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RADY: A FORTRAN PROGRAM FOR THE LEAST-SQUARES REFINEMENT OF CRYSTAL STRUCTURES

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RADY: A FORTRAN PROGRAM FOR THE LEAST-SQUARES REFINEMENT OF CRYSTAL STRUCTURES

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Abstract

This report is designed to give detailed descriptions and instructions for use of the least-squares program, RADY, for crystal-structure analyses. The program performs various calculations to obtain $|F_{obs}|$ from the observed intensity and to refine crystal structures by the full-matrix least-squares method.

KEYWORDS: Crystal-structure refinement; RADY; Computer program; Non-linear least-squares

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RADY99 版について

東京工業大学　　佐々木　聡

最小二乗プログラム RADY を少し改良しました。version 名は RADY99 です。お手元の入力説明書 (KEK Internal Report 87 - 3) での変更箇所を以下にお知らせします。
ソースファイル上で変更点がわかるようになっていますが、そちらについては、お送りしたメディアのソースリストをご参照下さい。

(1) 仮想記憶容量として 4MByte (機種によっては 4.1MB) 必要です。JCL の JOB 文に指定して下さい。尚、仮想記憶容量の小さいヴァージョンもあります (1989.10)。

(2) 配列を次の様に拡張しました。9 ページの配列の制限値は、パラメータの総数が 841 から 4040 に、動かすパラメータの数が 425 から 601 に、原子数が 99 から 302 に緩和されました (1989.10)。

--- Page 9, line 4 ---

Number of reflections	No Limit	(IWKREF = 1)
	10000	(IWKREF = 0)
Number of variables	4040	
Number of parameters refined (NV)	601	
Number of matrix elements	NV(NV+1)/2	
Number of atoms	302	
Number of symmetry operations	48	
Number of scales	20	
Number of scattering factors	25	
Number of k parameters	10	
Three dimensional array in REFAV/REFEX	(~~41,41,61~~) 91,91,101	

(3) h, k, l, F(obs), F(calc)-table の出力 FORMAT の選択：{2} File Control の中で。

--- Page 11, line 25 の後に ---

37 - 39　IKETA:　FORMAT of F(obs) and F(calc) for line printer and file output.
=0:　F8.3 on Lp; F12.5 on file.
=1:　F8.0 on Lp; F12.3 on file.

(4) **入力反射データの平均、最大値化、並べ変え、拡張ができます**(1989.10)：
13 ページの{3}項、カラム 52 - 54 以下が変更になります。この機能を使用される時は、Unit 15 の Working File が必要となります。International Tables の旧版に基づいたところがあり、三方晶系の一部などにバグがあります。忙しさを理由にまだ対処できていません。

--- Page 9, line 24 ---

(iii) Working files:　Unit 14 on reflection data (IWKREF = 1).
Unit 15 is used in REFAV and REFEX routines to average or expand the reflection data (IREFAV ≠ 0 or IREFEX ≠ 0).

52 - 54 IREFAV: Averaging equivalent reflections, maximizing among equivalent reflections, or rearanging reflection data. << 0 >>

A working file (unit 15) is needed when the value is not zero.

The maximum and minimum indices are limited by the 3-dimensional array (31,41,61) used in the subroutine REFEX.

=0: Skip this function.

>0: Averaging.

<0: Maximizing.

The following number on the symmetry is required for * IREFAV * .

=1: cubic m3m (1/48); =11: cubic m3 (1/24);

=2: tetragonal 4/mmm (1/16); =12: tetragonal 4/m (1/8);

=3: orthorhombic (1/8);

=4: monoclinic (b axis unique) (1/4);

=5: hexagonal 6/m (1/12); =15: hexagonal 6/mmm (1/24);

=6: trigonal (hexagonal set) -3 (1/16); =16: trigonal (hex) -3m (1/12);

=7: triclinic (1/2).

55 - 57 IREFEX: Expansion of reflection indices on the equivalent reflections. << 0 >>

A working file (unit 15) is needed when the value is not zero.

=0: Skip this function.

>0: Expanding the reflection data.

The following number on the symmetry is required for IREFEX.

=1: cubic m3m (1/48); =11: cubic m3 (1/24);

=2: tetragonal 4/mmm (1/16); =12: tetragonal 4/m (1/8);

=3: orthorhombic (1/8);

=4: monoclinic (b axis unique) (1/4);

=5: hexagonal 6/m (1/12); =15: hexagonal 6/mmm (1/24);

=6: trigonal (hexagonal set) -3 (1/16); =16: trigonal (hex) -3m (1/12);

=7: triclinic (1/2).

58 - 61 IOUTP: Region of reflection indices to be output in IREFEX routine. << 0 >>

=0: All reflections.

=1: Reflections within 1/2 space ($l \geq 0$).

=2: Reflections within 1/4 space ($k \geq 0$ & $l \geq 0$).

=3: Reflections within 1/8 space ($h \geq 0$, $k \geq 0$ & $l \geq 0$).

(5) **Windows Power FORTRAN にも対応させました** (1996.3)：

RADY (rady99ma.f) は、初期設定では WINDOWS 用になっています。rady99ma.f とダミーサブルーチン rady99s.f をコンパイルする必要があります。RADY は main frame でのダイナミック・リンクを想定して書かれているため、ダミーサブルーチンを変更するたびにこの作業が必要です。

また、その他の OS（たとえば main frame や UNIX 等）でコンパイルする時には以下の作業が必要です。

作業が必要な部分については、目印がつけられています。ソースの７３カラム目以降に、RADY0123 といったラベルの代わりに WIN-F1 あるいは WIN-F2 と書き込まれています。

（１）CALL RFOPN　文のある行をコメントアウトする．（WIN-F1 部分）
（２）SUBROUTINE RFOPN　文をコメントアウトする．　（WIN-F2 部分）

MS-WINDOWS では一般に GUI（Graphical User Interface)でコンパイルをおこないますが、コマンドベースでも実行可能です。そのためのバッチファイルを compile.bat として用意してあります。適当なオプション等を用いてコンパイルすることもできます。また、出力をファイル out.txt に出すバッチファイル RADY.BAT も便利です。

(6) **数値積分法による吸収補正** (1999.2.27)：

数値積分による吸収補正ルーチーン ACACA を RADY に組み込みました。
以下の **(A)** から **(E)** の作業が必要になります。

(A) working file を加える。

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(iii) Working files:　Unit 16 on integration method for absorption correction in the arbitory shape of a crystal (ICC = 2)

(B) {3}Parameter Control 内で、ICC を 2 に指定する。

-- page 12 --

13 - 15　ICC:　Absorption correction.　<< 0 >>

= 0:　No correction.
= 1:　Absorption correction for a spherical crystal.
= 2:　Absorption correction in the arbitory shape of a crystal (ACACA integration with small blocks).

(C) {6} Least-squares control の中で、常に IREF = 1 にする。

-- page 17 --

19 - 21　IREF　= 1 (always).

(D) {18}Variable FORAMT でのパラメータに TBAR を常に加えること。

-- page 25 --

Add TBAR term, according to the flag, IREF = 1.

(E) {18} と {19} の間の{18.5}に次のページに示す吸収補正 (ACACA) 用の入力を加える。

-- page 25 --

{18.5} ACACA absorption correction by integration. 以下の (a) から (f) まで

(a) **ACACA control** (10I3)

1 - 3	NPU:	Print out and/or file-out.	<< 2 >>
		All informations on the first 30 reflections are always printed out.	
		= 2: File output on unit 16.	
		= 3: File output on unit 16 with print on unit 6.	
4 - 6	MP:	Number of sections in the x direction.	
7 - 9	MQ:	Number of sections in the y direction.	
10 - 12	MR:	Number of sections in the z direction.	
13 - 15	NBF:	Number of plane equations.	
16 - 18	INC:	Crystal shape	
		= 1: Centric	
		= 2: Acentric	
19 - 21	INV:	Output with MH, MK, ML,Fo, sigF, SF, TBAR	
		= 3 always!	
22 - 24	INST:	Read reflections with MH, MK, ML,Fo, sigF, SF	<< 2 >>
		= 2: Rigaku	
		= 3: Philips	

(b) **Matrix for crystal orientation** (3F13.10)

Three lines (I = 1, 3) are required.

1 - 13	OM(I,1):	Components of the matrix OM in order to move a crystal
14 - 26	OM(I,2):	from the setting determined by UB matrix.
27 - 39	OM(I,3):	

(c) **Equations of the crystal faces** (10F7.4)

NBF sets are needed.

1 - 7	A:	The coefficients of the following equations:
8 - 14	B:	
15 - 21	C:	$Ax^2 + By^2 + Cz^2 + Dxy + Eyx + Gzx + Hx + Py + Qz = f$
22 - 28	D:	
29 - 35	E:	for each face in cm unit. Usually, f should be set to be 1.0.
36 - 42	G:	
43 - 48	H:	
49 - 56	P:	
57 - 63:	Q:	
64 - 70	f:	

(d) Definition of the integration area (10F8.4)

1 - 8	UA:	minimum x	(cm)
9 - 16	UB:	maximum x	(cm)
17 - 24	VA:	minimum y	(cm)
25 - 32	VB:	maximum y	(cm)
33 - 40	WA:	minimum z	(cm)
41 - 48	WB:	maximum z	(cm)

(e) Grid-interval
グラフライン 3行

(f) Grid weight
グラフライン 3行

(A) Description of RADY

(1) General Description

RADY is a FORTRAN program for structure factor calculation and full-matrix least-squares refinements of crystal structures. It makes possible to refine the following quantities in a normal treatment with x-ray or neutron diffraction data: these parameters are scale and extinction factors, atomic coordinates, overall temperature factors, isotropic, anisotropic and anharmonic thermal parameters, the multiplicities of sites of core and/or valence electrons, and the coefficients of $\sin\theta/\lambda$ in scattering form.

This program has been extended from the ORFLS (Busing and Levy, 1959, ORNL Central Files Memorandom, Oak Ridge National Laboratory, 1-43), the RADIEL (Coppens et al., 1979, Acta Crystallogr., A35, 63-72) and the RADY (Sasaki, 1982, XL Report, ESS, State Univ. of New York, 1-17). For further information on the method, the user may refer to their papers. The treatments of the extinction effect are based on the formalism of Becker and Coppens (1974, Acta Crystallogr., A30, 129-147). It is extended to refine anharmonic thermal parameters in the Gram-Charlier formalism up to sixth order (Haga, 1982, private communication; Johnson and Levy, 1974, International Tables for X-ray Crystallography, vol. IV). Also, several corrections such as Lorentz, polarization and absorption effects and thermal diffuse scattering (Skelton and Katz, 1969, Acta Crystallogr., A25, 563-568) are available in this program. Using the selection of control data, it is possible to choose the type of the radiation source including synchrotron x-rays, monochromators, F(obs)/I(obs) as input. This program has the functions to average the equivalent reflections on its symmetry requirement and to calculate geometical values needed for anisotropic extinction corrections.

By using replaceable dummy subroutines, the users can perform the refinements with various functions. The following subroutines may vary from refinement to refinement and should be supplied by the user in most cases: they are RESET, PATCH, SIGFO, PATCHS, EPATCH, ACCABS, CONV, and NSHIFT. The description for each subroutine will be given later.

(2) The Least-Squares Method

Although the function form of the crystal structure factor expressed as

$$F = \sum_{n=1}^{N} f_n \exp\{2\pi i(hx_n+ky_n+lz_n)\}\exp(-W) \qquad (1)$$

is transcendental, the form of the observation equations and the normal equations are not linear. Therefore, the form of the structure factor must be approximated by a truncated Taylor series. In such cases, the parameters of a structure model such as f_n and (x_n,y_n,z_n) can be varied in a non-linear least-squares procedures by minimizing the sum of the squares of the differences between the observed and calculated structure factors:

$$D = \sum_{i=1}^{m} w_i(|F_o| - |kF_c|)_i^2 = \Sigma\, w_i r_i^2 \ , \qquad (2)$$

where w_i is the weight of the observation, k is a scale factor and the summation are taken over all the observed reflections. In order to minimize D, the partial derivatives must be equal to zero. This gives the n normal equations

$$\frac{\partial D}{\partial p_j} = 2 \sum_{i}^{m} w_i r_i \frac{\partial r_i}{\partial p_j} = 0 \qquad (j = 1, 2, \cdots, n) \qquad (3)$$

and therefore,

$$\sum_{i=1}^{m} w_i(|F_o|-|kF_c(p_1,\cdots,p_j,\cdots)|) \frac{\partial|kF_c(p_1,\cdots,p_j,\cdots)|}{\partial p_j} = 0$$

$$(j = 1, 2, \cdots, n) \qquad (4)$$

where p_i is any of the scale, extinction, occupancy, positional or thermal parameters. There are n normal equations and m observational equations. In the cases that the approximate value of a parameter, t_j, is known for the true value, p_j, we can approximate the function as a Taylor series:

$$|kF_c(p_1,\cdots,p_j,\cdots)| = |kF_c(t_1,\cdots,t_j,\cdots)| + \frac{\partial|kF_c(t_1,\cdots t_j,\cdots)|}{\partial p_1}\Delta p_1 + \cdots + \frac{\partial|kF_c(t_1,\cdots t_j,\cdots)|}{\partial p_j}\Delta p_j + \cdots \quad (j = 1, 2, \cdots, n) \qquad (5)$$

with the parameter shift Δp_j given by

$$\Delta p_j = p_j - t_j \ . \qquad (6)$$

The higher terms in Δp_j than the first will be small compared with the linear term and can be neglected in the least-squares procedure. The substitution of eq. (5) in eq. (4) gives

$$\sum_{i=1}^{m} w_i\{\Delta F - \frac{\partial|kF_c|}{\partial p_1}\Delta p_1 - \cdots - \frac{\partial|kF_c|}{\partial p_j}\Delta p_j - \cdots\}\frac{\partial|kF_c|}{\partial p_j} = 0 \quad (j = 1, 2, \cdots, n) \qquad (7)$$

where ΔF operates as the observational quantity and is written by

$$\Delta F = |F_o| - |kF_c(p_1,\cdots,p_j,\cdots)| \qquad (8)$$

Expansion of eq. (7) leads to the following set of equations which are solvable for Δp_j's.

$$\begin{aligned}
&\sum_{i=1}^{m} w_i(\partial|kF_c|/\partial p_1)^2\Delta p_1+\cdots+\sum_{i=1}^{m} w_i(\partial|kF_c|/\partial p_1)(\partial|kF_c|/\partial p_n)\Delta p_n \\
&\qquad = \sum_{i=1}^{m} w_i\Delta F(\partial|kF_c|/\partial p_1) \\
&\qquad\qquad \cdots\cdots\cdots\cdots\cdots \\
&\qquad\qquad \cdots\cdots\cdots\cdots\cdots \\
&\sum_{i}^{m} w_i(\partial|kF_c|/\partial p_j)(\partial|kF_c|/\partial p_1)\Delta p_1+\cdots+\sum_{i}^{m} w_i(\partial|kF_c|/\partial p_j)(\partial|kF_c|/\partial p_n)\Delta p_n \\
&\qquad = \sum_{i=1}^{m} w_i\Delta F(\partial|kF_c|/\partial p_j) \\
&\qquad\qquad \cdots\cdots\cdots\cdots\cdots \\
&\qquad\qquad \cdots\cdots\cdots\cdots\cdots \\
&\sum_{i}^{m} w_i(\partial|kF_c|/\partial p_n)(\partial|kF_c|/\partial p_1)\Delta p_1+\cdots+\sum_{i}^{m} w_i(\partial|kF_c|/\partial p_n)^2\Delta p_n \\
&\qquad = \sum_{i=1}^{m} w_i\Delta F(\partial|kF_c|/\partial p_n)
\end{aligned} \tag{9}$$

Thus, the parameter shifts can be calculated by multiplying the inverse matrix, $((a_{jk}))^{-1}$, by the vector b_k:

$$\Delta p_j = ((a_{jk}))^{-1} b_k \tag{10}$$

where

$$a_{jk} = \Sigma w(\partial|kF_c|/\partial p_j)(\partial|kF_c|/\partial p_k)$$

and

$$b_k = \Sigma w\Delta F(\partial|kF_c|/\partial p_k) \ .$$

(3) Extinction Correction

In this program, the formalism of Becker and Coppens is used for the extinction correction (Becker and Coppens, 1974, Acta Crystallogr., A30, 129-147; A30, 148-153). According to their paper, the extinction-affected structure factor is defined as

$$|F| = y^{1/2}|kF_c| \ , \tag{11}$$

where k, y and F_c are scale, extinction correction and kinematical structure factors, respectively. The extinction correction factor can be written as follows:

$$y = y_p(x)y_s(X) \tag{12}$$

in the neutron diffraction case, and

$$y = \frac{y_p(x)y_s(X)\cos^2 2\theta_M + y_p(x\cos^2 2\theta)y_s(X\cos^2 2\theta)\cos^2 2\theta}{\cos^2 2\theta + \cos^2 2\theta_M} \tag{13}$$

in the polarized x-ray difraction case, where θ_M is the Bragg angle of the monochromator. $y_p(x)$ and $y_s(X)$ are primary and secondary extinction corrections, respectively, and are given by

$$y_p(x) = \left(1 + 2x + \frac{A(\theta)x^2}{1+B(\theta)x}\right)^{-1/2} \tag{14}$$

with $x = \left(\frac{3}{2}\frac{\lambda^4}{V^2}a^2\right)F_c^2\rho^2$ and

$$y_s(X) = \left(1 + cX + \frac{A(\theta)X^2}{1+B(\theta)X}\right)^{-1/2} \qquad (15)$$

with $X = \left(\frac{\lambda^3}{V^2}\, a^2 \overline{T}\right) F_c^2 \psi$,

where a is 10^{-12} cm or (e^2/mc^2) for the neutron or x-ray diffractions; λ, V and T are the wavelength of the radiation in Å, the volume of the unit cell in $Å^3$ and the absorption-weighted mean path length, respectively. ψ is the quantity on the mosaic distribution given by

$$\psi = \rho \,/\, \left(1 + \frac{\rho^2 \sin^2 2\theta}{g^2}\right)^{1/2} \qquad (16)$$

(a Gaussian distribution)

and

$$\psi = \rho \,/\, \left(1 + \rho\, \frac{\sin 2\theta}{g}\right) . \qquad (17)$$

(a Lorentzian distribution)

In the above equations, g and ρ (= r/λ) are the extinction parameters which should be refined in the least-squares procedure.

(4) Anharmonic Thermal Motion

The method to describe anharmonic thermal motion is based on the 3-dimensional Gram-Charlier expansion of the trivariate Gaussian probability density function. It is a Taylor-series-like expansion and can be related to the Fourier transformation of temperature factors. For details, see Johnson and Levy (1974, International Tables for X-ray Crystallography, vol. IV).

The probability density function, $\rho(x)$, of a trivariate normal distribution is given by

$$\rho(x) = \frac{\{\det(g)\}^{1/2}}{(2\pi)^{3/2}} \exp\{-\frac{1}{2} g_{mn}(x^m-x_0^m)(x^n-x_0^n)\} \qquad (18)$$

where the matrix $(\!(g_{mn})\!)$ is the inverse of variance-covariance matrix which represents the second moments about the mean position x_0 of the atom. Although the Gaussian probability density function provides an adequate model for thermal motion, more elaborate models are needed to describe anharmonicity. The Gram-Charlier series expansion is one of such models to correct the Gaussian expression. Using this expansion, the probability density function, $p(x)$, is approximated by

$$p(x) = \Big(1 - \frac{\partial a^p}{\partial x_p} + \frac{1}{2!}\frac{\partial}{\partial x_p}\frac{\partial}{\partial x_q} b^{pq} - \cdots$$

$$+ (-1)^j \frac{1}{j!}\frac{\partial}{\partial x_p}\cdots\frac{\partial}{\partial x_{p_j}} c^{pqr\cdots p_j}\Big)\, \rho(x) \qquad (19)$$

The coefficient tensors are symmetric for all permutations of indices. The Fourier transformation of the Gram-Charlier expansion for each atom is given by

$$t(\underset{\sim}{h}) = \left(1 + \frac{(2\pi i)^3}{3!} c^{pqr} h_p h_q h_r + \frac{(2\pi i)^4}{4!} d^{pqrs} h_p h_q h_r h_s \right.$$

$$\left. + \frac{(2\pi i)^5}{5!} e^{pqrst} h_p h_q h_r h_s h_t + \frac{(2\pi i)^6}{6!} f^{pqrstu} h_p h_q h_r h_s h_t h_u \right.$$

$$\left. + \cdots\cdot\right) \exp(-\beta^{pq} h_p h_q) \qquad (20)$$

where h is Miller indices and β is the anisotropic temperature factor. Therefore, the structure factor in eq. (1) is presented as:

$$F = \Sigma f \exp(2\pi i \underset{\sim}{h} \cdot \underset{\sim}{x}) t(\underset{\sim}{h}) \qquad (21)$$

By using the tensors up to the 6th rank, any point symmetry of crystals can be represented. The restriction on the coefficients for crystallographic site symmetries are given in published tables (see pp. 325-334 of International Tables for X-ray Crystallography, vol. IV, 1974).

(5) Requirements on Calculations

In the FACOM M360MP system, this program has the following limitation of arrays:

Number of reflections	No limit	(IWKREF = 1)
	10000	(IWKREF = 0)
Number of variables	841	
Number of parameters refined (NV)	425	
Number of matrix elements	NV(NV+1)/2	
Number of atoms	99	
Number of symmetry operations	48	
Number of scales	20	
Number of scattering factors	25	
Number of κ parameters	10	

The following files should be asigned during calculations:

(i) Input files:
- Unit 20 on reflection data (IFOFC ≠ 5)
- Unit 23 on initial atomic parameters (IAPS ≠ 5)
- Unit 24 on atomic scattering factors (IASFT ≠ 5)
- Unit 19 on derivatives (NOMAT = 1)

(ii) Output files:
- Unit 26 on structure factor tables (LFOFC ≠ 0)
- Unit 27 on variance-covariance matrix (LORFFE ≠ 0)
- Unit 28 on atomic parameters (LAPS ≠ 1 & ≠ 7)
- Unit 19 on derivatives (ITCH ≠ 0)

(iii) Working files:
- Unit 14 on reflection data (IWKREF = 1)
- Unit 15 See page S-1
- Unit 16 See page S-3

(6) Description of Input Data

{1} Title (18A4)

Any comments on this calculation.

{2} File Control (24I3)

Cols.	Variables	Contents	Default
1 - 3	IFOFC:	File unit number on reflection data. =5: Card (cards, FT05F001, or SYSIN). ≠5: File 20.	<< 20 >>
4 - 6	IASFT:	Unit no. on atomic scattering factor tables. Effective only when IASF=1 in {4}. =5: Card or its equivalent. ≠5: File 24.	<< 24 >>
7 - 9	IAPS:	Unit no. on initial atomic parameters. The details of atomic parameters are shown in {20} to {24}. =5: Card or its equivalent. ≠5: File 23.	<< 23 >>
10 - 12	LFOFC:	Output for final structure factors. =0: No output. ≠0: The values of h, k, l, Fo, Fc, Ac, Bc, and σF will be written on file 26 using the FORMAT(3I5,5F12.5).	<< 0 >>
13 - 15	LORFFE:	Output for variance-covariance matrix for function and error programs such as ORFFE. =0: No output. ≠0: Output on file 27.	<< 0 >>
16 - 18	LAPS:	Output for atomic parameters refined. =7: Output on card at final cycle. =1: No output. ≠1 & ≠7: Output on file 28 at each cycle.	<< 28 >>

19 - 21 NODIV: Calculation of derivatives. << 0 >>

=0: For normal operation.

=1: Derivatives are not calculated but are taken from file 19 written on previous cycle.

22 - 24 NOMAT: Calculation of inverse matrix. << 0 >>

=0: For normal operation.

=1: Inverse matrix is not calculated but is taken from file 19 written on previous cycle. When NODIV=0, the program will set NOMAT=0.

25 - 27 ITCH: Output for derivatives and inverse matrix. << 0 >>

=0: For normal operation.

≠0: Derivatives and inverse matrix are to be written on file 19 for later use.

28 - 30 IT: Output to line printers for atomic parameters. << 0 >>

=0: Only at the first and last cycles.

=1: At each cycle.

31 - 33 LPAM: Output to line printers for change-of-parameters. << 0 >>

=0: Only variable parameters will be picked up.

=1: All parameters will be printed out.

34 - 36 IWKREF: Working area to store the reflection data used for refinements. << 0 >>

=0: Use of memory array (≦10,000).

=1: Use of binary files (unit no. 14). No limit of the number of reflections.

{3} Parameter Control (24I3)

1 - 3 NA: The number of atoms in the asymmetric unit. << 6 >>

4 - 6 NS: The number of symmetry cards. << 4 >>

The number may be reduced to a half for the centrosymmetric structure with origin at center. The reduction may also be possible for the lattice type. See AMULT in {10}.

7 - 9 NQ: The number of scale factors. << 1 >>

Different scale factors will be applied according to SQ of reflection data (see {17}).

10 - 12 LITF: Selection of thermal parameters. << 0 >>

Only valid for reading and writing parameters. All calculations are based on u.

=-1: Read u, output β.

=0: Read u, output u.

=1: Read β, output u.

=2: Read β, output β.

13 - 15 ICC: Absorption correction for a spherical crystal. << 0 >>

=0: No correction.

=1: Yes. =2 (see page 5-3) ACACA使用(任意の形での吸収補正)

16 - 18 J4CD: The model of the four-circle diffractometer used. << 0 >>

=0: Philips PW1100 or PW1120.

=1: Syntex $P2_1$.

=2: Rigaku AFC-5.

=3: Picker (SUNY SB).

=4: PF-BL10A (or probably for the Busing & Levy's coordinates).

=5: Others.

19 - 21 IWBCD: Printing of structure factor tables << 0 >>

=-1: No print out.

=0: At every cycle.

=1: Following last cycle only.

22 - 24 JANN: Calculation of bond distances and angles. << 0 >>

=0: No calculation.

=n: The first n atoms will be considered. The extra input data are necessary in {29}.

25 - 27 JELIPS: Calculation of thermal ellipsoids. << 0 >>

=0: No calculation.

=n: The ellipsoids for the first n atoms will be calculated.

28 - 30 MONOCH: Type of the monochromator used. << 0 >>

=0: Vertical type.

=1: Horizontal type like Philips PW1100.

31 - 33 NEUT: Radiation used. << 0 >>

=0: X-rays.

=1: Neutron.

34 - 36 MAGNET: Magnetic scattering. << 0 >>

=0: No.

=1: Yes with NEUT=1

37 - 39 INTFOB: Input for structure factors. << 0 >>

=0: F(obs).

=1: I(obs).

40 - 42 LORENZ: Lorentz correction. << 0 >>

=0: No correction.

=1: Yes.

43 - 45 IPOR: Polarization correction for F(obs). << 0 >>

=0: No correction.

=1: Yes.

46 - 48 IPORE: Polarization correction on extinction terms. << 0 >>

=0: No correction.

=1: Yes.

49 - 51 ITDS: The correction for thermal-diffuse-scattering. << 0 >>

=0: No correction.

=1: Yes, for the isotropic TDS correction.

=2: Yes, for the anisotropic TDS correction.

52 - 54 IREFAV: Averaging or rearrangement of reflection data. << 0 >>

This is not correct. see page S-2

=0: Normal operation.

=1: Averaging into cubic independent reflections.

=2: Averaging into tetragonal.

=3: Averaging into orthorhombic.

=4: Averaging into monoclinic.

=5: Averaging into hexagonal.

=6: Averaging into trigonal (hexagonal setting).

55 - 57 IREFEX
58 - 61 IOUTP

See page S-2

{4} Atomic Scattering Factor Control (24I3)

1 - 3 NSF1: The number of atomic scattering factor tables << 3 >>
for core electrons. This number is also applied
for f' and f". In the case of neutron diffraction,

use NSF1 with ISF1=1, where NSF2 for f' and NSF3 for f" are set zero in the program. If magnetic scattering is considered, use NSF4 for such scattering factors.

4 - 6 NSF4: The number of atomic scattering factor tables of valence electrons. << 0 >>

7 - 9 ISF1: Form to read the f(core) tables. << 4 >>
The form is defined as follows:
=1: Scattering angle independent. (7F10.5)
=2: Scattering angle dependent. (8F9.6)
An atom has 32 values within the $\sin\theta/\lambda$ range from 0.0 to 1.55 at interval of 0.05.
=3: Scattering angle dependent. (10F7.3)
This format is based on the lists shown in International Tables for X-Ray Crystallography, vol. IV. There are 48 values per atom listed in tables up to $\sin\theta/\lambda$=1.0.
From 0.0 to 0.2 at interval of 0.01;
From 0.2 to 0.5 at interval of 0.02, including the values at each 0.05;
From 0.5 to 0.7 at interval of 0.05;
From 0.7 to 1.0 at interval of 0.1.
=4: Scattering angle dependent. (10F8.3)
There are 150 points per atom within the $\sin\theta/\lambda$ range from 0.0 to 1.49 at interval of 0.01.

10 - 12 ISF2: Form to read f'(core) tables. << 1 >>
The selection of format is given in ISF1.

13 - 15 ISF3: Form to read f"(core) tables. << 1 >>
The selection of format is given in ISF1.

16 - 18 ISF4: Form to read f(valence) tables. << 4 >>
The selection of format is given in ISF1.

19 - 21 IASF: Use of a special file "ASFT" for f(core) and f(valence) values. Valid only when ISF1=ISF4=4 and IASFT≠5 in {2}. << 0 >>
=0: No use.
=1: Yes, the file "ASFT" will be used.

22 - 24 NAS: The number of atoms for anomalous scattering correction. << 0 >>

=0: No correction.

=n: The first n atoms will be applied for this correction.

25 - 27 NAV: The number of atoms calculated using the models of anharmonic thermal vibration. << 0 >>

=0: Harmonic isotropic or anisotropic thermal models. (Normal operation)

=n: The Gram-Charlier expansion will be used for anharmonic motion of some of the first n atoms. The selection for each atom will be made using JTF(I) in {25}.

28 - 30 NAVRST: Anharmonic thermal calculations. << 0 >>

=0: Normal calculation.

=1: Only the reflections that h*k*l ≠ 0 will be considered for calculations.

{5} Reference of Scattering Factor Tables in File "ASFT" (24I3)

This card should be present only when IASF=1 in {4}.

NNN(I),I=1,(NSF1+NSF4)

NNN: The reference number of atomic scattering factor for the corresponding atom.

{6} Least-Squares Control (24I3)

1 - 3 NC: The number of cycles of least-squares calculations. << 0 >>

=0: Only structure factors will be calculated based on the input data. (No least-squares calculations will be made).

=n: For nomal least-squares calculations.

4 - 6 IWHPK: Switch of parameter selection cards in {27}. << 0 >>

IWHPS and IWHPK should not both be non-zero.
IWHPK cannot be used to change from isotropic
to anisotropic refinement.

=0: For normal operation.

=1: Separate parameter selection cards are supplied for each of the NC cycles.

7 - 9 IFSQ: Switch of refinements. << 1 >>

=1: For refinement on scaled F.

=2: For refinement on scaled F^2.

10 - 12 ICENT: Switch of centrosymmetry at origin. << 1 >>

=1: For centrosymmetric structures with origin at center.

=2: For all other structures.

13 - 15 IW: Weight for the least-squares calculations. << 1 >>

=1: Unit weight.

=2: $wt=1/\sigma F^2$.

=3: $wt=1/\sigma F^2$ and the SIGFO subroutine will be called.

=4: $wt=\sigma F^2$ and the SIGFO subroutine will be called.

=5: Unit weight and the SIGFO subroutine will be called.

When IW≧3, the replaceable subroutine, SIGFO(FH,FK,FL, FO,SIGF,SQ,TBAR,N) may be used for weighting and rejecting reflections. In order to reject reflections, set N=1 or wt^2<WMIN, where WMIN will be given in {10}. The special weighting scheme may be defined in this subroutine.

16 - 18 IRESET: Switch of RESET, PATCH, and ACCABS subroutines. << 0 >>

=0: No calling them.

=1: Replaceable RESET and PATCH subroutines are to be used for resetting of parameters and derivatives.

=2: Replaceable subroutine, ACCABS(FH,FK,FL,FO, SIGF,FC,N) is called following the calculation of structure factors for each reflection. It may be used to reject structure factors from the least-squares refinement (N=1 for

rejection; otherwise set N=0).

19 - 21 IREF: Form of reflection data read. << 0 >>

=0: Input FH,FK,FL,FO,SIGF,SQ.

=1: Input FH,FK,FL,FO,SIGF,SQ,TBAR.

=2: Input FH,FK,FL,FO,SIGF,SQ,(EFP(J),J=1,7).

FH: Miller indices.

FK:

FL:

FO: The observed structure factor |F(obs)|.

SIGF: The standard deviation of F(obs).

SQ: The reference number of scale factor.

TBAR: The mean path length (cm).

EFP(1): Components of the vector normal to

-(3) the plane containing the incident and diffracted beams.

EFP(4): Components of the unit vector

-(6) parallel to the incident beam.

EFP(7): =TBAR.

ACACAを使う場合(ICC=2)→IREF=1にする

see page S-3

22 - 24 IWHSO: Scale factor application. << 0 >>

=0: For normal operation.

25 - 27 MANYR1:

28 - 30 MANYR2: Limit of total reflections refined. << 0,0 >>

IMUCH is defined by 1000*MANYR1+MANYR2.

If IMUCH>0, only first IMUCH reflections will be used in the structure factor calculation and refinement. If IMUCH=0, all data are included.

31 - 33 IWHPS: Repetation of refinements. << 1 >>

=0: For normal operation.

=n: The refinement will be repeated for n different sets of initial parameters.

34 - 36 MAXIC: Limit for the number of cycles. << 0 >>

=0: For nomal operation (No maximums).

=n: The number of cycles for self submitting series of refinement.

37 - 39 ICONV: Convergence test. << 0 >>

=0: For no convergence test.

=1: A convergence test is applied.

40 - 42 ICORR: Output of correlation matrix. << 0 >>

=0: Complete correlation matrix is printed out.

=1: Only correlation matrix with the absolute value of p≧0.5 are printed out.

43 - 45 IWHTF: Run switch for thermal parameters. << 2 >>

=1: Program will not stop, but diagnostics will be printed out.

=2: Program will stop if non-real temperature factors encountered.

46 - 48 IWHEX: Run switch for extinction parameters. << 1 >>

=1: Program will not stop, but diagnostics will be printed out.

=2: Program will stop if non-real extinction factors encountered.

{7} Extinction Control (24I3)

1 - 3 JEXT: Extinction options. << 0 >>

=0: No extinction correction.

=1: Isotropic extinction correction.

=2: Anisotropic extinction correction for Type I.

=3: Anisotropic extinction correction for Type II.

=-2: Isotropic will be converted to anisotropic for Type I crystal.

=-3: Isotropic will be converted to anisotropic for Type II crystal.

=4: Anisotropic extinction correction for general case (simultaneous refinement on g and r).

4 - 6 ITYPE: Type of isotropic extinction. << 1 >>

ITYPE is only relevant when JEXT=1.

=1: Type I extinction correction dominated by mosaic spread (r.m.s. in seconds). $\eta=5.8186/g$.

=2: Type II extinction correction dominated by

particle size (cm).
Radius of mean particle is defined
by $10^{-4}g\lambda$, where λ is wavelength in 10^{-8} cm.

=3: General case (simultaneous refinement on g & r).
This should be attempted only for severe
extinction case.

7 - 9 IDISTR: The angular orientations of the crystalline. << 1 >>

=1: The diffracting power $\bar{\sigma}$ is approximated by
a Gaussian distribution. See expression 40a
of Becker and Coppens (1974).

=2: A Lorentzian distribution (See expression 41a).

10 - 12 IMOSC: Representation of g

=0: $g=\sqrt{D'ZD}$ by Coppens & Hamilton.

=1: $g=1/\sqrt{D'YD}$ by Nelmes & Thornley.

{8} L-Shell Control (24I3)

1 - 3 NKAPPA: The number of κ parameters. << 0 >>

4 - 6 NE: Constraint-switch for total number of electrons. << 0 >>

=0: Total number of electrons is not constrained.
Even if NE=0, the neutrality of atomic charges
can also be constrained by using RESET and
PATCH subroutines.

=1: Number of electrons is constrained to a constant.
When NE=1, either core or valence electron shell
must be varied.

7 - 9 IELEC: Set zero. << 0 >>

{9} Boundary Conditions for Rejecting Reflections (7F10.5)

1 - 10 STLMIN: Minimum limit of $\sin\theta/\lambda$. << 0.0 >>

11 - 20 STLMAX: Maximum limit of $\sin\theta/\lambda$. << 1.5 >>

21 - 30 FOMIN: Minimum limit of $|F(obs)|$ as input. << 0.0 >>
The $|F(obs)|$ must be the value before scalling.

IFMIN =0: Normal operation.
=1: If FOMIN<0, FOMIN=3*|FOMIN|/(1+2cos$^2\theta$).
=2: If FOMIN≧1000, F' should be greater than or equal to FOMIN/1000, where
F' = |F(obs)|/sin2θ(1+cos2θ)/(1+cos^{2}2θ).

31 - 40 FOMAX: Maximum limit of |F(obs)| as input. << 10^6 >>

41 - 50 OMTSIG: The reflections being |F(obs)| less than << 0.0 >>
OMTSIG*σF will be rejected in the calculation.

{10} Crystallographic Constants (7F10.5)

1 - 10 AMULT: Scale multiplied to the calculated structure << 0.0 >>
factor. This option can be saved the computing time in the CALC subroutine. The number of symmetry-operation cards must be related to the value of AMULT. For example, if AMULT=4.0, the number of operation cards must be reduced to 1/4.
General case = 1.0
Face-centered lattice = 4.0
Base-centered lattice = 2.0
Body-centered lattice = 2.0
Centrosymmetric structure with origin at center = Multiply the above number by 2.0.

11 - 20 AMIUR: Linear absorption coefficientm in cm^{-1} unit. << 1.0 >>
This value is necessary to calculate the absorption factors and the mean path lengths.
Reference: Table 3.2.2B (p.166-169) of International Tables for X-ray Crystallography, vol.III.

21 - 30 TTHM: The 2θ value of monochromator. << 0.0 >>
<ex> TTHM=12.185 for the Mo Kα ratiation from a graphite (001) monochromator.

31 - 40 WMIN: Minimum limit of a weight of $1/\sigma F^2$ or σF^2 for << 0.0 >>
the least-squares calculation.

41 - 50 TBARG: Mean pathlength in cm unit when IREF=0.
If TBARG≦0 & IREF=0 & NEUT=1, TBAR=TBARG=1.0.

If TBARG≦0 & IREF=0 & NEUT=0, TBAR=TBARG=0.01.

{11} Atomic Scattering Factors

(11-1) Atomic scattering factor tables for core electrons, f(core).
NSF1 sets of scattering factors are necessary (See NSF1 in {4}).
The FORMAT should be followed by ISF1 in {4}.

(11-2) Real parts of anomalous scattering terms for core electrons, f'.
NSF1 sets with the format given by ISF2. No input when NEUT=1.
Reference: Table 2.3.1 (pp.149-150) of International Tables for X-Ray Crystallography, vol. IV.

(11-3) Imaginary parts of anomalous scattering factors for core electrons, f". NSF1 sets with the format given by ISF3.
No input when NEUT=1.

(11-4) Atomic scattering factor tables for valence electrons, f(valence).
NSF4 sets with the format followed by ISF4. This card is necessary only when NSF4≠0.

When IASF=1 in {4}, the atomic scattering factors belonging to (11-1) and (11-4) will be read from file "ASFT", while (11-2) and (11-3) will be read on file unit 5 or 24 followed by IASFT in {2}.

{12} Symmetry Operation cards (9F5.2,3F9.6)

NS sets of operation cards are required in the form of the following matrices.

1 - 5	R11: Rotation matrix.
6 - 10	R12:
11 - 15	R13:
16 - 20	R21:
21 - 25	R22:
26 - 30	R23:
31 - 35	R31:
36 - 40	R32:

$$\begin{pmatrix} x' \\ y' \\ z' \end{pmatrix} = \begin{pmatrix} R11 & R12 & R13 \\ R21 & R22 & R23 \\ R31 & R32 & R33 \end{pmatrix} \begin{pmatrix} x \\ y \\ z \end{pmatrix} + \begin{pmatrix} T1 \\ T2 \\ T3 \end{pmatrix}$$

41 - 45 R33:

46 - 54 T1: Translation matrix.

55 - 63 T2:

64 - 72 T3:

{13} Cell Dimensions and Wavelength (7F10.6)

1 - 10	A1: Lattice constant a or a*	<< 0.0 >>
11 - 20	A2: Lattice constant b or b*	<< 0.0 >>
21 - 30	A3: Lattice constant c or c*	<< 0.0 >>
31 - 40	A4: Lattice constant α, α^*, cosα, or cosα^*	<< 90.0 or 0.0 >>
41 - 50	A5: Lattice constant β, β^*, cosβ, or cosβ^*	<< 90.0 or 0.0 >>
51 - 60	A6: Lattice constant γ, γ^*, cosγ, or cosγ^*	<< 90.0 or 0.0 >>
61 - 70	FLAME: Wavelength in angstrom.	<< 0.71069 >>

{14} Setting Parameters of Four-Circle Diffractometer (5E14.7)

The coordinates defined by Busing and Levy (1967) are essential. Two blank cards are necessary even in the case that setting parameters are not used in the calculation. Values are required if ($|JEXT| \geq 2$ and $IREF \neq 2$) or ITDS=2. << 9 * 0.0 >>

The 1st card:

1 - 14 SETP(1,1)

15 - 28 SEPT(1,2)

29 - 42 SETP(1,3)

43 - 56 SETP(2,1)

57 - 70 SETP(2,2)

The 2nd card:

1 - 14 SETP(2,3)

15 - 28 SEPT(3,1)

29 - 42 SETP(3,2)

43 - 56 SETP(3,3)

{15} Absorption Correction Factor A* (7F10.6)

The absorption correction factor should be given for successive scattering angle Θ (at intervals of Θ=5°) of spheres of radius R. Three cards are always used. Meaningful values are required if ICC=1 or (|JEXT|≧1 & IREF=0).

Reference: Table 5.3.6B (p.302-305) of International Tables for X-Ray Crystallography, vol.III.

The 1st card:

1 - 10	AS(1):	A* for Θ = 0° << 19 * 1.0 >>
11 - 20	AS(2):	A* for Θ = 5°
21 - 30	AS(3):	A* for Θ = 10°
31 - 40	AS(4):	A* for Θ = 15°
41 - 50	AS(5):	A* for Θ = 20°
51 - 60	AS(6):	A* for Θ = 25°
61 - 70	AS(7):	A* for Θ = 30°

The 2nd card:

1 - 10	AS(8):	A* for Θ = 35°
11 - 20	AS(9):	A* for Θ = 40°
21 - 30	AS(10):	A* for Θ = 45°
31 - 40	AS(11):	A* for Θ = 50°
41 - 50	AS(12):	A* for Θ = 55°
51 - 60	AS(13):	A* for Θ = 60°
61 - 70	AS(14):	A* for Θ = 65°

The 3rd card:

1 - 10	AS(15):	A* for Θ = 70°
11 - 20	AS(16):	A* for Θ = 75°
21 - 30	AS(17):	A* for Θ = 80°
31 - 40	AS(18):	A* for Θ = 85°
41 - 50	AS(19):	A* for Θ = 90°

{16} TDS correction control (7F10.3)

Present only when ITDS≧1.

1 - 10	APERV:	Detector aperture perpendicular to the scanning direction (in degree).	<< 1.0 >>
11 - 20	APERH:	Detector aperture parallel to the scanning.	<< 1.0 >>
21 - 30	SWDA:	Scan width in ω(deg), which is defined by	<< 0.0 >>
31 - 40	SWDB:	SWDA+SWDB*tanΘ.	<< 0.0 >>
41 - 50	SCTYPE:	Scan type.	<< 0.0 >>
		ω-2Θ scan when ω is greater than SCTYPE or equal to.	
		ω scan when ω is less than SCTYPE.	
51 - 60	TEMPR:	Temperature in Kelvin.	<< 298.2 >>
61 - 70	SPECW:	Specific weight in g/cm^3.	<< 1.0 >>

{17} Stiffness Tensor S_{ij} (6F10.3)

Present only when ITDS≧1.

Stiffness tensor in Mbar or 10^{12} dyne/cm^2 unit.

The valiables S(i,j) must be read as:

Each card contains six values (j=1,6); six cards are needed (i=1,6).

Diagonal elements and one of off-diagonal element pairs are necessary.

The 1st card:

1 - 10	S(1,1):	S_{11}
11 - 20	S(1,2):	S_{12}
21 - 30	S(1,3):	S_{13}
31 - 40	S(1,4):	S_{14}
41 - 50	S(1,5):	S_{15}
51 - 60	S(1,6):	S_{16}

The 2nd card:

1 - 10	S(2,1):	S_{21}
11 - 20	S(2,2):	S_{22}
...	....	
51 - 60	S(2,6):	S_{26}

The 6th card:

1 - 10	S(6,1):	S_{61}
11 - 20	S(6,2):	S_{62}
...	....	
51 - 60	S(6,6):	S_{66}

{18} Variable FORMAT for Reflection Data (Input) (18A4)

1 - 72 FM(I),I=1,18

When IREF=0 in {6}, for variables FH,FK,FL,FO,SIGF,SQ.
<< 3F4.0,F10,3,F6.3,11X,F1.0 >>

When IREF=1, for variables FH,FK,FL,FO,SIGF,SQ,TABR.
<< 3F4.0,F10,3,F6.3,2X,F5.0,F10,5,6F5.2 >>

When IREF=2, for variables FH,FK,FL,FO,SIGF,SQ,(EFP(J),J=1,7).
<< 3F4.0,F10,3,F6.3,2X,F5.0,F10,5,6F5.2 >>

ACACAを使って、任意の形での吸収補正を行う場合、IREF=1となり、TBAR 項の読み込みが必要

See page S-3

{19} Reflection Data

{18.5} ACACAを使う場合、そのコントロールを入れる。

See page S-4

The data should be read from file IFOFC in {2} with the FORMAT defined in {18}. All variables including hkl are defined as REAL. When IREF=0, variables "FH,FK,FL,F,SIGF,SQ" will be read. When IREF=1, variables "FH,FK,FL,F,SIGF,SQ,TBAR" will be read. When IREF=2, variables "FH,FK, FL,F,SIGF,SQ,(EFP(J),J=1,7)" will be read.

FH,FK,FL:	Miller indices of the reflection.
F:	The observed magnitude (crystal structure factor).
SIGF.	Standard deviation assigned to this observation.
SQ:	A reference number of scale factors. If this value is zero or blank, SQ=1.0 will be assigned. For the n-th scale factor, the number n should be input as REAL.
TBAR; EFP(7):	The mean path length (cm) in the crystal for the reflection. When a crystal is not sphere, the output from ACACA or an equivalent program should be used.
EFP(1)-(3):	Components of the vector normal to the plane containing the incident and diffracted beams. These are components of a unit vector defined with respect to unit triclinic axes parallel to the real crystal axes.

EFP(4)-(6): Components of the unit vector parallel to the incident beam. This information will be used for TYPE II refinements.

* Usually the values of EFP(1)-(6) will be automatically calculated in RADY, when the setting parameter (UB matrix) of the four-circle diffractometer is given in {14}. When IFOFC=5 in {2}, a card having FH≥100.0 is needed as the termination of input at the end of reflection data.

----- Atomic Parameters -----

A group of atomic parameters described in the following {20} to {24} will be read from file IAPS (see {2}).

The order of atomic parameter cards is:

(1) Scale factors.
(2) Overall temperature factor.
(3) Atomic coordinates for the 1st atom.
Temperature factors for the 1st atom.
Anharmonic temperature factors for the 1st atom (present only when NAV≥1).
Atomic coordinates for the 2nd atom.
Temperature factors for the 2nd atom.
Anharmonic temperature factors for the 2nd atom (present only when NAV≥2).
.....
.....
Atomic coordinates for the NAth atom.
Temperature factors for the NAth atom.
Anharmonic temperature factors for the NAth atom (present only when NAV≥NA).
(4) κ parameters (present only when NKAPPA≠0).
(5) Extinction parameters.

{20} Scale Factors (on file IAPS) (8F9.6)

Scale factors are applied to F(obs) for print-out and write-up to the file LFOFC. It should be noted that refinement is always done using F(calc)/SC(j), where j=1,NQ.

1 - 8	SC(1):	Scale factor for reflections having the reference number 1. (see SQ in {19})	<< 1.0 >>
9 - 16	SC(2):	Scale factor for the reference number 2.	
17 - 24	SC(3):		
.....	....		
	SC(NQ):	Scale factor for the reference number NQ given in {3}.	

{21} Overall Temperature Factor (on file IAPS) (8F9.6)

1 - 19	TO:	Overall temperature factor. TO=0.0 for normal operations.	<< 0.0 >>

{22-1} Atomic Coordinates (on file IAPS) (A4,2F2.0,I2,2F9.5,3F9.6)

The data cards from {22-1} to {22-3} must be arranged for each atom as follows:

(1) {22-1} for the 1st atom,
(2) {22-2} for the 1st atom,
(3) {22-3} for the 1st atom only when NAV≧1,
(4) {22-1} for the 2nd atom,
(5) {22-2} for the 2nd atom,
(6) {22-3} for the 2nd atom only when NAV≧2,
.....

(m) {22-1} for the NAth atom,
(n) {22-2} for the NAth atom,
(o) {22-3} for the NAth atom only when NAV≧NA.

1 - 4	ATOM(I):	Name or symbol to identify the atom

5 - 6	SF(I):	The reference number of scattering factor tables for core electrons; including f, f', and f".
7 - 8	SL(I):	The reference number of scattering factor tables for valence electrons.
9 - 10	IKAPPA(I):	The reference number of radial parameter (kappa) for the atom ATOM(I).
11 - 18	ATK(I):	Multiplicity of core electrons. For normal operations, this value corresponds to the multiplicity for this site. <ex> ATK=0.5, for the special position on a mirror.
19 - 26	ATL(I):	Multiplicity of valence electrons.
27 - 35	XYZ(1,I):	Fractional x coordinate for the atom.
36 - 44	XYZ(2,I):	Fractional y coordinate for the atom.
45 - 53	XYZ(3,I):	Fractional z coordinate for the atom. Although the standard deviations (SXYX(J,I),J=1,3) for x, y and z coordinates are written in the columns from 54 to 80 at each cycle of refinements, such values should be ignored on reading.

{22-2} Temperature Factors (on file IAPS) (6F9.6)

The input form depends on LITF in {3} and JTF(I) in {25}.
All calculations are based on u. Anisotropic temperature factor β_{ij} is defined by:

$$\exp\{-(h^2\beta_{11}+k^2\beta_{22}+l^2\beta_{33}+2hk\beta_{12}+2hl\beta_{13}+2kl\beta_{23})\},$$

and

$u_{ij}=\beta_{ij}/(2\pi^2 a_i a_j)$, where a_i is the i-th reciprocal vector.

1 - 9	BETA(1,I):	Isotropic B or anisotropic β_{11} or u_{11}.
10 - 18	BETA(2,I):	β_{22} or u_{22}.
19 - 27	BETA(3,I):	β_{33} or u_{33}.
28 - 36	BETA(4,I):	β_{12} or u_{12}.
36 - 45	BETA(5,I):	β_{13} or u_{13}.
46 - 54	BETA(6,I):	β_{23} or u_{23}.

{22-3} Anharmonic Temperature Factors (6E12.5)

The following cards are present only if NAV≧1.

(a) Component of the 3rd coefficient tensor (CCC(K,I),K=1,10)

The 1st card:

1 - 12	CCC(1,I):	C_{111}.
13 - 24	CCC(2,I):	C_{112}.
25 - 36	CCC(3,I):	C_{122}.
37 - 48	CCC(4,I):	C_{222}.
49 - 60	CCC(5,I):	C_{113}.
61 - 72	CCC(6,I):	C_{123}.

The 2nd card:

1 - 12	CCC(7,I):	C_{223}.
13 - 24	CCC(8,I):	C_{133}.
25 - 36	CCC(9,I):	C_{233}.
37 - 48	CCC(10,I):	C_{333}.

(b) Component of the 4th coefficient tensor (DDD(K,I),K=1,15)

The 1st card:

1 - 12	DDD(1,I):	D_{1111}.
....	....	
61 - 72	DDD(6,I):	D_{1113}.

The 2nd card:

1 - 12	DDD(7,I):	D_{1123}.
....	....	
61 - 72	DDD(12,I):	D_{2233}.

The 3rd card:

1 - 12	DDD(13,I):	D_{1333}.
13 - 24	DDD(14,I):	D_{2333}.
25 - 36	DDD(15,I):	D_{3333}.

(c) Component of the 5th coefficient tensor (EEE(K,I),K=1,21)

The 1st card:

1 - 12	EEE(1,I):	E_{11111}.
....	....	
61 - 72	EEE(6,I):	E_{22222}.

The 2nd card:

1 - 12	EEE(7,I):	E_{11113}.
....	....	
61 - 72	EEE(12,I):	E_{11133}.

The 3rd card:

1 - 12	EEE(13,I):	E_{11233}.
....	....	
61 - 72	EEE(18,I):	E_{22333}.

The 4th card:

1 - 12	EEE(19,I):	E_{13333}.
13 - 24	EEE(20,I):	E_{23333}.
25 - 36	EEE(21,I):	E_{33333}.

(d) Component of the 6th coefficient tensor (FFF(K,I),K=1,28)

The 1st card:

1 - 12	FFF(1,I):	F_{111111}.
....	....	
61 - 72	FFF(6,I):	F_{122222}.

The 2nd card:

1 - 12	FFF(7,I):	F_{222222}.
....	....	
61 - 72	FFF(12,I):	F_{122223}.

The 3rd card:

1 - 12	FFF(13,I):	F_{222223}.
....	....	
61 - 72	FFF(18,I):	F_{222233}.

The 4th card:

1 - 12	FFF(19,I):	F_{111333}.
....	....	
61 - 72	FFF(24,I):	F_{123333}.

The 5th card:

1 - 12	FFF(25,I):	F_{223333}.
....	....	
37 - 48	FFF(28,I):	F_{333333}.

{23} κ Parameters (on file IAPS) (9F8.4)

This card is present only when NKAPPA≠0.

FCAP(I),I=1,NKAPPA

{24} Extinction Parameters (on file IAPS) (7E10.3)

This card is always necessary even if JEXT=0.

1 - 11	G(1):	G11=G(iso) for IEXT=1, where IEXT=|JEXT| =r/λ for IEXT=1 and ITYPE=2 or 3.
12 - 22	G(2):	G_{22}.
23 - 33	G(3):	G_{33}.
34 - 44	G(4):	G_{12}.
45 - 55	G(5):	G_{13}.
56 - 66	G(6):	G_{23}.
67 - 77	G(7):	$G_{iso}=r/\lambda$; valid only when IEXT=4.

{25} Temperature Factor Selection (72I1)

JTF(I) is a temperature factor selection integer for each atom.
JTF(I), I=1,NA

=1: For isotropic temperature factor.
=2: For anisotropic temperature factor.
=3: For isotropic temperature factor to be converted to anisotropic before refining.

=4: For anharmonic temperature factor.

{26} Parameter Selection and Damping for Each Parameter (72I1)

KI(I): =0: The corresponding parameter is not to be varied.
=1: The corresponding parameter is varied.
=n: The corresponding parameter is varied and the damping factor on this parameter is set as 0.n (n=2,3,••,9).

There are 1+1+NA+(1)+(1) cards needed:

(1) The 1st card for: Scale factor: KI(I), I=1,NQ
(2) The 2nd card for: Overall temperature factor, TO(NQ+1)
(3) The 3rd card for: Multiplicity of atom (electrons), atomic coordinates and temperature factors for the 1st atom.

.....

.....

The (2+NA)th card for: Multiplicity of atom (electrons), atomic coordinates and temperature factors for the NAth atom.

The KI card for each atom has the following selections:
The 1st card:

column		
1	ATK(I):	On multiplicity of core electrons.
2	ATL(I):	On multiplicity of valence electrons.
3	XYZ(1,I):	On x coordinate for ATOM(I).
4	XYZ(2,I):	On y coordinate for ATOM(I).
5	XYZ(3,I):	On z coordinate for ATOM(I).
6	BETA(1,I):	On temperature factor β_{11}, u_{11}, B(iso) or U(iso).
7	BETA(2,I):	On temperature factor β_{22} or u_{22}.
8	BETA(3,I):	On temperature factor β_{33} or u_{33}.
9	BETA(4,I):	On temperature factor β_{12} or u_{12}.

10 BETA(5,I): On temperature factor β_{13} or u_{13}.
11 BETA(6,I): On temperature factor β_{23} or u_{23}.
12 CCC(1,I): On anharmonic temperature factor C_{111}.
13 CCC(2,I): On anharmonic temperature factor C_{112}.
.....
.....
21 CCC(10,I): On anharmonic temperature factor C_{333}.
22 DDD(1,I): On anharmonic temperature factor D_{1111}.
.....
.....
36 DDD(15,I): On anharmonic temperature factor D_{3333}.
37 EEE(1,I): On anharmonic temperature factor E_{11111}.
.....
.....
57 EEE(21,I): On anharmonic temperature factor E_{33333}.
58 FFF(1,I): On anharmonic temperature factor F_{111111}.
.....
.....
72 FFF(15,I): On anharmonic temperature factor F_{111233}.

The 2nd card (present only when NAV≧I):

1 FFF(16,I): On anharmonic temperature factor F_{112233}.
.....
.....
13 FFF(28,I): On anharmonic temperature factor F_{333333}.

**** If any atom is anisotropic, there are eleven parameters for each atom. For isotropic atoms, column 6 is B(iso) or u(iso), and columns 7-11 on the parameter selection card must contain zero or blank.
When NAV≧1 (anharmonic model), two cards are necessary for each atom up to the NAVth atom.

(4) The next card for: κ parameter.
Present only if NKAPPA≠0.

(5) The final card for: Extinction parameter.
Present only if JEXT≠0.

{27} Extra Parameter Selection Cards

Present only if IWHPK=1.

The group {27} must continue until there is a total of NC (Number of Cycles) sets of the following (a), (b) and (c).

(a) Cycle control (5I3)

1 - 3	NV:	Number of parameters varied.
4 - 6	NODIV:	See {2}.
7 - 9	NOMAT:	See {2}.
10 - 12	KIKI:	=0: The following KI cards will be read. =1: The KI cards on the previous cycle will be used. Also, the previous value of NV will be used.
13 - 15	ICHGP:	Exchange of parameters. =0: For normal operation. =10*m+n: Identification of parameters. m=1: Scale factors at the previous cycle will be changed by re-input. m=2: Multiplicity of scattering factors for core electrons will be changed. m=3: Multiplicity for valence electrons. m=4: Atomic coordinates. m=5: Temperature factors. m=6: Kappa parameters. m=7: Extinction parameters. n: Number of parameters changed.

(b) Parameter selection (KI) cards (72I1)

Present only when KIKI=0 in {27} (a).

The construction of KI cards is shown in {26}.

(c) Parameter re-input cards (2I3,F14.8)

Present only when ICHGP>10 in {27} (a).

n sets of cards must be used, where n has been defined in (a).

1 - 3	II:	The IIth parameter of the JJth atom will be considered as the target.
4 - 6	JJ:	The JJth atom will be considered as the target.
7 - 20	T:	Variable which should be changed. The array information: Two dimensional T(II,JJ): XYZ(II,JJ) or BETA(II,JJ). One dimensional T(II): SC(II), FKAP(II), or G(II). One dimensional T(JJ): ATK(JJ) or ATL(JJ).

{28} Refinement with Different Initial Parameters

Present only when IWHPS≧2.
The group of atomic parameters shown in {20} to {26} must continue until IWHPS sets of parameters have been provided.

{29} Calculation of Interatomic Distances and Angles

Present only when JANN≧1.
There are extra inputs needed for the calculation of interatomic distances and angles.

(a) Bond calculation control (2I5,4F10.6)

1 - 5	NSS:	Number of symmetry operations used in bond calculations. When NSS=0, NSS will be assumed to be equal to NS given in {3}.	<< 0 >>
6 - 10	IOMT:	Number of atoms, which will not be included in the bond calculations.	<< 0 >>
11 - 20	DMIN2:	The distances ranging DMIN2≦r≦DMAX2 will be used for angle calculations.	<< 0.05 >>
21 - 30	DMAX2:		<< 3.5 >>
31 - 40	DMIN3:	The distances ranging DMIN3≦r≦DMAX3 will be	<< 0.5 >>

used for distance calculations.

41 - 50 DMAX3: << 4.0 >>

(b) Elimination of the calculation for some atoms (26I3)

Present only when IOMT≧1 in (a).

JOMT(I): No bond calculations for the JOMT(I)th atom.
There are IOMT variables.

(c) Standard deviations of cell parameters (6F10.6)

1 - 10 SIGA: σa in angstrom.
11 - 20 SIGB: σb in angstrom.
21 - 30 SIGC: σc in angstrom.
31 - 40 SIGCA: σcosα.
41 - 50 SIGCB: σcosβ.
51 - 60 SIGCG: σcosγ.

(d) Symmetry operations (9F5.2,3F9.6)

Present only when NSS≠0 in (a).
The format should be the same to those given in {12}.
NSS sets will be required.

(7) Replaceable Subroutines

The following subroutines should be supplied in order to perform special and/or convenient calculations in most cases.

(7-1) Description

1) Subroutine RESET

This subroutine is used in the cases that there are parameters to be constrained. It is called once near the end of each cycle of least-squares calculation when IRESET = 1 in {6}. The variable name for each parameter is shown in the chapter (6).

2) Subroutine PATCH

This subroutine must be always supplied if the RESET subroutine is used. Using this subroutine, the user must give the derivatives for the parameters constrained in the RESET subroutine.

3) Subroutine ACCABS(FH,FK,FL,F,SIGF,FC,NREJ)

This subroutine is called following the calculation of structure factor for each reflection when IRESET = 2 in {6}. This may be used to modify, reject or pick-up the calculated strucure factors. Set the argument NREJ = 1 for rejection; otherwise set NREJ = 0.

4) Subroutine CONV(WRL,NCON,WR)

This subroutine is called at the completion of every structure factor calculation following a least-squares cycle when ICONV = 1 in {6}. The argument WRL is the weighted R factor prior to the refinement cycle, while WR is the weighted R factor following the cycle. This subroutine must return with NCON = 1, 2 or 3:

NCON = 1: the refinement continues in a normal way.

= 2: the RADY program will assume that the refinement has converged, and it will terminate after needed outputs.

= 3: the RADY program will assume that the refinement has diverged, and it will terminate without final outputs.

5) Subroutine EPATCH(EFP)

This subroutine is used to set constraints on the refinement of the extinction parameters. It is called only if the extinction correction is anisotropic.

6) Subroutine NSHIFT(PNEW,POLD,DELTA,I,K)

This subroutine is used to change PNEW in the convergence, where POLD is the value of the parameter prior to the refinement cycle, DELTA is the shift of the parameter and PNEW = POLD + DELTA. It is entered after the least-squares refinement as each parameter is being corrected with the sfift.

7) Subroutine PATCHS(S)

This subroutine is used when parameters are constrained. There is an entry in S for each parameter, not for each variable.

8) Subroutine SIGFO(FH,FK,FL,F,SIGF,SQ,TBAR,NREJ)

This subroutine is called for each reflection at the initial stage when IW ≧ 3 in {6}. To define the weight with SIGF, see IW in {6} and WMIN in {10}. Set NREJ = 1 in the case that the reflection should be completely omitted from the calculation. Set SIGF = 0, if unobserved reflections with F = 0 have been included.

(7-2) Lists of Replaceable Subroutines

```
      SUBROUTINE RESET
      COMMON/P1/ ATOM(99),SF(99),ATK(99),ATL(99),XYZ(3,99),BETA(6,99)
      COMMON/S7/ DFDEC(7),EC(7),EXTINC,IEXT,EFPIP(25)
      COMMON/H1/ CCC(10,99),DDD(15,99),EEE(21,99),FFF(28,99)
      WRITE(6,100)
  100 FORMAT(//1H ,15X,'+++ANHRM++ IN RESET ++++++++++')
      RETURN
      END
C --
      SUBROUTINE PATCH
      COMMON/P2/ DADAK(99),DBDAK(99),DADAL(99),DBDAL(99),DADX(3,99)
      COMMON/P3/ DBDX(3,99),DADB(6,99),DBDB(6,99),DFDAK(99),DFDAL(99)
      COMMON/P4/ DFDX(3,99),DFDB(6,99),JTF(99),SL(99)
      COMMON/H3/ DADC(10,99),DADD(15,99),DADE(21,99),DADF(28,99)
      COMMON/H4/ DBDC(10,99),DBDD(15,99),DBDE(21,99),DBDF(28,99)
      COMMON/H5/ DFDC(10,99),DFDD(15,99),DFDE(21,99),DFDF(28,99)
      RETURN
      END
C --
      SUBROUTINE ACCABS(H1,H2,H3,F,SIG,FC,NREJ)
      DATA IFIRST/0/
      NREJ=0
      IF(IFIRST.EQ.0) PRINT 100
      IFIRST=1
  100 FORMAT(//1H ,15X,'+++ANHRM++ IN ACCABS ++++++++++')
      RETURN
      END
C --
      SUBROUTINE CONV(WRL,NCONV,WR)
      GOO=WR/WRL
      IF(GOO-1.10)1,1,5
    1 IF(GOO-0.999)3,2,4
    4 IF(GOO-1.001)2,2,3
    2 NCONV=2
      RETURN
    3 NCONV=1
      RETURN
    5 NCONV=3
      RETURN
      END
C --
      SUBROUTINE EPATCH(EFPIP)
      DIMENSION EFPIP(25)
      DATA IFIRST/0/
      IF(IFIRST.EQ.0) WRITE(6,100)
  100 FORMAT(//1H ,15X,'+++ANHRM++ IN EPATCH ++++++++++')
      IFIRST=1
      RETURN
      END
C --
      SUBROUTINE NSHIFT(PNEW,POLD,DELTA,I,K)
      COMMON/FANCY/ IFFLAG,SMOOM,SPLAT,SPLASH,IEFLAG
      COMMON/FANCY2/ BLANKS
      COMMON/EXTIN/ ITYPEX,IDISTR,IMOSC,JEXT
      DATA IFIRST/0/
      RETURN
      END
```

```
C --
      SUBROUTINE PATCHS(S)
      DIMENSION S(841)
      RETURN
      END
C --
      SUBROUTINE SIGFO(FH,FK,FL,F,SIGF,SQ,TBAR,NREJ)
      COMMON/Q9/ SINL2,SINT,SIN2T,DTHE
      NREJ=0
      RETURN
      END
```

(8) JOB Example

(8-1) BATCH JOB Control for Al_2O_3

```
//XXXXXXX JOB USER=XXXXX,PASSWORD=YYYYY,REGION=4096K,CLASS=C,
//            TIME=(120,10)
//JOBPROC DD DSN=XXXXX.PROCLIB.CNTL
//  EXEC RADY,SUB='XXXXX.RADY.FORT77(SUB)'
//GO.FT06F001 DD SYSOUT=A
//GO.FT14F001 DD DSN=&&WORK1,UNIT=SYSDA,SPACE=(100,(1000))
//GO.FT15F001 DD DSN=&&WORK2,UNIT=SYSDA,SPACE=(100,(1000)),
//            DCB=(RECFM=FB,LRECL=80,BLKSIZE=3200)
//GO.FT20F001 DD DSN=XXXXX.BL10A.DATA(AL203),DISP=SHR
//GO.FT23F001 DD DSN=XXXXX.ATOMP.DATA(AL203),DISP=SHR
//GO.FT24F001 DD DSN=XXXXX.RADY.DATA(ASFT),DISP=SHR
//GO.FT26F001 DD DSN=XXXXX.WORKFOFC.DATA,DISP=SHR
//GO.FT28F001 DD DSN=XXXXX.WORKATMP.DATA,DISP=SHR
//GO.FT05F001 DD *
         --------------------
         ---  INPUT DATA  ---
         --------------------
/*
//
```

(8-2) Contents of PROCLIB.CNTL

```
//RADY PROC
//FORT EXEC PGM=JZK@FORT,PARM='OPTIMIZE(0)',REGION=4096K,COND=(4,LT)
//SYSIN DD DSN=&SUB,DISP=SHR
//SYSLIN DD DSN=&&LOADSET,UNIT=SYSSQ,SPACE=(TRK,(10,5)),
//          DISP=(MOD,PASS),DCB=BLKSIZE=3120
//SYSPRINT DD SYSOUT=*
//LKED EXEC PGM=LINKEDIT,PARM='LET,LIST,MAP',REGION=4096K,COND=(4,LT)
//SYSUT1 DD UNIT=SYSDA,SPACE=(TRK,(10,10))
//SYSLIB DD DSN=SYS1.FORTLIB,DISP=SHR
//SYSLMOD DD DSN=&&GOSET(MAIN),UNIT=SYSDA,DISP=(,PASS),
//           SPACE=(TRK,(10,10,1),RLSE)
//SYSLIN DD DSN=XXXXX.#RADY.OBJ,DISP=SHR
//       DD DSN=&&LOADSET,DISP=(OLD,PASS)
//SYSPRINT DD SYSOUT=*
//GO EXEC PGM=*.LKED.SYSLMOD,REGION=4096K,COND=(4,LT)
//*
```

(8-3) Input Data for Al_2O_3

```
AL2O3 CORUNDUM
 20 24 23 26  0 28  0  0  0  0  0  0
  2  6  1  2  0  2  1  0  0  1  0  0  0  0  0  0  0  0
  2  0  4  1  1  4  1  2  2
114  7
  7  0  1  1  1  1  0  0  0  0  0  0  0  1  1  1
  1  1  1  0
  0  0  0
 0.00       1.5         0.1        9000.    2.0
 6.        13.357     12.185     0.0
0.056      0.008
0.052      0.006
  1.0                    1.0                    1.0
      -1.0         1.0 -1.0                    1.0
 -1.0  1.0        -1.0                         1.0
      -1.0        -1.0                         1.0                    0.5
  1.0              1.0 -1.0                    1.0                    0.5
 -1.0  1.0             1.0                    1.0                    0.5
4.8400     4.8400     13.241     90.0       90.0       120.0      0.70926
0.223163       0.093287       0.025816       -.009533       -.208956
-.011057       0.083770       0.067395       -.070156
1.49       1.49       1.49       1.48       1.48       1.48       1.48
1.48       1.48       1.47       1.46       1.46       1.46       1.46
1.45       1.45       1.45       1.45
(3F3.0,2F9.3,F5.0)
44
1
0
  00110100011001000011000011001000 01
000
  10011100111001001001100110011011 011
000
1
 32  0  0  1
 32  0  0  1
 32  0  0  1
 32  0  0  1
 32  0  0  1
 32  0  0  1
 32  0  0  1
1
0
  000
000
  000
0
```

(8-4) Initial Atomic Parameters for Al_2O_3

```
 0.099693
 0.0
AL   1 0 1 0.33333 0.00001 0.0       0.0       0.353394
 0.027160 0.027160 0.002556 0.013580 0.0       0.0
-0.86414E-06-0.29738E-05-0.21097E-05 0.86414E-06-0.36819E-06-0.18410E-06
-0.36819E-06 0.0          0.0          0.11293E-06
 0.13635E-06 0.68174E-07 0.68174E-07 0.68174E-07 0.13635E-06 0.62282E-07
 0.10254E-06 0.40255E-07-0.62282E-07-0.27701E-07-0.13850E-07-0.27701E-07
 0.0          0.0         -0.72589E-08
 0.0          0.0          0.0          0.0          0.0          0.0
 0.0          0.0          0.0          0.0          0.0          0.0
 0.0          0.0          0.0          0.0          0.0          0.0
 0.0          0.0          0.0
 0.0          0.0          0.0          0.0          0.0          0.0
 0.0          0.0          0.0          0.0          0.0          0.0
 0.0          0.0          0.0          0.0          0.0          0.0
 0.0          0.0          0.0          0.0          0.0          0.0
 0.0          0.0          0.0          0.0
 0   2 0 1 0.50000 0.00001 0.306941 0.0       0.250000
 0.026418 0.026772 0.002721 0.013386 0.000530 0.001061
-0.21172E-05-0.51854E-05-0.51854E-05 0.0          0.51702E-05 0.51702E-05
 0.0          0.11760E-05 0.0          0.0
 0.95194E-07-0.56896E-06-0.33983E-06 0.11842E-06 0.23685E-06 0.26843E-06
 0.10059E-06-0.23510E-06-0.47020E-06-0.16686E-08-0.54638E-07-0.10928E-06
-0.18981E-07-0.37961E-07-0.31993E-08
 0.0          0.0          0.0          0.0          0.0          0.0
 0.0          0.0          0.0          0.0          0.0          0.0
 0.0          0.0          0.0          0.0          0.0          0.0
 0.0          0.0          0.0
 0.0          0.0          0.0          0.0          0.0          0.0
 0.0          0.0          0.0          0.0          0.0          0.0
 0.0          0.0          0.0          0.0          0.0          0.0
 0.0          0.0          0.0          0.0          0.0          0.0
 0.0          0.0          0.0          0.0
 0.280E+00
```

(8-5) Replaceable Subroutines for Al_2O_3

```
      SUBROUTINE RESET
      COMMON/P1/ ATOM(99),SF(99),ATK(99),ATL(99),XYZ(3,99),BETA(6,99)
      COMMON/S7/ DFDEC(7),EC(7),EXTINC,IEXT,EFPIP(25)
      COMMON/H1/ CCC(10,99),DDD(15,99),EEE(21,99),FFF(28,99)
      WRITE(6,100)
  100 FORMAT(//1H ,15X,'+++ANHRM++ IN RESET ++++++++++'/)
      BETA(2,1)=BETA(1,1)
      BETA(4,1)=0.5*BETA(1,1)
      BETA(5,1)=0.0
      BETA(6,1)=0.0
      BETA(4,2)=0.5*BETA(2,2)
      BETA(5,2)=0.5*BETA(6,2)
C * * * *       ALUMINUM
      CCC(4,1)=-CCC(1,1)
      CCC(2,1)=0.5*CCC(1,1)
      CCC(3,1)=-0.5*CCC(1,1)
      CCC(8,1)=0.0
      CCC(9,1)=0.0
      CCC(7,1)=CCC(5,1)
      CCC(6,1)=CCC(5,1)*0.5
      DDD(5,1)=DDD(1,1)
      DDD(2,1)=DDD(1,1)*0.5
      DDD(4,1)=DDD(1,1)*0.5
      DDD(9,1)=-DDD(6,1)
      DDD(3,1)=DDD(1,1)*0.5
      DDD(12,1)=DDD(10,1)
      DDD(7,1)=0.5*DDD(6,1)
      DDD(8,1)=-0.5*DDD(6,1)
      DDD(11,1)=DDD(10,1)*0.5
      DDD(13,1)=0.0
      DDD(14,1)=0.0
      EEE(2,1)=-EEE(1,1)
      EEE(6,1)=-0.4*EEE(1,1)+EEE(4,1)
      EEE(7,1)=0.5*EEE(5,1)
      EEE(9,1)=-0.6*EEE(1,1)+EEE(4,1)
      EEE(10,1)=0.5*EEE(5,1)
      EEE(13,1)=-EEE(1,1)+EEE(4,1)
      EEE(14,1)=0.5*EEE(5,1)
      EEE(15,1)=-EEE(8,1)+EEE(11,1)
      EEE(16,1)=0.5*EEE(12,1)
      EEE(17,1)=0.0
      EEE(18,1)=EEE(5,1)
      EEE(19,1)=-EEE(8,1)
      EEE(20,1)=EEE(12,1)
      EEE(21,1)=0.0
C * * * *      OXYGEN
      CCC(4,2)=0.0
      CCC(10,2)=0.0
      CCC(7,2)=0.0
      CCC(9,2)=0.0
      CCC(3,2)=CCC(2,2)
      CCC(6,2)=CCC(5,2)
      DDD(4,2)=0.5*DDD(5,2)
      DDD(13,2)=0.5*DDD(14,2)
      DDD(3,2)=DDD(5,2)/6.0 + 0.6666667*DDD(2,2)
      DDD(7,2)=DDD(9,2)/6.0 + 0.6666667*DDD(6,2)
      DDD(8,2)=0.5*DDD(9,2)
```

```
      DDD(11,2)=0.5*DDD(12,2)
      RETURN
      END
C --
      SUBROUTINE PATCH
      COMMON/P2/ DADAK(99),DBDAK(99),DADAL(99),DBDAL(99),DADX(3,99)
      COMMON/P3/ DBDX(3,99),DADB(6,99),DBDB(6,99),DFDAK(99),DFDAL(99)
      COMMON/P4/ DFDX(3,99),DFDB(6,99),JTF(99),SL(99)
      COMMON/H3/ DADC(10,99),DADD(15,99),DADE(21,99),DADF(28,99)
      COMMON/H4/ DBDC(10,99),DBDD(15,99),DBDE(21,99),DBDF(28,99)
      COMMON/H5/ DFDC(10,99),DFDD(15,99),DFDE(21,99),DFDF(28,99)
      DFDB(1,1)=DFDB(1,1)+DFDB(2,1)+0.5*DFDB(4,1)
      DFDB(6,2)=0.5*DFDB(5,2)+DFDB(6,2)
      DFDB(2,2)=DFDB(2,2)+0.5*DFDB(4,2)
C * * * * ALUMINUM
      DFDC(1,1)=DFDC(1,1)-DFDC(3,1)-DFDC(4,1)
      DFDC(2,1)=DFDC(2,1)+DFDC(3,1)
      DFDC(5,1)=DFDC(5,1)+DFDC(7,1)+0.5*DFDC(6,1)
      DFDD(1,1)=DFDD(1,1)+DFDD(5,1)+0.5*DFDD(2,1)+0.5*DFDD(4,1)+
     $          0.5*DFDD(3,1)
      DFDD(6,1)=DFDD(6,1)-DFDD(9,1)-DFDD(8,1)
      DFDD(10,1)=DFDD(10,1)+DFDD(12,1)+0.5*DFDD(11,1)
      DFDD(7,1)=DFDD(7,1)+DFDD(8,1)
C * * * *  OXYGEN
      DFDC(2,2)=DFDC(2,2)+DFDC(3,2)
      DFDC(5,2)=DFDC(5,2)+DFDC(6,2)
      DFDD(5,2)=DFDD(5,2)+0.5*DFDD(4,2)+0.1666667*DFDD(3,2)
      DFDD(2,2)=DFDD(2,2)+0.6666667*DFDD(3,2)
      DFDD(6,2)=DFDD(6,2)+0.6666667*DFDD(7,2)
      DFDD(14,2)=DFDD(14,2)+0.5*DFDD(13,2)
      DFDD(9,2)=DFDD(9,2)+0.5*DFDD(8,2)+0.1666667*DFDD(7,2)
      DFDD(12,2)=DFDD(12,2)+0.5*DFDD(11,2)
      RETURN
      END
C --
      SUBROUTINE CONV(WRL,NCONV,WR)
      GOO=WR/WRL
      IF(GOO-1.10)1,1,5
    1 IF(GOO-0.999)3,2,4
    4 IF(GOO-1.001)2,2,3
    2 NCONV=2
      RETURN
    3 NCONV=1
      RETURN
    5 NCONV=3
      RETURN
      END
C --
      SUBROUTINE SIGFO(FH,FK,FL,F,SIGF,SQ,TBAR,NREJ)
      COMMON/Q9/ SINL2,SINT,SIN2T,DTHE
      NREJ=0
      A=-FH+FK+FL
      B=AMOD(A,3.0)                    -h+k+l = 3n → ok
      IF(B.NE.0.0) GO TO 50
      NREJ=0
      GO TO 90
   50 NREJ=1
   90 RETURN
      END
```

(B) Contents of Atomic Scattering Factors' Tables

Atomic scattering factors for each atom (or ion) are taken from the reported tables[1)-5)] or calculated using the program ASF (Sasaki, unpublished). Most of data are interpolated to obtain 150 values per atom from $\sin\theta/\lambda$ = 0 to 1.49 ($\mathring{A}^{-1}$) at intervals of 0.01. Each atomic set in the ASFT file consists of one-line data for labelling and reference number and 15-lines data of the factors.

1) International Tables for X-ray Crystallography (1962) vol. III, Birmingham: kynoch Press.

2) International Tables for X-ray Crystallography (1974) vol. IV, Birmingham: kynoch Press.

3) Fukamachi (1971) Tech. Rep. B12, Inst. of Solid State Physics, Univ. Tokyo.

4) Tokonami (1965) Acta Crystallogr., 19, 486.

5) Sasaki, Fujino and Takéuchi (1979) Proc. Japan Acad., 55, 43-48.

The followings are the contents of the ASFT file including the reference number for atoms:

```
1     <H(O) *VOL.4*    HF>                                          HANDC  (  1)    1
        1.000   0.998   0.991   0.980   0.966   0.947   0.925   0.900   0.872   0.842
        0.811   0.778   0.744   0.710   0.676   0.641   0.608   0.574   0.542   0.511
        0.481   0.452   0.424   0.398   0.373   0.350   0.328   0.307   0.287   0.268
        0.251   0.235   0.220   0.206   0.193   0.180   0.169   0.158   0.148   0.139
        0.130   0.122   0.115   0.108   0.101   0.095   0.090   0.084   0.079   0.075
        0.071   0.067   0.063   0.059   0.056   0.053   0.050   0.047   0.044   0.042
        0.040   0.038   0.036   0.034   0.032   0.031   0.029   0.028   0.026   0.025
        0.024   0.023   0.022   0.021   0.020   0.019   0.018   0.017   0.017   0.016
        0.015   0.014   0.014   0.013   0.013   0.012   0.012   0.011   0.011   0.010
        0.010   0.010   0.009   0.009   0.009   0.008   0.008   0.008   0.007   0.007
        0.007   0.007   0.007   0.006   0.006   0.006   0.006   0.005   0.005   0.005
        0.005   0.005   0.004   0.004   0.004   0.004   0.003   0.003   0.003   0.003
        0.003   0.003   0.003   0.003   0.003   0.003   0.003   0.003   0.003   0.003
        0.003   0.003   0.003   0.003   0.003   0.003   0.002   0.002   0.002   0.002
        0.002   0.002   0.002   0.002   0.002   0.001   0.001   0.001   0.001   0.001

2     <H(BOND) *STEWART* >                                         HANDC  (  2)    2
        1.000   0.997   0.992   0.985   0.975   0.961   0.944   0.924   0.902   0.878
        0.853   0.827   0.801   0.773   0.744   0.713   0.682   0.651   0.622   0.594
        0.567   0.541   0.516   0.491   0.467   0.443   0.419   0.396   0.373   0.351
        0.330   0.310   0.291   0.274   0.258   0.244   0.231   0.218   0.206   0.195
        0.184   0.174   0.164   0.155   0.146   0.137   0.129   0.121   0.114   0.108
        0.102   0.098   0.092   0.087   0.082   0.078   0.074   0.070   0.066   0.062
        0.059   0.056   0.053   0.051   0.048   0.046   0.044   0.042   0.040   0.038
        0.036   0.034   0.033   0.031   0.030   0.028   0.027   0.026   0.024   0.024
        0.022   0.021   0.020   0.019   0.018   0.017   0.017   0.016   0.015   0.015
        0.014   0.013   0.013   0.012   0.012   0.011   0.011   0.010   0.010   0.010
        0.009   0.009   0.008   0.008   0.008   0.007   0.007   0.007   0.006   0.006
        0.006   0.006   0.006   0.005   0.005   0.005   0.005   0.005   0.045   0.004
```

```
5071                GOO=WR/WRL                                                    RADY5071
5072                IF(GOO-1.10) 1,1,5                                            RADY5072
5073              1 IF(GOO-0.999) 3,2,4                                           RADY5073
5074              4 IF(GOO-1.001) 2,2,3                                           RADY5074
5075              2 NCONV=2                                                       RADY5075
5076                RETURN                                                        RADY5076
5077              3 NCONV=1                                                       RADY5077
5078                RETURN                                                        RADY5078
5079              5 NCONV=3                                                       RADY5079
5080                RETURN                                                        RADY5080
5081                END                                                           RADY5081
5082          C --                                                                RADY5082
5083                SUBROUTINE EPATCH(EFPIP)                                      RADY5083
5084                DIMENSION EFPIP(25)                                           RADY5084
5085                DATA IFIRST/0/                                                RADY5085
5086                IF(IFIRST.EQ.0) PRINT 100                                     RADY5086
5087            100 FORMAT(//1H ,15X,'++++ RADY ++++   IN EPATCH SUBROUTINE'/)    RADY5087
5088                IFIRST=1                                                      RADY5088
5089                RETURN                                                        RADY5089
5090                END                                                           RADY5090
5091          C --                                                                RADY5091
5092                SUBROUTINE NSHIFT(PNEW,POLD,DELTA,I,K)                        RADY5092
5093                COMMON/FANCY/ IFFLAG,SMOOM,SPLAT,SPLASH,IEFLAG                RADY5093
5094                COMMON/FANCY2/ BLANKS                                         RADY5094
5095                COMMON/EXTIN/ ITYPEX,IDISTR,IMOSC,JEXT                        RADY5095
5096                DATA IFIRST/0/                                                RADY5096
5097                RETURN                                                        RADY5097
5098                END                                                           RADY5098
5099          C --                                                                RADY5099
5100                SUBROUTINE PATCHS(S)                                          RADY5100
5101                DIMENSION S(841)                                              RADY5101
5102                RETURN                                                        RADY5102
5103                END                                                           RADY5103
5104          C --                                                                RADY5104
5105                SUBROUTINE SIGFO(FH,FK,FL,F,SIGF,SQ,TBAR,NREJ)                RADY5105
5106                COMMON/Q9/ SINL2,SINT,SIN2T,DTHE                              RADY5106
5107                NREJ=0                                                        RADY5107
5108                RETURN                                                        RADY5108
5109                END                                                           RADY5109
```