20	197.41
15	1	0	1.91	1.35	-1.91	0.00	-1.35	0.00	180.00
15	2	1	2.36	1.83	2.15	0.97	1.67	0.75	24.30
15	3	2	1.37	0.77	1.27	-0.50	0.71	-0.28	338.53
15	4	3	1.39	1.18	-1.28	0.55	-1.08	0.46	156.88
15	5	2	1.96	1.76	1.30	-1.47	1.17	-1.32	311.49
15	6	3	1.69	1.69	1.65	0.36	1.65	0.36	12.42
15	7	2	2.00	1.23	1.98	0.25	1.22	0.16	7.22
15	8	3	1.43	0.40	-1.11	-0.89	-0.31	-0.25	218.82
15	9	2	1.42	0.74	1.09	0.91	0.57	0.47	39.81
15	9	6	1.41	1.13	1.40	-0.06	1.13	-0.05	357.59
15	10	1	1.44	1.20	1.32	-0.56	1.11	-0.46	337.25
15	10	3	1.42	0.69	1.38	-0.33	0.67	-0.16	346.54
15	11	6	1.36	0.88	1.30	-0.41	0.84	-0.27	342.42
15	12	5	1.35	0.78	-0.82	1.07	-0.48	0.62	127.67
16	0	0	3.39	3.34	3.39	-0.00	3.34	-0.00	360.00
16	2	2	1.40	1.01	-0.60	1.27	-0.43	0.91	115.26
16	3	1	1.70	1.32	-1.55	-0.71	-1.20	-0.55	204.58
16	4	0	2.21	2.22	2.21	0.00	2.22	0.00	0.00
16	4	2	1.41	1.29	1.41	0.04	1.29	0.03	1.51
16	4	4	2.47	2.90	2.46	-0.11	2.90	-0.13	357.51
16	5	1	1.43	0.76	-1.26	-0.67	-0.67	-0.36	208.16
16	6	0	2.00	1.53	2.00	0.00	1.53	0.00	0.00
16	6	2	1.42	0.91	-1.37	0.39	-0.87	0.25	164.16
16	7	0	1.42	0.02	-1.42	0.00	-0.02	0.00	180.00
16	8	0	3.17	2.27	3.17	-0.00	2.27	-0.00	360.00
16	8	2	1.42	0.77	1.28	0.61	0.70	0.33	25.53
16	10	0	1.40	0.54	1.40	0.00	0.54	0.00	0.00
16	10	2	1.40	0.49	-0.96	-1.02	-0.34	-0.36	226.67
16	11	1	1.39	0.81	-1.31	0.47	-0.76	0.27	160.18
16	12	0	1.89	1.68	1.89	0.00	1.68	0.00	0.00
17	2	1	1.42	0.57	1.38	-0.32	0.55	-0.13	346.85
17	3	2	1.42	1.26	1.42	-0.16	1.25	-0.14	353.55
17	4	0	1.42	0.06	1.42	-0.00	0.06	-0.00	360.00
17	4	3	1.43	1.00	-1.42	0.18	-0.99	0.13	172.60
17	5	2	1.42	0.71	1.42	-0.09	0.71	-0.04	356.47
17	7	2	2.00	0.63	1.99	-0.18	0.62	-0.06	354.80

TABLE VI (continued-9)

H	K	L	F0	FC	A0	B0	AC	BC	ALPHAC
17	8	0	1.40	0.08	-1.40	0.00	-0.08	0.00	180.00
17	9	0	1.95	0.80	-1.95	0.00	-0.80	0.00	180.00
17	9	2	1.38	0.88	1.38	0.00	0.88	0.00	0.12
18	2	2	1.42	1.61	-1.40	-0.22	-1.59	-0.26	189.11
18	6	2	1.39	1.93	-1.37	-0.22	-1.91	-0.31	189.08
18	10	0	1.82	0.22	-1.82	-0.00	-0.22	-0.00	180.00
19	2	1	1.39	0.87	1.15	-0.78	0.72	-0.49	325.96
20	0	0	1.89	1.89	1.89	0.00	1.89	0.00	0.00

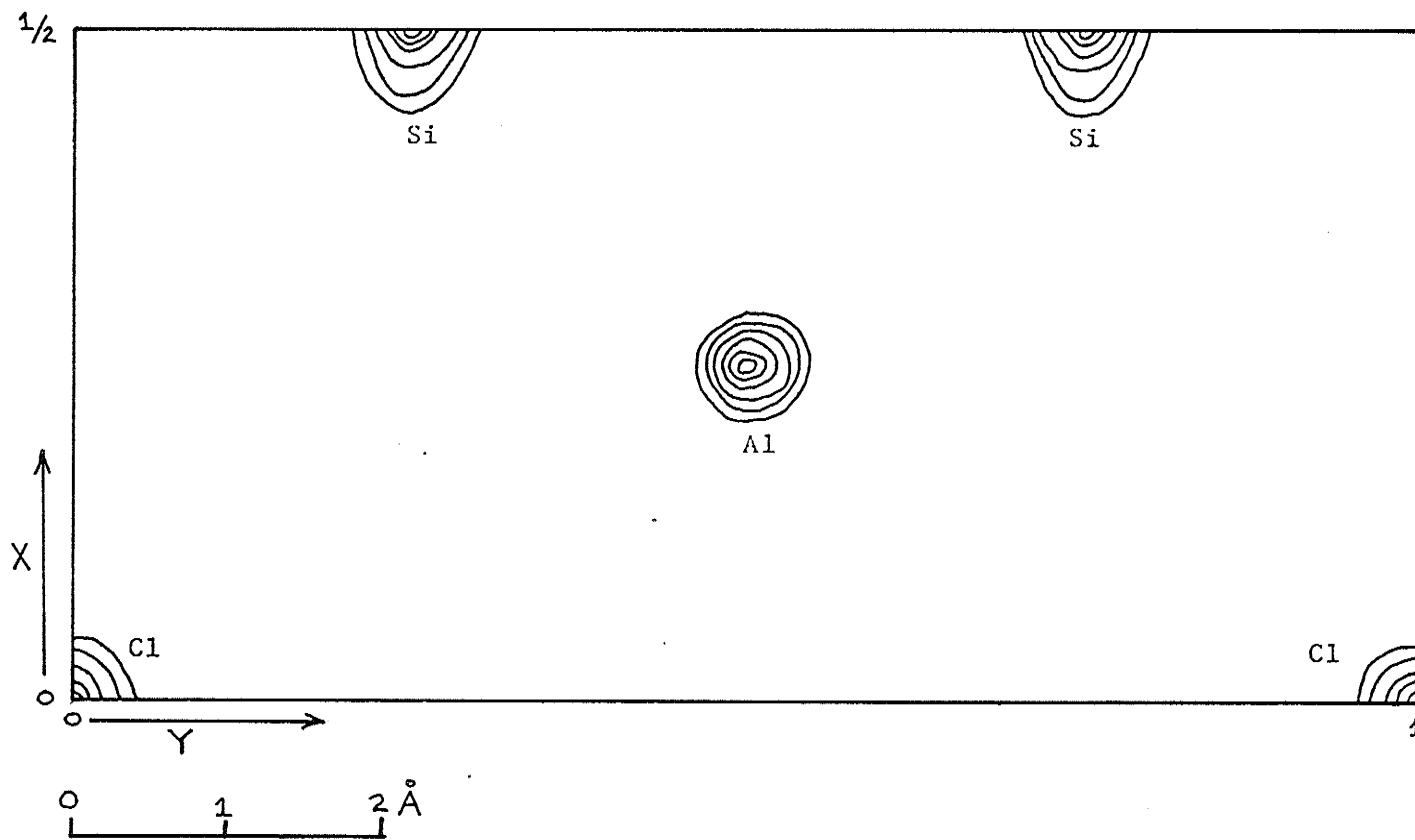


Fig. 6. ρ_0 : (001) Section at $z=0.0 \text{ \AA}$. The contours are drawn at intervals of 20 units around the atoms.

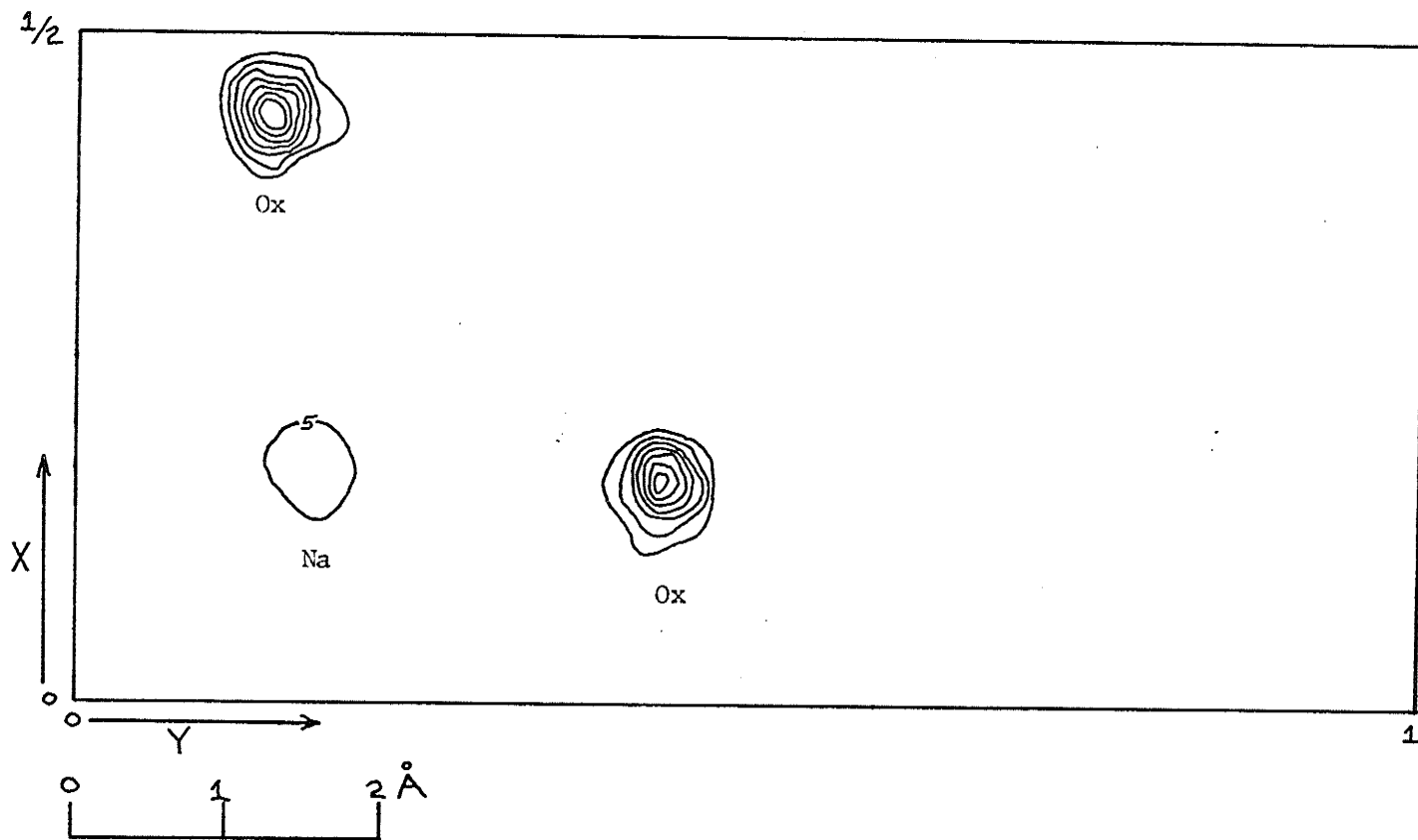


Fig. 7. ρ_0 : (001) section at $z = 1.211 \text{ \AA}$. The contours are drawn at intervals of 5 units around all atoms.

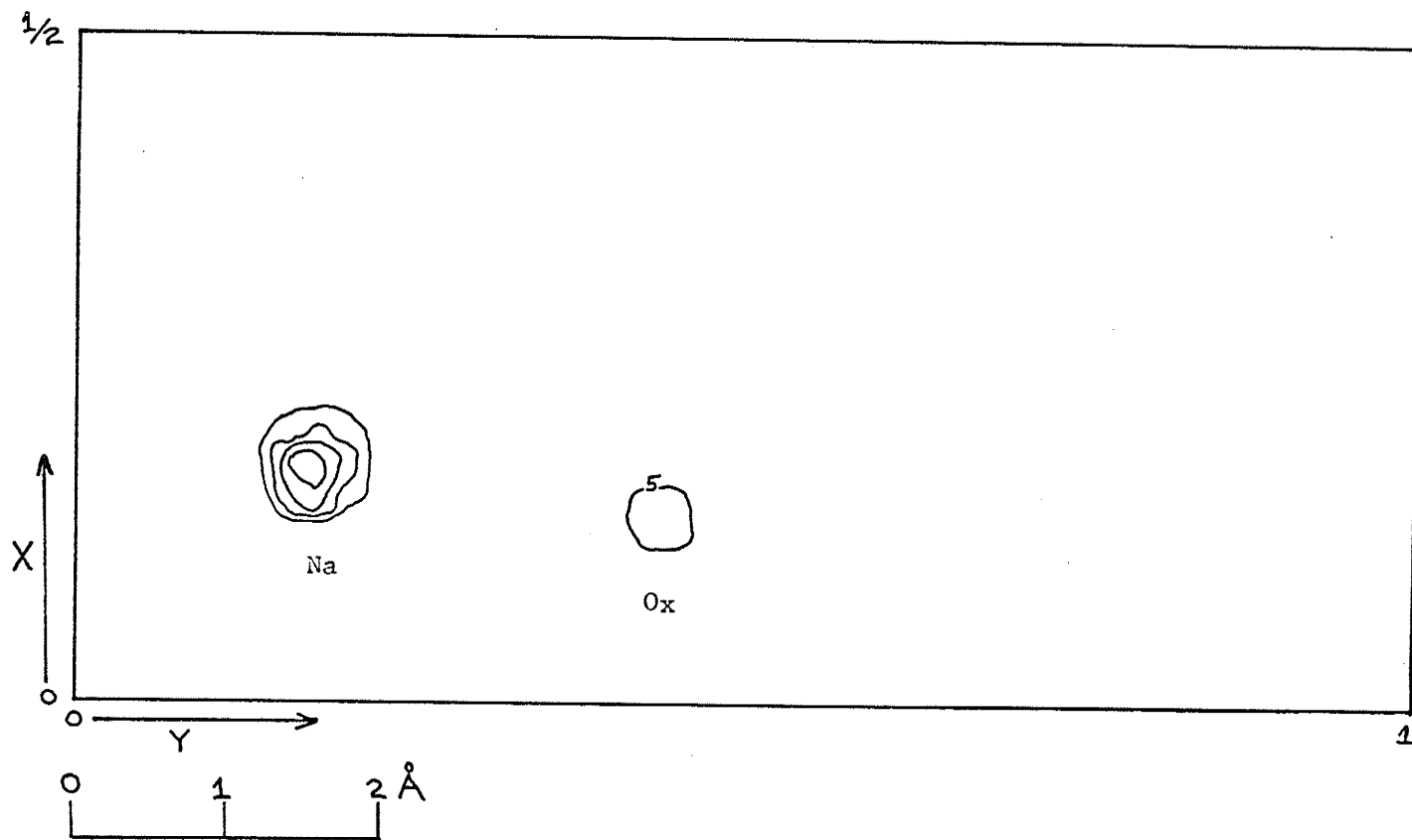


Fig. 8. ρ_o : (001) section at $z = 1.614 \text{ \AA}$. The contours are drawn at intervals of 10 units around the Na atom with one contour of 5 units around the O atom.

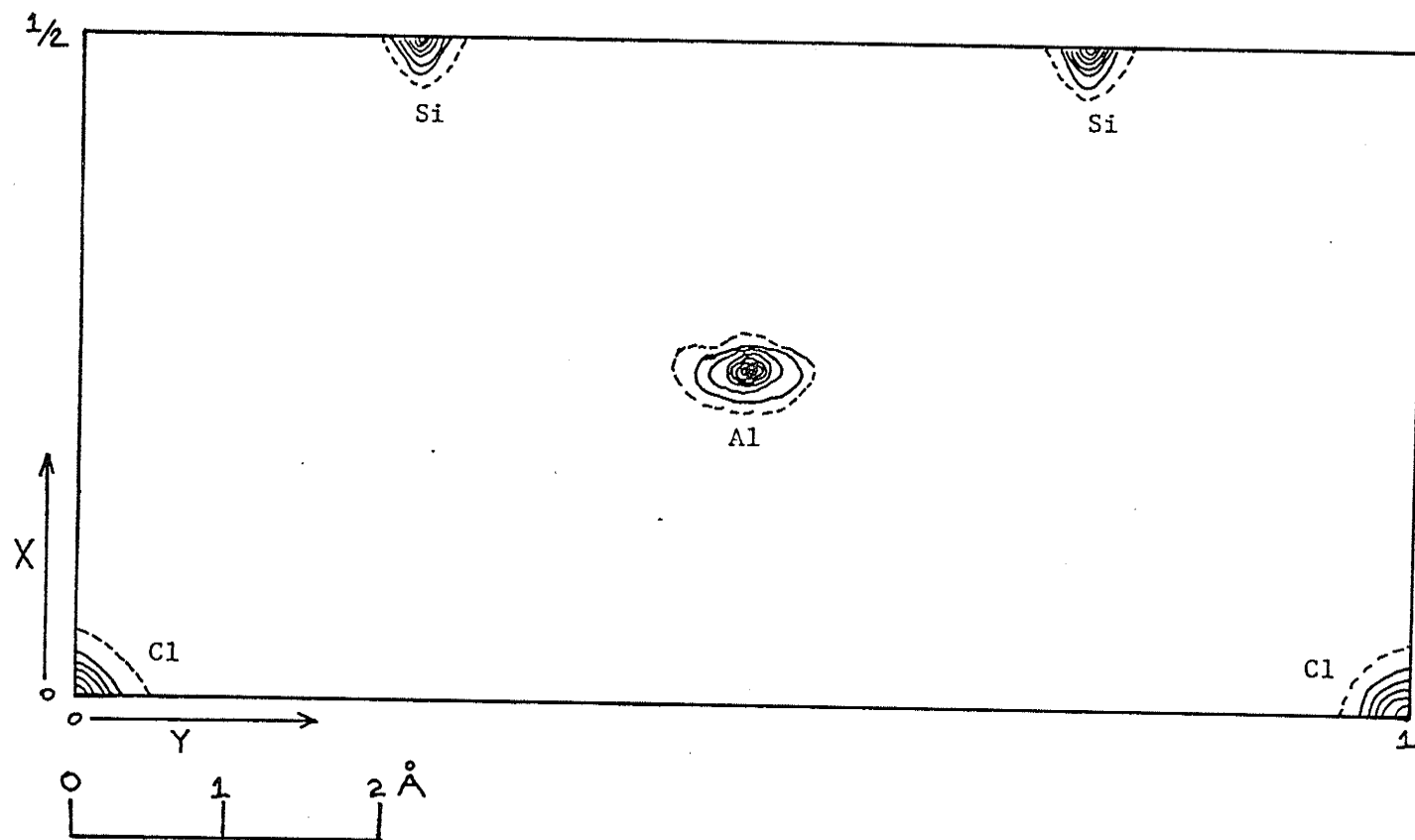


Fig. 9. $\rho_o - \rho_c$: (001) section at $z = 0.0 \text{ \AA}$. The contours are drawn at intervals of one unit with the zero contour dotted.

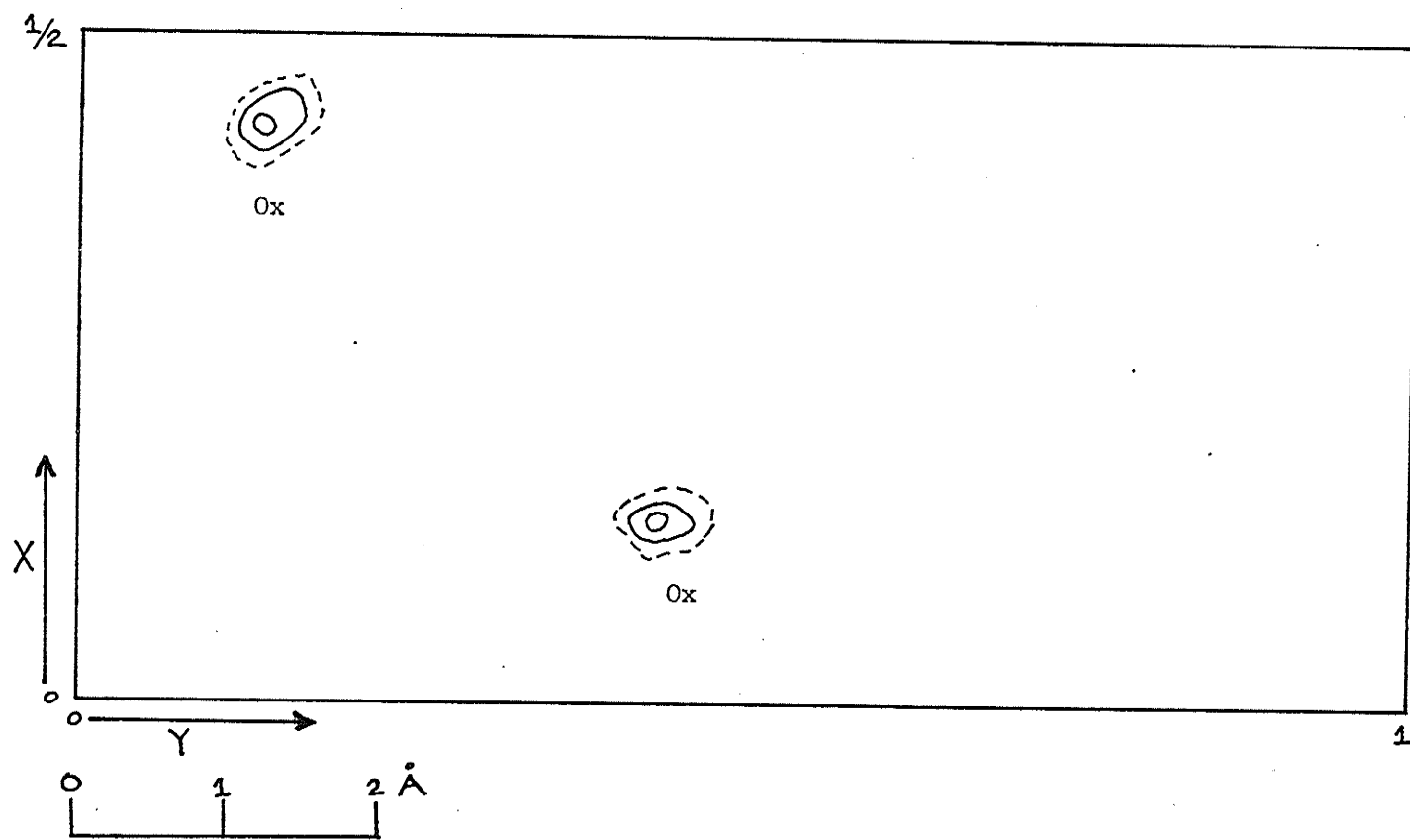


Fig. 10. $\rho_o - \rho_c$: (001) section at $z = 1.211\text{\AA}$. The contours are drawn at intervals of one unit with the zero line dotted.

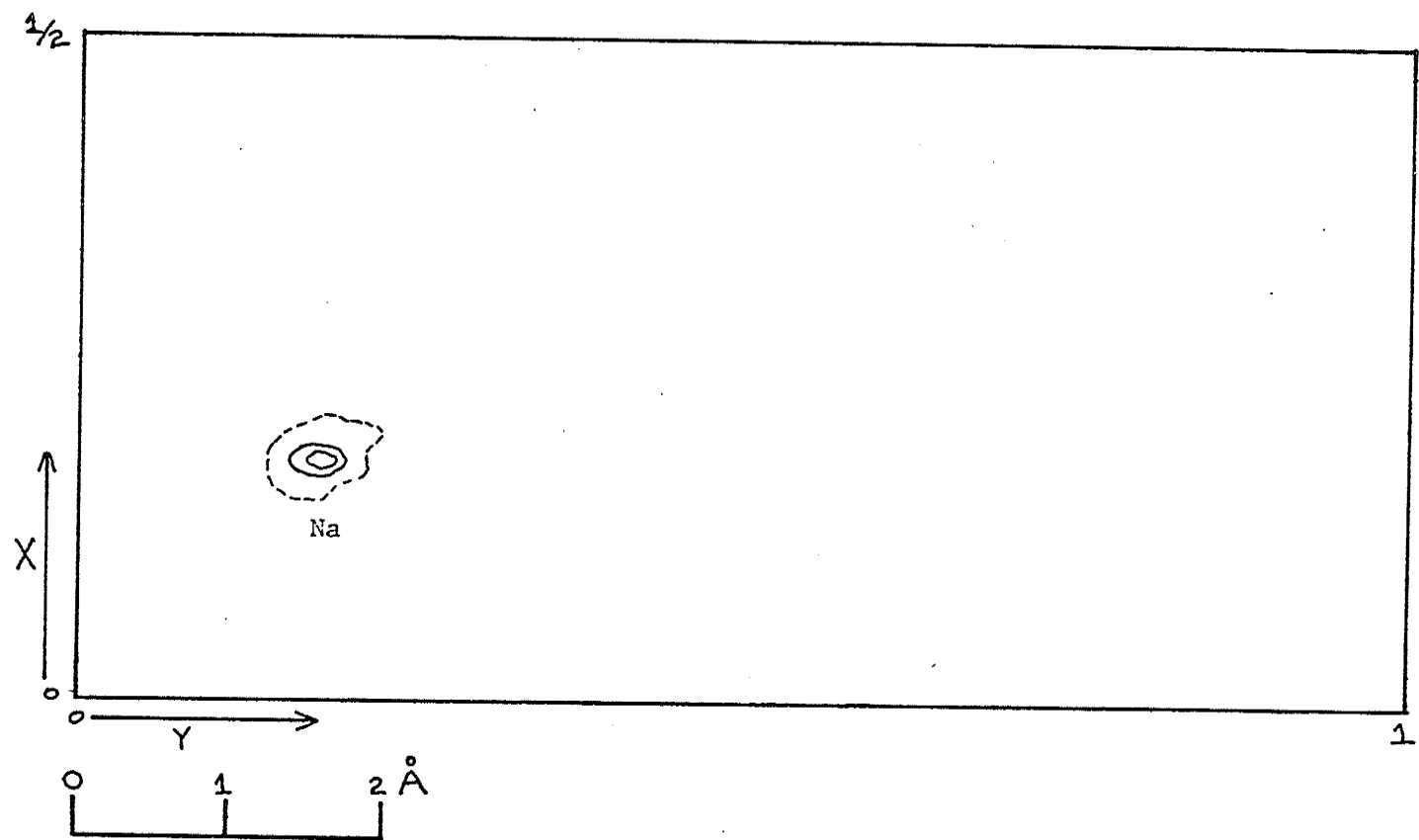


Fig. 11. $\rho_o - \rho_c$: (001) section at $z = 1.614\text{\AA}$. The contours are drawn at intervals of one unit with the zero line dotted.

TABLE VII

Interatomic Distances and Bond Angles

SiO₄ Tetrahedron

Si-O distance 1.629^oÅ

O-O distance 2.632^oÅ

O-O distance 2.717^oÅ

O-Si-O angle 107.74^o

O-Si-O angle 112.99^o

AlO₄ Tetrahedron

Al-O distance 1.730^oÅ

O-O distance 2.809^oÅ

O-O distance 2.857^oÅ

O-Al-O angle 108.55^o

O-Al-O angle 111.34^o

Other Angles and Distances

Si-O-Al angle 138.25^o

Na-Cl distance 2.733^oÅ

Na-Cl-Na angle 109.47^o

Na-O distance 2.353^oÅ

CHAPTER IX. CHARGE DISTRIBUTION CONSIDERATIONS

A. Introduction

An attempt^{was} made to see whether it is possible to interpret the sodalite structure in ionic terms. This involves the calculation of the positive electrostatic charges given by the metallic cations to their surrounding anions under different assumptions to see whether a reasonable charge distribution for the structure can be arrived at. This is essentially an attempt to apply Pauling's second rule relating to ionic structures.

This rule, as stated in Evans(1939), is: "In a stable coordination structure the total strength of the valency bonds which reach an anion from all the neighbouring cations is equal to the charge of the anion." The main idea underlying this rule is that one can interpret a crystal structure as a linking of polyhedra of anions around cations. Therefore, atoms are behaving as ions in this approach and one can consider the positive charge on the cation to be distributed to its surrounding anions. For an ionic structure viewed in this way, the sum of the positive charges received by one anion from all of its cations will nearly be equal to the negative charge on the anion.

The application of this rule in the 1920's and 30's was a big factor in solving the structures of many of the important rock-forming silicates such as feldspars, pyroxenes, amphiboles, and

micas. Gait & Ferguson (1968) have been able to interpret the structure of low albite, $\text{Na AlSi}_3\text{O}_8$, in these terms and it is an interpretation similar to theirs that is being attempted here.

When one attempts to interpret an aluminosilicate structure such as sodalite and albite in these terms there are three principal problems that arise. The first problem relates to the assumed pure Si-O and Al-O (T-O) distances, because these determine the proportion of Si and Al in a given tetrahedral site and hence the effective positive charge on that site. The second problem is in determining the coordination number of Na which is to be assumed in this interpretation. The last problem is that of deciding whether the positive charges should be distributed in some way that takes account of the differing cation-oxygen distances in one cation-oxygen polyhedron, as one might expect from Coulomb's law. Gait & Ferguson (1968) showed that low albite has virtually ideal charge distribution for an assumed Si-O distance of 1.602\AA , an assumed Al-O distance of 1.777\AA , and a coordination number (C.N.) of Na of 6, when the positive charges are distributed in amounts that are inversely proportional to the square of the cation-oxygen distances. Their procedure was followed here.

In the first instance it can easily be shown that the sodalite structure would have an ideal charge distribution if one assumes that:

- (a) The C.N. of Na is 4,

(b) The structure has not full ordering of Si and Al, but in fact any ordering down to complete disorder. (However, complete Si-Al disorder is precluded on structural evidence: completely disordered sodalite would have space^{group} symmetry $P\bar{4}3m$.)

(c) No account is taken of the differing cation-oxygen distances.

Three possible weaknesses exist in this interpretation. The first is that a 4 coordination of Na seems unusually small; Gait & Ferguson (1968) found a C.N. of 6 for Na in low albite. The next possible weakness is that the sizes of the two tetrahedra, as given by Löns & Schultz (1967) and confirmed by ourselves, suggest that the Si and Al are not quite fully ordered. Finally, Gait & Ferguson (1968) described strong evidence that account must be taken of differing cation-oxygen distances in distributing the positive charge.

The calculations on the sodalite structure were carried out in the manner described for low albite by Gait (1967).

B. Calculations

Assuming the Si-O and Al-O distances proposed by Smith & Bailey (1963), Löns & Schultz (1967) derived Al-contents of 0.1 and 0.9 for the two tetrahedra in sodalite based on their own derived tetrahedral "sizes". A consideration of the sodalite structure in ionic terms shows that because of the high symmetry of the structure the positive charges contributed to the oxygens by the tetrahedral cations are independent of the Si-Al distribution

because every O is linked to the two different tetrahedra. Thus on the face of it, it appears impossible to account for the Si-Al distribution in sodalite in terms of charge distribution as Gait & Ferguson (1968) were able to do in the case of low albite. Therefore, the author attempted to account only for the Na coordination by charge distribution calculations. Two calculations were made.

First Calculation

Na was assumed to be coordinated to the four closest anions, 3 oxygens at distances of 2.353\AA and by one chlorine at 2.733\AA , and the Na charge was distributed to the surrounding atoms as the inverse of the square of the distance. It was found that each oxygen would receive 0.267 e.s.u. and the chlorine 0.199 e.s.u. from Na^{+1} . Therefore, since each chlorine in the structure is tetrahedrally surrounded by 4 Na's, it would receive $4 \times 0.199 = 0.796$ e.s.u. of charge which is not sufficient to "balance" its unit negative charge. Hence the 4 coordination of Na may not be accounted for in this manner.

Second Calculation

In view of the result of the above calculation, an attempt was made to explain the structure using an Na coordination other than 4. The only other reasonable Na coordination appeared to be 8 in which the Na^{+1} charge is shared by 3 oxygens at 2.353\AA , 3 at 3.082\AA , a chlorine at 2.733\AA and another at 4.956\AA . For this coordination, each Cl receives a total charge of 0.676 e.s.u.

from the four surrounding Na's. Once again this approach had to be regarded as inadequate.

As a result of these calculations the author concluded that it was not possible to interpret the sodalite structure in this manner.

CHAPTER X. DISCUSSION OF THE RESULTS

The R-index of 0.1965 which was obtained is considerably larger than that of Löns & Schultz (1967) which was 0.082. Since the R-index is generally accepted as an indication of the accuracy of the refinement, one must conclude that the original intensities are not sufficiently accurate to yield a satisfactory low R-index. The reader will recall that the two reasons which were given by the author in the Introduction to the thesis for continuing the refinement after the paper of Löns & Schultz (1967) had appeared were:

- (a) To obtain Fourier maps which they had not obtained,
- (b) To investigate the charge distribution interpretation possibilities in the structure.

The author feels that both of these purposes have been fulfilled in this project. In addition, the author feels that a worthwhile contribution to the mineralogical data on the two Canadian sodalites has been made.

APPENDIX I. UPPER LEVEL WEISSENBERG INTENSITY CORRECTION

Subroutine (ELONGD): Correction for Intensities for Elongation
in the Upper Half of Equi-inclination Weissenberg Photographs

A. General Description

Phillips (1954) derives several formulae which govern the modification in reflection spot area observed on upper-level Weissenberg photographs. He also shows that these equations can be used in a routine correction of intensities visually estimated from such photographs. Phillips' equation (30) (for reflection-spot length with camera translation) reduces trigonometrically to

$$\frac{L \pm \Delta L}{L} = 1 + \frac{180}{2\pi} \frac{\sin\mu \cos\theta \cos\mu}{(\cos^2\mu - \cos^2\theta)(R_1 + R_2\cos\mu)}$$

where

- L - the length of the spot on the photograph,
- ΔL - change in length of the spot due to the geometrical effect,
- μ - the angle of inclination of the X-ray beam to the horizontal crystal axis,
- θ - the angle of reflection of the X-rays from a particular set of planes,
- R_1 - the radius of the cylindrical Weissenberg camera,
- R_2 - the crystal-to-source distance.

Singh (1967) gives a similar equation in a more recent publication.

This subroutine ELONGD was inserted as an additional subroutine in NRC-2 and it carries out the following steps for each reflection:

1. Calculates $\cos \theta$ from $\sin^2 \theta$ which has been brought in from the main program.
2. Sets μ equal to the proper angle (in radian) depending on the level of photograph from which the reflection has come.
3. Applies the correction to the original observed intensity which has not undergone any other correction up to this point.
4. Replaces the original uncorrected intensity with this corrected one and transfers it back to the main program.

Two runs with identical parameters were made on NRC-10 (SFLS) with and without the subroutine ELONGD. The R-indices for these two runs were 0.19 and 0.22 respectively which justifies the use of the elongation correction.

B. Listing

```
C *****
C SUBROUTINE TO CORRECT FOR ELONGATED REFLECTIONS
C IN UPPER HALF OF EQUI-INCLINATION WEISSENBERG
C PHOTOGRAPHS
C USES PHILLIPS EQUATION %30< OF ACTA CRYST 7,746
C 1954.....THIS EQUATION REDUCES TO FORM USED HERE
C BY GEOMETRY...SOME OF THE MOST IMPORTANT
C GEOMETRICAL EXPRESSIONS ARE
C %PSI<**2 # 4 SIN<THETA**2 - ZETA**2
C %PSI<MAXIMUM**2<# SQUAREROOT%4-ZETA**2<
C ZETA # -2SIN<%MU<
C SUBROUTINE ELONGD (SINSQ,IL,NET,PRINTR)
C INTEGER IL,PRINTR
C REAL MU,NET,ICORR,R1,R2,COSTA,SINSQ
C R1=28.7
C R2=75.0
C IF (1.0-SINSQ) 35,37,37
35 WRITE (PRINTR,36)
36 FORMAT (' IST SQRT IN ELONGD IS NEGATIVE@)
C GO TO 34
37 CONTINUE
C COSTA=SQRT(1-SINSQ)
C IF (IL.EQ.0) MU=0.0
C IF (IL.EQ.1) MU=2.2
C IF (IL.EQ.2) MU=4.6
C IF (IL.EQ.3) MU=6.9
C IF (IL.EQ.4) MU=9.2
C IF (IL.EQ.5) MU=11.4
C IF (IL.EQ.6) MU=13.8
C IF (IL.EQ.7) MU=16.1
C IF (IL.EQ.8) MU=18.4
C IF (IL.EQ.9) MU=21.0
C IF (IL.EQ.10) MU=23.5
C MU=MU*3.14159/180.00
C IF (COS(MU)**2-COSTA**2) 30,31,31
30 WRITE (PRINTR,32)
32 FORMAT (' SQRT NEG.-CORRECTION ELONGD NOT MADE@)
C GO TO 34
31 CONTINUE
C ICORR=NET*(1+(+28.648*SIN(MU)*COS(MU)*COSTA/
C 1(SQRT(COS(MU)**2-COSTA**2)*(R1+R2*COS(MU))))))
C NET=ICORR
34 CONTINUE
C RETURN
C END
```

APPENDIX II. SORTING SUBROUTINES

A. Introduction

Subroutines SORTA, SORTB, and SORTC: Sort and Average for Equivalent Reflections

The combination of three of these subroutines performs the operation of averaging the F_o and F_o^2 of all equivalent reflections, and of listing the resultant non-equivalent reflections in such a manner that $h \geq k \geq l$ for each reflection and in increasing order of l , k , and h respectively for the entire list of reflections. The description and listing of all three subroutines follows.

B. Subroutine SORTA

(a) General Description

This subroutine identifies all of the equivalent reflections as they are processed by NRC-2 and links them together by a key which is stored in the core memory with h , k , and l of the reflection. The remainder of the data for the reflection is stored on magnetic disk by the direct access method. The program list follows.

C. Subroutine SORTB

(a) General Description

This subroutine takes the equivalent reflections which have been linked together by an indicator in SORTA and averages

(b) Listing

```
C *****
  SUBROUTINE SORTA(IH,IK,IL,INITIA,IN,ARAY)
  INTEGER*2 ARAY, TABLE
C   THIS SUBROUTINE FINDS EQUIVALENT REFLECTIONS AS
C   THEY ARE PROCESSED BY THE NRC-2 PROGRAM AND LINKS
C   THESE REFLECTIONS TOGETHER BY AN INDICATOR
C   IN AN ARRAY WHICH IS CALLED ARAY...
  DIMENSION ARAY(4000), TABLE(18)
  J=0
  IP=3
  IN=IN+1
  ARAY(IN)=IH
  IN=IN+1
  ARAY(IN)=IK
  IN=IN+1
  ARAY(IN)=IL
  IN=IN+1
  IF (INITIA-1) 3,12,3
3  TABLE(1)=IH
  TABLE(2)=IL
  TABLE(3)=IK
  TABLE(4)=IL
  TABLE(5)=IK
  TABLE(6)=IH
  TABLE(7)=IL
  TABLE(8)=IH
  TABLE(9)=IK
  TABLE(10)=IK
  TABLE(11)=IL
  TABLE(12)=IH
  TABLE(13)=IK
  TABLE(14)=IH
  TABLE(15)=IL
  TABLE(16)=IH
  TABLE(17)=IK
  TABLE(18)=IL
7  IP=IP+4
  J=J+1
  K=1
  IF (INITIA-J) 12,12,6
12 ARAY(IN)=0
  RETURN
6  IF (ARAY(IN-IP)-TABLE(K)) 4,5,4
4  K=K+3
  IF (K-16) 6,6,7
5  L=K+1
  IT=IP-1
  IF (ARAY(IN-IT)-TABLE(L)) 4,9,4
9  IT=IT-1
  L=L+1
  IF (ARAY(IN-IT)-TABLE(L)) 4,10,4
10 ARAY(IN)=INITIA-J
  RETURN
  END
```

their F_o and F_c values. It then stores the resultant values on disk and stores the corresponding values of h , k , and l with $h \geq k \geq l$ in core memory. A listing of this program follows.

D. Subroutine SORTC

(a) General Description

This subroutine sorts all of the non-equivalent reflections from SORTB in such a way the l , k , and h increase in that $h \geq k \geq l$. It counts the number of non-equivalent reflections while sorting them. The parity (even/odd combination of the three indices) and order indicator (whether h , k , or l have varied from that of the previous reflection) are redetermined for the sorted reflections. The final hkl and associated data for each non-equivalent reflection sorted in the method described above are then stored on the output tape of NRC-2 and printed on the line printer. The number of non-equivalent reflections is printed on the line printer, and control is returned to the NRC-2 program. A listing of the subroutine follows.

(b) Listing

```
C *****
C THIS SUBROUTINE AVERAGES THE AMPLITUDES OF
C EQUIVALENT REFLECTIONS AND STORES THEM UNDER A
C RESULTANT REFLECTION WHICH HAS H GT. K GT. L.....
  SUBROUTINE SORTB(INITIA,IN,ARAY,PRINTR,IM,NUMBER)
  DEFINE FILE 11(1000,22,U,I8)
  DIMENSION ARAY(4000)
  INTEGER*2 ARAY
  DIMENSION AMPL(5),FFR(9)
  REAL AMP1,AMP2,AMP3,AMP4,AMP5
  INTEGER INITIA,IN,INDX,RECNO,COUNT,POINT,NUMBER
  INTEGER IH,IK,IL,INDR,IS,ORDIND,PARITY,MULT,PRINTR
  RECNO=0
  NAMPL=5
  NCRVES=10
  IS=IN
  INDX=INITIA
  IM=IS
  2 COUNT=1
  IF (ARAY(IS)) 43,4,3
43 WRITE (PRINTR,44) ARAY(IS)
44 FORMAT (' NEGATIVE RECORD NUMBER AT @,I4)
  GO TO 32
  3 IF (ARAY(IS)-32000) 5,32,5
  5 READ (11'INDX) INDR,IZ,ORDIND,PARITY,MULT,SINSQ,
  1RTW,(AMPL(M),M=1,NAMPL),(FFR(M),M=1,NCRVES)
  AMP1=AMPL(1)
  AMP2=AMPL(2)
  AMP3=AMPL(3)
  AMP4=AMPL(4)
  AMP5=AMPL(5)
  RECNO=ARAY(IS)
  7 IF (RECNO-INITIA) 45,46,46
46 WRITE (PRINTR,47) RECNO
47 FORMAT (' RECNO GT. NUMBER OF RECORDS@,I8)
  WRITE (PRINTR,50) ARAY(IS-3),ARAY(IS-2),
  1ARAY(IS-1),ARAY(IS)
50 FORMAT (4I8)
  GO TO 32
45 READ (11'RECNO) INDR,IZ,ORDIND,PARITY,MULT,SINSQ,
  1RTW,(AMPL(M),M=1,NAMPL),(FFR(M),M=1,NCRVES)
  POINT=RECNO*4
  IF (ARAY(POINT-3)-ARAY(IS-3)) 37,38,37
38 IF (ARAY(POINT-2)-ARAY(IS-2)) 37,39,37
39 IF (ARAY(POINT-1)-ARAY(IS-1)) 37,40,37
40 WRITE (PRINTR,41) RECNO
41 FORMAT ('//IDENTICAL HKL VALUES AT RECNO @,I4)
  GO TO 30
37 AMP1=AMP1+AMPL(1)
  AMP2=AMP2+AMPL(2)
  AMP3=AMP3+AMPL(3)
  AMP4=AMP4+AMPL(4)
  AMP5=AMP5+AMPL(5)
  COUNT=COUNT+1
30 RECNO=ARAY(POINT)
  ARAY(POINT)=32000
  IF (RECNO) 7,8,7
  8 AMPL(1)=AMP1/COUNT
```

(b) Listing (continued-2)

```
      AMPL(2)=AMP2/COUNT
      AMPL(3)=AMP3/COUNT
      AMPL(4)=AMP4/COUNT
      AMPL(5)=AMP5/COUNT
9  WRITE (11,INDX) INDR,IZ,ORDIND,PARITY,MULT,SINSQ,
      IRTW,(AMPL(M),M=1,NAMPL),(FFR(M),M=1,NCRVES)
C.  ONLY ONE REFLECTION OF THIS KIND
4  IH=ARRAY(IS-3)
      IK=ARRAY(IS-2)
      IL=ARRAY(IS-1)
      IF (IH-IK) 10,11,12
10  IF (IL-IK) 14,16,16
11  IF (IH-IL) 16,21,17
14  IF (IL-IH) 17,17,19
12  IF (IK-IL) 20,21,21
20  IF (IH-IL) 22,22,23
17  IA=IK
      IB=IH
      IC=IL
      GO TO 25
19  IA=IK
      IB=IL
      IC=IH
      GO TO 25
16  IA=IL
      IB=IK
      IC=IH
      GO TO 25
23  IA=IH
      IB=IL
      IC=IK
      GO TO 25
22  IA=IL
      IB=IH
      IC=IK
      GO TO 25
21  IA=IH
      IB=IK
      IC=IL
25  ARRAY(IM-3)=IA
      ARRAY(IM-2)=IB
      ARRAY(IM-1)=IC
      ARRAY(IM)=INDX
      IM=IM-4
32  INDX=INDX-1
      IF (INDX) 36,36,31
31  IS=IS-4
      GO TO 2
36  NUMBER=(IN-IM)/4
      RETURN
      END
```



```

C *****
SUBROUTINE SORTC(IM,NUMBER,ARAY,PRINTR,IN,TAPEB)
  DEFINE FILE 11(1000,22,U,I8)
  DIMENSION H(3),NH(3)
  DIMENSION ARAY(4000)
  INTEGER*2 ARAY
  INTEGER ORDIND,PARITY,H,NH
  INTEGER NUMBER,IM,TIME,IND,RECNO,PRINTR,HOUNT
  INTEGER TAPEB
144 FORMAT ('1 H K L A S O P M SIN SQ W
1 STRUCTURE AMPLITUDES AND SCATTERING FACTORS
PAGE@,I4/))

146 FORMAT(' @,I3,2I4,1X,5I2,F8.4,F7.3,6F9.2)
147 FORMAT(39X,10F9.3)
  IT=IM+5
C SORTS ALL REFLNS ACCORDING TO INCREASING H
  NAMPL=5
  NCRVES=10
  LINES=55
  NPAGES=1
  REAL AMPL(5),FFR(9),SINSQ,RTW
  LIM=0
  HIM=0
  HOUNT=0
  KOUNT=0
  HLOOP=0
  KLOOP=0
  KIW=IM+2
  2 TIME=0
  3 IND=0
  IP=0
  6 IP=IP+4
  IF (IT-IP-(IM+1)) 10,45,45
  45 IF (IND) 181,182,181
  182 IA=ARAY(IT)
  IB=ARAY(IT+1)
  IC=ARAY(IT+2)
  ID=ARAY(IT+3)
  181 IF (IA-ARAY(IT-IP)) 4,5,5
  4 IND=IND+1
  ARAY(IT-IP+4)=ARAY(IT-IP)
  ARAY(IT-IP+5)=ARAY(IT-IP+1)
  ARAY(IT-IP+6)=ARAY(IT-IP+2)
  ARAY(IT-IP+7)=ARAY(IT-IP+3)
  GO TO 6
  5 IF (IND) 10,11,10
  10 ARAY(IT-IP+4)=IA
  ARAY(IT-IP+5)=IB
  ARAY(IT-IP+6)=IC
  ARAY(IT-IP+7)=ID
  11 TIME=TIME+1
  IF (NUMBER-TIME-1) 12,12,13
  13 IT=IT+4
  GO TO 3
C NOW COUNTS ALL REFLNS OF A CERTAIN H VALUE
  12 IW=IM+1
  IZ=1
  180 HLOOP=0
  100 IF (ARAY(IW)-IZ) 101,101,102

```

(b) Listing (continued-2)

```
101 HLOOP=HLOOP+1
    IW=IW+4
    IF (IW-IN) 100,100,99
99 HOUNT=4
    IF (HLOOP) 139,139,104
102 IF (HLOOP) 103,103,104
103 IZ=IZ+1
    IF (IZ-30) 100,106,106
106 WRITE (PRINTR,170)
170 FORMAT (' ONE OF THE INDICES IS GREATER THAN 30@)
    GO TO 139
C   SORTS K IN THIS RANGE OF H
104 IF (HLOOP-1) 215,215,188
215 HIM=HIM-4
    GO TO 115
188 KIT=IM+6+HIM
    KIY=KIT-4
    KTIME=0
107 KIND=0
    KIP=0
108 KIP=KIP+4
    IF (KIT-KIP-KIY) 114,110,110
110 IF (KIND) 184,185,184
185 IA=ARAY(KIT-1)
    IB=ARAY(KIT)
    IC=ARAY(KIT+1)
    ID=ARAY(KIT+2)
184 IF (IB-ARAY(KIT-KIP)) 111,142,142
111 KIND=KIND+1
    ARAY(KIT-KIP+3)=ARAY(KIT-KIP-1)
    ARAY(KIT-KIP+4)=ARAY(KIT-KIP)
    ARAY(KIT-KIP+5)=ARAY(KIT-KIP+1)
    ARAY(KIT-KIP+6)=ARAY(KIT-KIP+2)
    GO TO 108
142 IF (KIND) 114,115,114
114 ARAY(KIT-KIP+4)=IB
    ARAY(KIT-KIP+3)=IA
    ARAY(KIT-KIP+5)=IC
    ARAY(KIT-KIP+6)=ID
115 KTIME=KTIME+1
    HIM=HIM+4
    IF (HLOOP-KTIME-1) 116,116,117
117 KIT=KIT+4
    GO TO 107
C   COUNTS K OF CERTAIN KIND FOR THIS VALUE OF H
116 HIM=HIM+4
    KIZ=0
    KLP=0
179 KLOOP=0
119 IF (ARAY(KIW)-KIZ) 120,120,121
120 KLOOP=KLOOP+1
    KLP=KLP+1
    KIW=KIW+4
    IF (HLOOP-KLP) 160,160,119
C   SETS KOUNT # 4 IF HAS READ ALL K IN RANGE OF H
160 KOUNT=4
    IF (KLOOP) 139,139,123
121 IF (KLOOP) 122,122,123
```

(b) Listing (continued-3)

```
122 KIZ=KIZ+1
    IF (KIZ-30) 119,106,106
C   SORTS L FOR CERTAIN VALUE OF H AND K
123 IF (KLOOP-1) 235,235,189
235 LIM=LIM-4
    GO TO 135
189 LIT=IM+7+LIM
    LIY=LIT-4
    LTIME=0
127 LIND=0
    LIP=0
128 LIP=LIP+4
    IF (LIT-LIP-LIY) 134,130,130
130 IF (LIND) 186,187,186
187 IA=ARRAY(LIT-2)
    IB=ARRAY(LIT-1)
    IC=ARRAY(LIT)
    ID=ARRAY(LIT+1)
186 IF (IC-ARRAY(LIT-LIP)) 131,152,152
131 LIND=LIND+1
    ARRAY(LIT-LIP+2)=ARRAY(LIT-LIP-2)
    ARRAY(LIT-LIP+3)=ARRAY(LIT-LIP-1)
    ARRAY(LIT-LIP+4)=ARRAY(LIT-LIP)
    ARRAY(LIT-LIP+5)=ARRAY(LIT-LIP+1)
    GO TO 128
152 IF (LIND) 134,135,134
134 ARRAY(LIT-LIP+4)=IC
    ARRAY(LIT-LIP+5)=ID
    ARRAY(LIT-LIP+2)=IA
    ARRAY(LIT-LIP+3)=IB
135 LTIME=LTIME+1
    LIM=LIM+4
    IF (KLOOP-LTIME-1) 136,136,137
137 LIT=LIT+4
    GO TO 127
136 LIM=LIM+4
    IF (KOUNT-4) 140,138,140
140 KIZ=KIZ+1
    GO TO 179
138 KOUNT=0
    IF (HOUNT-4) 141,139,141
141 IZ=IZ+1
    GO TO 180
C   OUTPUT ON PRINTER
139 WRITE (PRINTR,161)
161 FORMAT ('1 THIS IS THE HKL LIST AFTER EXECUTION OF PROGRAM
1 BETWEEN NRC-2 AND NRC-10)
    WRITE (PRINTR,144) NPAGES
    I1=1
    I2=2
    I3=3
    ICODE=1
    DO 150 I=1,NUMBER
    IM=IM+4
    RECNO=ARRAY(IM)
    IH=ARRAY(IM-3)
    IK=ARRAY(IM-2)
    IL=ARRAY(IM-1)
```

(b) Listing (continued-4)

```
      READ (IT,RECNO) INDR,IS,ORDIND,PARITY,MULT,SINSQ,  
      1RTW,(AMPL(M),M=1,NAMPL),(FFR(M),M=1,NCRVES)  
C     TEST SIGNS OF H,K,L, REVERSE SIGNS IF 2 OR 3  
C     INDICES ARE NEGATIVE  
      IF(IH) 280,281,281  
0280 NH(I1)=-IH  
      IS=1  
      GO TO 282  
0281 IS=0  
      NH(I1)=IH  
0282 IF(IK) 283,284,284  
0283 NH(I2)=-IK  
      IS=IS+2  
      GO TO 297  
0284 NH(I2)=IK  
0297 IF(IL) 285,299,299  
0285 NH(I3)=-IL  
      IS=IS+4  
      GO TO 298  
0299 NH(I3)=IL  
0298 GO TO(287,295,288,289,290,292,294),IS  
0286 IS=1  
      GO TO 296  
0287 IS=2  
      GO TO 296  
0288 IH=NH(I1)  
      IK=NH(I2)  
      IL=-IL  
0289 IS=4  
      GO TO 296  
0290 IS=3  
      IH=NH(I1)  
      IK=-IK  
0291 IL=NH(I3)  
      GO TO 296  
0292 IS=2  
      IH=-IH  
0293 IK=NH(I2)  
      GO TO 291  
0294 IS=1  
      IH=NH(I1)  
      GO TO 293  
0295 IS=3  
296 IF (ICODE-1) 257,261,257  
0261 ICODE=2  
      ORDIND=4  
      NREFNS=1  
      H(1)=NH(1)  
      H(2)=NH(2)  
      H(3)=NH(3)  
      GO TO 254  
C     ORDER OF REFLECTIONS  
0257 IF(NH(1)-H(1)) 247,248,247  
0248 IF(NH(2)-H(2)) 249,251,249  
0251 IF(NH(3)-H(3)) 252,253,252  
0253 ORDIND=0  
      GO TO 254  
0252 ORDIND=3
```

(b) Listing (continued-5)

```
0255 H(3)=NH(3)
      GO TO 254
0249 ORDIND=2
0256 H(2)=NH(2)
      GO TO 255
0247 ORDIND=1
      H(1)=NH(1)
      GO TO 256
C     PARITY AND MULTIPLICITY
0254 PARITY=4*MOD(H(I1),2)+2*MOD(H(I2),2)+MOD(H(I3),2)+1
      WRITE (PRINTR,146) IH,IK,IL,INDR,IS,ORDIND,PARITY,
      IMULT,SINSQ,RTW,(AMPL(M),M=1,NAMPL)
      WRITE (PRINTR,147) (FFR(M),M=1,NCRVES)
      LINES=LINES-2
      WRITE (TAPE8) IH,IK,IL,INDR,IS,ORDIND,PARITY,MULT,
      1SINSQ,RTW,(AMPL(M),M=1,NAMPL),(FFR(M),M=1,NCRVES)
      IF (LINES) 151,151,150
151  NPAGES=NPAGES+1
      LINES=55
      WRITE (PRINTR,144) NPAGES
150  CONTINUE
      WRITE (PRINTR,153) NUMBER
153  FORMAT ('NUMBER OF NON-EQUIVALENT REFLNS IS #0,I4)
      RETURN
      END
```

A.Main Program APPENDIX III. LISTING OF PROGRAM EXPAND

```
C THIS IS A PROGRAM WHICH WILL PRODUCE THE EQUIVALENT
C REFLECTIONS OUT OF A REFLECTION-----
C E.G.MAKES 123,132,213,231,321,AND 312 OUT OF 321
C MULTY IS AN ARRAY WHICH REPRESENTS THE RELATIVE
C MULTIPLICITIES OF HOO,HHO,HKO,HHH,HHL,LHH,HKL.....
  INTEGER MULTY(7),TAPEA,TAPEB,PRINTR,LISTA(12),SERA(12)
  INTEGER SERLA,CENTRE,SYSTEM,R(3,3,24),CENT(16),NPOS(16)
  INTEGER MATCES(24,16),W(32),SERL,SYMN(90),SYMB(90)
  INTEGER ATOMN(90),VIB(90),INDTRS(90),CRV(90)
  INTEGER SYMBOL(10),ORDIND,PARITY,CS(1280)
  INTEGER*2 ARAY
  DIMENSION ARAY(12000)
  REAL LAMBDA,GQT(9),T(3,24),X(90),Y(90),Z(90)
  REAL SX(90),SY(90),SZ(90),BISO(90),B11(90),B22(90)
  REAL B33(90),B23(90),B13(90),B12(90),OCCFR(90)
  REAL AMPL(10),FF(10)
C TO READ PAST FILES ON INPUT TAPE TO LOCATION OF PLANES RECORDS
  DEFINE FILE 11 (6000,22,U,18)
  TAPEA=8
  TAPEB=9
  PRINTR=6
  KOUNT1=0
  INITIA=0
  IN=0
  REWIND TAPEA
  REWIND TAPEB
  READ (TAPEA) IDT
  WRITE (TAPEB) IDT
  READ(TAPEA) JOBA,LSTA,SERLA,NLISTA,(LISTA(I),SERA(I),I=1,12)
  WRITE(TAPEB)JOBA,LSTA,SERLA,NLISTA,(LISTA(I),SERA(I),I=1,12)
  READ (TAPEA) LST,SERL,NRCRD,NWORD
  WRITE(TAPEB) LST,SERL,NRCRD,NWORD
  READ (TAPEA) (W(I),I=1,6),LAMBDA,W(7),W(8)
  WRITE(TAPEB) (W(I),I=1,6),LAMBDA,W(7),W(8)
  READ(TAPEA) (W(I),I=1,NWORD)
  WRITE(TAPEB) (W(I),I=1,NWORD)
  READ (TAPEA) (GQT(I),I=1,NWORD)
  WRITE(TAPEB) (GQT(I),I=1,NWORD)
  READ (TAPEA) (W(I),I=1,NWORD)
  WRITE(TAPEB) (W(I),I=1,NWORD)
  READ (TAPEA) LST,SERL,NRCRD,NWORD
  WRITE(TAPEB) LST,SERL,NRCRD,NWORD
  READ (TAPEA) NMATX,NSYM,CENTRE,LAT,SYSTEM
  WRITE(TAPEB) NMATX,NSYM,CENTRE,LAT,SYSTEM,I,I,I,I,I,I,I,I
  DO 130 K=1,NMATX
  READ(TAPEA) MATNO,((R(I,J,K),I=1,3),J=1,3),(T(I,K),I=1,3)
0130 WRITE(TAPEB) MATNO,((R(I,J,K),I=1,3),J=1,3),(T(I,K),I=1,3)
  DO 131 K=1,NSYM
  READ (TAPEA) CENT(K),NPOS(K),(MATCES(M,K),M=1,11)
  WRITE(TAPEB) CENT(K),NPOS(K),(MATCES(M,K),M=1,11)
  READ (TAPEA) (MATCES(M,K),M=12,24)
0131 WRITE(TAPEB) (MATCES(M,K),M=12,24)
  READ (TAPEA) LST,SERL,NRCRD,NWORD
  WRITE(TAPEB) LST,SERL,NRCRD,NWORD
  DO 133 K=1,NRCRD
  READ (TAPEA) (W(I),I=1,NWORD)
0133 WRITE(TAPEB) (W(I),I=1,NWORD)
  READ (TAPEA) LST,SERL,NRCRD,NWORD
```

A. Main Program (continued-2)

```
WRITE(TAPEB) LST,SERL,NRCRD,NWORD
DO 137 K=1,NRCRD
  READ (TAPEA) SYMN(K),SYMB(K),ATOMN(K),X(K),Y(K),Z(K),SX(K),SY(K)
  1 SZ(K), BISO(K),VIB(K),INDTRS(K),B11(K),B22(K),B33(K),
  2B23(K),B13(K),B12(K),OCCFR(K)
0137 WRITE(TAPEB) SYMN(K),SYMB(K),ATOMN(K),X(K),Y(K),Z(K),SX(K),SY(K)
  1 SZ(K), BISO(K),VIB(K),INDTRS(K),B11(K),B22(K),B33(K),
  2B23(K),B13(K),B12(K),OCCFR(K)
  READ (TAPEA) LST,SERL,NRCRD,NWORD
  WRITE (TAPEB) LST,SERL,NRCRD,NWORD
  MIN=1
  DO 354 M=1,NRCRD
  MAX=MIN+31
  READ (TAPEA) (CS(L),L=MIN,MAX)
  WRITE(TAPEB) (CS(L),L=MIN,MAX)
354 MIN=MAX+1
  READ (TAPEA) LST,SERL,NRCRD,NWORD
  WRITE (TAPEB) LST,SERL,NRCRD,NWORD
  READ (TAPEA) FOOO,NPLNS,I1,I2,I3,NAMPL,NCRVES,C1,C2,C3,(W(L),
  1L=1,NAMPL),(SYMBOL(N),N=1,NCRVES)
  WRITE(TAPEB) FOOO,NPLNS,I1,I2,I3,NAMPL,NCRVES,C1,C2,C3,(W(L),
  1L=1,NAMPL),(SYMBOL(N),N=1,NCRVES)
C HAS READ TO PLACE WHERE PLANES RECORDS ARE LOCATED
  MULTY(1)=1
  MULTY(2)=2
  MULTY(3)=3
  MULTY(4)=4
  MULTY(5)=5
  MULTY(6)=6
  MULTY(7)=7
  DO 500 I=1,478
  READ (TAPEA) IH,IK,IL,INDR,IS,ORDIND,PARITY,MULT,SINSQ,RTW,
  1(AMPL(N),N=1,NAMPL),(FF(M),M=1,NCRVES)
  CALL SETUP (IH,IK,IL,INDR,IS,ORDIND,PARITY,MULT,SINSQ,RTW,
  IAMPL,FF,NAMPL,NCRVES,IN,INITIA,ARAY)
  NZONE=7
  IF (IH) 10,11,10
  11 NZONE=NZONE-4
  IF (IK) 12,13,12
  13 NZONE=NZONE-2
  GO TO 75
  12 IF (IL) 14,13,14
  14 IF (IK-IL) 75,17,75
  17 NZONE=NZONE-1
  GO TO 75
  10 IF (IK) 20,21,20
  21 NZONE=NZONE-4
  IF (IL) 22,13,22
  22 IF (IH-IL) 75,17,75
  20 IF (IL) 23,24,23
  24 NZONE=NZONE-4
  IF (IH-IK) 75,17,75
C ENTER HERE FOR NO ZERO INDICES
  23 IF (IH-IL) 25,26,32
  26 IF (IL-IK) 17,28,13
  28 NZONE=NZONE-3
  GO TO 75
  25 IF (IK-IL) 40,17,75
```

A. Main Program (continued-3)

```
40 IF (IH-IK) 75,13,75
30 IF (IH-IL) 75,13,75
32 IF (IK-IL) 75,13,39
39 IF (IH-IK) 75,17,75
75 MALT=MULTY(NZONE)
   IF (MALT-3) 100,101,102
100 CALL SETUP (IL,IH,IK,INDR,IS,ORDIND,PARITY,MULT,SINSQ,RTW,
   IAMPL,FF,NAMPL,NCRVES,IN,INITIA,ARAY)
   CALL SETUP (IK,IL,IH,INDR,IS,ORDIND,PARITY,MULT,SINSQ,RTW,
   IAMPL,FF,NAMPL,NCRVES,IN,INITIA,ARAY)
   KOUNT1=KOUNT1+3
   GO TO 500
101 CALL SETUP (IK,IL,IH,INDR,IS,ORDIND,PARITY,MULT,SINSQ,RTW,
   IAMPL,FF,NAMPL,NCRVES,IN,INITIA,ARAY)
   CALL SETUP (IH,IL,IK,INDR,IS,ORDIND,PARITY,MULT,SINSQ,RTW,
   IAMPL,FF,NAMPL,NCRVES,IN,INITIA,ARAY)
   CALL SETUP (IL,IK,IH,INDR,IS,ORDIND,PARITY,MULT,SINSQ,RTW,
   IAMPL,FF,NAMPL,NCRVES,IN,INITIA,ARAY)
   CALL SETUP (IL,IH,IK,INDR,IS,ORDIND,PARITY,MULT,SINSQ,RTW,
   IAMPL,FF,NAMPL,NCRVES,IN,INITIA,ARAY)
   CALL SETUP (IK,IH,IL,INDR,IS,ORDIND,PARITY,MULT,SINSQ,RTW,
   IAMPL,FF,NAMPL,NCRVES,IN,INITIA,ARAY)
   KOUNT1=KOUNT1+6
   GO TO 500
C   ENTER IF MALT IS GREATER THAN 3
102 IF (MALT-5) 600,100,105
600 KOUNT1=KOUNT1+1
   GO TO 500
105 IF (MALT-6) 100,100,101
500 CONTINUE
   WRITE (6,400) KOUNT1
400 FORMAT ('THE NUMBER OF PLANES GENERATED BY THE SMALL PROGRAM
   1 BETWEEN NRC-2 AND NRC-10 IS @,I8)
C   HAVE NOW LOADED ALL EQUIVALENT REFLECTIONS ONTO
C   DISK AND INTO MEMORY
   NUMBER=KOUNT1
   IM=0
   CALL SORGEN (IM,NUMBER,ARAY,PRINTR,IN,TAPEB)
   END FILE TAPEB
   END FILE TAPEA
   REWIND TAPEA
   REWIND TAPEB
   RETURN
   END
```


B. Subroutine Setup

```
C THIS SUBROUTINE IS TO STORE THE HKL VALUE OF A REFLECTION  
C IN THE MEMORY AND TO STORE THE OTHER DATA IN MAGNETIC  
C DISK BY THE DIRECT ACCESS METHOD  
SUBROUTINE SETUP (IA, IB, IC, INDR, IS, ORDIND, PARITY, MULT, SINSQ,  
RTW, AMPL, FF, NAMPL, NCRVES, IN, INITIA, ARAY)  
  DEFINE FILE 11 (6000,22,U,I8)  
  INTEGER ORDIND, PARITY, MULT, IN, INITIA  
  INTEGER*2 ARAY  
  DIMENSION ARAY(12000)  
  REAL AMPL(5), FF(10), SINSQ, RTW  
  INITIA=INITIA+1  
  IN=IN+1  
  ARAY(IN)=IA  
  IN=IN+1  
  ARAY(IN)=IB  
  IN=IN+1  
  ARAY(IN)=IC  
  IN=IN+1  
  ARAY(IN)=INITIA  
  WRITE (11,'INITIA) INDR, IS, ORDIND, PARITY, MULT, SINSQ, RTW,  
1 (AMPL(M), M=1, NAMPL), (FF(M), M=1, NCRVES)  
  RETURN  
  END
```

C. Subroutine SORGEN

```
C *****
SUBROUTINE SORGEN (IM,NUMBER,ARAY,PRINTR,IN,TAPEB)
DEFINE FILE 11 (6000,22,U,I8)
DIMENSION H(3),NH(3)
DIMENSION ARAY(12000)
INTEGER TAPEB
INTEGER*2 ARAY
INTEGER ORDIND,PARITY,H,NH
INTEGER NUMBER,IM,TIME,IND,RECNO,PRINTR,HOUNT
144 FORMAT ('1 H K L A S O P M SIN SQ W
1 STRUCTURE AMPLITUDES AND SCATTERING FACTORS
1 PAGE@,I4/ )
146 FORMAT(' @,I3,2I4,1X,5I2,F8.4,F7.3,6F9.2)
147 FORMAT(39X,10F9.3)
IT=IM+5
C SORTS ALL REFLNS ACCORDING TO INCREASING H
NAMPL=5
NCRVES=10
LINES=55
NPAGES=1
REAL AMPL(5),FFR(9),SINSQ,RTW
LIM=0
HIM=0
HOUNT=0
KOUNT=0
HLOOP=0
KLOOP=0
KIW=IM+2
2 TIME=0
3 IND=0
IP=0
6 IP=IP+4
IF (IT-IP-(IM+1)) 10,45,45
45 IF (IND) 181,182,181
182 IA=ARAY(IT)
IB=ARAY(IT+1)
IC=ARAY(IT+2)
ID=ARAY(IT+3)
181 IF (IA-ARAY(IT-IP)) 4,5,5
4 IND=IND+1
ARAY(IT-IP+4)=ARAY(IT-IP)
ARAY(IT-IP+5)=ARAY(IT-IP+1)
ARAY(IT-IP+6)=ARAY(IT-IP+2)
ARAY(IT-IP+7)=ARAY(IT-IP+3)
GO TO 6
5 IF (IND) 10,11,10
10 ARAY(IT-IP+4)=IA
ARAY(IT-IP+5)=IB
ARAY(IT-IP+6)=IC
ARAY(IT-IP+7)=ID
11 TIME=TIME+1
IF (NUMBER-TIME-1) 12,12,13
13 IT=IT+4
GO TO 3
C NOW COUNTS ALL REFLNS OF A CERTAIN H VALUE
12 IW=IM+1
IZ=0
180 HLOOP=0
```

C. Subroutine SORGEN (continued-2)

```
100 IF (ARAY(IW)-IZ) 101,101,102
101 HLOOP=HLOOP+1
    IW=IW+4
    IF (IW-IN) 100,100,99
99 HOUNT=4
    IF (HLOOP) 139,139,104
102 IF (HLOOP) 103,103,104
103 IZ=IZ+1
    IF (IZ-30) 100,106,106
106 WRITE (PRINTR,170)
170 FORMAT (' ONE OF THE INDICES IS GREATER THAN 30@)
    GO TO 139
C   SORTS K IN THIS RANGE OF H
104 IF (HLOOP-1) 215,215,188
215 HIM=HIM-4
    GO TO 115
188 KIT=IM+6+HIM
    KIY=KIT-4
    KTIME=0
107 KIND=0
    KIP=0
108 KIP=KIP+4
    IF (KIT-KIP-KIY) 114,110,110
110 IF (KIND) 184,185,184
185 IA=ARAY(KIT-1)
    IB=ARAY(KIT)
    IC=ARAY(KIT+1)
    ID=ARAY(KIT+2)
184 IF (IB-ARAY(KIT-KIP)) 111,142,142
111 KIND=KIND+1
    ARAY(KIT-KIP+3)=ARAY(KIT-KIP-1)
    ARAY(KIT-KIP+4)=ARAY(KIT-KIP)
    ARAY(KIT-KIP+5)=ARAY(KIT-KIP+1)
    ARAY(KIT-KIP+6)=ARAY(KIT-KIP+2)
    GO TO 108
142 IF (KIND) 114,115,114
114 ARAY(KIT-KIP+4)=IB
    ARAY(KIT-KIP+3)=IA
    ARAY(KIT-KIP+5)=IC
    ARAY(KIT-KIP+6)=ID
115 KTIME=KTIME+1
    HIM=HIM+4
    IF (HLOOP-KTIME-1) 116,116,117
117 KIT=KIT+4
    GO TO 107
C   COUNTS K OF CERTAIN KIND FOR THIS VALUE OF H
116 HIM=HIM+4
    KIZ=0
    KLP=0
179 KLOOP=0
119 IF (ARAY(KIW)-KIZ) 120,120,121
120 KLOOP=KLOOP+1
    KLP=KLP+1
    KIW=KIW+4
    IF (HLOOP-KLP) 160,160,119
C   SETS KOUNT # 4 IF HAS READ ALL K IN RANGE OF H
160 KOUNT=4
    IF (KLOOP) 139,139,123
121 IF (KLOOP) 122,122,123
```

C. Subroutine SORGEN (continued-3)

```
122 KIZ=KIZ+1
    IF (KIZ-30) 119,106,106
C   SORTS L FOR CERTAIN VALUE OF H AND K
123 IF (KLOOP-1) 235,235,189
235 LIM=LIM-4
    GO TO 135
189 LIT=IM+7+LIM
    LIY=LIT-4
    LTIME=0
127 LIND=0
    LIP=0
128 LIP=LIP+4
    IF (LIT-LIP-LIY) 134,130,130
130 IF (LIND) 186,187,186
187 IA=ARAY(LIT-2)
    IB=ARAY(LIT-1)
    IC=ARAY(LIT)
    ID=ARAY(LIT+1)
186 IF (IC-ARAY(LIT-LIP)) 131,152,152
131 LIND=LIND+1
    ARAY(LIT-LIP+2)=ARAY(LIT-LIP-2)
    ARAY(LIT-LIP+3)=ARAY(LIT-LIP-1)
    ARAY(LIT-LIP+4)=ARAY(LIT-LIP)
    ARAY(LIT-LIP+5)=ARAY(LIT-LIP+1)
    GO TO 128
152 IF (LIND) 134,135,134
134 ARAY(LIT-LIP+4)=IC
    ARAY(LIT-LIP+5)=ID
    ARAY(LIT-LIP+2)=IA
    ARAY(LIT-LIP+3)=IB
135 LTIME=LTIME+1
    LIM=LIM+4
    IF (KLOOP-LTIME-1) 136,136,137
137 LIT=LIT+4
    GO TO 127
136 LIM=LIM+4
    IF (KOUNT-4) 140,138,140
140 KIZ=KIZ+1
    GO TO 179
138 KOUNT=0
    IF (HOUNT-4) 141,139,141
141 IZ=IZ+1
    GO TO 180
C   OUTPUT ON PRINTER
139 WRITE (PRINTR,161)
161 FORMAT (' THIS IS THE HKL LIST AFTER SUBROUTINES
    I SORTA, SORTB, SORTC @)
    WRITE (PRINTR,144) NPAGES
    I1=1
    I2=2
    I3=3
    ICODE=1
    DO 150 I=1,NUMBER
    IM=IM+4
    RECNO=ARAY(IM)
    IH=ARAY(IM-3)
    IK=ARAY(IM-2)
    IL=ARAY(IM-1)
```

C. Subroutine SORGEN (continued-4)

```
      READ (11'RECNO) INDR,IS,ORDIND,PARITY,MULT,SINSQ,  
      1RTW,(AMPL(M),M=1,NAMPL),(FFR(M),M=1,NCRVES)  
C     TEST SIGNS OF H,K,L, REVERSE SIGNS IF 2 OR 3  
C     INDICES ARE NEGATIVE  
      IF(IH) 280,281,281  
0280 NH(I1)=-IH  
      IS=1  
      GO TO 282  
0281 IS=0  
      NH(I1)=IH  
0282 IF(IK) 283,284,284  
0283 NH(I2)=-IK  
      IS=IS+2  
      GO TO 297  
0284 NH(I2)=IK  
0297 IF(IL) 285,299,299  
0285 NH(I3)=-IL  
      IS=IS+4  
      GO TO 298  
0299 NH(I3)=IL  
0298 GO TO(287,295,288,289,290,292,294),IS  
0286 IS=1  
      GO TO 296  
0287 IS=2  
      GO TO 296  
0288 IH=NH(I1)  
      IK=NH(I2)  
      IL=-IL  
0289 IS=4  
      GO TO 296  
0290 IS=3  
      IH=NH(I1)  
      IK=-IK  
0291 IL=NH(I3)  
      GO TO 296  
0292 IS=2  
      IH=-IH  
0293 IK=NH(I2)  
      GO TO 291  
0294 IS=1  
      IH=NH(I1)  
      GO TO 293  
0295 IS=3  
296 IF (ICDDE-1) 257,261,257  
0261 ICODE=2  
      ORDIND=4  
      NREFNS=1  
      H(1)=NH(1)  
      H(2)=NH(2)  
      H(3)=NH(3)  
      GO TO 254  
C     ORDER OF REFLECTIONS  
0257 IF(NH(1)-H(1)) 247,248,247  
0248 IF(NH(2)-H(2)) 249,251,249  
0251 IF(NH(3)-H(3)) 252,253,252  
0253 ORDIND=0  
      GO TO 254  
0252 ORDIND=3
```

C. Subroutine SORGEN (continued-5)

```
0255 H(3)=NH(3)
      GO TO 254
0249 ORDIND=2
0256 H(2)=NH(2)
      GO TO 255
0247 ORDIND=1
      H(1)=NH(1)
      GO TO 256
C     PARITY AND MULTIPLICITY
0254 PARITY=4*MOD(H(I1),2)+2*MOD(H(I2),2)+MOD(H(I3),2)+1
      WRITE (PRINTR,146) IH,IK,IL,INDR,IS,ORDIND,PARITY,
1MULT,SINSQ,RTW,(AMPL(M),M=1,NAMPL)
      WRITE (PRINTR,147) (FFR(M),M=1,NCRVES)
      LINES=LINES-2
      WRITE (TAPEB) IH,IK,IL,INDR,IS,ORDIND,PARITY,MULT,
1SINSQ,RTW,(AMPL(M),M=1,NAMPL),(FFR(M),M=1,NCRVES)
      IF (LINES) 151,151,150
151 NPAGES=NPAGES+1
      LINES=55
      WRITE (PRINTR,144) NPAGES
150 CONTINUE
      WRITE (PRINTR,153) NUMBER
153 FORMAT ('1 NUMBER OF EQUIVALENT REFLECTIONS IS @,I4)
      RETURN
      END
```

REFERENCES

- Barth, T.W.F. (1926): Die Kristallographische Beziehung Zwischen Helvin und Sodalit. Norsk. Geol. Tidsskrift, 9, 40.
- Barth, T.W.F. (1932): The Structures of the Minerals of the Sodalite Family. Zeit. Krist., 83, 405.
- Evans, R.C. (1939): Crystal Chemistry, Cambridge Press
- Ferguson, R.B. (1960): The Low Temperature Phases of the Alkali Feldspars and Their Origin. Canadian Mineralogist, 6, 415.
- Ferguson, R.B.; Traill, R.; and Taylor ^{W.H.} (1958): The Crystal Structures of Low Temperature and High Temperature Albites. Acta Cryst., 11, 331.
- Gait, R.I. (1967): Computer Calculated Electrostatic Charge Distributions of the Structure of the Feldspars Low Albite, High Albite, and Anorthite. Thesis, Ph.D., University of Manitoba.
- Gait, R.I. and Ferguson, R.B. (1968): Electrostatic Charge Distributions in the Structure of Low Albite, $\text{NaAlSi}_3\text{O}_8$. Acta Cryst., accepted for publication.
- Gruner, J.W. (1931): Structures of Some Silicates. Amer. Min., 16, 452.

- Internationale Tabellen zur Bestimmung von Kristallstrukturen
(1935). Vol. II,; Gebruder Borntraeger, Berlin.
- International Tables for X-Ray Crystallography (1952). Vol. I.
Birmingham: Kynoch Press.
- International Tables for X-Ray Crystallography (1959). Vol. II.
Birmingham: Kynoch Press.
- International Tables for X-Ray Crystallography (1962). Vol. III.
Birmingham: Kynoch Press.
- Jaeger, F.M. (1929): Trans. Far. Soc., 25, 320.
- James, R.S. (1968): An Occurrence of Sodalite at the Princess
Quarry, Bancroft, Ontario. Canadian Mineralogist,
9, 574.
- Kotackowska, M. (1935): Arch. Min. Soc. Sci. Varsovie, 2, 170.
- Lons, V.J. and Schultz, H. (1967): Strukturverfeinerung von
Sodalith. Acta Cryst., 23, 434.
- Miser, H.D. and Glass, J.J. (1941): Fluorescent Sodalite and
Hackmanite from Magnet Cove, Arkansas. Amer. Min.,
26, 437.
- Nelson, J.B. and Riley, D.P. (1945): Proc. Phys. Soc. (London),
57, 160.
- Nuffield, E.W. (1966): X-Ray Diffraction Methods, Wiley and
Sons, New York.

- Pauling, L. (1930): The Structure of Sodalite and Helvite, Zeit. Krist., 74, 213.
- Phillips, D.C. (1954): On the Visual Estimation of X-ray Reflection Intensities from Upper-Level Weissenberg Photographs. Acta Cryst., 7, 746.
- Singh, A.K. (1967): Zeit. Krist., 124, 367.
- Smith, J.V. and Bailey, S.W. (1963): Acta Cryst., 16, 801.
- Taylor, W.H. (1934): Proc. Roy. Soc. London, Ser. A, 145, 80.
- Thomson, T. (1810): Chemical Analysis of Sodalite, a New Mineral from Greenland, Royal Soc. Edin. Trans., 5, 387.
- Walker, T.L. and Parsons, A.L. (1935): Evanescent Pink Sodalite and Associated Minerals from Dungannon Township, Ontario, University of Toronto Studies, Geol. Series, 20, 5.