

Lawrence Berkeley National Laboratory

Recent Work

Title

THE CRYSTAL STRUCTURE OF VANADYL BISACETYLACETONATE

Permalink

<https://escholarship.org/uc/item/28q580hp>

Author

Dodge, Richard Patrick.

Publication Date

1958-03-31

UNIVERSITY OF
CALIFORNIA

*Radiation
Laboratory*

THE CRYSTAL STRUCTURE OF VANADYL
BISACETYLACETONATE

TWO-WEEK LOAN COPY

*This is a Library Circulating Copy
which may be borrowed for two weeks.
For a personal retention copy, call
Tech. Info. Division, Ext. 5545*

DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.

UNIVERSITY OF CALIFORNIA

Radiation Laboratory
Berkeley, California

Contract No. W-7405-eng-48

THE CRYSTAL STRUCTURE OF
VANADYL BISACETYLACETONATE

Richard Patrick Dodge
(Thesis)

March 31, 1958

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

- A. Makes any warranty or representation, express or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or
- B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission to the extent that such employee or contractor prepares, handles or distributes, or provides access to, any information pursuant to his employment or contract with the Commission.

THE CRYSTAL STRUCTURE OF
VANADYL BISACETYLACETONATE

Contents

Abstract	3
Introduction	4
Experimental	4
Structure Determination	5
Discussion of Structure	8
Summation of Fourier Series	11
Acknowledgments	16
Appendix	17
Bibliography	27

THE CRYSTAL STRUCTURE OF
VANADYL BISACETYLACETONATE

Richard Patrick Dodge

Radiation Laboratory
and

Department of Chemistry and Chemical Engineering
University of California, Berkeley, California

March 31, 1958

ABSTRACT

The crystal structure of the chelate vanadyl bisacetylacetonate has been determined by x-ray diffraction. The compound is not a polymer in the solid but forms separate molecules, and there is no intermolecular bonding other than the usual Van der Waals type. The main feature of interest in the result is the geometrical arrangement of oxygen atoms about the central vanadium atom. The ligand oxygens form a square plane perpendicular to the vanadyl ion; however, the vanadium atom is not in this plane but slightly above it.

The structure determination was carried out by Fourier methods and refinement of the parameters was done by the least-squares method. Fourier summations were computed on the IBM 701 calculator, and the program used for these calculations is briefly described.

INTRODUCTION

The structure of the compound vanadyl bisacetylacetonate was studied to establish the geometry of the bonds about five-coordinated vanadium (IV). Jones has reported work on the VO^{++} complexes with β -diketones, and in his discussion of vanadyl bisacetylacetonate and its reactions he assumed a square planar arrangement of four oxygen atoms about the central vanadium atom, with the bond to the fifth oxygen presumably perpendicular to the plane of the other four.¹ From the chemical evidence, it was impossible to rule out the trigonal bipyramid arrangement of the five bonds. Recent work on the electron spin-resonance spectra of the compound indicates that the bipyramid structure is improbable.² The geometry of the five central bonds of vanadyl bisacetylacetonate can also be expected for similar compounds such as vanadyl tetraphenylporphine.³

EXPERIMENTAL

Single crystals of vanadyl bisacetylacetonate were provided by Rue L. Belford. The crystals were blue-green in color and those selected for study averaged 0.1 mm in thickness. The crystals belonged to the class $\bar{1}$ and exhibited only the principal faces, so that the crystals looked very much like the unit cell.

Multiple-film Weissenberg pictures were taken with rotation about the chosen *a* axis. A second set of pictures taken about the *b* axis was used to scale the intensities of the first set of pictures. Precession photographs were made of all three of the principal planes of the reciprocal lattice. The unit-cell constants were obtained from these photographs and quartz-calibrated zero-level Weissenberg pictures. The intensities of the reflections were evaluated by visual comparison with a set of standard intensities.

The lattice was found to be triclinic with the following values for the reciprocal cell:

$$\begin{array}{lll} a^* = 0.1459 & b^* = 0.1349 & c^* = 0.0957 \\ \alpha^* = 101.0^\circ & \beta^* = 103.5^\circ & \gamma^* = 109.5^\circ \end{array}$$

If we apply the transformation formulas, the following values are found for the direct triclinic cell:

$$\begin{array}{ll} a = 7.53 \pm 0.02 \text{ \AA} & \alpha = 73.0^\circ \\ b = 8.23 \pm 0.03 \text{ \AA} & \beta = 71.3^\circ \\ c = 11.24 \pm 0.04 \text{ \AA} & \gamma = 66.6^\circ \end{array}$$

The specific gravity of the compound was found by suspension to be 1.49. Using the molecular weight of 265.15 and the cell constants, we obtain a value of 2.01 molecules per unit cell.

STRUCTURE DETERMINATION

Early work on the structure determination involved working with projections along the [100] and [010] axes. The Patterson function was calculated for both of these projections for the purpose of locating the vector between two vanadium atoms. Further work by Fourier methods in two dimensions met with no success largely because of lack of resolution especially along [010].

Successful approach to the solution of the problem began with a calculation of the Patterson function in three dimensions using the scaled data and the Fourier program coded for the IBM 701 calculator. This calculation gave a very high maximum which located the vector between the two vanadium atoms unambiguously. The origin of the unit cell used for further calculations was chosen as halfway between the vanadium atoms. This origin was assumed to be a center of symmetry and one molecule was therefore related to the other molecule in the unit cell by symmetry. An attempt was then made to assign correct signs to at least a part of the terms in the Fourier series for the electron density. The method adopted is the "heavy atom" method.

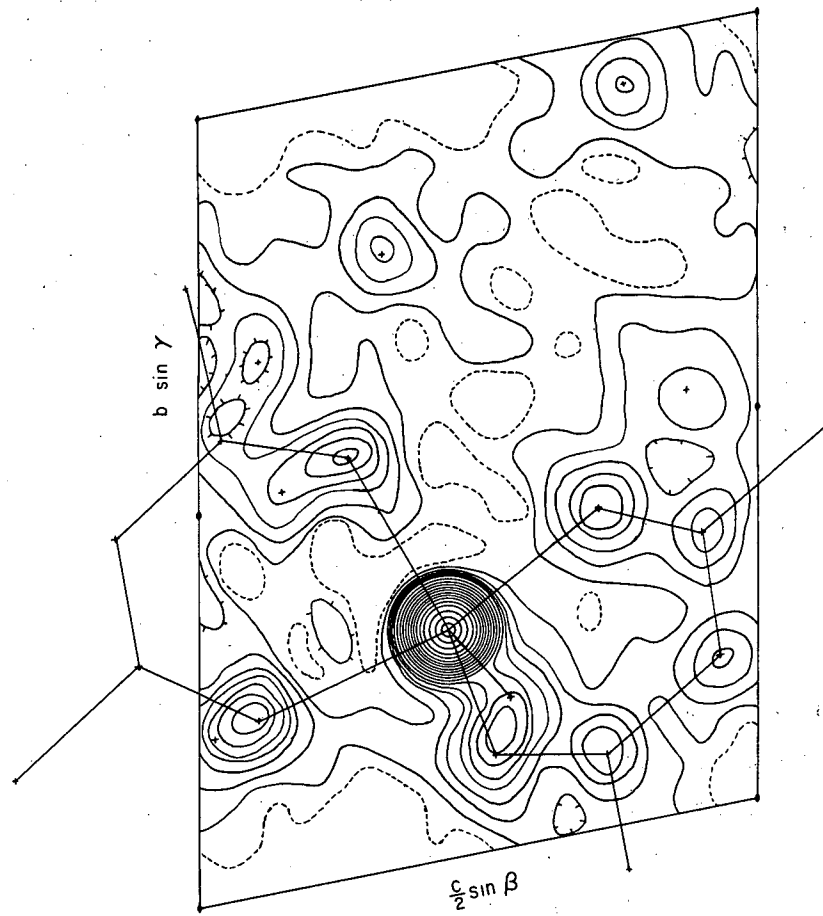
By the use of only the known position of the vanadium atom, structure factors were calculated for all of the 1164 observed reflections. Calculation of structure factors was accomplished on the IBM 650

machine by modifying the least-squares II program of M. Senko for the space group $P\bar{1}$.⁴ An arbitrary criterion was adopted to select terms that appeared to have a good probability of having the correct sign. If a calculated magnitude for F_c satisfied the relation $2 > F_c/F_o > 0.5$ the calculated sign for this value was used as the sign of F_o in the Fourier series for the electron density. Otherwise the term was omitted from the calculation.

The success of this method depended in part upon knowing the correct value of the scaling factor between the calculated and observed structure factors. The scaling factor used at this stage was obtained by comparison with calculated structure factors and turned out to be within 25% of the correct value. The terms thus chosen were then used in calculating the electron density Fourier series, $\rho(xyz)$.

The resulting calculation showed several peaks within a 2.0\AA sphere of the vanadium atom which were used in constructing a five-atom trial structure. Structure factors were computed for this partial trial structure but using only the $F(Ok1)$ data. Again the arbitrary criterion mentioned above was used to select terms to be used in another electron density projection along $[100]$. This projection showed some of the features expected in the final structure and only two more $\rho(yz)$ calculations were needed to give the final result shown in Fig. 1. Because the structure is not completely resolved in this direction, the bonding shown in the figure was established only after completion of three dimensional refinement.

An attempt was then made to solve the projection along $[010]$ in the same manner as was done with the projection along $[100]$. This attempt failed because the molecules in this projection are seen practically on edge. With this knowledge, i. e. that the chelate rings are planar and also coplanar or nearly so, a rough assignment of x parameters was made. Structure factors based on these rough atomic parameters were then calculated for all of the 1164 observed reflections. By comparing the calculated and observed structure factors from the successful projection along $[100]$, it was possible to use an accurate scaling factor. Individual temperature factors were not used at this stage.



MU-14978

Fig. 1. Fourier projection along the [100] axis. Contours are drawn at $2 e/A^2$ intervals. Dotted contour is at one e/A^2 .

A large majority of the F_c from this structure-factor calculation met the requirement $2 > F_c/F_o > 0.5$. The terms thus selected were then used in a calculation of $\rho(xyz)$. This was not done for the complete asymmetric unit, but $\rho(xyz)$ was calculated in a fine grid only in the regions of interest for the purpose of obtaining good x parameters for all atoms. The structure thus obtained gave for the value of $R = \Sigma(|F_o| - |F_c|) / \Sigma|F_o| = 0.207$. At this point the structure parameters, individual isotropic temperature factors, and scaling factor were refined by the least squares II program of M. Senko⁴ modified for the space group $P\bar{1}$. Five cycles of refinement were needed to give a final value of $R = 0.126$. Throughout the refinement, weighting factors were assigned to the various reflections as proposed by Hughes.⁵ The assigned factors were $w = 1/F_o^2$ if $I_o > 16 I_{min}$ and, if $I_o < 16 I_{min}$, $w = 1/16 F_{min}^2$, where F_{min} is a representative value, and I_o is the uncorrected observed intensity. Unobserved reflections were used in the calculation; for values of $F_{min} > F_c$, $F_{min} - F_c$ was set equal to zero for the purpose of the calculation and for values of $F_c > F_{min}$, this difference was set equal to $-F_c$.

The program was modified, and the final refinement was repeated to evaluate the standard deviations of atomic coordinates taking account of cross terms between all three coordinates of each atom. The modification consisted of computing $\Sigma F_x F_y$ and $\Sigma F_y F_z$ in addition to the relevant sums already calculated by Senko's program, and solving the determinants as given by Cruickshank⁶ for the standard deviations of the atomic coordinates.

The final atomic parameters with temperature factors and standard deviations are given in Table I. The final values for calculated and observed structure factors will be found in the Appendix.

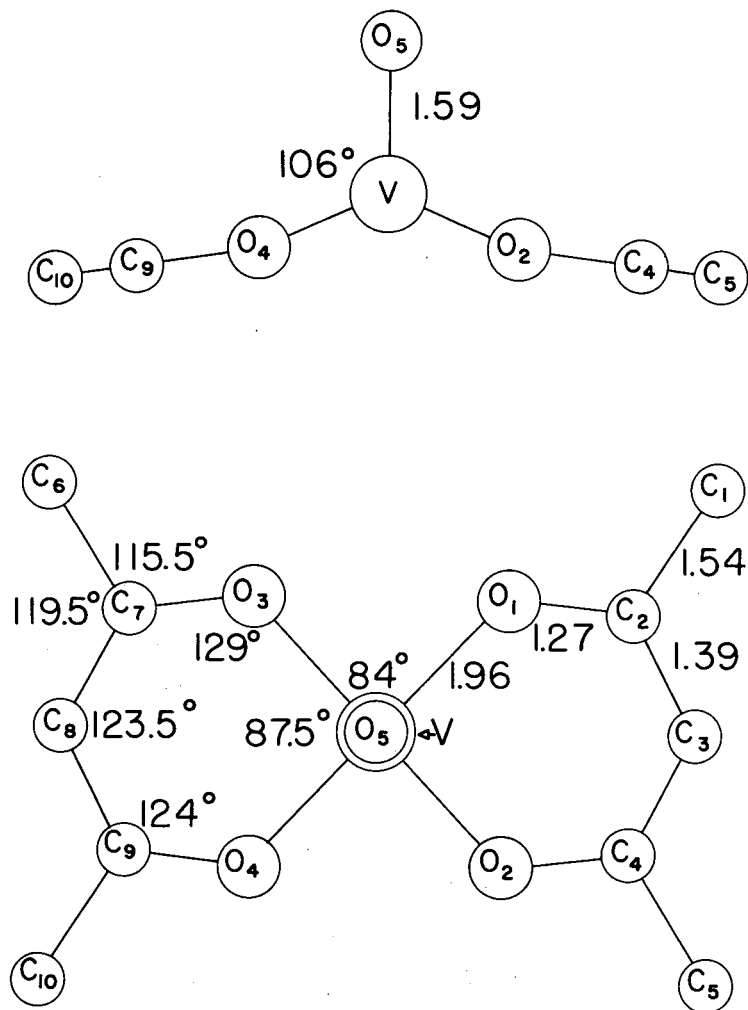
DISCUSSION OF STRUCTURE

Figure 2 gives two views of the molecule together with the average structure parameters for the entire molecule. The distances given for a bond length are the average values for all such bond types in the molecule if we assume C_{2v} symmetry, and the bond angles in Fig. 2 were

Table I

Final Structure Parameters for Vanadyl Bisacetylacetonate

Atom	B_n	x	y	z	σ_x	σ_y	σ_z
V	2.61	0.1440	0.2901	0.2227	0.0004	0.0003	0.0002
O ₁	3.12	0.9478	0.4110	0.3588	0.0016	0.0012	0.0008
O ₂	2.97	0.9941	0.1228	0.2671	0.0015	0.0011	0.0008
O ₃	3.15	0.1486	0.5359	0.1349	0.0015	0.0011	0.0008
O ₄	3.34	0.2287	0.2277	0.0540	0.0016	0.0012	0.0008
O ₅	3.58	0.3350	0.1894	0.2807	0.0016	0.0012	0.0008
C ₁	3.91	0.2798	0.5409	0.4369	0.0027	0.0020	0.0014
C ₂	2.84	0.8194	0.3529	0.4509	0.0022	0.0016	0.0011
C ₃	3.48	0.7930	0.1942	0.4655	0.0025	0.0018	0.0013
C ₄	3.01	0.8703	0.0923	0.3676	0.0024	0.0017	0.0012
C ₅	3.42	0.7913	0.9444	0.3816	0.0025	0.0018	0.0013
C ₆	4.25	0.7818	0.2090	0.0126	0.0029	0.0022	0.0015
C ₇	2.73	0.2244	0.5926	0.0193	0.0023	0.0016	0.0011
C ₈	3.01	0.7059	0.5084	0.0759	0.0024	0.0017	0.0012
C ₉	2.96	0.7149	0.6813	0.0550	0.0023	0.0016	0.0012
C ₁₀	3.91	0.6234	0.7880	0.1662	0.0027	0.0020	0.0014



MU-14979

Fig. 2. Interatomic distances (right) and intramolecular angles (left).

calculated from these average values. The individual bond lengths are given in Table II together with the estimated standard deviations. Because no bond length deviates from the average by more than twice the standard deviation, the molecule has the symmetry C_{2v} within the error of the determination. This symmetry did not occur crystallographically so that all the bond lengths were measured four times with the exception of the closer V-O bond.

The bond distance in the vanadyl ion ($V-O_5$) is of special interest, as no other measurement of the vanadyl ion is known to the author. A bond distance of 1.56\AA in V_2O_5 occurs between vanadium and oxygen, however, the oxidation state is not the same.

There are no close approaches between molecules of vanadyl bisacetylacetonate in the solid, the nearest approach being 3.50\AA . The vanadium atom has its nearest neighboring vanadium atom at 5.71\AA ; the next distance being 6.96\AA . The vanadyl oxygen (O_5) has its nearest nonbonded vanadium atom at a distance of 6.27\AA ; the next being at 6.72\AA .

The main structural feature of vanadyl bisacetylacetonate is the geometrical arrangement of the oxygen atoms that gives rise to a bending of the rings away from the coplanarity that might otherwise be expected. The acetylacetonate molecules in the structure (minus one proton apiece) are themselves planar as expected, but the two planes form an angle of 17.5° . The molecular parameters are rather similar to those of copper bisacetylacetonate, which has a crystallographic center of symmetry at the copper position with the oxygen atoms forming a square in the planar molecule.⁷ The fifth oxygen in the vanadium compound destroys this symmetry and also repels the other oxygen atoms to an average distance of 2.8\AA , which gives the vanadium the appearance of being pushed up out of the plane of the oxygen atoms.

SUMMATION OF FOURIER SERIES

During the course of this problem a program was planned and written in collaboration with Dr. D. H. Templeton and Dr. R. E. Jones to calculate Fourier Series on the IBM 701 calculator. This machine has a magnetic-core type high-speed memory of 4096 full words (35

Table II

Interatomic Bond Distances in Angstrom Units		
Bond	Distance	σ
V - O ₅	1.588	0.011
V - O ₁	1.947	0.012
V - O ₂	1.971	0.012
V - O ₃	1.982	0.012
V - O ₄	1.945	0.012
C ₂ - O ₁	1.292	0.017
C ₄ - O ₂	1.246	0.017
C ₇ - O ₃	1.270	0.017
C ₉ - O ₄	1.274	0.017
C ₇ - C ₈	1.387	0.020
C ₉ - C ₈	1.399	0.020
C ₂ - C ₃	1.353	0.020
C ₄ - C ₃	1.413	0.021
C ₇ - C ₆	1.551	0.022
C ₉ - C ₁₀	1.559	0.021
C ₂ - C ₁	1.561	0.022
C ₄ - C ₅	1.503	0.021

binary bits and sign) with additional drum storage of 8192 full words. Input is by punched card, and output is by type-716 alphabetical printer. Tape units are available on this machine, but they were not needed for this problem.

The problem calculates the following:

$$\frac{1}{2} \rho (X_1 X_2 X_3) = \frac{1}{2} A (000) + \sum_{H_1=0}^{\infty} \sum_{H_2=-\infty}^{\infty} \sum_{H_3}^{\infty} \left[A (H_1 H_2 H_3) \cos 2\pi (H_1 X_1 + H_2 X_2 + H_3 X_3) + B (H_1 H_2 H_3) \sin 2\pi (H_1 X_1 + H_2 X_2 + H_3 X_3) \right].$$

The summation extends over one-half of reciprocal space, so that if hkl is used, $\bar{h}\bar{k}\bar{l}$ is omitted. The primes on the summation signs indicate that special reflections are omitted such as $0\bar{k}l$, $0\bar{k}0$, $00\bar{l}$, and 000 .

Input to the program consists of data cards, with one reflection to a card, and cards that modify the basic program for a particular problem. The modifying cards specify the range of the indices H_i and the range and fineness of the coordinates X_i over which the series is to be evaluated. Since the program calculates the sum by the general formula, higher symmetries than triclinic require a redundant input because the reflections have a Laue symmetry that is not used in the formula. As a part of the basic program, there is a program to generate just half of the reflections related by Laue symmetry from a single independent input-data card. The space group of the problem is specified by a system of code numbers on the modifying cards.

The electron density is calculated in a manner similar to that of Trueblood and coworkers, i. e., the result is obtained from the expanded form of the triclinic formula and in three distinct stages.⁸ After the data have been stored in the memory and the equivalent reflections have been generated, the coefficients T_1 and T_2 are calculated:

$$T_1(H_1H_2X_3) = \sum_{H_3=-\infty}^{\infty} [A(H_1H_2H_3) \cos 2\pi H_3 X_3 + B(H_1H_2H_3) \sin 2\pi H_3 X_3]$$

and

$$T_2(H_1H_2X_3) = \sum_{H_3=-\infty}^{\infty} [B(H_1H_2H_3) \cos 2\pi H_3 X_3 - A(H_1H_2H_3) \sin 2\pi H_3 X_3].$$

These coefficients form a table in the second frame of the high-speed memory. The value of X_3 is specified by the modifying cards, and is the level of the cut in the unit cell. Thus, a complete three-dimensional calculation consists of a series of slices perpendicular to a chosen axis. The basic program is the complete calculation of one of these cuts. Provision is made for storing the input data in drum storage so that card reading is eliminated for additional calculations.

The coefficients T_1 and T_2 are located in the second frame, depending on the index limits specified for the problem, and are placed so that the resulting table can be scanned sequentially for the second stage of the calculation. This stage evaluates and stores the coefficients C and S as follows:

$$C(H_1X_2X_3) = \sum_{H_2=-\infty}^{\infty} [T_1(H_1H_2X_3) \cos 2\pi H_2 X_2 + T_2(H_1H_2X_3) \sin 2\pi H_2 X_2]$$

and

$$S(H_1X_2X_3) = \sum_{H_2=-\infty}^{\infty} [T_2(H_1H_2X_3) \cos 2\pi H_2 X_2 - T_1(H_1H_2X_3) \sin 2\pi H_2 X_2].$$

Since storage space in fast memory at this stage is limited, the table of C and S coefficients is computed in sections and stored in drum memory. After the table is complete, it is transferred to the second frame of fast memory replacing the table that is no longer needed. The final stage of the calculation computes the electron density:

$$\frac{1}{2} \rho(X_1X_2X_3) = \sum_{H_1=0}^{\infty} [C(H_1X_2X_3) \cos 2\pi H_1 X_1 + S(H_1X_2X_3) \sin 2\pi H_1 X_1].$$

This final table is calculated in sections and printed out on the printer.

The format of the output is a rectangular table with the $\rho(X_1X_2X_3)$ values easily found by means of headings identifying the coordinates X_1 and X_2 . Because the IBM 701 is a fixed-point machine, there is some difficulty in the possibility of overflow of the final answers. A provision is made in the program to rescale the answers and repeat the calculation in the case of overflow.

The over-all scheme of calculating the Fourier series allows the calculation of special cases such as Patterson series or Fourier projections with no change in the basic code and only a small change in the modifying cards. Thus, a two-dimensional summation results if only zero-level data are used. The only difference is that the first table is formed directly from the data with no trigonometric factors or summations, and this is done automatically.

The advantage in using a machine like the IBM 701 for this problem is the machine's high speed. In the structure determination of vanadyl bisacetylacetonate, each Fourier projection required 5 minutes or less for completion. A typical three-dimensional calculation like the Patterson function calculated at 8125 points required only 35 minutes to complete.

ACKNOWLEDGMENTS

I wish to express my thanks to Professor David H. Templeton for his guidance and suggestions throughout the course of this research, and also to other members of the research group who made the task easier, particularly, Mrs. Helena Ruben, Dr. M. Senko, and Dr. R. E. Jones.

For their part in the coding of the Fourier program, special thanks go to Dr. D. H. Templeton again for his general insight and continued encouragement and to Dr. R. E. Jones who coded the difficult problem of the calculation of equivalent reflections. This research was supported by the Atomic Energy Commission.

APPENDIX

Observed and Calculated Structure Factors

An unobserved F_o value, that is, a value less than the minimum possible value for the particular reflection to be observed, appears as a zero in this table. In a few cases, a reflection that was originally thought to be unobserved actually was found later to lie outside the region of reciprocal space that was photographed. This error accounts for discrepancies in cases such as $15\bar{3}$, $1\bar{5}2$, and $2\bar{4}2$.

h	k	l	F _o	F _c	h	k	l	F _o	F _c	h	k	l	F _o	F _c
0	0	2	25	38-	0	2	10-	0	25-	0	5	2-	118	120
0	0	3	45	57	0	2	11	55	41	0	5	3	104	98
0	0	4	130	121	0	2	11-	48	35	0	5	3-	0	30
0	0	5	97	97	0	2	12-	44	34	0	5	4	70	64-
0	0	6	78	73-	0	3	0	113	75-	0	5	4-	0	2
0	0	7	78	64-	0	3	1	54	46	0	5	5	60	49-
0	0	8	34	32	0	3	1-	114	106-	0	5	5-	0	21-
0	0	9	0	28	0	3	2	62	75-	0	5	6	0	7
0	0	10	58	46-	0	3	2-	162	150-	0	5	6-	129	121
0	0	11	0	8-	0	3	3	27	36-	0	5	7	64	74
0	0	12	52	42-	0	3	3-	0	10-	0	5	7-	68	63
0	0	13	42	29	0	3	4	0	6-	0	5	8	0	20
0	1	0	151	174-	0	3	4-	81	70	0	5	8-	0	25-
0	1	1	306	478-	0	3	5	228	248	0	5	9	41	32-
0	1	1-	181	222	0	3	5-	0	12-	0	5	9-	36	32-
0	1	2	33	25-	0	3	6	0	26	0	5	10	0	18-
0	1	2-	179	199-	0	3	6-	74	62-	0	5	11	51	46
0	1	3	226	248	0	3	7	100	103-	0	5	12	38	30
0	1	3-	81	86-	0	3	7-	86	82-	0	6	0	0	14-
0	1	4	53	61	0	3	8	0	24-	0	6	1	68	71
0	1	4-	22	15	0	3	8-	0	19	0	6	1-	70	59-
0	1	5	202	203-	0	3	9	68	58	0	6	2	0	14
0	1	5-	159	153	0	3	9-	53	50	0	6	2-	0	2-
0	1	6	77	63-	0	3	10	79	80	0	6	3	159	162-
0	1	6-	64	56	0	3	10-	57	48-	0	6	3-	59	61
0	1	7	67	49	0	3	11-	58	54-	0	6	4	40	28-
0	1	7-	0	11-	0	3	12	74	59-	0	6	4-	65	56
0	1	8	120	112	0	4	0	45	68	0	6	5	41	36-
0	1	8-	68	60-	0	4	1	51	29	0	6	5-	35	41-
0	1	9	97	92-	0	4	1-	176	166	0	6	6	51	53
0	1	9-	0	29-	0	4	2	94	96-	0	6	6-	72	75-
0	1	10-	53	44	0	4	2-	76	72	0	6	7	0	28-
0	1	11-	58	41	0	4	3	0	4-	0	6	7-	53	46
0	1	12-	0	16-	0	4	3-	133	132-	0	6	8	64	75-
0	1	13-	40	35-	0	4	4	152	152	0	6	9	0	5-
0	2	0	185	178-	0	4	4-	82	96-	0	6	10	0	26
0	2	1	152	134-	0	4	5	0	16	0	6	11	0	11
0	2	1-	122	85-	0	4	5-	51	33	0	7	0	0	19
0	2	2	83	73	0	4	6	51	55-	0	7	1	0	27
0	2	2-	135	142	0	4	6-	0	1	0	7	1-	0	16-
0	2	3	30	19	0	4	7	0	16-	0	7	2	79	84-
0	2	3-	81	77	0	4	7-	65	53-	0	7	2-	75	71-
0	2	4	380	394-	0	4	8	0	30	0	7	3	0	25
0	2	4-	111	98-	0	4	8-	63	58-	0	7	3-	59	51-
0	2	5	135	142-	0	4	9	0	26	0	7	4	62	67
0	2	5-	0	1-	0	4	9-	0	16	0	7	4-	0	24
0	2	6	100	115	0	4	10	70	64-	0	7	5	0	21
0	2	6-	0	29-	0	4	10-	52	56	0	7	5-	38	44
0	2	7	68	51	0	4	11	58	49-	0	7	6	0	12-
0	2	7-	74	70	0	4	11-	32	30-	0	7	7	55	51-
0	2	8	74	61-	0	4	13	31	32	0	7	8	0	34
0	2	8-	101	99	0	5	0	68	49-	0	7	9	64	56
0	2	9	50	50-	0	5	1	0	9	0	7	10	0	5-
0	2	9-	0	14-	0	5	1-	36	35-	0	7	11	54	51-
0	2	10	36	30	0	5	2	103	83	0	8	0	0	35-

h	k	l	F _o	F _c	h	k	l	F _o	F _c	h	k	l	F _o	F _c
0	8	1	38	43-	1	1-	7	113	106-	1	3-	0	160	152-
0	8	1-	51	50	1	1-	7-	63	54-	1	3	1	66	48
0	8	2	0	14	1	1	8	89	91	1	3	1-	88	65
0	8	2-	34	33	1	1	8-	44	38-	1	3-	1	106	87-
0	8	3	54	46	1	1-	8	77	75-	1	3-	1-	60	55-
0	8	4	33	27	1	1	9-	32	26-	1	3	2	55	25-
0	8	5	0	22-	1	1-	9-	91	89-	1	3	2-	200	170-
0	8	6	0	23-	1	1-	10	51	54	1	3-	2	129	135-
0	8	7	36	34	1	1-	10-	50	42-	1	3-	2-	179	161
0	9	1	33	34	1	1	11	113	104	1	3	3	0	19
0	9	2	0	23	1	1	11-	53	38	1	3	3-	110	90-
0	9	3	0	6-	1	1-	11-	73	58-	1	3-	3	87	88
0	9	4	29	28-	1	1	12	39	38	1	3-	3-	130	112-
					1	1-	12	35	19-	1	3	4	54	56
1	0	2	250	283-	1	1-	13-	21	19	1	3	4-	93	80
1	0	3	268	252	1	2	0	78	56-	1	3-	4	104	87
1	0	3-	179	181-	1	2-	0	48	37	1	3-	4-	116	120-
1	0	4	81	61	1	2	1	205	192	1	3	5	139	122
1	0	5	47	25-	1	2	1-	118	136-	1	3	5-	71	62
1	0	5-	169	149	1	2-	1	67	65	1	3-	5	57	39-
1	0	6	60	49-	1	2-	1-	169	150-	1	3	6	103	99-
1	0	7	37	33-	1	2	2	104	92-	1	3	6-	0	2-
1	0	7-	103	93-	1	2	2-	45	16	1	3-	6	68	67-
1	0	8	74	75	1	2-	2	95	95	1	3-	6-	102	93
1	0	8-	56	51	1	2-	2-	0	25	1	3	7	61	60-
1	0	9	59	58	1	2	3	119	97-	1	3	7-	90	77-
1	0	9-	45	40	1	2	3-	120	107	1	3	8	0	29
1	0	10	108	112-	1	2-	3	53	43-	1	3	8-	45	47-
1	0	10-	79	64	1	2-	3-	196	177	1	3-	8	60	60
1	0	11	31	28-	1	2	4	0	21-	1	3-	8-	81	85-
1	0	12-	55	39-	1	2	4-	108	84-	1	3	9	50	50
1	0	13	38	29	1	2-	4	81	68-	1	3	9-	42	51
1	1-	0	124	114	1	2-	4-	230	214-	1	3	10-	31	25-
1	1-	1	369	417	1	2	5	64	52	1	3-	10	32	27-
1	1-	1-	181	148-	1	2	5-	92	80-	1	3-	10-	110	88
1	1	2	219	236	1	2-	5-	127	131-	1	3	11	64	44-
1	1	2-	321	274	1	2	6	229	240	1	3	11-	37	27-
1	1-	2	270	230-	1	2	6-	156	115-	1	3-	11-	31	35
1	1-	2-	207	183-	1	2	7	90	86-	1	3	12	58	45-
1	1	3	184	157	1	2	7-	0	29	1	3-	12	45	47
1	1	3-	0	17-	1	2-	7	65	57	1	3-	12-	44	34-
1	1-	3	34	22-	1	2-	7-	189	184	1	4	0	209	184-
1	1-	3-	32	41	1	2	8	109	98-	1	4-	0	110	92
1	1	4	68	66-	1	2	8-	103	101	1	4	1	150	132-
1	1	4-	21	3	1	2	9	70	70-	1	4	1-	181	163
1	1-	4-	272	256	1	2-	9	77	75-	1	4-	1	65	45
1	1	5	168	151-	1	2-	9-	52	51-	1	4-	1-	65	65
1	1	5-	52	39	1	2	10-	37	26-	1	4	2	72	55-
1	1-	5	42	25	1	2-	11	46	47	1	4	2-	54	26
1	1	6	0	8-	1	2-	11-	46	43	1	4-	2	43	38
1	1	6-	117	100	1	2	12	49	44-	1	4-	2-	56	37-
1	1-	6	52	40-	1	2	12-	38	33	1	4	3	72	63
1	1-	6-	203	201-	1	2	13	53	44-	1	4	3-	95	82-
1	1	7	109	110	1	2-	13-	14	17-	1	4-	3	110	109-
1	1	7-	106	96	1	3	0	317	300	1	4-	3-	30	22-

h	k	l	F _o	F _c	h	k	l	F _o	F _c	h	k	l	F _o	F _c
1	4	4	51	36	1	5	13	37	29-	1	7	11	41	32-
1	4	4-	89	76-	1	6	0	0	7-	1	8	0	42	42-
1	4-	4	67	62	1	6-	0	70	62-	1	8-	0	26	29
1	4-	4-	54	40	1	6	1	81	75	1	8	1-	30	28
1	4	5	161	166-	1	6	1-	35	34-	1	8-	1-	53	59-
1	4	5-	0	19	1	6-	1	65	58-	1	8	2	31	31
1	4-	5	96	96	1	6-	1-	54	50	1	8	2-	28	30
1	4-	5-	82	80	1	6	2	0	24	1	8-	2-	38	43-
1	4	6	0	24-	1	6	2-	48	42-	1	8	3	36	43
1	4	7	69	61	1	6-	2	52	62	1	8	3-	30	34
1	4-	7	52	58-	1	6-	2-	0	10-	1	8-	3	46	53-
1	4-	7-	44	39-	1	6	3	109	101-	1	8-	3-	27	24
1	4	8	0	15	1	6	3-	0	18-	1	8-	4-	39	35
1	4	8-	74	64-	1	6-	3	60	68	1	8	5-	31	33-
1	4	9	51	50-	1	6-	3-	99	99-	1	8-	7-	32	29
1	4-	9	61	53	1	6	4	123	118	1	8-	8-	38	38
1	4-	9-	98	92	1	6	4-	63	71	1	8	9	85	42-
1	4	10	55	48-	1	6-	4	31	32-	1	9	1	46	35
1	4	10-	51	45	1	6-	4-	88	77-	1	9	1-	31	37-
1	4-	11	42	39-	1	6	5	161	171	1	9-	2-	32	31
1	4-	11-	85	77-	1	6	5-	56	53-	1	9	3	52	48-
1	4	12	72	48	1	6	6	36	31	1	9-	3-	46	51
1	4	13	51	40	1	6	6-	50	45-	1	9	4	33	26-
1	5	0	107	99-	1	6-	6-	41	34	1	9	8	38	40-
1	5-	0	55	45-	1	6	7	52	36-	1	10	2	21	28-
1	5	1	56	48	1	6-	7	79	85	1	10	3	27	23
1	5	1-	53	47-	1	6	8	45	41-	1	10	4	38	26
1	5-	1	41	26	1	6-	8-	37	29-					
1	5-	1-	74	68-	1	6	9	64	67	2	0	0	298	317-
1	5	2	145	139	1	6-	9-	42	41-	2	0	1	205	248-
1	5	2-	85	82	1	6	10	50	40	2	0	1-	31	7-
1	5-	2	0	82	1	6	11	30	29-	2	0	2	373	581-
1	5-	2-	82	76	1	6-	11-	22	20	2	0	2-	89	83
1	5	3	0	12-	1	6	12	30	20-	2	0	3	120	103
1	5	3-	0	98	1	7	0	48	35	2	0	3-	88	98-
1	5-	3	50	39-	1	7-	0	61	61	2	0	4	0	13
1	5-	3-	106	106	1	7	1-	37	36	2	0	4-	133	126-
1	5	4	214	217-	1	7-	1	50	46-	2	0	5	107	95-
1	5	4-	35	30	1	7-	1-	84	89	2	0	5-	0	7
1	5-	4	99	115-	1	7	2	68	64-	2	0	6	79	62-
1	5-	4-	77	76	1	7	2-	0	12-	2	0	6-	102	98
1	5	5	45	47-	1	7-	2	49	53-	2	0	7	39	29
1	5	5-	37	41-	1	7-	2-	57	69-	2	0	7-	0	8-
1	5	6	29	34	1	7	3	37	37-	2	0	8	89	98
1	5	6-	72	54	1	7	3-	66	54-	2	0	8-	23	8-
1	5	7	0	36	1	7	4	81	93	2	0	9	0	1-
1	5	7-	60	46	1	7-	4	51	49	2	0	9-	0	23-
1	5	8	51	55-	1	7	5	52	47-	2	0	10	83	78-
1	5-	8	81	69-	1	7	5-	31	41	2	0	10-	90	68
1	5-	8-	44	49	1	7-	5-	35	42	2	0	11	0	21
1	5	9	0	10	1	7-	6	28	28-	2	0	11	0	21
1	5	10	31	39	1	7-	7-	53	56-	2	0	11-	0	13
1	5	10-	27	26-	1	7	8	63	58	2	0	12	86	82
1	5-	10	27	29	1	7	9	37	37	2	0	12-	35	29-
1	5	11	46	34	1	7-	9-	23	21	2	1-	0	68	51

h	k	l	F _o	F _c	h	k	l	F _o	F _c	h	k	l	F _o	F _c
2	1	1-	78	87-	2	2-	2-	114	106	2	3-	3-	123	106-
2	1-	1	221	210	2	2	3	42	43	2	3	4	54	47-
2	1-	1-	29	28	2	2	3-	0	2-	2	3	4-	0	11
2	1	2	177	295	2	2-	3	111	106-	2	3-	4	0	6
2	1	2-	49	45	2	2-	3-	195	197	2	3-	4-	70	66-
2	1-	2	65	62-	2	2	4	156	157	2	3	5	154	141-
2	1-	2-	133	113-	2	2	4-	159	161	2	3	5-	131	131
2	1	3	26	38	2	2-	4	96	109-	2	3-	5	89	95-
2	1	3-	30	26	2	2-	4-	104	109	2	3-	5-	42	35-
2	1-	3	120	220-	2	2	5	93	75	2	3	6	133	128-
2	1-	3-	89	91-	2	2	5-	0	7	2	3	6-	64	68
2	1	4	37	29-	2	2-	5	0	21	2	3-	6	0	18
2	1	4-	0	6-	2	2-	5-	78	64-	2	3-	6-	59	46
2	1-	4	199	195	2	2	6	179	191	2	3	7	60	58-
2	1-	4-	119	98	2	2	6-	159	149-	2	3	7-	66	62-
2	1	5	65	63-	2	2-	6	122	118	2	3-	7	82	88
2	1	5-	156	146-	2	2-	6-	60	62-	2	3-	7-	57	57
2	1-	5	20	8	2	2	7	41	26-	2	3	8	62	58
2	1-	5-	111	108	2	2	7-	65	68-	2	3	8-	0	8-
2	1	6	65	46	2	2-	7	49	58-	2	3-	8	0	4
2	1	6-	0	11	2	2-	7-	45	52	2	3-	8-	74	73-
2	1-	6	84	102-	2	2	8	76	76-	2	3	9	32	32
2	1-	6-	109	112-	2	2	8-	0	50	2	3	9-	75	59
2	1	7	150	166	2	2-	8	85	78-	2	3-	9	0	15-
2	1	7-	115	115	2	2-	8-	49	45	2	3-	9-	71	69-
2	1-	7	127	127-	2	2	9	0	25	2	3	10	49	41-
2	1-	7-	0	7-	2	2	9-	55	5	2	3-	10-	35	33
2	1	8	56	62-	2	2-	9	0	25-	2	3	11	56	57-
2	1	8-	0	4-	2	2-	9-	0	1-	2	3-	11	20	20
2	1-	8	0	11	2	2	10	57	52	2	3-	11-	51	37
2	1-	8-	0	32-	2	2	10-	37	32-	2	3	12	0	12-
2	1	9	61	56-	2	2-	10	0	2	2	3	13	51	41
2	1	9-	72	57-	2	2-	10-	63	48-	2	4	0	334	311-
2	1-	9	87	99	2	2	11	0	21-	2	4-	0	45	48
2	1-	9-	73	54	2	2	11-	29	21-	2	4	1	220	182-
2	1	10	0	10-	2	2-	11	0	1	2	4	1-	52	45
2	1	10-	59	39-	2	2-	11-	0	1	2	4-	1	102	85-
2	1-	10	0	9	2	2	12	59	49-	2	4-	1-	103	102
2	1-	10-	0	14	2	2-	12	31	35-	2	4	2	138	130
2	1	11	97	99	2	2-	12-	20	17	2	4	2-	172	177
2	1	11-	39	28	2	2	13	0	7-	2	4-	2	0	87-
2	1-	11	71	59-	2	2	14	35	35	2	4-	2-	68	57-
2	1-	11-	61	56-	2	3	0	193	169	2	4	3	0	16
2	1	12	0	17-	2	3-	0	115	100-	2	4	3-	36	33
2	1	12-	43	37	2	3	1	209	206-	2	4-	3	34	30-
2	1	13	48	44-	2	3	1-	245	242	2	4-	3-	70	82-
2	2	0	153	166	2	3-	1	24	33	2	4	4	99	97-
2	2-	0	41	17	2	3-	1-	30	38-	2	4	4-	59	47-
2	2	1	180	167	2	3	2	123	63	2	4-	4	79	91
2	2	1-	52	67-	2	3	2-	64	61-	2	4-	4-	31	24-
2	2-	1	160	149	2	3-	2	106	107	2	4	5	89	76-
2	2-	1-	76	65-	2	3-	2-	105	109	2	4	5-	0	1
2	2	2	117	125-	2	3	3	93	99	2	4-	5	40	49
2	2	2-	138	121-	2	3	3-	108	95-	2	4-	5-	35	38
2	2-	2	121	90	2	3-	3	130	129	2	4	6	0	16

h	k	l	F _o	F _c	h	k	l	F _o	F _c	h	k	l	F _o	F _c
2	4	6	37	16	2	5-	9-	0	16	2	7	4	36	39
2	4	6-	57	62	2	5	10	43	39	2	7	4-	37	37-
2	4-	6	0	12-	2	5-	10-	42	33-	2	7-	4-	38	38-
2	4-	6-	46	48	2	5	11	0	6-	2	7	5	108	115-
2	4	7	49	57	2	5	12	48	35-	2	7	5-	0	1-
2	4	7-	33	35	2	5	13	28	25-	2	7-	5-	43	43
2	4-	7	0	11	2	6	0	68	64	2	7	6	57	62-
2	4-	7-	37	26-	2	6-	0	57	79-	2	7	6-	40	28
2	4	8-	26	35-	2	6	1	36	3-	2	7-	6-	0	12
2	4-	8	65	67	2	6	1-	0	2	2	7	7	47	53
2	4-	8-	0	29-	2	6-	1	0	2	2	7-	7-	31	29-
2	4	9	46	40-	2	6-	1-	33	35	2	7	8	40	37
2	4-	9	34	27	2	6	2	49	53-	2	7-	8-	31	25-
2	4-	9-	63	52	2	6	2-	76	74-	2	7	9	0	33-
2	4	10	0	27	2	6-	2	56	56	2	7	10	79	65-
2	4-	10-	55	50	2	6-	2-	52	54	2	7	11	0	7
2	4	11	28	27	2	6	3	36	39-	2	7	12	39	35
2	4-	11-	35	28-	2	6	3-	28	29-	2	8	0	45	38-
2	4	12	63	51	2	6-	3	0	1-	2	8-	0	52	63
2	4	14	36	32-	2	6-	3-	73	72	2	8	1	0	3-
2	5	0	49	46-	2	6	4	126	126	2	8	1-	35	33-
2	5-	0	0	19	2	6	4-	74	74	2	8	2	59	66
2	5	1	133	133	2	6-	4	72	70-	2	8	2-	0	1
2	5	1-	112	89-	2	6-	4-	0	27-	2	8-	2-	36	39-
2	5-	1	62	51	2	6	5	181	194	2	8	3	0	4
2	5-	1-	98	107-	2	6	5-	0	3	2	8	3-	72	63
2	5	2	70	60	2	6-	5-	0	15-	2	8-	3-	25	23-
2	5	2-	89	81-	2	6	6	86	101-	2	8	4	46	44-
2	5-	2	0	33-	2	6	6-	40	34	2	8	4-	25	25
2	5-	2-	0	21-	2	6-	6-	0	17	2	8-	4-	23	29
2	5	3	159	161-	2	6	7	0	12	2	8	5	0	19-
2	5	3-	77	76	2	6	7-	47	39-	2	8	5-	26	24-
2	5-	3	52	44-	2	6-	7-	45	41	2	8	6	0	17
2	5-	3-	0	12	2	6	8	40	42	2	8	7	0	20
2	5	4	128	119-	2	6-	8-	0	21	2	8	8	29	29-
2	5	4-	93	95-	2	6	9	64	60	2	8	9	57	54-
2	5-	4	40	50-	2	6-	9-	28	25-	2	8	10	35	32
2	5-	4-	49	45	2	6	10	0	16	2	8	11	23	17
2	5	5	116	111	2	6	11	46	43-	2	9	2	51	43-
2	5	5-	77	80-	2	6	12	0	5	2	9	2-	43	36-
2	5-	5	46	53	2	6	13	32	25	2	9	3	54	43-
2	5-	5-	0	11-	2	7	0	47	45	2	9	3-	20	16-
2	5	6	89	91	2	7-	0	0	24	2	9	4	0	16
2	5	6-	0	30-	2	7	1	36	35-	2	9	5	42	34
2	5-	6	0	22-	2	7	1-	62	60	2	9	9	34	32
2	5-	6-	52	49-	2	7-	1	61	66-	2	10	2	24	26-
2	5	7	68	76-	2	7-	1-	0	32	2	10	3	30	23
2	5	7-	0	7-	2	7	2	0	16	2	10	6	27	27-
2	5-	7	67	64-	2	7	2-	52	43					
2	5-	7-	35	39-	2	7-	2	0	19	3	0	0	111	117-
2	5	8	45	48-	2	7-	2-	0	3-	3	0	1	29	35-
2	5	8-	47	37	2	7	3	32	24	3	0	2	71	80-
2	5-	8	61	60-	2	7	3-	0	8-	3	0	2-	61	39
2	5-	8-	42	31	2	7-	3	63	71	3	0	3	158	162
2	5	9	0	1-	2	7-	3-	0	34-	3	0	3-	35	43

h	k	l	F _o	F _c	h	k	l	F _o	F _c	h	k	l	F _o	F _c
3	0	4	42	15	3	2	0	100	106	3	3-	2-	90	103
3	0	4-	79	63-	3	2-	0	55	48	3	3	3	92	77
3	0	5	210	188-	3	2	1	67	79-	3	3	3-	56	53-
3	0	5-	67	57-	3	2	1-	87	60	3	3-	3	0	2-
3	0	7	108	106	3	2-	1	110	102	3	3-	3-	56	50
3	0	9	83	92-	3	2-	1-	117	96-	3	3	4	118	104-
3	0	11	46	40	3	2	2	76	84-	3	3	4-	143	140-
3	0	11-	58	44	3	2	2-	31	20	3	3-	4	59	44-
3	0	12	81	77	3	2-	2	56	58-	3	3-	4-	39	32-
3	0	13	43	36-	3	2-	2-	30	30	3	3	5	72	70-
3	1-	0	94	70	3	2	3	34	34	3	3	5-	61	52
3	1	1-	39	23-	3	2	3-	84	75-	3	3-	5	30	41-
3	1-	1	62	27-	3	2-	3	137	151-	3	3-	5-	49	50-
3	1-	1-	108	92	3	2-	3-	87	81	3	3	6	0	13-
3	1	2-	217	186-	3	2	4	105	130	3	3	6-	129	130
3	1-	2	115	109-	3	2	4-	177	138	3	3-	6	48	56
3	1-	2-	118	116-	3	2-	4	0	1	3	3-	6-	0	20
3	1	3-	40	29	3	2-	4-	102	104	3	3	7	0	1
3	1-	3	161	161-	3	2	5	29	26	3	3	7-	0	21-
3	1-	3-	97	102-	3	2	5-	133	136	3	3-	7	41	37
3	1	4	112	124-	3	2-	5	130	151	3	3-	7-	55	56
3	1	4-	72	71	3	2-	5-	86	81-	3	3	8	88	102
3	1-	4	250	261	3	2	6	66	67-	3	3	8-	0	7
3	1-	4-	30	21	3	2	6-	79	79-	3	3-	8	33	42-
3	1	5	33	17	3	2-	6	0	18-	3	3	9	41	37-
3	1	5-	129	124-	3	2-	6-	38	32-	3	3	10	63	61-
3	1-	5	42	34-	3	2	7	86	75-	3	3	10-	32	25
3	1-	5-	90	78	3	2	7-	92	88-	3	3	11	0	2
3	1	6	101	78	3	2-	7	82	99-	3	3	12	51	39
3	1	6-	90	85-	3	2-	7-	58	45-	3	3	13	35	26
3	1-	6	93	91-	3	2	8	53	56-	3	4	0	84	64-
3	1-	6-	84	80	3	2	8-	0	11	3	4-	0	47	53-
3	1	7	67	61	3	2	8	64	61-	3	4	1	55	53
3	1	7-	22	14	3	2	8-	40	19	3	4	1-	141	117-
3	1-	7	31	39	3	2	9	135	147	3	4-	1	121	120-
3	1-	7-	79	74	3	2	9-	49	38	3	4-	1-	95	96
3	1	8	113	110-	3	2-	9	48	47	3	4	2	250	239
3	1	8-	76	74	3	2-	9-	40	39	3	4	2-	119	111
3	1-	8	103	108	3	2	10	48	48	3	4-	2	0	1-
3	1-	8-	36	36-	3	2-	10-	39	32-	3	4-	2-	0	23
3	1	9	53	58-	3	2	11	34	21-	3	4	3	52	41-
3	1	9-	24	16-	3	2	11-	33	29-	3	4	3-	80	78
3	1-	9	45	53	3	2-	11-	21	20-	3	4-	3	44	56
3	1-	9-	79	64	3	2	12	0	4	3	4-	3-	63	70-
3	1	10	29	27	3	2	13	49	45	3	4	4	87	66-
3	1	10-	56	45-	3	2	14	32	29	3	4	4-	29	28-
3	1-	10	33	35-	3	3	0	55	59-	3	4-	4	0	19-
3	1-	10-	57	52	3	3-	0	135	138-	3	4-	4-	0	37-
3	1	11	26	22-	3	3	1	233	228-	3	4	5	49	53
3	1-	11	31	34-	3	3	1-	185	158	3	4	5-	52	45-
3	1	12	81	81-	3	3-	1	119	118	3	4-	5	41	33-
3	1-	12	33	20	3	3-	1-	0	1-	3	4-	5-	0	3
3	1	13	52	53-	3	3	2	31	27-	3	4	6	69	70
3	1-	13	23	25	3	3	2-	100	89	3	4	6-	0	21
3	1	14	24	19	3	3-	2	172	186	3	4-	6	41	46-

h	k	l	F _o	F _c	h	k	l	F _o	F _c	h	k	l	F _o	F _c
3	4-	6-	49	46	3	6-	1	42	44	3	8	4-	23	14
3	4	7	60	70	3	6	2	36	29-	3	8	5	42	40
3	4	7-	50	47	3	6	2-	31	35-	3	8	5-	13	15-
3	4-	7-	47	42-	3	6	3	59	48	3	8	6	34	34
3	4	8	65	67-	3	6	3-	0	3	3	8	10	61	48
3	4	8-	0	10-	3	6-	3-	87	77	3	8	11	40	37
3	4-	8-	53	57-	3	6	4	23	8	3	9	0	37	39
3	4	9	43	44-	3	6	4-	0	18	3	9	1-	20	23
3	4	9-	32	22-	3	6	5	29	29-	3	9	2	58	57-
3	4	10	45	45	3	6	5-	61	53	3	9	2-	26	19-
3	4-	10	28	30-	3	6-	5-	33	26-	3	9	3-	30	31-
3	4-	10-	34	29	3	6	6	179	191-	3	9	4	67	65
3	4	11	63	61	3	6	6-	44	36	3	9	5	36	42
3	4	13	50	38-	3	6-	6	29	38	3	10	3	37	37
3	4	14	35	28-	3	6	7	43	48-	3	10	5	66	66-
3	5	0	99	89	3	6	7-	47	35-					
3	5-	0	45	43	3	6-	7-	37	37	4	0	0	50	32-
3	5	1	74	75	3	6	8	63	59	4	0	1	81	71
3	5	1-	121	107-	3	6	8-	12	31-	4	0	1-	58	42-
3	5-	1	0	14	3	6-	8-	28	28	4	0	2	244	265
3	5-	1-	0	29-	3	6	9	0	1-	4	0	2-	76	64
3	5	2	88	76-	3	6	10	0	19-	4	0	3	112	115
3	5	2-	153	146-	3	6	11	44	42-	4	0	3-	95	104
3	5-	2	61	62-	3	6	13	39	38	4	0	4	73	62
3	5-	2-	88	110-	3	7	0	45	45-	4	0	4-	40	31-
3	5	3	112	99-	3	7	1	22	25-	4	0	5	55	60-
3	5	3-	0	31	3	7	1-	0	6	4	0	5-	50	39-
3	5-	3	0	1-	3	7-	1	42	52-	4	0	6	101	97
3	5-	3-	65	69-	3	7	2	40	44	4	0	6-	69	62-
3	5	4	0	4-	3	7	2-	81	88	4	0	7	48	29
3	5	4-	53	35-	3	7-	2-	46	45	4	0	7-	58	48
3	5-	4	0	29	3	7	3	67	74	4	0	8	57	64-
3	5-	4-	22	21	3	7	3-	0	12	4	0	8-	0	2
3	5	5	129	126	3	7-	3	27	36	4	0	9	53	54-
3	5	5-	0	9-	3	7	4	31	36-	4	0	9-	44	39-
3	5-	5	39	35	3	7	4-	41	35-	4	0	10	33	16
3	5-	5-	53	51	3	7-	4-	31	35-	4	0	10-	27	25-
3	5	6	101	98	3	7	5	0	8-	4	0	11	22	30
3	5	6-	69	58-	3	7	5-	41	42-	4	0	13	40	39-
3	5-	6-	26	30-	3	7	6	0	21	4	1	0	31	18
3	5	7	91	89-	3	7	6-	25	20	4	1-	0	35	16
3	5	7-	40	32-	3	7-	6-	24	28	4	1	1	28	20
3	5-	7-	22	34-	3	7	7	78	77	4	1	1-	20	19
3	5	8	30	26	3	7	8	0	4-	4	1-	1	137	116-
3	5	8-	37	32	3	7	9	47	40-	4	1-	1-	132	120
3	5-	8-	27	16	3	7	11	40	31	4	1	2	113	165-
3	5	9	0	9	3	7	12	33	28	4	1	2-	115	100-
3	5	9-	26	21	3	8	0	0	6	4	1-	2	0	12
3	5-	9-	29	25	3	8	1	22	16	4	1-	2-	19	16
3	5	10	0	22	3	8	1-	62	63-	4	1	3	152	184-
3	5	11	55	47-	3	8	2	22	28	4	1	3-	58	38
3	5	14	32	29	3	8	2-	40	39-	4	1-	3	26	18
3	6	0	46	41	3	8	3	67	80-	4	1-	3-	78	65-
3	6	1	74	69-	3	8	3-	37	38	4	1	4	90	106-
3	6	1-	88	90	3	8	4	39	36-	4	1	4-	86	74

h	k	l	F _o	F _c	h	k	l	F _o	F _c	h	k	l	F _o	F _c
4	1-	4	50	45	4	2-	7	31	32-	4	4-	0	52	68-
4	1-	4-	0	14	4	2-	7-	61	59-	4	4	1	144	137
4	1	5	108	121	4	2	8	29	13	4	4	1-	128	116-
4	1	5-	0	21-	4	2	8-	41	25-	4	4-	1	59	67-
4	1-	5	72	68-	4	2-	8	0	21	4	4-	1-	0	28
4	1-	5-	0	20-	4	2	9	45	47	4	4	2	150	158
4	1	6	15	16-	4	2	9-	17	14	4	4	2-	34	38-
4	1	6-	52	49-	4	2-	9	53	59	4	4-	2	71	78
4	1-	6	96	95-	4	2-	9-	36	28	4	4-	2-	79	79
4	1-	6-	90	79	4	2	10	46	45-	4	4	3	28	16
4	1	7	83	89-	4	2	10-	37	35	4	4	3-	30	25
4	1	7-	28	25-	4	2-	10	22	18	4	4-	3	53	48-
4	1-	7	102	115	4	2	11	23	16-	4	4-	3-	0	19
4	1-	7-	0	12	4	2	12	42	33	4	4	4	74	58-
4	1	8	43	27-	4	2	13	39	33	4	4	4-	51	42
4	1	8-	80	61	4	3	0	86	62-	4	4-	4	31	39-
4	1-	8	59	75	4	3-	0	41	40-	4	4-	4-	68	69-
4	1-	8-	50	42-	4	3	1	81	98-	4	4	5	61	45
4	1	9	0	2-	4	3-	1	100	114	4	4	5-	40	36-
4	1	9-	39	32	4	3-	1-	27	27-	4	4-	5	0	16-
4	1-	9	23	22-	4	3	2	23	2	4	4-	5-	31	30-
4	1	10	59	70	4	3	2-	94	79	4	4	6	92	94
4	1	10-	37	31-	4	3-	2	39	31	4	4	6-	51	46-
4	1-	10	44	40-	4	3-	2-	0	1	4	4-	6-	48	46
4	1-	10-	26	22	4	3	3	118	150	4	4	7	0	9-
4	1	11	45	41-	4	3	3-	0	2	4	4	8	76	84-
4	1	12	32	28-	4	3-	3	114	130-	4	4	8-	49	44
4	1-	12	29	23	4	3-	3-	97	90	4	4-	8-	47	42-
4	2	0	44	46	4	3	4	112	122-	4	4	9	0	7
4	2-	0	75	73	4	3	4-	126	114-	4	4-	9	35	41-
4	2	1	58	67-	4	3-	4	58	61-	4	4	10	59	68
4	2	1-	77	67	4	3-	4-	0	2	4	4	11	0	8
4	2-	1	16	12-	4	3	5	44	44-	4	4	12	85	80-
4	2-	1-	77	72-	4	3	5-	44	41-	4	4	13	28	23-
4	2	2-	0	7-	4	3-	5	31	28	4	5	0	137	134
4	2-	2	125	103-	4	3-	5-	52	54-	4	5-	0	32	18
4	2-	2-	139	149-	4	3	6	104	118	4	5	1	46	50-
4	2	3	20	21	4	3	6-	45	30	4	5	1-	0	10
4	2	3-	119	112-	4	3-	6	33	38	4	5-	1	19	13-
4	2-	3	50	47-	4	3-	6-	0	4	4	5-	1-	62	65
4	2-	3-	26	17-	4	3	7	125	144	4	5	2	115	93-
4	2	4	89	121	4	3-	7	0	5-	4	5	2-	65	53
4	2	4-	0	1-	4	3-	7-	57	42	4	5-	2	27	25-
4	2-	4	63	67	4	3	8	57	63	4	5-	2-	0	15-
4	2-	4-	50	41	4	3	8-	17	16-	4	5	3	0	3-
4	2	5	81	88-	4	3-	8	39	41-	4	5	3-	0	3-
4	2	5-	133	130	4	3	9	68	75-	4	5-	3	26	28
4	2-	5	86	107	4	3	9-	53	46-	4	5-	3-	50	45-
4	2-	5-	0	7	4	3-	9-	22	11-	4	5	4	60	38
4	2	6	140	159-	4	3	10	28	30-	4	5	4-	45	43
4	2	6-	31	28	4	3-	10	29	32	4	5-	4	38	38
4	2-	6	63	59-	4	3	11	70	65	4	5-	4-	0	3-
4	2-	6-	0	2	4	3	12	43	29	4	5	5	66	68
4	2	7	33	22-	4	3	13	23	16-	4	5	5-	94	84
4	2	7-	20	16-	4	4	0	0	1	4	5-	5-	71	68

h	k	l	F _o	F _c	h	k	l	F _o	F _c
4	5	6	0	9	4	8	1	57	53
4	5	6-	37	26-	4	8	1-	46	40-
4	5	7	118	125-	4	8	2	26	28-
4	5	7-	51	36-	4	8	2-	78	74-
4	5-	7-	34	35-	4	8	3	45	46-
4	5	8	0	13	4	8	4	23	13
4	5	9	74	94	4	8	4-	26	23
4	5	10	20	12-	4	8	5	52	48
4	5	11	52	39-	4	8	6	0	8-
4	5	13	49	49	4	8	7	36	37-
4	6	0	54	50-	4	8	8	39	20
4	6-	0	19	20	4	8	9	0	32
4	6	1	67	58-	4	8	12	39	35-
4	6	1-	67	68	4	9	1	26	33-
4	6-	1-	40	35-	4	9	1-	58	51
4	6	2	23	25	4	9	2	29	24-
4	6	2-	0	16	4	9	3	48	41
4	6-	2-	43	41-	4	9	4	81	71
4	6	3	46	38	4	9	5	21	26-
4	6	3-	0	15-	4	9	6	64	57-
4	6-	3	28	26-	4	9	9	35	9-
4	6	4	46	33-	4	9	10	36	28-
4	6	4-	107	94-	4	10	2	33	33
4	6-	4-	31	39	4	10	3	28	38
4	6	5	95	83-	4	10	4	24	21-
4	6	6	78	72-	4	10	5	51	44-
4	6	6-	46	40	4	10	6	20	23
4	6-	6	15	26	4	10	7	34	36
4	6-	6-	33	45-					
4	6	7	68	70-					
4	6	8	75	75					
4	6	9	59	60-					
4	6	10	52	56-					
4	6	11	0	9-					
4	6	12	35	26					
4	7	0	74	69-					
4	7	1	40	38					
4	7	1-	82	84-					
4	7	2	20	8-					
4	7	2-	39	42					
4	7-	2-	20	24					
4	7	3	28	23-					
4	7	3-	51	42					
4	7	4	60	63-					
4	7	4-	0	10-					
4	7	5	51	56					
4	7	5-	44	38-					
4	7	6	71	65					
4	7	7	59	58					
4	7	8	0	20-					
4	7	9	0	20-					
4	7	11	61	48					
4	7	12	30	22					
4	7	13	41	42-					
4	8	0	75	70					

BIBLIOGRAPHY

1. Mark M. Jones, J. Am. Chem. Soc. 76, 5995 (1954).
2. Robert D. Feltham, Bonding and Spectra of Coordination Compounds (Thesis), UCRL-3867, July 1957.
3. K. Ueno and A. Martell, J. Phys. Chem. 60, 934-8 (1956).
4. Michael E. Senko, The Crystal Structures of a Triazole and Choline Chloride (Thesis), UCRL-3521, September 1956.
5. E. Hughes, J. Am. Chem. Soc. 63, 1737 (1941).
6. D. Cruickshank and A. Robertson, Acta Cryst. 6, 698 (1953).
7. Koyama, Saito, and Kuroya, J. Inst. Polytech. Osaka City Univ., Ser. C 4, 43-9 (1953).
8. Trueblood, Sparks, Prosen, and Kruse, Acta Cryst. 9, 350 (1956).