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A COMPUTER PROGRAM FOR THE REDUCTION OF DIFFUSE X-RAY DATA FROM SOLID SOLUTIONS

R. O. Williams

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R. O. Williams

OCTOBER 1972

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ABSTRACT

This report documents a computer program to reduce the diffuse x-ray scattering data from cubic alloys by a multiple regression analysis. The solution provides information on the short-range order and the atomic displacements. The advantages of the present method are that it provides increased freedom in making observations, fewer approximations are required, and a complete error analysis is obtained.

INTRODUCTION

Over the past 25 years it has been realized that metallic solid solutions are rather complex in that the atoms need not be randomly distributed nor sit precisely on the lattice sites. These irregularities produce diffuse diffraction of x-rays, which is quite distinct from the sharp Bragg diffraction. A number of experimental methods are capable of studying such effects, but the most extensive and unambiguous information comes from this diffuse scattering. Although only a few systems have been extensively studied, the results are very interesting in the considerable variety of observed effects. Ultimately such studies should be very important in developing theories of solutions concerning, for example, the heat of mixing, size effects, flow strength, anelastic properties, ordering, and precipitation.

The total coherent diffuse scattering is the sum of three contributions: short-range order, first-order displacements, and second-order displacements (which include thermal diffuse scattering, TDS). The observations are to be restricted such that higher terms can be ignored. Borie and Sparks[1] showed that as a result of the different symmetries of

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[1]B. S. Borie and C. J. Sparks, Jr., *Acta Cryst.* A27: 198–201 (1971).

these contributions the total intensity can be separated into components that depend only on a single effect such that the desired parameters can be obtained from a Fourier analysis. This treatment is a recent development, and only a few systems have been fully analyzed.[2-5]

The intensity is a linear function of parameters that describe each contribution. It follows, therefore, that a solution can also be obtained by a standard multiple regression analysis (also known as least-squares fitting). This report documents the program for carrying out this analysis. A principle advantage of this method over the separation technique[1] is that one obtains estimates of the parameter errors and their correlation.

The method has been applied to a reanalysis of the data on Cu—16 at. % Al by Borie and Sparks,[6] and the results are to be covered elsewhere.[7]

X-RAY THEORY

The intensity, in electron units, of x rays scattered from any material is given by

$$I = \Sigma_u \Sigma_v f_u f_v e^{i\underset{\sim}{k}\cdot(\underset{\sim}{R}_u - \underset{\sim}{R}_v)} , \tag{1}$$

where the atoms at sites u and v have atomic scattering factors of f_u and f_v and a relative position of $\underset{\sim}{R}_u - \underset{\sim}{R}_v$, and $\underset{\sim}{k}$ is defined below. For a cubic lattice we may define a position in reciprocal space by the vector $\underset{\sim}{h}$ as

$$\underset{\sim}{h} = \underset{\sim}{h}_1 + \underset{\sim}{h}_2 + \underset{\sim}{h}_3 , \tag{2}$$

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[2]W. Lin, J. E. Spruiell, and R. O. Williams, *J. Appl. Cryst.* 3: 297—305 (1970).

[3]W. Lin and J. E. Spruiell, *Acta Met.* 19: 451—61 (1971).

[4]J. E. Gragg, Jr., and J. B. Cohen, *Acta Met.* 19: 507—19 (1971).

[5]T. Ericsson and J. B. Cohen, *Acta Cryst.* A27: 97 (1971).

[6]B. Borie and C. J. Sparks, Jr., *Acta Cryst.* 17: 827—35 (1964).

[7]R. O. Williams, "Short-Range Structure in a Copper 16 Atomic Percent Aluminum," to be submitted for publication.

where the components are in the directions of the cube edges. This vector is given by

$$\underset{\sim}{h}_1 + \underset{\sim}{h}_2 + \underset{\sim}{h}_3 = (\underset{\sim}{S} - \underset{\sim}{S}_0)a_0/2\lambda \ , \tag{3}$$

where $\underset{\sim}{S}_0$ and $\underset{\sim}{S}$ are the unit vectors defining the incident and diffracted beam of wave length λ and a_0 is the lattice parameter. Since

$$\underset{\sim}{k} = 2\pi(\underset{\sim}{S} - \underset{\sim}{S}_0)/\lambda \ , \tag{4}$$

it follows that

$$\underset{\sim}{k} = 4\pi(\underset{\sim}{h}_1 + \underset{\sim}{h}_2 + \underset{\sim}{h}_3)/a_0 \ . \tag{5}$$

The relative location of a pair of atoms in an fcc or bcc lattice can be expressed as

$$\underset{\sim}{R}_u - \underset{\sim}{R}_v = \tfrac{1}{2}\left[(l + \delta_l)\underset{\sim}{a}_1 + (m + \delta_m)\underset{\sim}{a}_2 + (n + \delta_n)\underset{\sim}{a}_3\right] , \tag{6}$$

where l, m, and n are integers corresponding to the average lattice, the $\underset{\sim}{a}$'s are the cube edges, and the δ's are the components of the deviation from the average lattice vector.

Since we obtain an overall average from diffraction experiments, the double sum in Eq. (1) may be replaced by a single sum over N average sites. We may further expand the exponential term through the quadratic term in δ, the imaginary parts being zero because of symmetry. For a binary alloy we have

$$\begin{aligned}\frac{I}{Nf_A^{\,2}} = \sum_{i=0}^{\infty} & \left[(A + Bf_B/f_A)^2 + \alpha AB(1 - f_B/f_A)^2\right] \Sigma \cos 2\pi(h_1 l + h_2 m + h_3 n) \\ & + 2\pi AB(f_B/f_A - 1) \sum_{i=1}^{\infty} \gamma \Sigma h_j \sin 2\pi(h_1 l + h_2 m + h_3 n) \\ & - 2\pi^2 AB \sum_{i=1}^{\infty} \varepsilon \sum h_j^{\,2} \cos 2\pi(h_1 l + h_2 m + h_3 n) \\ & - 4\pi^2 AB \sum_{i=1}^{\infty} \rho \sum h_j h_k \cos 2\pi(h_1 l + h_2 m + h_3 n) \ ,\end{aligned} \tag{7}$$

where A and B are the atomic fractions and α is the Warren-Cowley order parameter defined as

$$\alpha_i = 1 - P_i/B \ ,$$

where P_i is the probability of finding a B atom in a particular site in the ith shell around an A atom. Necessarily $\alpha_0 = 1$, and for a random alloy all the α's for $i \neq 0$ are zero. The outer sums are over successive shells (i), while the inner sums are over the cosine or sine terms corresponding to the permutations and combinations of l, m, and n that describe the positions of all the sites in the particular shell. Also

$$\gamma_j = \left(\frac{A}{B} + \alpha_i\right) \delta^j_{AA} - \left(\frac{B}{A} + \alpha_i\right) \frac{f_B}{f_A} \delta^j_{BB} \ , \tag{8}$$

where the δ^j_{AA} is now the average of one of the three δ's defined in Eq. (6) for A-A pairs in the ith shell; similarly for δ^j_{BB}. For each kind of pair there will be three δ's for each shell, but symmetry requires that $\delta_l = 0$ if $l = 0$ and $\delta_l = \delta_m$ if $l = m$. The displacement between unlike pairs is eliminated by the requirement of volume conservation. The particular h_j in the sum is that h in the sine term multiplied by the index l, m, or n that corresponds to the particular δ.

The quadratic term is given as

$$\varepsilon = \left(\frac{A}{B} + \alpha\right)\delta^2_{AA} + 2(1 - \alpha) \frac{f_B}{f_A} \delta^2_{AB} + \left(\frac{B}{A} + \alpha\right) \frac{f^2_B}{f^2_A} \delta^2_{BB} \ , \tag{9}$$

where there will in general be a different ε for each component of a shell and $\varepsilon_l = \varepsilon_m$ only if $l = m$. Note that δ^2 is in reality $\langle\delta^2\rangle$ such that $\varepsilon_l \neq 0$ even when $l = 0$. The ε's cannot be negative since none of the terms on the right side of Eq. (9) can be negative. The h_j in the second sum is identified as in the previous term. One sees that the displacement between unlike pairs must be retained in this expression.

The cross term is given as

$$\rho_{lm} = \left(\frac{A}{B} + \alpha\right) \delta^l_{AA}\delta^m_{AA} + 2(1 - \alpha) \frac{f_B}{f_A} \delta^l_{AB}\delta^m_{AB} + \left(\frac{B}{A} + \alpha\right)\frac{f^2_B}{f^2_A} \delta^l_{BB}\delta^m_{BB} \ . \tag{10}$$

There will in general be three such terms for each shell, but $\rho_{lm} = 0$ if l or $m = 0$. The h_j and h_k in the second sum are identified as those multiplying the index l and m as for the previous terms.

It is of interest to physically identify the terms in Eq. (7). The first term is the relatively uniform Bragg intensity, which is collected by the cosine sum into the sharp peaks. The first member of the alpha series is the monotonic Laue intensity, which is redistributed by the cosine series. Normally the alpha series converges such that this intensity is not sharp. However, for perfect long-range order this intensity is collected into sharp superlattice peaks. The gamma terms are called the first-order size effect. Since thermal motion has a zero time-average displacement, it cannot contribute to these terms. One expects that these linear terms will converge because this is a necessary condition for the existence of an average lattice that gives sharp Bragg peaks.

Both static and dynamic (thermal) displacements contribute to quadratic terms, and these terms become greater with larger shells. Since these terms do not converge, they correspond to a negative delta function at the Bragg peaks and in effect take some of the Bragg intensity and redistribute it in reciprocal space. If the displacements were all independent then all the ε's would have the same value, so the quadratic diffuse intensity would be uniform except for the delta function.

SOLUTION OF THE DIFFRACTION EQUATION

Borie and Sparks[8] showed that the observations can be decomposed into separate functions that depend only on α, γ, ε, and ρ as a result of the particular h dependence of Eq. (7). These parameters are then obtained by a Fourier transform. This method has been used by several investigators[2–5] and represents the most detailed analysis of diffuse scattering that has been made. Alternately, since the intensity is a linear combination of α, γ, ε, and ρ as shown in Eq. (7), then in theory we can apply a multiple regression analysis to fit the observations in terms of these series. The α and γ series are well behaved

[8] B. S. Borie and C. J. Sparks, Jr., *Acta Cryst.* A27: 198–201 (1971).

for short-range order, but unfortunately the ε and ρ series do not converge; hence this method is not strictly applicable. Before a sufficient number of ε and ρ terms can be evaluated to properly represent this component of intensity the solution becomes so ill conditioned that useful values are not obtained. The long-wavelength TDS, which has maxima at the Bragg peaks, is the primary cause of this difficulty. This intensity can be represented as follows.

Long-wavelength TDS is properly treated in terms of an elastic continium and is defined by only three elastic constants for cubic materials. A discrete lattice gives rise to dispersion such that the diffuse intensity decreases less rapidly with decreasing wavelength than predicted by the continium treatment. It also shows the symmetry dictated by the lattice. The entire diffraction problem can be treated in terms of the Born-von Karman model in which the lattice dynamics are derived in terms of near neighbor interactions.[9,10] We want our expression to approach the long-wavelength limit, so we can use only three independent interaction constants. For the fcc lattice we may use the three parameters for the first neighbor. A rigorous application requires the solution of the secular equations for each position in reciprocal space, but, following Houston,[11] we are able to adequately represent the angular dependence by simple geometric expressions.

The relation between the three first neighbor interaction parameters and the elastic constants are given as

$$\alpha_1 = aC_{11}/4 \ , \qquad \beta_1 = aC_{44}/2 - aC_{11}/4 \ , \qquad \gamma_1 = aC_{12}/4 + aC_{44}/4 \ . \tag{11}$$

These may be used in Eqs. 11.65—.67 in Warren[9] to describe the three elastic waves in each of the three symmetry directions. The more general

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[9]B. E. Warren, *X-ray Diffraction*, Addison-Wesley, Reading, Massachusetts, 1969.

[10]C. B. Walker, *Phys. Rev.* 103: 547 (1956).

[11]W. V. Houston, *Rev. Mod. Phys.* 20: 161 (1948).

expression is now approximated as

$$D_1 = \frac{\omega_l^2 m}{aC_{11}} = \left[1 + (Y-1)P_L - P_{123}\right] S1 + \left[(1+X+Y)P_L + (1+2X+4Y)P_{123}\right] S2 \,, \quad (12a)$$

$$D_2 = \frac{\omega_{t1}^2 m}{aC_{11}} = Y(1 - P_{123})S1 + \left[(1 - X - Y)P_{t1} + (1 - X + Y)P_{123}\right] S2 \,, \quad (12b)$$

$$D_3 = \frac{\omega_{t2}^2 m}{aC_{11}} = \left[(1 - Y)P_{t2} + (1 - P_{123})Y\right] S1$$

$$+ \left[(2Y - 1)P_{t2} + (1 - X + Y)P_{123}\right] S2 \,, \quad (12c)$$

where

$$X = C_{12}/C_{11}, \quad Y = C_{44}/C_{11} \,; \quad (13)$$

$$P_{t1} = 4 \cos^2 \phi_1 \cos^2 \phi_2 \, (1 - 3 \cos^2 \phi_3) \,,$$

$$P_{t2} = 4 \cos^2 \phi_3 \, (\cos^2 \phi_1 + \cos^2 \phi_2 - 6 \cos^2 \phi_1 \cos^2 \phi_2) \,,$$

$$P_L = P_{t1} + P_{t2} \,, \text{ and} \quad (14)$$

$$P_{123} = 27 \cos^2 \phi_1 \cos^2 \phi_2 \cos^2 \phi_3 \,;$$

with angles defined in Fig. 1, and

$$S1 = 4 \sin^2 \pi g_x \,, \quad S2 = \sin^2 2\pi g_x \,. \quad (15)$$

Our $\underset{\sim}{g}$ vector is in the same units as our $\underset{\sim}{h}$ vector, Eq. (3), g_x being the largest component.

The thermal diffuse intensity is now given by Eq. (11.35) in Warren[9] as

$$\frac{I}{Nf_A^2} = \left(\frac{Af_A + Bf_B}{f_A}\right)^2 C|\underset{\sim}{h}|^2 \sum_{j=1,3} \frac{\cos^2(\underset{\sim}{h}, \underset{\sim}{e}_j)}{D_j} \,, \quad (16)$$

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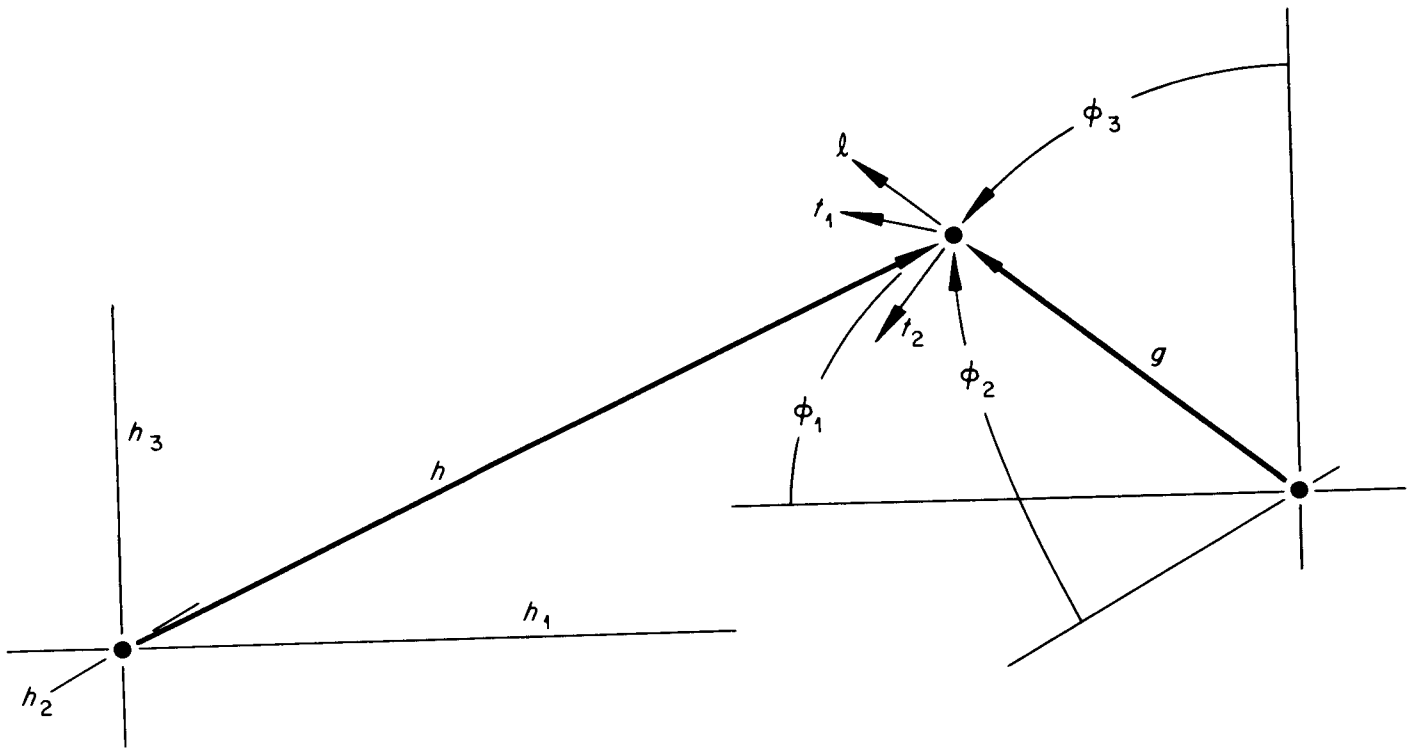

Fig. 1. The Relation Between an Observation at Reciprocal Lattice Point $\underset{\sim}{h}$, Which is in a Brillouin Zone Centered at $\underset{\sim}{g} = 0$. The longitudinal wave is taken to be parallel to $\underset{\sim}{g}$. For the two transverse waves t_1 has zero h_3 displacement and t_2 is tangent to arc ϕ_3 at $\underset{\sim}{g}$.

where $(\underset{\sim}{h},\underset{\sim}{e}_j)$ is the angle between $\underset{\sim}{h}$ and the displacement of the jth wave, which is defined in Fig. 1. The value of C is to be obtained by the regression analysis; from this result,

$$C_{11} = 16\pi^2 \kappa T/a_0^3 C \ , \tag{17}$$

where κ is the Boltzmann constant and T the temperature.

Methods for solving for nonlinear terms such as X and Y are known but are relatively more involved than evaluating linear coefficients. The included program uses the simpler approach of providing solutions for various combinations of X and Y based upon initial estimates. Since one must in general resort to several solutions to obtain a more or less optimum combination of terms, there should be ample opportunity to establish satisfactory choices for X and Y.

If this were a complete treatment, Eq. (16) would replace the ε and the ρ series in Eq. (7). Since it is not, one can still include low-order terms of these two series to improve the fit where the terms are now understood to represent that quadratic intensity not represented in our TDS expression. We would also include a term

$$2\pi\left(A + Bf_B/f_A\right)^2 \ |\underset{\sim}{h}|^2 \varepsilon_0 \ , \tag{18}$$

where ε_0 is to be evaluated in the solution. The rationale for this term is that if, for example, all the displacements were random, then we would only have a single term, which increased simply as $|\underset{\sim}{h}|^2$. There would still be a negative delta function at the Bragg peaks, but this region is excluded from our observations.

We would hope that the low-order terms of ε and ρ would compensate for the contribution by the static displacements, for the fact that we used only first neighbor interactions, and possibly in part for the way we approximated the TDS.

We have intentionally left the e^{-2m} term out of Eq. (16). Presumably one restricts the observations such that higher order TDS, which is not included in Eq. (16), is unimportant, in which case e^{-2m} possibly may also be neglected. It should be included if it gives a significantly better

fit and should also be included in the ε_0 term. Note that the value of m is not independently adjustable since it must have a value such that the intensity lost from the Bragg peaks is just that intensity in the TDS and ε_0 term. We would not require an e^{-2m} term in the α series in Eq. (7), in contrast to the expression by Walker and Keating,[12] since the ε and the ρ terms now represent this effect. However, when we replace the ε and ρ series by the TDS expression, such a term might now be in order, again an improved fit being sufficient justification.

STATISTICS

The present solution gives the values of the parameters, an estimate of their errors, and the correlation between the errors. Also a necessary part of the computations is the calculated intensity and the residuals, the differences between the observed intensity and the calculated intensity. Although it is not included in the present program, one can readily calculate the error in the observed intensity, or in any of its components. This information helps to indicate where the observations are in greater error, but the residuals also give essentially the same information and also indicate the probable sign of the errors. The statistics that are used are generally known and may be found in Hamilton,[13] for example. Certain statistical aspects of the present problem require special consideration.

The magnitudes of the parameter errors are proportional to the residuals, but further, the present calculation is based upon the assumption that these residuals are random. Formally, there is no problem if this is not so since then one needs only to include in the weighting matrix the off-diagonal terms that result from the correlation between the errors in the data. For the present problem this is of no help since in the first place very little is known about the correlations, and in the second place a full weighting matrix makes the size of the problem prohibitive. One could undoubtedly reformulate the program to accommodate correlations between

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[12] C. B. Walker and D. T. Keating, *J. Appl. Phys.* 34: 2309 (1963).

[13] W.C.C. Hamilton, *Statistics in Physical Science*, Ronald, New York, 1964.

intensity that are close in reciprocal space, but this does not solve the first problem. A desirable solution would be the identification and reduction of such nonrandom errors. Dropping the weighting factor entirely would not seriously degrade the solution, but its inclusion does permit certain conveniences.

The reader should realize that the parameter errors are to be taken as random, and should the observations be biased, because of improper beam standardization for example, then the parameters will also contain additional biased errors. Gragg et al.[14] have recently considered the overall problem of errors in such measurements.

It is important to realize that there is a basic difference between using a regression analysis and a Fourier analysis on the separated functions. For a given number of terms the regression analysis will give a superior fit, but as the number of terms increases at some place the solution becomes ill conditioned such that the terms in the solution will take on large, unreal magnitudes. This means that the solution is developing considerable small-scale structure to better fit the observations. Thus ultimately the solution must become unsatisfactory. We have shown elsewhere[15] how to include very powerful supplementary conditions that restrict such behavior and permit the solution to be carried to almost any length. A solution using a Fourier analysis cannot exhibit such improper behavior and in that sense is superior. In a very real sense the use of supplementary conditions accomplishes the same things that one may do in a Fourier analysis, such as smoothing and filling in data at the Bragg peaks. Such supplementary conditions degrade the degree of fit only very little and hence do not introduce any particular uncertainty in the estimation of the errors. They do, however, greatly reduce the magnitude of the error correlation, and thus constitute a very useful device. As a general rule one would include sufficient terms in each series until the coefficients have magnitudes similar to their errors.

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[14] J. E. Gragg, Jr., M. Hayakawa, and J. B. Cohen, ACA Winter Meeting, Albuquerque, N.M., February 1972.

[15] R. O. Williams, "Short-Range Structure in a Copper 16 Atomic Percent Aluminum," to be submitted for publication.

A detailed examination of the residuals can prove to be of considerable value. Any systematic deviation between the observed and calculated intensities over the field of measurement must be due to limitations in the data, in the expression being fit to the data, or both. Obviously the elimination of such problems can materially improve the final results. Further, the residuals should show no significant correlation with any variable; such correlations provide a clue as to the nature of the difficulty. The documented program provides information about the behavior of the residuals as functions of 2θ.

DESCRIPTION OF THE PROGRAM

Because of the difficulties involved no attempt has been made to write a general program that would accept data in any format. Rather, the listed program was written to handle a specific set of observations, and this section provides the documentation such that the user can make such changes as required. For problems of this complexity it is important that the user have a reasonable understanding of the program; else the chances of major errors are great. Having gained this understanding one can readily make the required modifications.

The listed program, given in Appendix A, was designed to evaluate the observations[16] on a Cu–16 at. % Al crystal at 77°K using monochromatic copper radiation. It accepts 4032 observations and computes 23 values of α, 17 values of γ, 2 values of ε, and one each of ρ, ε_0, and C, as defined by Eqs. (7) and (16). Each solution is repeated nine times, using the combinations of three different values of X and Y. The observations consisted of the following sets:

Data Set	Region	Range, deg.*	
		2θ	ϕ
A	$H_1 - H_2 - H_3 = 0$, $\phi = 0$ at $[2\bar{1}\bar{1}]$	20–136	0–30
B	$H_1 - H_2 - H_3 = 0$, $\phi = 0$ at $[2\bar{1}\bar{1}]$	50–136	1–31
C	$H_3 = 0$, $\phi = 0$ at [100]	20–136	0–44†
D	$H_3 = 0$, $\phi = 0$ at [100]	50–136	1–43

*Increments of 2θ and ϕ are 2°.
†Last trace at 45°.

[16] B. Borie and C. J. Sparks, Jr., *Acta Cryst.* 17: 827–35 (1964).

We speak of the values of α, γ, etc. collectively as the solution and the terms that they multiply in Eqs. (7) and (16) as the coefficients, which consist of two parts, the presum part and the geometric part.

The program consists of three main parts: DIFFUSE, which handles all the input and part of the output and which calls the subroutines; SERIES, which calculates the coefficients; and STAT, which carries out the regression analysis. STAT uses a subroutine MATQ for a matrix inversion, which is not included since the user can supply an equivalent subroutine. Programming is considerably simplified by the use of six function-type subprograms that calculate the sines and cosines as required in the geometric coefficients. A final function IRND(X) rounds X to the closest integer. In this section FORTRAN statements, variables, and programs are capitalized.

Table 1 identifies the presum parts of the coefficients and gives the variable names used in the program. Two names may be used in DIFFUSE; the dimensioned variable contains all required values, which are calculated in loop 6. The second name is one that occurs in COMMON/C/, which provides access by SERIES. The final three variables locate the position of the observation.

Table 1. Variables Transferred Between Programs by COMMON/C/

Variable	DIFFUSE	SERIES
$2AB\left(1 - f_B/f_A\right)^2$	FB, APL	E
$AB\left(1 - f_B/f_A\right)^2$	FBO, APO	APO
$4\pi AB\left(f_B/f_A - 1\right)$	FBA, GAM	F
$-4\pi^2 AB$	EP	G
$-8\pi^2 AB$	RHO	H
$\left(\frac{Af_A + Bf_B}{f_A}\right)^2 \lvert \underset{\sim}{h} \rvert^2$	TDS, TD	TD
$2\pi\left(\frac{Af_A + Bf_B}{f_A}\right)^2 \lvert \underset{\sim}{h} \rvert^2$	FQ, EPO	EPO
h_1, h_2, h_3	H_1, H_2, H_3	X, Y, Z

Labeled common blocks are used to provide access to the different variables in the different parts of the program. Extensive use is made of the facility for breaking up a common block differently in the different parts of the program. This simplifies the program, but the user must understand how this works when making changes and may require professional help. The common blocks are listed in Table 2 along with the names of the variables they contain in different parts of the program. As a specific example COMMON/A/ contains the observed intensity as the four data sets listed in DIFFUSE, while in STAT these values are treated as a singly dimensioned variable A.

We now proceed through the program, indicating what is being done and identifying the variables.

FAL and FCU are the atomic scattering factors of aluminum and copper in increments of 0.05 sin θ/λ starting with zero and are provided by the DATA statements. ALAM is the wavelength of the radiation; AO is the lattice parameter. The three values of X, Eq. (12) that are used are X12, X12+DX12, and X12+2DX12; similarily for Y, Y44 and DY44. XAB is the product of the compositions, and BG will be used in the weighting function. Variables LL, MM, and NN are the integers that locate the atoms in the shells. They are of the form $l00$, $ll0$, lll, $lm0$, lmm, and lmn and are indicated by values of 1 through 6, respectively, in INDEX. These values are multiplied by 2π in loop 39.

The next four loops read in the data. In loop 6 we calculate those parts of the coefficients that are a function of 2θ and ϕ. WR is proportional to the area associated with each observation and will become part of the weighting function. The factor 0.002 simply controls the size of the numbers generated during the matrix inversion, thus reducing the possibility of overflows, but otherwise does not affect the solution. The term 59/(I + 43) drops from 1 to 0 when I exceeds 16 to compensate for the way in which the observations interlace. Loop 20 sets WA, WB, WC, and WD to zero.

The next four loops proceed to calculate the reciprocal lattice coordinates for each observation, H1, H2, and H3, assign the proper values to the variables in COMMON/C/, and then call SERIES. These loops are

Table 2. Contents of Labeled Common Blocks

Block	Variable	Name of Variable in		
		DIFFUSE	SERIES	STAT
A	I/Nf_A^2 observed	A, B, C, D		A
B	I/Nf_A^2 calculated	AI, BI, CI, DI		B
F	Weighting factor	WA, WB, WC, WD		W
D	Coefficients for			
	α_0, α		AD, D	D
	γ		B	
	ε_0, ε, ρ		C	
	TDS		A	
G	α, γ, ε, ρ, C	AX		AX

so arranged that the observations are considered sequencially as stored in COMMON/A/. The weighting function WA etc. is taken to be proportional to WR and inversely proportional to the observed intensity plus BG.

Certain observations will be missing, in particular those too close to the Bragg peaks, and such missing values are excluded from the analysis by the simple expediency of setting the weighting factor to zero. However, values for the calculated intensity will be obtained for all points.

Before considering in detail how the coefficients are calculated in SERIES, we need to consider how many terms of each kind are required for each shell. Obviously only one α applies, but for the other variables there may be more than one, as shown in the following:

Type of Shell	γ	ε	ρ
$l00$	l	$l,0$	
$ll0$	l	$l,0$	ll
lll	l	l	ll
$lm0$	l,m	$l,m,0$	lm
lmm	l,m	l,m	lm,mm
lmn	l,m,n	l,m,n	lm,ln,mn

Thus there is one linear term for each different nonzero component of the shell, a quadratic term for each different component, and a cross term for each distinct pair having nonzero components. Multiple terms for a specific shell are calculated in the order given in the table. The names of the variables that will contain the coefficients are listed in Table 2. Loop 7 calculates the complete set of coefficients for the first 11 shells. The quadratic and cross terms are temporarily stored as variable C because more are evaluated than will be used. Loop 42 transfers the required ones into CD. Loop 27 calculates the coefficients for the α terms for shells 12 through 22. Variables D, B, and CD must be properly dimensioned such that each position is filled with a proper coefficient. It is permissible to calculate more linear terms in loop 7 than required to fill B since the extra ones, which overflow into CD, are subsequently written over with the correct coefficient. The functions SL, SLM, and SLMN calculate a sine and cosine term as required in the inner sums, the sine corresponding to S1, the cosine as C1, for example. We make no distinction between the ε and ρ terms apart from position, but the ρ terms may be identified as having factors H and two reciprocal lattice components. For loop 27 we only require the cosine terms which are calculated by functions R, S, or T. Before any of these six functions are called the components of the shell (times 2π) are loaded into unlabeled COMMON, which is then accessed in calculating the sines and cosines.

Immediately following statement 27, XX, YY, and ZZ are the components for the closest Brillouin zone having even indices; X2, Y2, and Z2, the closest one with odd indices. The observation belongs to that zone having the smallest $\underset{\sim}{g}$ vector; hence the comparison of G1 and G2. COS1, COS2, and COS3 are equal to $|\underset{\sim}{h}|^2 \cos(\underset{\sim}{h}, \underset{\sim}{e}_j)$ for the three different waves as given in Eq. (16). P1, P2, P3, PT1, PT2, and PL have the same meaning as defined in Eq. (14). Loop 41 loads into A the nine different coefficients corresponding to the combinations of X and Y.

Equation (7) applies equally well to either a bcc or fcc structure; one simply has to load the values of LL, MM, and NN that define the positions for the desired structure. However, the TDS formulation is specific to the fcc lattice and would require modification for a bcc lattice.

Once loop 9 is completed in DIFFUSE, all the coefficients have been set up and STAT is called. The dimensions of IX, AS, X, AJ, and AX must be at least NUM, the number of unknowns. Loop 15 forms what is known as the coefficients of the normal equations, AAA. Loop 50 is executed nine times for the different combinations of X and Y, but a complete solution is obtained only for the fifth pass. Loop 62 modifies AAA for subsequent solution. Loop 61 loads AAA into AA. Loops 2, 3, and 4 calculate the correlation between the various terms in the observations. XX must be made an identity matrix; then MATQ inverts AA leaving the inverse in XX. DET is the value of the determinant. Loops 20 and 17 calculate the correlation between the errors. The solution is in AX, the calculated intensities in B, and the errors in AS. Variable ER1 sums the residuals for each value of 2θ. Loop 31 calculates the various components of intensity: specifically A1 is due to the alpha series, A2 the linear terms, A3 the quadratic and cross terms, and A4 the TDS term for selected traces.

On returning to DIFFUSE, a complete printout of the observed intensity, the calculated intensity, and the difference is executed. Roughly 2 min of IBM 360/91 time is required for a solution of 44 values from 4032 observations.

CHANGES IN THE PROGRAM

Some care has been used in writing this program to make it reasonably efficient in the use of computer time. Alternate methods of handling the large numbers of sines and cosines could materially reduce the time required to calculate the coefficients, giving perhaps a 10% overall savings. However, a wiser expenditure of time would be to put these coefficients on tape such that for successive runs these coefficients are not recalculated.

The program must be modified to accept data in other formats, so one may be tempted to redesign the program to accept many different formats. Offhand this would seem so difficult as to be ill advised, unless of course one had with each observation its coordinates; in that case the modifications are trivial.

In general several runs must be made to ensure that more or less optimum combinations of parameters are being calculated. In the present program this requires making dimensional changes in common blocks and in dimension statements, a process that is prone to error. Since one may use variable dimensions in subroutines, one can rather easily make the changes to simplify this problem. One simply uses a master program to define the overall size of the common blocks and specify the variables that define the way in which the blocks are used in the subroutines. Subsequent changes are entirely restricted to the main program and can be very simple.

The way in which the TDS was represented appeared to be adequate for the copper-aluminum data. However, one might well want to use a more rigorous representation if for no other reason than to judge the adequacy of the present representation. Provided one retains the elastic constant ratios, X and Y, then it appears entirely feasible to solve the secular equations for each observation and thus reduce the approximations. One must obtain a different solution for each combination of X and Y, and should this represent appreciable computer time then the results could be stored on tape for later runs. One could also use additional interaction parameters, but this tends to complicate the solution. It is simpler to use low-order ε and ρ terms instead, and they should be just as satisfactory.

There are other possible problems in trying to represent the TDS: at reduced temperatures the different modes are not equally excited; at higher temperatures one will have higher orders of TDS. How to best handle these problems remains to be worked out. It would be practical to simply use Eq. (7) and include restraints to restrict the curvature in the ε and ρ series in those regions where this intensity was slowly varying, as was done for the α series for the copper-aluminum data.[17] One would now have to calculate many more terms, and this could pose a problem.

---

[17]R. O. Williams, "Short-Range Structure in a Copper 16 Atomic Percent Aluminum," to be sumitted for publication.

Many alloy systems show extra intensity near the Bragg peaks, which on precipitation leads to the so-called side bands. The present representation of the TDS is not capable of fitting this intensity. If one were to include enough terms in the ε and ρ series to represent this feature, then ill conditioning will very likely result, and one must use supplementary conditions as noted above, possibly leaving out the TDS expression entirely.

TESTING

The program must be properly implemented to give correct results. A fairly comprehensive initial test can be made as follows. First the program is implemented just as listed in this report and then run after removing the C's from the first column of the two test cards listed early in the program. When this is done the observations are random numbers between zero and unity. The solution should give correlation matrices very similar to those listed in Appendix B. The first coefficient, α_0, should have a value close to 5.0, and all the other coefficients should be zero within about 2 standard errors. The determinant should be approximately 1.2×10^{13}, and the variance should be 8.3×10^{-5}. And finally the results should be insensitive to values of X and Y.

We checked this program very carefully, and hopefully it is free of significant errors. The author would appreciate any errors being brought to his attention and in turn will notify other known users.

DISCUSSION

We have already indicated that certain problems can arise in a regression analysis but do not occur in a Fourier analysis, but these may be handled in a straightforward manner. Two minor advantages of the present method are that the correct x-ray scattering factors are used and weighting is used to correct for the varying precision of the observations. These will result in only modest improvements in the solution.

This method is considered to have several major advantages. The first is that the observations may be made with considerable freedom, and all observations can be included in the analysis. And secondly we obtain a complete error analysis. What might turn out to be the greatest advantage is that the method necessarily computes the residuals. If from these data one establishes the presence of systematic errors and is able to identify and reduce them, then a significant improvement in the solution may result. One could do the same thing after having obtained a solution by separation, but it is less likely to be as complete and to cover the same range in reciprocal space.

ACKNOWLEDGMENT

Among the several people at the laboratory who have contributed to this work, Cullie J. Sparks, Jr., deserves special mention.

APPENDIX A

```
      PROGRAM DIFFUSE
      DIMENSION RAD(60),PH1(60),PH2(60),WR(60),FBA(60),FQ(60),
     1FCU(15),FB(60),FAL(15),TDS(60),LL(40),MM(40),NN(40),FBO(60)
      COMMON/A/A(16,59),B(16,44),C(24,59),D(22,44)/B/AI(16,59),BI(16,
     144),CI(24,59),DI(22,44)/F/WA(16,59),WB(16,44),WC(24,59),WD(22,44)
      COMMON/C/H1,H2,H3,ISUM,APO,APL,GAM,EPO,EP,RHO,TD
      COMMON /E/AL(40),AM(40),AN(40),INDEX(40)/EL/X12,DX12,Y44,DY44
      COMMON/G/IR1(4032),AX(50)
      DATA FAL/13.,12.46,11.32,10.20,9.30,8.58,7.97,7.41,6.87,6.34,5.82
     1,5.33,4.86,4.42,4.02/
      DATA FCU/26.8,26.23,25.,23.36,21.59,19.78,18.,16.27,14.66,13.16
     1,11.81,10.81,9.56,8.64,7.84/
      ALAM=1.54178$AO=3.643
      X12=0.7$DX12=0.063$Y44=0.500$DY44=0.016
      XAB=0.16*0.84$BG=.02$RT6=SQRT(1./6.)$RT2=SQRT(1./2.)$PI=3.1415927
      EP=-4.*PI**2*XAB$RHO=2.*EP
      READ 109,((LL(I),MM(I),NN(I),INDEX(I)),I=1,40)
      DO 39 I=1,40$AL(I)=LL(I)*6.283185$AM(I)=MM(I)*6.283185
   39 AN(I)=NN(I)*6.283185$DO 1 I=1,16
    1 READ 100,(A(I,J),J=1,59)$DO 2 I=1,16
    2 READ 100,(B(I,J),J=1,44)$DO 3 I=1,24
    3 READ 100,(C(I,J),J=1,59)$DO 4 I=1,22
    4 READ 100,(D(I,J),J=1,44)
C  25 A(I)=RANF(0.)                                                    TEST
C     DO 25 I=1,4032                                                   TEST
      DO 6 I=1,60$X=(I-1)*.0174533$PH1(I)=COS(X)$PH2(I)=SIN(X)
      Y=SIN((I+9)*.01745329)/ALAM$Z=Y*20.$IZ=IFIX(Z)+1$Z=AMOD(Z,1.)
      TCU=FCU(IZ)*(1.-Z)+FCU(IZ+1)*Z$TAL=FAL(IZ)*(1.-Z)+FAL(IZ+1)*Z
      HH=RAD(I)=Y*AO$Z=TDS(I)=((.16*TAL+.84*TCU)/TCU)**2
      FQ(I)=2.*PI*Z*HH**2$Z=(TAL-TCU)/TCU
      FBA(I)=4.*PI*XAB*Z$YY=FB(I)=Z*Z*XAB*2.$FBO(I)=YY*.5
    6 WR(I)=Y/(2.-59/(I+43))*.002*COS((I+9)*.01745329)
      DO20I=1,4032
   20 WA(I)=0$ISUM=0
      DO5 J=1,59$X=RAD(J)$Y=WR(J)$APO=FBO(J)$APL=FB(J)$EPO=FQ(J)
      TD=TDS(J)$GAM=FBA(J)$DO5I=1,16$II=2*I-1$ISUM=ISUM+1$IR1(ISUM)=J
      Z=A(I,J)$IF(Z.GT.0)WA(I,J)=Y/(Z+BG)
      XX=X*PH1(II)*RT6$YY=X*PH2(II)*RT2$H1=2.*XX$H2=XX+YY$H3=XX-YY
    5 CALL SERIES
      DO 7 J=1,44$K=J+15$X=RAD(K)$Y=WR(K)$APO=FBO(K)$APL=FB(K)$EPO=FQ(K)
      TD=TDS(K)$GAM=FBA(K)$DO 7 I=1,16$II=2*I$ISUM=ISUM+1
      IR1(ISUM)=K$Z=B(I,J)$IF(Z.GT.0)WB(I,J)=Y/(Z+BG)
      XX=X*PH1(II)*RT6$YY=X*PH2(II)*RT2$H1=2.*XX$H2=XX+YY$H3=XX-YY
    7 CALL SERIES$H3=0.
      DO 8J=1,59$X=RAD(J)$Y=WR(J)$APO=FBO(J)$APL=FB(J)$EPO=FQ(J)
      TD=TDS(J)$GAM=FBA(J)$DO8I=1,24$II=2*I-1$IF(I.EQ.24)II=46
      H1=PH1(II)*X$H2=PH2(II)*X$ISUM=ISUM+1$IR1(ISUM)=J
      Z=C(I,J)$IF(Z.GT.0)WC(I,J)=Y/(Z+BG)
    8 CALL SERIES
      DO 9 J=1,44$K=J+15$X=RAD(K)$Y=WR(K)$APO=FBO(K)$APL=FB(K)$EPO=FQ(K)
      TD=TDS(K)$GAM=FBA(K)$DO9I=1,22$II=2*I$H1=PH1(II)*X$H2=PH2(II)*X
      ISUM=ISUM+1$IR1(ISUM)=K$Z=D(I,J)$IF(Z.GT.0)WD(I,J)=Y/(Z+BG)
    9 CALL SERIES$CALL STAT
      PRINT105$PRINT 102,A$PRINT 105$PRINT102,AI$PRINT105$PRINT102,WA
      PRINT105$PRINT 102,B$PRINT 105$PRINT102,BI$PRINT105$PRINT102,WB
      PRINT105$PRINT 101,C$PRINT 105$PRINT101,CI$PRINT105$PRINT101,WC
      PRINT105$PRINT 104,D$PRINT 105$PRINT104,DI$PRINT105$PRINT104,WD
  100 FORMAT(26F3.3)
  101 FORMAT(1H0,3P24F5.0)
```

```
102 FORMAT(1H0,20X,3P16F5.0)
104 FORMAT(1H0,5X,3P22F5.0)
105 FORMAT(1H1)
109 FORMAT(8(I3,2I1,I2,3X))
    END


    SUBROUTINE SERIES
    DIMENSION C(80)
    CCMMON AA,BB,AB
    COMMON /E/AL(40),AM(40),AN(40),INDEX(40)/EL/X12,DX12,Y44,DY44
    COMMON/D/AD(4032),D(4032,22),B(4032,17),CD(4032,3),A(4032,9)
    COMMON/C/X,Y,Z,J,APO,E,F,EPO,G,H,TD
    AD(J)=APO$CD(J,1)=EPO$IZ=IY=1
    DO 7 IX=1,11$AA=AL(IX)$BB=AM(IX)$AB=AN(IX)
    GOTO(1,2,3,4,5,6)INDEX(IX)
  1 B(J,IY)=F*(X*SL(X,C1)+Y*SL(Y,C2)+Z*SL(Z,C3))$D(J,IX)=E*(C1+C2+C3)  LOO*****
    C(IZ)=G*(X*X*C1+Y*Y*C2+Z*Z*C3)$C(IZ+1)=G*(X*X*(C2+C3)+Y*Y*(C1+C3)+
   1Z*Z*(C1+C2))$IZ=IZ+2$IY=IY+1$GOTO7
  2 S1=SL(X+Y,C1)$S2=SL(X-Y,C2)$S3=SL(X+Z,C3)$S4=SL(X-Z,C4)$S5=SL(     LLO*****
   1Y+Z,C5)$S6=SL(Y-Z,C6)$D(J,IX)=E*(C1+C2+C3+C4+C5+C6)$B(J,IY)=
   2F*(X*(S1+S2+S3+S4)+Y*(S5+S6+S1-S2)+Z*(S3-S4+S5-S6))$IY=IY+1
    C(IZ)=G*(X*X*(C1+C2+C3+C4)+Y*Y*(C1+C2+C5+C6)+Z*Z
   2*(C3+C4+C5+C6))$C(IZ+1)=G*(X*X*(C5+C6)+Y*Y*(C3+C4)+Z*Z*(C1+C2))
    C(IZ+2)=H*(X*Y*(C1-C2)+Y*Z*(C5-C6)+X*Z*(C3-C4))$IZ=IZ+3$GOTO7
  3 S1=SL(X+Y+Z,C1)$S2=SL(X+Y-Z,C2)$S3=SL(X-Y+Z,C3)$S4=SL(X-Y-Z,C4)    LLL*****
    D(J,IX)=E*(C1+C2+C3+C4)$B(J,IY)=F*(X*(S1+S2+S3+S4)+Y*(S1+
   1S2-S3-S4)+Z*(S1-S2+S3-S4))
    C(IZ+1)=H*(X*Y*(C1+C2-C3-C4)+Y*Z*(C1-C2-C3+C4)+X*Z*(C1-C2+C3-C4))
    C(IZ)=G*(X*X+Y*Y+Z*Z)*(C1+C2+C3+C4)$IZ=IZ+2$IY=IY+1$GOTO7
  4 S1=SLM(X,Y,C1)$S2=SLM(X,-Y,C2)$S3=SLM(X,Z,C3)$S4=SLM(X,-Z,C4)      LMO*****
    S5=SLM(Y,X,C5)$S6=SLM(Y,-X,C6)$S7=SLM(Y,Z,C7)$S8=SLM(Y,-Z,C8)$S9=
   1SLM(Z,X,C9)$S10=SLM(Z,-X,C10)$S11=SLM(Z,Y,C11)$S12=SLM(Z,-Y,C12)
    CA=C1+C2+C3+C4$CB=C5+C6+C7+C8$CC=C9+C10+C11+C12$XX=X*X$YY=Y*Y
    ZZ=Z*Z$D(J,IX)=(CA+CB+CC)*E
    B(J,IY)=(X*(S1+S2+S3+S4)+Y*(S5+S6+S7+S8)+Z*(S9+S10+S11+S12))*F
    B(J,IY+1)=F*(X*(S5-S6+S9-S10)+Y*(S1-S2+S11-S12)+Z*(S3-S4+S7-S8))
    C(IZ+3)=H*(X*Y*(C1-C2+C5-C6)+X*Z*(C3-C4+C9-C10)+Y*Z*(C7-C8+C11-C12
   1))$C(IZ)=G*(XX*CA+YY*CB+ZZ*CC)$C(IZ+1)=G*(XX*(
   2C5+C6+C9+C10)+YY*(C1+C2+C11+C12)+ZZ*(C3+C4+C7+C8))$C(IZ+2)=G*
   4(XX*(C7+C8+C11+C12)+YY*(C3+C4+C9+C10)+ZZ*(C1+C2+C5+C6))$IZ=IZ+4
    IY=IY+2$GOTO7
  5 S1=SLM(X,Y+Z,C1)$S2=SLM(X,Y-Z,C2)$S3=SLM(X,Z-Y,C3)                  LMM*****
    S4=SLM(X,-Y-Z,C4)$S5=SLM(Y,X+Z,C5)$S6=SLM(Y,X-Z,C6)
    S7=SLM(Y,-X+Z,C7)$S8=SLM(Y,-X-Z,C8)$S9=SLM(Z,X+Y,C9)
    S10=SLM(Z,X-Y,C10)$S11=SLM(Z,-X+Y,C11)$S12=SLM(Z,-X-Y,C12)
    CA=C1+C2+C3+C4$CB=C5+C6+C7+C8$CC=C9+C10+C11+C12
    D(J,IX)=E*(CA+CB+CC)$XX=X*X$YY=Y*Y$ZZ=Z*Z
    B(J,IY)=F*(X*(S1+S2+S3+S4)+Y*(S5+S6+S7+S8)+Z*(S9+S10+S11+S12))
    B(J,IY+1)=F*(X*(S5+S6-S7-S8+S9+S10-S11-S12)+Y*(S1+S2-S3-S4+S9-S10
   1+S11-S12)+Z*(S1-S2+S3-S4+S5-S6+S7-S8))
    C(IZ+2)=H*(X*Y*(C1+C2-C3-C4+C5+C6-C7-C8)+Y*Z*(C5-C6+C7-C8+C9-C1
   40+C11-C12)+X*Z*(C1-C2+C3-C4+C9+C10-C11-C12))$C(IZ+3)=H*(X*Y*(C9
   5-C10-C11+C12)+Y*Z*(C1-C2-C3+C4)+X*Z*(C5-C6-C7+C8))
    C(IZ)=G*(XX*CA+YY*CB+ZZ*CC)$C(IZ+1)=G*(XX*(CB+CC)+YY*(CA+CC)+
   1ZZ*(CA+CB))$IZ=IZ+4$IY=IY+2$GOTO7
  6 S1=SLMN(X,Y,Z,C1)$S2=SLMN(X,Y,-Z,C2)$S3=SLMN(X,-Y,Z,C3)             LMN*****
```

```
      S4=SLMN(X,-Y,-Z,C4)$S5=SLMN(X,Z,Y,C5)$S6=SLMN(X,Z,-Y,C6)
      S7=SLMN(X,-Z,Y,C7)$S8=SLMN(X,-Z,-Y,C8)$S9=SLMN(Y,X,Z,C9)
      S10=SLMN(Y,X,-Z,C10)$S11=SLMN(Y,-X,Z,C11)$S12=SLMN(Y,-X,-Z,C12)
      S13=SLMN(Y,Z,X,C13)$S14=SLMN(Y,Z,-X,C14)$S15=SLMN(Y,-Z,X,C15)
      S16=SLMN(Y,-Z,-X,C16)$S17=SLMN(Z,X,Y,C17)$S18=SLMN(Z,X,-Y,C18)
      S19=SLMN(Z,-X,Y,C19)$S20=SLMN(Z,-X,-Y,C20)$S21=SLMN(Z,Y,X,C21)
      S22=SLMN(Z,Y,-X,C22)$S23=SLMN(Z,-Y,X,C23)$S24=SLMN(Z,-Y,-X,C24)
      CA=C1+C2+C3+C4+C5+C6+C7+C8$CB=C9+C10+C11+C12+C13+C14+C15+C16
      CC=C17+C18+C19+C20+C21+C22+C23+C24$D(J,IX)=E*(CA+CB+CC)
      B(J,IY)=F*(X*(S1+S2+S3+S4+S5+S6+S7+S8)+Y*(S9+S10+S11+S12+S13+
     1S14+S15+S16)+Z*(S17+S18+S19+S20+S21+S22+S23+S24))
      B(J,IY+1)=F*(X*(S9+S10-S11-S12+S17+S18-S19-S20)+Y*(S1+S2-S3-S4
     2+S21+S22-S23-S24)+Z*(S5+S6-S7-S8+S13+S14-S15-S16))
      E(J,IY+2)=F*(X*(S13-S14+S15-S16+S21-S22+S23-S24)+Y*(S5-S6+S7-S8
     3+S17-S18+S19-S20)+Z*(S1-S2+S3-S4+S9-S10+S11-S12))$IY=IY+3
      XX=X*X$YY=Y*Y$ZZ=Z*Z$C(IZ)=G*(XX*CA+YY*CB+ZZ*CC)$C(IZ+1)=G*(XX*(
     5C9+C10+C11+C12+C17+C18+C19+C20)+YY*(C1+C2+C3+C4+C21+C22+C23+C24)
     6+ZZ*(C5+C6+C7+C8+C13+C14+C15+C16))$C(IZ+2)=G*(XX*(C13+C14+C15
     6+C16+C21+C22+C23+C24)+YY*(C5+C6+C7+C8+C17+C18+C19+C20)+ZZ*(C1+C2
     7+C3+C4+C9+C10+C11+C12))$XY=X*Y$YZ=Y*Z$XZ=X*Z$C(IZ+3)=H*(XY*(
     8C1+C2-C3-C4+C9+C10-C11-C12)+YZ*(C13+C14-C15-C16+C21+C22-C23-C24)+
     9XZ*(C5+C6-C7-C8+C17+C18-C19-C20))$C(IZ+4)=H*(XY*(C17-C18-C19
     1+C20+C21-C22-C23+C24)+YZ*(C1-C2-C3+C4+C5-C6-C7+C8)+XZ*(C9-C10-C11
     2+C12+C13-C14-C15+C16))$C(IZ+5)=H*(XY*(C5-C6+C7-C8+C13-C14+C15
     3-C16)+YZ*(C9-C10+C11-C12+C17-C18+C19-C20)+XZ*(C1-C2+C3-C4+C21-C22
     4+C23-C24))$IZ=IZ+6
    7 CONTINUE$DO42 I=2,3
   42 CD(J,I)=C(I)$DO 27 IX=12,22$AA=AL(IX)$BB=AM(IX)$AB=AN(IX)
      GOTO(21,22,23,24,25,26)INDEX(IX)
   21 XX=R(X)+R(Y)+R(Z)$GOTO 27                                        LOO-----
   22 XX=R(X+Y)+R(X-Y)+R(X+Z)+R(X-Z)+R(Y+Z)+R(Y-Z)$GOTO 27             LLO-----
   23 XX=R(X+Y+Z)+R(X+Y-Z)+R(X-Y+Z)+R(X-Y-Z)$GOTO 27                   LLL-----
   24 XX=S(X,Y)+S(X,-Y)+S(X,Z)+S(X,-Z)+S(Y,X)+S(Y,-X)+S(Y,Z)+S(Y,-Z)   LMO-----
     1+S(Z,X)+S(Z,-X)+S(Z,Y)+S(Z,-Y)$GOTO 27
   25 XX=S(X,Y+Z)+S(X,Y-Z)+S(X,-Y-Z)+S(X,-Y+Z)+S(Y,X+Z)+S(Y,X-Z        LMM-----
     1)+S(Y,-X-Z)+S(Y,-X+Z)+S(Z,X+Y)+S(Z,X-Y)+S(Z,-X-Y)+S(Z,-X+Y)$GOTO27
   26 XX=T(X,Y,Z)+T(X,Y,-Z)+T(X,-Y,-Z)+T(X,-Y,Z)+T(X,Z,Y)+T(Z,-Y,X)+   LMN-----
     1T(X,Z,-Y)+T(X,-Z,-Y)+T(X,-Z,Y)+T(Y,X,Z)+T(Y,X,-Z)+T(Y,-X,-Z)
     2+T(Y,-X,Z)+T(Y,Z,X)+T(Y,Z,-X)+T(Y,-Z,-X)+T(Y,-Z,X)+T(Z,X,Y)+
     3T(Z,X,-Y)+T(Z,-X,-Y)+T(Z,-X,Y)+T(Z,Y,X)+T(Z,Y,-X)+T(Z,-Y,-X)
   27 D(J,IX)=E*XX
      XX=X-FLOAT(IRND(X))$YY=Y-FLOAT(IRND(Y))$ZZ=Z-FLOAT(IRND(Z))
      X2=X-FLOAT(IFIX(X))-.5$Y2=Y-FLOAT(IFIX(Y))-.5$Z2=Z-FLOAT(IFIX(Z))
     1-.5$G1=XX*XX+YY*YY+ZZ*ZZ$G2=X2*X2+Y2*Y2+Z2*Z2$IF(G2.GT.G1)GOTO40
      XX=X2$YY=Y2$ZZ=Z2$G1=G2
   40 ZB=XX*XX+YY*YY$ZA=X*XX+Y*YY$COS1=(Z*ZZ+ZA)**2/G1
      COS2=(X*YY-Y*XX)**2/ZB$COS3=(ZA*ZZ-Z*ZB)**2/(ZB*(ZZ*ZZ+ZB))
      GX=AMAX1(ABS(XX),ABS(YY),ABS(ZZ))$S1=4.*SIN(3.141592*GX)**2
      S2=SIN(6.283184*GX)**2$P1=XX*XX/G1$P2=YY*YY/G1$P3=ZZ*ZZ/G1
      P12=P1*P2$PT1=4.*P12*(1.-3.*P3)$PT2=4.*P3*(P1+P2-6.*P12)
      P123=P12*P3*27.$PL=PT1+PT2$P0=1.-PL-P123
      DO41 I=1,9$IX=(I-1)/3$X=X12+DX12*IX$Y=Y44+DY44*(I-1-3*IX)
      VP=(1.-X+Y)*P123
   41 A(J,I)=(COS1/(S1*(P0+PL*Y)+S2*(PL*(1.+X+Y)+P123*(1.+2.*X+Y*4.)))+
     1COS2/(S1*(Y*(P0+PT1)+PT2)+S2*((1.-X-Y)*PT1+(2.*Y-1.)*PT2+VP))+
     2COS3/(S1*(Y*(P0+PT2)+PT1)+S2*((1.-X-Y)*PT2+(2.*Y-1.)*PT1+VP)))*TD
      RETURN$END
```

```
      SUBROUTINE STAT
      DIMENSIONIX(50),AA(44,44),XX(44,44),AS(50),AAA(44,44),AJ(60)
      DIMENSION ER1(59)
      COMMON/A/A(4032)/B/B(4032)/F/W(4032)
      COMMON/G/IR1(4032),AX(50)/D/D(4032,52)
CAUTION---NUM MUST AGREE WITH DIMENSION STATEMENT FOR AAA, AA AND XX
      NUM=44$NX=NUM*NUM$DO 201 I=1,59
  201 ER1(I)=0.$DO 15 I=1,NUM$DO15J=I,NUM$TS=0.$DO 14 K=1,4032
   14 TS=TS+D(K,I)*D(K,J)*W(K)
   15 AAA(I,J)=AAA(J,I)=TS
      DO 50 IEL=1,9$IF(IEL.EQ.1)GOTO52$J=NUM-1+IEL$DO51 I=1,4032
   51 D(I,NUM)=D(I,J)
      DO 62 I=1,NUM$TS=0.$DO63J=1,4032
   63 TS=TS+D(J,I)*D(J,NUM)*W(J)
   62 AAA(I,NUM)=AAA(NUM,I)=TS
   52 DO61I=1,NX
   61 AA(I)=AAA(I)$IF(IEL.NE.5)GOTO19
      DO 2 I=1,NUM$AX(I)=Y=1./SQRT(AA(I,I))$DO 2 J=1,NUM
    2 XX(I,J)=AA(I,J)*Y$PRINT 111$DO3I=1,NUM$Y=AX(I)$DO4J=1,I
    4 IX(J)=IRND(XX(J,I)*Y*100.0)$IT=I-1
    3 PRINT 100, I,(IX(J),J=1,IT)
   19 DO 18 I=1,NX
   18 XX(I)=0.$DO21 I=1,NUM
   21 XX(I,I)=1.
      CALLMATQ(AA,XX,NUM,NUM,DET,NUM,NUM)$IF(IEL.NE.5)GOTO54
      DO 20 I=1,NUM$AX(I)=Y=1./SQRT(XX(I,I))$DO 20 J=1,NUM
   20 AA(I,J)=XX(I,J)*Y$PRINT 112
      DO 16 I=1,NUM$Y=AX(I)$IT=I-1$DO17J=1,I
   17 IX(J)=IRND(AA(J,I)*100.*Y)
   16 PRINT 100, I,(IX(J),J=1,IT)$PRINT103
   54 DO 9 J=1,NUM$TA=0.$DO8 K=1,4032
    8 TA=TA+A(K)*W(K)*D(K,J)
    9 AJ(J)=TA$DO7 I=1,NUM$TA=0.$DO65 J=1,NUM
   65 TA=TA+XX(I,J)*AJ(J)
    7 AX(I)=TA$VAR=0.$DO 11 I=1,4032$TS=0.$DO 12 J=1,NUM
   12 TS=TS+AX(J)* D(I,J)$ZZ=A(I)-TS$X=ZZ*W(I)
      ER1(IR1(I))=ER1(IR1(I))+X$VAR=VAR+X*ZZ
      IF(IEL.EQ.9.AND.W(I).GT.0.)W(I)=ZZ
   11 B(I)=TS$VAR=VAR/(4032.-NUM)$PRINT 102,DET,VAR
      DO 13 I=1,NUM
   13 AS(I)=SQRT(VAR*XX(I,I))
      PRINT 101,((I,AX(I),AS(I)),I=1,NUM)$IF(IEL.NE.5)GOTO50
      NZ=NUM-1$PRINT105,ER1$DO 31 II=1,6$PRINT116,II$DO31 J=1,59
      GOTO(41,42,43,44,45,46)II
   41 K=J*16-15$GOTO47
   42 K=J*16- 5$GOTO47
   43 K=J*16$GOTO47
   44 K=J*24+1625$GOTO47
   45 K=J*24+1632$GOTO47
   46 K=J*24+1638
   47 A1=A2=A3=0.$DO 32 I=1,23
   32 A1=A1+D(K,I)*AX(I)$DO 33 I=24,40
   33 A2=A2+AX(I)*D(K,I)$DO 34 I=41,NZ
   34 A3=A3+AX(I)*D(K,I)$A4=AX(NUM)*D(K,NUM)$A5=A1+A2$A6=A4+A3$A7=A5+A6
   31 PRINT 109,J,A1,A2,A5,A4,A3,A6,A7,A(K)
   50 CONTINUE
  100 FORMAT(I3,1X,43I3)
  101 FORMAT(/,5(3(I10,2F10.5)/))
  103 FORMAT(1H1)
```

```
102 FORMAT(16H0  DETERMINATE  ,E10.4, 10H  VARIANCE  ,E12.4,31H     SOL
   1UTION, STANDARD ERROR    ,/)
105 FORMAT(//,24H  RESIDUAL DISTRIBUTION    ,/10(/10F10.4))
109 FORMAT(I20,3P3(F13.0,2F9.0))
111 FORMAT(45H1              CORRELATION BETWEEN PARAMETERS    ,//)
112 FORMAT(43H1     CORRELATION BETWEEN PARAMETER ERRORS  ,//)
116 FORMAT(17H1        TRACE NO.,I3, 86H         ALPHA      GAMMA      SUM
   1           TDS       QUAD     SUM          TOTAL      OBS.    ,/)
    RETURN$END


    FUNCTION SL(X,W)
    COMMON A,B,C
    D=X*A$SL=SIN(D)$W=COS(D)$RETURN$END

    FUNCTION SLM(X,Y,W)
    COMMON A,B,C
    D=X*A+Y*B$SLM=SIN(D)$W=COS(D)$RETURN$END

    FUNCTION SLMN(X,Y,Z,W)
    COMMON A,B,C
    D=X*A+Y*B+Z*C$SLMN=SIN(D)$W=COS(D)$RETURN$END

    FUNCTION R(X)
    COMMON A,B,C
    R=COS(X*A)$RETURN$END

    FUNCTION S(X,Y)
    COMMON A,B,C
    S=COS(A*X+B*Y)$RETURN$END

    FUNCTION T(X,Y,Z)
    COMMON A,B,C
    T=COS(A*X+B*Y+C*Z)$RETURN$END

    FUNCTION IRND(X)
    IF(X.GT.0)GOTO1
    IRND=IFIX(X-.5)$RETURN
  1 IRND=IFIX(X+.5)$RETURN$END
```

APPENDIX B

CORRELATION BETWEEN PARAMETERS

| | | |

CORRELATION BETWEEN PARAMETER ERRORS

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36	37	38	39	40	41	42	43	44
1																																												
2		4																																										
3	-81		6																																									
4		-1	-21	1																																								
5	81	-7	-98		0																																							
6		3	-48	-9		5	9																																					
7	-81		5	95		0	-97	-7																																				
8		5	-13	-8	-29		8	-10	-5																																			
9	64	-8	-89		0	88	11	-86		9																																			
10	-1	19		5	-8	-5	-52		4	24	-6																																		
11		4	-37	-8	-23		8	39	-6	22	10	-18																																	
12	-77		5	92	-1	-96	-8	96	-6	-85		5	-7																															
13	-7	9		8	13	-8		3	6	-57	-9	-23	-22	6																														
14	73	-2	-84		1	86		3	-92		2	75	-3		2	-91	-1																											
15		6	-37	-5	10		5	3	-4	22		7	17	25	-4	-32		2																											
16	-9	42	14		9	-14	-48	11	-42	-16	19	-62	11	35		-5	-30																											
17	-6	24		7	-29	-6	-7		5	-36	-8	-18	-7		6	26		-3	-36	21																									
18	39		4	-28		1	35	-5	-40	-6		0	0	-4	-44		5	49	-3		5	4																							
19	-1	25		2	5	-3	-20		2	-28	-5	-32	-29		2	22		0	-46	29	26		7																						
20		7	-36	-13	-5	12	34	-10	34	13	-23	44	-9	-44		4	27	-65	-22	-5	-19																								
21	-46		3	45	-4	-48	-2	51		0	-53	-1	-1	49		2	-49	-4		5		4	-8		2	-3																			
22	-41	-3	39	-2	-43		4	53		4	-27		0	3	49	-5	-71		2	-5		0	-57	-2		3	20																		
23		0	-8	-1	-16		0	1	-1		7		1	6	-22		0	-3		1		3		0		0		0	-1	-4	-1		1												
24		1		8		3		4	-2	-6		2	-7	-3		1	-8		1		6		0	-2		9		1		4		3	-7		0		0		2						
25	-5		3		3		0	-3	-3		3	-6	-3		0	-4		3		4	-2	-6		6		7		0		3	-5		2		1		0		6						
26		0		3		3		0	-3	-5		3	-1	-6	-2		-7		2		3	-2	-3	10		1		5		8	-6		3		0	-1		3		4					
27		1		0	-2	-1		2	-1		0		6		2		4		2	-1	-6		0		1	-5	-2		0	-2		6		0		0	-1	-57		-9		-9				
28		3	-2	-1	-1		2		2	-2		4		1		0		1	-2	-1		3		2	-3	-4		3	-1		3	-1	-3		0	-7	-77		2		4					
29	-2		2		1	-6	-1	-6		1		4	-1		0		1		1		0		1	-4		0		2		3		5		0	-1	-4		1		2		2	-5	-11	-5		
30		0	-4	-2	-4		2		3	-1		7		1	-1		6		1	-3	-1		1	-8	-2	-5	-3		5	-2		2		0	-74		-5	-16	64		3		4			
31	-1		3		1		0	-2	-2		2	-2	-1		1	-3		2		3	-3	-1		2		1	-2		2	-3	-1		3		2		6	59	-4	-2	-77		4	-2		
32		0		0		0		1		0		3		0	-5		0	-2		0		1		0	-1		3		2		2	-4	-2		0		2		3		0	-3	-6		1		5		4	-41	-8	-11		
33	-1	-3	-1	-3		1		4		0		4		4		3		4		0	-1		0		1	-8	-1	-5	-5		7		0		1	-3	-11	-1	-57	12	-5	-1	20		3	-4		
34		1		2		0		2		1	-1	-1		-4		0	-4	-3	-3		6		3	-2		7		0		6		2	-6	-2	-3	-1	60		8	14	-71	-2	-6	-71	-2		9	-22		
35		1	-2		0		0		0		1		0		0	-1		1		1		1	-1		4		1	-3	-2	-2		0		0		2	-2	-2	-44		1	3	55	-6	-3	-61		8		0	-1	
36		3	-3	-2		5		2		6	-3	-7		2	-2		1	-3		1		2		1		2		1		2	-3	-2	-1		1		2		1		1	-2	-6		2	-53	-8	-1	20		1		8		2
37		2	-4	-1		0		1		0	-2	-5		1		1		3	-1		0		1		5	-3		3		1		1		0	-2	-1		1		2		0	-4	-4		2		3		1		3	-7	-12		0		0	-5	
38		2	-4	-1		0		0		4	-1		2		1	-2		5		1	-2		1		0	-2		1	-6	-5		2		1		2	-5	-7		5		7	-35	-2		6	-12		1	-10	-11	21	-4	11		5	
39	-2		2		1	-1	-1	-1		0	-1		0	-2		1		0	-4		0		0		0		4		1		1		0		1		0		1		1	49		2		0	-61		2		3	55	-5		4		0	-71	-3	-2		2
40		2	-4	-3		1		2		4	-1		1		4		2		3	-2	-1		0		2	-3	-4	-4		-2		3	-1		1		0		0	-21	-9		1	-20		3		0	12		4		6	-5	-4	-9		2	-6		2	
41	-43	-15		1		3	-1		-1		1	-1		-1	-5		1		1		7		0	-7		3		4	-2		6		1	-1		0	-1	-9		10		2	-3	-6		8		3		1	-5	-3		0	-3	-2	-1	-1		4	-1	
42		10	47	-3	-3		2		4	-2		0		4	-2	-3	-2	-6		0		8	-1		0	-2	-6	-1		2	-1	-3		6	-2		0		4	-2	-7	-2		5		3		2		1	-3		5	-4		1		5		2	-24	
43	-2		7		4	-14	-3	-10		3	22	-3	10		1		4	-18	-4		2	-3	-5	-4		0		1		2		3		4	-7	-11		7	17		5	-5		9		0		1	-5	-13		1		3	-5	-7	-1	-1	-9		3	
44		2	-2	-2		0		2		0	-2	-1		2		0		0	-3	-4		1	-1	-4	-7	-1	-5	-1	-4	-3	-5		0		0		0		0		0		0		0		0		0		0		0		0		0		0	-1		1	1

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