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LUCAS HEIGHTS

A GENERAL IBM360 COMPUTER PROGRAM FOR DETERMINING THE MULTIPLICITIES OF CRYSTAL FORMS, TO BE USED IN CONJUNCTION WITH THE RIETVELD POWDER PROFILE REFINEMENT PROGRAM

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ABSTRACT

An IBM360 computer program has been written which generates multiplicity data for the crystal forms in any space group. The input data consist of the unit cell dimensions, lattice centring indicator, system type, equivalent positions of the space group, and the range of h, k, l and 2θ. The program calculates structure factors for all reflexions over the powder pattern, and sorts them into forms using the IBM OS SORT/MERGE Program. The form indices {hkl} are taken as those of the last member of the form generated, and the multiplicity m{hkl} as the number of reflexions encountered through the form. The {hkl}, m{hkl} data serve as input for the Rietveld powder profile refinement program for the refining of structural parameters with powder data. Previously, these had to be deduced from the International Tables for X-ray Crystallography Volume 1, which is difficult to use with the higher symmetry systems. As the {hkl} and multiplicity generation is a major part of the effort needed to set up the Rietveld program, the present program reduces significantly the work involved in these refinements. The program gives [{hkl}, m{hkl}] cards in about 30-50 seconds of CPU time.

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The following descriptors have been selected from the INIS Thesaurus to describe the subject content of this report for information retrieval purposes. For further details please refer to IAEA-INIS-12 (INIS: Manual for Indexing) and IAEA-INIS-13 (INIS: Thesaurus) published in Vienna by the International Atomic Energy Agency.

COMPUTER CODES; SPACE GROUPS; CRYSTAL LATTICES; CRYSTAL STRUCTURE; CRYSTALLOGRAPHY; MULTIPLICITY; NEUTRON DIFFRACTION + DATA; X-RAY DIFFRACTION + DATA

CONTENTS

1. INTRODUCTION

At present, there is considerable interest in the refinement of crystal structure parameters with neutron or X-ray powder diffraction profile data. This can be done with the Rietveld computer program [Rietveld 1969] which accepts the data in the form of a series of measured intensities along the 2θ scale of the pattern. All octants of the reciprocal lattice in the 2θ range contribute to the powder pattern. However, the (hkl) reflexion input is simplified by grouping reflexions into 'forms' {hkl}, with the same 2θ and structure amplitude $|F_{hkl}|$. The number of reflexions in a form {hkl} is the multiplicity, m_{hkl}.

After correction for background and geometrical factors, the intensity at a particular 2θ angle is proportional to $\sum_{hkl} m_{hkl} F^2_{hkl}$, the sum being over the number of different forms present. Each form is represented by one {hkl} in the form with a multiplicity m_{hkl}; the forms are weighted by a Gaussian or other peak shape function.

Table 3.5.1 on pages 32-35 of the International Tables for X-ray Crystallography, Volume I [1965] gives multiplicity factors for forms in all crystal systems. Although this table is simple to use for the low-symmetry triclinic, monoclinic, and orthorhombic systems, it becomes difficult to use when the symmetry is higher, due to cyclic permutations of indices within forms, and the occurrence of forms with the same 2θ angle but different $|F_{hkl}|$ (e.g. $|F_{hkl}| \neq |F_{hk\bar{l}}|$ in some trigonal and hexagonal cases). Also, as pointed out in the Additional Note on page 34 of Table 3.5.1, different conventions in choosing the unit cell axes relative to the symmetry elements can change the multiplicities of the forms (cf. Laue Groups $\bar{3}$ml and $\bar{3}$lm).

The Rietveld program requires as input all {hkl} and corresponding m_{hkl} in the range of the pattern. Since these must be worked out for the particular space group with the help of a table such as Table 3.5.1, correctly assigning multiplicities is a major task in setting up a Rietveld refinement. Often a few $|F_{hkl}|$ must be calculated with a structure factor program such as ORFLS [Busing, Martin & Levy 1962] to check that the interpretation of the table is correct. The author was requested by colleagues working in higher symmetry systems, (who had experienced difficulty with Table 3.5.1) to work out multiplicity data for the Rietveld program for special cases.

A program for determining multiplicities of Bragg reflexions in any space group has recently been written at the AERE, Harwell [Rouse & Cooper 1976]. It is essentially a table look-up program and makes use of a table based on Table 3.5.1 stored in the computer. This program requires as

input values of the relevant {hkl}s punched one to a card. As there may be several hundred {hkl}s in a powder pattern, this approach is not labour saving, compared with the present AAEC program which generates each {hkl} required. The AAEC program also derives the m {hkl}s from first principles, thus removing the possibility of errors in the interpretation of Table 3.5.1. One such error appears to have occurred in the Harwell program: it does not distinguish between Laue groups $\bar{3}$ml and $\bar{3}$1m, as required in the Additional Note to Table 3.5.1 and therefore gives incorrect results for space groups based on $\bar{3}$1m. (Errors in Table 3.5.1 itself for these Laue groups have also recently been found (M.M. Elcombe and J. Nimmo, private communication).) As the Harwell program was not immediately adaptable to Rietveld refinements and was a table look-up program, the following general program for profile work was therefore written.

2. PROGRAM STRATEGY

The program generates all possible indices in the reflexion sphere up to the specified 2θ limits, and writes on disc a record (sorted on h, k, l) for each reflexion which contains:

$$h,k,l\ \sin^2\theta,\ 2\theta,\ |F| \text{ and NNN}$$

where $|F|$ is a structure factor calculated from the general formula $F^2 = A^2 + B^2$ (see below) and NNN is a check on the equivalence of forms in the hexagonal, tetragonal and cubic cases, respectively $(h^4 + k^4 + i^4)$, $(h^4 + k^4)$ and $(h^4 + k^4 + l^4)$. Although non-equivalent reflexions with the same quadratic form integers (e.g. $h^2 + k^2 + l^2$ for cubic) should be differentiated by their structure factors (e.g. 510 and 431 in primitive cubic), NNN is included as an additional check.

Systematic absences are eliminated when their structure factors are calculated to be zero. If a reflexion calculates $|F| = 0$, the structure factor is recalculated with a new set of positional parameters to make sure it is an absence and not accidentally zero. To eliminate false symmetry in the $|F|$ values, two dummy atoms at random locations (x_1,y_1,z_1) and (x_2,y_2,z_2) in the unit cell are used to calculate

$$A = \sum_n \cos 2\pi\ (hx_n + ky_n + lz_n) \quad \text{and}$$

$$B = \sum_n \sin 2\pi\ (hx_n + ky_n + lz_n).$$

Without the sum (over two atoms), false higher symmetry in the $|F|$ values can occur in some systems, e.g. rhombohedral, R3m.

The $\sin^2\theta$, 2θ and $|F|$ values are calculated in REAL*8, (to about 17

decimal places), and the IBM OS SORT/MERGE Program is used to sort the list of structure factors on $\sin^2\theta$(major), $|F|$(intermediate), and NNN(minor). The IBM OS SORT/MERGE Program (Program No. 360S-SM-023, File No. S360-330S, Order No. GC28-6543-8), is referenced in the catalogued procedure SORT, which is a member of the AAEC system data set SYS1.PROCLIB.

Finally, the reflexions in the sorted list are examined to see if the following conditions are met simultaneously for all reflexions to be given the same multiplicity:

(1) the $\sin^2\theta$ values all agree to within 1×10^{-6};

(2) the $|F|$ values all agree to within 1×10^{-5}; and

(3) the reflexions have the same NNN value.

The Rietveld {hkl} cards are punched out as (h,k,l,multiplicity), the {hkl} value being the {hkl} of the last reflexion in a group (it is immaterial which {h,k,l} in the group is chosen). For a computer with precision higher than REAL*8, e.g. the IBM370 with REAL*16, it would be worthwhile modifying this program for the higher precision.

Note that the error limits on $|F|$ and $\sin^2\theta$ in Appendix B may have to be altered for different computers.

3. PROGRAM DATA CARDS

Card 1 Title

Card 2 λ, a, b, c, α, β, γ, NS, ICENT, ISYS, NN (7F10.5, I2, 3I1)

where λ = wavelength

a,b,c,α,β,γ are direct cell constants

NS = number of symmetry cards

ICENT = 1 for centric, 0 for acentric

ISYS = lattice indicator : 1 for body-centred (I), 2 for all face-centred (F),3 for face-centred (A), 4 for face-centred (B), 5 for face-centred (C), 6 for rhombohedral based on hexagonal axes, and 7 for primitive

NN = system type : 1 for hexagonal, or rhombohedral on hexagonal axes, 2 for tetragonal, 3 for cubic, and 4 for all others.

It has been assumed that rhombohedral crystals will be based on a hexagonal lattice. If the rhombohedral cell is to be used, then the cell can be treated as triclinic (NN=4).

Cards 3 Symmetry Cards (12 F6.3)

For a symmetry position J, the card reads:

X(1,J) Y(1,J) Z(1,J) T(1,J), X(2,J), Y(2,J), Z(2,J) T(2,J),
X(3,J) Y(3,J) Z(3,J) T(3,J)

There is one card for each position including (x,y,z), e.g. (y-x, -x,z) should be -1. 1. 0. 0. -1. 0. 0. 0. 0. 0. 1. 0. (This is the same type of symmetry card input as in the Rietveld program itself.)

Card 4 H_{min} K_{min} L_{min} H_{max} K_{max} L_{max} $2\theta_{max}$ (6I5, F10.4)

These are the minimum and maximum values of the Miller indices and the maximum 2θ value (> 1.5 halfwidths past the end of the pattern).

4. PROGRAM ASSEMBLY AND CONTROL CARDS

```
//JCTU     JØB ('********/D51CT007',G3),J.C.TAYLØR,
//             CLASS=B,TIME=1
//EXEC FØRTGCLG
//FØRT.SYSIN DD *
```

[Program I plus SUBRØUTINE CALL (see Appendix A).

```
/*
//GØ.FT20F001 DD UNIT=SYSDA,DISP=(,PASS),DSN=&&T,SPACE=(TRK,(10,10)), *
//             DCB=(RECFM=FB,BLKSIZE=800,LRECL=80)
//GØ.SYSIN  DD *
```

[DATA CARDS, as above.

```
/*
// EXEC SØRT,REGIØN=240K
//SØRTIN DD DSN=&&T,DISP=(ØLD,PASS)
//SØRTØUT DD DSN=&&TT,DISP=(,PASS),DCB=(RECFM=FB,LRECL=80,BLKSIZE=800),*
//             SPACE=(TRK,(10,10)),UNIT=SYSDA
//SØRTWK01 DD UNIT=SYSDA,SPACE=(TRK,40)
//SØRTWK02 DD UNIT=SYSDA,SPACE=(TRK,40)
//SØRTWK03 DD UNIT=SYSDA,SPACE=(TRK,40)
//SØRTWK04 DD UNIT=SYSDA,SPACE=(TRK,40)
SØRT FIELDS=(11,8,CH,A,51,7,CH,A,71,10,CH,A)
/*
// EXEC FØRTGCLG
//FØRT.SYSIN DD *
```

[Program II (see Appendix B).

```
/*
//GØ.FT20F001 DD UNIT=SYSDA,DISP=(ØLD,DELETE),DSN=&&TT
/*
```

5. OUTPUT OF PROGRAM

(i) From PROGRAM I

The input data are printed out, followed by a list of {h,k,l}, A and B for all {h,k,l} in the pattern.

A typical output follows:

```
    CUBIC,FD3 NO 203
         NO OF SYMMETRY CARDS IS   12      ICENT(1 FOR CENTRIC,   0 FOR ACENTRIC) IS    1      LATTICE ( 1 FOR I

 2 FOR F,         3 FOR A, 4 FOR B, 5 FOR C, 6 FOR RHOMBOHEDRAL

BASED ON         HEXAGONAL AXES, 7 FOR PRIMITIVE) IS    2
         SYSTEM IS  (1 FOR HEXAGONAL OR RHOMB ON HEXAGONAL      LATTICE,2 FOR TETRAGONAL,3 FOR CUBIC,4 FOR ALL OTHER) IS      3
          SPACE GROUP POSITIONS LISTED BELOW
          1.000 0.0    0.0    0.0    0.0    1.000 0.0    0.0    0.0    0.0    1.000 0.0
          1.000 0.0    0.0    0.0    0.0   -1.000 0.0    0.250 0.0    0.0   -1.000 0.250
         -1.000 0.0    0.0    0.250 0.0    1.000 0.0    0.0    0.0    0.0   -1.000 0.250
         -1.000 0.0    0.0    0.250 0.0   -1.000 0.0    0.250 0.0    0.0    1.000 0.0
          0.0    0.0    1.000 0.0    1.000 0.0    0.0    0.0    0.0    1.000 0.0    0.0
          0.0    0.0    1.000 0.0   -1.000 0.0    0.0    0.250 0.0   -1.000 0.0    0.250
          0.0    0.0   -1.000 0.250 1.000 0.0    0.0    0.0    0.0   -1.000 0.0    0.250
          0.0    0.0   -1.000 0.250-1.000 0.0    0.0    0.250 0.0    1.000 0.0    0.0
          0.0    1.000 0.0    0.0    0.0    0.0    1.000 0.0    1.000 0.0    0.0    0.0
          0.0    1.000 0.0    0.0    0.0    0.0   -1.000 0.250-1.000 0.0    0.0    0.250
          0.0   -1.000 0.0    0.250 0.0    0.0    1.000 0.0   -1.000 0.0    0.0    0.250
          0.0   -1.000 0.0    0.250 0.0    0.0   -1.000 0.250 1.000 0.0    0.0    0.0
         JHMIN=  -13   JKMIN=  -13   JLMIN=  -13   JHMAX=   13   JKMAX=   13   JLMAX=   13
    WAV=  1.0000000  A=  8.00000000  B=  8.00000000  C=  8.00000000  ALF= 90.00000000  BET= 90.00000000  GAM= 90.00000000
         RECIPROCAL CELL IS
           0.1250000  0.1250000  0.1250000  0.0000001  0.0000001  0.0000001
               -6         -2         -2      1.40491    0.0
               -6         -2          0     -0.24751    0.0
               -6         -2          2      1.40491    0.0
               -6          0         -2      3.80706    0.0
               -6          0          0     -0.00000    0.0
               -6          0          0     -0.00000    0.0  , etc.
```

(ii) From PROGRAM II

A list is printed in the order h,k,l,$\sin^2\theta$,2θ, $|F_{hkl}|$ sorted on $\sin^2\theta$, $|F|$ and NNN. After sorting, the program finds the end (hkl) of each form and prints and punches this {hkl} and the multiplicity of the form, the punched cards being in the Rietveld program format.

In the above case, the program gave the list:

{h, k, l}	m	2θ
1,-1,-1	8	12.429
0,-2,-2	12	20.364
-3,-1,-1	24	23.927
-2,-2,-2	8	25.008
0, 4, 0	6	28.955
-1,-3, 3	24	31.618
-2,-2,-4	24	35.659
3,-3, 3	8	37.902
-1, 5,-1	24	37.902
0,-4,-4	12	41.410
3,-1, 5	24	43.401
-1, 3,-5	24	43.401
	etc.	

6. TESTS OF THE PROGRAM

The program was tested for typical space groups in all crystal systems and gave correct answers for these cases. The input parameters and results are given below.

(i) Triclinic, $P\bar{1}$ (No. 2)

Unit cell: a = 5.0, b = 6.0, c = 7.0Å, alpha = 95.0, beta = 100.0, gamma = 105.0°, λ = 1.0Å.

Program output:

{h, k, l}	m	2θ
0, 0,-1	2	8.400
0, 1, 0	2	9.996
0,-1, 1	2	12.140
-1, 0, 0	2	12.141
1, 0,-1	2	13.289
1,-1, 0	2	13.407
0,-1,-1	2	13.940
-1, 1, 1	2	15.249
1, 0, 1	2	16.139

(continued)

{h, k, l}	m	2θ (continued)
1,-1, 1	2	16.414
0, 0, 2	2	16.846
1, 1, 0	2	17.793

etc.

(ii) Monoclinic, C2/C, No. 15

Unit cell: a = 6.0, b = 7.0, c = 8.0Å, beta = 100.0°, λ = 1.0Å.

Program output:

{h, k, l}	m	2θ
1,-1, 0	4	12.715
-1, 1, 1	4	13.796
0, 0,-2	2	14.584
-1,-1,-1	4	15.487
0, 2, 0	2	16.426
0,-2,-1	4	17.987
-1,-1, 2	4	18.063
2, 0, 0	2	19.487
-1,-1,-2	4	20.643
0, 2, 2	4	22.034
-2, 0, 2	2	22.269
1,-1,-3	4	23.912

etc.

(iii) Orthorhombic, Pnma No. 62

Unit cell: a = 6.0, b = 7.0, c = 8.0Å, λ = 1.0Å.

Program output:

{h, k, l}	m	2θ
0,-1,-1	4	10.893
1, 0,-1	4	11.958
0, 0,-2	2	14.362
1, 1,-1	8	14.512
0, 2, 0	2	16.426
1, 0,-2	4	17.281
-1, 1, 2	8	19.151
2, 0, 0	2	19.188
-1,-2,-1	8	20.367
2, 0, 1	4	20.506

etc.

(iv) Tetragonal, I $4_{1/a}$

Unit cell: a = 12.0, b = 12.0, c = 15.0Å, λ = 1.0Å.

Program output:

{h, k, l}	m	2θ
-1, 0,-1	8	6.117
-2, 0, 0	4	9.560
-1,-1,-2	8	10.209
2,-1,-1	8	11.358
-2,-1,-1	8	11.358
2, 0,-2	8	12.252
-1, 0,-3	8	12.438
-2,-2, 0	4	13.536
0,-3,-1	8	14.866
0, 0,-4	2	15.325
-2,-2,-2	8	15.564
-1, 2,-3	8	15.711
2,-1,-3	8	15.711
1, 1, 4	8	16.764

etc.

(v) Hexagonal, P$\bar{3}$m1, No. 164

Unit cell: a = 6.0, b = 6.0, c = 8.0Å, λ = 1.0Å.

Program output:

{h, k, l}	m	2θ
0, 0,-1	2	7.167
1, 0, 0	6	11.044
0, 1,-1	6	13.177
1, 0,-1	6	13.177
0, 0,-2	2	14.362
1, 0,-2	6	18.152
0,-1, 2	6	18.152
-2, 1, 0	6	19.188
-1, 2, 1	12	20.507
0, 0,-3	2	21.614
-2, 0, 0	6	22.192
0, 2, 1	6	23.348
-2, 0,-1	6	23.348
-1, 2,-2	12	24.049
1, 0,-3	6	24.333
-1, 1, 3	6	24.333

etc.

(vi) Rhombohedral,R3m, No. 160 (Hexagonal Axes)

Unit cell: a = 13.095, c = 5.658Å, λ = 1.0Å.

Program output:

{h, k, l}	m	2θ
-2, 1, 0	6	9.426
0,-1, 1	6	12.200
2, 0,-1	6	15.439
-3, 0, 0	6	16.364
2,-3,-1	12	18.120
-4, 2, 0	6	18.918
3,-4, 1	12	22.571
0, 1, 2	6	22.606
-4, 4, 1	6	24.509
-2, 2, 2	6	24.541
-5, 1, 0	12	25.113
-2, 5,-1	12	26.312
	etc.	

(vii) Rhombohedral, R3m, No. 160 (Rhombohedral Axes)

Unit cell: a = 8.0Å, alpha = 56.0°, λ = 1.0Å.

Program output:

{h, k, l}	m	2θ
-1,-1,-1	2	8.531
0,-1, 0	6	9.265
-1,-1, 0	6	10.496
2, 1, 1	6	14.416
-1, 1, 0	6	15.301
2, 2, 1	6	16.774
-2,-2,-2	2	17.109
2, 0, 1	12	17.543
-1,-1, 1	6	17.916
0, 0,-2	6	18.591
0,-2,-2	6	21.082
3, 2, 2	6	21.883
-1,-3,-1	6	22.779
3, 2, 1	12	23.030
-1, 2, 0	12	23.643
	etc.	

(viii) Cubic, Space Group Fd3, No. 203

This case has been considered in Section 5 (i) and (ii).

At the Lucas Heights computer installation, the present multiplicity program has been concatenated with the Rietveld program and has performed satisfactorily in a year's usage for Rietveld refinements of real crystal structures in different systems.

7. COMPUTATION TIMES

The computation times (CPU times) for the above test runs were between 28 seconds and 45 seconds, except for the tetragonal case, where the large unit cell generated many reflexions, and the time was 1 minute and 3 seconds.

8. ACKNOWLEDGEMENTS

The author wishes to thank Dr. G. W. Cox for help in using SORT and Dr. R. L. Davis for discussions.

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APPENDIX A

PROGRAM I AND THE SUBROUTINE, CALC

```
      IMPLICIT REAL*8(A-G),REAL*8(S-Z)
      COMMON X(3,24),Y(3,24),Z(3,24),T(3,24)
      COMMON F,JH,JK,JL,ICENT,NS
         DIMENSION TITLE(20)
      REWIND 20
      III=0
      READ(1,1006)(TITLE(I),I=1,20)
 1006 FORMAT(20A4)
      WRITE(3,1007)(TITLE(I),I=1,20)
 1007 FORMAT(5X,20A4)
 1001 READ(1,1)WAV,A,B,C,ALF,BET,GAM,NS,ICENT,ISYS,NN
       WRITE(3,360)NS,ICENT,ISYS
  360 FORMAT(10X,'NO OF SYMMETRY CARDS IS',I5,5X,'ICENT(1 FOR CENTRIC,
     * 0 FOR ACENTRIC) IS',I5,5X,'LATTICE ( 1 FOR I'//', 2 FOR F,
     * 3 FOR A, 4 FOR B, 5 FOR C, 6 FOR RHOMBOHEDRAL'//' BASED ON
     * HEXAGONAL AXES, 7 FOR PRIMITIVE) IS',I5)
       WRITE(3,850)NN
  850 FORMAT(10X,'SYSTEM IS  (1 FOR HEXAGONAL OR RHOMB ON HEXAGONAL
     * LATTICE,2 FOR TETRAGONAL,3 FOR CUBIC,4 FOR ALL OTHER) IS  ',I5)
    1 FORMAT(7F10.5,I2,3I1)
C  READ SYMMETRY CARDS
      WRITE(3,528)
  528 FORMAT(10X,' SPACE GROUP POSITIONS LISTED BELOW')
      DO 200 J=1,NS
      READ(1,201)((X(I,J),Y(I,J),Z(I,J),T(I,J)),I=1,3)
        WRITE(3,350)((X(I,J),Y(I,J),Z(I,J),T(I,J)),I=1,3)
  350 FORMAT(10X,12F6.3)
  201 FORMAT(12F6.3)
  200 CONTINUE
      READ(1,4)JHMIN,JKMIN,JLMIN,JHMAX,JKMAX,JLMAX,T2M
    4 FORMAT(6I5,F10.4)
        WRITE(3,361)JHMIN,JKMIN,JLMIN,JHMAX,JKMAX,JLMAX
  361 FORMAT(10X,'JHMIN=',I5,3X,'JKMIN=',I5,3X,'JLMIN=',I5,3X,
     * 'JHMAX=',I5,3X,'JKMAX=',I5,3X,'JLMAX=',I5,3X)
      WRITE(3,3) WAV,A,B,C,ALF,BET,GAM
    3 FORMAT(5X,'WAV=',F12.8,2X,'A=',F12.8,2X,'B=',F12.8,2X,'C=',F12.8,
     * 2X,'ALF=',F12.8,2X,'BET=',F12.8,2X,'GAM=',F12.8,2X)
  605  ALF=ALF/57.2957795131
      BET=BET/57.2957795131
      GAM=GAM/57.2957795131
      CA=DCOS(ALF)
      CB=DCOS(BET)
      CG=DCOS(GAM)
      SA=DSIN(ALF)
      SB=DSIN(BET)
      SG=DSIN(GAM)
      V=A*B*C*DSQRT(1.+(2.*CA*CB*CG)-CA**2-CB**2-CG**2)
      AS=(B*C*SA)/V
      BS=(A*C*SB)/V
      CS=(A*B*SG)/V
      CAS=(CB*CG-CA)/(SB*SG)
      CBS=(CG*CA-CB)/(SG*SA)
      CGS=(CA*CB-CG)/(SA*SB)
      WRITE(3,821)AS,BS,CS,CAS,CBS,CGS
  821 FORMAT(10X,'RECIPROCAL CELL IS'/10X,6F11.7)
  603 JH=JHMIN
      JK=JKMIN
      JL=JLMIN
   51 CONTINUE
   50 SINSQT=(WAV**2/4.)*(JH**2*AS**2+JK**2*BS**2+JL**2*CS**2
```

(continued)

APPENDIX A (continued)

```
   1+2.*JK*JL*BS*CS*CAS+2.*JL*JH*CS*AS*CBS+2.*JH*JK*AS*BS*CGS)
     COSSQT=1.-SINSQT
      SINT=DSQRT(SINSQT)
    IF(JH.EQ.0.AND.JK.EQ.0.AND.JL.EQ.0)GO TO 505
    IF(SINT.GT.,93)GO TO 505
    IF(SINT-.999)40,505,505
 40 CONTINUE
    IF(SINT-.01)505,505,41
 41 CONTINUE
 61 D=(.5*WAV)/SINT
    TH=DARSIN(SINT)*57.2957795131
    TH2=2.*TH
    IF(TH2-T2M)5,505,505
  5 CONTINUE
    GO TO (703,704,705,706.707,708,709),ISYS
703 JN=JH+JK+JL
    JJN=MOD(JN,2)
    IF(JJN.NE.0)GO TO 505
     GO TO 709
704 JN1=JH+JK
    JN2=JK+JL
    JJN1=MOD(JN1,2)
    JJN2=MOD(JN2,2)
    IF(JJN1.NE.0.OR.JJN2.NE.0)GO TO 505
     GO TO 709
705 JN3=JK+JL
    JJN3=MOD(JN3,2)
    IF(JJN3.NE.0)GO TO 505
     GO TO 709
706 JN4=JH+JL
    JJN4=MOD(JN4,2)
    IF(JJN4.NE.0)GO TO 505
     GO TO 709
707 JN5=JH+JK
    JJN5=MOD(JN5,2)
    IF(JJN5.NE.0)GO TO 505
     GO TO 709
708 JN6=-JH+JK+JL
    JJN6=MOD(JN6,3)
    IF(JJN6.NE.0)GO TO 505
709 CONTINUE
    CALL CALC
    IF(F.LT.0.00001) GO TO 505
     GO TO (830,831,832,833),NN
830 IN=-JH-JK
    NNN=JH**4+JK**4+IN**4
    GO TO 834
831 NNN=JH**4+JK**4
    GO TO 834
832 NNN=JH**4+JK**4+JL**4
    GO TO 834
833 NNN=1
834 CONTINUE
     WRITE(20,6)III,JH,JK,JL,SINSQT,TH2,F,NNN
  6 FORMAT(I1,3I3,F20.18,F20.16,F20.15,I10)
505 CONTINUE
    IF(JL.GE.JLMAX)GO TO 501
504 JL=JL+1
    GO TO 51
501 IF(JK.GE.JKMAX)GO TO 502
```

(continued)

APPENDIX A (continued)

```
      JL=JLMIN
      JK=JK+1
      GO TO 51
  502 IF(JH.GE.JHMAX)GO TO 503
      JL=JLMIN
      JK=JKMIN
      JH=JH+1
      GO TO 51
  503 CONTINUE
  413  WRITE(20,7)
    7 FORMAT('1', 9X,'9.9')
      ENDFILE 20
      REWIND 20
      STOP
      END
      SUBROUTINE CALC
       IMPLICIT REAL*8(P-Z),REAL*8(A-G)
      COMMON X(3,24),Y(3,24),Z(3,24),T(3,24)
      COMMON F,JH,JK,JL,ICENT,NS
       DIMENSION P(2),Q(2),R(2)
C  CALCULATE GEOMETRICAL PART OF STRUCTURE FACTOR
        M=0
      F=0.
      A=0.
      B=0.
      P(1)=.123456789
      Q(1)=.437281416
      R(1)=.763407159
      P(2)=.642356842
      Q(2)=.321658975
        R(2)=.652398412
       S=6.283185307
    4 CONTINUE
         DO 1  K=1,2
      DO 1 J=1,NS
       FA=JH*S*(P(K)*X(1,J)+Q(K)*Y(1,J)+R(K)*Z(1,J)+T(1,J))
      FB=JK*S*(P(K)*X(2,J)+Q(K)*Y(2,J)+R(K)*Z(2,J)+T(2,J))
       FC=JL*S*(P(K)*X(3,J)+Q(K)*Y(3,J)+R(K)*Z(3,J)+T(3,J))
      F1=FA+FB+FC
       A=A+DCOS(F1)
      B=B+DSIN(F1)
    1 CONTINUE
      IF(ICENT.EQ.1) B=0.

      WRITE(3,7)JH,JK,JL,A,B
    7 FORMAT(10X,3I8,2F10.5)
       F=DSQRT(A**2+B**2)
      IF(F.GT..00001)GO TO 3
      IF(M.EQ.1)GO TO 3
       P(1)=.987654321
      Q(1)=.321654987
       R(1)=.357246891
      P(2)=.426589214
      Q(2)=.683572415
      R(2)=.753689241
       M=1
       GO TO 4
    3 CONTINUE
      RETURN
      END
```

APPENDIX B

PROGRAM II

```
      DIMENSION J1(50),J2(50),J3(50),FA(50),SINSQT(50),TH2(50),NNN(50)
      REAL*8 FA,FF,TH2,SINSQT
      REWIND 20
   15 FORMAT(I5)
      KNTR = 0
      I=1
      M=0
      READ(20,1)II,J1(I),J2(I),J3(I),SINSQT(I),TH2(I),FA(I),NNN(I)
    1 FORMAT(I1,3I3,F20.18,F20.16,F20.15,I10)
      WRITE(3,10)J1(I),J2(I),J3(I),SINSQT(I),TH2(I),FA(I)
   10 FORMAT(10X,3I4,10X,F20.18,5X,F20.16,5X,F20.15)
      IF(II.NE.0) GO TO 5
    4 I=I+1
      READ(20,1)II,J1(I),J2(I),J3(I),SINSQT(I),TH2(I),FA(I),NNN(I)
      IF(II.NE.0) GO TO 5
      J=I-1
      M=M+1
      FF=FA(J)
      IF(DABS(FA(I)-FF).LT.1.0D-05.AND.(SINSQT(I)-SINSQT(J)).LT
     * .1.0D-06.AND.NNN(I).EQ.NNN(J))GO TO 102
        GO TO 101
  102 WRITE(3,10)J1(I),J2(I),J3(I),SINSQT(I),TH2(I),FA(I)
      GO TO 4
  101 WRITE(3,2)J1(J),J2(J),J3(J),M
      WRITE(3,10)J1(I),J2(I),J3(I),SINSQT(I),TH2(I),FA(I)
      KNTR = KNTR+1
    2 FORMAT('        1',4I8)
      J1(1)=J1(I)
      J2(1)=J2(I)
      J3(1)=J3(I)
      FA(1)=FA(I)
      SINSQT(1)=SINSQT(I)
       NNN(1)=NNN(I)
      M=0
      I=1
```

```
      GO TO 4
    5 CONTINUE
      REWIND 20
      WRITE (3,6) KNTR
    6 FORMAT ('0******',I5,' CARDS PUNCHED ******')
      STOP
      END
```