EXPERIMENTAL DETERMINATION OF STRUCTURE FACTORS OF
TiAl AND SILICON

DISSERTATION
Presented in Partial Fulfillment of the Requirements for
the Degree Doctor of Philosophy in
the Graduate School of The Ohio State University

By

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* * * * *

The Ohio State University
1994

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Advisor
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Copyright
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1994
Dedicated
to

Late Mr. P. N. Subramanian
My tenth grade teacher
Setupati Higher Secondary School
Madurai
India

and

All teachers of the world
ACKNOWLEDGMENTS

I would like to thank my advisor Dr. Hamish Fraser for his support and guidance throughout this research project. Without his capabilities to identify, understand and solve the problem this project would not have been possible.

I would like to thank Dr. Zaluzec of ANL for allowing me to use Transmission Electron Microscope facilities located at the Electron Microscopy Center (EMC) at the Argonne National Laboratories (ANL) and teaching me everything about experimental energy filtering and convergent beam electron microscopy. I would also like to thank all the staff members at EMC at ANL, Argonne for their support and help during my visits there. I would also like to specially thank Dr. 'Bob' Wheeler IV for his participation, ideas, enthusiasm and support during his tenure at ANL and now at OSU.

I would also like to thank Dr. Dennis Maher, University of North Carolina, Raleigh for his ideas, enthusiasm and excellent insights in the application of Convergent Beam Electron Diffraction Technique. I would like to thank Dr. Ian Jones of University of Birmingham, UK, for teaching me principles of computational electron diffraction and writing the rocking curve matching program for the systematic case.

In the X-ray diffraction front, I would like to thank Dr. Downs for his expert guidance in the single crystal X-ray diffraction experiments, data analysis
and electron charge density studies. I would also like to thank Chiquito Crasto for his help in maintaining the X-ray equipment.

In the Fraser group, I would like to thank original group members Dr. Ahuja, Dr. Hou and Steve Johnson for being so resourceful during my Ph.D at OSU. I would also like to thank my diligent colleague, Sergui Altynov, for his ideas and input in developing the routines electron diffraction calculations.

I would also like to thank Dr. Clark, Dr. Wheeler, Dr. Ringnalda, Rajarshi and Balanethiram for helping me to proofread this thesis.

Last but not the least, I would like to thank my lovely wife 'Kamala' for enduring the tough times and my parents for letting me do whatever I wanted to do.
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D. 1 Output of the program SYSPAT.
# LIST OF SYMBOLS

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
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<tbody>
<tr>
<td>( m )</td>
<td>Relativistic mass of electron</td>
</tr>
<tr>
<td>( m_e )</td>
<td>Rest mass of electron</td>
</tr>
<tr>
<td>( \varepsilon_0 )</td>
<td>Permittivity of free space</td>
</tr>
<tr>
<td>( c )</td>
<td>Velocity of light</td>
</tr>
<tr>
<td>( t )</td>
<td>Foil thickness</td>
</tr>
<tr>
<td>( E_0 )</td>
<td>Accelerating voltage in KV</td>
</tr>
<tr>
<td>( e )</td>
<td>Electronic charge</td>
</tr>
<tr>
<td>( h )</td>
<td>Planck's constant</td>
</tr>
<tr>
<td>( \lambda )</td>
<td>Relativistic electron wavelength in Å</td>
</tr>
<tr>
<td>( d_g )</td>
<td>Interplanar spacing</td>
</tr>
<tr>
<td>( \mathbf{g} )</td>
<td>Reciprocal lattice vector ((</td>
</tr>
<tr>
<td>( \mathbf{K} )</td>
<td>Electron wave vector ((</td>
</tr>
<tr>
<td>( \mathbf{K}_i )</td>
<td>Incident wave vector</td>
</tr>
<tr>
<td>( \mathbf{K}_o )</td>
<td>Scattered wave vector</td>
</tr>
<tr>
<td>( \theta_B )</td>
<td>Bragg angle</td>
</tr>
<tr>
<td>( s )</td>
<td>Scattering vector defined by (2s = (\mathbf{K}_o - \mathbf{K}_i))</td>
</tr>
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\[
\mathbf{s} = |\mathbf{s}| = \frac{\sin \theta_B}{\lambda} = \frac{1}{2d_g}
\]
\[ f^e(s) : \text{Electron atomic scattering factor} \]

\[ f^X(s) : \text{X-ray atomic scattering factor} \]

\[ \Omega : \text{Unit cell volume} \]

\[ B : \text{Isotropic Debye-Waller (D-W) factor in } \text{Å}^2 \]

\[ B = 8\pi^2<u^2> \text{ Å}^2 \]

\[ <u^2> : \text{Mean square vibrational amplitude of atoms } \text{Å}^2 \]

\[ F_g^E : \text{Dynamic x-ray structure factor (or) Structure factor} \]

\[ F_g^C : \text{Theoretically calculated dynamic x-ray structure factor} \]

\[ F_g^C = \sum_j f^X_j(s) \exp(-B_j s^2) \exp(-2\pi i g \cdot r_j) \]

\[ F_g^E : \text{Experimental dynamic x-ray structure factor} \]

\[ F(s) : \text{Static x-ray structure factor} \]

\[ F^C(s) : \text{Theoretically calculated static x-ray structure factor} \]

\[ F^C(s) = \sum_j f^X_j(s) \exp(-2\pi i g \cdot r_j) \]

\[ F^E(s) : \text{Experimental static x-ray structure factor} \]

\[ V_g : \text{Fourier coefficient of the crystal potential in volt} \]

\[ V_g = \frac{1}{\Omega} \sum_j f^e_j(s) \exp(-B_j s^2) \exp(-2\pi i g \cdot r_j) \]

\[ U_g : \text{Dynamic structure factor a dimension of (length)}^2 \]

\[ U_g = \frac{2m |e| V_g}{\hbar^2} \]
\( \xi_g \) : Extinction distance with a dimension of (length)

\[
\xi_g = \frac{1}{\lambda |U_g|}
\]

\( \xi_g \) : Mean absorption with a dimension of (length)

\( V(r) \) : Total crystal electrostatic potential as seen by the electron

\[
V(r) = \sum_g V_g \exp(2\pi i g \cdot r)
\]

\( s_g \) : Vector in the reciprocal space indicating the deviation from the exact Laue condition

\( w \) : Deviation parameter or dynamical interaction parameter

\[
w = |s_g \xi_g|
\]

\( \rho(r) \) : Electronic charge density at point \( r \) in electrons/\( \text{Å}^3 \)

\( \rho_s(r) \) : Static electronic charge density at point \( r \) in electrons/\( \text{Å}^3 \)

\[
\rho_s(r) = \frac{1}{\Omega} \sum_g F(s) \exp(2\pi i g \cdot r)
\]

\( \rho_d(r) \) : Dynamic electronic charge density at point \( r \) electrons/\( \text{Å}^3 \)

\[
\rho_d(r) = \frac{1}{\Omega} \sum_g F_g \exp(2\pi i g \cdot r)
\]

\( \rho_n(r) \) : Nuclear charge density
Brittleness of TiAl has been attributed to strong directional bonding by a number of researchers. Their predictions have been based on theoretical calculations of electron charge density distribution. It is necessary to complement these predictions by experimental measurements. The work described in this thesis, aimed towards that end, involves measurement of Debye-Waller factors by four circle x-ray diffraction and of structure factors by energy filtered convergent beam electron diffraction CBED methods.

Stoichiometric single crystals are required for the measurement of Debye-Waller factors by the four circle x-ray diffraction method. Because of constraints imposed by the phase diagram only non-stoichiometric single crystal of TiAl are available. Measurement of Debye-Waller parameters have been attempted by using aluminum rich TiAl single crystals of compositions Ti54at%Al and Ti56at%Al. The symmetry of L10 structure of TiAl dictates that all reflections with Miller indices (hkl) not satisfying the condition h+k=2n should be extinct. However, during the x-ray diffraction experiments diffuse diffracted intensities were observed for reflections of h+k=2n+1 type. This indicate the possibility of occupation of the excess Al atoms on the Ti-sites. If the excess Al atom preferentially occupies one of the Ti-sites, it would lead to the formation of L12 type TiAl3 unit cells within the TiAl lattice. This notion has been further verified by least-squares refinement of the data obtained from Ti54at%Al single crystal. Also Debye-Waller factor values were different for equivalent Ti-sites in TiAl.
The CBED method has been developed for accurate structure factor measurement. Factors such as limitation due to the angular resolution of the aperture and complex matrix and perturbation treatment of absorption have been considered. Computer routines, incorporating these factors, have been developed for the calculation of CBED patterns and for matching the rocking curves. The $220$ structure factor of silicon has been measured, as a test case, to an accuracy $< 0.1\%$. The temperature dependence of the coefficient of $220$ absorption potential has been determined. The results indicate the Einstein phonon model [Bird and King (1990)] underestimates the coefficient of $220$ absorption potential of silicon.
CHAPTER - I

INTRODUCTION

TiAl is a low density intermetallic compound which has a high melting temperature and exhibits a high yield strength. In spite of these attractive properties, practical applications of TiAl are limited because it is brittle at room temperature. In order to understand the factors contributing to the lack of ductility, the deformation mechanisms (for example, slip systems) operative in TiAl have been the subject of considerable study over a number of years [e.g., Shechtman, Blackburn and Lipsitt (1974), Lipsitt, Schechtman and Schafrik (1975), Hug, Loiseau and Lasalmonie (1986) and Court, Vasudevan and Fraser (1990)]. In the main, these studies have attributed the brittleness of TiAl to the reduced mobility, or blocking, of certain types of dislocations. The explanations for the blocking of dislocations in TiAl have been based on either the possible non-coplanar nature of the dislocation core or the presence of significant Peierls valleys for dislocations arising from strong directional bonding. The former explanation for the blocking of dislocations, when the dislocation cores spread in planes and directions out of the slip plane, was proposed for Ni$_3$Al by Paidar, Pope and Vitek (1984). The latter explanation for the reduced mobility of dislocations was first proposed by Greenberg et al. (1988) by using the theoretical calculations of the electron charge density of Anisimov et al. (1987). Since then a number of electron charge density calculations [(e.g.) Morinaga et al. (1990), Fu and Yoo (1990) and Eberhart, Clougherty and
MacLaren (1993)] using different theoretical methods have been performed for TiAl. In this latter work, not only has the anisotropy of bonding charge densities been calculated, but also a comparison between these distributions in TiAl and the ductile intermetallic CuAu, both L1₀ structures been made, by using the Layered Korringa Kohn Rostoker (LKKR) method. The topology of the calculated electron charge density distribution of CuAu was found to be similar to that of ductile FCC metals, whereas the topology of charge density for TiAl was unique and showed strong directionality. This suggests that the explanation for lack of ductility in TiAl, based on the reduced mobility of dislocations due to directional bonding is probably more appropriate than that based on the non-planar nature of dislocation cores.

As is described below in section 2.1, these various computations of the anisotropy of bonding charge densities have indicated that some degree of covalent bonding exists in TiAl. However, it is also noted that the results of the various calculations do not always agree, and in fact in some cases contradictory results are obtained. Since the argument advanced by Greenberg, et al. (1988) depends critically on the directional effects of bonding, it is obviously necessary to establish credibility in the computational methods by experimental verification, and so distinguish between the various theoretical approaches. Indeed, the methods used to calculate the bonding charge density distributions in TiAl have been used generally in the area of computational materials science, and there is a more global need to provide experimental determinations of physical quantities that are derived from this computational effort. Thus, the work involved here is motivated not only by a desire to determine the factors influencing the ductility in TiAl, but also by the need to provide experimental verification of the methods used in computational science.
The main aim at the outset of the present study was to determine experimentally the anisotropy of bonding charge densities in TiAl. Such determinations of the electron charge density distribution requires accurate measurement of the Debye-Waller and the x-ray structure factors. The experimental studies described in this thesis have involved the use of four circle single crystal x-ray diffraction and the convergent beam electron diffraction (CBED) techniques. In order to obtain meaningful results, experiments must be made with a precision of less than 1%. It is for this reason that the CBED technique was used, since such precision has been demonstrated in the past (Zuo, Spence and O'Keefe (1988)) for GaAs. During the course of the work, it was found that CBED measurements on TiAl did not yield results which would permit sufficient precision to be obtained, for reasons described below, and it became necessary to perfect the techniques by studying a sample which has been characterized in detail by other techniques. The choice of Si was made, and the bulk of the work described has involved the determination of the 220 structure factor in Si. The result of this work is an extremely refined method of determining structure factors, and also a very precise determination of the 220 structure factor in Si, determined for the first time accurately using the CBED technique. The lack of a sample of TiAl with a precisely known composition of Ti-50at.%Al has prevented at this time the determination of the charge densities in TiAl.
CHAPTER - II

BACKGROUND AND LITERATURE REVIEW

2.1: Theoretical Predictions of Electron Charge Density Distributions in TiAl

Theoretical calculations of the electron charge density distribution in TiAl have been performed using various methods. Calculation of charge densities around Ti atoms was first performed using the method of linear combinations of muffin-tin orbitals (LMTO) by Anisimov et al. (1987). These results suggested that significant anisotropy in the charge density about Ti atoms exists, for example, in the (001) (using parentheses appropriate to a tetragonal structure [Hug, Loiseau and Lasalmonie (1986)]) layers containing only Ti atoms (Figure 2.1). Here directional bonding is expected between the Ti atoms along the <110> directions. In addition, such directional bonding is expected to develop between Ti atoms lying along the <001> directions. It was concluded by Anisimov et al. (1987) that, while there is a general tendency for bonding to be directional in TiAl, strong directional bonds are expected to form between atoms in any plane that contain only Ti atoms, such as (001), (110) and (112), resulting in a rigid framework in the Ti sublattice of this compound.

Using the results from the LMTO calculation, Greenberg et al. (1988) proposed that, in general, any dislocation segment lying parallel to a direction containing a row of Ti atoms such as <001>, <110> and <112> (Figure 2.2) will have
a small core-width. According to Peierls-Nabarro theory, dislocations with small core-width will experience a large Peierls stress. This analysis has been used to predict that certain dislocation segments in TiAl will tend to be sessile or exhibit reduced mobility because of the anisotropy in the Peierls stress [Court, Vasudevan and Fraser (1990)]. It is then expected that dislocation segments lying close to <001>, <110> and <112> will lie in deep Peierls valleys and therefore tend to be relatively sessile. The influence of temperature is to reduce the effective depths of the Peierls valleys by thermal activation. Hence, these dislocation segments are expected to exhibit increased mobility at elevated temperature. These predictions are supported by the experimental observations of dislocation structure made in this compound by Court, Vasudevan and Fraser (1990). It appears, then, that there is correlation between the predictions of Greenberg et al. (1988) based on the influence of bonding charge densities and the experimental observations of Court, Vasudevan and Fraser (1990).

Figure 2.1: The L1$_6$ unit-cell of TiAl shown above consist of layers of titanium and aluminum layers along [001] direction.
Figure 2.2: The [111] slip-plane of TiAl showing rows of similar atoms along [110] and [112] directions. It has been predicted that dislocations with line-direction parallel to these directions may encounter a large Peierls stress.
In another independent study, Woodward, MacLaren and Rao (1992) have supported the predictions by Anisimov et al. (1987) through the calculated variation in the density of states in TiAl. Their results indicate directional bonding, arising from electrons with energies near the Fermi level (d electrons), between Ti atoms lying along <110>. In addition, these authors also proposed directional bonding between second nearest neighbor Ti-Ti atoms in the (002) plane and second nearest neighbor Al-Al atoms along <001>.

Recently Morinaga et al. (1990) have opposed the theory of Greenberg et al. (1988) by proposing that p-d orbital bonding (Al-Ti) caused TiAl to become brittle; using the cluster variation method to calculate the electronic structure of TiAl, they predict that any alloying addition to stoichiometric TiAl which enhances p-d orbital (Al-Ti) bonding will cause the alloy to become brittle. These arguments are supported by the observation that TiAl becomes more brittle as the Al content is increased. Aluminum concentration in excess of stoichiometry is supposed to substitute at the Ti-site and hence lead to an increase in the number of p-d orbital bonds. Supporting arguments for the p-d orbital (Al-Ti) bonding theory have been provided by Fu and Yoo (1990) using their Full Potential, Linearized Augmented Plane Wave (FLAPW) calculations. FLAPW calculations, like LMTO calculations by Anisimov et al. (1987) also indicated that the charge distribution is anisotropic in (001) containing only Ti atoms, indicating Ti-Ti directional bonding along <110> directions. However, Fu and Yoo (1990) have stated that they noted p_z-type charge polarization about Al-sites resulting in strong cohesion between Al and Ti layers.
in TiAl. These authors attribute the strong cohesion between Al and Ti layers to the \( p-d \) (Al-Ti) type directional bonding.

2.2: Experimental Determination of Electron Charge Densities

Experimental methods for the determination of the electron charge density distribution are based on the theory of elastic scattering of x-rays. The fundamental physical quantity which describes the elastic scattering power of an atom is known as the x-ray atomic scattering factor, \( f^\text{A}(s) \), where \( s = |s| = \sin\theta/\lambda \), \( \omega \) is the Bragg angle and \( \lambda \) is the wavelength of incident x-ray beam) is the scattering vector shown in Figure 2.3. The x-ray atomic scattering factor is represented in units of number of electrons.

![Diagram of scattering vector](image)

Figure 2.3: Definition of the scattering vector \( s \).

In a diffraction experiment, x-rays are scattered by a crystal consisting of an aggregate of atoms arranged with regular periodicity. In such cases, the scattering power of an unit-cell is given by a quantity known as the static structure factor, \( F(s) \), in units of number of electrons. The quantity \( F(s) \) is referred to as the static structure factor because atomic vibrational effects are not included in its
definition. The relationship between $F(s)$ and the static electron density distribution, $\rho_s(r)$, can be derived from first principles of quantum mechanics [Feil (1977)],

$$F(s) = \int_{Unit-Cell} \rho_s(r) \exp(-4\pi is \cdot r) dr$$

(2.1)

According to equation (2.1), the static structure factor, $F(s)$, is the Fourier transform of the electron charge density, $\rho_s(r)$. This suggests, if structure factors are measured accurately, $\rho_s(r)$ can be determined by the Fourier transformation of $F(s)$. Then the electron charge density, $\rho_s(r)$, can be written in the form of a summation of series, as shown in equation (2.2)

$$\rho_s(r) = \frac{1}{\Omega} \sum_g F(g) \exp(4\pi is \cdot r)$$

(2.2)

where $g = 2s (|g| = 2 |s| = 2 \sin \theta / \lambda)$ and $\Omega$ is the volume of the unit cell. It should be noted that the quantity $F(s)$ is a complex number. However, for a crystal with a center of symmetry, the phase term of $F(s)$ is either 0 or $\pi$ and the imaginary part of $F(s)$ is equal to zero. Hence, only the amplitude of $F(s)$ has to be measured for determining the electron density distribution of a centrosymmetric crystal.

In a diffraction experiment, thermal vibration cannot be avoided. Hence, a modified structure factor known as the dynamic structure factor, $F_g$, is measured. The dynamic structure factor is related to the static structure factor through the Debye-Waller (D-W) factor. For a single elemental crystal, the relationship between the static and the dynamic structure factors is given by,
\[ F_g = F(s) \ast \exp(-Bs^2) \]  

(2.3)

where \( B \) is the isotropic D-W factor. The dynamic charge distribution, \( \rho_d(r) \), can be then derived by the Fourier transformation of the dynamic structure factor,

\[ \rho_d(r) = \frac{1}{\Omega} \sum_g F_g \exp(4\pi i s \cdot r) \]  

(2.4)

In this thesis, the dynamic x-ray structure factor, \( F_g \), hereafter will be simply referred to as the structure factor for convenience. Structure factors can be determined experimentally by x-ray, electron or \( \gamma \)-ray diffraction methods. Once \( F_g \) is measured, the static charge density distribution can be obtained by properly modeling the vibrational effects through the D-W factors.

In order to determine the electron charge density distribution by using equation (2.2) or (2.4), an infinite number of structure factors have to be determined. However, depending on the experimental technique, typically only a limited number of structure factors can be measured. As a result, any attempt to determine the electron charge density distribution would have to use a truncated series from equation (2.4). In order to understand how many terms of the series in equation (2.4) would be required to obtain a complete description of the electron charge density distribution, certain properties of Fourier relations (2.2) and (2.4) need to be analyzed.
2.2.1: Analysis of Fourier Relationship between \( F_8 \) and \( \rho_d(r) \)

The fundamental property of the Fourier relation is to transform a diffuse function into a compact one and vice versa. Since \( F_8 \) is the Fourier transform of \( \rho_d(r) \), information related to the compact core electrons in real space are represented by high-angle structure factors (large \( \sin \theta / \lambda \) or \( s \) value) in reciprocal space. Similarly, information relevant to diffuse bonding electrons, i.e. the valence electrons, is mainly contained in low-angle structure factors. It may be adequate to use low-angle structure factors to determine the bonding electron density distribution. However, it should be noted that the cutoff limit of \( s \) value for the low-angle structure factors depends on the composition, crystal structure and bonding characteristics. For instance, in some materials the electron charge density may accumulate in the compact region of the covalent bond [Coppens (1982)] and as a result its effect may extend beyond the cutoff limit of low-angle structure factors in reciprocal space. In such cases, it is essential to accurately determine the maximum number of structure factors possible, in order to obtain a full description of the electron charge density distribution. However, such charge accumulations in compact regions of bonds are highly unlikely in cases of the intermetallics where the nature of bonding is intermediate between covalent and metallic. In spite of this, it is still difficult to define a specific value of \( s \) for truncation of equation (2.4) without loss of details in measured electron charge density distribution. For these reasons, instead of depending on equation (2.3), different modeling and mapping methods have been developed to extract information about the electron density distribution from the structure factors. These methods are discussed in the following section.
2.2.2: Mapping of Electron Charge Density Distribution

Modeling of electron charge density distributions is unavoidable in the case of non-centrosymmetric crystals due to the 'Phase problem' mentioned earlier. Thus, the structure factor, \( F_g \), is a complex number with an amplitude, \(|F_g|\), and a phase, \( \phi \). For centrosymmetric crystals the phase, \( \phi \), is equal to either 0 or \( \pi \) radians, but it is an unknown for non-centrosymmetric crystals. Only the structure factor amplitude, \(|F(s)|\), is measured by most of the established experimental techniques. In a non-centrosymmetric crystal the phase term is usually determined by theoretical modeling of the electron charge density distribution. Fortunately, the current material of interest, TiAl, has a center of symmetry and does not require specific theoretical models to deal with the phase problem. However, in any material, only a fraction (>1% Zuo (1989)) of the total number of electrons in an atom participate in bonding while the remainder behave similar to the electrons in a spherical free-atom and the charge density distribution plotted using (2.4) will be dominated by the core electron density. Even in centrosymmetric crystals, in order to understand minor details about the nature of bonding, different mapping and modeling techniques, described in the following subsections, will have to be used.

2.2.2.1: Charge Deformation Maps

A total charge density distribution map plotted using a truncated Fourier series described by (2.2) or (2.4) would be dominated by the spherical atom densities and details about the distribution of bonding electrons would not be visible. To overcome this problem a 'deformation density' can be plotted to highlight the rearrangement of the electron density due to the interatomic bonding. The 'static
deformation density', \( \Delta \rho_s(r) \), given in equation (2.5), is the difference between the total electron density given by equation (2.2) and the electron density obtained by the superposition of the spherical free-atom density.

\[
\Delta \rho_s(r) = \frac{1}{\Omega} \sum_g (F_E^g(s) - F_C^g(s)) \exp(2\pi ig \cdot r)
\]  

(2.5)

where \( F_C^g(s) \) is the free-atom structure factor calculated from free atom scattering factors [Doyle and Turner (1968)] as given below

\[
F_C^g(s) = \sum_j f_j^s(s) \exp(-2\pi ig \cdot r_j)
\]  

(2.6)

Here \( f_j^s(s) \) is the calculated spherical free-atom scattering factors [(e.g.) Doyle and Turner (1968)]. Similarly, the 'dynamic deformation density', \( \Delta \rho_d(r) \), is given by

\[
\Delta \rho_d(r) = \frac{1}{\Omega} \sum_g (F_E^g - F_C^g) \exp(2\pi ig \cdot r)
\]

(2.7)

where \( F_E^g \) is the experimental structure factor and \( F_C^g \) is calculated from the free atom scattering factors as given below

\[
F_C^g = \sum_j f_j^s(s) \exp(-B_j s^2) \exp(-2\pi ig \cdot r_j)
\]  

(2.8)

Here \( B_j \) is the D-W factor. The deformation map is quite popular in the community involved in charge density calculations because of its simplicity and its ability to
qualitatively bring out the bonding features in crystals. The drawbacks of using a deformation map are that it is difficult to characterize the nature of bonding using these maps [Downs (1990)] and little quantitative information can be obtained from them.

In a number of metals and intermetallics, because of the experimental difficulties, only a few low-angle structure factors have been measured. In such cases, only the first few terms of the Fourier series in equation (2.6) would be available. In such cases, the nature of bonding has been determined by simply using the low-angle structure factors (e.g.) in Al and Cu by Fox, Tabbernor and Fisher (1989), in Be by Fox and Fisher (1988), in GaAs by Zuo, Spence and O'Keefe (1988), and in NiAl by Fox and Tabbernor (1991). There are two arguments in favor of deformation density mapping using the low-angle structure factors.

1. The bonding charge distribution is comparatively diffuse in real space. This suggests that the information would be contained within the low-angle structure factors in reciprocal space.

2. Because of experimental limitations and effect of thermal vibrations, for most materials the difference \((F^E_g - F^C_g)\) for the high-angle structure factors are less than the error in the measured value of \(F^E_g\).

These arguments have been challenged by Bernard and Zunger (1989) and Lu, Wei and Zunger (1992). Using the Local Density Approximation (LDA) calculations Bernard and Zunger (1989) and Lu, Wei and Zunger (1992) have calculated higher order structure factors in GaAs and NiAl respectively. A comparison of theoretical deformation maps plotted using an infinite number of structure factors with the experimental deformation maps plotted with just a few
low-angle structure factors revealed that features pertinent to the electronic contributions to bonding were lost in the maps plotted using the experimental low-angle structure factors. However, arguments presented in favor of using low-angle structure factors to plot a deformation map cannot be completely ruled out because of two reasons. Firstly, it is practically impossible to measure an infinite number of structure factors and secondly, an experimental charge deformation map is still an useful tool for obtaining information about charge density distribution. Another approach to avoid these problems is to use a pseudoatom model to extract information from the experimental data.

2.2.2 : Rigid Pseudoatom Model

Errors introduced in the electron charge density distribution due to the truncation of Fourier series, (2.3) and (2.4), can be averted by fitting a model electron charge density to the experimental structure factors. A rigid Pseudoatom model for electron charge density determination has been developed by Stewart (1976) to perform such modeling. This model involves direct determination of individual scattering factors of all the atoms in the unit cell by modeling the electron distribution around every atom or the scattering center. The electron distribution around each atom is modeled into a rigid pseudoatom by using multipole functions. The Fourier transform of the rigid pseudoatom gives a generalized atomic scattering factor of that pseudoatom in the unit cell. The electronic charge density distribution in real space is then derived by superposition of multipole functions refined using the experimental structure factor data. This model has been applied to electron charge density analysis of silicon by Spackman (1986).
2.3: Experimental Determination of Structure Factors

Experimental determination of electron charge density requires the measurement of x-ray structure factors. Structure factors should be determined to an accuracy of 0.1% [Hart and Deustch (1990)] in order to obtain accurate details about the bonding electron charge density distribution. In recent years x-ray, electron and gamma-ray diffraction techniques have been used to measure $F_G$. A brief review of some of the electron and the x-ray diffraction techniques used for measurement of structure factors will be presented in following sections.

2.3.1: Electron diffraction methods

Structure factors can be measured to an accuracy of 0.1% to 0.5% [Fox (1993)] by different electron diffraction techniques. Even though similar accuracy can be obtained by using the x-ray pendellösung and the single crystal diffraction methods, these methods require single crystals with accurate shapes and dimensions. Because of the practical difficulties in such single crystals, electron diffraction methods employing polycrystalline thin foils are preferred for the determination of structure factor of intermetallic compounds.

The major disadvantage of the electron diffraction techniques is that they are restricted to the measurement of low-angle structure factors. Electron diffraction techniques employing dynamical diffraction theory calculations also require accurate knowledge of the accelerating voltage, lattice parameters, foil thickness, orientation and the D-W factors. Measurement of accurate accelerating voltage requires a separate procedure every time an experiment is conducted. Attempts to measure D-W factors by convergent beam electron diffraction (CBED)
techniques, using the Higher Order Laue Zone (HOLZ) intensities [Holmestad et al. (1993)], is still in the developmental stage. As a result, electron diffraction techniques depend on x-ray or neutron diffraction techniques to measure D-W factors. However, the electron diffraction techniques are still popular because these disadvantages are outweighed by the difficulties in obtaining a perfect single crystal specimen for the x-ray diffraction experiment.

A brief review of the electron diffraction methods used for determination of x-ray structure factors will be presented later in this section. More detailed reviews of the electron diffraction methods can be found in Fox and Fisher (1988), Spence and Zuo (1992) and Spence (1992). All of the approaches reviewed here, for structure factor measurement, exploit the dynamical effects of electron diffraction. Electron diffraction experiments and subsequent dynamical theory calculations usually yield a quantity known as the Fourier coefficient of the crystal potential, \( V_g \) (volts). The experimentally measured \( V_g \) is then converted to \( F_g \) by using the Mott-Bethe relationship [Mott (1930)]. The experimental error in \( V_g \) is transferred to the x-ray structure factor during this conversion. The principle behind the Mott-Bethe relationship and the advantage of measuring \( V_g \) and then converting it to \( F_g \) are discussed in the following section.

Relation between \( V_g \) and \( F_g \)

In order to avoid any confusion in terminology, in this section the nomenclature and the symbols for the physical parameters have been adopted from Spence and Zuo (1992). By analogy to the x-ray atomic scattering factor, \( f^x(s) \), the electron scattering power of an atom is represented by the electron atomic scattering factor, \( f^e(s) \). The equations for \( f^x(s) \) and \( f^e(s) \) are defined as below
\[ f^x(s) = \int \rho_s(r) \exp(-4\pi is \cdot r) dr \] (2.9)

\[ f^e(s) = \int V(r) \exp(-4\pi is \cdot r) dr \] (2.10)

where \( V(r) \) is the total electrostatic potential as seen by the electron beam and \( s \) is the scattering vector as defined in Figure 2.3. Since in an actual electron diffraction experiment electrons are diffracted by a periodically arranged group of atoms, the Fourier coefficient of the crystal potential is used to represent the scattering power of the unit cell. The Fourier coefficient of the crystal potential, \( V_g \), is represented by a Fourier equation given in (2.11).

\[ V_g = \int_{Unit-cell} V(r) \exp(-B s^2) \exp(-2\pi ig \cdot r) dr \] (2.11)

where \( g = 2s \) and \( B \) is the D-W factor.

In an experimental electron or x-ray diffraction experiment, the parameter that is observed is the diffracted intensity. In order to determine the relationship between the \( f^e(s) \) and \( f^x(s) \), physical properties that influence the diffracted intensities in x-ray and electron diffraction have to be considered. The diffracted intensity in the case of electron diffraction is a function of \( f^e(s) \), which, in turn, is obtained by the Fourier transformation of \( V(r) \). The total electrostatic potential, \( V(r) \), includes the contribution from the nuclear charge and the electronic charge from electrons surrounding the nucleus.
The diffracted intensity in the case of x-ray diffraction is a function of \( f^x(s) \), which, in turn, is obtained by the Fourier transformation of \( \rho_s(r) \). The static electronic charge density, \( \rho_s(r) \), includes only the contribution from the electronic charge from electrons surrounding the nucleus. The unpolarized nuclear contribution from the nuclear charge density, \( \rho_n(r) \), is excluded in the case of x-rays. The relationship between \( V(r) \) and \( \rho_s(r) \) is then given by Poisson’s equation,

\[
\nabla^2 V(r) = -\frac{|e|}{\varepsilon_o} [\rho_n(r) - \rho_s(r)]
\]

(2.12)

The relation between \( f^e(s) \) and \( f^x(s) \) can be obtained by substituting equation (2.12) in (2.10) and assuming \( V(r) = 0 \) at large \( r \),

\[
f^e(s) = \frac{|e|}{16\pi^2 \varepsilon_o s^2} [Z - f^x(s)]
\]

(2.13)

where \( Z \) is the atomic number and \( s \) is the scattering vector. By analogy to equation (2.8), the Fourier coefficient of the crystal potential, \( V_g \), for an unit cell containing atoms of different kind can be obtained by superposition of the free atom scattering factors, \( f^q(s) \), as given in (2.14)

\[
V_g = \frac{1}{\Omega} \sum_j f^e_j(s) \exp(-B_j s^2) \exp(-2\pi i \mathbf{g} \cdot \mathbf{r}_j)
\]

(2.14)

where \( B_j \) is the isotropic D-W factor of \( j \)'th atom. Substituting (2.13) in (2.14) gives the Mott-Bethe relationship [Mott (1930)]
\[
V_g = \frac{|e|}{16\pi^2\varepsilon_0\Omega} \sum_j \left[ \frac{Z_j - f_j^x(s)}{s^2} \right] \exp(-B_j s^2) \exp(-2\pi i g \cdot r_j)
\] (2.15)

where \(\Omega\) is the volume of the unit cell. Two other quantities commonly used in the literature are the dynamic structure potential, \(U_g\), and the extinction distance, \(\xi_g\). Here

\[
U_g = \frac{2m|e|V_g}{\hbar^2}
\] (2.16)

\(U_g\) has the dimension of \((\text{length})^2\), and is usually represented in units of Å\(^2\).

\[
\xi_g = \frac{1}{\lambda|U_g|}
\] (2.17)

where \(\lambda\) is the relativistic electron wavelength in Å. The extinction distance \(\xi_g\) has the dimension of \((\text{length})\) and is usually represented in units of Å. Combining equations (2.8), (2.13), (2.15), (2.16) and (2.17), the relation between \(F_g\) and \(\xi_g\) can be derived.

\[
F_g = \sum_j Z_j \exp(-B_j s^2) \exp(2\pi i g \cdot r_j) - \left( \frac{8\pi^2\varepsilon_0\hbar^2\Omega s^2}{\gamma m_e e^2 \lambda} \right) \frac{1}{\xi_g}
\] (2.18)

where \(m_e\) is the rest mass of the electron, and \(\gamma\) is the relativistic constant given by

\[
\gamma = \frac{1}{\sqrt{1 - \frac{v^2}{c^2}}} = 1 + 1.956934 \times 10^{-3} E_0
\] (2.19)
where $c$ is the velocity of light, $v$ is the velocity of electron and $E_o$ is the accelerating voltage in kilovolts (KV).

The unique advantage of using equation (2.18) to convert measured $\xi_g$ to $F_g$ is that the experimental errors in $\xi_g$ are transferred to $F_g$ through the factor dependent on the $s$ ($s = 1/2d_g$). The ratio of the error in $F_g$ to that in $\xi_g$ can be obtained by differentiating $F_g$ (equation 2.18) with respect to $\xi_g$:

$$\frac{dF_g}{F_g} \frac{d\xi_g}{\xi_g} = \left(\frac{8\pi^2 \varepsilon_0 e^2 \hbar^2 \Omega_s^2}{\gamma m_e e^2 \lambda}\right) \frac{1}{\xi_g}$$ (2.20)

This ratio has been calculated for the low-angle structure factors by using the free atom scattering factors [Doyle and Turner (1968)] for Si and TiAl. Calculated results are tabulated in Tables 2.1 and 2.2. It can be seen from Table 2.1 for Si, the experimental error in $F_{III}$ is approximately 1/3 of the experimental error in $\xi_g$ and from Table 2.2 for TiAl, the experimental error in $F_{001}$ is 1/7 of the experimental error in $\xi_{001}$. The conversion factor increases as $s$ increases and after some point the experimental error in $\xi_g$ is magnified when it is converted to $F_g$ (e.g., 422 of Si and 003 of TiAl from Tables 2.1 and 2.2 respectively. This suggests that enhanced accuracy is achieved only for the first five low-angle structure factors in Si and the first ten low-angle structure factors in TiAl by using the Mott-Bethe relationship. It should be pointed out that, in the above analysis, the accuracy of $\xi_g$ is assumed to be independent of $g$.

Error analysis of the relationship between $\xi_g$ and $F_g$ also assumed accurate values for material properties such as the lattice parameters, the D-W factors, and
the relativistic electron wavelength. However, it should be noted that errors in lattice parameters, the relativistic electron wavelength and D-W factors would introduce an additional systematic error in $F_g$ value. Hence, accurate values of lattice parameters, the D-W factors, and the relativistic electron wavelength should be used for converting $\xi_g$ to $F_g$.

Table 2.1: The ratio of error in $F_g$ to the error in $\xi_g$, given by (2.20), for first six low-angle reflections of silicon. Ratios have been calculated using the room temperature D-W factor ($B_{si} = 0.4632 \text{ Å}^2$), room temperature lattice parameter ($a = 5.431 \text{ Å}$) and free atom scattering factor values [Doyle and Turner (1968)].

<table>
<thead>
<tr>
<th>(h k l)</th>
<th>$s = \frac{1}{2d_g} \text{ Å}$</th>
<th>$\frac{dF_g}{F_g} / \frac{d\xi_g}{\xi_g}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>(1 1 1)</td>
<td>0.16</td>
<td>0.33</td>
</tr>
<tr>
<td>(2 2 0)</td>
<td>0.26</td>
<td>0.61</td>
</tr>
<tr>
<td>(3 1 1)</td>
<td>0.31</td>
<td>0.71</td>
</tr>
<tr>
<td>(4 0 0)</td>
<td>0.37</td>
<td>0.86</td>
</tr>
<tr>
<td>(3 3 1)</td>
<td>0.40</td>
<td>0.95</td>
</tr>
<tr>
<td>(4 2 2)</td>
<td>0.45</td>
<td>1.09</td>
</tr>
</tbody>
</table>
Table 2.2: The ratio of error in $F_g$ to the error in $\xi_g$, given by (2.20), for first twelve low-angle reflections of TiAl. Ratios have been calculated using the room temperature D-W factor ($B_{Al} = 0.71$ Å$^2$ and $B_{Ti} = 0.42$ Å$^2$), room temperature lattice parameter ($a = 3.99$ Å and $c = 4.07$ Å) and free atom scattering factor values [Doyle and Turner (1968)].

<table>
<thead>
<tr>
<th>(h k l)</th>
<th>$s = \frac{1}{2d_g}$ Å</th>
<th>$\frac{dF_g}{F_g} / \frac{d\xi_g}{\xi_g}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>(0 0 1)</td>
<td>0.12</td>
<td>0.13</td>
</tr>
<tr>
<td>(1 1 0)</td>
<td>0.18</td>
<td>0.23</td>
</tr>
<tr>
<td>(1 1 1)</td>
<td>0.22</td>
<td>0.44</td>
</tr>
<tr>
<td>(0 0 2)</td>
<td>0.25</td>
<td>0.52</td>
</tr>
<tr>
<td>(2 0 0)</td>
<td>0.25</td>
<td>0.53</td>
</tr>
<tr>
<td>(2 0 1)</td>
<td>0.28</td>
<td>0.55</td>
</tr>
<tr>
<td>(1 1 2)</td>
<td>0.30</td>
<td>0.64</td>
</tr>
<tr>
<td>(2 0 2)</td>
<td>0.35</td>
<td>0.84</td>
</tr>
<tr>
<td>(2 2 0)</td>
<td>0.35</td>
<td>0.86</td>
</tr>
<tr>
<td>(0 0 3)</td>
<td>0.37</td>
<td>0.92</td>
</tr>
<tr>
<td>(2 2 1)</td>
<td>0.38</td>
<td>0.94</td>
</tr>
<tr>
<td>(3 1 0)</td>
<td>0.40</td>
<td>1.03</td>
</tr>
</tbody>
</table>
2.3.1.1: Convergent Beam Methods

The convergent beam electron diffraction (CBED) method of structure factor refinement involves matching of the experimental CBED intensities to that calculated by the dynamical theory of electron diffraction. The Fourier coefficient of the crystal potential can be determined by matching either one or two dimensional CBED patterns. Most of the attempts to measure $V_g$ have restricted themselves to one dimensional pattern matching i.e. making use of the variation of intensity along a given systematic row. Thus, one-dimensional CBED pattern matching is known as the 'systematic' or the 'rocking curve' method. In this method, a CBED pattern is recorded with the crystal oriented such that the electron beam is parallel to a direction (beam direction, B) reasonably away from a low-order zone axis. The aim is to minimize interaction from the non-systematic reflections. Usually the crystal is also oriented such that the beam direction is exactly or close to the Bragg condition of the reflection whose structure factor is being determined. An example of the energy filtered CBED pattern at systematic orientation with 220 silicon at the Bragg condition is shown in Figure 2.4. When the crystal is oriented close to the Bragg condition of the reflection of interest, the CBED disk contains parallel-fringes, perpendicular to the $g$-vector. The rocking curve shown in Figure 2.4b is the intensity profile taken along the direction parallel to $g$-vector (along the line AB in Figure 2.4a). The fringe spacing and the relative intensity of the fringes in a rocking curve are used fit $V_g$.

The CBED technique, developed by Kossel and Mollenstedt (1939), was first utilized by MacGillvary (1940) to determine $V_g$ by using the two beam approximation. Eight years later, Ackermann (1948a,b) demonstrated that the two
beam approximation is not accurate enough for structure factor determination. With the advent of computers, many beam calculations using dynamical theory of electron diffraction were attempted by Goodman and Lemphfuhl (1967) and Voss, Lehmpfuhl and Smith (1980) in MgO and Silicon, respectively. However, from the experimental standpoint all of the above followed the same procedure; the CBED pattern was recorded on to a film and the rocking curve was obtained by digitizing the recorded intensities. The problem with such a procedure is that, the rocking curve contained both elastically and inelastically scattered intensities. Unfortunately, the diffuse inelastic background cannot be accounted for by the dynamical theory of electron diffraction. Goodman and Lemphfuhl (1967) was the first to recognize the problem caused by the inelastically scattered diffuse intensities. Voss, Lehmpfuhl and Smith (1980) developed a solution by subtracting the inelastic background on the assumption that the intensity is a constant in the direction parallel to the fringes in a CBED disk (parallel to line CD in Figure 2.4a). However, this method is empirical and does not involve subtraction of all inelastic background. Zuo, Spence and O'Keeffe (1988) were the first to actually filter inelastically scattered electrons by scanning the CBED patterns over an electron spectrometer i.e. making use of electron energy loss spectroscopy (EELS). Zuo, Spence and O'Keeffe (1988) then used the Bloch wave formulation of the dynamical theory of electron diffraction to refine the low-angle structure factors in GaAs to an accuracy of 0.2%. The CBED systematic method was later extended to accurate determination of the structure factor phases in the non-centrosymmetric crystals by Zuo, Spence and Hoier (1989) and Zuo et al. (1993). This technique has been applied to the determination of $F_{220}$ of Si in the present work. More detailed description of this technique will be presented in Chapter IV.
A two dimensional CBED pattern matching method was first proposed by Bird and Saunders (1992). This method is commonly known as the 'zone-axis method' because match is made to a two dimensional pattern recorded along a direction of a low index pole. An energy-filtered zone-axis pattern recorded with the beam direction along the [110] pole of silicon is shown in Figure 2.5. In principle, two-dimensional zone axis CBED pattern contains sufficient details to fit structure factors of many of the reflections in the given zone. The sensitivity and the accuracy of the two dimensional CBED pattern matching was tested by Bird and Saunders (1992) by using a simulated CBED pattern as idealized experimental data. This test neglected the effect of Higher Order Laue Zone (HOLZ) reflection and assumed zero diffuse inelastic background. The results indicated that, in order to obtain structure factors values of 0.1% accuracy from the zone-axis patterns the lattice parameters must be known to the accuracy of 0.1%, D-W factors to 0.2% accuracy and the accelerating voltage to 1% accuracy.

The 'zone-axis method' was not attempted before the advent of the energy-filtering. Even if an energy filtering system is used, from the experimental standpoint, it is difficult to take care of the limitation in angular resolution of the electron counting system. More detailed discussion of the angular resolution of the electron counting system is given in Chapter IV. Computationally, the zone-axis method requires refinement of more than a few structure factors simultaneously. This increases the number of fitted parameters in the calculation and complicates the fitting procedure.
Figure 2.4: (a) Energy-filtered CBED systematic pattern of silicon 220 reflection. (b) Rocking-curve obtained by measuring the intensity profile along AB from (a).
Figure 2.5: Energy-Filtered [100] zone axis CBED pattern of silicon.
2.3.1.2: Critical-Voltage Method

The critical-voltage technique for the measurement of the low-angle structure factor was first developed by Nagata and Fukuhara (1967). Since then it has been applied frequently for the measurement of the low-angle structure factor. In comparison with the convergent beam electron diffraction technique, the critical-voltage method has the advantage of not requiring any sophisticated energy filtering or intensity counting device to record elastically scattered electrons. However, since the critical-voltage of the low order reflection may be anywhere in the range of 100-1500 KV, an high voltage electron microscope (HVEM) is required for structure factor measurement. The highest order of structure factor that can be determined by the critical-voltage technique is limited by the maximum accelerating voltage of the microscope. In addition, since the electron probe size is usually not converged, the area sampled is considerably larger than that examined in the CBED technique. Even though smaller diffraction areas can be achieved by using the high voltage CBED technique developed by Sellar, Imeson and Humphereys (1980), the use of converging probes at 1 MeV would lead to heavy contamination problems.

In the critical-voltage method, to determine the structure factor of the 'n'th order reflection, the crystal is oriented such that the 'n+1'th order reflection is at the Bragg condition. The accelerating voltage of the microscope is then varied until the intensity of the 'n+1'th order reflection reaches the minimum. The accelerating voltage under this condition is known as the 'critical-voltage'. The critical-voltage can be determined exactly by monitoring the disappearance of Kikuchi lines associated with the second order reflection [Thomas et al. (1974)] or
the disappearance of the maxima in a dark field bend contour [Lally et al. (1972)]. The intensity minima at the critical-voltage occurs because of destructive interference of the Bloch waves contributing to the second or higher order reflection. The ratio of ‘n’th to ‘n+1’th order structure factor value is then determined by analysis of the critical-voltage using the many beam dynamical theory.

The accuracy of the critical-voltage method for x-ray structure factor measurement is of the order of 0.1% to 0.6%. The accuracy to which the critical-voltage can be measured depends on the available instrumentation and the scattering properties of the crystal. Using modern HVEM’s and materials with favorable scattering properties, the critical voltage can be determined to an accuracy of ±1 KV. In the critical-voltage method the ratio of ‘n’th to ‘n+1’th order structure factors are determined, so the accuracy of the ‘n+1’th order structure factor plays an important role in the accuracy of ‘n’th order structure factor derived from this ratio. It is an usual practice to use the free atom structure factor values for the higher order structure factors. In such case, the deviation of the real crystal from the free atom model introduces large errors in the measured lower order structure factor. In addition, the accuracy of this technique also depends on the D-W factors because they are required for the calculation of the higher order structure factors and conversion of the coefficients of the crystal potential to the x-ray structure factors. In the case of intermetallic compound TiAl, the D-W factors have not been measured accurately. Under these circumstances it is impossible to achieve 0.1% accuracy as claimed by Fox (1993) for low-angle structure factors in TiAl.
2.3.1.3: Intersecting Kikuchi line method

The Intersecting Kikuchi Line (IKL) method for the structure factor measurement was developed by Gjonnes and Hoier (1971). Unlike the convergent beam method or the critical-voltage method, the IKL method does not require any intensity or voltage measurement. The IKL method uses the width of the separation that appears when an high-angle Kikuchi line intersects the Kikuchi lines associated with the low-angle systematic row. A more detailed description of the IKL method can be obtained from Spence (1992). Matsuhata et al. (1984) have measured the low-angle structure factors of the Cu and Cu₃Au using this IKL method to an accuracy of 0.5%. In order to measure structure factors using this technique, a suitable orientation is required where the splitting of intersecting Kikuchi lines is unperturbed by the other interactions. These diffraction conditions are difficult to attain using commonly used microscope accelerating voltages. As a result this method has not applied with enthusiasm in spite of its simplicity.

2.3.2: X-ray Diffraction methods

The most obvious choice for measurement of x-ray structure factors would be to use x-ray diffraction methods. Even though the structure factors can be measured by using either powder or single crystal diffraction, the powder method is not used because accuracy attainable is not sufficiently high. Single crystal x-ray diffraction techniques have been applied to a number of minerals, semiconductors and metals. The major advantage of x-ray diffraction technique is that, both the low and the high order structure factor amplitudes can be determined. Unfortunately, it is very difficult to obtain single crystals specimens, with required size and shape, of intermetallic compound such as TiAl. As a result, application of
the single crystal method to intermetallic compounds has been limited. A brief review of the single crystal x-ray diffraction methods will be presented in this section.

2.3.2.1: Four-Circle Single Crystal Diffraction

The four-circle single crystal x-ray diffraction is a fast and accurate method for structure factor determination. In this method, a well collimated monochromatic x-ray beam is made incident on a small a single crystal, the integrated intensities of diffraction peaks of various reflections are then collected in the sphere of reciprocal space. Structure factors are then determined from these integrated intensities after the application of proper corrections and analyses. The highest order structure factor that can be determined depends on the intensity of the incident beam, scattering power of the crystal and the crystal dimensions. In general, the integrated intensities, and thus the structure factors, of reflections with a s value \( s = \sin \theta / \lambda \) given by \( s = 1.3 \) can be measured using this technique. The four-circle method has been applied to electronic charge density distribution in beryllium by Larsen and Hansen (1984) and to the D-W factor determination (from high order structure factors) in NiAl by Georgeopoulous and Cohen (1977). More detailed description of the experimental data collection would be given in Chapter-III.

The data analysis procedure of four-circle method of structure factor determination assumes kinematic x-ray scattering conditions. A large defect free crystal would result in multiple scattering of x-rays and cause deviation from the kinematic scattering conditions. This phenomenon is known as ‘extinction’ [Downs (1990)] because multiple scattering tends to reduce diffracted intensities of certain
reflections. Kinematic diffraction conditions can be achieved by reducing the size of sample or the radiation wavelength, or both. The wavelength of the x-ray radiation is almost never varied in a x-ray diffraction experiment. The minimum diffracting volume (size) is determined by the scattering power of the crystal and the detector sensitivity. Because of these reasons, extinction is usually avoided indirectly by introducing defect population (dislocation, point defects) in the lattice. A large single crystal with an uniform defect distribution can be considered to contain large number of small crystals (mosaicty). In other words, in this method the crystal size is effectively reduced by defects to ensure kinematic scattering conditions.

The intermetallic compound TiAl is soft in comparison to minerals like quartz and exhibits a reasonable defect population, and as a result extinction is not a potential problem in TiAl. However, it should also be noted that excessive defect population will result in widening of x-ray peaks (mosaic spread) and introduce a dampening effect similar to that caused by thermal vibration. Excessive mosaicty results in over estimation of the D-W factors. Because of this reason, it is necessary to maintain optimum defect population so as to avoid extinction and minimize mosaic spread.

Under kinematic and extinction free conditions the relationship between the integrated intensity, \( I_g \), and the structure factor, \( F_g \), is given by [Feil (1977)],

\[
I_g = k \frac{(1 + \cos^2 2\theta)}{2} |F_g|^2
\]

(2.21)

where \( k \) is a constant known as the scale factor, \((1+\cos^2 2\theta)/2\) is the Lorentz
polarization factor and \( \theta \) the Bragg angle. The scale factor, \( k \), is dependent upon
the strength of the incident beam, counting period and the size of the crystal. The
accuracy of the experimental data in the single crystal method is usually estimated
by the agreement (R-factor) between the integrated intensities of equivalent
reflections after the Lorentz polarization and the absorption corrections. The R-
factor is given by

\[
R\text{-factor} = \frac{\sum_{g} \sum_{i=1}^{n} |F_{g,i}^2 - <F_{g}^2>|}{\sum_{g} n <F_{g}^2>} \times 100\%
\] (2.22)

where \( <F_{g}^2> \) is the average of corrected integrated intensities of the equivalent
reflections and \( n \) is the number of equivalent reflections. The experimental data
set is considered accurate enough for charge density analysis when the R-factor is
less than 0.5% [Downs (1991)]. In most materials, the R-factor and thus the accuracy
of the experimental data is dependent on the extent of the absorption correction.
Absorption correction is trivial in the case of spherical samples because all the
equivalent reflections are absorbed to the same extent and a simple spherical
absorption correction can be applied. However, as the single crystal specimen
deviates from a sphere, the agreement between the integrated intensity of
crystallographically equivalent reflections is degraded drastically and a different
method of absorption correction [Rees (1977)] has to be applied. More detailed
discussion on the role of the absorption correction and different absorption
correction methods will be presented in chapter III.
2.3.2.2: Pendellösung Methods

The x-ray pendellösung method is by far the most accurate method for the measurement of structure factors. Aldred and Hart (1973) were the first to determine the low-angle atomic scattering factors of silicon to an accuracy of 0.07%. In this method, a narrow, monochromatic x-ray beam is incident upon a wedge shaped single crystal. The hyperbolic pendellösung fringe pattern which result from the interference between different propagation modes is recorded on a high resolution nuclear emulsion. The fringe spacing in this pattern is related to the structure factor amplitude, |Fg|, through the spherical-wave theory of pendellösung fringes [Kato (1968)].

The pendellösung method is both simple and accurate, but in spite of that this method is not popular because it is very difficult to obtain wedge shaped single crystal for most materials. A number of modifications to overcome this difficulty has been suggested by various researchers. In a modified pendellösung technique, Saka and Kato (1986) measured Si structure factors with 0.05 to 0.1% accuracy using parallel-sided crystals and continuous radiation. The basic difference between monochromatic and white radiation method is that, in the former the wavelength, \( \lambda \), is kept constant and the dependence of the intensity on thickness is determined whereas in the latter the dependence of intensity as a function of \( \lambda \) is measured. Takama et al. (1984) and Takama and Sato (1988) have determined the structure factors of Al, Cu and Zn using white radiation. The accuracy achieved by this technique is of the order of 0.25% to 0.4%.
2.4: Discussion

Experimental determination of the electron density distribution requires accurate measurement of structure factors. A number of x-ray and electron diffraction techniques can be used to measure structure factors. The accuracy of the measured $F_g$ values depends mainly on the choice of the technique. Even though accurate measurement of $F_g$ is the goal, the choice of the technique is influenced mainly by the applicability of the technique to the material of interest. In addition, for TiAl, only inaccurate D-W factors values measured by powder diffraction after assuming $B_{Al} = B_{Ti}$ are available [Fox, Stoner and Cade (1992)]. In view of this, the D-W factors of TiAl need to be determined independently.

The intermetallic compound TiAl exists over a large compositional range (48-56at% Al content), has high melting point and is relatively ductile (compared to minerals). With available resources it seems practically impossible to obtain a perfect wedge shaped or parallel sided single crystal sample of TiAl. Because of this reason, the *pendellösung* method cannot be used for structure factor measurement. The four circle single crystal x-ray diffraction can be applied for structure factor measurement provided, small relatively defect free single crystals are obtained. The advantage of using four-circle method is that, accurate values of D-W factors can also be obtained by refining the high-angle structure factor data.

The Ti-Al phase diagram, close to stoichiometry, promotes nucleation of a second phase whenever there is a slight compositional fluctuation. As a result, realistically only non-stoichiometric single crystals TiAl with high Al content are available. A non-stoichiometric single crystal can be used for structure factor measurement by the four circle method, provided the site occupancies are refined
carefully. However, it is still difficult to measure structure factors with 100% confidence level using a non-stoichiometric single crystal. This complication can be avoided by selecting the electron diffraction routes which use a polycrystalline sample. In principle, the accuracy of electron diffraction techniques is also much better than the 0.5% accuracy [Coppens (1991)] of the four-circle method. Because of these reasons the electron diffraction route was chosen for structure factor measurement and the four circle method was chosen for D-W factor measurement.

The choice of the electron diffraction techniques was made after considering the following factors; the applicability of the technique to TiAl, the availability of instrumentation and accuracy of the technique. The Intersecting Kikuchi Line (IKL) was not chosen because this technique requires a specific orientation for every reflection and has at best an accuracy of only 0.5%. The critical-voltage method is more accurate than the IKL method and simpler than CBED method. However, the critical voltage method is limited by the maximum attainable accelerating voltage of the microscope. Even if a microscope with an unlimited accelerating voltage is made available, the critical-voltage cannot be determined accurately after a certain voltage because of radiation damage to the sample by the high-energy electron probe. Fox (1993) has measured critical-voltages in TiAl for first five structure factors (up to the 201) using a 1.5 MeV HVEM. The higher angle structure factors (above the 201) were not measured because the critical-voltages exceeded 1.5 MeV. Fox (1993) has also reported a poor accuracy (3%) in the measured value of the F_{201} and an ambiguous value for F_{110}. The error in F_{201} was caused by the radiation damage (around 1 MeV) to the sample and the accuracy of F_{110} is not determined, because of the inaccuracies in F_{220} value. It should be
noted that the radiation damage problem is amplified further for the higher-angle reflections because, the critical-voltage increases with decreasing d-spacing (or increasing s). Because of the above drawbacks, the critical-voltage method was not chosen for the determination of structure factors in TiAl.

The need for a HVEM and problems due to the radiation damage can be averted if the CBED method is used for $F_g$ measurement. A nominal voltage (120 KV) scanning transmission electron microscope (STEM), coupled with an energy-filtering system, can be used for structure factor measurement by the CBED method. The maximum number of structure factors that can be measured by the CBED method is limited by the dynamic range of the intensity counting system and the column of the microscope. If the dynamic range of the intensity counting system is small, the relative intensities in the CBED disk of a weak reflection with respect to the bright field disk cannot be measured accurately. However, the latest electron counting devices offer a large dynamic range for the accurate measurement of weak intensities. Since the accelerating voltage is not a limiting factor, the structure factors of relatively high-angle and strong reflections can be measured by the CBED technique.

As discussed in section (2.2.1.1) of this chapter, in the CBED technique, structure factor can be refined by matching either an one-dimensional rocking curve from a CBED pattern, recorded off the zone axis, or a two-dimensional zone axis CBED pattern. The rocking curve method is experimentally time consuming, because CBED patterns at systematic orientations have to be recorded for each structure factors individually, whereas a single zone axis pattern is sufficient for measuring all of the low-angle structure factors in that zone. However, the two-
dimensional pattern matching is computationally intensive, due to the large number of parameters included in the minimization procedure and requires efficient algorithms to attain a proper minimum. Complicated and efficient algorithms are in the developmental stage [Saunders (1994)] to carry out such minimization. The rocking curve method, on the other hand, requires relatively little computational power, because only a few variables have to be matched and standard routines from mathematical libraries can be used to perform the minimization. In addition, the rocking curve method is relatively well established, in comparison to the zone axis method [(e.g.) Zuo, Spence and O'Keeffe (1988), Zuo et al. (1993) and Zuo (1993)]. Because of these reasons, the CBED rocking curve method was chosen for measurement of structure factors in the intermetallic compound TiAl. Theoretical principles, experimental procedure, analytical methods and results of the four circle single crystal x-ray diffraction and the CBED techniques are discussed in Chapters III and IV respectively.
CHAPTER-III

MEASUREMENT OF DEBYE-WALLER FACTORS IN TiAl

3.1: Introduction

The four-circle single crystal x-ray diffraction method has been used extensively for crystal structure analysis [(e.g.) Collins and Mahar (1982)], Debye-Waller (D-W) factor determination [(e.g.) Saravanan, Mohanlal and Chandrasekaran (1992) and Georgeopoulos and Cohen (1977)] and electron density distributions [(e.g.) Staudenmann, Coppens and Miller (1976) and Larsen and Hansen (1984)]. In this technique, one integrated intensity data set can be used to determine the crystal structure, D-W factors and the electron charge density distribution. Of these three, the electron charge density analysis requires the most accurate (0.5% accuracy) data set. Even though the goal of this work is to obtain an accurate integrated intensity data set for modeling and mapping of electron charge density distribution, the work described in the thesis focuses on the determination of the D-W factors. The principles behind the experimental procedure, data analysis, and the results of x-ray diffraction experiments of TiAl will be discussed in this chapter.
3.2: Overview

3.2.1: Four-Circle Diffractometry

The geometry of the four-circle stage is shown in Figure 3.1. The four-circle stage has two base circles, $\omega$ and $2\theta$, mounted about the common axis passing through the diameter of a third circle $\chi$. The $\omega$ and $2\theta$ circles can be adjusted independently or geared together. The fourth circle $\phi$ is mounted on the $\chi$ circle and rides on the perimeter of the $\chi$ circle. When $\chi$ is equal to 0°, the $\phi$ axis coincides with the axes of $\omega$ and $2\theta$ circles as shown in Figure 3.1. Usually a single crystal, of approximately 25 to 75 $\mu$m size, is first mounted on a glass fiber and the glass fiber in turn is mounted on a goniometer head. The position of the crystal on the goniometer head is adjusted by using the setting screws in the goniometer such that the crystal is positioned exactly on the point of intersection of axes of four-circles at all $\omega$, $\phi$, $\chi$ and $\theta$ values.

Figure 3.1: A schematic representation of a four-circle stage in a four circle diffractometer. The $2\theta$ and $\omega$ circles are always coaxial and $2\theta$, $\omega$ and $\phi$ will be coaxial when $\chi = 0$. 
The experimental setup consists of a fixed x-ray source and a movable detector mounted at the end of a radial arm fixed to the 0\(^\circ\) mark of the 2\(\theta\) circle. A well collimated beam of x-rays from the source is aligned such that it passes through the point of the intersection of the axes of all four circles, (i.e.) through the crystal and the detector when the 2\(\theta\) arm is at 0\(^\circ\). The detector moves along with the 2\(\theta\) circle in a plane containing the incident and diffracted beams.

In a diffraction experiment an automatic x-ray diffraction peak search is performed, for every new crystal mounted on the diffractometer, by varying \(\omega\), \(\chi\), \(\phi\), and \(\theta\). Twenty five high angle peaks are centered and the corresponding \(\omega\), \(\chi\), \(\phi\), and \(\theta\) values are recorded. The cell parameters (lattice constants: a, b and c and angles: \(\alpha\), \(\beta\) and \(\gamma\)) are then refined by a least-squares method using the values of the \(\omega\), \(\chi\), \(\phi\), and \(\theta\) data set. The x-ray peaks are then indexed and an orientation matrix relating the \(hkl\) indices of the planes to the angular coordinates \(\omega\), \(\chi\), \(\phi\), and \(\theta\) values is obtained. The orientation matrix is then systematically used to collect diffracted intensity data from different \(hkl\) planes. During data collection, the \(hkl\) planes corresponding to the reflection of interest are oriented into the Bragg condition, with respect to the beam, by using the orientation matrix. The intensity profile \(i\) is then measured by first rotating the 2\(\theta\) circle along with the detector to the Bragg angle of the reflection and then symmetrically scanning the intensity profile of the diffracted beam. The integrated intensity is obtained by integrating the intensity profile over the scan width. The accuracy of the integrated intensity data depends on the accuracy of the orientation matrix.
3.2.2: Absorption Correction and Maximum Crystal Size Determination

Absorption of diffracted x-ray beams is a major source of systematic error in the determination of structure factors by the four circle method. It is not a major problem for a spherical single crystal, because all the equivalent reflections are absorbed to the same extent and a simple absorption correction can be applied. However in a non-spherical crystal, diffracted beams of different equivalent reflections will be absorbed to a different extents because of the difference path in lengths. For of this reason, the absorption correction for a non-spherical crystal is a complicated. In principle, for both spherical and non-spherical crystals the measured x-ray integrated intensities are corrected for absorption by dividing them by a transmission factor, \( \Lambda \), given by

\[
\Lambda = \frac{1}{\nabla} \int_{\nabla} \exp(-\mu T) dv
\] (3.1)

where \( \mu \) is the linear absorption coefficient, \( \nabla \) is the volume of the crystal and \( T \) is the sum of the incident and diffracted path lengths.

In equation (3.1), \( \mu \) for a compound can be determined from the calculated atomic mass absorption coefficients, \((\mu_i/\rho_i)\), of individual elements for the specific characteristic radiation of the source [International Tables for X-ray Crystallography: Vol-IV (1974)] by

\[
\mu = \rho \sum_i g_i \left[ \frac{\mu_i}{\rho_i} \right]
\] (3.2)

where \( \rho \) is the density of the compound, \( g_i \) is weight fraction of the individual
elements. The transmission factor for a spherical sample can be obtained by numerical integration of (3.1). For a non-spherical polyhedral crystal, the analytical absorption correction suggested by de Meulenaer and Tompa (1965) can be applied. De Meulenaer and Tompa (1965) first solved analytically equation (3.1) for a tetrahedral crystal. They then went on to apply the solution to a polyhedral crystal by dividing the polyhedron into a number of tetrahedra. This correction is usually referred to as the analytical absorption correction.

The spherical absorption correction is much simpler from the experimental point of view because the only experimental data required is the radius of the sphere. For the analytical absorption correction, the Miller indices of the crystal facets and the perpendicular distances of the crystal facets from an arbitrary origin have to be determined experimentally. The Miller indices of the crystal faces can be determined by using an optical microscope with a goniometer stage attachment. In this method two angular coordinates, $\phi_1$ and $\chi_1$, are determined for each facet of the crystal in a 'basis' (coordinate system) defined for the goniometer stage attached to the optical microscope. These angular coordinates are then transformed to the 'basis' of the diffractometer stage ($\phi$ and $\chi$) by using a transformation matrix. The Miller indices of the crystal faces are then determined from the angular coordinates by using the orientation matrix of the diffractometer. The accuracy to which the angular coordinates of the facets can be measured depends on the size of the crystal, and the areas of the facets of the crystal. The perpendicular distance of a crystal facet, from an arbitrary origin, can be measured using an image-splitting eyepiece attachment to an optical microscope. Usually one of the corners of the crystal is chosen as the arbitrary origin so that the perpendicular distances of the
from the origin facets, intersecting at that corner is zero.

Errors in the measured values of perpendicular distance, angular coordinates and the calculated absorption coefficients will introduce systematic errors into the structure factors. These systematic errors can be kept to minimum by minimizing the absorption correction. Absorption of x-rays, in turn, can be minimized by decreasing \( \mu \) or \( T \), or both. In fact in equation (3.1), when \( \mu = 0 \) and / or \( T = 0 \), \( A \) will be equal to 1 and there will be zero absorption. Since \( \mu \) is a material property, it cannot be changed. However, the total path length, \( T \), can be decreased by decreasing the size of the crystal. As a rule of thumb, the maximum size of the crystal is determined by placing a minimum limit of \( A = 0.8 \) [Downs (1991)] for any reflection by using equation (3.1). In other words, for charge density studies, absorption above 20% is not recommended.

The maximum crystal size for a single crystal x-ray diffraction experiment can be determined by assuming a spherical geometry. For a sphere, \( T \) is maximum when \( 2\theta = 180^\circ \). But in a normal data collection process the x-ray diffraction intensities of reflections with a \( \sin\theta/\lambda \) above 1 Å\(^{-1}\) are dampened by thermal vibrations, and are not observed. Then, for all practical purposes, the sum of the incident and the diffracted path length, \( T \), within a spherical crystal can be decided by assuming a limiting \( \sin\theta/\lambda \) value of 1 Å\(^{-1}\). A \( \sin\theta/\lambda \) of 1 Å\(^{-1}\) for Mo-K\(\alpha\) radiation is equivalent to a \( 2\theta \) value of 90\(^\circ\). For a sphere of diameter, \( d \), the maximum value of \( T \) is \( d \sqrt{2} \) when \( 2\theta \) is equal to 90\(^\circ\). The maximum diameters for Be and TiAl have been calculated for Mo-K\(\alpha\) radiation by substituting \( A = 0.8 \) and \( T = d \sqrt{2} \) into equation (3.1), and are tabulated in Table 3.1. In practice, in addition the above analysis, the diffracting power of the crystal, the intensity of the source and the
sensitivity of the detector are also taken into consideration before deciding the crystal size.

Table 3.1: Linear x-ray absorption coefficients of different elements and the maximum diameters estimated using equation (3.1) after assuming A = 0.8 and T = \( \sqrt{2} \times \) diameter. Required \( \mu \) values were obtained from the International Tables for X-ray Crystallography: Vol-IV (1974).

<table>
<thead>
<tr>
<th>Material</th>
<th>Absorption Coefficient ( \mu ) in cm(^{-1} )</th>
<th>Diameter in ( \mu )m</th>
</tr>
</thead>
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<tr>
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<td>3300.0</td>
</tr>
<tr>
<td>TiAl</td>
<td>65.00</td>
<td>33.0</td>
</tr>
</tbody>
</table>

3.3: Experimental Procedure

3.3.1: Experimental Setup

A Rigaku-AFC6 four-circle x-ray diffractometer, located in the Department of Geological Sciences, the Ohio State University, was used for the x-ray diffraction experiments. The four circles of the Rigaku-AFC6 diffractometer are driven by stepping motors controlled by micro-VAX computer. A molybdenum rotating anode source was used to generate x-rays. This source requires an high vacuum system and a sophisticated cooling system for operation. In spite of this, the rotating anode source is preferred because a relatively intense x-ray beam can be produced (60 KV, 200 mA filament current) when compared with an x-ray tube source (50 KV, 40 mA filament current), without melting the target. This allows one to obtain measurable diffracted intensities from a small volume of diffracting specimen.
3.3.2: Specimen Preparation

The major barrier for accurate data collection in a single crystal diffraction experiment is the availability of a good quality single crystal specimen. In order to maintain a minimum 80% transmission factor (\(A = 0.8\)) for TiAl, a single crystal of approximately 33 \(\mu\)m thickness is required (Table 3.1). Through preliminary experiments it was found that the diffracted intensity of high-angle reflections from a 33 \(\mu\)m TiAl single crystal were very difficult to detect, even when the full capacity of 55 KV accelerating voltage and 180 mA current of the source of the AFC6 diffractometer was utilized. However, intense high order reflections are required for the accurate determination of the orientation matrix and D-W factors. To achieve this the size of the crystal used in the x-ray diffraction experiment has to be increased (which will increase the absorption correction).

The ideal specimen shape for a single crystal x-ray diffraction experiment is a perfect sphere. A spherical single crystal for x-ray diffraction can be obtained by using the Nonius crystal spherizer [Saravanan, Mohanlal and Chandrasekaran (1992)]. In this technique, a large quantity of small cubic crystals is loaded into a small, semi-closed cylindrical container whose interior wall is coated with abrasive material. A constant flowing stream of air is directed into the cylinder so that the cubic crystals are ground to spheres by the wall of the container. The disadvantages are; this technique is slow (it takes few months for grinding) and requires a large number of small, perfectly cubic, or close to cubic, single crystals as a starting material.

A new method for producing spherical single crystal specimens of metals and intermetallic compounds was developed in the present research. This method
involves heat treating gas atomized spherical powders, at temperatures close to the melting point, to allow recrystallization and grain growth. The heat treated material is then washed in an acid solution to remove the oxide layer. The particles were chosen randomly and tested for single-crystallinity by performing a small step-size peak search over large angular ranges of \( \omega, \phi, \chi, \) and \( 2\theta \) circles, and determining an orientation matrix from the angular coordinates of these x-ray peaks. This orientation matrix is then tested by orienting the crystal to bring any other randomly chosen reflection of the material to the diffracting condition. The major advantage of this technique is that the crystal is never physically sheared; as a result, perfect, deformation-free, spherical single crystals of small diameter (> 50 \( \mu \)m) can be produced. The major disadvantage of this method is that it is difficult to confirm unequivocally whether a particle is a single crystal or not. In many cases, a particle may contain one large grain and a number of relatively very small grains. The question as to whether such a particle can be used in a single crystal x-ray diffraction experiment is still an open research issue.

The technique described above has been used to produce almost spherical Ti\(_3\)Al single crystal. About 50 grams of > 60 \( \mu \)m sized gas-atomized, two phase Ti 48at\% Al powder was encapsulated in an evacuated quartz tube back filled with low pressure, high purity, argon. The powders were then heat-treated for 72 hours at 1325 °C. The excess Al in the Ti 48at\% Al was lost during the heat treatment and the powder mainly contained the Ti\(_3\)Al phase. The oxide layer was washed off in a dilute (HF + HNO\(_3\)) solution. X-ray peak searches were performed on a number of spherical particles until a single crystal was obtained. In order to apply this method to TiAl, high Al content, gas atomized, TiAl powders will have to be used.
as a starting material to compensate for the Al evaporation. This method was not attempted for TiAl because high Al content gas atomized TiAl powders were unavailable.

When spherical single crystals are not available a multifaceted specimen close to cube shape can be used. In this technique, a cubic specimen or a polyhedral specimen closely shaped to a cube is cleaved from a large thin sheet by using a sharp single edged razor blade. This technique is very successful in the case of hard minerals with specific cleavage planes. In a ductile intermetallic like TiAl, cutting using sharp razor blade leads to heavy plastic deformation of the single crystal. The x-ray peaks from the such crystals are broad because of the mosaic spread and are difficult to center. The accuracy of the orientation matrix is affected when the peaks are not properly centered. In addition, the x-ray intensities are also dampened as a result of deformation. As a first approximation a single crystal of Ti56at%Al crystal specimen of size $\approx 75\times 75\times 150\ \mu m$ was cut, using a razor blade, from large single crystal kindly supplied by Drs. Kad and Oliver of The University of Tennessee, Knoxville.

In order to minimize the deformation of the single crystal, different approaches to cutting perfect small single crystals from large ones have been tried in ductile materials. Larsen and Hansen (1984) used electric arc machining to produce beryllium crystals of size $100\times 250\times 300\ \mu m$ for their single crystal diffraction experiment. Using the same principle, a Ti54at%Al rod of $\approx 80\times 80\times 500\ \mu m$ size was machined from a large grained Ti54at%Al material (also supplied by Dr.’s Kad and Oliver of The University of Tennessee, Knoxville) by electric discharge machining (EDM). A fine, 75 $\mu m$ diameter, wire was used to minimize the depth
of the oxide layer formed during machining. A number of small crystals of size ≈ 80x80x100 µm were broken from these rods, using a razor blade, and the crystal with best looking x-ray peaks was chosen for the experiment. Examples of the broad x-ray peak (2θ = 20.1°) of the Ti54at.%Al single crystal produced by cleaving the electric discharge machined rods, together with the sharp x-ray peak (2θ = 18.6°) from undeformed Ti3Al, produced by heat treatment, are shown in Figures 3.2 and 3.3. As per Laue interference function, for a perfect defect free crystal, the width of the x-ray peak decreases as the volume increases. The Ti3Al crystal (≈ 40 µm) produced by heat-treatment of gas atomized powder had a smaller volume than the Ti54at.%Al single crystal (≈ 75 µm). In spite of that, because of the artifacts introduced during specimen preparation, the width of Ti54at.%Al peak in Figure 3.2 is much larger than that of the x-ray peak from the Ti3Al crystal as shown in Figure 3.3.
Figure 3.2: Wide x-ray diffraction peak ($2\theta = 20.1^\circ$) from Ti54at%Al single crystal produced by cleaving the electric discharge machined rods.
Figure 3.3: Sharp x-ray peak (2θ = 18.6°) from an undeformed Ti₃Al crystal produced by heat treatment of gas atomized powders.
3.4: Data Collection and Analysis

The integrated intensity data sets were collected at ambient temperature (22 °C) from single crystal specimens of compositions Ti54at%Al and Ti56at%Al prepared by techniques described in the previous section. A Mo-Kα rotating anode source, with a Zr-filter, was used as the source in the x-ray experiment. An automatic peak search was performed to determine the orientation matrix for each crystal independently. The orientation matrix was further refined by centering the high-angle reflections ($\sin \theta / \lambda > 0.65$) by least-squares refinement. The lattice parameter of the material was determined by a least-squares method, using both the angular coordinates ($\omega$, $\phi$ and $\chi$) and the $hkl$ indices of twenty five reflections ($\sin \theta / \lambda > 0.65$). All the above calculations are features of the Rigaku AFC6 diffractometer software and are menu driven. The intensity profile of various reflections (up to a $\sin \theta / \lambda = 1.0$) were then collected by symmetrically scanning over the center of x-ray peaks.

The crystal structure of TiAl [Duwez and Taylor (1952)] dictates that only those reflections with indices satisfying the reflection condition $h+k = 2n$ are present. However, during the data collection, as an independent test for the space group and atomic positions, the integrated intensities of reflections with all possible combinations of $h$, $k$ and $l$ within the $\sin \theta / \lambda$ of 1.0 Å$^{-1}$ were collected. In spite of the fact that the Ti56at%Al crystal was larger than the Ti54at%Al, the x-ray peaks from the former crystal were comparatively more diffuse than those obtained from the latter. The increase in width might have resulted from a compositional change or from deformation (artifacts) introduced during specimen preparation.
After the completion of the data collection, angular coordinates of facets of the crystal, and perpendicular distances of facets from an arbitrary origin, were measured by using an optical microscope coupled with an image splitting eyepiece and a goniometer mounting stage. The facets were then indexed by using the orientation matrix. The integrated intensity data sets were then processed by using the TEXSAN\textsuperscript{1} structure analysis software package. The net integrated intensities were corrected for Lorentz-polarization by using the routine PROCESS. The routine HKL was used for the absorption correction and averaging of the equivalent reflections. An analytical correction option in the routine HKL, based on the methodology by de Meulenaer and Tompa (1965), was applied to the data set. A total of 193 independent reflections were obtained after averaging. The agreement factor (R-factor) between the equivalent reflections for the Ti54at\%Al single crystal was 3.7\% and for the Ti56at\%Al single crystal was 7.3\%. Due to poor accuracy, the integrated intensity data set from the Ti56at\%Al single crystal was not used to extract any quantitative information. The averaged intensities of the first 15 low-angle reflections obtained from the Ti54at\%Al and Ti56at\%Al single crystals, after Lorentz-polarization and absorption corrections, are listed in Tables 3.2 and 3.3.

\textsuperscript{1}TEXSAN is trademark of Molecular Structure Corporation, Texas, USA.
Table 3.2: Averaged integrated intensities, $I_g$, of 15 low-angle reflections from Ti54at%Al after Lorentz-polarization and absorption corrections.

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<th>$\sigma(I_g)$</th>
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Table 3.3: Averaged integrated intensities, $I_g$, of 15 low-angle reflections from Ti56at%Al after Lorentz-polarization and absorption corrections.

<table>
<thead>
<tr>
<th>h</th>
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3.5: Results and Discussions

The L1₀ crystal structure of TiAl, referred to as the 'large unit cell' and the smallest possible unit cell of TiAl referred to as the 'small unit cell', are shown in Figure 3.4. A detailed crystallographic description of the two different unit cells of TiAl is given in Tables 3.4 and 3.5. The L1₀ unit cell or the large unit cell is the unit cell most commonly used by the materials science community to describe the TiAl crystal structure. In this work, unless otherwise specified, the reflection conditions, atomic arrangements and lattice parameters are based on the L1₀ structure, or the large unit cell, given in Table 3.5.

The symmetry of L1₀ unit cell dictates that the reflections with Miller indices that do not satisfy the condition \( h+k = 2n \), will be extinct. However, from Tables 3.4 and 3.5, it can been seen that weak diffraction intensities for reflections with indices satisfying the condition \( h+k = 2n+1 \), are present in both in Ti54at%Al and Ti56at%Al. The diffuse intensity profile of the (010) reflection satisfying the condition \( h+k = 2n+1 \) from the Ti54at%Al single crystal is shown in Figure 3.5. To understand this anomaly, the following factors should be considered; the reflection condition \( h+k = 2n \) is valid only for a perfectly ordered, stoichiometric, L1₀ type unit cell, but both of the TiAl specimens used in the experiment were non-stoichiometric and had excess Al atoms. These excess Al atoms can either occupy interstitial sites (improbable) or substitute for a Ti atom in the Ti sublattice. If the excess Al atoms occupy the interstitial sites, or are randomly distributed in the Ti sublattice, there will be no intensity for the reflections with Miller indices satisfying the condition \( h+k = 2n+1 \). If Al atoms substituted for Ti atoms in Ti sublattice, then reflections with Miller indices satisfying the condition \( h+k = 2n+1 \) need not be extinct.
Figure 3.4: Large (L1₀) and small unit cell of the TiAl structure.
Table 3.4: Crystallographic data on TiAl; Small unit cell

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Table 3.5: Crystallographic data on TiAl; \( L_1 \) \(_0 \) structure (Large unit cell)

<table>
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<th>Multiplicity</th>
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</table>
Figure 3.5: The intensity profile of the weak and diffuse $010$ reflection breaking the symmetry rule $h+k = 2n+1$ from single crystal Ti54at%Al.
This point can be tested by comparing the intensities of \( h+k = 2n+1 \) reflections obtained from specimens with different Al contents; thus if Al atoms substitute for Ti, the intensity of \( h+k = 2n+1 \) type reflections should increase with increasing Al content. Since the intensity of the diffracted x-rays is also a function of the volume of the single crystal and counting period, the intensities from two different crystals cannot be directly compared, however, the ratios of the intensities can be compared. The ratios \((I_{(010)}/I_{(020)}),(I_{(101)}/I_{(202)})\) and \((I_{(110)}/I_{(220)})\) from the single crystals of two different compositions are listed in Table 3.6. When the Al content is increased from 54at% to 56at% there is about 99% increase in the \((I_{(010)}/I_{(020)})\) ratio and about 120% increase in the \((I_{(101)}/I_{(202)})\) ratio, but there is only 4% change (within error range of 3 to 9%) in the \((I_{(110)}/I_{(220)})\) ratio. In other words, the intensity of \( h+k = 2n+1 \) type reflections are drastically increased with increasing Al content, while the intensity of \( h+k = 2n \) type reflections remains practically unchanged. This again suggests that the excess Al atoms may be occupying the Ti-sites.

Table 3.6  The ratios \((I_{x}/I_{y})\) of different x-ray reflection from single crystals of two different compositions Ti54at%Al and Ti56at%Al. The percentage deviation is defined as \( \frac{y-x}{x} \times 100\% \)

<table>
<thead>
<tr>
<th>Ratio</th>
<th>Ratio for Composition: Ti54at%Al</th>
<th>Ratio for Composition: Ti56at%Al</th>
<th>Percentage Deviation</th>
</tr>
</thead>
<tbody>
<tr>
<td>((I_{(010)}/I_{(020)}))</td>
<td>0.0065</td>
<td>0.0129</td>
<td>98.5</td>
</tr>
<tr>
<td>((I_{(101)}/I_{(202)}))</td>
<td>0.0020</td>
<td>0.0044</td>
<td>120.0</td>
</tr>
<tr>
<td>((I_{(110)}/I_{(220)}))</td>
<td>0.1483</td>
<td>0.1425</td>
<td>-3.9</td>
</tr>
</tbody>
</table>
3. 5. 1: Debye-Waller (D-W) Factor Determination

Under kinematic conditions, the relationship between the averaged integrated intensity \( I_g \) and the isotropic D-W factors can be given by,

\[
I_g = k|F_g^C|^2
\]  

(3.3a)

where \( k \) is the scale factor and \( F_g^C \) is the theoretical structure factor. The equation for \( F_g^C \) of a compound with multiply occupied sites can be given by,

\[
F_g^C = \sum_j \left[ \sum_i f_i^{X}(s)x_i\exp(-B_is^2) \right] \exp(-2\pi i g \cdot r_j)
\]  

(3.3b)

where \( s \) is equal to \( \sin\theta/\lambda \), \( B_i \) is the D-W factor of the \( i \)-th element, and \( x_i \) is the atom fraction of element ‘\( i \)’ in the ‘\( j \)’th site. For stoichiometric and perfectly ordered TiAl, \( x_i = 1 \) and \( F_g^C \) is given by,

\[
F_g^C = f_n^{X}(s)[1 + \exp(-\pi i [h+k])\exp(-B_n s^2) + f_{Al}^{X}(s)[\exp(-\pi i [h+l]) + \exp(-\pi i [k+l])\exp(-B_{Al} s^2)]
\]  

(3.3c)

where \( B_{Ti} \) and \( B_{Al} \) are the isotropic D-W factors of Ti and Al respectively, and \( f_n^{X}(s) \) and \( f_{Al}^{X}(s) \) are calculated free-atomic scattering factors. Equation (3.3) neglects bonding effects and assumes atoms to be isotropically vibrating inside the crystal. This assumption regarding bonding is valid, especially in the case of high-order structure factors, because the main contribution to the scattering process comes from the non-bonding core electrons close to the vibrating nucleus. In addition, the deviation of experimental structure factors from those calculated
from the neutral atom scattering factor is usually smaller than the experimental errors in the measured integrated intensities. The D-W factors are usually determined using equations (3.3a, b and c) by Wilson plot or least-squares technique.

3. 5. 1. 1 : Wilson Plot Method

Equations (3.3b) and (3.3c) can be simplified by simply assuming equal D-W factors for all the sites within the unit cell \( (B = B_i = B_{Al} = B_{Ti}) \)

\[
F^C_g = \left\{ \sum_j \left[ \sum_i f_i^X (s)x_i \right] \exp(-2\pi ig \cdot r_j) \right\} \exp(-Bs^2) \quad (3.4a)
\]

\[
F^C_g = \left\{ \frac{f_{Ti}^X (s)[1+\exp(-\pi i[hl+kl])]}{f_{Al}^X (s)[\exp(-\pi i[hl+kl]) + \exp(-\pi i[kl])]} \right\} \exp(-Bs^2) \quad (3.4b)
\]

The terms enclosed in the flower brackets in the above equations are simply equal to the theoretically calculated static structure factors, \( F^C(s) \). Then

\[
F^C_g = F^C(s) \exp(-Bs^2) \quad (3.5)
\]

The relation, given in (3.6), between the integrated intensities and the equi-isotropic D-W factor, \( B \), \((B = B_{Ti} = B_{Al})\) can be obtained by combining equations (3.5) and (3.3a) and taking logarithm on both sides. Thus
\[
\ln \left( \frac{I_g}{\left| F_C(s) \right|^2} \right) = \ln K - 2Bs^2
\] (3.6)

From equation (3.6), the equi-isotropic D-W factor \( B \) is one half of the negative slope of the line obtained by plotting the left hand side of the equation (3.6) as a function of \( s^2 \). This method of D-W factor determination is known as the Wilson plot method [Willis and Pryor (1975)]. The Wilson plots for the x-ray data from Ti-54at%Al are given in Figures 3.6 and 3.7. The free-atom static structure factors \( (F_C(s)) \) were calculated using the Hartree-Fock free-atom scattering factors after Doyle and Turner (1968). A D-W factor value of 0.67 Å\(^2\) was obtained from Figure 3.6 when the excess Al atoms were assumed to be randomly distributed in the interstitial sites. The equi-isotropic D-W factor value increased to 0.70 Å\(^2\) when the excess Al atoms were assumed to be distributed on one of the Ti-sites (Figure 3.7). In both cases, the equi-D-W factor \( B \) values are in between the D-W factor of pure Ti, \( B_{Ti}^{ref} = 0.56 \) Å\(^2\) (average of c-component and a-component [Schoening and Witt (1965)]), and pure Al, \( B_{Al}^{ref} = 0.85 \) Å\(^2\) [Dingle and Medlin (1972)].
Figure 3.6: The Wilson plot of x-ray integrated intensity data from a Ti54at%Al. All the excess Al atoms are assumed to be randomly distributed in the interstitial locations.
Figure 3.7: The Wilson plot of x-ray integrated intensity data from a Ti54at%Al. All the excess Al atoms are assumed to be distributed on one of the Ti-sites.
3.5.1.2: Least-squares Refinement

A number of standard software packages, such as XTAL-3.0 [Hall (1990)], RFINE-90 [Finger and Prince (1975)], and VALRAY [Stewart and Spackman (1983)], can be used to perform least-squares refinement of the D-W factors and the structure factors. VALRAY and RFINE-90 were used for least-squares refinement of the x-ray data in the present study. The software VALRAY is designed for the refinement of the electronic population of individual sites in the unit cell and the software RFINE-90 is designed for the refinement of the site occupancy of individual atoms in the unit cell. During the refinement process both programs minimize the difference between the model structure factor and the scaled experimental structure factor obtained from the integrated intensity data, by a nonlinear least-squares method. The agreement between theory and experiment is measured in terms of the R\textsubscript{IS}-factor defined by

\[ R_{IS} = \sum_{i=1}^{n} \omega_i \left[ k \sqrt{I_i} - F_i^e \right]^2 \] (3.7)

where \( n \) is the number of structure factors included in the least-square refinement, \( k \) is the scale factor, \( F_i^e \) is the structure factor calculated from a model and \( \omega_i \) is the weighting factor. In most refinement problems, \( \omega_i \) is equal to \( 1/\sigma(I) \), where \( \sigma(I) \) is the estimated standard deviation in the experimental integrated intensity.

A preliminary least-squares refinement of the isotropic D-W factors was performed by assuming that all of the excess Al atoms were distributed on interstitial locations. The program VALRAY was used to perform the refinement (the kind assistance of Dr. Downs, Department of Geological Sciences, The Ohio
State University being acknowledged). The theoretical structure factors were calculated by using the Hartree-Fock spherical free-atom model, refined values of the D-W factors are tabulated in Table 3.7.

Table 3.7 Results of the D-W factor refinement using the software package VALRAY. R\(_{\text{B}}\)-factor = 3.29%.

<table>
<thead>
<tr>
<th>Atoms</th>
<th>Isotropic D-W factors B in Å(^2)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ti1</td>
<td>0.66±0.01</td>
</tr>
<tr>
<td>Ti2</td>
<td>0.95±0.01</td>
</tr>
<tr>
<td>Al</td>
<td>0.65±0.01</td>
</tr>
</tbody>
</table>

In spite of the fact that the site symmetry of the Ti1-sites and the Ti2-sites in the (1,1,0) unit cell of TiAl are equivalent, the VALRAY refinement gave two entirely different D-W factors for the Ti-atoms, as seen from Table 3.7. More interestingly, when the electronic population of one of the Ti2-sites was included as a variable, the D-W factors of the Ti sites approached each other, however, the electronic population of the Ti2 sites was lowered from 22 to about 18. The decrease in the electron population of the Ti2 sites supports our preliminary analysis that this Ti-site is partially occupied by the lower atomic number element Al. In such a case, more accurate values of the D-W factors could be obtained if the population of Al atoms in Ti-sites was refined. Since VALRAY did not have the facility to perform site occupancy refinement, the program RFINE-90 was chosen for this purpose.
In the RFINE-90 refinement, the theoretical free-atom structure factors of TiAl were calculated from the Hartree-Fock free atom scattering factors [Doyle and Turner (1968)]. The dispersion correction was obtained from Kissel and Pratt (1990). Since the x-ray peaks of reflections with Miller indices satisfying the condition \( h+k = 2n+1 \) were very weak and diffuse, the integrated intensities of these reflections could not be measured accurately. For of these reasons these reflections were not included in the refinement. In addition, in order to ensure a statistically sound analysis, only 102 reflections satisfying the condition \( \frac{I_g}{\sigma(I_g)} > 4 \) were included in the site occupancy and the D-W factor refinement.

A preliminary RFINE-90 refinement was performed by using the small unit cell structure for TiAl and considering three different cases. In the first case (Table 3.8), all excess Al atoms were assumed to be distributed on interstitial locations, in the second case (Table 3.9) all the excess Al atoms were assumed to be evenly distributed on the Ti-sites, and in the third case (Table 3.10) the excess Al atoms were assumed to be partitioned between the Ti-sites and interstitial sites. From Tables 3.8-3.10, it can be seen that the agreement between the model and the experimental structure factors (lower R_{pF}-Factor) is better in the case of multiple site occupancy for Ti-sites (cases 2 and 3), rather than for single site occupancy. However, the D-W factor of Ti (B_{Ti}) is greater than the D-W factor of Al (B_{Al}) in all of the three cases. This is, by intuition, unacceptable because the atomic weight of Ti is much higher than that of Al and the Debye-Waller factor of elemental titanium is 34% lower than that of elemental aluminum. This implies that the assumption about the equal distribution of Al atoms in the Ti1 and Ti2 sites in TiAl is probably incorrect and it is be necessary to consider some other way to model the distribution of excess Al atoms.
Table 3.8: **Case-1**: Stoichiometric TiAl with single occupancy on Ti and Al sites.

<table>
<thead>
<tr>
<th>Atom</th>
<th>Occupancy</th>
<th>B Å²</th>
<th>R_{ls}-factor (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ti</td>
<td>1</td>
<td>0.845±0.006</td>
<td>2.6</td>
</tr>
<tr>
<td>Al</td>
<td>1</td>
<td>0.691±0.012</td>
<td></td>
</tr>
</tbody>
</table>

Table 3.9: **Case-2**: Non-stoichiometric TiAl with single occupancy in Al-site and fixed multiple occupancy in Ti-site.

<table>
<thead>
<tr>
<th>Atom</th>
<th>Occupancy</th>
<th>B Å²</th>
<th>R_{ls}-factor (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ti</td>
<td>0.92Ti +0.08Al</td>
<td>0.774±0.004</td>
<td>1.8</td>
</tr>
<tr>
<td>Al</td>
<td>1</td>
<td>0.700±0.008</td>
<td></td>
</tr>
</tbody>
</table>

Table 3.10: **Case-3**: Non-stoichiometric TiAl with single occupancy in Al-site and variable multiple occupancy at Ti-site.

<table>
<thead>
<tr>
<th>Atom</th>
<th>Occupancy</th>
<th>B Å²</th>
<th>R_{ls}-factor (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ti</td>
<td>0.913Ti +0.087Al</td>
<td>0.769±0.007</td>
<td>1.7</td>
</tr>
<tr>
<td>Al</td>
<td>1</td>
<td>0.701±0.008</td>
<td></td>
</tr>
</tbody>
</table>

The pattern for the distribution of excess Al atoms in TiAl may be predicted by using the results from the theoretical phase stability and bond energy calculations. Results from a number of theoretical electron density and phase stability calculations [(e.g.) Asta et al. (1992), Hong et al. (1990), Fu (1990), Fu and Yoo (1990) and Morinaga et al. (1990)] indicates that p-d type bonding is more stable than p-p or d-d bonding in TiAl. If this prediction is true, then the excess Al atoms in TiAl lattice will be distributed in Ti-sites in a manner which maximizes
the $p$-$d$ type bonding. The $p$-$d$ type bonding is maximized when the excess Al atoms are distributed partially on one of the Ti-sites of the $L1_0$ structure rather than evenly on both of the Ti-sites. This type of distribution would result in a short-range $L1_2$ type (TiAl$_3$) ordering within the TiAl lattice. The problem with this notion is that the pure titanium tri-aluminide (TiAl$_3$) phase is stabilized in the $D0_{22}$ structure rather than the $L1_2$ structure. However, it should also be noted that the phase stability calculations performed by FLAPW method [Fu (1990)] and Full Potential LMTO method [Asta et al. (1992)] predict that the total energy difference between $D0_{22}$ and $L1_2$ structures at equilibrium volume is comparatively low ($\approx 2$ mRy/atom), and a metastable $L1_2$ type TiAl$_3$ compound is highly feasible. In fact, the $L1_2$ structure of TiAl$_3$ has been stabilized by partially replacing some Al atoms with Cu, Ni or Fe [Kumar (1993)]. In consideration of these factors, it is appropriate to confine excess Al atoms to one of the Ti sites, rather than to distribute them among both Ti-sites. In such a case, the excess Al atoms in the Ti54at%Al can be either constrained to one of the Ti-sites in the $L1_0$ unit cell (compositional constraint) or partitioned between one of the Ti-sites (in the $L1_0$ unit cell) and the interstitial sites.

The results tabulated in Tables 3.11 and 3.12 have been obtained by constraining all the excess Al atoms to one of the Ti sites. In other words, the Ti1-site has a fixed composition of 16at% Al and 84at% Ti, while the Ti2-site and the Al-sites contain 100% of their respective atoms. An R-factor value of 1.6 was obtained in this refinement; this value being lower than the R-factor values obtained in Tables 3.8 through 3.10. D-W factors of 0.46 and 0.71 Å$^2$ for singly occupied Ti2 and Al sites are comparable to the pure elemental values of 0.56 and 0.85 Å$^2$. 
However, the D-W factor of the multiply occupied Ti1-site is 1.28 Å², is which is unreasonably high. It should be noted here that in RFINE-90, the D-W factor of a particular site is refined irrespective of the composition of that site. Here the scattering factor of multiply occupied sites is defined by the linear combination of the scattering factors of the different elements occupying that site. Such a representation is rather questionable, but unavoidable in the absence of any other established procedure to represent multiple occupancy. In essence, this suggests that the error in the definition of the scattering factors of the multiply occupied site is the most probable cause for unusually higher D-W factors for that site. The results of the refinement when all of the excess Al is moved to the Ti2-site, instead of Ti1-site, are is shown in Table 3.12. Identical results obtained in Tables 3.11 and 3.12 indicate the Ti1 and Ti2 sites are completely interchangeable, which is in complete agreement with the site symmetry associated with these atoms.

Results obtained by least-squares refinement, considering the distribution of excess Al atoms between one of the Ti-sites and the interstitial sites is shown in Tables 3.13 and 3.14. Here the composition of one of the Ti-sites is also included as a variable during the R-factor minimization. Results indicate that the R-factor remains unchanged in this case, but there is only 11.7at% of Al instead of the 16at% in the multiply occupied Ti-site. The D-W factors obtained from the singly occupied method are \( B_{Al} = 0.71 \text{ Å}^2 \) and \( B_{Ti} = 0.42 \text{ Å}^2 \). These D-W factors are again comparable with the elemental values of 0.85 Å² and 0.56 Å² for pure Al and Ti respectively, however, the D-W factor of the multiply occupied Ti-site is again unreasonably high.
Table 3.11: Debye-Waller Factors of non-stoichiometric TiAl assuming single occupancy in Al-site and fixed multiple occupancy in Ti1-site: Compositional Constraint.

<table>
<thead>
<tr>
<th>Atom</th>
<th>Occupancy</th>
<th>( B \ \text{Å}^2 )</th>
<th>( R_{1S} )-factor (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ti1</td>
<td>0.84Ti+0.16Al</td>
<td>1.208±0.062</td>
<td></td>
</tr>
<tr>
<td>Ti2</td>
<td>1</td>
<td>0.460±0.035</td>
<td>1.6</td>
</tr>
<tr>
<td>Al</td>
<td>1</td>
<td>0.705±0.007</td>
<td></td>
</tr>
</tbody>
</table>

Table 3.12: Debye-Waller Factors of non-stoichiometric TiAl assuming single occupancy in Al-site and fixed multiple occupancy in Ti2-site: Compositional Constraint.

<table>
<thead>
<tr>
<th>Atom</th>
<th>Occupancy</th>
<th>( B \ \text{Å}^2 )</th>
<th>( R_{1S} )-factor (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ti1</td>
<td>1</td>
<td>0.460±0.035</td>
<td></td>
</tr>
<tr>
<td>Ti2</td>
<td>0.84Ti+0.16Al</td>
<td>1.208±0.062</td>
<td>1.6</td>
</tr>
<tr>
<td>Al</td>
<td>1</td>
<td>0.705±0.007</td>
<td></td>
</tr>
</tbody>
</table>

Table 3.13: Debye-Waller Factors of non-stoichiometric TiAl assuming single occupancy in Al-site and variable multiple occupancy in Ti2-site.

<table>
<thead>
<tr>
<th>Atom</th>
<th>Occupancy</th>
<th>( B \ \text{Å}^2 )</th>
<th>( R_{1S} )-factor (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ti1</td>
<td>1</td>
<td>0.417±0.032</td>
<td></td>
</tr>
<tr>
<td>Ti2</td>
<td>0.883Ti+0.117Al</td>
<td>1.343±0.075</td>
<td>1.6</td>
</tr>
<tr>
<td>Al</td>
<td>1</td>
<td>0.705±0.007</td>
<td></td>
</tr>
</tbody>
</table>
Table 3.14: Debye-Waller Factors of non-stoichiometric TiAl assuming single occupancy in Al-site and variable multiple occupancy in Ti1-site.

<table>
<thead>
<tr>
<th>Atom</th>
<th>Occupancy</th>
<th>B Å²</th>
<th>R_{is}-factor (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ti1</td>
<td>0.883Ti+0.117Al</td>
<td>1.343±0.075</td>
<td></td>
</tr>
<tr>
<td>Ti2</td>
<td>1</td>
<td>0.417±0.032</td>
<td>1.6</td>
</tr>
<tr>
<td>Al</td>
<td>1</td>
<td>0.705±0.007</td>
<td></td>
</tr>
</tbody>
</table>

3.6: Summary and Conclusion

A summary of the Debye-Waller factor values obtained by different approaches (after neglecting those approaches giving $B_{Ti} < B_{Al}$) using the x-ray diffraction data from Ti54at%Al is given in Table 3.15. The D-W factor values of Al is almost constant, and ranges from 0.67 to 0.71 Å². However, the D-W factor of Ti ranges from 0.42 to 0.70 Å² (without considering the multiply occupied site). The D-W factor combination of $B_{Al}$ = 0.71 Å² and $B_{Ti}$ = 0.42 Å², obtained for the singly occupied Ti and Al sites in TiAl by site occupancy refinement, falls in the reasonable range, however, the large D-W factor value obtained for the multiply occupied Ti-site (1.34 Å²) is not well understood.

The problems due to the site occupancies may be avoided if a stoichiometric single crystal of TiAl (Ti-50at%Al) were used. Because of the reasons discussed in Chapter-II it is practically impossible to obtain such a stoichiometric single crystal. In essence, the current status of the x-ray results indicates that it will be difficult to determine accurate values of D-W factors or structure factors of TiAl by using the
single crystal x-ray diffraction. From the point of view of understanding the
deformation behavior, x-ray work provides the first experimental evidence
regarding the site occupancy of Al atoms in Ti-sites. This might offer an explanation
for the increased brittleness of TiAl with increasing Al content [Greenberg et al.
(1992)]. More detailed results will have to be obtained by site occupancy refinement
of x-ray data collected from series of TiAl single crystal specimens with increasing
Al content.

Table 3.15: Summary of the D-W factors obtained by using the x-ray data from
Ti54at%Al single crystal by different data analysis procedures

<table>
<thead>
<tr>
<th>Method</th>
<th>Occupancy of excess Al</th>
<th>$B_{Al}$ Å$^2$</th>
<th>$B_{Ti1}$ Å$^2$</th>
<th>$B_{Ti2}$ Å$^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wilson Plot</td>
<td>Confined to interstitial sites</td>
<td>0.67</td>
<td>0.67</td>
<td>0.67</td>
</tr>
<tr>
<td>Wilson Plot</td>
<td>Confined to titanium sites</td>
<td>0.70</td>
<td>0.70</td>
<td>0.70</td>
</tr>
<tr>
<td>Least squares:</td>
<td>Confined to the Ti2-site</td>
<td>0.71</td>
<td>0.46</td>
<td>1.21</td>
</tr>
<tr>
<td>RFINE-90</td>
<td>Partitioned between the Ti-2 and interstitial sites</td>
<td>0.71</td>
<td>0.42</td>
<td>1.34</td>
</tr>
</tbody>
</table>
CHAPTER - IV

CONVERGENT BEAM ELECTRON DIFFRACTION (CBED) METHOD

4.1: Introduction

The energy-filtered CBED rocking curve method has been previously applied to the determination of the structure factors in GaAs by Zuo, Spence and O'Keefe (1988), in BeO by Zuo et al. (1993) and in MgO by Zuo and Spence (1991). In this thesis, a Philips EM420 Transmission Electron Microscope (TEM) coupled with a custom built energy-filtering attachment, which permitted single electron counting, was used for CBED experiments. The theoretical pattern matching was performed using FORTRAN programs developed as a part of this thesis at The Ohio State University. This is the first attempt to refine structure factors using this particular experimental setup and the fitting routines. At first, the experimental plan involved an attempt to refine structure factors in TiAl directly, and the results of this preliminary work have been presented elsewhere [Swaminathan (1993)]. In subsequent work, it was found that it was not possible to obtain results with sufficient precision, i.e. less than 0.1%, from samples of differing thickness. It is imperative that such consistency of results be obtained, and it is of interest to consider the reasons why such precision was not obtained. Thus, in the first instance, the sample used was nominally Ti-52Al, and there is the uncertainty concerning the actual composition, and then the problem of knowing the detailed
site occupancy of the compound when off-stoichiometry. Secondly, as explained in the previous chapter, there is some ambiguity over the exact values of the D-W factors which should be used for simulations of diffraction in this compound. For these reasons, until a suitable TiAl sample is produced, corresponding to exactly stoichiometric composition, it is unlikely that realistic determinations of the structure factors of TiAl may be made. Moreover, it is essential that the technique to be used to perform refinement of the structure factors be robust and accurate. Therefore, the emphasis of the present study has been placed on the development of experimental and theoretical techniques which will permit such refinement to be done. For such research, it is important to make measurements on a sample which has been well characterized by other techniques, and hence Si has been chosen. Therefore, the primary goal of this chapter has been to establish the experimental data collection and the fitting procedures involved in the structure factor refinement, by using the silicon 220 reflection as a test case. A detailed description of the theoretical background, experimental procedure, data analysis and fitting procedure involved in structure factor determination by the CBED rocking curve method will be presented in this chapter.

4.2: Background

The aim of this section is to explain the theoretical background behind the computer programs for the calculation of CBED patterns and the matching of rocking curves. The computer programs are based on the quantum mechanical formulation of the dynamical theory of electron diffraction for calculating of the CBED patterns and rocking curves. Excellent reviews of this theory can be found in Hirsch et al. (1965), Reimer (1984), Humphreys and Bithell (1992), and Spence and Zuo (1992). A short
review of relevant portions of the Bloch wave formulation, and the principles behind its application to structure factor determination, will be presented in this section.

4.2.1: Dynamical Theory of Electron Diffraction

4.2.1.1: Bloch Wave Formulation and Solution of the Schrödinger Equation

The wavefunction of electrons moving with energy $eE_0$ ($E_0$ is the accelerating voltage) in vacuum can be assumed to behave as a plane wave with a wavefunction $\psi(r)$ defined by

$$\psi(r) = \exp(2\pi i K \cdot r) \quad (4.1)$$

where $K$ is the electron wavevector defined by the following equation

$$|K|^2 = K^2 = \frac{2me}{\hbar^2}E_0 \quad (4.2)$$

Similarly, the wavefunction of an electron moving within a crystal with a periodic potential, $V(r)$, can be represented using the Bloch theorem as below

$$\psi(r) = C(r)\exp(2\pi i k \cdot r) \quad (4.3)$$

where $k$ is the Bloch wavevector and $C(r)$ is the Bloch function. The Bloch function, $C(r)$, has the periodicity of the lattice and hence equation (4.3) may be expanded as a Fourier series based on the reciprocal lattice to give

$$\psi(r) = \sum_g C_g \exp(2\pi i [k + g] \cdot r) \quad (4.4)$$
where $C_g$ is known as the Bloch wave coefficient.

In the Bloch wave formulation of the dynamical theory, the total wave function, $\Psi(r)$, of fast electrons within the crystal is represented by the linear superposition of Bloch waves given by (4.4)

$$\Psi(r) = \sum_j \alpha_j \psi_j(r) = \sum_j \alpha_j \sum g C_g^{(j)} \exp(2\pi i l \cdot k^{(j)} + g \cdot l \cdot r)$$  \hspace{1cm} (4.5)

where $\alpha_j$ is the excitation amplitude of the $j$'th Bloch wave. The $\alpha_j$'s are obtained by applying the proper boundary conditions for continuity of the total wavefunction, the $C_g^{(j)}$'s can be estimated using the Schrödinger equation.

The Schrödinger equation for the wavefunction, $\Psi(r)$, of the fast electron moving within the periodic crystal potential $V(r)$ is given by

$$\nabla^2 \Psi(r) + \frac{8\pi^2 m e^2}{\hbar^2} [E_0 + V(r)] \Psi(r) = 0$$  \hspace{1cm} (4.6)

where $m$ is the relativistic mass of the fast electron and $E_0$ is the accelerating voltage. The periodic crystal potential ($V(r)$) can be expanded as a Fourier series in the reciprocal lattice as

$$V(r) = \sum_{-\infty}^{+\infty} V_h \exp(2\pi i h \cdot r) = \frac{2me^2}{\hbar^2} \sum_{-\infty}^{+\infty} U_h \exp(2\pi i h \cdot r)$$  \hspace{1cm} (4.7)

where $V_h$ is a complex constant known as the coefficient of the crystal potential and $U_h$ is the coefficient of the modified crystal potential, $U(r)$; $U_h$ is related to $V_h$
by

\[ U_h = \frac{2m_\text{el}}{\hbar^2} V_h \]  

(4.8)

The accelerating voltage \((E_0)\) is related to the mean electron wavevector \((K)\) by the following equation

\[ |K| = K = \frac{2m_\text{el}}{\hbar^2} (E_0 + V_o) = \frac{2m_\text{el}}{\hbar^2} (E_o) \]  

(4.9)

where \(V_o\) is the mean crystal potential. The mean crystal potential, \(V_o\), is of the order of 10 V and is negligible in the case of high energy electron diffraction where, \(E_o\) is of the order of \(10^5\) volts. Substitution of equations (4.5), (4.7) and (4.9) in the Schrödinger equation (4.6) yields the following standard set of dispersion equations for high energy electron diffraction

\[ \left[ K^2 - (k^{(j)} + g_1)^2 \right] C_g^{(j)} + \sum_{h \neq g_1} U_{g_1 - h} C_h^{(j)} = 0 \]

\[ \left[ K^2 - (k^{(j)} + g_2)^2 \right] C_g^{(j)} + \sum_{h \neq g_2} U_{g_2 - h} C_h^{(j)} = 0 \]

\[ \left[ K^2 - (k^{(j)} + g_n)^2 \right] C_g^{(j)} + \sum_{h \neq g_n} U_{g_n - h} C_h^{(j)} = 0 \]  

(4.10)
Figure 4.1: The two beam dispersion surface and reflecting sphere construction.
In general there is one such equation for each value of \( g \), or for each reflection, considered. An exact solution for the Schrödinger equation is obtained if \( n = \infty \), however, usually an approximate solution is obtained by considering a finite number of appropriate reflections. The equation for \( K^2 - (k^{(j)} + g)^2 \) can be obtained by using the dispersion surface for the two beam case, as shown in Figure 4.1 [Hirsch et al. (1965)]. From the geometry of Figure 4.1 we have

\[
\begin{align*}
K^2 - (k^{(j)})^2 &= K^2 - (K + \gamma^{(j)} \cos \theta_0)^2 = -2K\gamma^{(j)} \cos \theta_0 \\
K^2 - (k^{(j)} + g)^2 &= K^2 - (K + (\gamma^{(j)} - s_g) \cos \theta_g)^2 \\
&= 2K(s_g - \gamma^{(j)} \cos \theta_g)
\end{align*}
\]  
(4.11)

where

\[
s_g = K^2 - (K + g)^2
\]  
(4.12)

When \( \theta_g \) is small, \( \cos \theta_g \) is approximately equal to one. Using this approximation and by substituting equations (4.11) and (4.12) in (4.10) we obtain

\[
\left[ K^2 - (K + g_i)^2 \right] C^{(j)}_{g_i} + \sum_{h \neq g_i} U_{g_i} C^{(j)}_h = 2K\gamma^{(j)} C^{(j)}_{g_i} : i = 1, ..., n
\]  
(4.13)

These sets of linear equations can be written in the matrix form as below
\[
\frac{1}{2K} \begin{bmatrix}
K^2 - (K + g_1)^2 & U_{g_1 - g_2} & \cdots & U_{g_1 - g_n} \\
U_{g_2 - g_1} & K^2 - (K + g_2)^2 & \cdots & \cdots \\
\cdots & \cdots & \cdots & \cdots \\
U_{g_n - g_1} & \cdots & \cdots & \cdots \end{bmatrix} \begin{bmatrix}
C_{1}^{(j)} \\
C_{2}^{(j)} \\
\cdots \\
C_{n}^{(j)} \end{bmatrix} = \gamma^{(j)} \begin{bmatrix}
C_{1}^{(j)} \\
C_{2}^{(j)} \\
\cdots \\
C_{n}^{(j)} \end{bmatrix}
\]

or as

\[
AC_g = \gamma^{(j)} C_g
\]

where \( A \) is a \( n \times n \) matrix (corresponding to \( n \) beams), referred to as a ‘structure matrix’. The off-diagonal elements of the structure matrix are given by \( A_{gh} \)

\[
A_{gh} = \frac{U_{g-h}}{2K} = \frac{1}{2\xi_{g-h}}
\]

where \( \xi_g \) is the extinction distance, and the diagonal elements \( A_{gg} \) are given by

\[
A_{gg} = K^2 - (K + g)^2
\]

Equation (4.15) is an eigenvalue problem. The structure matrix, \( A \), has \( n \) eigenvalues \( \gamma^{(j)} : (j = 1, \ldots, n) \), with accompanying eigenvectors \( C_g^{(j)} : (g = g_1, g_2, \ldots, g_n) \). Since the coefficient of the crystal potential, \( V_g \), and hence \( U_g \) are complex constants, the structure matrix \( A \) is in general complex. However, for a centrosymmetric crystal without absorption, the phase term of \( U_g \) is either 0 or \( \pi \); as a result \( U_g = U_g \) so that \( A \) will be real symmetric. Since the crystal structures of TiAl and Si both have a center of symmetry, work in later sections of this thesis focuses only on centrosymmetric crystals.
4.2.1.2: Determination of Diffracted intensities

The diffracted amplitudes, \( \Phi(t) \), can be determined using \( \gamma^{(j)} \) and \( C_g^{(j)} \) and the boundary conditions for the continuity of the wavefunction given in (4.5) \( (\psi(r) | z \text{ and } \nabla \psi(r) | z) \) are continuous across the top and bottom surfaces of the thin foil [Humphreys and Bithell (1992)]. Hence

\[
\Phi(t) = \begin{bmatrix} \phi_0(t) \\ \phi_g(t) \end{bmatrix} = C \begin{bmatrix} \exp(2\pi i \gamma^{(1)} t) & 0 & 0 & 0 \\ 0 & \exp(2\pi i \gamma^{(2)} t) & 0 & 0 \end{bmatrix} C^{-1} \begin{bmatrix} \phi_0(0) \\ \phi_g(0) \end{bmatrix}
\] (4.18)

where \( C \) is a matrix formed by eigenvectors as shown below

\[
C = \begin{bmatrix} C_0^1 & C_0^2 & \cdots & C_0^n \\ C_g^1 & C_g^2 & \cdots & C_g^n \\ \cdots & \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots & \cdots \end{bmatrix}
\] (4.19)

\( \phi_i(0) \) is the incident amplitude of the \( i \)’th beam, and \( \phi_i(t) \) is the diffracted amplitude of the \( i \)’th beam at thickness \( t \). In a normal electron diffraction experiment \( \phi_0(0) = 1 \) and \( \phi_{iso}(0) = 0 \). The diffracted intensity of the beam \( g \) at thickness \( t \) is given by

\[
I_g(K) = |\phi_g(t)|^2
\] (4.20)
4.2.1.3: Phenomenological treatment of Absorption

In dynamical theory, absorption due to inelastic scattering is usually treated by the phenomenological theory of absorption [Yoshioka (1957)]. In this theory, an imaginary potential \( V'(r) \) is added to the imaginary part of \( V(r) \). Since \( V'(r) \) is associated with the crystal lattice it can be expanded as a Fourier series similar to equation (4.7). In such a case we have

\[
V(r) \rightarrow V(r) + iV'(r) \tag{4.21}
\]

\[
V_g \rightarrow V_g + iV_g' \tag{4.22}
\]

\[
U_g \rightarrow U_g + iU_g' \tag{4.23}
\]

\[
\frac{1}{\xi_g} \rightarrow \frac{1}{\xi_g} + i\frac{1}{\xi_g} \tag{4.24}
\]

where \( V_g' \) is known as the coefficient of the absorption potential. The absorption potential \( V'(r) \) and \( V_g' \) are a function of the accelerating voltage and temperature. In electron diffraction three sources, namely phonon scattering \((\Delta E_o \sim 0.02 \text{ eV})\), plasmon scattering \((\Delta E_o \sim 20 \text{ eV})\), and single electron excitation \((\sim 200 \text{ eV})\), contribute to \( V'(r) \). By employing energy filtering \((< 5 \text{ eV})\), all contributions from inelastic scattering except phonon scattering may be minimized. In such a case, \( V_g' \) is dominated by phonon scattering, and so its contribution can be determined by the Einstein phonon model [Bird and King (1990)].
In a centrosymmetric crystal with absorption, the imaginary part of $U_g$ is not zero. This means that the structure matrix, $A$, is complex but still symmetric. The diffracted intensities can be obtained by diagonalizing the complex symmetric structure matrix. However, diagonalization of the complex symmetric matrix is computationally more intensive when compared to a real matrix of the same dimension. Because of this reason, a perturbation theory [Metherell (1975)] method described below can be used to incorporate absorption.

The Schrödinger equation for a fast electron moving inside a complex potential $(V(r) + iV'(r))$ yields a complex Bloch wave vector given by

$$k^{(j)} \rightarrow k^{(j)} + i q^{(j)}$$

(4.25)

and the total wavefunction, $\Psi(r)$, of the fast electron within the crystal is rewritten as

$$\Psi(r) = \sum_j \sum_g \alpha^{(j)} \sum \chi^{(j)} \exp(2\pi i [k^{(j)} + g \cdot r]) \exp(-2\pi q^{(j)} \cdot r)$$

(4.26)

In other words, each Bloch wave is exponentially attenuated as it propagates through the crystal. The equation (4.27) for $q^{(j)}$ [Humphreys and Bithell (1992)] is derived starting from (4.26) and following a similar procedure to that described for a real crystal potential.

$$q^{(j)} = \mathbf{C}^{(j)\ast} \begin{bmatrix} U_0' & U_{-g}' & 0 & 0 \\ U_g' & U_0' & 0 & 0 \\ 0 & 0 & 0 & \bullet \\ 0 & 0 & 0 & \bullet \end{bmatrix} \mathbf{C}^{(j)}$$

(4.27)
Figure 4.2: (a) A schematic representation of CBED in systematic orientation. The incident converged cone of electrons can be considered to contain a finite number of incident beams (K). Then for each intensity point (Io(K)) in the bright field disk there is a conjugate intensity point (Ig(K)) in each dark field disk. (b) Different parts of the experimental Bright Field (BF) and Dark Field (DF) rocking curves that are affected by t, ξg and ξ'g.
where
\[
C^{(j)} = \begin{bmatrix}
C^{(j)}_a \\
\bullet \\
\bullet \\
\bullet
\end{bmatrix}
\]
(4.28)

and \(C^{(j)*}\) is the complex conjugate of \(C^{(j)}\). The amplitude (\(\Phi(t)\)) is then estimated by substituting
\[
\gamma^{(j)} \rightarrow \gamma^{(j)}_{\text{real}} + i q^{(j)}
\]
(4.29)
in equation (4.18), where \(\gamma^{(j)}_{\text{real}}\) is the eigenvalue of the real symmetric structure matrix of a centrosymmetric crystal without any absorption (note: \(\gamma^{(j)}_{\text{real}}\) may be complex).

4.2.2: Structure Factor Determination

A schematic diagram of a CBED experiment is shown in Figure 4.2(a). A converged electron probe incident on the specimen may be assumed to consist of a specified number of electron beams, each corresponding to a specific incident beam direction. For every incident beam direction there is one intensity point in the (000) disk and a conjugate intensity point in all of the dark field disks. From equation (4.15), it can be seen that the diagonal elements of the structure matrix are different for different \(K\)'s. This suggests that the calculation of a CBED pattern requires determination of eigenvalues and eigenvectors of the structure matrix \(A\) for each incident beam wavevector '\(K\)'s. Once \(A\) is diagonalized, the intensity \(I_0(K)\) in the (000) CBED disk beam and the corresponding conjugate intensity point \(I_g(K)\) in any other \('g\)'th dark field CBED disk can be determined using equation (4.20).
In the CBED rocking curve method of structure factor determination, the crystal is oriented with respect to the electron beam in such a way that the reflection \((g)\) of interest is at the Bragg condition. A CBED pattern recorded with the crystal oriented at the systematic orientation is shown in Figure 2.2. In this orientation, the reflection at the Bragg condition and reflections in that systematic row are strongly excited in comparison to the off-systematic reflections. In fact, in most cases the reflection \((g)\) at Bragg condition is strongly excited with respect to the other reflections in the systematic row. This means the amplitudes, \(\phi_i(t)\), given by (4.18) of all the higher order systematic and off-systematic reflections are low when compared to \(\phi_o(t)\) and \(\phi_g(t)\). Because of this reason it can be assumed that the errors in the structure factor values of the higher order systematic and off-systematic reflections included in the Bloch wave calculations will not drastically affect the result, and so theoretically calculated free-atom values of the structure factor can be used in Bloch wave calculations for these reflections. A more detailed explanations, using the Bethe potential can be found in Electron Microdiffraction by Spence and Zuo (1992).

In the systematic orientation, those parameters associated with the '0'\(^{th}\) and 'g'\(^{th}\) beams will be very sensitive to the intensity distribution in the these CBED disks. Three important parameters that determine the intensity distribution in the bright field and dark field disks are the foil thickness, \(t\), the extinction distance, \(\xi_g\), and the absorption part of the extinction distance, \(\xi_g\). These parameters were fitted to simulate the theoretical rocking curve and thus create a match with the experimental rocking curve. The thickness of the thin foil enters the calculation through the equation (4.18), \(\xi_g\) enters the calculation through (4.14), and \(\xi_g\) enters
the calculation through (4.14) or (4.29), depending on whether the complex matrix formulation or perturbation theory is used to treat absorption. Different parts of the bright field and dark field rocking curves that are sensitive to \( t, \xi_g \) and \( \xi_g' \) are shown in Figure 4.2(b).

4.3: FORTRAN Codes for CBED Calculations

As discussed in the previous section, the theoretical calculation of CBED patterns and matching of rocking curves requires setting up a \( n \)-dimensional structure matrix, diagonalization of this matrix, and finally calculating the diffraction intensity from the eigenvalues and eigenvectors for each incident beam direction in the converging probe. Structured computer programs have been written as a part of this thesis to perform these steps. These programs can handle a tetragonal unit cell with a center of symmetry and can be readily altered for any other centrosymmetric crystal structure by making simple changes in specific subroutines. The input parameters required by the program are the Debye-Waller factors, unit cell parameters, coefficients of free atom scattering factors [Doyle and Turner (1968)], the orientation of the crystal with respect to the electron beam and the diameter of the CBED disk as a fraction of the \( g \)-vector.

Reflections that have to be included in the Bloch wave calculations are either given as inputs to the program or automatically determined by using the routine beams by specifying the number of reflections to be included in the calculation as zero. The principle behind the automatic beam selection process is shown in Figure 4.3. The subroutine beams would then include all the reflections with a \( \xi_{sh} \) (\( \xi_{sh} \) is the extinction distance of the reflection calculated from free atom scattering factors...
Figure 4.3: Ewald's sphere construction. The selection procedure used in the routine **beams** to include reflections in the Bloch wave calculations. All those beams which have a $\xi_{sh}<w$ are included in the calculation. The limiting value of the dynamical interaction parameter $w$ is given as an input to the calculation.
and $s_h$ is the deviation parameter for the given beam direction) value less than the input value of $w$. The extinction distances, $\xi_h$, required for calculating the real part of the off-diagonal elements of the structure matrix were calculated by using the input free-atom scattering factor values. The complex part of the off-diagonal elements (absorption potential $V_g$) for CBED are calculated from the D-W factors by using Einstein's phonon model [Bird and King (1990)]. In all of the calculations standard mathematical routines from IMSL\textsuperscript{2} FORTRAN libraries were used for diagonalization of real symmetric and complex symmetric matrices. A description of the FORTRAN programs for simple calculation of CBED pattern (SYSPAT) and rocking curve matching (FIT) are given in the following sections.

4.3.1: SYSPAT - Program for calculation of CBED patterns

The program SYSPAT is used to calculate the two-dimensional intensities in the bright field CBED disk and a specified number of dark field CBED disks. If the exact values of the lattice parameters are known, the position of the HOLZ lines can be used to determine either the exact crystal orientation or the accelerating voltage. The flow chart of the program SYSPAT for the calculation of the systematic CBED patterns is shown in Figure 4.4. In addition to the input parameters mentioned earlier, the thickness, $t$, of the sample and values of experimental extinction distance, $\xi_g$, as well as the absorption part of the extinction distance, $\xi'_g$, are also read in from the input file by the main program. The subroutine \texttt{diffrt} is used to set up the diffraction calculation. The off-diagonal elements of the structure matrix, $A$, are set up by calling subroutine \texttt{off\_diagonal}. As seen from equation (4.14), the off-diagonal elements are independent of the incident beam

\textsuperscript{2} IMSL FORTRAN library is distributed by Visual Numerics, Houston, TX.
Figure 4.4: Flow chart of the program **SYSPAT**. The FORTRAN program **SYSPAT** is used to calculate CBED patterns in zone axis and systematic orientations.
direction and need to be set up only once in the calculations. The subroutine `upoff_diagonal` is used to update the off-diagonal elements with any experimental \( \xi_g \) and \( \xi'_g \), input to the program. The function of subroutine `fcn` is to set up the diagonal elements of the structure matrix for every incident beam direction, diagonalize the matrix and determine corresponding intensities in each of the disks to be calculated. The FORTRAN code for SYSPAT, additional subroutines, example input file and example output files are given in Appendices A, B, C and I.

The two-dimensional CBED patterns calculated using SYSPAT are mainly used for the determination of the orientation and the accelerating voltage. These are not high precision calculations, so in SYSPAT, anomalous absorption was treated by perturbation theory, i.e. only a real symmetric matrix is diagonalized. The calculated CBED intensities are then written into a file which can be plotted using proper plotting routines.

4.3.1: **FIT - Program for matching the rocking curves**

The purpose of the program FIT is to match the theoretically calculated rocking curve to the experimentally measured one by varying the \( \xi_g \) and \( \xi'_g \) of the reflection at Bragg condition, the thickness, \( t \), of the sample and the background intensities underneath the bright field and the dark field rocking curves. The thickness, \( t \), of the sample and \( \xi_g \) are sensitive to the spacing and the height of the dark field fringes. The absorption part of the extinction distance, \( \xi'_g \), is derived from the asymmetry of the bright field rocking curve. The sources of the background intensity are instrumental noise and phonon scattering.
The basic structure of the program FIT is same as that of the SYSPAT except that it calculates only an one-dimensional pattern (rocking curve) and has an automatic fitting routine to enable high precision structure factor refinement. The complex matrix treatment of absorption is usually recommended for this purpose, however, diagonalization of the complex matrix is a time consuming process when compared to a real matrix required by perturbation theory. Also there is no quantitative evidence that there is improvement in the precision of the structure factors by using the complex matrix formulation. In order to verify whether the complex matrix formulation is an important requirement, different codes, incorporating the complex matrix formulation and perturbation theory treatment of absorption, have been written for matching the rocking curves. The codes incorporating the complex matrix formulation and perturbation theory treatment of absorption along with example input and output are given in Appendices D through I. The flowcharts of the program FIT and fitting subroutine are shown in Figures 4.5 and 4.6 respectively. Even though some of the subroutines have the same name as those used in SYSPAT, they are slightly modified to include the fitting routines.

In FIT, the theoretical rocking curve is automatically matched to the experimental rocking curve by minimizing quantity $\chi^2$ defined by

$$\chi^2 = \frac{1}{m \cdot n} \sum_{j=1}^{m} \sum_{i=1}^{n} \left( \text{factor} \cdot \frac{I_{\text{calc}(i,j)} - I_{\text{expt}(i,j)}}{I_{\text{expt}(i,j)}} \right)^2$$

(4.30)

where 'm' is the total number of rocking curves fitted, 'n' is the number of points
Read INPUT

If $nbeams=0$

Determine the number of variables

Call `diffrc`:
Set off-diagonal elements of dynamical matrix
Call `off_diagonal`
Return

Set up the variables for minimization

Minimization routine
Call `BCONF`
Minimize $\chi^2$
Minimized

Output minimized variables and calculated rocking curve
Stop

Figure 4.5: Flow chart of the program FIT. The program FIT is used to match rocking curves obtained from CBED patterns recorded at systematic orientation.
Figure 4.6: The flow chart of the procedures involved in the fitting routine used in the program FIT.
included in the rocking curve of each beam, $I_{\text{calc}(i,j)}$ is the experimentally calculated rocking curve intensity of the $'i'$th point in the $'j'$th beam, $I_{\text{expt}(i,j)}$ is the experimentally measured intensity of the $'i'$th point in the $'j'$th beam and $\text{factor}$ is defined by

$$\text{factor} = \frac{I_{\text{expt}(x,y)}}{I_{\text{calc}(x,y)}}$$

(4.31)

where $x$ is the normalizing point on the $'y'$th beam in the systematic row. The values of $x$ and $y$ are given as an input to the program. The purpose of multiplying by $\text{factor}$ is to normalize the experimental intensities, so that they can be compared to the theoretical intensities. Experience indicates that any point in the rocking curve which has approximately 50% of the maximum intensity can be used as a normalizing point. The choice of the normalizing intensity is very important when the first guess of the fitted parameters are far off from the best fit. The subroutine $\text{fcn}$ is used to calculate the rocking curve intensities and the $\chi^2$ minimization. A quasi-Newton finite difference gradient method of minimization [Dennis and Schnabel (1983)], incorporated in the subroutine BCONF of the IMSL² FORTRAN library, was used to carry out the minimization.

4. 4 : Experimental CBED Data Collection and Analysis

4. 4. 1 : Energy-Filtered CBED Intensity Measurement

Energy-filtered CBED patterns were recorded using a Philips EM420 transmission/scanning transmission electron microscope (TEM/STEM) with a LaB₆ electron source and interfaced to a combination of commercial and custom
built ancillary equipment in the Electron Microscopy Center of Argonne National Laboratory. The commercial equipment used in this study includes: a Gatan Model 607 serial electron energy loss spectrometer, a Gatan Model 636-N double tilt cooling holder, a Gatan Model 652 double tilt heating holder and an EDAX 9900 data acquisition/computer system.

Energy-filtered diffracted intensities were measured by digitized serial scanning of CBED patterns over an entrance aperture into an uniform field sector magnet electron spectrometer as illustrated Figure 4.7. The function of the uniform field sector magnet is to transfer the electron beam from the entrance aperture, containing elastically and inelastically scattered electrons, into an energy-loss spectrum. The energy resolution limit of the spectrometer employed during these measurements was less than 0.75 eV and was limited by stray AC magnetic fields in the room. A mechanically adjustable post-spectrometer slit is then used to filter inelastically scattered electrons in the energy loss spectrum. The energy resolution of the measured diffraction intensity is determined by the width of the post spectrometer slit. This was preset to measure only those electrons which were scattered within an energy window of 5 eV of the incident electron energy of ~ 120 KV. This resolution was sufficient for most of the inelastically scattered electrons short of phonon scattering. Also inelastically scattered electrons within the energy window of 5 eV were taken into account by including background intensities as one of the variables in the calculation.

The electron intensity emerging from the post-spectrometer slit is incident on to a photo multiplier tube (PMT), where it was processed either by analog to digital converter (ADC) or by direct pulse counting. The zero-level electron count
Figure 4.7: A schematic diagram of the experimental setup used to collect energy filtered CBED patterns (courtesy Dr. Zaluzec). This system is located at the Argonne National Laboratories, Argonne, IL.
resulting from instrumental noise was determined by mechanically closing the entrance aperture (by placing a viewing screen over the entrance aperture), and measuring the intensity. When the signal is processed by ADC, the zero-level is usually set to around 50 counts/sec by adjusting the gain of the amplifier. The dynamic range of the EELS system, when the signal is processed by ADC, is from 0 to 4000 counts/sec⁻¹, and in the direct pulse counting mode from 0-32000 counts/sec⁻¹.

In the serial scanning mode, the CBED pattern is scanned line by line with each line divided into a number of steps. The total time required to record a pattern depends on the number of lines scanned in, the number of steps in each line, the time period spent on counting electrons at each step and the time required to process the signal. The maximum time period that might be spent on recording a single CBED pattern depends on the overall system stability and the degree of contamination of the specimen surface. The former is governed by the combined stabilities of the probe position, beam energy and beam current, and the latter depends on the specimen temperature. The overall system stability was insured for the longest data acquisition times (upto 20 minutes) by recording matching data in duplicate runs. Examples of filtered and unfiltered CBED patterns from a silicon crystal oriented with 220 at the Bragg condition, along with the rocking curves taken along the line AB are shown in Figures 4.8 and 4.9. The inelastic background in the unfiltered pattern is clearly visible in both the unfiltered pattern and under the rocking curve in Figure 4.9. Experimental measurements were recorded at specimen stage temperatures of -165 °C, the actual specimen temperature is being expected to differ from these values by no more than ± 10 °C due to local electron beam heating.
Figure 4.8: Filtered silicon 220 systematic CBED pattern along with the bright field and dark field rocking curves. The pattern was recorded at a stage temperature of 108 K. The pattern is sharp and inelastic background is very low (only the phonon scattering).
Figure 4.9: Unfiltered silicon 220 systematic CBED pattern along the bright field and dark field rocking curves. The pattern was recorded at a stage temperature of 108 K. Inelastic background can be seen in the pattern and underneath the rocking curves.
4. 4. 2 : Angular Resolution of the Entrance Aperture

The angular resolution of all the diffraction measurements is controlled by the size of the entrance aperture (Figure 4.7) convoluted with the camera length of the diffraction lenses. Angular resolution may be improved by increasing the camera length or decreasing the size of the entrance aperture or both. For a given specimen thickness, both of these actions would reduce the number of electrons passing through the entrance aperture and, as a result, would decrease the signal to noise ratio. More optimum counting statistics may be obtained either by increasing the strength of the incident beam, or by decreasing the spot size (by reducing the field strength of the C1 lens), or by increasing the counting period at each step. The former would increase the probe size and a larger area from the specimen would be illuminated. The latter would increase the total time required to record a pattern and is limited by overall system stability and contamination problems. After careful analysis of preliminary results, an entrance aperture size of 200 µm and camera length of 600 mm with a limited angular resolution of 0.33 mrad were chosen for the CBED experiments.

The angular resolution effect is pronounced when there are sharply varying features in the diffraction pattern. In the systematic case, as pointed out in Figure 4.2(b), the angular spacing between the dark field fringes, and the relative intensity of dark field fringes, are used for structure factor refinement. The angular spacing between the dark field fringes of any specific reflection is a function of the crystal structure and the thickness of the sample. The average angular spacing between the fringes decreases as the thickness increases, and the fringes will not be resolved properly when the average angular spacing of fringes is less than the angular
resolution (0.33 mrad). An example showing a dark field rocking curve of the 113 reflection from a thick sample (236 nm) of TiAl, together with the curve fitted using the systematic reflections, is shown in Figure 4.10. It can be seen from Figure 4.9 that the minima of the theoretical curve reach the constant background level while those of the experimental curve do not. There are approximately 16 sharp intensity variations (8 maxima and 8 minima) in this dark field rocking curve over an angular range of \( \approx 5 \) mrad. In other words, on an average there is a sharp intensity variation for every 0.31 mrad. This is less than 0.33 mrad, the angular resolution of the detection system. This problem could be overcome by recording CBED patterns from a thinner region and thus reducing the number of fringes in the given angular range. An 113 dark field rocking curve from a thinner (67 nm) region of the same TiAl specimen along with its theoretical fit is shown in Figure 4.11.

The aperture effect can be simulated perfectly by integrating calculated two dimensional CBED intensities, corresponding to an infinite number of incident beams, within the solid angle enclosed by the 200 \( \mu \)m entrance aperture. Since it is impossible to calculate intensity for an infinite number of incident beam directions, CBED intensities for a finite number of incident beam direction (points) can be calculated and the region in between points can be linearly interpolated. When the angular spacing between the neighboring points of the rocking curve is greater than 0.33 mrad then an array of CBED intensities (represented by small circles Figure 4.12) has to be calculated individually for each pixel in the CBED pattern, as shown in Figure 4.12. These intensities can be summed to get the intensity in the experimental CBED pattern.
Figure 4.10: Experimental and fitted 113 dark field rocking curves from a thick (236 nm) region of TiAl foil. There is approximately one sharp intensity variation for every 0.31 mrad. This is less than the angular resolution of the detecting system (0.33 mrad), as a result, the minima in the experimental curve do not reach the background level.
Figure 4.11: Experimental and fitted 113 dark field rocking curves from a thin (67 nm) region of TiAl foil. The fringes are clearly resolved in the experimental curve and minima in both experimental and theoretical curves almost reach the background level.
Figure 4.12: The intensity of each pixel in the CBED pattern is given by the integrated sum of diffracted electrons enclosed within a solid angle defined by the diameter of the aperture and the camera length. To simulate this situation a finite array of intensity points (represented by small circles) can be calculated within this solid angle and then summed over to get the total intensity. In the systematic case, the intensity is constant along Y direction, so that only those intensity points along the diameter of the aperture and parallel to g vector have to be computed.
The intensity integration method described above requires the computationally intensive calculation of two dimensional CBED intensities. However, for rocking curve matching, the characteristics of the systematic CBED patterns can be used to reduce the computational requirement. At the systematic orientation, CBED disks have fringes parallel to the Y-direction, as shown in Figure 4.12, and if the scan direction is parallel to the g-vector, the intensity along the Y-direction can be assumed to be a constant within the aperture. Under this condition only a row of \( m \) intensity points along the diameter of the aperture shown in Figure 4.12(b) and parallel to the g-vector has to be computed. If CBED intensities for \( m \) directions are calculated over a radius, \( r \), of the circular aperture, then the structure matrix has to be diagonalized \( 2m+1 \) times for calculating each intensity point in the rocking curve. In a typical calculation, when \( m=5 \), the structure matrix has to be diagonalized only 11 times by this approximation, when compared to 79 times \( (\pi m^2) \) in the two dimensional calculation.

The principle behind the numerical integration of the calculated intensities over the circular aperture is shown in Figure 4.13. If two intensity points \( I_k \) and \( I_i \) are calculated at points \( r_k \) and \( r_i \) respectively, then the number of electrons (intensity) passing through the angular range \( r_k \) and \( r_i \) would be given by the shaded volume element. The total intensity of a pixel is given by sum of all such volume of elements within the aperture. This principle was applied to fit the 113 rocking curve shown in Figure 4.10. The fitted result is shown in Figure 4.14. The calculated curve in this case agrees well with the experimental curve.
Figure 4.13: The principle behind the aperture correction. In the systematic case, intensity is constant along the Y direction. Under this situation, if two intensity points $I_k$ and $I_i$ are calculated at $r_k$ and $r_i$ respectively, the number of electrons (intensity) that passed through the angular range $r_k$ and $r_i$ would then be given by the shaded elemental volume. The total number of electrons that passed through the angular range of the aperture per unit time would be given by the sum of all such volume elements.
Figure 4.14: Experimental and fitted dark field 113 rocking curves from a thick (236 nm) region of TiAl foil. An aperture correction has been applied to the calculated rocking curve. There is much better agreement between the calculated and experimental curve than the one shown in Figure 4.10.
4.4.3: Data Collection and Processing

A rocking curve for structure factor refinement can be obtained from a CBED pattern recorded at any systematic orientation along the pair of Kikuchi lines corresponding to the reflection of interest. However, the fringes in a systematic CBED patterns will be perturbed to a different extents at different orientations by the HOLZ line intensities. Even though HOLZ lines are useful for pinpointing exactly the orientation, they introduce sharp intensity variations into the rocking curve and therefore exacerbate the problem associated with the angular resolution. In addition, CBED intensities at HOLZ line perturbations cannot be accurately simulated because the extinction distances of the HOLZ reflections are not known accurately. The extent of HOLZ perturbations at a given orientation can be reduced by decreasing the foil thickness, but the minimum foil thickness is usually limited by the minimum number of fringes (at least four) required for fitting the thickness of the sample. Because of these reasons systematic patterns for structure factor refinement have to be recorded at orientations with minimum off-systematic interactions and a reasonable inter-fringe angular spacing.

A number of energy-filtered CBED patterns from a single crystal Si foil kindly supplied by Dr. Maher of the North Carolina State University, Raleigh, NC were recorded with the sample oriented in each case, at the Bragg condition (along the Kikuchi lines) for the 220 reflection. The patterns were recorded with the camera length set to 600 mm and accelerating voltage of the microscope set to 120 KV. To aid orientation determination, with every energy-filtered CBED pattern an unfiltered low camera length pattern, including the nearest low-order zone, was either recorded on a photographic film or scanned into the computer through the entrance aperture.
The approximate orientation of the crystal was determined first by using
the Kikuchi lines in the low-camera length unfiltered pattern. The procedure
involved measurement of the distance Y (Figure 4.15) of the closest low index
pole from the line joining the centers of the bright field and dark field disks in
units of the length of g-vector (equal to 2θ degrees). In the above case, the Ewald’s
sphere is assumed to be flat over the reciprocal distance Y. This assumption is
valid because the radius of the Ewald’s sphere at 120 KV, is large and only a few
degrees of tilt is included in the distance Y. A more accurate orientation,
corresponding to the rocking curve used in the calculation, can be derived by
calculating a series of systematic CBED patterns using the program SYSPAT and
exactly matching the HOLZ pattern. An example of a calculated CBED systematic
pattern along with an experimental pattern at the same orientation is shown in
Figures 4.16 (a and b). Once the direction corresponding to point P (Figure 4.15)
is known the beam direction, B, corresponding to the center of the bright field
disk O can be obtained by simply tilting the direction (P) corresponding to point
P (Figure 4.15) by a Bragg angle θ B degrees along the -g direction. This might not
be acceptable in cases when the crystal is not oriented exactly at Bragg condition.
In such cases, the rocking curves would be asymmetric and the beam direction
corresponding to the center of the bright field disk can be obtained by adding a Δg
to K ( K is mean wavevector antiparallel to B) during the fitting process. The
program FIT has the provision to automatically fit the magnitude of Δg in terms
of fraction of pixels in a rocking curve.

Rocking curves for structure factor refinement were obtained by taking a
profile through the CBED disks as shown in Figure 4.16c. The distance between
Figure 4.15: A schematic diagram of a low camera length CBED pattern recorded at the systematic orientation. An approximate orientation can be determined by measuring $Y$ in terms of lengths of $g$-vector as shown above.
Figure 4.16: Accurate orientation determination by matching the HOLZ lines in the experimental CBED pattern to the calculated CBED pattern. (a) CBED pattern calculated using the program 
SYSPAT. (b) Experimental CBED pattern. (c) Intensity profile taken at orientation [1117 1117 240].
the centers of the Bright Field (BF) and the Dark Field (DF) disks (1/d-spacing) was determined in terms of pixels by superimposing the BF and DF rocking curves such that the minima in the BF rocking curve coincide with the maxima in the DF rocking curve or vice versa as shown in Figure 4.17. In many cases it is difficult to position the condenser aperture C2 symmetrically about the Bragg condition of the DF disk. In such cases, the number of data points on either side of the Bragg condition of the DF rocking curve will not be equal. The DF rocking curve can be made perfectly symmetrical by simultaneously cutting-off points in the DF and the BF rocking curve when they are overlaid on each other as shown in Figure 4.17. The rocking curves are then written on to a formatted file so that they can be read by the program FIT during the structure factor refinement.

4. 5 : Voltage Determination

Accurate values of the relativistic electron wavelength are required for the calculation of CBED patterns and the rocking curves. The relativistic electron wavelength is determined from the accelerating voltage. The accelerating voltage is usually determined by matching the relative positions of the deficient HOLZ lines in the zero disk of the CBED pattern obtained from the kinematical theory of electron diffraction. In the kinematical approximation, the HOLZ line positions are predicted by the Bragg's law where the locus of the HOLZ line is the locus of the Bragg condition. The geometric construction of the HOLZ line is shown in Figure 4.18. In three dimensions, for any given g-vector, all the incident beam directions (K) and the diffracted beam directions (K') satisfying the Bragg condition form two separate cones as shown in Figure 4.18. The HOLZ lines are then given by the projection of the HOLZ pair on the observation plane. The perimeter of the
Figure 4.17: The number of pixels separating any intensity point $I_0(K)$ in the (000) disk and the conjugate point $I_g(K)$ in Figure 4.2(a) is equal to the length of the $g$ in number of pixels. It can be determined by first overlaying the Bright Field (BF) and Dark Field (DF) rocking curve such that the minima in the BF rocking curve coincide with maxima of the DF rocking curve and vice versa, as shown above. Then the length of $g$ in pixels is the number of pixels separating $I_{BF}$ and $I_{DF}$ in Figure 4.16c.
Figure 4.18: Construction of HOLZ lines using the kinematical theory. The incident and diffracted beam directions are given by $K$ and $K'$. The locus of the Bragg condition forms two cones, corresponding to $K$ and $K'$. 
base circle of the cone is very large when compared to the length of a typical $g$-vector and in high energy electron diffraction (small $\lambda$ and $|K|=1/\lambda$) only a small portion of the cone is observed. In such a case the HOLZ lines can be approximated to a pair of straight lines perpendicular to the $g$-vector. The position of the HOLZ line is defined in Figure 4.18 by $\alpha$, the angle between the direction perpendicular to the observation plane (in most cases, the zone axis) and the incident beam direction, $K$. From Figure 4.18

$$\alpha = \theta_B - \beta = \sin^{-1}\left(\frac{g\lambda}{2}\right) - \sin^{-1}\left(\frac{g_z}{g}\right)$$ (4.32)

where $g = |g| = \frac{1}{d_g}$, $d_g$ is the d-spacing of plane $g$, $g_z$ is magnitude of the z-component of the $g$ vector defined in the Figure 4.18, and $\theta_B$ is the Bragg angle and $\lambda$ is the relativistic wavelength. The relativistic wavelength, $\lambda$, is given by Spence and Zuo (1992)

$$\lambda = \frac{0.38783}{\sqrt{E_o(1 + 0.97845 \times 10^{-3} E_o)}} \text{Å}$$ (4.33)

where $E_o$ is the accelerating voltage in KV. In equation (4.32), the term that is affected directly by change in voltage is $\theta_B$. The relationship between the wavelength and $\theta_B$ is given by Bragg's law

$$\sin \theta_B = \frac{g}{2} \lambda$$ (4.34)
When $\theta_B$ is small

\[ \theta_B = \frac{g}{2} \lambda \]  \hspace{1cm} (4.35)

differentiating (4.35) we have

\[ d\theta_B = \frac{1}{2d_g} d\lambda \]  \hspace{1cm} (4.36)

For reflections in higher order Laue zones, $d_g << 1$, and so a small change in $\lambda$ would lead to large variations in $\theta_B$ and thus in the relative position of the HOLZ line.

In practice, it is impossible to monitor the movement of a single HOLZ line, but it is much easier to monitor a triangle formed by three HOLZ lines (which are not in the same zone) [Hoier (1969)]. Unfortunately, this method requires determination of the exact orientation where a HOLZ triangle is sensitive to the nominal accelerating voltage of the microscope. To avoid this, Fitzgerald and Johnson (1984) have calibrated the accelerating voltage, using a Si specimen, in terms of ratios of distances between points of intersection between specific HOLZ lines for different ranges of KV's. In this thesis both of these kinematical methods have been used for accelerating voltage determination.

At a given accelerating voltage, the relative positions of the HOLZ lines may change with foil thickness due to dynamical interactions (e.g. zero layer of the [111] zone axis in silicon as demonstrated by Mansfield, Bird and Saunders (1992)). This means that when there is a strong dynamical interaction, kinematical methods
may not be accurate. This problem is avoided if HOLZ lines from a weak zone axis, such as the [013] zone of silicon, are matched. However, it is difficult to predict the extent of the dynamical interaction. Because of this, the accelerating voltage determined by kinematical methods should also be verified using dynamical theory calculations.

4.5.1: Voltage Determination by Measuring Ratio of Distances

An electron-transparent single crystal specimen of semiconductor grade silicon, with the foil normal close to the [111] zone-axis and prepared by ion-beam thinning was used for the voltage determination experiments. The microscope was aligned properly and the voltage was set nominally at 120 KV. A number of Kikuchi patterns for different exposure timings, at three orientations close to three equivalent <13 14 15> type zones were recorded on a photographic film. These orientations are marked by filled circles in the Kikuchi map shown in Figure 4.19. Enlarged prints were made on F5 grade photographic paper using the best negative obtained at each of the three equivalent <13 14 15> zones and the prints were then digitized into a computer file. The distances OA, OB, SX and SY shown in Figure 4.20(a) were measured using the profile tool in IMAGE\textsuperscript{3} software for Macintosh. An example of such a digitized Kikuchi pattern is shown in Figure 4.20(b). The polynomial fits [Fitzgerald and Johnson (1984)] for \( E_0 \) in terms ratios \( R_1 = \frac{OA}{OB} \) and \( R_2 = \frac{SX}{SY} \) in the voltage range of 115 to 125 KV, given in equations (4.37) and (4.38), were then used to determine \( E_0 \).

\[
E_0 = 126.317 - 24.572 \ R_1 \tag{4.37}
\]

\[
E_0 = 126.318 - 21.710 \ R_2 \tag{4.38}
\]

\textsuperscript{3} IMAGE is a public domain software distributed by National Institutes of Health, Washington, DC.
Figure 4.19: Silicon [111]-zone axis Kikuchi pattern. Open and closed circles indicate the equivalent orientations (approximately $<13\,14\,15>$) used to record the diffraction pattern for accelerating voltage determination.
Figure 4.20: (a) An expanded view of the kinematic Kikuchi (HOLZ) pattern close to the $<13\ 14\ 15>$ orientation. (b) A sample of the experimental Kikuchi pattern recorded on a photographic film at 98 K. The pattern has been printed on photographic paper and digitized.
The ratios of distances $R_1$ and $R_2$ along with the accelerating voltages obtained from them for three equivalent orientations are shown in Table 4.1.

**Table 4.1:** The ratios of distances $R_1 = OA/OB$ and $R_2 = SX/SY$, and the corresponding values of accelerating voltage $E_0$ in KV have been tabulated below.

<table>
<thead>
<tr>
<th>Equivalent Orientation</th>
<th>$R_1 = OA/OB$</th>
<th>$E_0$ in KV</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.27869</td>
<td>119.47</td>
</tr>
<tr>
<td>2</td>
<td>0.27778</td>
<td>119.49</td>
</tr>
<tr>
<td>3</td>
<td>0.27778</td>
<td>119.49</td>
</tr>
<tr>
<td>$R_2 = SX/SY$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>0.30841</td>
<td>119.62i</td>
</tr>
<tr>
<td>2</td>
<td>0.32407</td>
<td>119.495</td>
</tr>
<tr>
<td>3</td>
<td>0.31429</td>
<td>119.282</td>
</tr>
<tr>
<td>Averaged $E_0$</td>
<td></td>
<td>119.48±0.2</td>
</tr>
</tbody>
</table>
4. 5. 2 : HOLZ pattern matching using the kinematical and the dynamical calculations

Uncertainties in the kinematical HOLZ pattern calculation for voltage determination have been pointed out by Lin, Bird and Vincent (1989). It is necessary to verify the accelerating voltage, measured by kinematical calculations in the previous section, by dynamical theory calculations. The HOLZ pattern in the (0 0 0) disk of [0 1 3] zone of Si was chosen for this purpose. The shape of the triangle formed by \{1 5 13\}, \{2 4 14\} and \{3 7 11\} HOLZ deficiency lines in the zero disk of [0 1 3] zone-axis CBED pattern is sensitive to $E_0$ around the voltage range of 120 KV, and was used for determination of the accelerating voltage using kinematical and dynamical theory calculations.

The kinematical theory calculation of the HOLZ pattern was carried out using Desktop Microscopist$^4$. Various parameters used in Desktop Microscopist to calculate the HOLZ pattern are listed in Table 4.2.

Table 4.2 : Parameters used for the kinematical theory calculation of the HOLZ pattern using Desktop Microscopist.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lattice Parameter (a)</td>
<td>0.54294 nm [Shah and Strumanis (1972)]</td>
</tr>
<tr>
<td>Dynamic Range</td>
<td>4.5</td>
</tr>
<tr>
<td>Convergence angle ($\alpha$)</td>
<td>0.25 Degrees</td>
</tr>
<tr>
<td>Camera Constant</td>
<td>300 mmÅ</td>
</tr>
<tr>
<td>Laue zones included</td>
<td>1, 2 and 3</td>
</tr>
<tr>
<td>Zone axis</td>
<td>[0 1 3]</td>
</tr>
</tbody>
</table>

$^4$ Desktop Microscopist is distributed by Virtual Laboratories, Albuquerque, NM 87191.
Figure 4.21: [0 1 3]-silicon HOLZ pattern calculated using the kinematical theory of electron diffraction for three different accelerating voltages along with the experimental (000) CBED disk. The movement of the intersection of (14 4 2) and (13 5 1) HOLZ lines with respect to (11 7 3) HOLZ line is indicated by points A and B. The possible range of positions of point B in the experimental pattern is shown by the flower bracket. The accelerating voltage is then deduced to be 119.45±0.15 KV by comparing this range with the calculated patterns.
The energy-filtered (000) disk of the [013] zone-axis CBED pattern, recorded at a stage temperature of 101 K, along with the HOLZ pattern in the (000) disk of the [013] zone-axis, calculated for various voltages by kinematical theory of electron diffraction, are shown in Figure 4.21. The segment formed by the intersection of \((14 \overline{4} 2)\) and \((13 5 \overline{1})\) HOLZ deficiency lines with the \((11 7 3)\) HOLZ deficiency line is marked by A, and the intersection of \((14 4 2)\) and \((13 5 \overline{1})\) HOLZ deficiency lines is marked by B. These points coincide with each other at an accelerating voltage of 119.3 KV and move apart as the accelerating voltage is increased to 119.45 and 119.6 KV. The HOLZ lines in the experimental patterns are relatively thick and it is difficult to locate exactly the intersection B, but the range over which the intersection is likely to be located is shown by "flower" brackets. A comparison of this range with the kinematical theory calculations gives an accelerating voltage of 119.45±0.15 KV.

This result was further verified by recalculating the [013] zone-axis pattern of silicon using the dynamical theory of electron diffraction. A total of 110 beams selected by using the routine \texttt{beams} (\(\xi_{hs} \leq 315\)) were included in the three dimensional Bloch wave calculations. The coefficient of the crystal potentials \((V_g)\) for these beams were determined from the free atom scattering factors calculated by Doyle and Turner (1968). The absorption part of the crystal potentials, \(V_g'\), were calculated, using the Debye-Waller factor of 0.2357 Å², by the Einstein phonon model [Bird and King (1990)]. The calculated and experimental patterns are shown in Figure 4.22, along with the experimental pattern. In spite of the dynamical interaction, points A and B behave almost exactly in the kinematical calculation. A comparison of the experimental pattern to three theoretically calculated patterns
Figure 4.22: [0 1 3]-silicon HOLZ pattern calculated using the dynamical theory of electron diffraction for three different accelerating voltages along with the experimental (000) CBED disk. The movement of the intersection of (14 4 2) and (13 5 1) HOLZ lines with respect to (11 7 3) HOLZ line is indicated by points A and B. The possible range of positions of point B in the experimental pattern is shown by the flower bracket in the experimental pattern. The accelerating voltage is then deduced to be 119.45±0.15 KV by comparing this range with the calculated patterns.
indicates that the pattern calculated using an accelerating voltage of 119.45 KV is in best agreement with the experimental pattern. This means all three methods of voltage determinations agree very well with each other. So an accelerating voltage of 119.45±0.2 KV was used in structure factor refinement calculations.

4. 6 : Results and Discussion

4. 6. 1 : Silicon 220 Structure Factor Refinement : A test case

In Bloch wave calculations, the anomalous absorption can be treated by perturbation theory or simply by adding the absorption potential to the imaginary part of the coefficient of the crystal potential. For a centrosymmetric crystal, the perturbation treatment will result in a real symmetric structure matrix, whereas, adding the absorption potential to the imaginary part of the crystal potential will result in a complex symmetric matrix. The complex matrix formulation has been recommended for high precision structure factor measurements by Hewat and Humphreys (1974), Zuo, Spence and Hoier (1989) and Humphreys and Bithell (1992). However, the complex matrix formulation also drastically increases the computational requirement (CPU time).

The error in the structure factors measured by the CBED method is a function of extraneous parameters, such as the accuracy of Debye-Waller factors, the angular resolution of the detection system, the number of beams included in the calculation, the accuracy of the absorption coefficient, and the error in the measured accelerating voltage. There is no quantitative evidence that the precision achieved by using the complex matrix formulation would outweigh extraneous errors resulting from these factors. The best way to determine whether the complex matrix formulation
is necessary is to compare the results of complex matrix to the results obtained through the perturbation theory by using a standard test case.

The experimental rocking curves can be simulated exactly only if the limitation due to the angular resolution of the detection system is properly modeled. Similar to the complex matrix formulation, incorporating the aperture correction procedure described in section 4.4.2 in fitting calculations will drastically increase the computational requirement (time). The effect of the aperture can be considerably reduced by avoiding HOLZ lines and maintaining a reasonably large inter-fringe angular spacing. However, it is still necessary to verify if the aperture correction makes any difference in the final value of the structure factor and its precision.

Si was chosen as a standard test material to verify whether the complex matrix formulation and aperture corrections are required for structure factor refinement. The main advantage of Si is that it is one of the most widely studied materials and its structure factors have been already measured by a number of different techniques. The 220 structure factor (F_{220}) was chosen for the test structure factor refinement because, within the available range of the foil thickness, the angle of convergence in TEM, and orientations of the thin foil, the 220 CBED disk had low HOLZ interactions and a reasonable number of fringes (4 to 8) in the given angular range (6 mrad). A summary of the past experimental measurements of F_{220} of Si together with the experimental techniques and references, are given in Table 4.3. The experimental ξ_{220} values range between 82.5 to 83.6 nm, however as discussed in Chapter II, the x-ray Pendellösung method is by far the most accurate method for x-ray structure factor measurement, and the values of ξ_{220} measured by this method range between 82.9 to 83.4 nm.
Table 4.3: Experimental $F_{220}$ values available in the literature. Parameters used for conversion of electron quantities to x-ray quantities and vice versa are given in Table 4.4.

<table>
<thead>
<tr>
<th>Technique</th>
<th>$V_{220}$</th>
<th>$\xi_{220}$</th>
<th>$F_{220}$</th>
<th>$F_{220}^S$ electrons</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>x-ray - <em>Pendellösung</em></td>
<td>4.375</td>
<td>82.88</td>
<td>67.068</td>
<td>69.208±0.007</td>
<td>Aldred and Hart (1973)</td>
</tr>
<tr>
<td>x-ray - <em>Pendellösung</em></td>
<td>4.368</td>
<td>83.00</td>
<td>67.130</td>
<td>69.272±0.005</td>
<td>Spackman (1986)</td>
</tr>
<tr>
<td>x-ray - <em>Pendellösung</em></td>
<td>4.346</td>
<td>83.35</td>
<td>67.343</td>
<td>69.492±0.001</td>
<td>Saka and Kato (1986)</td>
</tr>
<tr>
<td>CBED-rocking curve</td>
<td>4.396</td>
<td>82.48</td>
<td>66.866</td>
<td>69.000±0.280</td>
<td>Voss, Lehmpfuhl and Smith (1980)</td>
</tr>
<tr>
<td>IKL</td>
<td>4.335</td>
<td>83.65</td>
<td>67.448</td>
<td>69.600±0.08</td>
<td>Terasaki, Watenabe and Gionnes (1979)</td>
</tr>
</tbody>
</table>

* [*Spackman (1986)] has re-analyzed [Aldred and Hart (1973)]'s x-ray *Pendellösung* data.

The rocking curves for structure factor refinement were extracted from five different CBED patterns, corresponding to five different thicknesses, recorded at room temperature (291 K) by the procedure described in section 4.4.3. The orientation of the crystal, corresponding to the central point of the bright field rocking curve, was determined accurately by the procedure described in section 4.4.3. Experimental rocking curves were then matched to the theoretically calculated ones by varying the thickness, $\xi_{220}$, $\xi_{220}$, and the backgrounds under the BF and DF rocking curves. The absorption part of the extinction distance, $\xi_g$, for higher order reflections was calculated using Einstein's phonon model [Bird and King (1990)] from the D-W factor. Reflections for the Bloch wave calculations were
selected by the routine beams using \( w = 175 \). The theoretical rocking curves for matching were calculated by four methods listed below.

I. Anomalous absorption treated by perturbation (real symmetric matrix) and no correction for entrance aperture. This method requires the least computer CPU time.

II. Anomalous absorption coefficient, \( 1/\xi_g' \), is added to \( 1/\xi_g \) (complex symmetric matrix) and no correction for entrance aperture.

III. Anomalous absorption treated by perturbation (real symmetric matrix) and correction for entrance aperture using the model described in section 4.4.2.

IV. Anomalous absorption coefficient, \( 1/\xi_g' \), is added to \( 1/\xi_g \) (complex symmetric matrix) correction for entrance aperture using the model described in section 4.4.2. This method requires the maximum CPU time for fitting.

The Debye-Waller factor of Si, and other relevant experimental parameters required for the CBED calculations, are given in Table 4.4.

Table 4.4: Parameters used in the dynamical theory matching of 220 rocking curves of silicon in the program FIT.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lattice Parameter ((a))</td>
<td>0.5431 nm</td>
<td>Spackman (1986)</td>
</tr>
<tr>
<td>Debye-Waller Factor</td>
<td>0.4632 Å²</td>
<td>Spackman (1986)</td>
</tr>
<tr>
<td>Accelerating Voltage</td>
<td>119.45 KV</td>
<td>Present Measurement</td>
</tr>
</tbody>
</table>
The fitted values of different parameters are given in Table 4.5. The errors are estimated using the equation

$$\text{Error} = \frac{\sigma_{n-1}}{\sqrt{n}}$$  \hspace{1cm} (4.39)

where $\sigma_{n-1}$ is the standard deviation and $n$ is number of sample points.

Averaged values of the $\xi_{220}$ fitted by four different methods range between 82.2 to 82.3 nm. This means it makes no difference whether absorption is treated by the complex matrix formulation or by perturbation theory. Also the aperture correction seems to be unnecessary in the thickness range (124 to 200 nm) considered for the 220 reflection. All these predictions can be reconfirmed by re-tabulating and re-averaging the data in Table 4.6. The estimated error in $\xi_{220}$ values obtained by different fitting procedures range between 0.003 to 0.08%. This means that the final result is independent of the fitting procedure. In other words, in this foil thickness range for this reflection, the least time consuming computational procedure involving least time for computation, which incorporate the perturbation treatment of absorption and neglecting the aperture effect (Method-I) can be used for theoretical calculation of the rocking curves.
Table 4.5. Fitted values of the thickness (t) the extinction distance (ξ_{220}) and the absorption coefficient (ξ_{220}). Five different rocking curves numbered 1 to 5 have been fitted by four different methods described above. A dynamical interaction parameter value, w, of 175 was used to select beams (~ 50 beams).

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<th>Fitting Method</th>
<th>Fitted t in nm</th>
<th>Fitted ξ_{220} in nm</th>
<th>Fitted ξ_{220} in nm</th>
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The output in Table 4.5 is re-tabulated such that parameters fitted by different methods are grouped for every rocking curve. Five different rocking curves numbered 1 to 5 have been fitted by four different methods described in the text. A dynamical interaction parameter value, $w$, of 175 was used to select reflections using the routine beams.

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<th>Fitted $\xi_{220}$ nm</th>
<th>Fitted $\xi_{220}$ nm</th>
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<td>Error</td>
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<td>0.003</td>
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</table>
Even though the results tabulated in Tables 4.5 and 4.6 show that the fitted values of the $\xi_{220}$ are not affected by the fitting procedure, there is still no explanation as to why the averaged values of $\xi_{220}$ listed in Table 4.6 differ from the x-ray Pendellösung measurements by 6 to 11 Å. In fact, the fitted $\xi_{220}$ values obtained from five different rocking curves are scattered over a range of 10 Å (817 to 827 Å). There could be two reasons for this behavior; firstly, as discussed earlier this is the first attempt to measure $\xi_{220}$ from the energy filtered rocking curve measured by this experimental set up and this could have given rise to some instabilities and thus systematic errors. This is highly unlikely, because identical CBED patterns could be reproduced experimentally in duplicate runs even after days. Secondly, the reflections that are included in the calculations were determined using the routine beams. In all of above calculations routine beams selected all the beams which have a $\xi_{hsh} \leq 175$. In other words, approximately 50 beams were included in the Bloch wave calculations. It is possible that insufficient number of appropriate beams have been included in the calculations for the Bloch wave calculations to converge. The calculations were then repeated after increasing the $\xi_{hsh}$ limit to 500 and using Method-I. There were about 140 reflections included in the Bloch wave calculations. The results of these calculations are given in Table 4.7. An example of a rocking curve fitted by this method is shown in Figure 4.23.
Table 4.7. Fitted values of thickness (t) extinction distance ($\xi_{220}$) and the absorption coefficient ($\xi_{220}$). Five rocking curves numbered 1 to 5 have been fitted by Method-I with $\omega = 500$ (Approximately 140 beams included in the calculation).

<table>
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<tr>
<th>Rocking Curve No.</th>
<th>Orientation B</th>
<th>Fitting Method</th>
<th>Fitted t in nm</th>
<th>Fitted $\xi_{220}$ nm</th>
<th>Fitted $\xi_{220}$ nm</th>
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Figure 4.23: (a) A bright field 220 rocking curve along with the fitted curve calculated using approximately 140 beams, which were selected using a $w$ of 500. (b) A dark field 220 rocking curve along with the fitted curve calculated using approximately 140 beams selected using a $w$ of 500.
Results of the calculations made with $w = 500$ calculations tabulated in Table 4.7 indicate that the $\xi_{220}$ values are still differ from other experimental measurements by approximately 6 to 11 Å and are scattered over a range of 11 Å. Unfortunately $w$ cannot be increased further because even a 140 beams minimization calculation using the method-I takes as long as 3 hours in the CRAY-YMP, and 70 hours in a SUN SPARC 670 MP server. Because of this, a modified beam selection criterion was developed. In this method, instead of selecting all of the low and higher order beams which have a $\xi_{hsh}$ less than a specified $w$ value, an additional restriction based on the perpendicular (reciprocal) distance of the reflection from the zero layer is also applied. Thus, only those reflections ($h$) satisfying the following conditions are included in the calculations

1. $\xi_{hsh} < w$ and $B \cdot h < nlaue$ where $w$ and $nlaue$ are the input values of the dynamical interaction parameter and perpendicular (reciprocal) distance of the diffraction spot $h$ from the zero layer (see Figure 4.24).

2. $s_h < s_{max}$ where $s_{max}$ is the maximum deviation from the Bragg condition and is calculated from the radius of the CBED disk as shown in Figure 4.25.

$$s_{max} = |g| \tan \alpha_{max}$$  \hspace{1cm} (4.40)

where

$$\alpha_{max} = 2 \theta_B \frac{r}{|g|}$$  \hspace{1cm} (4.41)

where $r$ is the radius of CBED disk (see Figure 4.25). This criterion would include any reflection $h$, provided the magnitude of the deviation vector $|s_h|$
Figure 4.24: A schematic representation of the modified beam selection criterion: \( \zeta_h s_h < \omega \) and \( |B \cdot h| < nlaue \). The beam direction \( B \) is antiparallel to \( K \) but \( |B| \neq |K| \). In other words, only those reflections which satisfy the condition \( \zeta_h s_h < \omega \), and additionally those that satisfy the condition \( |B \cdot h| < nlaue \), are included in the calculations. An additional restriction is imposed based on whether the reflection is located above or below the reciprocal lattice layer \( nlaue \).
Figure 4.25: A schematic representation of the second beam selection criterion: $s_h < s_{\text{max}}$. This criterion would include any reflection $h$, provided the magnitude of the deviation vector $|sh|$ (see Figure 4.23 for definition of $s_h$) is less than the maximum deviation in bright field disk. The maximum deviation ($s_{\text{max}}$) of the $'g'$th disk is estimated from (a) as $|g| \tan \alpha_{\text{max}}$ and $\alpha_{\text{max}}$ is obtained from (b).
(see Figure 4.23 for definition of $s_b$) is less than the maximum deviation in
the bright field disk. The purpose is to include all the HOLZ lines that are
passing through the (000) CBED disk which might have been eliminated by
the first condition.

The same rocking curves were fitted again using this new beam selection
criteria. The limiting dynamical interaction parameter, $w$, value was given by $w = 
4000$ and $nlaue$ was varied. The fitted results are tabulated in Table 4.8. An example
fit for every $nlaue$ value used are given in Figures 4.26 through 4.29.

The fitted results shown in Table 4.8 indicate that the estimated error, $\Delta P$, in
$\xi_{220}$ and $\xi_{220}$ decreases as the $nlaue$ increases. When $nlaue = 9800$, there is only
0.07% error in the fitted values of $\xi_{220}$ and 1.2% error in the fitted values of $\xi_{220}'$.
Also the fitted value of the $\xi_{220}$ differs by less than 1 Å from the nearest x-ray
Pendellösung measurement listed in Table 4.3. This confirms that the Bloch wave
calculations have converged.

4.6.1.1: Error Analysis

It should be noted that the error in $\xi_{220}$ estimated from the $\sigma_{n-1}$ of the five
fitted $\xi_{220}$ values include only the random errors, $\Delta R$. In our case random errors
are:

1. The error in the orientation determination.

2. The error in the $\xi_g$'s and $\xi_g'$s of higher order reflections included in the Bloch
   wave calculations. Depending on the orientation, a different numbers of
   reflections have been included in the calculations and thus $\xi_g$'s and $\xi_g'$s are
Table 4.8. Fitted values of thickness (t) extinction distance ($\xi_{220}$) and the absorption coefficient ($\xi'_{220}$). Five rocking curves numbered 1 to 5 have been fitted by Method-I. All of the calculations were performed using a $\omega$ value of 4000. Reflections included in the calculations were chosen by an modified selection procedure described in the text.

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<td>0.27</td>
<td>23</td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>[11170 11170 2400]</td>
<td>7200</td>
<td>125.1</td>
<td>83.93</td>
<td>2934</td>
<td>2.43</td>
</tr>
<tr>
<td>2</td>
<td>[12131 12131 2400]</td>
<td>7200</td>
<td>136.8</td>
<td>82.79</td>
<td>3281</td>
<td>3.06</td>
</tr>
<tr>
<td>3</td>
<td>[12131 12131 2400]</td>
<td>7200</td>
<td>145.4</td>
<td>83.49</td>
<td>3301</td>
<td>3.07</td>
</tr>
<tr>
<td>4</td>
<td>[11060 11060 2400]</td>
<td>7200</td>
<td>147.8</td>
<td>83.01</td>
<td>3295</td>
<td>5.04</td>
</tr>
<tr>
<td>5</td>
<td>[11003 11003 2400]</td>
<td>7200</td>
<td>192.9</td>
<td>82.96</td>
<td>3135</td>
<td>4.22</td>
</tr>
<tr>
<td>Average</td>
<td></td>
<td></td>
<td></td>
<td>83.24</td>
<td>3189</td>
<td></td>
</tr>
<tr>
<td>Error</td>
<td></td>
<td></td>
<td></td>
<td>0.21</td>
<td>71</td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>[11170 11170 2400]</td>
<td>9600</td>
<td>124.8</td>
<td>83.01</td>
<td>3290</td>
<td>3.34</td>
</tr>
<tr>
<td>2</td>
<td>[12131 12131 2400]</td>
<td>9600</td>
<td>137.0</td>
<td>82.65</td>
<td>3414</td>
<td>3.10</td>
</tr>
<tr>
<td>3</td>
<td>[12131 12131 2400]</td>
<td>9600</td>
<td>144.6</td>
<td>82.70</td>
<td>3477</td>
<td>2.41</td>
</tr>
<tr>
<td>4</td>
<td>[11060 11060 2400]</td>
<td>9600</td>
<td>147.8</td>
<td>82.79</td>
<td>3350</td>
<td>6.36</td>
</tr>
<tr>
<td>5</td>
<td>[11003 11003 2400]</td>
<td>9600</td>
<td>193.4</td>
<td>82.88</td>
<td>3263</td>
<td>4.15</td>
</tr>
<tr>
<td>Average</td>
<td></td>
<td></td>
<td></td>
<td>82.81</td>
<td>3359</td>
<td></td>
</tr>
<tr>
<td>Error (AR)</td>
<td></td>
<td></td>
<td></td>
<td>0.06</td>
<td>39</td>
<td></td>
</tr>
<tr>
<td>$F^s_{220}$</td>
<td></td>
<td></td>
<td></td>
<td>69.171</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Error</td>
<td></td>
<td></td>
<td></td>
<td>0.037</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Figure 4.26: (a) A bright field 220 rocking curve along with the curve fitted using a $w$ of 4000 and $B.h = 2400$. (b) A dark field 220 rocking curve along with the curve fitted using a $w$ of 4000 and $B.h = 2400$. 
Figure 4.27: (a) A bright field 220 rocking curve along with the curve fitted using a $w$ of 4000 and $B.h = 4800$. (b) A dark field 220 rocking curve along with the curve fitted using a $w$ of 4000 and $B.h = 4800$. 

Angular dispersion in mrad.
Figure 4.28: (a) A bright field 220 rocking curve along with the curve fitted using a $\theta$ of 4000 and $B_h = 7200$. (b) A dark field 220 rocking curve along with the curve fitted using a $\theta$ of 4000 and $B_h = 7200$. 

\[ t = 124 \text{ nm} \]
Figure 4.29: (a) A bright field 220 rocking curve along with the curve fitted using a $w$ of 4000 and $B.h = 9600$. (b) A dark field 220 rocking curve along with the curve fitted using a $w$ of 4000 and $B.h = 9600$. 
also different.

3. The error in experimental intensity measurement.

4. Errors in the other fitted parameters, such as the thickness (t), $\xi_{220}$, $\xi'_{220}$, and background intensities.

5. Any fluctuations in TEM accelerating voltage.

These errors are referred to as random errors because they are different for different rocking curves or CBED patterns. The estimated error in $\xi_{220}$ does not include the systematic error in the accelerating voltage. The systematic error in $\xi_{220}$ resulting from the accelerating voltage, is given by Zuo (1989) as $\Delta E_o/2E_o$, where $E_o$ is the accelerating voltage. The overall error in $\xi_g$ would be given by

$$\left( \frac{\Delta \xi_g}{\xi_g} \right)^2 = \left( \frac{\Delta R}{\xi_g} \right)^2 + \left( \frac{\Delta E_o}{2E_o} \right)^2$$

(4.42)

By substituting $\Delta R = 0.06$ nm, $\Delta E_o = 0.2$ KV, $\xi_g = 82.81$ nm and $E_o = 119.45$ KV in (4.42) we obtain the overall error in $\xi_{220}$ as below:

Overall error in $\xi_{220} = \frac{\Delta \xi_{220}}{\xi_{220}} \times 100 = 0.11 \%$  

(4.43)

Using the conversion factors tabulated in Table 2.1, the overall error in the x-ray structure factor, $F_g$, is given by

Overall error in $F_g = 0.61 \times 0.11 = 0.07 \%$  

(4.44)
A summary of the present and past experimental measurements of $F_{220}$ of silicon along with the experimental techniques and references are given in Table 4.9. Values of $\xi_{220}$ determined by Pendellösung method range from 82.88 to 83.35 nm and the $\xi_{220}$ value of 82.81 nm obtained from the present measurement agree closely with Aldred and Hart's (1973) value. The present approach has been extremely successful in determining the value 220 structure factor in Si. The experimental approach used together with the fitting routines are robust.

Table 4.9  A comparison of current $F_{220}$ value with past experimental measurements. Parameters used for the conversion of electron quantities to x-ray quantities and vice versa are given in Table 4.4.

<table>
<thead>
<tr>
<th>Technique</th>
<th>$V_{220}$</th>
<th>$\xi_{220}$</th>
<th>$F_{220}$</th>
<th>$F_{s220}^e$</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>CBED-rocking curve</td>
<td>4.379</td>
<td>82.81</td>
<td>67.032</td>
<td>69.171±0.050</td>
<td>Present</td>
</tr>
<tr>
<td>x-ray - Pendellösung</td>
<td>4.375</td>
<td>82.88</td>
<td>67.668</td>
<td>69.208±0.007</td>
<td>Aldred and Hart (1973)</td>
</tr>
<tr>
<td>x-ray - Pendellösung</td>
<td>4.368</td>
<td>83.00</td>
<td>67.130</td>
<td>69.272±0.005</td>
<td>Spackman (1986)</td>
</tr>
<tr>
<td>x-ray - Pendellösung</td>
<td>4.361</td>
<td>83.14</td>
<td>67.200</td>
<td>69.343±0.165</td>
<td>Takama, Iswasaki and Sato (1980)</td>
</tr>
<tr>
<td>x-ray - Pendellösung</td>
<td>4.346</td>
<td>83.35</td>
<td>67.343</td>
<td>69.492±0.001</td>
<td>Saka and Kato (1986)</td>
</tr>
<tr>
<td>CBED-rocking curve</td>
<td>4.396</td>
<td>82.48</td>
<td>66.866</td>
<td>69.000±0.280</td>
<td>Voss, Lehmpfuhl and Smith (1980)</td>
</tr>
<tr>
<td>IKL</td>
<td>4.335</td>
<td>83.65</td>
<td>67.448</td>
<td>69.600±0.08</td>
<td>Terasaki, Waterabe and Gjonnes (1979)</td>
</tr>
</tbody>
</table>
4.6.2: Comparison of Experimental and Theoretical Absorption Coefficients

In all the above calculations the coefficient of the absorption potential, $V_g^\varepsilon$, and hence $\xi_g^\varepsilon$ of the higher order reflections has been calculated theoretically from Einstein's phonon model [Bird and King (1990)]. However, the fitted room temperature values of $\xi_{220}^\varepsilon$ of 3359 nm were found to be far smaller than the value of 3952 nm estimated from the theory using a D-W factor of 0.4632 Å². This means that the theory is probably underestimating the absorption potential. The coefficient of the absorption potential, $V_g^\varepsilon$, is dependent on the temperature. By studying the temperature dependence of the $V_g^\varepsilon$ one can prove whether the theory is underestimating the absorption or not. For this purpose the rocking curves obtained from the energy-filtered CBED patterns recorded at different temperatures were matched using the program FIT. For Si, experimental Debye-Waller factors are available at 92 K [Cummings and Hart (1988)] and at 293 K [Spackman (1986)]. Debye-Waller factors of Si for other temperatures were estimated from the fits for phonon dispersion curves provided by Sears and Shelley (1991). Lattice parameters at various temperatures were estimated using the linear coefficient of thermal expansion [Shah and Strumanis (1972)]. The specimen temperature was assumed to be equal to the stage temperature in all the calculations. Errors in the room temperature values were estimated by fitting five different rocking curves at five different thicknesses as listed in Table 4.8. The error in $V_{220}^\varepsilon$ at other temperatures was assumed to be equal to the room temperature value.

The (stage) temperature dependence of the fitted values for the coefficient of the absorption potential, $V_{220}^\varepsilon$, along with those calculated from experimental and theoretical D-W factors, are shown in Figure 4.30. The 98 K data point was not
Figure 4.30: Temperature dependence of the coefficient of absorption potential ($V'_{220}$). The fitted values are approximately 0.01 V high than the theoretical calculation.
connected to the remainder of the curve because there is a discontinuity in the temperature dependence of the D-W factor (hence $V_{220}^\prime$) curve at 0.2T$_{\text{melting}}$ of silicon [Sears and Shelley (1991)].

The shape of the temperature dependence curve of fitted $V_{220}^\prime$ values is similar to that calculated from the Einstein phonon model. This is an expected result because all the inelastically scattered electrons, except those scattered by phonons, have been filtered. At temperatures $\leq 298$ K, the fitted value of $V_{220}^\prime$ is shown in Figure 4.23 and is approximately 0.01 V higher than that predicted by the phonon model [Bird and King (1990)] at room temperature and above. This suggests a correction should be applied to higher order $V_g^\prime$ values that have been calculated using the phonon model. The nature of the correction would depend on the $g$ vector and is currently unpredictable using available data.
CHAPTER - V

SUMMARY AND CONCLUSIONS

5.1: Summary

A number of theoretical charge density calculations have predicted strong directional bonding to be the major contributing factor for the brittleness of TiAl. However, it would be difficult to arrive at any conclusion unless the electron charge density distribution in TiAl is determined experimentally. In order to do this, the x-ray structure factors of TiAl have to be measured accurately. In view of this, the work described in this thesis focused on various methodologies involved in the experimental determination of x-ray structure factors of TiAl. To this effect, studies on Si have been undertaken, where Si is considered to be a well characterized material.

TiAl is stable over a compositional range of 48 to 56 at% Al content. At present, because of the constraints imposed by the phase diagram only aluminum rich TiAl single crystals are available. Without a stoichiometric single crystal it is difficult to obtain unambiguous structure factor values by the single crystal x-ray diffraction methods. Because of this reason, electron diffraction techniques using polycrystalline thin foils have to be used for the structure factor measurement. There are two main difficulties with electron diffraction methods, firstly, as seen from Tables 2.2 and 2.3, the accuracy of the electron diffraction method deteriorates
with increasing sinθ/λ value, as a result it is difficult to measure medium and high angle reflections as accurately as the low-angle ones. Secondly, the electron diffraction methods require accurate values of the Debye-Waller (D-W) factors, which have to be measured independently by conventional single crystal x-ray or neutron diffraction methods.

In this work, the D-W factor measurement was attempted by the four-circle single crystal x-ray diffraction method. The four-circle method requires a small (> 40 μm size) single crystal specimen of TiAl for accurate integrated intensity measurement. With the available specimen preparation techniques, it seems, it would be difficult to obtain a good quality single crystal of 40 μm size. All of the attempts to start from large single crystal and reduce the size of the crystal by cleaving or machining have been largely unsuccessful. Most of the crystals are either deformed or lost during the size reduction process before reaching 40 μm size. Because of these reasons single crystal Ti54at%Al specimen of ~ 80 μm size was used for the determination of D-W factors. The results of four circle x-ray diffraction, discussed in Chapter III, lead to an unexpected problem caused by multiple occupancy in the Ti sites.

The experimental procedure and computer routines for automatic structure factor refinement by matching rocking curves have been developed for future use. The computer programs and experimental data collection system have been successfully tested after considering various theoretical formulations and experimental limitations. The 220 structure factor of silicon has been measured, as a test case, to an accuracy that is comparable to the x-ray Pendellösung method. The temperature dependence of the coefficient of the absorption potential for the
220 silicon has also been measured. Results indicate that the coefficient of the absorption potential values used in CBED calculations might have been underestimated by Einstein's model for phonon scattering. This might possibly introduce an additional (unknown) systematic error into the fitted values of structure factors.

5.2: Conclusions

1. Attempts to measure D-W factors by using a non-stoichiometric single crystal have yielded ambiguous values for the D-W factors for different sites in TiAl. Because of this reason, the structure factors and the electron charge density distribution in TiAl have not been measured.

2. Results of the single crystal x-ray diffraction experiments provide the first experimental proof for the occupancy of excess Al atoms on the Ti sublattice. This is supported by the fact that the intensity of reflections with indices satisfying the condition \( h+k = 2n+1 \) increased with increasing Al content.

3. Least-squares refinement results indicate that there is a high probability that the excess Al atoms preferentially occupy one of the Ti sites in the L10 TiAl, rather than equally on both of the Ti sites. In fact, this hypothesis is consistent with the phase stability calculations of Fu (1990) and Asta et al. (1992); however, it is difficult to draw a final conclusion using the data from one composition.

4. The procedure for the CBED rocking curve method of structure factor determination has been established using the silicon 220 as a test case. Application of this technique to structure factor determination in TiAl is pending upon determination of the D-W factors.
CHAPTER VI

FUTURE WORK

The current status of this program indicates that it will be practically impossible to measure D-W factors or structure factors of TiAl unambiguously by techniques that use a single crystal specimen. In view of this, new techniques employing polycrystalline material will have to be developed for the measurement of D-W factors in TiAl. At present, the CBED method is being evaluated for the measurement of D-W factor of silicon by using 220 reflections. The computer routines for the CBED method use a finite difference gradient method for the multidimensional $\chi^2$ minimization. This procedure is not very efficient and requires a large number of iterations (CPU time) to reach the global minimum. This will be an important drawback as the number of fitted parameters is increased e.g. the D-W factors for TiAl introduces two more variables. Because of this, more efficient algorithms employing analytical gradients, similar to the one developed for zone-axis pattern matching by Saunders (1994), will have to be developed for future use.

Even though the single crystal x-ray diffraction experiments did not serve the goal of the present project, it has opened new avenues of research from the point of understanding the deformation behavior of non-stoichiometric TiAl. For example, the decreasing ductility of TiAl with increasing Al content can be
explained on the basis of increasing metastable (L12) TiAl3 within TiAl lattice. In order to prove this, x-ray data will have to be collected systematically from a series of TiAl samples with changing Al content. For this purpose it would be advisable to prepare single crystal samples by heat-treating gas atomized powders rather than starting from a large crystal and reducing its size.
APPENDIX A: SYSPAT - Program for the Calculation CBED Patterns

The main routine of the program, SYSPAT, for the calculation CBED patterns is given in this section. This program utilizes the routine EVCSF from the IMSL FORTRAN library for the diagonalization of the structure matrix. In addition to the IMSL library, routines given in Appendix I should also be linked during compilation.
PROGRAM SYSFAT
C CALCULATION OF DISKS IN THE SYSTEMATIC PATTERN
C ABSORPTION IS TREATED BY PERTURBATION:
C BEAM DIRECTION IS INTO THE SPECIMEN: DOWNWARDS IN A TEM
C APERTURE CORRECTION OPTION INCLUDED: CAMARA LENGTH = 600MM
C APERTURE CORRECTION IS NOT PERFORMED IN DIRECTION PARALLEL
C TO THE KIKUCHI LINES INTENSITY IS ASSUMED CONSTANT IN THIS
C DIRECTION
C COMPATIBLE WITH: SUN-OS, CRAY-UNICOS, AND VAX-VMS
C LIBRARIES REQUIRED: IMSL-MATH LIBRARY
C OPTION: IFLAGEXTN = 2 IS DESIGNED ESPECIALLY FOR SI ROOMT
C
C PROGRAM WORKS FOR CUBIC AND TETRAGONAL CRYSTAL STRUCTURES
C
C VARIABLES:
C NATOMS NO. OF ATOM TYPES IN THE UNIT CELL
C IPOS NO. OF ATOMIC POSITIONS IN THE UNIT CELL
C IZ ATOMIC NUMBER
C NAME ATOMIC SYMBOL
C XSI(1 -200) EXTINCTION DISTANCES
C XSIDSH(1 -200) ABSORPTION DISTANCES
C ALATT,CLATT LATTICE PARAMETER
C T FOIL THICKNESS
C V BEAM VOLTAGE (KV)
C IDPIXEL NUMBER OF EXPERIMENTAL POINTS (PER BEAM)
C
CHARACTER*2 ATONMNAME(10),NAME(3)
DIMENSION P1(10),P2(10),P3(10),DW1(10),BEAM(3)
EXTERNAL FCN

COMMON /BLK1/ IBEAM(30,3),XSI(30),XSIDSH(30)
COMMON /BLK1/ XSI(200),XSIDSH(200),W(200),ALATT,CLATT,T,V,
A NUMBM(5),NUMBM,IFLGNZON,DEVRRG,
B CALC(300,5),IVAR(35),IFLAGEXTN,IAPR,IFLGLP
COMMON /BLK2/ NREAD1,NRITE1,NRITE2
COMMON /BLK3/ BMDIR(3),IG(3),IVECY(3),IBEAM(200,3),
A IBRAGG,NBEAMS,IPXEL,IPXEL,IZAXIS(3)
COMMON /BLK4/ P1,P2,P3,P4,DW1,IPOS
COMMON /BLK5/ IZ(2),AX(2,4),BX(2,4),CC(2),NATOMS
COMMON /BLK7/ATOMNAME,NAME
COMMON /H/NLAUE

C SET UP INPUT AND OUTPUT CHANNELS
C
NREAD1=7
NRITE1=9
NRITE2=10
OPEN(UNIT=NREAD1,FILE='FITNSYS.DAT1',STATUS='OLD')
OPEN(UNIT=NRITE1,FILE='FIT.OUT1',STATUS='UNKNOWN')
WRITE(NRITE1,895)

C
C INPUT DIFFRACTION DATA ON FIRST CHANNEL (NREAD1)
READ(NREAD1,700)
C READ NO. OF ATOMIC POSITIONS AND THE NATOMS
READ(NREAD1,705)IPOS,NATOMS
WRITE(NRITE1,896)IPOS,NATOMS
C AX, BX, C : H-F ATOMIC SCATTERING FACTORS
READ(NREAD1,700)
WRITE(NRITE1,897)
C READ ATOMIC NUMBER AND FREE ATOM SCATTERING FACTORS FOR "NATOMS":
D0120I=1,NATOMS
READ(NREAD1,706)NAME(I),IZ(I),(AX(I,J),BX(I,J),J=1,4),CC(I)
WRITE(NRITE1,896)NAME(I),IZ(I),(AX(I,J),BX(I,J),J=1,4),CC(I)
120 CONTINUE
READ(NREAD1,700)
WRITE(NRITE1,898)
C READ ATOMNAME, ATOMIC POSITIONS P1,P2,AND P3,
C DEBYE WALLER FACTORS IN "NM"
D0121I=1,IPOS
READ(NREAD1,707)ATOMNAME(I),P1(I),P2(I),P3(I),DW1(I)
WRITE(NRITE1,899)ATOMNAME(I),P1(I),P2(I),P3(I),DW1(I)
121 CONTINUE
C READ LATTICE PARAMETERS, THICKNESS IN NM,VOLTAGE IN KV
C DEVRBG: DEVIATION FROM BRAGG CONDITION IN PIXELS
READ(NREAD1,700)
READ(NREAD1,702)ALATT,CLATT,T,V,DEVRBG
WRITE(NRITE1,903)ALATT,CLATT,T,V,DEVRBG
C
C READ BASIC IG = G-VECTOR: IBRAGG = THE G IN BRAGG:
C IZAXIS = ZONE AXIS: IGPIXEL = NO. OF PIXEL IN G:
C IDPIXEL = NO. OF PIXELS IN DIAMETER OF DISK:
C NBEAMS = NO. OF BEAMS OR REFLECTIONS
C
C : => IF NBEAMS = 0 THEN GOTO 119 AND
C USE ROUTINE BEAMS TO DETERMINE REFLECTIONS:
C IFLAGEXTN IS THE FLAG FOR PROPER EXTINCTION DISTANCE ROUTINE
C IF IFLAGEXTN=0, THEN USE EXTN (BIRD'S ROUTINE);
C IF IFLAGEXTN=1, THEN USE EXTN1 (H-F FREE ATOM MODEL);
C IF IFLAGEXTN=2, THEN USE EXTN2 (EXP. STR. FACTORS);
C
READ(NREAD1,700)
READ(NREAD1,920)IG,IBRAGG,IZAXIS,IGPIXEL,IDPIXEL,NBEAMS,
AIFLAGEXTN,IAPR
WRITE(NRITE1,921)IG,IBRAGG,IZAXIS,IGPIXEL,IDPIXEL,NBEAMS,
A1APR,IFLAGEXTN
C
C WHEN IBRAGG=0, RESET IBRAGG=1 FOR ANGULAR RESOLUTION CALCULATIONS
C AFTER SETTING IFLAGZON >0
IF(IBMAGG.EQ.0)THEN
IFLGZON=1
IBRAGG=1
ENDIF

C
READ(NREAD1,700)
IF(NBEAMS.EQ.0)GOTO119
C IF NBEAMS .NE.0 THEN READ IBEAM: ARRAY IN WHICH BEAMS ARE STORED;
DO122I=1,NBEAMS
READ(NREAD1,708) (IBEAM(I,J),J=1,3)
BEAM(1)=FLOAT(IBEAM(I,1))
BEAM(2)=FLOAT(IBEAM(I,2))
BEAM(3)=FLOAT(IBEAM(I,3))
C
C IF IFLAGEXTN=0, THEN USE EXTN (BIRD'S ROUTINE);
C IF IFLAGEXTN=1, THEN USE EXTN1 (H-F FREE ATOM MODEL);
C IF IFLAGEXTN=2, THEN USE EXTN2 (EXP. STR. FACTORS);
C
CALL EXTN(ATOMNAME,P1,P2,P3,DW1,IPOS,BEAM,ALATT,CLATT,V
$,$XSI(I),$XSIDSH(I))
C
C AVOID EXTINCTION CALCULATION FOR 000 REFLECTION
IF(I.EQ.1)GOTO 122
C
IF (IFLAGEXTN .EQ. 1) THEN
CALL EXTN1(ATOMNAME,NAME,P1,P2,P3,DW1,IPOS,BEAM,ALATT,CLATT,V,
$XSI(I))
ELSEIF (IFLAGEXTN .EQ. 2 ) THEN
CALL EXTN2(ATOMNAME,NAME,P1,P2,P3,DW1,IPOS,BEAM,ALATT,CLATT,V,
$XSI(I))
ENDIF
122 CONTINUE
C
:119 READ (NREAD1,700)
C
C READ WMAX = SG*XSI, THE DYNAMICAL INTERACTION PARAMETERS
C NLAUE(FORMAT 18) (FIG 4.24): THE NUMBER OF LAUE ZONES
C USE VERY HIGH NLAUE ZONE IF YOU WANT TO USE JUST
C WMAX = SG*XSI CRITERIA (UNMODIFIED BEAM SELECTION CRITERIA);
C IFLOGEXP = 0 IF NO EXPERIMENTAL EXTINCTION DISTANCES ARE USED
C IFLOGEXP > 0 IF THERE EXPERIMENTAL EXTINCTION DISTANCE TO BE INPUT
C AND THE VALUE OF IFLOGEXP INDICATES NO. OF THE EXTINCTION DISTANCES

READ (NREAD1,709) WMAX,NLAUE,IFLOGEXP
C
READ(7,700)
READ(NREAD1,705) (IMGBM(I),I=1,5)
C
C DETERMINE HOW MANY IMAGING BEAMS: NIMGBM
C
DO1I=2,5
IF (IMGBM(I).EQ.0) GOTO 2
1 CONTINUE
   I=6
2 NIMGBM=I-1
   WRITE(NRITE1,906) (IMGBM(I),I=1,NIMGBM)

   IF (NBEMS.GT.0) GO TO 123
   CALL BEAMS(WMAX)
   WRITE(NRITE1,931)NBEMS,WMAX

C
123 WRITE(NRITE1,925)IFLGEXP
   WRITE(NRITE1,928)
   DO 124 I=1,NBEMS
124 WRITE(NRITE1,926) (IBEAM(I,J),J=1,3),XSI(I),XSIDS(I),W(I),I
   IF (IFLGEXP.EQ.0) GOTO 126
   READ(NREAD1,700)
   READ(NREAD1,700)
   READ(NREAD1,700)
   READ(NREAD1,700)
   READ(NREAD1,700)
   READ(NREAD1,700)
   READ(NREAD1,700)
   READ(NREAD1,700)
   READ(7,700)

C
C READ BEAMS INCLUDED IN THE SCAN, INITIAL VALUES OF
C XSI, XSIDS AND BKG:
C
C   WRITE(NRITE1,924)
   DO 125 I=1,IFLGEXP
      READ(NREAD1,708)(IBEAM1(I,J),J=1,3),XSI1(I),XSIDS1(I)
   125 WRITE(NRITE1,926)(IBEAM1(I,J),J=1,3),XSI1(I),XSIDS1(I)

C SET UP DIFFRACTION CALCULATION
C
126 CALL DIFFRT
   CALL FCN
C
   STOP

C
700 FORMAT(A80)
701 FORMAT(5F10.0)
702 FORMAT(5F10.0)
704 FORMAT(2F10.0)
705 FORMAT(15I5)
706 FORMAT(A2,I4,3F7.4,F8.4,3F7.4,F8.4,F7.4)
707 FORMAT(A2,3F6.0,F9.0)
708 FORMAT(3I5,2F10.0,F6.0)
709 FORMAT(F5.0,I8,I5)
801 FORMAT(5F10.0)
895 FORMAT('OUTPUT OF SYSPAT')
896 FORMAT(3X,'NO. OF ATOMIC POSTIONS : ',I5/,3X,
A 'NO. OF ATOM TYPES : ',I5/)
897 FORMAT('AX, BX, C :H-F ATOMIC SCATTERING FACTORS')
898 FORMAT('/' ELE X Y Z D-W FACTOR ')
899 FORMAT(A2,3F7.3,F9.4)
903 FORMAT('/' LATTICE PARAMETER A='',F8.6,' NM'/
A ' C='',F8.6,' NM'/
B ' FOIL THICKNESS =', F8.3,
C ' NM'/ BEAM VOLTAGE =',F8.2,' KV'
D ',/ DEVIATION FROM BRAGG=',F9.3,' PIXELS'/)
906 FORMAT('/' IMAGING BEAMS DISKS TO BE CALCULATED :',5I4)
920 FORMAT(4I5,3I10,6I5)
921 FORMAT(3X,'G VECTOR : ',3I5/,3X,
A 'BEAM IN BRAGG : 'I5/,3X
B 'ZONE AXIS : ',3I10/,3X,
C 'NO. OF PIXEL IN G : ', I5/,3X, 'NO. OF PIXEL IN THE D : ',
D I5/,3X,'NUMBER OF BEAMS : ',I5,
E ' (IF NBEMS=0, USE WMX CRITERIA)/,3X,
F 'APERTURE CORR TN. FLAG : ',I5, ' (IF =0 NO CORRECTION)'
G '/3X,'SUBROUTINE EXTN',I1,' IS USED FOR XSI CALCULATION',
H '/3X, 'BEAMS : ')
924 FORMAT('/ VALUES OF EXPERIMENTAL PARAMETERS INCLUDED',
A ' IN THE PROFILE:')
925 FORMAT('/' IFLGEXP (IF =0, USE ONLY THEORETICAL VALUES): ',I5/)
926 FORMAT(3I5,3F12.4,I5)
927 FORMAT('/' INCIDENT BEAM DIRECTION : ',3F7.2,/)
928 FORMAT('/' H',' K',' L',' XSI NM ',' XSIDASH NM'
A )
929 FORMAT('/' H',' K',' L',' XSI NM ',' XSIDSH NM ',',
A ' BKG')
931 FORMAT
A (' NUMBER OF BEAMS INCLUDED IN CALCULATIONS, NBEMS = ',I6
B ' DYNAMICAL INTERACTION PARAMETER, WMAX = ',
C F8.2/)  
C
C END
C
C SUBROUTINE FOR SETTING UP THE DYNAMICAL MATRIX
C
C SUBROUTINE DIFFRT
C
C A(200,200) = DYNAMICAL MATRIX
C XDSH = MATRIX WITH XSIDH FOR BEAMS IN DYNAMICLA MATRIX
C BMDIR(3) = BEAMDIRECTION VECTOR
C GV = G VECTOR
C ZAXIS = ZONE AXIS
C VECY = DEVIATION VECTOR PERPENDICULAR G AND ON ZONE AXIS
C BEAM = BEAMS INCLUDED IN THE CALCULATION
C IGPIXEL = NUMBER OF PIXELS IN A 'G' LENGTH
C IDPIXEL = NUMBER OF PIXEL IN A CBED DISC DIAMETER
C IBRAGG = REFLECTION ON THE BRAGG CONDITION
C IBRAGG=0, INDICATES ZONE AXIS PATTERN: OR SYMMETRIC PATTERN
C STBMDR = STARTING BEAM DIRECTION
C BMDSRT = SCALAR DESCRIBING THE BEAM STARTING POINT
C BMEND = SCALAR DESCRIBING THE BEAM ENDING POINT
C
DIMENSION A(200,200),TEMP1(3),XDISH(200,200),TEMP2(3),VECY(3)
A,STBMDR1(3)
COMMON /BLK1/ XSI(200),XSIDSH(200),W(200),ALATT,CLATT,T,V,
A IMGBM(5),NIMGBM,IFLGZON,DEVBRG,
B CALC(300,5),IVAR(35),IFLAGEXTN,IAPR,IFLGEXP
COMMON /BLK3/ BMIR(3),IG3,IVEC3,IBEAM(200,3),
A IBRAGG,NBEAMS,IPXPIXEL,IPXPIXEL,IZAXIS(3)
COMMON /BLK6/ V1,AK,BMDSRT,BMEND,DAKX,STBMDR3(3),GV(3),ZAXIS(3)
A,A,BEAMS(200,3),G,XDISH,ANG,RISTEP,DIF,VECY,STBMDR1
C
DATA MAXB/100/
C
C CONVERT KV TO VOLTS
C
V1=V*1000.
C
WAVE VECTOR (CONSTANTS FROM EADES ET AL.)
C
AK=SQR(T(V1*(1.+0.97948E-6*V1))/1.2264
C
SIZE OF G
C
G=SQR(T((IG1)**2+IG2)**2)/((ALATT**2)+(IG3)**2)/(CLATT**2))
C
SIZE OF GIBRAGG = I1BRAGG*GV1
C
GIBRAGG=SQR(T((IG1*IBRAGG)**2+(IG2*IBRAGG)**2)/(ALATT**2)+
A((IG3*IBRAGG)**2)/(CLATT**2))
C
CALCULATE THE BRAGG ANGLE
C
BRAGG = ASIN(GIBRAGG/2/AK)
COSBRAG= COS(BRAGG)*AK
SINBRAG=-1*SIN(BRAGG)*AK
C
CALCULATE THE ANGULAR LENGTH IN MRAD BETWEEN PIXELS
ANG=2.0*BRAGG*1000./IPXPIXEL
C
ANGULAR RADIUS R IS SET FOR 600 MM CAMARA LENGTH &
C 0.2NM APERTURE DIAMETER IN MRAD
C
R =0.333333333/2
C NO. OF STEPS ON EITHER SIDE OVER WHICH THE INTEGRATION IS PERFORMED
C
ISTEP = IAPR*(0.3333333/ANG/2.)

C DIF IS QUANTITY LEFT IN EXCESS OF THE STEPS
C
IF(IAPR.EQ.0) GO TO 1
DIF=0.3333333/2. - FLOAT(ISTEP)*ANG/IAPR

C DETERMINE BEAM DIRECTION
C
BMDIR = COSBRAG*ZAXIS + SINBRAG*GV

1 CALL FLOATVEC(IZAXIS,ZAXIS)
   CALL NORM(ZAXIS)
   CALL FLOATVEC(IG,GV)
   CALL NORM(GV)
   IF(IFLGZON.LE.0) THEN
      CALL SCALEVEC(ZAXIS,COSBRAG)
      CALL SCALEVEC(GV,SINBRAG)
      CALL ADDVEC(ZAXIS,GV,BMDIR)
      CALL NORM(ZAXIS)
   ELSE
      CALL SCALENEW(ZAXIS,BMDIR,AK)
   ENDIF

C NOW WE HAVE VECTOR BMDIR(3) WHICH REPRESENTS THE INCIDENT BEAM DI-
C RECTION
C WITH THE LENGTH OF 1./LAMBDAM [NM**(-1)]
C
DO 30 I=1,NBEAMS
C "BEAMS(I,3)" ARE RL VECTORS (THOSE, THAT WE INCLUDE IN CALCULATIONS),
C IN THE UNITS OF NM**(-1)
C
BEAMS(I,1) = FLOAT(IBEAM(I,1)) / ALATT
BEAMS(I,2) = FLOAT(IBEAM(I,2)) / ALATT
BEAMS(I,3) = FLOAT(IBEAM(I,3)) / CLATT
30 CONTINUE

C CALCULATE BMDSRT AND BMDEND:
C "BMDSRT" IS LENGTH OF VECTOR FROM THE CENTER TO THE LEFT END OF BF
C DISK (-)
C "BMDEND" IS LENGTH OF VECTOR FROM THE CENTER TO THE RIGHT END OF BF
C DISK (+)
C ORIGIN IS TAKEN IN THE MIDDLE OF BF DISK (000).
C
BMDSTR = - 0.5 * FLOAT(IDPIXEL) / FLOAT(IGPIXEL) * G
BMDEND = + 0.5 * FLOAT(IDPIXEL) / FLOAT(IGPIXEL) * G

C    DAKX = (BMDEND-BMDSTR)/(IDPIXEL-1)
C
C DETERMINE THE STARTING INCIDENT BEAM DIRECTION
C
CALL RECMN(IG,GV)
CALL NORM(GV)
CALL SCALNEW(GV,TEMP1,BMDRT)

C    NOW TEMP1(3) IS A VECTOR FROM CENTER OF 000 DISK TO LEFT (STARTING)
C    EDGE OF THE DISK. UNITS => NM**(-1) == TEMP2
C
CALL ADDVEC(TEMP1,BMDIR,TEMP2)

C FIND DIRECTION PERPENDICULAR TO TEMP2 AND G
CALL CROSS(GV,TEMP2,VECY)
CALL NORM(VECY)
FACTOR=0.5*G*(FLOAT(IDPIXEL)/FLOAT(IGPIXEL))
CALL SCALVEC(VECY,FACTOR)
CALL ADDVEC(VECY,TEMP2,STBMORL)
CALL SCALVEC(VECY,-1.0)
CALL NORM(VECY)

C
C STBMORL(3) IS A VECTOR FROM THE EWALD SPHERE CENTER TO
C THE STARTING POINT (LEFT EDGE OF BF 000 DISK).
C
THOPI=6.283185307
CALL OFF_DIAGONAL(NBEAMS,IBEAM,A,XDHS)

C
IF(IFLGEXP.NE.0)CALL UOFF_DIAGONAL(NBEAMS,IBEAM,A,XDHS)
C
RETURN
END

C SUBROUTINE FOR INTENSITY CALCULATION
C
SUBROUTINE FCN

REAL TEMP(3),BEAM(3),TEMP1(3)

COMPLEX CE(200),CU

DIMENSION A(200,200),AC(200,200),GAMMA(200),Q(200)
C.A,XDHS(200,200),CAL(900,5),CBED(512,512),VECY(3),STBMORL(3)
COMMON /BLK1/ XST(200),XSIDSH(200),W(200),ALATT,CLATT,T,V,
      A
      IMGBM(5),NIMGBM,IFLGZON,DEVBG,
      B
      CALC(300,5),IVAR(35),IFLAGEXTN,IAPR,IFLGEXP
COMMON /BLK2/ NREAD1,NRITE1,NRITE2
COMMON /BLK3/ BMDIR(3),IG(3),IVECY(3),IBEAM(200,3),
A IBRAGG,NBEAMS,IGPIXEL,IDPIXEL,IZAXIS(3)
COMMON /BLK6/ V1,AK,BMDSRT,BMDEND,DAKX,STBMDR(3),GV(3),ZAXIS(3),
A A,BEAMS(200,3),G,XDSH,ANG,R,ISTEP,DIF,VECY,STBMDR1
C
DATA MAXBM/200/
TWOPI=6.283185307
C
C SET UP THE OFF-DIAGONAL ELEMENTS OF THE DYNAMICAL MATRIX
C
C SET UP THE DIAGONAL ELEMENTS
C
C ORIENTATION LOOP
C
C IF IAPR = 0 NO APERTURE INTEGRATION
C
IFAC=IAPR
IF(IAPR.EQ.0)IFAC=1
F=0.
C
C DEPENDING 'IAPR' INCREASE THE NO. OF ORIENTATIONS TO BE CALCULATED
C IF (IAPR =0) THEN 'ISTEP' IS ALSO ZERO.
C
DO1?IL=1,IDPIXEL
AKY= (IL-1)*DAKX
CALL SCALENEW(VECY,TEMP1,AKY)
CALL ADDVEC(TEMP1,STBMDR1,STBMDR)
C
DO2I=1,IDPIXEL*IFAC+2*ISTEP
C
C ADD PERCENTAGE DEVIATION FROM BRAGG :DEVERG
C
AKX=(I-1-ISTEP)*(DAKX/IFAC) + DEVERG/IGPIXEL
C
C DETERMINE THE K VECTOR
C GV IS IN UNIT ( 1 NM**(-1) LONG ) IN THE G-DIRECTION
C AKX IS CURRENT INCREMENT OF ORIENTATION (IN NM**(-1) ).
C GV*AKX IS THE INCREMENTAL VECTOR
C
CALL SCALENEW(GV,TEMP,AKX)
C
C ADD INCREMENTAL VECTOR TO STBMDR TO GET NEW BMDIR
C
CALL ADDVEC(STBMDR,TEMP,BMDIR)
C
C SET THE LENGTH OF NEW BMDIR TO AK
CALL NORM(BMDIR)
CALL SCALEVEC(BMDIR,AK)

C SET UP THE DIAGONAL ELEMENTS OF DYNAMICAL MATRIX
C
DO3J=1,NBEAMS
BEAM(1)= BEAMS(J,1)
BEAM(2)= BEAMS(J,2)
BEAM(3)= BEAMS(J,3)
C
C SIZE OF THE BEAM
C
CALL ADDVEC(BMDIR,Beam,TEMP)
C
C DETERMINE EXCITATION PARAMETER-SG
C
SG= (AK**2 - SCALAR(TEMP,TEMP) ) / (2. * AK)
C
IF(J,EQ.1)SG=0.
A(J,J)=SG
C
C SET DIAGONAL ELEMENTS OF XDHS TO (1/MEAN ABSORPTION POTENTIAL)=
C XSIDSH0
C
XDHS(J,J)=XSIDSH(1)
3 CONTINUE
C
CALL EVCSF(NBEAMS,A,KAXBM,GAMMA,AC,MAXB)
C
C WORK OUT ABSORPTION COEFFICIENTS (REF: ELECTRON
C DIFFRACTION, BY HUMPHREYS AND BITHELL, IN ELECTRON
C DIFFRACTION TECHNIQUES, ED. BY J. M. COWLEY)
C
DO5J=1,NBEAMS
Q(J)=0.
DO5K=1,NBEAMS
DO5L=1,NBEAMS
Q(J)=Q(J)+AC(K,J)*AC(L,J)/2./XDHS(K,L)
5 CONTINUE
C
C COMPLEX PHASE FACTOR
C
DO6J=1,NIMGBM
6 CE(J)=CEXP(CMPLX(-TWOPI*T*Q(J),TWOPI*T*GAMMA(J)))
C
C DETERMINE INTENSITIES : ELECTRON MICRODIFFRACTION BY ZOU & SPENCE
C
DO7J=1,NIMGBM
CU=CMPLX(0.,0.)
DO7K=1,NBEAMS
CU=CU+AC(J,K)*CE(K)*AC(1,K)
7 CAL(I,J)=REAL(CU*CONJG(CU))

2 CONTINUE

C IF IA PR = 0 NO APERTURE INTEGRATION
C
C IF (IA PR.EQ.0) GOTO 9
C
C INTEGRATE OVER THE CIRCULAR APERTURE
C
KOUNT=0
DO81=ISTEP+1, (IA PR*IDPIXEL+ISTEP), IA PR
KOUNT =KOUNT+1
DO8J=1, NIMG BM
CALC(KOUNT,J)=0.0
DO8JL=ISTEP,-(ISTEP-1),-1
HH1=CAL(I-JL,J)
HH2=CAL(I-JL+1,J)
X1=JL*ANG/IA PR
X2=(JL-1)*ANG/IA PR
M=(HH1-HH2)/(X1-X2)
H1=HH1-M*X1
H2=N*R+H1
IF(JL.EQ.-ISTEP)THEN
   X1= X1-DIF
ELSEIF(JL.EQ.(ISTEP-1))THEN
   X2= X2+DIF
ENDIF

C FOR MORE DETAILS ABOUT INTEGRATION HERE SEE SWAMINATHAN'S THESIS
C
CALC(KOUNT,J)=CALC(KOUNT,J)+H1*AREA(X1,X2)+
A((H1-H2)/R)*(R**2*(X1-X2)-(X1-X2)**3/3)

8 CONTINUE

C GOTO 10
C
9 DO11J=1, NIMG BM
   DO11I=1, IDPIXEL
11   CALC(I,J)=CALC(I,J)

10 CONTINUE
   DO18J=1, NIMG BM
   DO18I=1, IDPIXEL
   K=IDPIXEL*(J-1)+I
18   CBED(IL,K)=CALC(I,J)
   WRITE(*,*) ' LINE =', IL , (CBED(IL,IJ),I,J=2,82,20)
   WRITE(13,1003)(CBED(IL,J),J=1,512)

17 CONTINUE
   REWIND(13)
CALL WRITEC(CBED)
RETURN

901 FORMAT(' F =',E15.5)
902 FORMAT(' NVAR, (XGUESS(I),I=1,NVAR)',I5,5E15.7)
1001 FORMAT(2I5,2F10.5)
1002 FORMAT(10(2X,F13.5))
1003 FORMAT(8(E10.4))
END

C CALCULATE THE AREA OF APERTURE
C
FUNCTION AREA(X1,X2)
R=0.33333333/2
AREA=ABS(2.*(X2*SQRT(R**2-X2**2)/2.-X1*SQRT(R**2-X1**2))/2.
  +R**2/2.*(ASIN(X2/R)-ASIN(X1/R)))
RETURN
END

C SUBROUTINE FOR CALCULATION OF OFF-DIAGONAL ELEMENTS OF THE
C DYNAMICAL MATRIX
C
SUBROUTINE OFF_DIAGONAL(NBEAM,IBEAM,A,XDISH)
CHARACTER*2 ATOMNAME(10),NAME(3)
REAL XSIDIS,XSIABS
DIMENSION A(200,200),IBEAM(200,3),BEAM(3),XDISH(200,200)
DIMENSION P1(10),P2(10),P3(10),DW1(10)

COMMON /BLK1/ XSI(200),XSIDSH(200),W(200),ALATT,CLATT,T,V,
  A IMGBM(5),NIMGBM,IFLGZON,DEVB,4
  B CALC(300,5),IVAR(35),IFLAGEXTN,IAPR,IFLGEXP
COMMON /BLK4/ P1,P2,P3,P4,DW1,IPOS
COMMON /BLK7/ATOMNAME,NAME
INTEGER NBEAM

DO2K=1,NBEAM
DO2L=K+1,NBEAM

C CALCULATE THE BEAM = G1-G2 VECTOR
C
BEAM(1)= FLOAT( IBEAM(K,1)-IBEAM(L,1) )
BEAM(2)= FLOAT( IBEAM(K,2)-IBEAM(L,2) )
BEAM(3)= FLOAT( IBEAM(K,3)-IBEAM(L,3) )

C CALCULATE THE EXTINCTION DISTANCE FOR BEAM
C IF IFLAGEXTN=0, THEN GO TO EXTN (BIRD'S ROUTINE);
C IF IFLAGEXTN=1, THEN GO TO EXTN1 (FREE ATOM MODEL);
C IF IFLAGEXTN=2, THEN GO TO EXTN2 (EXP. STR. FACTORS);
C
CALL EXTN(ATOMNAME,P1,P2,P3,DW1,IPOS,BEAM,ALATT,CLATT,T,V
\$ ,XSIDIS,XSIABS;
IF (IFLAGEXTN .EQ. 1) THEN
CALL EXTN1(ATOMNAME,NAME,P1,P2,P3,DW1,IPOS,BEAM,ALATT,CLATT,V,
\$XSIDIS)
ELSEIF (IFLAGEXTN .EQ. 2) THEN
CALL EXTN2(ATOMNAME,NAME,P1,P2,P3,DW1,IPOS,BEAM,ALATT,CLATT,V,
\$XSIDIS)
ENDIF

C SET UP THE DYNAMICAL MATRIX ELEMENTS : 1/(2*XSI)

A(K,L)=0.5/XSIDIS
A(L,K)=0.5/XSIDIS

C SET UP THE ABSORPTION MATRIX ELEMENTS: 1/(2*XSIDSH)

XSIDSH(K,L)=XSIABS
XSIDSH(L,K)=XSIABS

CONTINUE
RETURN
END

C SUBROUTINE FOR UPDATING THE OFF_DIAGONAL ELEMENTS OF THE DYNAMICAL
C MATRIX

SUBROUTINE UPOFF_DIAGNIAL(NBEAM,Ibeam,A,XSIDH)
REAL XSIDIS
INTEGER NBEAM
DIMENSION A(200,200),Ibeam(200,3),BEAM(3),XSIDH(200,200)
DIMENSION P1(10),P2(10),P3(10),DW1(10),ABSBM(10)
COMMON /BLK/ Ibeam(10),XSII(10),XSIDSH(10),A
COMMON /BLK1/ XSI(200),XSIDSH(200),W(200),ALATT,CLATT,T,V,
A IMGBM(5),NIMBM,IFLgzon,DEVBG,
B CALC(300,5),IVAR(35),IFLAGEXTN,IAPR,IflGexp
COMMON /BLK4/ P1,P2,P3,P4,DW1,IPOS
COMMON /BLK7/ATOMNAME,NAME

C CALCULATE THE (1/D-SPACING) FOR ALL IMAGING BEAMS

DO1M=1,IFLGexp
ABSBM(M)=(Ibeam1(M,1)**2 + Ibeam1(M,2)**2)/ALATT**2 +
(Ibeam1(M,3)**2)/CLATT**2
IF (ABSBM(M) .EQ. 0.0) THEN
XSI(1)=XSII(M)
XSIDSH(1)=XSIDSH1(M)
ENDIF
CONTINUE

C COMPARE FIRST THE (1/D-SPACING) VALUES AND THE HKL VALUES
C OF IMAGING BEAM WITH THE (G1-G2) BEAMS OF THE DYNAMICAL
C MATRIX TO FIND THE EQUIVALENT REFLECTION IN THE OFF DIAGONAL
C AND UPDATE IT WITH NEW VALUE:
C **WARNING** JUST D-SPACING COMPARISON IS NOT FOOL PROOF:
C (E.G) (511) AND (333) FOR SI HAVE SAME D-SPACING:
C
DO2K =L, NBEAM
DQ2L= K+1,NBEAM
BEAM(1) = FLOAT( IBEAM(K,1)-IBEAM(L,1) )
BEAM(2) = FLOAT( IBEAM(K,2)-IBEAM(L,2) )
BEAM(3) = FLOAT( IBEAM(K,3)-IBEAM(L,3) )
BM = (BEAM(1)**2 + BEAM(2)**2)/ALATT**2 + (BEAM(3)**2)/CLATT**2
DO3M=1,IFLGEXP
  IF ( ABSBM(M).EQ.BM ) THEN
    IF ((ABS(BEAM(1)).EQ.ABS(IBEAM1(M,1))).AND. ABS(BEAM(2)).EQ.
      AABS(IBEAM1(M,2)).AND. ABS(BEAM(3)).EQ. AABS(IBEAM1(M,3))).OR.
      B(ABS(BEAM(1)).EQ.ABS(IBEAM1(M,2))).AND. ABS(BEAM(2)).EQ.
      CABS(IBEAM1(M,3)).AND. ABS(BEAM(3)).EQ. AABS(IBEAM1(M,1))).OR.
      D(ABS(BEAM(1)).EQ.ABS(IBEAM1(M,3))).AND. ABS(BEAM(2)).EQ.
      EABS(IBEAM1(M,1))).AND. ABS(BEAM(3)).EQ. AABS(IBEAM1(M,2))) THEN
      SIGN=A(K,L)/ABS(A(K,L))
      XSIDIS=SIGN*ABS(XSID1(M))
      A(K,L)=0.5/XSIDIS
      A(L,K)=0.5/XSIDIS
      SIGNDSh=XDSH(K,L)/ABS(XDSH(K,L))
      XDSH(K,L)=SIGNDSh*ABS(XSIDSH1(M))
      XDSH(L,K)=SIGNDSh*ABS(XSIDSH1(M))
    ENDIF
  ENDIF
3  CONTINUE
2  CONTINUE
RETURN
END
APPENDIX B : Example Input to the Program SYSPAT

File: FITNSYS.DAT1

SILICON DATA: ipos, natoms, 215
  8 1
AX, BX, C : H-F ATOMIC SCATTERING FACTORS: A2, A4, 3f7.4, f8.4, 3f7.4, f8.4, f7.4
SI 14 6.2915 2.4396 3.0353 32.3337 1.9891 0.6785 1.5410 81.6937 1.1407
Atom, Position, D-W factor: a2, 3f6.0, f9.0
SI -.125 -.125 -.125  0.4632
SI  .375 .375  -.125  0.4632
SI  .375  -.125 .375  0.4632
SI -.125  .375  .375  0.4632
SI  .125  .125  .125  0.4632
SI  .625  .625  .125  0.4632
SI  .125  .625  .625  0.4632
SI  .625  .125  .625  0.4632
a, c (nm) 2f10.0; t, V (nm, kV) 2f10.0;
.543102028 .543102028 124.8428 119.45 0.00 0.73606
ig. idragg, ibmdir igpixel, idpixel, nbeams, iflagextn, iaper{13i5}
 2 2 0 1 -11170 11170 2400 115 115 0 1 0
Beams included:  ! Dummy
Wmax, nlaue, number_of_experimental extinction distances (f5.0,i8,i5)
4000. 9600 2  ! If nbeams=0, beams is called
input(1-9ngbm) 5i5 (number of systematic reflections in this case 0 and g)
 0 1 0 0 0
Dummy line  ! These dummy lines are
Dummy line  ! used here to facilitate
Dummy line  ! direct use of
Dummy line  ! input files to FIT_COMP and FIT_PERT
Dummy line  ! as input file
Dummy line  ! to SYSPAT, with
Dummy line  ! slight modification
Beam, extinction distances and background: 3i5, 2f10.0, f6.0
 0 0 0 0.0000 534.2700 59.82
 2 2 0 -83.008-3390.1631 46.78
APPENDIX C : Example Output of the Program SYSPAT

File : FIT.OUT1

OUTPUT OF NSYSIM

   NO. OF ATOMIC POSTIONS :  8
   NO. OF ATOM TYPES       :  1

AX, BX, C : H-F ATOMIC SCATTERING FACTORS
SI 14 6.2915 2.4386 3.0353 32.3337 1.9891 0.6785 1.5410 81.6937 1.1407

ELE  X   Y   Z   D-W FACTOR
SI -0.125 -0.125 -0.125  0.4632
SI  0.375  0.375 -0.125  0.4632
SI  0.375 -0.125  0.375  0.4632
SI -0.125  0.375  0.375  0.4632
SI  0.125  0.125  0.125  0.4632
SI  0.625  0.625  0.125  0.4632
SI  0.125  0.625  0.625  0.4632
SI  0.625  0.125  0.625  0.4632

LATTICE PARAMETER A = 0.543102 NM
                  C = 0.543102 NM
FOIL THICKNESS    = 124.843 NM
BEAM VOLTAGE       = 119.45 KV
DEVIATION FROM BRAGG=  0.000 PIXELS

G VECTOR :  2  2  0
BEAM IN BRAGG :  1
ZONE AXIS :  -11170  11170  2400
NO. OF PIXEL IN G :  115
NO. OF PIXEL IN THE D :  115
NUMBER OF BEAMS  :  0 (IF NBEAMS=0, USE WMAX CRITERIA)
APERURE CORRTN. FLAG :  0 (IF =0 NO CORRECTION)

SUBROUTINE EXTN1 IS USED FOR XSI CALCULATION

BEAMS :

IMAGING BEAMS DISKS TO BE CALCULATED :  0  1

NUMBER OF BEAMS INCLUDED IN CALCULATIONS, NBEAMS =  177
DYNAMICAL INTERACTION PARAMETER, \( W_{\text{MAX}} = 4000.90 \)

IFLGEQ (IF = 0, USE ONLY THEORETICAL VALUES): 2

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VALUES OF EXPERIMENTAL PARAMETERS INCLUDED IN THE PROFILE:

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Figure D.1: Output of the program SYSPAT.
APPENDIX D : FIT_PERT - Program for Matching Rocking Curves

The main routine of the program, FIT_PERT, for matching rocking curves is given in this section. FIT_PERT uses perturbation theory treat anomalous absorption. This program utilize routines EVCSF and BCONF (U4INF) from the IMSL FORTRAN library for the diagonalization of the structure matrix and the $\chi^2$ minimization. In addition to the IMSL library, routines given in Appendix I should also be linked during compilation.
PROGRAM FIT_PERT
C ABSORPTION IS TREATED BY PERTURBATION:
C BEAM DIRECTION IS INTO THE SPECIMEN: DOWNWARDS IN A TEM
C APERTURE CORRECTION OPTION INCLUDED: CAMARA LENGTH = 600MM
C : APERTURE SIZE=0.2MM
C APERTURE CORRECTION IS NOT PERFORMED IN DIRECTION PARALLEL
C TO THE KIKUCHI LINES INTENSITY IS ASSUMED CONSTANT IN THIS
C DIRECTION
C COMPATIBLE WITH: SUN-OS, CRAY-UNICOS, AND VAX-VMS
C LIBRARIES REQUIRED : IMSL-MATH LIBRARY
C OPTION: IFLAGEXTN = 2 IS DESIGNED ESPECIALLY FOR SI ROOMT
C
C PROGRAM WORKS FOR CUBIC AND TETRAGONAL CRYSTAL STRUCTURES
C
C VARIABLES:
C NATOMS    NO. OF ATOM TYPES IN THE UNIT CELL
C IPOS      NO. OF ATOMIC POSITIONS IN THE UNIT CELL
C IZ        ATOMIC NUMBER
C NAME      ATOMIC SYMBOL
C XSI(1-100) EXTINCTION DISTANCES
C XSIDSH(1-100) ABSORPTION DISTANCES
C ALATT,CLATT LATTICE PARAMETER
C T         FOIL THICKNESS
C V         BEAM VOLTAGE (KV)
C MASK      DEFINES WHICH PARAMETERS VARY
C
1 THICKNESS
C 2-11 XSI(1-50)
C 12-21 XSIDSH(1-50)
C 22-32 BKG(1-11)
C
C EXPTL    HOLDS EXPERIMENTAL INTENSITIES
C MAXEXP   MAXIMUM NUMBER OF EXPERIMENTAL INTENSITIES (PER BEAM)
C IDPIXEL  NUMBER OF EXPERIMENTAL POINTS (PER BEAM)
C
DIMENSION XGUESS(35),XSSCALE(35),IPARAM(7),RPARAM(7),X(35),
A MASK(35),XLB(35),XUB(35)
C
CHARACTER*2 ATOMNAME(10),NAME(3)
DIMENSION P1(10),P2(10),P3(10),DW1(10),BEAM(3)
EXTERNAL UMINF,U4INF,FCN
C
COMMON /BLK1/ XSI(200),XSIDSH(200),W(200),ALATT,CLATT,T,V,
A IMGBM(5),NIMGBM,EXPTL(300,5),DEVBRG,
B CALC(300,5),MAXEXP,NORMBM,IVAR(35),
C ILOR(5,5),IHIR(5,5),BKG(11),IFLAGEXTN,IPR
COMMON /BLK2/ NREAD1,NREAD2,NRITE1,NRITE2
COMMON /BLK3/ BMNDR(3),IG(3),IVECY(3),IBEAM(200,3),
A IBRAGG,NBEAMS,IGPIXEL,IDPIXEL,IZAXIS(3)
COMMON /BLK4/ P1,P2,P3,P4,DW1,IPOS
COMMON /BLK5/ IZ(2),AX(2,4),BX(2,4),CC(2),NATOMS
COMMON /BLK7/ATOMNAME,NAMEN
COMMON /H/NLAUE

C
DATA MAXEXP/1000/

C SET UP INPUT AND OUTPUT CHANNELS

NREAD1=7
NREAD2=8
NRITE1=9
NRITE2=10
OPEN(UNIT=NREAD1,FILE='FITNSYS.DAT1',STATUS='OLD')
OPEN(UNIT=NREAD2,FILE='FIT.DAT2',STATUS='OLD')
OPEN(UNIT=NRITE1,FILE='FIT.OUT1',STATUS='UNKNOWN')
OPEN(UNIT=NRITE2,FILE='FIT.OUT2',STATUS='UNKNOWN')
WRITE(NRITE1,895)

C INPUT DIFFRACTION DATA ON FIRST CHANNEL (NREAD1)
READ(NREAD1,700)

C READ NO. OF ATOMIC POSITIONS AND THE NATOMS
READ(NREAD1,705)IPOS,NATOMS
WRITE(NRITE1,896)IPOS,NATOMS

C AX, BX, C :H-F ATOMIC SCATTERING FACTORS
READ(NREAD1,700)
WRITE(NRITE1,897)

C READ ATOMIC NUMBER AND FREE ATOM SCATTERING FACTORS FOR "NATOMS"
DO120I=1,NATOMS
READ(NREAD1,706)NAME(I),IZ(I),(AX(I,J),BX(I,J),J=1,4),CC(I)
WRITE(NRITE1,706)NAME(I),IZ(I),(AX(I,J),BX(I,J),J=1,4),CC(I)
120 CONTINUE
READ(NREAD1,700)
WRITE(NRITE1,898)

C READ ATOMNAME, ATOMIC POSITIONS P1,P2, AND P3,
C DEBYE WALLER FACTORS IN "NM"
DO121I=1,IPOS
READ(NREAD1,707)ATOMNAME(I),P1(I),P2(I),P3(I),DW1(I)
WRITE(NRITE1,899)ATOMNAME(I),P1(I),P2(I),P3(I),DW1(I)
121 CONTINUE

C READ LATTICE PARAMETERS, THICKNESS IN NM, VOLTAGE IN KV
C DEVBREG: DEVIATION FROM BRAGG CONDITION IN PIXELS
READ(NREAD1,700)
READ(NREAD1,702)ALATT,CLATT,T,V,DEVBREG
WRITE(NRITE1,903)ALATT,CLATT,T,V,DEVBREG

C
C READ BASIC IG = G-VECTOR: IBRAGG = THE G IN BRAGG:
C IZAXIS = ZONE AXIS: IGPIXEL = NO. OF PIXEL IN G:
C IDPIXEL= NO. OF PIXELS IN DIAMETER OF DISC:
C NBEAMS = NO. OF BEAMS OR REFLECTIONS
C : ==> IF NBEAMS = 0 THEN GOTO 119 AND
C USE ROUTINE BEAMS TO DETERMINE REFLECTIONS:
C IFLAGEXTN IS THE FLAG FOR PROPER EXTINCTION DISTANCE ROUTINE
C IF IFLAGEXTN=0, THEN USE EXTN (BIRD'S ROUTINE);
C IF IFLAGEXTN=1, THEN USE EXTN1 (H-F FREE ATOM MODEL);
C IF IFLAGEXTN=2, THEN USE EXTN2 (EXP. STR. FACTORS);
C
READ(NREAD1,700)
READ(NREAD1,920) IG,IBRAGG,IZAXIS,IGPIXEL,IDPIXEL,NBEAMS,
AILAGEXTN,IAPR
WRITE(NRITE1,921) IG,IBRAGG,IZAXIS,IGPIXEL,IDPIXEL,NBEAMS,
AIAPR,IFLAGEXTN
READ(NREAD1,700)
IF(NBEAMS.EQ.0)GOTO119
C IF NBEAMS .NE.0 THEN READ IBEAM: ARRAY IN WHICH BEAMS ARE STORED;
DO122I=1,NBEAMS
READ(NREAD1,708) (IBEAM(I,J),J=1,3)
BEAM(1)=FLOAT(IBEAM(I,1))
BEAM(2)=FLOAT(IBEAM(I,2))
BEAM(3)=FLOAT(IBEAM(I,3))
C
C IF IFLAGEXTN=0, THEN USE EXTN (BIRD'S ROUTINE);
C IF IFLAGEXTN=1, THEN USE EXTN1 (H-F FREE ATOM MODEL);
C IF IFLAGEXTN=2, THEN USE EXTN2 (EXP. STR. FACTORS);
C
CALL EXTN(ATOMNAME,P1,P2,P3,DW1,IPOS,BEAM,ALATT,CLATT,V
$,XSI(I),XSIDSH(I))
C
C AVOID EXTINCTION CALCULATION FOR 000 REFLECTION
IF(I.EQ.1)GOTO 122
C
IF (IFLAGEXTN .EQ. 1) THEN
CALL EXTN1(ATOMNAME,NAME,P1,P2,P3,DW1,IPOS,BEAM,ALATT,CLATT,V,
$XSI(I))
ELSEIF (IFLAGEXTN .EQ. 2 ) THEN
CALL EXTN2(ATOMNAME,NAME,P1,P2,P3,DW1,IPOS,BEAM,ALATT,CLATT,V,
$XSI(I))
ENDIF
122 CONTINUE
C
119 READ (NREAD1,700)
C
C READ WMAX = SG*XSI, THE DYNAMICAL INTERACTION PARAMETERS
C NLAUE(FORMAT 18)(FIG 4.24): THE NUMBER OF LAUE ZONES
C USE VERY HIGH NLAUE ZONE IF YOU WANT TO USE JUST
C WMAX = SG*XSI CRITERIA (UNMODIFIED BEAM SELECTION CRITERIA)
C
C READ (NREAD1,709) WMAX,NLAUE
C
READ(7,700)
READ(NREAD1,705) (IMGBM(I), I=1,5)

C DETERMINE HOW MANY IMAGING BEAMS: NIMGBM
C
DO1 I=2,5
   IF(IMGBM(I).EQ.0) GOTO2
1 CONTINUE
   I=6
2 NIMGBM=I-1
   WRITE(NRITE1,906) (IMGBM(I), I=1,NIMGBM)

   IF(NBEAMS.GT.0) GO TO 123
   CALL BEAMS(WMAX)
   WRITE(NRITE1,931) NBEAMS, WMAX

C 123 WRITE(NRITE1,928)
   DO 124 I=1,NBEAMS
   124 WRITE(NRITE1,925) (IBEAM(I,J), J=1,3), XSI(I), XSIDSH(I), W(I), I
C
C READ MASK: MASK(1) = THICKNESS;
C   MASK(2-11) = XSI;
C   MASK(12-22) = XSIDSH;
C   MASK(23-32) = BKG;
C   MASK(33) = BRAGG ANGLE VARIATION;
C   MASK(34-35) = FOR FUTURE USE;
C
READ(NREAD1,700)
READ(NREAD1,703) MASK
WRITE(NRITE1,904)
   IF(MASK(1).NE.0) WRITE(NRITE1,914)
   DO13 I=2,11
      IF(MASK(I).NE.0) WRITE(NRITE1,915) I-1
      J=I+10
   IF(MASK(J).NE.0) WRITE(NRITE1,916) J-1
      K=I+20
   IF(MASK(K).NE.0) WRITE(NRITE1,922) K-22
13 CONTINUE
   L=33
   IF(MASK(L).NE.0) WRITE(NRITE1,923) L-32

C C INPUT ROCKING CURVE DATA ON SECOND CHANNEL (NREAD2) C
C
   READ(7,700)
   WRITE(NRITE1,929)
C
C READ BEAMS INCLUDED IN THE SCAN, INITIAL VALUES OF C XSI, XSIDSH AND BKG:
C
   WRITE(NRITE1,924)
   DO 125 I=1,NIMGBM
READ(NREAD1,708) (IBEAM(I,J),J=1,3),XSI(I),XSIDSH(I),BKG(I)
125 WRITE(NRITE1,708) (IBEAM(I,J),J=1,3),XSI(I),XSIDSH(I),BKG(I)

C READ IN EXPERIMENTAL RANGES OF POINTS TO BE EXCLUDED FROM THE FIT:
C
READ(NREAD1,700)
READ(NREAD1,705) (( ILOR(JB,IE), IE=1,5 ), JB=1,NIMGBM)
READ(NREAD1,700)
READ(NREAD1,705) (( IHIR(JB,IE), IE=1,5 ), JB=1,NIMGBM)
DO16I=1,NIMGBM
16 WRITE(9,918)I-1,( ILOR(I,JE),IHIR(I,JE), JE=1,5)
READ(NREAD2,700)

C READ DATA IN THE INVERTED ORDER:
DO18I=IDPIXEL,1,-1
18 READ(NREAD2,801) (EXPTL(I,J),J=1,NIMGBM)

C CHECK VALIDITY OF DATA
C
DO3I=1,NIMGBM
DO 3 JE=1,5
IF(ILOR(I,JE).LT.0.OR.IGOR(I,JE).GT.IDPIXEL)GOTO5
IF(IHIR(I,JE).LT.0.OR.IHIR(I,JE).GT.IDPIXEL)GOTO5
GOTO3
5 WRITE(NRITE1,917)
STOP
3 CONTINUE
IF(IDPIXEL.GT.1)GOTO17
WRITE(NRITE1,908)
STOP
17 IF(IDPIXEL.LE.MAXEXP)GOTO12
WRITE(NRITE1,907)
STOP
12 CONTINUE

C LOCATE MAXIMUM INTENSITY
C
EXPMAX=0.
DO20I=1,NIMGBM
DO11J=1,IDPIXEL
IF(EXPTL(J,I).LT.EXPMAX)GOTO11
EXPMAX=EXPTL(J,I)
MAXEXP=J
NORMBM=I
11 CONTINUE
20 CONTINUE

C IF PREFERRED, READ IN
C
READ(NREAD1,700)
READ(NREAD1,705)ITEMP1,ITEMP2
IF (ITEMP1.EQ.0 .AND. ITEMP2.EQ.0) GOTO21
DO22I=1,NIMGBM
IF(ITEMP1.NE.IMGBM(I))!GOTO22
NORMBM=I
22 CONTINUE
MAXEXP=ITEMP2
21 CONTINUE
C
WRITE(NRITE1,919)MAXEXP,IMGBM(NORMBM)
C
C SET UP DIFFRACTION CALCULATION
C
CALL DIFPRT
WRITE(NRITE1,927)BMDIR
C
C IDENTIFY WHICH PARAMETERS ARE TO VARY
C
J=0
DO6I=1,33
IF(MASK(I).EQ.0)GOTO6
J=J+1
IVAR(J)=I
6 CONTINUE
IF(J.GT.0)GOTO7
WRITE(NRITE1,909)
STOP
7 NVAR=J
C
C INITIALISE ORIGINAL GUESSED VALUES
C
DO8I=1,NVAR
IF(IVAR(I).EQ.1)XGUESS(I)=T
IF(IVAR(I).GT.1 .AND. IVAR(I).LT.12)XGUESS(I)=XI(IVAR(I))
IF(IVAR(I).GT.11 .AND. IVAR(I).LT.22)XGUESS(I)=XSIDSH(IVAR(I)-10)
IF(IVAR(I).GT.21 .AND. IVAR(I).LT.33)XGUESS(I)=BKG(IVAR(I)-21)
8 IF(IVAR(I).EQ.33)XGUESS(I)=DEVBKG
C
SET UP REST OF MINIMISATION ROUTINE
C
C CALL U4INF TO SET UP PARAMETERS IPARAM AND RPARAM ITERATIONS
C REFER IMSL MANUAL
CALL U4INF(IPARAM,RPARAM)
C
DO9I=1,NVAR
9 XSCALE(I)=1.0
C
FScale=1.0
READ(NREAD1,700)
READ(NREAD1,705)IPARAM(3),IPARAM(4),IPARAM(5)
WRITE(*,*)IPARAM
C READ STEPL==RPARAM(2)
C
READ(NREAD1,700)
READ(NREAD1,*)RPARAM(2)
C
C RPARAM(5) FALSE CONVERGENCE TOLERANCE
C
RPARAM(5)=1.0E-02
C
C IBTYPE =0; ==> USER WILL SUPPLY THE BOUNDS
C
IBTYPE=0
C
C SET UP THE BOUNDS FOR THE VARIABLES
C
DO15I=1,NVAR
SIGN=XGUESS(I)/ABS(XGUESS(I))
IF(SIGN.GT.0)THEN
XLB(I)=XGUESS(I)/2.
XUB(I)=XGUESS(I)*2.
ELSE
XUB(I)=XGUESS(I)/2.
XLB(I)=XGUESS(I)*2.
ENDIF
C IF DEVBNG IS VARIED BOUNDS ARE BETWEEN +1 OR -1
IF(IVAR(I).EQ.33)THEN
XLB(I)=-1.
XUB(I)=1.
ENDIF
15 CONTINUE
C
C MINIMISATION
C
IF (MASK(35).EQ.0) THEN
CALL BCONF(FCN,NVAR,XGUESS,IBTYPE,XLB,XUB,XSCALE,FScale,
AIPARAM,RPARAM,X,FVALUE)
ELSE
CALL FCN(NVAR,XGUESS,FVALUE)
ENDIF
C WRITE MINIMISED PARAMETERS ON FIRST OUTPUT CHANNEL
C
DO10I=1,NVAR
IF(IVAR(I).EQ.1)WRITE(NRITE1,910)T
IF(IVAR(I).GT.1.AND.IVAR(I).LT.12)WRITE(NRITE1,911)IVAR(I)-1,
AXSI(IVAR(I))
IF(IVAR(I).GT.11.AND.IVAR(I).LT.22)
AWRITE(NRITE1,912)IVAR(I)-11,XSIDSH(IVAR(I)-10)
IF(IVAR(I).GT.21.AND.IVAR(I).LT.33)
AWRITE(NRITE1,930)IVAR(I)-22,BKG(IVAR(I)-21)
IF(IVAR(I).EQ.33)WRITE(NRITE1,932)DEVBNG
10 CONTINUE
   WRITE(NRITE1,913)FVALUE
C
C WRITE CALCULATED PROFILE ON SECOND CHANNEL
C
   WRITE(NRITE2,1001)NIMGBM, IDPIXEL
   DO4I=1, IDPIXEL
   4 WRITE(NRITE2,1002) (EXPTL(I,J), J=1, NIMGBM), (CALC(I,J), J=1, NIMGBM)
C
   STOP
C
700 FORMAT(A80)
701 FORMAT(5F10.0)
702 FORMAT(5F10.0)
703 FORMAT(I2/10I2/10I2/I2/10I2/10I2)
704 FORMAT(2F10.0)
705 FORMAT(15I5)
706 FORMAT(A2, I4, 3F7.4, F8.4, 3F7.4, F8.4, F7.4)
707 FORMAT(A2, 3F6.0, F9.0)
708 FORMAT(3I5, 2F10.0, F6.0)
709 FORMAT(F5.0, 18, I5)
801 FORMAT(5F10.0)
895 FORMAT('OUTPUT OF FIT_PERT: ABSORPTION TREATED BY PERTURBATION')
896 FORMAT(3X, 'NO. OF ATOMIC POSTIONS : ', I5, 3X,
      A 'NO. OF ATOM TYPES : ', I5)
897 FORMAT('AX, BX, C : H-F ATOMIC SCATTERING FACTORS')
898 FORMAT('/' ELE X Y Z D-W FACTOR ')
899 FORMAT(A2, 3F7.3, F9.4)
901 FORMAT(' INITIAL EXTINCTION DISTANCES: '/(5F10.1))
902 FORMAT(' INITIAL ABSORPTION DISTANCES: '/(5F10.1))
903 FORMAT('/', ' LATTICE PARAMETER A =', F8.6, ' NM/'
      A  ' C =', F8.6, ' NM/'
      B ' FOIL THICKNESS =', F8.3,
      C ' NM/ ' BEAM VOLTAGE =', F8.2, ' KV'
      D ' DEVIATION FROM BRAGG=', F9.3, ' PIXELS')
904 FORMAT('/' MASK DETAILS ')
906 FORMAT('/' IMAGING BEAMS INCLUDED IN THE PROFILE: ', 5I4)
907 FORMAT(' TOO MANY EXPERIMENTAL POINTS')
908 FORMAT(' TOO FEW EXPERIMENTAL POINTS')
909 FORMAT(' NO PARAMETERS SET TO VARY')
910 FORMAT(' OPTIMISED FOIL THICKNESS =', F12.4)
911 FORMAT(' THE BEST FIT ', I2,' TH EXTINCTION DISTANCE IS ', F12.4)
912 FORMAT(' THE BEST FIT ', I2,' TH ABSORPTION DISTANCE IS ', F14.4)
913 FORMAT(' THE MINIMIZED SUM OF SQUARED DEVIATIONS =', E15.5)
914 FORMAT(' THICKNESS IS BEING OPTIMISED')
915 FORMAT(' THE', I2,' TH EXTINCTION DISTANCE IS BEING ',
      A ' OPTIMISED')
916 FORMAT(' THE', I2,' TH ABSORPTION DISTANCE IS BEING ',
      A ' OPTIMISED')
917 FORMAT(/ ' THE NUMBER OF EXPERIMENTAL POINTS (PER BEAM) IS',I4)
918 FORMAT(/ ' FOR THE ',II,'TH BEAM USED RANGES OF POINTS EXCLUDED',
       A
   5(/I3, ' TO ',I3))
919 FORMAT(/ ' THE NORMALISING POINT IS NUMBER ',I3,' OF THE ',
       A
   II,'TH BEAM')
920 FORMAT(415,3110,715)
921 FORMAT(3X,'G VECTOR' : ',315,/,3X,
       A'BEAM IN BRAGG' : ',I5,/,3X,
       B'ZONE AXIS' : ',3110,/,3X,
       C'NO. OF PIXEL IN G' : ',I5,/,3X,'NO. OF PIXEL IN THE D' : ',
       D I5,/,3X,'NUMBER OF BEAMS' : ',I5,
       E' (IF NBEAMS=0, USE WMAX CRITERIA)' : ',/3X,
       F'APERTURE CORRTN. FLAG' : ',I5,' (IF =0 NO CORRECTION)
       G'/3X,'SUBROUTINE EXTN',II,' IS USED FOR XSI CALCULATION',
       H'/,3X,'BEAMS :')
922 FORMAT(/ ' THE',II,'TH BACKGROUND IS BEING OPTIMISED')
923 FORMAT(/ ' THE BRAGG ANGLE IS BEING OPTIMISED')
924 FORMAT(/ ' INITIAL VALUES OF VARIABLES FOR THE BEAMS INCLUDED',
       A' IN THE PROFILE:')
925 FORMAT(3X,A2,1X,4F7.4)
926 FORMAT(315,3F12.4,15,2X,F6.1)
927 FORMAT(/,' INCIDENT BEAM DIRECTION : ',3F7.2,/) 
928 FORMAT(/,' H' ,' K' , ' L' , ' XSI NM ' , ' XSIDASH NM' 
       A)
929 FORMAT(/,' H' , ' K' , ' L' , ' XSI NM ' , ' XSIDSH NM ' ,
       A'BKG')
930 FORMAT(/ ' THE BEST FIT ',II,' TH BEAM BACKGROUND :',F12.4)
931 FORMAT(/ ' THE BEST FIT FOR DEVIATION FROM BRAGG CONDITION 
       :',F12.4,
       A'MRAD')
1001 FORMAT(315,2F10.5)
1002 FORMAT(10(2X,F13.5) )
C
C END
C 
C SUBROUTINE FOR SETTING UP THE DYNAMICAL MATRIX
C 
C SUBROUTINE DIFFRT
C 
C A(200,200) = DYNAMICAL MATRIX
C XDISH = MATRIX WITH XSIDSH FOR BEAMS IN DYNAMICLA MATRIX
C BMNDR(3) = BEAMDIRECTION VECTOR
C GV = G VECTOR
C ZAXIS = ZONE AXIS
C VECY = DEVIATION VECTOR PERPENDICULAR G AND ON ZONE AXIS
C BEAM = BEAMS INCLUDED IN THE CALCULATION
C IGPIXEL = NUMBER OF PIXELS IN A 'G' LENGTH
C IDPIXEL = NUMBER OF PIXEL IN A CBED DISC DIAMETER
C IBRAGG = REFLECTION ON THE BRAGG CONDITION
C STBMDR = STARTING BEAM DIRECTION
C BMDSRT = SCALAR DESCRIBING THE BEAM STARTING POINT
C BMEND = SCALAR DESCRIBING THE BEAM ENDING POINT
C
DIMENSION A(200,200),TEMP1(3),XDSH(200,200)
COMMON /BLK1/ XSI(200),XSIDSH(200),W(200),ALATT,CLATT,T,V,
A IMGBM(5),NIMGBM,EXPFL(300,5),DEVRG,
B CALC(300,5),MAXEXP,NORMBM,IVAR(35),
C ILOR(5,5),IHIR(5,5),BKG(11),IFLAGEXTN,IAFR
COMMON /BLK3/ BMDIR(3),IG(3),IVECY(3),IBEAM(200,3),
A IBRAGG,NBEAMS,IGPIXEL,IDPIXEL,IZAXIS(3)
COMMON /BLK6/ V1,AK,BMDSRT,BMEND,DAKX,STBMDR(3),GV(3),ZAXIS(3)
A ,A,BEAMS(200,3),G,XDSH,ANG,R,ISTEP,DIF
C
DATA MAXBM/200/
C
C CONVERT K7 TO VOLTS
C
V1=V*1000.
C
C WAVE VECTOR (CONSTANTS FROM EADES ET AL.)
C
AK=sqrt(V1*(1.0+0.97848E-6*V1))/1.2264
C
C SIZE OF G
C
G=sqrt((IG(1)**2+IG(2)**2)/(ALATT**2)+(IG(3)**2)/(CLATT**2))
C
C SIZE OF GIBRAGG = |IBRAGG*GV|
C
GIBRAGG=sqrt(((IG(1)*IBRAGG)**2+(IG(2)*IBRAGG)**2)/(ALATT**2)+
A((IG(3)*IBRAGG)**2)/(CLATT**2))
C
C CALCULATE THE BRAGG ANGLE
C
BRAGG = ASIN(GIBRAGG/2/AK)
COSBragg = COS(BRAGG)*AK
SINBRAGG = -1*SIN(BRAGG)*AK
C
C CALCULATE THE ANGULAR LENGTH IN MRAD BETWEEN PIXELS
ANG=2.0*BRAGG*1000./IGPIXEL
C
C ANGULAR RADIUS R IS SET FOR 600 MM CAMARA LENTGY &
C 0.2MM APERTURE DIAMETER IN MRAD
C
R =0.333333333/2
C NO. OF STEPS ON EITHER SIDE OVER WHICH THE INTEGRATION IS PERFORMED
C
ISTEP = IAPR*(0.33333333/ANG/2.)
C
C DIF IS QUANTITY LEFT IN EXCESS OF THE STEPS
C
IF(IAPR.EQ.0)GO TO 1
DIF=0.33333333/2.- FLOAT(ISTEP)*ANG/IAPR
C
C DETERMINE BEAM DIRECTION
C
C BMDIR = COSBRAG*ZAXIS + SINBRAG*GV
  1 CALL DIRNM(IZAXIS,ZAXIS)
    CALL NORM(ZAXIS)
    CALL RECNM(IG,GV)
    CALL NORM(GV)
    CALL SCALVEC(ZAXIS,COSBRAG)
    CALL SCALVEC(GV,SINBRAG)
    CALL ADDVEC(ZAXIS,GV,BMDIR)
C
C NOW WE HAVE VECTOR BMDIR(3) WHICH REPRESENTS THE INCIDENT BEAM DI-
C WITH THE LENGTH OF 1./LAMBDA [NM**(-1)]
C
DO 30 I=1,NBEAMS
C
C "BEAMS(I,3)" ARE RL VECTORS (THOSE, THAT WE INCLUDE IN CALCULATIONS),
C IN THE UNITS OF NM**(-1)
C
BEAMS(I,1) = FLOAT( IBEAM(I,1) ) / ALATT
BEAMS(I,2) = FLOAT( IBEAM(I,2) ) / ALATT
BEAMS(I,3) = FLOAT( IBEAM(I,3) ) / CLATT
30 CONTINUE
C
C CALCULATE BMDSRT AND BMEND:
C "BMDSRT" IS LENGTH OF VECTOR FROM THE CENTER TO THE LEFT END OF BF
DISK (-)
C "BMEND" IS LENGTH OF VECTOR FROM THE CENTER TO THE RIGHT END OF BF
DISK (+)
C ORIGIN IS TAKEN IN THE MIDDLE OF BF DISK (000).
C
BMDSRT = - 0.5 * FLOAT(IDPIXEL) /FLOAT(IGPIXEL) * G
BMEND = + 0.5 * FLOAT(IDPIXEL) /FLOAT(IGPIXEL) * G
C
DAKX = (BMEND-BMDSRT)/(IDPIXEL-1)
C
C DETERMINE THE STARTING INCIDENT BEAM DIRECTION
C
CALL RECNM(IG,GV)
CALL NORM(GV)
CALL SCALENEW(GV, TEMP1, BMDSR)
C
C NOW TEMP1(3) IS A VECTOR FROM CENTER OF 000 DISK TO LEFT (STARTING)
C EDGE OF THE DISK. UNITS => NMI(-1)
C
CALL ADDVEC(TEMP1, BMDIR, STBMDR)
C
STBMDR(3) IS A VECTOR FROM THE EWALL SPHERE CENTER TO
C THE STARTING POINT (LEFT EDGE OF BF 000 DISK).
C
TWOPI=6.283185307
CALL OFF_DIAGN(LENGTH, IBEAM, A, XDSH)
C
RETURN
END
C
C SUBROUTINE FOR INTENSITY CALCULATION CHISQ DETERMINATION
C
SUBROUTINE FCN(NVAR, XGUESS, F)
C
REAL TEMP(3), BEAM(3)
C
COMPLEX CE(200), CU
C
DIMENSION A(200,200), AC(200,200), GAMMA(200), Q(200), XGUESS(35)
A, XDSH(200,200), CAL(900,5);
COMMON /BLK1/ XSI(200), XSIDS(200), W(200), ALATT, CLATT, TV,
A IMGBM(5), NIMBM, EXPTL(300,5), DEVBKG,
B CALC(300,5), MAXEXP, NORMBM, IVAR(35),
C ILOR(5,5), IHIR(5,5), BKG(11), IFLAGEXTN, IAPR
COMMON /BLK2/ NREAD1, NREAD2, NRITE1, NRITE2
COMMON /BLK3/ BMDIR(3), IG(3), IVECY(3), IBEAM(200,3),
A IBRAGG, NBEAMS, IGPIXEL, IDPIXEL, IZAXIS(3)
COMMON /BLK6/ V1, AK, BMDSR, BMDEND, DAKX, STBMDR(3), CV(3), ZAXIS(3),
A A, BEAMS(200,3), G, XDSH, ANG, R, ISTEP, DIF
C
DATA MAXBM/200/
TWOPI=6.283185307
C
C REMOVE
C
WRITE(9,902)NVAR, (XGUESS(I), I=1, NVAR)
C
C ASSIGN VARYING PARAMETERS
C
DO1=1,NVAR
IF(IVAR(I), EQ.1)T=XGUESS(I)
IF(IVAR(I), GT.1.AND. IVAR(I), LT.12)XSI(IVAR(I))=XGUESS(I)
IF(IVAR(I), GT.11.AND. IVAR(I), LT.22)XSIDS(IVAR(I)-10)=XGUESS(I)
IF(INVARI.GT.21.AND.INVAR.LT.33)BKG(INVARI-21)=XGUSS(I)
1 IF(INVARI.EQ.33)DEVBG=XGUSS(I)

C SET UP THE OFF-DIAGONAL ELEMENTS OF THE DYNAMICAL MATRIX
C
CALL UOFFSET_DVAGINAL(NBEAMS,IBEAM,A,XDSH)

C SET UP THE DIAGONAL ELEMENTS
C
C ORIENTATION LOOP
C
C IF IAPR = 0 NO APERTURE INTEGRATION
C
IFAC=IAPR
IF(IAPR.EQ.0)IFAC=1
F=0.

C DEPENDING 'IAPR' INCREASE THE NO. OF ORIENTATIONS TO BE CALCULATED
C IF (IAPR =0) THEN 'ISTEP' IS ALSO ZERO.
C
DO2I=1,IDPIXEL*IFAC+2*ISTEP

C ADD PERCENTAGE DEVIATION FROM BRAGG :DEVBG
C
AKX=(I-1-ISTEP)*(DAXX/IFAC) + DEVBG/IGPIXEL

C DETERMINE THE K VECTOR
C GV IS IN UNIT ( 1 NM**(-1) LONG) IN THE G-DIRECTION
C AKX IS CURRENT INCREMENT OF ORIENTATION IN NM**(-1) .
C GV*AKX IS THE INCREMENTAL VECTOR
C
CALL SCALEVE(new(GV,TEMP,AKX)

C ADD INCREMENTAL VECTOR TO STBMDR TO GET NEW BMIDR
C
CALL ADDVECC(STBMDR,TEMP,BMIDR)

C SET THE LENGTH OF NEW BMIDR TO AK
CALL NORM(BMIDR)
CALL SCALEVECC(2MDR,AK)

C SET UP THE DIAGONAL ELEMENTS OF DYNAMICAL MATRIX
C
DO3J=1,NBEAMS
BEAM(1)= BEAMS(J,1)
BEAM(2)= BEAMS(J,2)
BEAM(3)= BEAMS(J,3)

C SIZE OF THE BEAM
C
CALL ADDVEC(BMDIR, BEAM, TEMP)
C
C DETERMINE EXCITATION PARAMETER-SG
C
SG = (AK**2 - SCALAR(TEMP, TEMP)) / (2. * AK)
C
IF(J.EQ.1) SG = 0.
A(J,J) = SG
C
C SET DIAGONAL ELEMENTS OF XDSH TO (1/Mean ABSORPTION POTENTIAL) = XSIDS0
C
XDSH(J,J) = XSIDS0(1)
3 CONTINUE
C
WRITE(*,1000)((A(L,J),L=1,NBEAMS),J=1,NBEAMS)
C
STOP
1000 FORMAT(10F8.4)
C
CALL EVCSF(NBEAMS, A, MAXBM, GAMMA, AC, MAXSM)
C
C WORK OUT ABSORPTION COEFFICIENTS (Ref: HUMPHREYS PAPER IN TEM TECHNIQUES
C BOOK EDITED BY J. M. COWLEY)
C
DO5J=1,NBEAMS
Q(J) = 0.
DO5K=1,NBEAMS
DO5L=1,NBEAMS
Q(J) = Q(J) + AC(K,J)*AC(L,J) / 2. / XDSH(K,L)
5 CONTINUE
C
C COMPLEX PHASE FACTOR
C
DO6J=1,NBEAMS
6 CE(J) = CEXP(COMPLEX(-TWOPI*T*Q(J), TWOPI*T*GAMMA(J)))
C
C DETERMINE INTENSITIES: SEE ZUO AND SPENCE BOOK ON MICRODIFFRACTION
C
DO7J=1,NIMGBM
CU = COMPLEX(0., 0.)
DO7K=1,NBEAMS
CU = CU + AC(J,K)*CE(K)*AC(1,K)
7 CAL(I,J) = REAL(CU*CONJG(CU))
C
2 CONTINUE
C
C IF IAPR = 0 NO APERTURE INTEGRATION
C
IF(IAPR.EQ.0) GOTO 9
C
INTEGRATE OVER THE CIRCULAR APERTURE

KOUNT=0
DO8I=ISTEP+1,(IAPR*IDPIXEL+ISTEP),IAPR
KOUNT =KOUNT+1
DO8J=1,NIMGBM
CALC(KOUNT,J)=0.0
DO8JL=ISTEP,-(ISTEP-1),-1
HH1=CAL(I-JL,J)
HH2=CAL(I-JL+1,J)
X1=JL*ANG/IAPR
X2=(JL-1)*ANG/IAPR
M=(HH1-HH2)/(X1-X2)
H1=HH1-M*X1
N2=M*H+H1
IF(JL.EQ.-ISTEP)THEN
X1= X1+DIF
ELSEIF(JL.EQ.(ISTEP-1))THEN
X2= X2-DIF
ENDIF

FOR MORE DETAILS ABOUT INTEGRATION HERE SEE SWAMINATHAN'S THESIS

CALC(KOUNT,J)=CALC(KOUNT,J)+H1*AREA(X1,X2)+
A((H1-H2)/R)*R**2*(X1-X2)-(X1-X2)**3/3
8 CONTINUE

GOTO 10

DO1J=1,NIMGBM
DO1I=1,IDPIXEL
11 CALC(I,J)=CAL(I,J)

NORMALISE TO EXPERIMENTAL DATA

FACTOR=(EXPTL(MAXEXP,NORMBM)-BKG(NORMBM))/CALC(MAXEXP,NORMBM)

EVALUATE SUM OF SQUARES OF DEVIATIONS:

F= SUM((EXPTL-CALC)**2/EXPTL)/NPOIN

NPOIN=0
DO12I=1,IDPIXEL
DO12J=1,NIMGBM
DO13IE=1,5
IF(I.GE.ILOR(J,IE).AND.I.LE.IHIR(J,IE))THEN
CALC(I,J)=CALC(I,J)*FACTOR+BKG(J)
GOTO12
ENDIF
13 CONTINUE
CALC(I,J)=CALC(I,J)*FACTOR+BKG(J)
F=F+(EXPTL(I,J)-CALC(I,J))**2/EXPTL(I,J)
NPOIN=NPOIN+1
12 CONTINUE
F=F/NPOIN
C
C REMOVE
C
WRITE(9,901)F
IF(.TRUE.) RETURN
C
C WRITE CALCULATED PROFILE ON SECOND CHANNEL
C
WRITE(NRITE2,1001)NIMGBM,IDPIXEL,IGPIXEL
C
DO41I=1,IDPIXEL
14 WRITE(NRITE2,1002)EXPTL(I,J),J=1,NIMGBM,CALC(I,J),J=1,NIMGBM
IF(.TRUE.) STOP
C
RETURN
901 FORMAT( F =',E15.5)
902 FORMAT( NVAR,(XGUESS(I),I=1,NVAR),I5,5E15.7)
1001 FORMAT(2I5,2F10.5)
1002 FORMAT(10(2X,F13.5))
END
C
C CALCULATE THE AREA OF APERTURE
FUNCTION AREA(X1,X2)
R=0.3333333/2
AREA=ABS(2.*(X2*SQRT(R**2-X2**2)/2.-X1*SQRT(R**2-X1**2)/2.+$+R**2/2*(ASIN(X2/R)-ASIN(X1/R))))
RETURN
END
C
C SUBROUTINE FOR CALCULATION OF OFF DIAGONAL ELEMENT OF THE DYNAMICAL
C MATRIX
C
SUBROUTINE OFF_DIAGONAL(NBEAM,IBEAM,A,XDSH)
CHARACTER*2 ATOMNAME(10),NAME(3)
REAL XSID1S,XSIABS
DIMENSION A(200,200),IBEAM(200,3),BEAM(3),XDSH(200,200)
DIMENSION P1(10),P2(10),P3(10),DW1(10)
COMMON /BLK1/ XSI(200),XSIDSH(200),W(200),ALATT,CLATT,?,V,
A IMGBM(5),NIMGBM,EXPTL(300,5),DEVBG,
B CALC(300,5),MAXEXP,NORMBM,IVAR(35),
C IOR(5,5),IHIR(5,5),BKG(11),IFLAGEXTN,IAPR
COMMON /BLK4/ P1,P2,P3,P4,DW1,IPOS
COMMON /BLK7/ATOMNAME,NAME
INTEGER NBEAM
C
DO2K=1, NBEAM
DO2L=K+1,NBEAM
C
C CALCULATE THE BEAM = G1-G2 VECTOR
C
BEAM(1) = FLOAT( IBEAM(K,1)-IBEAM(L,1) )
BEAM(2) = FLOAT( IBEAM(K,2)-IBEAM(L,2) )
BEAM(3) = FLOAT( IBEAM(K,3)-IBEAM(L,3) )
C
C CALCULATE THE EXTINCTION DISTANCE FOR BEAM
C IF IFLAGEXTN=0, THEN GO TO EXTN (BIRD'S ROUTINE);
C IF IFLAGEXTN=1, THEN GO TO EXTN1 (FREE ATOM MODEL);
C IF IFLAGEXTN=2, THEN GO TO EXTN2 (EXP. STR. FACTORS);
C
CALL EXTN(ATOMNAME,P1,P2,P3,DW1,IPOS,BEAM,ALATT,CLATT,V
  $,XSIDIS,XSIABS)
  IF (IFLAGEXTN .EQ. 1) THEN
    CALL EXTN1(ATOMNAME,NMNAME,P1,P2,P3,DW1,IPOS,BEAM,ALATT,CLATT,V,
             $,XSIDIS)
  ELSEIF (IFLAGEXTN .EQ. 2) THEN
    CALL EXTN2(ATOMNAME,NMNAME,P1,P2,P3,DW1,IPOS,BEAM,ALATT,CLATT,V,
             $,XSIDIS)
  ENDIF
C
C SET UP THE DYNAMICAL MATRIX ELEMENTS :1/(2*XSI)
C
A(K,L)=0.5/XSIDIS
A(L,K)=0.5/XSIDIS
C
C SET UP THE ABSORPTION MATRIX ELEMENTS:1/(2*XSIDSH)
C
XDSH(K,L)=XSIABS
XDSH(L,K)=XSIABS
2 CONTINUE
RETURN
END
C
C SUBROUTINE FOR UPDATING THE OFF_DIAGONAL ELEMENTS OF THE DYNAMICAL
C MATRIX
C EVERY ITERATION
C
SUBROUTINE UPOFF_DIAGONAL(NBEAM,IBEAM,A,XDSH)
  CHARACTER*2 ATOMNAME(10),NAME(3)
  REAL XSIDIS
  INTEGER NBEAM
  DIMENSION A(200,200),IBEAM(200,3),BEAM(3),XDSH(200,200)
  DIMENSION P1(10),P2(10),P3(10),DW1(10),ABSBM(10)
  COMMON /BLK1/ XS(200),XSIDSH(200),W(200),ALATT,CLATT,T,V.
  A IMGM(5),NIMGBM,EXPTL(300,5),DEVBRG,
  B CALC(300,5),MAXEXP,NORMB,MIVAR(35),
  C ILOR(5,5),IHIR(5,5),BKKG(11),IFLAGEXTN,IAPR
COMMON /BLK4/ P1,P2,P3,P4,DLW1,IPOS
COMMON /BLK7/ATOMNAME,XNAME
C
C CALCULATE THE (1/D-SPACING) FOR ALL IMAGING BEAMS
C
DO1M=1,NIMGBM
  ABSBM(M) = (IBEAM(M,1)**2 + IBEAM(M,2)**2)/ALATT**2 +
             $(IBEAM(M,3)**2)/CLATT**2
C
C COMPARE THE (1/D-SPACING) VALUES OF IMAGING BEAM WITH ALL THE
C (61-G2) BEAMS OF THE DYNAMICAL MATRIX TO FIND THE EQUIVALENT
C REFLECTION IN THE OFF DIAGONAL AND UPDATE IT WITH NEW VALUE:
C **WARNING** D-SPACING COMPARISON IS NOT FOOL PROOF:
C (E.G) (511) AND (333) FOR SI HAVE SAME D-SPACING:
C
DO2K = 1, NBEAM
DO2L= K+1,NBEAM
  BEAM(1) = F (IBEAM(K,1)-IBEAM(L,1))
  BEAM(2) = F (IBEAM(K,2)-IBEAM(L,2))
  BEAM(3) = F (IBEAM(K,3)-IBEAM(L,3))
  BM = (BEAM(1)**2 + BEAM(2)**2)/ALATT**2 + (BEAM(3)**2)/CLATT**2
  DO3M=1,NIMGBM
  IF(ABSBM(M).EQ.BM) THEN
    IF((ABS(BEAM(1)).EQ.ABS(IBEAM(M,1))).AND.(ABS(BEAM(2)).EQ.
      ABS(IBEAM(M,2))).AND.(ABS(BEAM(3)).EQ.ABS(IBEAM(M,3))).OR.
      B(ABS(BEAM(1))).EQ.ABS(IBEAM(M,2)).AND.(ABS(BEAM(2)).EQ.
      ABS(IBEAM(M,3))).AND.(ABS(BEAM(3)).EQ.ABS(IBEAM(M,1))).OR.
      D(ABS(BEAM(1))).EQ.ABS(IBEAM(M,2)).AND.(ABS(BEAM(2)).EQ.
      ABS(IBEAM(M,3))).AND.(ABS(BEAM(3)).EQ.ABS(IBEAM(M,1))).THEN
      SIGM=K(L)/ABS(A(K,L));
      XSIDI=SIGN*ABS(XSI(M))
      A(K,L)=0.5/XSIDIS
      A(L,K)=0.5/XSIDIS
      SIGDSH=XSISH(K,L)/ABS(XDISH(K,L))
      XDISH(K,L)=SIGDSH*ABS(XSIDH(M))
      XDISH(L,K)=SIGDSH*ABS(XSIDH(M))
    ENDIF
    ENDF
  CONTINUE
C
RETURN
END
APPENDIX E : FIT_COMP - Program for Matching Rocking Curves

The main routine of the program, FIT_COMP, for matching rocking curves is given in this section. FIT_COMP uses complex structure matrix formulation to treat anomalous absorption. This program utilizes routine EVCCG and BCONF (U4INF and LINCG) from the IMSL FORTRAN library for the diagonalization of the structure matrix and the $\chi^2$ minimization. In addition to the IMSL library, routines given in Appendix I should also be linked during compilation.
PROGRAM FIT_COMP
C COMPLEX MATRIX FORMULATION:
C BEAM DIRECTION IS INTO THE SPECIMEN: DOWNWARDS IN A TEM
C APERTURE CORRECTION OPTION INCLUDED: CAMARA LENGTH =600MM
C : APERTURE SIZE=0.2MM
C APERTURE CORRECTION IS NOT PERFORMED IN DIRECTION PARALLEL
C TO THE KIKUCHI LINES INTENSITY IS ASSUMED CONSTANT IN THIS
C DIRECTION
C COMPATIBLE WITH: SUN-OS, CRAY-UNICOS, AND VAX-VMS
C LIBRARIES REQUIRED : IMSL-MATH LIBRARY
C OPTION: IFLAGEXTN = 2 IS DESIGNED ESPECIALLY FOR SI ROOMT
C
C PROGRAM WORK FOR CUBIC AND TETRAGONAL CRYSTAL STRUCTURES
C
C VARIABLES:
C NATOMS NO. OF ATOM TYPES IN THE UNIT CELL
C IPOS NO. OF ATOMIC POSITIONS IN THE UNIT CELL
C IZ ATOMIC NUMBER
C NAME ATOMIC SYMBOL
C XSI(1 -100) EXTINCTION DISTANCES
C XSIDSH(1 -100) ABSORPTION DISTANCES
C ALATT,CLATT LATTICE PARAMETER
C T FOIL THICKNESS
C V BEAM VOLTAGE (KV)
C MASK DEFINES WHICH PARAMETERS VARY
C 1 THICKNESS
C 2-11 XSI(1-50)
C 12-21 XSIDSH(1-50)
C 22-32 BKG(1-11)
C
C EXPTL HOLDS EXPERIMENTAL INTENSITIES
C MAXEXP MAXIMUM NUMBER OF EXPERIMENTAL INTENSITIES (PER BEAM)
C IDPIXEL NUMBER OF EXPERIMENTAL POINTS (PER REFLECTION)
C
DIMENSION XGUESS(35),XSCLAE(35),IPARAM(7),RPARAM(7),X(35),
A MASK(35),XLB(35),XUB(35)
C
CHARACTER*2 ATOMNAME(10),NAME(3)
DIMENSION P1(10),P2(10),P3(10),DW1(10),BEAM(3)
EXTERNAL UMINF.U4INF, FCN
C
COMMON /BLK1/ XSI(200),XSIDSH(200),W(200),ALATT,CLATT,T,V,
A IMGBM(5),NIMGBM,EXPTL(300,5),DEVBRG,
B CALC(300,5),MAXEXP,NORMBM,IVAR(35),
C ILOR(5,5),IHIR(5,5),BKG(11),IFLAGEXTN,IAPR
COMMON /BLK2/ NREAD1,NREAD2,NRTE1,NRTE2
COMMON /BLK3/ BMDIR(3),IG(3),IVECY(3),IBEAM(200,3),
A IBRAGG,NBEAMS,IGPIXEL,IPDIXEL,IZAXIS(3)
COMMON /BLK4/ P1,P2,P3,P4,DW1,IPOS
COMMON /BLK5/ IZ(2),AX(2,4),BX(2,4),CC(2),NATOMS
COMMON /BLK7/ATOMNAME,NAME
COMMON /H/NLAUE
C
DATA MAXEXP/1000/
C
C SET UP INPUT AND OUTPUT CHANNELS
C
NREAD1=7
NREAD2=8
NRITE1=9
NRITE2=10
OPEN(UNIT=NREAD1,FILE='FITNSYS.DAT1',STATUS='OLD')
OPEN(UNIT=NREAD2,FILE='FIT.DAT2',STATUS='OLD')
OPEN(UNIT=NRITE1,FILE='FIT.OUT1',STATUS='UNKNOWN')
OPEN(UNIT=NRITE2,FILE='FIT.OUT2',STATUS='UNKNOWN')
WRITE(NRITE1,895)
C
C INPUT DIFFRACTION DATA ON FIRST CHANNEL (NREAD1)
READ(NREAD1,700)
C READ NO. OF ATOMIC POSITIONS AND THE NATOMS
READ(NREAD1,705)IPOS,NATOMS
WRITE(NRITE1,896)IPOS,NATOMS
C AX, BX, C : H-F ATOMIC SCATTERING FACTORS
READ(NREAD1,700)
WRITE(NRITE1,897)
C READ ATOMIC NUMBER AND FREE ATOM SCATTERING FACTORS FOR "NATOMS"
DO120I=1,NATOMS
READ(NREAD1,706)NAME(I),IZ(I),(AX(I,J),BX(I,J),J=1,4),CC(I)
WRITE(NRITE1,706)NAME(I),IZ(I),(AX(I,J),BX(I,J),J=1,4),CC(I)
120 CONTINUE
READ(NREAD1,700)
WRITE(NRITE1,898)
C READ ATOMNAME, ATOMIC POSITIONS P1,P2,AND P3,
C DEBYE WALLER FACTORS IN "NM"
DO121I=1,IPOS
READ(NREAD1,707)ATOMNAME(I),P1(I),P2(I),P3(I),DW1(I)
WRITE(NRITE1,899)ATOMNAME(I),P1(I),P2(I),P3(I),DW1(I)
121 CONTINUE
C READ LATTICE PARAMETERS, THICKNESS IN NM,VOLTAGE IN KV
C DEVRG: DEVIATION FROM BRAGG CONDITION IN PIXELS
READ(NREAD1,700)
READ(NREAD1,702) ALATT,CLATT,T,V,DEVRG
WRITE(NRITE1,903)ALATT,CLATT,T,V,DEVRG
C
C READ BASIC IG = G-VECTOR: IBRAGG = THE G IN BRAGG:
C IZAXIS = ZONE AXIS: IGPIXEL = NO. OF PIXEL IN G:
C IDPIXEL= NO. OF PIXELS IN DIAMETER OF DISC:
C NBEAMS = NO. OF BEAMS OR REFLECTIONS
C :
==> IF NBEAMS = 0 THEN GOTO 119 AND
C USE ROUTINE BEAMS TO DETERMINE REFLECTIONS:
C IFLAGEXTN IS THE FLAG FOR PROPER EXTINCTION DISTANCE ROUTINE
C IF IFLAGEXTN=0, THEN USE EXTN (BIRD'S ROUTINE);
C IF IFLAGEXTN=1, THEN USE EXTN1 (H-F FREE ATOM MODEL);
C IF IFLAGEXTN=2, THEN USE EXTN2 (EXP. STR. FACTORS);
C
READ(NREAD1,700)
READ(NREAD1,920) IG,IBRAGG,IZAXIS,IGPIXEL,IPPIXEL,NBEAMS,
AIFLAGEXTN,IAPR
WRITE(NRITE1,921) IG,IBRAGG,IZAXIS,IGPIXEL,IPPIXEL,NBEAMS,
AIAPR,IFLAGEXTN
READ(NREAD1,700)
IF(NBEAMS.EQ.0)GOTO119
C IF NBEAMS .NE.0 THEN READ IBEAM: ARRAY IN WHICH BEAMS ARE STORED;
DO1221=1,NBEAMS
READ(NREAD1,708) (IBEAM(I,J),J=1,3)
BEAM(1)=FLOAT(IBEAM(I,1))
BEAM(2)=FLOAT(IBEAM(I,2))
BEAM(3)=FLOAT(IBEAM(I,3))
C
C IF IFLAGEXTN=0, THEN USE EXTN (BIRD'S ROUTINE);
C IF IFLAGEXTN=1, THEN USE EXTN1 (H-F FREE ATOM MODEL);
C IF IFLAGEXTN=2, THEN USE EXTN2 (EXP. STR. FACTORS);
C
CALL EXTN(ATOMNAME,P1,P2,P3,DW1,IPCS,BEAM,ALATT,CLATT,V
$ ,XSI(I),XSIDSH(I))
C
C AVOID EXTINCTION CALCULATION FOR 000 REFLECTION
IF(I.EQ.1)GOTO 122
C
IF (IFLAGEXTN .EQ. 1) THEN
CALL EXTN1(ATOMNAME,NAME,P1,P2,P3,DW1,IPOS,BEAM,ALATT,CLATT,V,
$ ,XSI(I))
ELSEIF (IFLAGEXTN .EQ. 2 ) THEN
CALL EXTN2(ATOMNAME,NAME,P1,P2,P3,DW1,IPOS,BEAM,ALATT,CLATT,V,
$ ,XSI(I))
ENDIF
122 CONTINUE
C
119 READ (NREAD1,700)
C
C READ WMAX = SG*XSI, THE DYNAMICAL INTERACTION PARAMETERS
C NLAUE FORMAT (18) (FIG 4.24): THE NUMBER OF LAUE ZONES
C USE VERY HIGH NLAUE ZONE IF YOU WANT TO USE JUST
C WMAX = SG*XSI CRITERIA (UNMODIFIED BEAM SELECTION CRITERIA)
C
C
READ (NREAD1,709) WMAX,NLAUE
C
READ(7,700,
READ(NREAD1,705) (IMGBM(I),I=1,5)
C DETERMINE HOW MANY IMAGING BEAMS: NIMGBM
C
DO1I=2,5
IF(IMGBM(I).EQ.0)GO TO 2
CONTINUE
I=6
2 NIMGBM=I-1
WRITE(NRITE1,906) (IMGBM(I),I=1,NIMGBM)

IF (NBEAMS.GT.0) GO TO 123
CALL BEAMS(WMAX)
WRITE(NRITE1,931)NBEAMS,WMAX
C
123 WRITE(NRITE1,928)
DO 124 I=1,NBEAMS
124 WRITE(NRITE1,926) (IBEAM(I,J),J=1,3),XSI(I),XSIDSH(I)
C
C READ MASK: MASK(1)  = THICKNESS;
C                  MASK(2-11) = XSI;
C                  MASK(12-22) = XSIDSH;
C                  MASK(23-32) = BKG;
C                  MASK(33)  = BRAGG ANGLE VARIATION;
C                  MASK(34-35) = FOR FUTURE USE;
C
READ(NREAD1,700)
READ(NREAD1,703)MASK
WRITE(NRITE1,904)
IF(MASK(1).NE.0)WRITE(NRITE1,914)
DO13I=2,11
IF(MASK(I).NE.0)WRITE(NRITE1,915)I-1
J=I+10
IF(MASK(J).NE.0)WRITE(NRITE1,916)J-11
K=I+20
IF(MASK(K).NE.0)WRITE(NRITE1,922)K-22
L=I+32
IF(MASK(L).NE.0)WRITE(NRITE1,923)L-32
13 CONTINUE
C
C INPUT ROCKING CURVE DATA ON SECOND CHANNEL (NREAD2)
C
READ(7,700)
WRITE(NRITE1,929)
C
C READ BEAMS INCLUDED IN THE SCAN, INITIAL VALUES OF
C XSI,XSIDSH AND BKG:
C
WRITE(NRITE1,924)
DO 125 I=1,NIMGBM
READ(NREAD1,708) (IBEAM(I,J),J=1,3),XSI(I),XSIDSH(I),BKG(I)
125 WRITE(NRITE1,926) (IBEAM(I,J),J=1,3),XSI(I),XSIDSHE(I),BKG(I)
C READ IN EXPERIMENTAL RANGES OF POINTS TO BE EXCLUDED FROM THE FIT:
C
READ(NREAD1,700)
READ(NREAD1,705) (( ILOR(JB,IE), IE=1,5 ), JB=1,NIMGBM)
READ(NREAD1,700)
READ(NREAD1,705) (( IHIR(JB,IE), IE=1,5 ), JB=1,NIMGBM)
DO16I=1,NIMGBM
16 WRITE(9,918)I-1, ( ILOR(I,JE),IHIR(I,JE), JE=1,5)
READ(NREAD2,700)
C READ DATA IN THE INVERTED ORDER:
DO18I=IDPIXEL,1,-1
19 READ(NREAD2,801)(EXPTL(I,J),J=1,NIMGBM)
C CHECK VALIDITY OF DATA
C
DO3I=1,NIMGBM
DO 3 JE=1,5
IF(ILOR(I,JE).LT.0.OR.IHOR(I,JE).GT.IDPIXEL)GOTO5
IF(IHOR(I,JE).LT.0.OR.IHOR(I,JE).GT.IDPIXEL)GOTO5
GOTO3
5 WRITE(NRITE1,917)
STOP
3 CONTINUE
IF(IDPIXEL.GT.1)GOTO17
WRITE(NRITE1,908)
STOP
17 IF(IDPIXEL.LE.MAXEXP)GOTO12
WRITE(NRITE1,907)
STOP
12 CONTINUE
C LOCATE MAXIMUM INTENSITY
C
EXPMAX=0.
DO20I=1,NIMGBM
DO1IJ=1,IDPIXEL
IF(EXPTL(J,I).LT.EXPMAX)GOTO11
EXPMAX=EXPTL(J,I)
MAXEXP=J
NORMBM=I
11 CONTINUE
20 CONTINUE
C IF PREFERRED, READ IN
C
READ(NREAD1,700)
READ(NREAD1,705)ITEMP1,ITEMP2
IF(ITEMP1.EQ.0.AND.ITEMP2.EQ.0)GOTO21
DO22 I=1,NIMGBM
IF (ITEMP1.NE.IMGBM(I)) GOTO22
NORMBM=I
22 CONTINUE
MAXEXP=ITEMP2
21 CONTINUE
C
WRITE(NRITE1,919)MAXEXP,IMGBM(NORMBM)
C
C SET UP DIFFRACTION CALCULATION
C
CALL DIFFR
WRITE(NRITE1,927)BMDIR
C
C IDENTIFY WHICH PARAMETERS ARE TO VARY
C
J=0
DO6 I=1,35
IF (MASK(I).EQ.0) GOTO6
J=J+1
IVAR(J)=I
6 CONTINUE
IF (J.GT.0) GOTO7
WRITE(NRITE1,909)
STOP
7 NVAR=J
C
C INITIALISE ORIGINAL GUESSED VALUES
C
DO8 I=1,NVAR
IF (IVAR(I).EQ.1) XGUESS(I)=T
IF (IVAR(I).GT.1 .AND. IVAR(I).LT.12) XGUESS(I)=XSI(IVAR(I))
IF (IVAR(I).GT.11 .AND. IVAR(I).LT.22) XGUESS(I)=XSIDSH(IVAR(I)-10)
IF (IVAR(I).GT.21 .AND. IVAR(I).LT.33) XGUESS(I)=BKG(IVAR(I)-21)
8 IF (IVAR(I).EQ.33) XGUESS(I)=DEVBRG
C
C SET UP REST OF MINIMISATION ROUTINE
C
C CALL U4INF TO SET UP PARMETERS IPARAM AND RPARAM ITERATIONS
C REFER IMSL MANUAL
C
CALL U4INF(IPARAM,RPARAM)
C
DO9 I=1,NVAR
9 XSCALE(I)=1.0
C
FSIZE=1.0
READ(NREAD1,700)
READ(NREAD1,705)IPARAM(3),IPARAM(4),IPARAM(5)
WRITE(*,*)IPARAM
C
C READ STEPL=>RPARAM(2)
C
  rEAD(NREAD1,700)
  READ(NREAD1,*)RPARAM(2)
C
C  RPARAM(5) FALSE CONVERGENCE TOLERANCE
C
C  RPARAM(5)=1.0E-02
C
C  IBTYPE =0; ===> USER WILL SUPPLY THE BOUNDS
C
C  IBTYPE=0
C
C  SET UP THE BOUNDS FOR THE VARIABLES
C
  DO15I=1,NVAR
    SIGN=XGUESS(I)/ABS(XGUESS(I))
    IF(SIGN.GT.0)THEN
      XLB(I)=XGUESS(I)/2.
      XUB(I)=XGUESS(I)*2.
    ELSE
      XUB(I)=XGUESS(I)/2.
      XLB(I)=XGUESS(I)*2.
    ENDF
C  IF DEVRG IS VARIED BOUNDS ARE BETWEEN +1 OR -1
  IF(IVAR(I).EQ.33)THEN
    XLB(I)=-1.
    XUB(I)=1.
  ENDF
15 CONTINUE
C
C  MINIMISATION
C
  CALL BCONF(FCN,NVAR,XGUESS,IBTYPE,XLB,XUB,XSCALE,FSCALE,
      A
      IPARAM,RPARAM,X.FVALUE)
C
C  WRITE MINIMISED PARAMETERS ON FIRST OUTPUT CHANNEL
C
  DO10I=1,NVAR
    IF(IVAR(I).EQ.1)WRITE(NRITE1,910)T
    IF(IVAR(I).GT.1.AND.IVAR(I).LT.12)WRITE(NRITE1,911)IVAR(I)-1,
      AXSI(IVAR(I))
    IF(IVAR(I).GT.11.AND.IVAR(I).LT.22)
      AWRITE(NRITE1,912)IVAR(I)-11,XSIDSH(IVAR(I)-10)
    IF(IVAR(I).GT.21.AND.IVAR(I).LT.33)
      AWRITE(NRITE1,930)IVAR(I)-22,BKG(IVAR(I)-21)
    IF(IVAR(I).EQ.33)WRITE(NRITE1,932)DEVRG
10  CONTINUE
    WRITE(NRITE1,913)FVALUE
C
C  WRITE CALCULATED PROFILE ON SECOND CHANNEL
WRITE(NRITE2,1001)NIMGMB, IDPIXEL
DO41=1, IDPIXEL
4 WRITE(NRITE2,1002) (EXPTL(I,J), J=1, NIMGMB), (CALC(I,J), J=1, NIMGMB)

STOP

700 FORMAT(A80)
701 FORMAT(5F10.0)
702 FORMAT(5F10.0)
703 FORMAT(I2/1012/1012/I2/1012/1012)
704 FORMAT(2F10.0)
705 FORMAT(I5I5)
706 FORMAT(A2, I4, 3F7.4, F8.4, 3F7.4, F8.4, F7.4)
707 FORMAT(A2, 3F6.0, F9.0)
708 FORMAT(I5, 2F10.0, F6.0)
709 FORMAT(F5.0, I8, I5)
801 FORMAT(5F10.0)
895 FORMAT('OUTPUT OF FIT_COMP: ABSORPTION BY COMPLEX MATRIX')
896 FORMAT(3X, 'NO. OF ATOMIC POSTIONS : ', I5, 3X,
A, 'NO. OF ATOM TYPES : ', I5)
897 FORMAT('AX, BX, C : N-F ATOMIC SCATTERING FACTORS')
898 FORMAT('/ELE X Y Z D-W FACTOR/')
899 FORMAT(A2, 3F7.3, F9.4)
901 FORMAT(' INITIAL EXTINCTION DISTANCES: '/(5F10.1))
902 FORMAT(' INITIAL ABSORPTION DISTANCES: '/(5F10.1))
903 FORMAT('/ LATTICE PARAMETER A =', F8.6, ' NM /
A   ', C =', F8.6, ' NM /
B   ' ' FOIL THICKNESS =', F8.3,
C   ' NM '/ BEAM VOLTAGE =', F8.2, ' KV'
D   ', ' DEVIATION FROM BRAGG=', F9.3, ' PIXELS // /)
904 FORMAT('/ MASK DETAILS :')
906 FORMAT('/ IMAGING BEAMS INCLUDED IN THE PROFILE: ', 5I4)
907 FORMAT('/ TOO MANY EXPERIMENTAL POINTS')
908 FORMAT('/ TOO FEW EXPERIMENTAL POINTS')
909 FORMAT('/ NO PARAMETERS SET TO VARY')
910 FORMAT('/ OPTIMISED FOIL THICKNESS =', F12.4)
911 FORMAT('/ THE BEST FIT ', I2, 'TH EXTINCTION DISTANCE IS ', F12.4)
912 FORMAT('/ THE BEST FIT ', I2, 'TH ABSORPTION DISTANCE IS ', F14.4)
913 FORMAT('/ THE MINIMISED SUM OF SquARED DEVIATIONS = ', E15.5)
914 FORMAT('/ THICKNESS IS BEING OPTIMISED')
915 FORMAT('/ THE', I2, 'TH EXTINCTION DISTANCE IS BEING ',
A   'OPTIMISED')
916 FORMAT('/ THE', I2, 'TH ABSORPTION DISTANCE IS BEING ',
A   'OPTIMISED')
917 FORMAT('/ THE NUMBER OF EXPERIMENTAL POINTS (PER BEAM) IS ', I4)
918 FORMAT('/ FOR THE ', I1, 'TH BEAM USED RANGES OF POINTS EXCLUDED',
A   5(/I3, ' TO ', I3))
919 FORMAT('/ THE NORMALISING POINT IS NUMBER ', I3, ' OF THE ')
A I1,'TH BEAM')
920 FORMAT(4I5,3I10,7I5)
921 FORMAT(3X,'G VECTOR : ',3I5/,3X,
A 'BEAM IN BRAGC : ',I5/,3X
B 'ZONE AXIS : ',3I10/,3X,
C 'NO. OF PIXEL IN G : ',I5/,3X, 'NO. OF PIXEL IN THE D : ',
D I5/,3X, 'NUMBER OF BEAMS : ',I5,
E' (IF NBEAMS=0, USE WMAX CRITERIA)/,3X,
F'APERTURE CORRTN. FLAG : ',I5, ' (IF =0 NO CORRECTION)'
G//3X,'SUBROUTINE EXTN',I1,' IS USED FOR XSI CALCULATION',
H ' //3X, 'BEAMS ::')
922 FORMAT(/ ' THE',I2,' TH BACKGROUND IS BEING OPTIMISED')
923 FORMAT(/ ' THE BRAGG ANGLE IS BEING OPTIMISED')
924 FORMAT(/ ' INITIAL VALUES OF VARIABLES FOR THE BEAMS INCLUDED',
A ' IN THE PROFILE:')
925 FORMAT(3X,A2,1X,4F7.4)
926 FORMAT(3I5,2F12.4,2X,F6.1)
927 FORMAT(/, ' INCIDENT BEAM DIRECTION : ',3F7.2,/) /
928 FORMAT(/,' H',' K',' L',' XSI NM ',' XSIDASH NM',
A)
929 FORMAT(/,' H',' K',' L',' XSI NM ',' XSIDSH NM ',
A ' BRG')
930 FORMAT('/ THE BEST FIT ',I2,' TH BEAM BACKGROUND :',F12.4)
931 FORMAT(
A (' NUMBER OF BEAMS INCLUDED IN CALCULATIONS, NBEAMS = ',I6
B ' DYNAMICAL INTERACTION PARAMETER, WMAX = ',
C F8.2/)
932 FORMAT('/ THE BEST FIT FOR DEVIATION FROM BRAGG CONDITION :
A ' MRAD')
1001 FORMAT(3I5,2F10.5)
1002 FORMAT(10(2X,F13.5) )
C C END
C C SUBROUTINE FOR SETTING UP THE DYNAMICAL MATRIX
C C SUBROUTINE DIFFRT
C C A(200,200) = DYNAMICAL MATRIX
C BMDIR(3) = BEAMDIRECTION VECTOR
C GV = G VECTOR
C ZAXIS = ZONE AXIS
C VECY = DEVIATION VECTOR PERPENDICULAR G AND ON ZONE AXIS
C BEAM = BEAMS INCLUDED IN THE CALCULATION
C IGPIXEL = NUMBER OF PIXELS IN A 'G' LENGTH
C IDPIXEL = NUMBER OF PIXEL IN A CBED DISC DIAMETER
C IBRAGG = REFLECTION ON THE BRAGG CONDITION
C STBDR = STARTING BEAM DIRECTION
C BMDSRC = SCALAR DESCRIBING THE BEAM STARTING POINT
C BMEND = SCALAR DESCRIBING THE BEAM ENDING POINT
C
COMPLEX A(200,200)
DIMENSION TEM(3)
COMMON /BLK1/ XI(200),XSIDSH(200),W(200),ALATT,CLATT,T,V,
A IMGEM(5),NIMGEM,EXPTL(300,5),DEVBGRG,
B CALC(300,5),MAXEXP,NORMEM,IVAR(35),
C ILOR(5,5),IHIR(5,5),BKGT(11),IFLAGEXTN,IPR
COMMON /BLK3/ BMDIR(3),IG(3),IVECY(3),IBEAM(200,3),
A IBRAGR,NBEAMS,IGPIXEL,IDPIXEL,IZAXI(3)
COMMON /BLK6/ V1,A,AK,BMDRT,BMDEND,DAK,X,BMDR(3),GV(3),ZAXI(3)
A A,BEAMS(200,3),G,ANG,R,ISTEP,DIF
C
DATA MAXBM/200/
C
C CONVERT KV TO VOLTS
C
V1=V*1000.
C
WAVE VECTOR (CONSTANTS FROM EADES ET AL.)
C
AK=SQR(T(V1*1.0978*1E-6*V1))/1.2264
C
SIZE OF G
C
G=SQR(T((IG(1)**2+IG(2)**2)/(ALAT**2)+IG(3)**2)/(CLAT**2))
C
SIZE OF GBRAGG = |IBRAGR*GV|
C
GIBRAGG=SQR(T(((IG(1)*IBRAGR)**2+(IG(2)*IBRAGR)**2)/(ALAT**2)+
A((IG(3)*IBRAGR)**2)/(CLAT**2))
C
CALCULATE THE BRAGG ANGLE
C
BRAGG = ASIN(GIBRAGG/2/AK)
COSBRAG= COS(BRAGG)*AK
SINBRAG= -1*SIN(BRAGG)*AK
C
CALCULATE THE ANGULAR LENGTH IN MRAD BETWEEN PIXELS
ANG=2.0*BRAGG*1000./IGPIXEL
C
ANGULAR RADIUS R IS SET FOR 600 MM CAMARA LENGTH &
C 0.2MM APERTURE DIAMETER IN MRAD
C
R =0.333333333/2
C
NO. OF STEPS ON EITHER SIDE OVER WHICH THE INTEGRATION IS PERFORMED
C
ISTEP =IAPR*(0.333333333/ANG/2.)
DIF IS QUANTITY LEFT IN EXCESS OF THE STEPS

IF(IAPR.EQ.0) GO TO 1
DIF=0.33333333/2.- FLOAT(ISTEP)*ANG/IAPR

Determine beam direction

BMDIR = COSBRAG*ZAXIS + SINBRAG*GV
1 CALL FLOATVEC(IZAXIS,ZAXIS)
   CALL NORM(ZAXIS)
   CALL FLOATVEC(IG,GV)
   CALL NORM(GV)
   CALL SCALEVEC(ZAXIS,COSBRAG)
   CALL SCALEVEC(GV,SINBRAG)
   CALL ADDVEC(ZAXIS,GV,BMDIR)

Now we have vector BMDIR(3) which represents the incident beam direction with the length of 1./LAMBDAMM(-1)

DO 30 I=1,NBEAMS

"BEAMS(I,3)" are RL vectors (those, that we include in calculations), in the units of NM**(-1)

BEAMS(I,1) = FLOAT(Ibeam(I,1)) / ALATT
BEAMS(I,2) = FLOAT(Ibeam(I,2)) / ALATT
BEAMS(I,3) = FLOAT(Ibeam(I,3)) / CLATT
30 CONTINUE

Calculate BMDSR and BMDEND:
"BMDSR" is length of vector from the center to the left end of BF disk (-)
"BMDEND" is length of vector from the center to the right end of BF disk (+)
Origin is taken in the middle of BF disk (000).

BMDSR = - 0.5 * FLOAT(IDPIXEL) / FLOAT(IDPIXEL) * G
BMDEND = + 0.5 * FLOAT(IDPIXEL) / FLOAT(IDPIXEL) * G

DAKX = (BMDEND-BMDSR)/(IDPIXEL-1)

Determine the starting incident beam direction

CALL RECNM(IG,GV)
   CALL NORM(GV)
   CALL SCALENEW(GV,TEMP1,BMDSR)

Now TEMP1(3) is a vector from center of 000 disk to left (starting) edge of the disk. Units => NM**(-1)
CALL ADDVEC(TEM1,BMDIR,STBMDR)

STBMDR(3) IS A VECTOR FROM THE EWALD SPHERE CENTER TO
THE STARTING POINT (LEFT EDGE OF BF 000 DISK).

TWOPI=6.283185307
CALL OFF_DIAGONAL(NBEAMS,IBEAM,A)

RETURN
END

SUBROUTINE FOR INTENSITY CALCULATION CHISQ DETERMINATION

SUBROUTINE FCN(NVAR,XGUESS,F)

REAL TEMP(3),BEAM(3)

COMPLEX CE(200),CU,A(206,200),AC(200,200),GAMMA(200),
AACINV(200,200)

DIMENSION XGUESS(35),CAL(900,5)
COMMON /BLK1/ XSI(200),XSIDSH(200),W(200),ALATT,CLATT,T,V,
A IMGBM(5),NIMGBM,EXPTL(300,5),DEVBRG,
B CALC(300,5),MAXEXP,NORMBM,IVAR(35),
C IORL(5,5),IHIR(5,5),BKG(11),IFLAGEXTN,IAFR
COMMON /BLK2/ NREAD1,NREAD2,NRITE1,NRITE2
COMMON /BLK3/ BMDIR(3),IG(3),IVECY(3),IBEAM(200,3),
A IBRAGG,NBEAMS,IGPIXEL,IPDPIXEL,IZAXIS(3)
COMMON /BLK6/ V1,AK,BMDSRT,BMDEND,DAXX,STBMDR(3),GV(3),ZAXIS(3),
A A,BEAMS(200,3),G,ANG,R,ISTEP,DIF

DATA MAXBE/200/
TWOPI=6.283185307

REMOVE

WRITE(9,902)NVAR,(XGUESS(I),I=i,NVAR)

ASSIGN VARYING PARAMETERS

DO11=1,NVAR
IF(IVAR(I),EQ.1)T=XGUESS(I)
IF(IVAR(I),GT.1.AND.IVAR(I),LT.12)XSI(IVAR(I))=XGUESS(I)
IF(IVAR(I),GT.11.AND.IVAR(I),LT.22)XSIDSH(IVAR(I)-10)=XGUESS(I)
IF(IVAR(I),GT.21.AND.IVAR(I),LT.33)BKG(IVAR(I)-21)=XGUESS(I)
1 IF(IVAR(I),EQ.33)DEVBRG=XGUESS(I)

SET UP THE OFF-DIAGONAL ELEMENTS OF THE DYNAMICAL MATRIX
CALL UPOFF_DIAGONAL(NBEAMS,I BEAM,A)

C

C SET UP THE DIAGONAL ELEMENTS

C

C ORIENTATION LOOP

C

C IF IAPR = 0 NO APERTURE INTEGRATION

C

IF (IAPR.EQ.0) IFAC=1
F=0.

C

C DEPENDING 'IAPR' INCREASE THE NO. OF ORIENTATIONS TO BE CALCULATED
C IF (IAPR =0) THEN 'ISTEP' IS ALSO ZERO.

C

DO2I=1,IDPIXEL*IFAC+2*ISTEP

C

C ADD PERCENTAGE DEVIATION FROM BRAGG : DEVBRRG

C

AKX=(I-1-ISTEP)*(DAKX/IFAC) + DEVBRRG/IDPIXEL

C

C DETERMINE THE K VECTOR

C GV IS IN UNIT ( 1 NM**(-1) LONG ) IN THE G-DIRECTION
C AKX IS CURRENT INCREMENT OF ORIENTATION ( IN NM**(-1) ).
C GV*AKX IS THE INCREMENTAL VECTOR

C

CALL SCALENEW(GV,TEMP,AKX)

C

C ADD INCREMENTAL VECTOR TO STBMDIR TO GET NEW BMDIR

C

CALL ADDVEC(STBMDIR,TEMP,BMDIR)

C

C SET THE LENGTH OF NEW BMDIR TO AK

C CALL NORM(BMDIR)

C CALL SCALEVEC(BMDIR,AK)

C

C SET UP THE DIAGONAL ELEMENTS OF DYNAMICAL MATRIX

C

DO3J=1,NBEAMS
BEAM(1)= BEAMS(J,1)
BEAM(2)= BEAMS(J,2)
BEAM(3)= BEAMS(J,3)

C

C SIZE OF THE BEAM

C

CALL ADDVEC(BMDIR,BEAM,TEMP)

C

C DETERMINE EXCITATION PARAMETER-SG

C

SG= (AK**2 - SCALAR(TEMP,TEMP) ) /(2. * AK)
C
IF(J.EQ.1)SG=0.
A(J,J)=CMPLX(SG,0.5/XS1DSH(1))
C
3 CONTINUE
WRITE(*,1000)((A(L,J),L=1,NBEAMS),J=1,NBEAMS)
STOP
1000 FORMAT(10F8.4)
C
CALL EVCCG(NBEAMS,A,MAXBM,GAMMA,AC,MAXBM)
CALL LINCG(NBEAMS,AC,MAXBM,ACINV,MAXBM)
C
C COMPLEX PHASE FACTOR
C
DO6J=1,NBEAMS
6 C(J)=CEXP(CMPLX(-TWOPI*T*AIMAG(GAMMA(J)),TWOPI*T*
AREAL(GAMMA(J))))
C
DO7J=1,NMGGBM
CU=CMPLX(0.,0.)
DO7K=1,NBEAMS
CU=CU+AC(J,K)*CE(K)*(ACINV(K,1))
7 CAL(I,J)=REAL(CU*CONJG(CU))
C
2 CONTINUE
C
IF IAPR = 0 NO APERTURE INTEGRATION
C
IF(IAPR.EQ.0) GOTO 9
C
C INTEGRATE OVER THE CIRCULAR APERTURE
C
KOUNT=0
DO8I=ISTEP+1,(IAPR*IDPIXEL+ISTEP),IAPR
KOUNT =KOUNT+1
DO8J=1,NMGGBM
CALC(KOUNT,J)=0.0
DO8JL=ISTEP,-(ISTEP-1),-1
HH1=CAL(I-JL,J)
HH2=CAL(I-JL+1,J)
X1=JL*ANG/IAPR
X2=(JL-1)*ANG/IAPR
M=(HH1-HH2)/(X1-X2)
H1=HH1-M*X1
H2=M*R+H1
IF(JL.EQ.-ISTEP)THEN
X1= X1-DIF
ELSEIF(JL.EQ.(ISTEP-1))THEN
X2= X2+DIF
ENDIF
C
ENDIF
C
C FOR MORE DETAILS ABOUT INTEGRATION HERE SEE SWAMINATHAN'S THESIS
C
CALC(KOUNT,J) = CALC(KOUNT,J) + H1 * AREA(X1,X2) +
A((H1-H2)/R)*(R**2*(X1-X2)-(X1-X2)**3/3)
8  CONTINUE
C
GOTO 10
C
9  DO11J=1,NIMGMB
   DO11I=1,IDPIXEL
   11  CALC(I,J) = CAL(I,J)
C
C NORMALISE TO EXPERIMENTAL DATA
C
10  FACTOR = (EXPTL(MAXEXP,NORMBM) - BKG(NORMBM))/CALC(MAXEXP,NORMBM)
C
C EVALUATE SUM OF SQUARES OF DEVIATIONS:
C F = SUM((EXPTL-CALC)**2/EXPTL)/NPOIN
C
NPOIN=0
DO12I=1,IDPIXEL
DO12J=1,NIMGMB
DO13IE=1,5
IF(I.GE.ILOR(J,IE).AND.I.LE.IHIR(J,IE))THEN
   CALC(I,J) = CALC(I,J)*FACTOR+BKG(J)
GOTO12
ENDIF
13  CONTINUE
CALC(I,J) = CALC(I,J)*FACTOR+BKG(J)
F = F + (EXPTL(I,J) - CALC(I,J))**2/EXPTL(I,J)
NPOIN=NPOIN+1
12  CONTINUE
F = F / NPOIN
C
C REMOVE
C
WRITE(9,901) F
IF(.TRUE.) RETURN
C
C WRITE CALCULATED PROFILE ON SECOND CHANNEL
C
WRITE(NRITE2,1001) NIMGMB, IDPIXEL, IGPIXEL
C
DO14I=1,IDPIXEL
14  WRITE(NRITE2,1002) (EXPTL(I,J), J=1,NIMGMB), (CALC(I,J), J=1,NIMGMB)
IF(.TRUE.) STOP
C
RETURN
C C CALCULATE THE AREA OF APERTURE
 FUNCTION AREA(X1,X2)
   R=0.33333333/2
   AREA=ABS(2.*(X2*SQRT(R**2-X2**2)/2.-X1*SQRT(R**2-X1**2)/2.+$4*R**2/2.*(ASIN(X2/R)-ASIN(X1/R))))
   RETURN
 END
C
C C SUBROUTINE FOR CALCULATION OF OFF DIAGONAL ELEMENT OF THE
C DYNAMICAL MATRIX
C
SUBROUTINE OFF_DIAGNOL(NBEAM,IBEAM,A)
 CHARACTER*2 ATOMNAME(10),NAME(3)
 REAL XSIDIS,XSIABS
 COMPLEX A(200,200)
 DIMENSION IBEAM(200,3),BEAM(3)
 DIMENSION P1(10),P2(10),P3(10),DW1(10)
 COMMON /BLK1/ XS1(200),XSIDSH(200),W(200),ALATT,CLATT,T,V,
 A IMGBM(5),NIMGBM,EXPTL(300,5),DEVBRG,
 B CALC(300,5),MAXEXP,NORMBM,IVAR(35),
 C ILOR(5,5),IHIR(5,5),BKG(11),IFLAGEXTN,IAPR
 COMMON /BLK4/ P1,P2,P3,P4,DW1,IPOS
 COMMON /BLK7/ATOMNAME,NAME
 INTEGER NBEAM

 DO2K=1, NBEAM
 DO2L=K+1,NBEAM

 C C CALCULATE THE BEAM = G1-G2 VECTOR
 C
 BEAM(1)= FLOAT( IBEAM(K,1)-IBEAM(L,1) )
 BEAM(2)= FLOAT( IBEAM(K,2)-IBEAM(L,2) )
 BEAM(3)= FLOAT( IBEAM(K,3)-IBEAM(L,3) )

 C C CALCULATE THE EXTINCTION DISTANCE FOR BEAM
 C IF IFLAGEXTN=0, THEN GO TO  EXTN (BIRD’S ROUTINE);
 C IF IFLAGEXTN=1, THEN GO TO  EXTN1 (FREE ATOM MODEL);
 C IF IFLAGEXTN=2, THEN GO TO  EXTN2 (EXP. STR. FACTORS);
 C
 CALL EXTN(ATOMNAME,P1,P2,P3,DW1,IPOS,BEAM,ALATT,CLATT,V $
 XSIDIS,XSIABS)
 IF (IFLAGEXTN .EQ. 1) THEN
  CALL EXTN1(ATOMNAME,NAME,P1,P2,P3,DW1,IPOS,BEAM,ALATT,CLATT,V, $
 XSIDIS)
ELSEIF (IFLAGEXTN .EQ. 2) THEN
    CALL EXTN2(ATOMNAME,NAMES,P1,P2,P3,DW1,IPSO,BEAM,ALATT,CLATT,V,$XSIDIS)
ENDIF
C
C SET UP THE DYNAMICAL MATRIX ELEMENTS : 1/(2*XI)
A(K,L) = CMPLX(0.5/XSIDIS, 0.5/XSIABS)
A(L,K) = A(K,L)
2 CONTINUE
RETURN
END

C SUBROUTINE FOR UPDATING THE OFF-DIAGONAL ELEMENTS OF THE
C DYNAMICAL MATRIX EVERY ITERATION
C
SUBROUTINE UOFF_DIAGNOL(NBEAM,IBEAM,A)
CHARACTER*2 ATOMNAME(10), NAME(3)
REAL XSIDIS
COMPLEX A(200,200)
INTEGER NBEAM
DIMENSION IBEM(200,3), BEAM(3)
DIMENSION P1(10), P2(10), P3(10), DW1(10), ABSBM(10)
COMMON /BLK1/ XSI(200), XSIDSH(200), W(200), ALATT, CLATT, T, V,
A IMGBM(5), NIMGBM, EXPTL(300,5), DEVRG,
B CALC(300,5), MAXEXP, NORMBM, IVAR(35),
C ILOR(5,5), IHIR(5,5), BKG(11), IFLAGEXTN, IAPR
COMMON /BLK4/ P1,P2,P3,P4,DW1,IPSO
COMMON /BLK7/ ATOMNAME, NAME
C
C CALCULATE THE (1/D-SPACING) FOR ALL IMAGING BEAMS
C
DO1M=1,NIMGBM
ABSBM(M)=(IBEAM(M,1)**2 + IBEAM(M,2)**2)/ALATT**2 +
$(IBEAM(M,3)**2)/CLATT**2
C
C COMPARE THE (1/D-SPACING) VALUES OF IMAGING BEAM WITH ALL THE
C (G1-G2) BEAMS OF THE DYNAMICAL MATRIX TO FIND THE EQUIVALENT
C REFLECTION IN THE OFF-DIAGONAL AND UPDATE IT WITH NEW VALUE:
C **WARNING** D-SPACING COMPARISON IS NOT FOOL PROOF:
C (E.G.) (511) AND (333) FOR SI HAVE SAME D-SPACING:
C
DO2K =1, NBEAM
DO2L= K+1, NBEAM
BEAM(1)= FLOAT( IBEAM(K,1)-IBEAM(L,1) )
BEAM(2)= FLOAT( IBEAM(K,2)-IBEAM(L,2) )
BEAM(3)= FLOAT( IBEAM(K,3)-IBEAM(L,3) )
BM = (BEAM(1)**2 + BEAM(2)**2)/ALATT**2 + (BEAM(3)**2)/CLATT**2
DO3M=1, NIMGBM
IF (ABSBM(M).EQ.BM) THEN
IF ((ABS(BEAM(1))).EQ.ABS(IBEAM(M,1)) .AND. ABS(BEAM(2)).EQ.
AABS(IBEAM(M,2)).AND.ABS(IBEAM(3)).EQ.ABS(IBEAM(M,3))).OR.
B(ABS(IBEAM(1)).EQ.ABS(IBEAM(M,2)).AND.ABS(IBEAM(2))).EQ.
CABS(IBEAM(M,3)).AND.ABS(IBEAM(3)).EQ.ABS(IBEAM(M,1))).OR.
D(ABS(IBEAM(1)).EQ.ABS(IBEAM(M,3)).AND.ABS(IBEAM(2))).EQ.
EABS(IBEAM(M,1)).AND.ABS(IBEAM(3)).EQ.ABS(IBEAM(M,2))).THEN
SIGN=REAL(A(K,L))/ABS(REAL(A(K,L)))
XSIDIS=SIGN*ABS(XSI(M))
XSIABS=SIGN*ABS(XSIDSH(M))
A(K,L)=CMPLX(0.5/XSIDIS,0.5/XSIABS)
A(L,K)=A(K,L)
ENDIF
ENDIF
3 CONTINUE
2 CONTINUE
RETURN
END
APPENDIX F: Example Input to Programs FIT_PERT and FIT_COMP

File: FITNSYS.DAT1

SILICON DATA: ipos, natoms, 2i5
  8  1
AX, BX, C :H-F ATOMIC SCATTERING FACTORS: A2, i4, 3f7.4, f8.4, 3f7.4, f8.4, f7.4
SI  14 6.2915 2.4386 3.0353 32.3337 1.9891 0.6785 1.5410 81.6937 1.1407
Atom, Position, D-W factor: a2, 3f6.0, f9.0
SI -0.125 -0.125 0.125 0.4632
SI -0.375 -0.375 0.375 0.4632
SI -0.125 -0.375 0.375 0.4632
SI -0.125 -0.125 -0.375 0.4632
SI 0.625 0.625 0.625 0.4632
SI 0.625 0.125 0.125 0.4632
SI 0.625 -0.125 -0.125 0.4632

a, b (nm) 2f10.0; t, V (nm, kV) 2f10.0;
.54310 2028 .54310 2028 124.8263 119.45 0.00 0.73606
ig, ibragg ibmdir igpixel, idpixel, nbexam, iflagextn, iaper(13i5)
  2 2 0 1 -1170 1170 2400 115 80 0 1 0

Beams included: #dummy
Wmax, nlaue (f5.0, 18)
5000. 2400 If nbeams=0, BEAMS is called
imgbm(1-ningbm) 5i5
  0 1 0 0 0
mask(1-21) 12/1012/1012
  1
  1 0 0 0 0 0 0 0 0
  1 0 0 0 0 0 0 0 0
  1
  1 0 0 0 0 0 0 0 0
  0 0 0 0 0 0 0 0 0

Beam, extinction distances and background: 3i5, 2f10.0, f6.0
  0 0 0 0.0000 534.2700 59.82
  2 2 0 -83.191-3290.1631 46.78

Low end of excluding ranges 5(5i5) (5 for bf, 5 for df)
  0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0

High end of excluding ranges 5(5i5)
  0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0

Normalising point (beam, point) if required
  1 37
Maximun num. of iterations
5000 5000 5000
STEPL rparam(2)
1.0e-07

220
File: FIT.DAT2

Bright and Dark field rocking curves! Dummy line

1301.000  137.000
1309.000  103.000
1427.000  53.000
1400.000  49.000
1256.000  67.000
1202.000  123.000
1207.000  178.000
1031.000  272.000
 976.000  312.000
 982.000  307.000
1123.000  270.000
1202.000  212.000
1277.000  140.000
1292.000  72.000
1365.000  57.000
1297.000  88.000
1193.000  180.000
1053.000  313.000
1017.000  441.000
 906.000  581.000
 825.000  665.000
 810.000  695.000
 834.000  644.000
 982.000  511.000
1147.000  344.000
1284.000  204.000
1339.000  106.000
1373.000  62.000
1354.000  91.000
1191.000  235.000
 975.000  360.000
 771.000  560.000
 656.000  720.000
 572.000  909.000
 401.000 1060.000
 233.000 1207.000
 201.000 1310.000
 149.000 1433.000
 127.000 1541.000
 128.000 1473.000
 169.000 1499.000
 219.000 1507.000
 340.000 1429.000
 430.000 1359.000
 599.000 1282.000
 755.000 1116.000
 965.000  942.000
| 1084.000 | 712.000 |
| 1300.000 | 561.000 |
| 1477.000 | 425.000 |
| 1623.000 | 258.000 |
| 1747.000 | 110.000 |
| 1751.000 | 74.000  |
| 1635.000 | 84.000  |
| 1675.000 | 168.000 |
| 1398.000 | 291.000 |
| 1364.000 | 458.000 |
| 1318.000 | 522.000 |
| 1242.000 | 673.000 |
| 1277.000 | 608.000 |
| 1348.000 | 547.000 |
| 1447.000 | 450.000 |
| 1501.000 | 318.000 |
| 1623.000 | 168.000 |
| 1711.000 | 109.000 |
| 1796.000 | 62.000  |
| 1702.000 | 76.000  |
| 1644.000 | 141.000 |
| 1452.000 | 238.000 |
| 1426.000 | 304.000 |
| 1459.000 | 336.000 |
| 1473.000 | 364.000 |
| 1527.000 | 294.000 |
| 1488.000 | 175.000 |
| 1566.000 | 123.000 |
| 1581.000 | 70.000  |
| 1690.000 | 42.000  |
| 1634.000 | 77.000  |
| 1519.000 | 100.000 |
| 1500.000 | 157.000 |
APPENDIX G: Example Output of the Program FIT_PERT

File: FIT.OUT1

OUTPUT OF FIT_PERT: ABSORPTION TREATED BY PERTURBATION
NO. OF ATOMIC POSITIONS  :  8
NO. OF ATOM TYPES  :  1

AX, BX, C : H-F ATOMIC SCATTERING FACTORS
SI  14 6.2915 2.4386 3.0353 32.3337 1.9891 0.6785 1.5410 81.6937 1.1407

ELE  X    Y    Z    D-W FACTOR
SI -0.125 -0.125 -0.125  0.4632
SI  0.375  0.375 -0.125  0.4632
ST  0.375 -0.125  0.375  0.4632
SI -0.125  0.375  0.375  0.4632
SI  0.125  0.125  0.125  0.4632
SI  0.625  0.625  0.125  0.4632
SI  0.125  0.625  0.625  0.4632
SI  0.625  0.125  0.625  0.4632

LATTICE PARAMETER A = 0.543102 NM
C = 0.543102 NM
FOIL THICKNESS  = 124.826 NM
BEAM VOLTAGE = 119.45 KV
DEVIAITON FROM BRAGG= 0.000 PIXELS

G VECTOR  :  2 2 0
BEAM IN BRAGG  :  1
ZONE AXIS  :  -11170 11170 2400
NO. OF PIXEL IN G  :  115
NO. OF PIXEL IN THE D  :  86
NUMBER OF BEAMS  :  0 (IF NBEAMS=0, USE WMAX CRITERIA)
APERTURE CORRNL. FLAG  :  0 (IF =0 NO CORRECTION)

SUBROUTINE EXTN1 IS USED FOR XI CALCULATION

BEAMS :

IMAGING BEAMS INCLUDED IN THE PROFILE:  0  1

NUMBER OF BEAMS INCLUDED IN CALCULATIONS, NBEAMS = 64
DYNAMICAL INTERACTION PARAMETER, \[ WMAX = 5000.00 \]

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MASK DETAILS:

THICKNESS IS BEING OPTIMISED

THE 1 TH EXTINCTION DISTANCE IS BEING OPTIMISED

THE 1 TH ABSORPTION DISTANCE IS BEING OPTIMISED

THE 0 TH BACKGROUND IS BEING OPTIMISED

THE 1 TH BACKGROUND IS BEING OPTIMISED

H   K   L   XSI NM   XSIDSH NM   BKG

INITIAL VALUES OF VARIABLES FOR THE BEAMS INCLUDED IN THE PROFILE:

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2   2   0   -83.   -3290.   47.

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0 TO 0
0 TO 0
0 TO 0
0 TO 0

FOR THE 1TH BEAM USED RANGES OF POINTS EXCLUDED

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0 TO 0
0 TO 0
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0 TO 0
THE NORMALISING POINT IS NUMBER 37 OF THE 1TH BEAM

INCIDENT BEAM DIRECTION : -210.04 206.36 44.73

OPTIMISED FOIL THICKNESS = 124.7482
THE BEST FIT 1TH EXTINCTION DISTANCE IS -84.3795
THE BEST FIT 1TH ABSORPTION DISTANCE IS -3290.1597
THE BEST FIT 0 TH BEAM BACKGROUND : 59.7767
THE BEST FIT 1 TH BEAM BACKGROUND : 46.7637
THE MINIMISED SUM OF SQUARED DEVIATIONS = 0.26210E+01

File: FIT.OUT2

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APPENDIX H: Example Output of the Program FIT_COMP

File: FIT.OUT1

OUTPUT OF NSYSR: ABSORPTION TREATED BY PERTURBATION
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NO. OF ATOM TYPES: 1

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NUMBER OF BEAMS: 0 (IF NBEAMS=0, USE WMAX CRITERIA)
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SUBROUTINE EXTN: IS USED FOR XSI CALCULATION

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MASK DETAILS:

THICKNESS IS BEING OPTIMISED

THE 1 TH EXTINCTION DISTANCE IS BEING OPTIMISED

THE 1 TH ABSORPTION DISTANCE IS BEING OPTIMISED

THE 0 TH BACKGROUND IS BEING OPTIMISED

THE 1 TH BACKGROUND IS BEING OPTIMISED

H K L XSI NM XSIDSH NM BKG

INITIAL VALUES OF VARIABLES FOR THE BEAMS INCLUDED IN THE PROFILE:

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FOR THE 1TH BEAM USED RANGES OF POINTS EXCLUDED

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THE NORMALISING POINT IS NUMBER 37 OF THE 1TH BEAM

INCIDENT BEAM DIRECTION : -210.04 206.36 44.73

OPTIMISED FOIL THICKNESS = 124.7760
THE BEST FIT 1TH EXTINCTION DISTANCE IS -84.4152
THE BEST FIT 1TH ABSORPTION DISTANCE IS -3290.1333
THE BEST FIT 0 TH BEAM BACKGROUND = 58.4795
THE BEST FIT 1 TH BEAM BACKGROUND = 50.3041
THE MINIMISED SUM OF SQUARED DEVIATIONS = 0.30153E+01

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APPENDIX I: Common Routines used in SYSPAT, FIT_PERT and FIT_COMP

This section contains subroutines that are common to SYSPAT, FIT_PERT and FIT_COMP. The subroutine ATOM, that is used for the calculation of extinction distance and absorption part of the extinction distance has been obtained from Bird and King (1990).
SUBROUTINE EXTN: CALCULATION OF EXTINCTION DISTANCES AND
XSIDSH OF ANY BEAM GIVEN BY VARIABLE :BEAM, USING ATOM SUBROUTINE BY

SUBROUTINE EXTN_ATOMNAME(P1,P2,P3,DW,IPOS,BEAM,ALAT,CLAT,V0,XSI
$ ,XSIDSH!
CHARACTER ATOMNAME(10)*2
C FORMREAL,FE - REAL PART OF THE FORM FACTOR
C FORMIMAG,FA - IMAG PART OF THE FORM FACTOR
C P1,P2,P3 - ATOMIC POSITIONS
C KV - VOLTAGE IN KV
C S - SIN(THETA)/WAVELENGTH
C ALATT,CLATT - LATTICE PARAMETERS IN NM
C VOL - VOLUME OF UNIT CELL
C VREAL - REAL PART OF THE FOURIER COEFFICIENTS OF CRYSTAL POT.
C VIMAG - IMAG PART OF THE FOURIER COEFFICIENTS OF CRYSTAL POT.
C
INTEGER H, K, L
DIMENSION P1(10),P2(10),P3(10),DW(10),FORMREAL(10),FORMIMAG(10)
DIMENSION BEAM(3)
!
REAL VREAL,VIMAG
DOUBLE PRECISION KV,DW(10),FE,FA,S
PI = 3.141592654
INT =0
H=BEAM(1)
K=BEAM(2)
L=BEAM(3)
!
CALCULATE UNIT CELL VOLUME
DO 13 I=1,10
   DW(I)=DBLE(DW(I))
13 CONTINUE
!
A=ALAT*1.0D+01
C=CLAT*1.0D+01
!
VOL = A*A*C
KV=DBLE(V0)
!
CALCULATE THE ELECTRON STRUCTURE FACTORS
!
CALCULATE THE FORM FACTORS FOR ALL ATOMS/POSITIONS
S= 0.25D0*((H**2+K**2)/(DBLE(A)**2)+(L**2)/(DBLE(C)**2))
S=DSQRT(S)
!
DO 15 I=1,IPOS
FE=0.0D0
FA=0.0D0
CALL ATOM_ATOMNAME(I),DW(I),S,KV,FE,FA,INT
FORMREAL(I)=SNGL(FE)
FORMIMAG(I)=SNGL(FA)
15 CONTINUE
C
VREAL = 0.0
VIMAG = 0.0
DO 16 KM = 1, IPOS
PHAS = 2.0*PI*(H*P1(KM)+K*P2(KM)+L*P3(KM))
VREAL = VREAL + FORMREAL(KM)*COS(PHAS) +
$ FORMIMAG(KM)*SIN(PHAS)
VIMAG = VIMAG - FORMREAL(KM)*SIN(PHAS) +
$ FORMIMAG(KM)*COS(PHAS)
16 CONTINUE
C
VREAL=47.87801152*VREAL/VOL
VIMAG=47.87801152*VIMAG/VOL
C
CONVERT CRYSTAL POTENTIAL (VG) VALUES TO EXTINCTION DISTANCE
V=V0*1000.
C
REF: ELECTRON MICRODIFFRACTION - ZUO & SPENCE BOOK PAGE.33
GAMMA=(1+1.9569346E-06*V)
C
REF: ELECTRON MICRODIFFRACTION - ZUO & SPENCE BOOK PAGE.7
WAVEL=12.2643/SQRT(V*(1.0+0.97845E-06*V))
UGREAL= 0.006648352*GAMMA*VREAL
UGIMAG= 0.006648352*GAMMA*VIMAG
C
EXTINCTION DISTANCE IN NANO METER
C
IF(ABS(UGREAL).LE.1.E-10)THEN
XSI=1.0E+10
ELSE
XSI = 0.1/(UGREAL*WAVEL)
ENDIF
IF(ABS(UGIMAG).LE.1.E-10)THEN
XSISH=1.0E+10
ELSE
XSISH= 0.1/(UGIMAG*WAVEL)
ENDIF
RETURN
END
C
-----------------------------------------------
- C --------------------------------- ATOM - ATOMIC FORM FACTORS SUBROUTINE ---------
-----------------------------------------------
C -------------------------------------------- AUTHOR: QUENTIN KING, SCHOOL OF PHYSICS, BATH UNIVERSITY ----
THIS PROGRAM IS MADE FREELY AVAILABLE TO ALL IN THE ELECTRON
DIFFRACTION COMMUNITY. WE WOULD ASK THAT USERS MAKE REFERENCE
TO THE PAPER THAT DESCRIBES THE BACKGROUND TO THIS SUBROUTINE:

CALL ATOM( ELNAME, M, S, KVOLTS, FE, FA, STATUS )

IN: ELNAME - CH*2 - ELEMENT NAME (UPPER CASE ONLY)
M - D.P. - 8.(PI^2).U^2; U^2 = MEAN SQUARE ATOMIC VIBRATION
S - D.P. - SCATTERING VECTOR (AS IN DOYLE AND TURNER)
KVOLTS - D.P. - ACCELERATING VOLTAGE IN KVOLTS
STATUS - INT. - ERROR CONTROL

OUT: FE - D.P. - ELASTIC PART OF ATOMIC FORM FACTOR * EXP(-MS^2)
FA - D.P. - IMAG PART OF FORM FACTOR (INC. 1/BETA) * EXP(-
MS^2)
STATUS - INT. - RETURNED STATUS

NOTE: IF MS^2 > 6.0 OR S > 6.0 BOTH FE AND FA RETURN 0.0D0
IF M < 0.05 THEN FA RETURNS 0.0D0

ELNAME: ELEMENT NAME, IN UPPER CASE, LEFT JUSTIFIED,
E.G. 'GA', 'AS', 'S', 'P'

KVOLTS: ACCELERATING VOLTAGE IN KEV.
IF KVOLTS = 0.0D0, BETA IS TAKEN AS 1.

STATUS: IF STATUS IS PASSED WITH A NEGATIVE VALUE, THEN SHOULD AN
ERROR OCCUR, THE SUBROUTINE WILL WRITE THE RELEVANT ERROR
MESSAGE TO UNIT -STATUS AND EXECUTION WILL BE TERMINATED.
IF STATUS IS POSITIVE, A CODE NUMBER IS RETURNED IN STATUS
AND EVEN IF AN ERROR HAS OCCURRED, EXECUTION WILL NOT BE
TERMINATED. THE CODES ARE:

0 - SUCCESSFUL
1 - UNKNOWN ELEMENT
2 - M OUT OF RANGE (M < 0 OR M > 2)
3 - S LESS THAN ZERO
4 - KVOLTS OUT OF RANGE (KVOLTS < 0.0 OR KVOLTS > 10.^4)

-----------------------------------------------------------------------------------

SUBROUTINE ATOM( ELNAME, M, S, KVOLTS, FE, FA, STATUS )
-----------------------------------------------------------------------------------

INTEGER IN, JN, DTN
PARAMETER ( IN = 6, JN = 10, DTN = 27 )

DOUBLE PRECISION GM( IN ), GMS2( JN )

INTEGER NUMELS
CHARACTER*294 BLNAMS

PARAMETER ( NUMELS = 98,
               ELNAMS = 'H HE LI BE B C N O F NE NA MG ' +
               ' AL SI P S CL AR K CA SC TI V CR ' +
               ' MN FE CO NI CU ZN GA GE AS SE BR KR ' +
               ' RB SR Y ZR NB MO TC RU RK PD AG CD ' +
               ' IN SN SB TE I XE CS BA LA CE PR ND ' +
               ' PM SM EU GD TB DY HO ER TM YB LU HF ' +
               ' TA W RE OS IR PT AU HG TL PB BI PO ' +
               ' AT RN FR RA AC TH PA U NP PU AM CM ' +
               ' BK CF ' )

INTEGER STATUS, ISTAT, I, I0, I1, J, J0, J1, ELNUM
INTEGER DT( DTN, NUMELS ), MLTFCT( NUMELS ), ADDFCT( NUMELS )
INTEGER COUNT, BFLAG, RFLAG, LFLAG

REAL RF( 4 ), QF( 4, 5 ), FATM( 4 ), F( IN, JN, NUMELS )
REAL RMLT, RADD, EXP, EXPMS2

DOUBLE PRECISION M, S, KVOLTS, FE, FA, DWF, DTS( DTN )
DOUBLE PRECISION MS2, BETA, EIQINT, CINT, DEXP, SQRT

CHARACTER*2 ELNAME
CHARACTER*20 ERRMSG( 4 )

SAVE

DATA GM / 0.05, 0.15, 0.30, 0.70, 1.30, 2.00/
DATA GMS2/ 0.00, 0.005, 0.025, 0.07, 0.20, 0.50, 1.20, 2.00, 3.50, 6.00/

DATA ERRMSG / 'UNKNOWN ELEMENT. ' ,
               ' M OUT OF RANGE. ' ,
               ' S LESS THAN ZERO. ' ,
               ' KVOLTS OUT OF RANGE. ' /

DATA DTS / 0.00, 0.05, 0.10, 0.15, 0.20, 0.25, 0.30, 0.35, 0.40, 0.45, 0.50, 0.60, 0.70, 0.80, 0.90, 1.00, 1.20, 1.40, 1.60, 1.80, 2.00, 2.50, 3.00, 3.50, 4.00, 5.00,
DATA MLTFCST( 1), ADDFCST( 1), (DT(I, 1), I=1, DTN)/3459366, 00672,
+00529, 00510, 00450, 00382, 00311, 00249, 00199, 00160, 00130,
+00107, 00089, 00064, 00048, 00037, 00029, 00024, 00017, 00012,
+00010, 00008, 00007, 00004, 00003, 00002, 00001, 00001, 00001/
DATA ((F(I, J, 1), I=1, IN), J=1, JN)/
+563., 571., 585., 156., 000, 121., 226., 236., 256., 23.2, 54.5, 26.8, 89.2,
+107., 106., 168., 184., 125., 168., 181., 180./

DATA MLTFCST( 2), ADDFCST( 2), (DT(I, 2), I=1, DTN)/1831398, 00993,
+00418, 00410, 00390, 00359, 00323, 00286, 00250, 00217, 00189,
+00164, 00143, 00110, 00086, 00068, 00055, 00046, 00032, 00024,
+00019, 00015, 00012, 00008, 00005, 00004, 00003, 00002, 00001/
DATA ((F(I, J, 2), I=1, IN), J=1, JN)/
+546., 581., 608., 89.0, 109., 128., 193., 245., 280., 000., 721, 16.0, 44.0,
+75.0, 95.1, 115., 116., 114., 117., 128., 134./

DATA MLTFCST( 3), ADDFCST( 3), (DT(I, 3), I=1, DTN)/1270060, 01501,
+03286, 02860, 01879, 01166, 00753, 00526, 00396, 00314, 00259,
+00219, 00186, 00145, 00115, 00093, 00077, 00064, 00046, 00035,
+00028, 00022, 00017, 00011, 00008, 00006, 00004, 00003, 00002/
DATA ((F(I, J, 3), I=1, IN), J=1, JN)/
+568., 599., 617., 118., 116., 163., 213., 267., 294., 000., 22.2, 37.3, 51.0,
+94.6, 115., 122., 131., 118., 133., 136., 147./

DATA MLTFCST( 4), ADDFCST( 4), (DT(I, 4), I=1, DTN)/0967722, 01920,
+03052, 02807, 02237, 01635, 01161, 00832, 00614, 00469, 00371,
+00302, 00253, 00186, 00145, 00117, 00096, 00081, 00059, 00045,
DATA MLTFC(5),ADDFCT(5),(DT(I,5),I=1,DTN)/0787931,02428,
+02794,02638,02250,01791,01377,01048,00803,00625,00497,
+00402,00333,00239,00182,00144,00118,00098,00072,00055,
+00043,00035,00028,00019,00013,00010,00007,00005,00003/
DATA ((F(I,J,5),I=1,IN),J=1,JN)/
+575.,587.,583.,117.,138.,190.,232.,258.,269.,000,20.0,44.6,71.0,
+93.9,107.,121.,128.,121.,136.,141.,147./

DATA MLTFC(6),ADDFCT(6),(DT(I,6),I=1,DTN)/0653425,02832,
+02509,02406,02138,01796,01460,01168,00932,00748,00606,
+00497,00413,00297,00223,00175,00141,00117,00085,00064,
+00051,00041,00034,00022,00016,00012,00009,00006,00004/
DATA ((F(I,J,6),I=1,IN),J=1,JN)/
+943.,888.,858.,852.,932.,995.,999.,970.,931.,906.,444.,520.,545.,
+567.,568.,565.,101.,148.,193.,228.,245.,255.,000,10.7,45.9,69.7,
+87.5,96.2,115.,116.,121.,132.,137.,140./

DATA MLTFC(7),ADDFCT(7),(DT(I,7),I=1,DTN)/0575485,03181,
+02211,02144,01963,01718,01458,01216,01006,00831,00689,
+00575,00484,00353,00266,00208,00167,00137,00098,00074,
+00058,00047,00039,00026,00018,00013,00010,00007,00005/
DATA ((F(I,J,7),I=1,IN),J=1,JN)/
+561.,566.,573.,114.,141.,180.,220.,237.,253.,000,16.5,29.1,64.9,
+78.8,89.3,113.,117.,119.,128.,134.,136./

DATA MLTFC(8),ADDFCT(8),(DT(I,8),I=1,DTN)/0507865,03564,
```
+01983, 01937, 01808, 01625, 01422, 01222, 01040, 00881, 00747,
+00635, 00542, 00403, 00307, 00241, 00193, 00159, 00113, 00085,
+00066, 00053, 00044, 00029, 00020, 00015, 00012, 00008, 00005/
DATA ((F(I,J,8),I=1,IN),J=1,JN)/
+556.,573.,587.,109.,158.,178.,213.,238.,262.,000.,13.7,34.0,59.0,
+74.2,89.3,110.,111.,120.,124.,130.,133./
C
----- F ----- C

DATA MLTFTC( 9), ADDFCT( 9), (DT(I, 9),I=1,DTN)/0456837,04056,
+01801,01767,01671,01532,01371,01206,01049,00908,00784,
+00677,00586,00445,00344,00272,00219,00180,00128,00095,
+00074,00060,00049,00032,00023,00017,00013,00009,00006/
DATA ((F(I,J,9),I=1,IN),J=1,JN)/
+936.,931.,954.,987.,980.,999.,996.,977.,969.,968.,483.,525.,544.,
+567.,590.,609.,118.,156.,188.,219.,250.,289.,000.,19.3,39.5,60.2,
+80.3,98.4,114.,118.,122.,130.,133.,138./
C
----- NE ----- C

DATA MLTFTC(10), ADDFCT(10), (DT(I,10),I=1,DTN)/0402885,04463,
+01652,01626,01552,01443,01313,01176,01043,00918,00805,
+00706,00619,00479,00376,00300,00244,00201,00143,00106,
+00083,00066,00054,00036,00025,00019,00015,00009,00007/
DATA ((F(I,J,10),I=1,IN),J=1,JN)/
+563.,590.,610.,110.,162.,186.,219.,258.,291.,000.,15.5,40.0,61.0,
+82.7,103.,110.,112.,120.,125.,130.,135./
C
----- NA ----- C

DATA MLTFTC(11), ADDFCT(11), (DT(I,11),I=1,DTN)/0361753,04957,
+04778,04138,02967,02099,01594,01295,01095,00946,00827,
+00727,00642,00505,00403,00325,00266,00221,00158,00117,
+00092,00073,00059,00039,00028,00021,00016,00010,00007/
DATA ((F(I,J,11),I=1,IN),J=1,JN)/
+563.,592.,610.,114.,157.,188.,222.,265.,298.,000.,19.4,43.1,61.4,
+88.4,111.,110.,114.,119.,126.,130.,136./
C
----- MG ----- C
```
DATA MLTFCT(12), ADDFCT(12), (DT(I, 12), I=1, DTN) / 0339037, 05314,
+05207, 04717, 03656, 02657, 01953, 01502, 01211, 01012, 00868,
+00757, 00667, 00528, 00425, 00347, 00286, 00239, 00172, 00129,
+00100, 00080, 00065, 00043, 00030, 00022, 00017, 00011, 00008/
DATA ((F(I, J, 12), I=1, IN), J=1, JN) /
+208, 241, 276, 338, 397, 444, 299, 322, 340, 382, 428, 468, 449,
+501, 532, 543, 549, 562, 660, 683, 724, 766, 759, 740, 951, 936,
+940, 975, 998, 989, 973, 979, 979, 983, 993, 999, 484, 506, 539,
+577, 608, 625, 612, 152, 182, 230, 276, 308, 300, 216, 329, 625,
+91.7, 116, 110, 115, 118, 125, 131, 137 /

C  -----  AL  -----  

C DATA MLTFCT(13), ADDFCT(13), (DT(I, 13), I=1, DTN) / 0313151, 05684,
+95889, 05371, 04237, 03132, 02299, 01737, 01363, 01111, 00932,
+00801, 00700, 00551, 00445, 00356, 00304, 00255, 00185, 00139,
+00109, 00087, 00070, 00046, 00032, 00024, 00019, 00012, 00009/
DATA ((F(I, J, 13), I=1, IN), J=1, JN) /
+205, 236, 269, 328, 384, 431, 292, 306, 324, 366, 412, 451, 453,
+497, 508, 506, 515, 532, 659, 690, 726, 734, 706, 686, 952, 936,
+946, 991, 977, 945, 991, 971, 979, 986, 999, 996, 471, 518, 538,
+580, 609, 624, 118, 162, 184, 232, 279, 308, 300, 198, 367, 616,
+946, 118, 108, 112, 119, 123, 129, 137 /

C  -----  SI  -----  

C DATA MLTFCT(14), ADDFCT(14), (DT(I, 14), I=1, DTN) / 0289074, 06202,
+05828, 05421, 04467, 03437, 02589, 01969, 01534, 01231, 01016,
+00861, 00743, 00578, 00465, 00383, 00320, 00270, 00198, 00150,
+00117, 00093, 00076, 00050, 00035, 00026, 00020, 00013, 00009/
DATA ((F(I, J, 14), I=1, IN), J=1, JN) /
+208, 242, 277, 340, 402, 453, 288, 301, 323, 371, 424, 470, 460,
+490, 489, 490, 509, 534, 662, 700, 722, 703, 674, 663, 948, 941,
+958, 978, 946, 902, 971, 982, 980, 995, 999, 983, 481, 514, 546,
+584, 612, 625, 120, 157, 189, 241, 285, 311, 300, 215, 385, 695,
+100, 123, 110, 115, 118, 125, 133, 140 /

C  -----  P  -----  

C DATA MLTFCT(15), ADDFCT(15), (DT(I, 15), I=1, DTN) / 0269398, 06675,
+05488, 05192, 04457, 03586, 02796, 02169, 01702, 01362, 01115,
+00933, 00797, 00610, 00487, 00401, 00335, 00284, 00210, 00160,
+00125, 00100, 00082, 00053, 00037, 00028, 00021, 00014, 00010/
DATA ((F(I, J, 15), I=1, IN), J=1, JN) /
+212, 250, 290, 361, 431, 489, 283, 300, 328, 387, 449, 503, 463,
+479, 475, 486, 512, 555, 663, 703, 711, 678, 659, 661, 936, 942,
+964, 966, 916, 870, 971, 972, 980, 999, 994, 965, 497, 516, 541,
+586, 614, 623, 126, 158, 189, 245, 288, 311, 300, 256, 414, 721,
+104, 126, 111, 118, 118, 126, 135, 141 /
+594., 605., 601., 124., 166., 202., 251., 285., 299., .000, 25.0, 43.4, 76.1,
+107., 122., 108., 114., 118., 126., 133., 141./

----- CA ----- 

DATA MTFCT (20), ADDFCT (20), (DT(I,20), I=1,DTN)/0202344, 08756,
+09913, 08703, 06388, 04550, 03409, 02695, 02206, 01838, 01548,
+01314, 0123, 00838, 00647, 00515, 00422, 00354, 00262, 00202,
+00162, 00132, 00107, 00071, 00050, 00037, 00028, 00018, 00013/ 
DATA ((F(I,J,20), I=1,IN), J=1,JN)/
+592., 598., 592., 126., 164., 205., 253., 282., 295., .000,26.5, 45.8, 79.6,
+107., 120., 108., 115., 117., 127., 134., 141./

----- SC ----- 

DATA MTFCT (21), ADDFCT (21), (DT(I,21), I=1,DTN)/0192461, 09141,
+05307, 08318, 06328, 04633, 03509, 02783, 02281, 01905, 01612,
+01374, 01179, 00885, 00684, 00544, 00444, 00371, 00273, 00211,
+00169, 00137, 00112, 00075, 00052, 00039, 00030, 00019, 00014/ 
DATA ((F(I,J,21), I=1,IN), J=1,JN)/
+590., 593., 587., 123., 168., 206., 253., 280., 292., .000,24.7, 46.8, 80.7,
+106., 119., 117., 113., 117., 126., 134., 139./

----- TI ----- 

DATA MTFCT (22), ADDFCT (22), (DT(I,22), I=1,DTN)/0184009, 09612,
+08776, 07937, 06199, 04643, 03564, 02844, 02311, 01964, 01668,
+01428, 01230, 00930, 00721, 00573, 00467, 00389, 00285, 00219,
+00175, 00143, 00117, 00078, 00055, 00041, 00031, 00020, 00014/ 
DATA ((F(I,J,22), I=1,IN), J=1,JN)/
+933., 886., 877., 880., 967., 984., 999., 936., 918., 896., 492., 528., 562.,
+589., 591., 597., 126., 168., 210., 254., 279., 293., .000,26.5, 49.1, 82.1,

----- V ----- 

DATA MTFCT (23), ADDFCT (23), (DT(I,23), I=1,DTN)/0176552, 09963,
+08305, 07581, 06045, 04616, 03588, 02885, 02386, 02011, 01716,
+01476, 01277, 00973, 00757, 00602, 00450, 00408, 00297, 00228,
+00182, 00148, 00122, 00081, 00057, 00043, 00033, 00021, 00015/ 
DATA ((F(I,J,23), I=1,IN), J=1,JN)/
DATA (F(I,J,27),I=1,IN),J=1,JN)/
+913.,905.,921.,930.,979.,999.,987.,946.,926.,921.,481.,539.,565.,
+102.,119.,105.,109.,117.,125.,132.,136./

C ----- NI ----- C

DATA MLTFCX(28),ADDFCT(28), (DT(I,28),I=1,DTN)/0146163,11936,
+06569,06169,05249,04283,03500,02914,02474,02133,01858,
+01631,01440,01136,00909,00737,00605,00504,00364,00275,
+00217,00176,00146,00097,00069,00052,00040,00026,00018/
DATA ((F(I,J,28),I=1,IN),J=1,JN)/
+104.,122.,105.,111.,117.,126.,133.,137./

C ----- CU ----- C

DATA MLTFCX(29),ADDFCT(29), (DT(I,29),I=1,DTN)/0140764,12429,
+05600,05287,04585,03862,03267,02800,02428,02123,01868,
+01651,01464,01163,00935,00761,00626,00523,00378,00285,
+00224,00182,00150,00101,00072,00054,00041,00027,00019/
DATA ((F(I,J,29),I=1,IN),J=1,JN)/
+914.,918.,941.,959.,975.,999.,983.,947.,935.,931.,495.,539.,570.,
+106.,126.,106.,113.,117.,127.,133.,138./

C ----- ZN ----- C

DATA MLTFCX(30),ADDFCT(30), (DT(I,30),I=1,DTN)/0136903,12626,
+06065,05735,04962,04123,03421,02880,02468,02144,01882,
+01633,01478,01181,00955,00781,00646,00541,00391,00296,
+00232,00188,00155,00104,00074,00055,00043,00028,00019/
DATA ((F(I,J,30),I=1,IN),J=1,JN)/
+918.,929.,951.,962.,984.,999.,983.,951.,943.,941.,486.,539.,568.,
+105.,126.,104.,109.,116.,125.,131.,137./

C ----- GA ----- C

DATA MLTFCX(31),ADDFCT(31), (DT(I,31),I=1,DTN)/0132790,13077,
+07108,06629,05564,04486,03629,02997,02531,02179,01903,
+01679, 01492, 01197, 00973, 00800, 00665, 00558, 00405, 00306,
+00240, 00194, 00160, 00107, 00076, 00057, 00044, 00029, 00020/
DATA ((F(I,J,31), I=1, IN), J=1, JN)/
+500., 541., 588., 625., 656., 668., 676., 711., 761., 782., 790., 965., 938.,
+590., 602., 610., 129., 180., 215., 258., 293., 317., 000, 27.7, 51.9, 82.1,
+108., 129., 105., 111., 117., 126., 132., 138./

C ------ GE ------ C

DATA MLTFCT(32), ADDPCT(32), (DT(I,32), I=1, DTN)/0128270, 13445,
+07378, 06935, 05902, 04783, 03849, 03143, 02623, 02235, 01938,
+01702, 01510, 01212, 00989, 00817, 00681, 00574, 00418, 00317,
+00248, 00200, 00165, 00110, 00079, 00059, 00046, 00029, 00021/
DATA ((F(I,J,32), I=1, IN), J=1, JN)/
+925., 946., 955., 945., 982., 999., 979., 957., 955., 951., 486., 544., 569.,
+590., 603., 611., 126., 181., 218., 259., 295., 318., 000, 25.8, 52.7, 82.2,
+108., 130., 104., 109., 116., 125., 131., 137./

C ------ AS ------ C

DATA MLTFCT(33), ADDPCT(33), (DT(I,33), I=1, DTN)/0124644, 13890,
+07320, 06953, 06048, 04992, 04048, 03299, 02733, 02308, 01986,
+01734, 01533, 01228, 01004, 00832, 00697, 00589, 00431, 00327,
+00256, 00206, 00170, 00113, 00081, 00061, 00047, 00030, 00021/
DATA ((F(I,J,33), I=1, IN), J=1, JN)/
+591., 606., 614., 129., 182., 218., 261., 298., 321., 000, 27.5, 54.2, 83.3,
+111., 133., 104., 110., 117., 126., 132., 138./

C ------ SE ------ C

DATA MLTFCT(34), ADDPCT(34), (DT(I,34), I=1, DTN)/0121773, 14189,
+07205, 06895, 06105, 05132, 04211, 03446, 02849, 02393, 02045,
+01776, 01562, 01245, 01018, 00847, 00711, 00603, 00444, 00337,
+00264, 00212, 00175, 00116, 00083, 00062, 00048, 00031, 00022/
DATA ((F(I,J,34), I=1, IN), J=1, JN)/
+940., 959., 945., 919., 989., 999., 979., 970., 970., 957., 498., 544., 569.,
+593., 610., 618., 131., 181., 216., 262., 299., 322., 000, 28.5, 51.8, 82.8,
+111., 134., 104., 110., 116., 125., 132., 138./

C ------ BR ------ C
+604., 601., 591., 137., 187., 227., 276., 301., 314., 0.00, 31.7, 56.0, 93.8,
+119., 133., 100., 108., 113., 124., 133., 138. /

C ----- SB ----- 

DATA MLFCT(51), ADDFC(51), (DT(I,51), I=1, DTN) /0080497, 20471,
+10974, 10387, 08976, 07386, 06010, 04940, 04131, 03514, 03030,
+02640, 02318, 01819, 01453, 01181, 00976, 00821, 00608, 00472,
+00379, 00311, 00259, 00173, 00122, 00091, 00071, 00046, 00033/
DATA ((F(I,J,51), I=1, IN), J=1, JN) /
+468., 492., 527., 564., 598., 676., 663., 667., 685., 697., 706., 939., 948.,
+594., 591., 582., 135., 186., 224., 271., 296., 308., 0.00, 30.3, 54.0, 91.5,
+115., 130., 97.8, 105., 111., 121., 130., 135. /

C ----- TE ----- 

DATA MLFCT(52), ADDFC(52), (DT(I,52), I=1, DTN) /0080072, 20913,
+11003, 10470, 09160, 07608, 06209, 05090, 04234, 03583, 03078,
+02677, 02350, 01847, 01479, 01205, 00997, 00838, 00619, 00480,
+00385, 00316, 00264, 00176, 00125, 00093, 00072, 00047, 00033/
DATA ((F(I,J,52), I=1, IN), J=1, JN) /
+603., 598., 589., 137., 188., 229., 276., 300., 312., 0.00, 31.1, 55.7, 93.6,

C ----- I ----- 

DATA MLFCT(53), ADDFC(53), (DT(I,53), I=1, DTN) /0078680, 21331,
+10905, 10434, 09235, 07764, 06375, 05234, 04341, 03658, 03130,
+02715, 02380, 01871, 01503, 01227, 01016, 00855, 00630, 00487,
+00391, 00321, 00268, 00179, 00127, 00095, 00073, 00048, 00034/
DATA ((F(I,J,53), I=1, IN), J=1, JN) /
+602., 597., 589., 139., 189., 230., 276., 300., 312., 0.00, 32.3, 56.9, 94.1,

C ----- XE ----- 

DATA MLFCT(54), ADDFC(54), (DT(I,54), I=1, DTN) /0076705, 21695,
+10794, 10371, 09274, 07884, 06529, 05374, 04454, 03742, 03191,
+02759, 02414, 01897, 01526, 01248, 01036, 00871, 00642, 00495,
+00397, 00325, 00272, 00183, 00130, 00097, 00075, 00049, 00034/  
DATA (F(I,J,54), I=1, IN), J=1, JN)/  
+598., 592., 585., 136., 189., 230., 275., 297., 309., 000., 30.8, 57.1, 94.1.,  
+116., 131., 98.8, 106., 112., 123., 131., 136./  
C  
C ----- CS -----  
C  
DATA MLTFC5 (55), ADDFC5 (55), (DT(I,55), I=1, DTN)/  
0076222, 21948,  
+16508, 14106, 10525, 08201, 06649, 05471, 04547, 03822, 03255,  
+02809, 02453, 01923, 01548, 01269, 01055, 00888, 00654, 00502,  
+00405, 00332, 00272, 00186, 00132, 00098, 00076, 00050, 00035/  
DATA (F(I,J,55), I=1, IN), J=1, JN)/  
+474..490., 524., 559., 582., 684., 676., 681., 683., 691., 702., 973., 955.,  
+600., 595., 590., 139., 192., 232., 273., 298., 311., 000., 32.2, 57.1, 92.2,  
C  
C ----- BA -----  
C  
DATA MLTFC5 (56), ADDFC5 (56), (DT(I,56), I=1, DTN)/  
0074750, 22284,  
+18267, 15854, 11675, 08682, 06829, 05570, 04628, 03895, 03318,  
+02861, 02494, 01951, 01570, 01288, 01073, 00904, 00666, 00511,  
+00411, 00337, 00277, 00189, 00134, 00100, 00078, 00051, 00036/  
DATA (F(I,J,56), I=1, IN), J=1, JN)/  
+598., 593., 589., 138., 193., 232., 273., 297., 310., 000., 31.5, 57.5, 92.0,  
+115., 130., 98.6, 106., 113., 123., 130., 136./  
C  
C ----- LA -----  
C  
DATA MLTFC5 (57), ADDFC5 (57), (DT(I,57), I=1, DTN)/  
0072813, 22726,  
+17805, 15790, 12010, 09017, 07053, 05714, 04730, 03974, 03381,  
+02911, 02535, 01979, 01592, 01308, 01090, 00920, 00678, 00521,  
+00415, 00340, 00284, 00192, 00137, 00102, 00079, 00051, 00036/  
DATA (F(I,J,57), I=1, IN), J=1, JN)/  
+593., 588., 584., 137., 190., 231., 272., 294., 307., 000., 31.2, 57.5, 92.9,  
+115., 130., 98.0, 106., 112., 123., 130., 135./  
C  
C ----- CE -----  
C  
DATA MLTFC5 (58), ADDFC5 (58), (DT(I,58), I=1, DTN)/  
0071607, 23142,
\begin{verbatim}
+17378, 15460, 11850, 08958, 07041, 05728, 04759, 04010, 03420,
+02949, 02570, 02008, 01917, 01329, 01109, 00936, 00690, 00530,
+00421, 00345, 00288, 00195, 00119, 00104, 00080, 00052, 00037/
DATA ((F(I,J,58), I=1,IN), J=1, JN)/
+591., 558., 585., 139., 191., 232., 273., 295., 308., 000., 32.1, 58.4, 93.2,
+115., 130., 98.3, 106., 112., 123., 130., 135.1/
C
C ----- PR ----- C

DATA MLTFC(59), ADFCT(59), (DT(I,59), I=1, DTN)/0070600, 23411,
+16987, 14940, 11290, 08556, 06815, 05620, 04719, 04006, 03434,
+02973, 02598, 02037, 01643, 01351, 01128, 00953, 00702, 00539,
+00428, 00350, 00292, 00198, 00141, 00105, 00081, 00053, 00037/
DATA ((F(I,J,59), I=1, IN), J=1, JN)/
+591., 589., 587., 140., 191., 231., 272., 295., 309., 000., 32.5, 57.2, 92.5,
+114., 130., 97.8, 106., 112., 123., 130., 134.1/
C
C ----- ND ----- C

DATA MLTFC(60), ADFCT(60), (DT(I,60), I=1, DTN)/0069240, 23752,
+16606, 14670, 11150, 08486, 06785, 05615, 04731, 04029, 03462,
+03003, 02628, 02064, 01666, 01372, 01146, 00969, 00715, 00548,
+00435, 00355, 00296, 00201, 00144, 00107, 00083, 00054, 00038/
DATA ((F(I,J,60), I=1, IN), J=1, JN)/
+589., 588., 586., 138., 191., 232., 271., 295., 309., 000., 31.4, 57.7, 92.1,
+114., 130., 97.2, 105., 111., 122., 129., 134.1/
C
C ----- FM ----- C

DATA MLTFC(61), ADFCT(61), (DT(I,61), I=1, DTN)/0068188, 24144,
+16242, 14390, 11000, 08413, 06749, 05605, 04737, 04046, 03486,
+03029, 02655, 02089, 01688, 01391, 01164, 00985, 00727, 00557,
+00442, 00360, 00301, 00203, 00146, 00109, 00084, 00055, 00039/
DATA ((F(I,J,61), I=1, IN), J=1, JN)/
+589., 589., 588., 139., 192., 232., 272., 296., 310., 000., 32.3, 58.6, 92.3,
+115., 131., 97.4, 105., 111., 122., 129., 134.1/
C
C ----- SM ----- C
\end{verbatim}
DATA MLTFCT(62), ADDFCT(62), (DT(I, 62), I=1, DTN) /0067193, 24472, 
+15897, 14120, 10860, 08336, 06710, 05589, 04739, 04059, 03506, 
+03053, 02680, 02113, 01709, 01411, 01181, 01000, 00739, 00566, 
+00449, 00366, 00305, 00206, 00148, 00111, 00086, 00056, 00039/ 
DATA ((F(I, J, 62), I=1, IN), J=1, JN) / 
+590., 590., 589., 139., 194., 233., 272., 296., 312., 300., 31.8, 59.3, 92.1, 
+115., 132., 97.1, 105., 112., 122., 129., 134./ 

----- EJ ----- 

DATA MLTFCT(63), ADDFCT(63), (DT(I, 63), I=1, DTN) /0066597, 24394, 
+15563, 13867, 10722, 08267, 06673, 05574, 04740, 04072, 03525, 
+03075, 02703, 02136, 01730, 01429, 01198, 01016, 00752, 00575, 
+00457, 00372, 00307, 00209, 00150, 00113, 00087, 00057, 00040/ 
DATA ((F(I, J, 63), I=1, IN), J=1, JN) / 
+590., 591., 591., 138., 193., 232., 270., 296., 312., 300., 29.8, 57.6, 88.9, 
+113., 130., 94.7, 103., 109., 120., 126., 132./ 

----- GD ----- 

DATA MLTFCT(64), ADDFCT(64), (DT(I, 64), I=1, DTN) /0065629, 24686, 
+15266, 13810, 10950, 08505, 06828, 05664, 04796, 04113, 03559, 
+03105, 02730, 02158, 01749, 01446, 01123, 01030, 00763, 00585, 
+00463, 00377, 00313, 00212, 00152, 00114, 00088, 00057, 00040/ 
DATA ((F(I, J, 64), I=1, IN), J=1, JN) / 
+918., 904., 886., 875., 990., 999., 974., 941., 917., 893., 511., 553., 579., 
+589., 592., 591., 139., 192., 230., 270., 296., 312., 300., 30.0, 56.2, 89.0, 
+113., 130., 94.4, 103., 109., 119., 126., 132./ 

----- TB ----- 

DATA MLTFCT(65), ADDFCT(65), (DT(I, 65), I=1, DTN) /0064526, 24966, 
+14974, 13370, 10450, 08121, 06591, 05534, 04731, 04086, 03554, 
+03113, 02745, 02178, 01770, 01465, 01231, 01045, 00776, 00595, 
+00470, 00382, 00318, 00214, 00155, 00116, 00090, 00058, 00041/ 
DATA ((F(I, J, 65), I=1, IN), J=1, JN) / 
+917., 905., 891., 883., 986., 999., 970., 939., 917., 894., 505., 555., 578., 
+588., 592., 592., 137., 192., 231., 270., 297., 313., 300., 28.8, 56.6, 88.6, 
+113., 131., 93.6, 101., 108., 119., 125., 131./
+594.600.601.143.198.235.276.305.323...000.33.2.60.9.92.3,
+119.138.96.5.105.111.121.129.135."

----- YB -----

```
DATA MLTFCT(70), ADDFCFT(70), (DT(I, 70), I=1, DTN)/0059472, 26401,
+13557, 12280, 09800, 07746, 06360, 05398, 04669, 04078, 03568,
+03170, 02818, 02263, 01854, 01545, 01305, 01114, 00833, 00642,
+00507, 00411, 00341, 00228, 00165, 00124, 00097, 00063, 00044/
DATA ((P(I, J, 70), I=1, IN), J=1, JN)/
+204.252.300.376.443.492.293.343.380.435.485.524.448.489.527.
+581.611.629.656.673.692.726.753.764.969.917.911.905.901.893.
+999.981.954.932.911.890.500.550.569.583.590.590.147.192.228.
+269.298.315.300.28.0.55.0.86.6.113.133.91.0.98.0.106.115.122.128."
```

----- LU -----

```
DATA MLTFCT(71), ADDFCFT(71), (DT(I, 71), I=1, DTN)/0059509, 27451,
+13486, 12380, 10080, 08000, 06520, 05483, 04712, 04103, 03604,
+03187, 02834, 02278, 01868, 01558, 01317, 01126, 00844, 00651,
+00515, 00417, 00345, 00231, 00167, 00126, 00098, 00064, 00045/
DATA ((P(I, J, 71), I=1, IN), J=1, JN)/
+924.916.910.999.998.972.952.930.908.513.562.582.596.
+603.603.143.199.235.277.308.325.300.32.9.60.4.92.2.
+120.139.95.8.104.111.121.128.135."
```

----- HF -----

```
DATA MLTFCT(72), ADDFCFT(72), (DT(I, 72), I=1, DTN)/0058688, 27861,
+13177, 12230, 10160, 08163, 06660, 05578, 04772, 04143, 03632,
+03209, 02853, 02294, 01882, 01571, 01329, 01138, 00854, 00660,
+00522, 00423, 00350, 00234, 00169, 00128, 00099, 00065, 00045/
DATA ((P(I, J, 72), I=1, IN), J=1, JN)/
+537.579.607.628.670.691.710.739.754.758.966.935.932.
+926.916.906.999.997.972.953.931.908.516.563.582.597.
+604.604.144.199.236.278.309.326.300.33.5.61.0.92.9.
+121.140.96.0.104.111.121.129.135."
```

----- TA -----

```
DATA MLTFCT(73), ADDFCFT(73), (DT(I, 73), I=1, DTN)/0057820, 28041,
+12856, 12010, 10150, 08266, 06776, 05672, 04840, 04191, 03668,
+03237, 02876, 02311, 01896, 01583, 01341, 01148, 00864, 00668,
+00530, 00429, 00355, 00237, 00171, 00129, 00101, 00065, 00046/
DATA ((P(I, J, 73), I=1, IN), J=1, JN)/
DATA MLTFCT(89), ADDFCT(89), (DT(I,85), I=1, DTN)/0047695,33166,
+20484,18410,14400,11058,08780,07196,06025,05122,04410,
+03842,03381,02687,02193,01824,01541,01318,00999,00784,
+00632,00518,00432,00287,00205,00154,00121,00079,00056/
DATA ((F(I,J,89), I=1, IN), J=1, JN)/
+502.,601.,591.,146.,200.,237.,284.,310.,321.,000,33.1,58.8,96.7,
+124.,139.,91.7,100.,108.,117.,127.,132.(/

DATA MLTFCT(90), ADDFCT(90), (DT(I,90), I=1, DTN)/0047128,33583,
+20115,18330,14680,11388,39022,07353,06129,05196,04466,
+03885,03416,02712,02212,01840,01554,01330,01006,00791,
+00637,00523,00436,00291,00207,00156,00122,00080,00056/
DATA ((F(I,J,90), I=1, IN), J=1, JN)/
+484.,496.,515.,530.,543.,679.,689.,691.,684.,677.,672.,947.,948.,
+602.,600.,590.,147.,200.,238.,285.,310.,321.,000,33.5,59.3,97.4,
+124.,139.,91.9,100.,107.,118.,127.,133.(/

DATA MLTFCT(91), ADDFCT(91), (DT(I,91), I=1, DTN)/0046651,33825,
+19568,17770,14200,11102,08906,07341,06167,05252,04524,
+03938,03462,02744,02235,01857,01568,01342,01016,00797,
+00643,00528,00440,00294,00209,00157,00123,00081,00057/
DATA ((F(I,J,91), I=1, IN), J=1, JN)/
+601.,599.,589.,147.,200.,237.,284.,309.,320.,000,33.6,58.7,97.0,
+124.,139.,91.5,100.,107.,117.,126.,132.(/

DATA MLTFCT(92), ADDFCT(92), (DT(I,92), I=1, DTN)/0046265,34222,
+19119,17436,14052,11069,08928,07387,06221,05307,04576,
+03985,03503,02775,02257,01875,01582,01353,01024,00803,
+00649,00534,00443,00297,00212,00159,00124,00082,00058/
DATA ((F(I,J,92), I=1, IN), J=1, JN)/
+933.,897.,862.,839.,999.,985.,984.,947.,906.,871.,520.,559.,582.,
+603.,600.,589.,148.,201.,239.,285.,310.,320.,000,34.0,59.7,97.6,
+926.887.857.841.999.988.982.940.896.862.520.558.582.+
+601.596.585.149.201.239.285.308.318.000.34.0.59.3.98.2.+
+123.138.90.9.98.8.107.117.126.132./

C

----- BK ----- C

DATA MLTFCT(97), ADFACT(97), (DT(I,97), I=1,DTN)/0044025,35809,
+17406,16079.13330,10784,08878,07468,06378,05502,04784,
+04189,03693,02927,02376,01968,01657,01415,01067,00836,
+00674,00555,00465,00313,00223,00167,00130,00086,00061/;
DATA ((F(I,J,97),I=1,IN),J=1,JN)/
+481.506.545.575.598.683.683.682.691.705.713.953.951.+
+603.597.586.150.202.241.287.305.319.000.34.3.60.1.99.1.+
+124.138.91.3.100.118.127.132./

C

----- CF ----- C

DATA MLTFCT(98), ADFACT(98), (DT(I,98), I=1,DTN)/0043537,35963,
+16841,15850,13200,10716,08850,07466,06393,05528,04815,
+04222,03726,02957,02400,01987,01673,01427,01076,00843,
+00679,00560,00469,00316,00225,00168,00132,00087,00061/;
DATA ((F(I,J,98),I=1,IN),J=1,JN)/
+924.885.859.846.999.991.983.938.893.861.519.559.583.+
+601.594.584.149.202.240.286.308.318.000.33.6.59.3.98.3.+
+123.137.90.4.99.0.106.117.126.131./

C

------------------------ INITIALISE VARIABLES ------------------------

C

ISTAT = 0
BETA = 1.0D0
FE = 0.0D0
FA = 0.0D0
MS2 = M*S*S
DWF = DEXP(-MS2)
ELNUM = ( 2 + INDEX( ELNAMS, ELNAME//"" ) ) / 3

C

------------------------ CHECK PARAMETERS ------------------------

C

IF ( ELNUM.EQ.0 ) ISTAT = 1
IF ( M.LT.0.DO.OR.M.GT.GM( IN ) ) ISTAT = 2
IF ( S.LT.0 ) ISTAT = 3
IF ( KVOLTS.LT.0.DO.OR.KVOLTS.GT.1.D4 ) ISTAT = 4
IF ( ISTAT.GT.0.AND.STATUS.LT.0 ) THEN
    WRITE ( -STATUS, "/(2A/)//'')

`ATOM ERROR -- ', ERRMSG( ISTAT ),
`EXECUTION TERMINATED.'

STOP
ENDIF
IF ( STATUS.GE.0 ) STATUS = ISTAT
IF ( STATUS.GT.0.OR.
+ MS2.GT.GMS2( JN ).OR.S.GT.DTS( DTN ) ) GOTO 9999

C
----- CALCULATE BETA IF VOLTAGE ISN'T ZERO -----
C
IF ( KVOLTS.GT.0 ) BETA = SQRT( 1.D0 - 261127.D0 /
+ ( 261127.D0 +
+ KVOLTS * ( KVOLTS + 1032.D0 ) ) )

C
----- INTERPOLATE REAL PART -----  
C
1010
I0 = 2
I0 = I0 + 1
IF ( S.GT.DTS( I0 ).AND.I0.LT.( DTN - 1 ) ) GOTO 1010

C
RF( 1 ) = REAL( DT( I0 - 2, ELNUM ) )
RF( 2 ) = REAL( DT( I0 - 1, ELNUM ) )
RF( 3 ) = REAL( DT( I0 , ELNUM ) )
RF( 4 ) = REAL( DT( I0 + 1, ELNUM ) )
FE = DWF * CINT( DTS( I0 - 2 ), S, RF ) / 1.D3

C
IF ( M.LT.GM( 1 ) ) GOTO 9999

C
RMLT = 10. / REAL( MLTFCT( ELNUM ) )
RADD = 1.E-6 * REAL( ADDFCT( ELNUM ) )

C
3020
I0 = 0
I0 = I0 +1
IF ( M.GE.GM (I0 ).AND.I0.LT.IN ) GOTO 3020

C
IF ( MS2.GE.GMS2( JN - 2 ) ) THEN

C
----- INTERPOLATE ABSORPTIVE PART USING CUBIC -----  
C
IF ( I0.LT.3 ) I0 = 3
IF ( I0.EQ.IN ) I0 = IN - 1
I0 = I0 - 3
J0 = JN - 4

C
DO 1050 J = 1, 4
EXPMS2 = EXP( REAL( GMS2( J0 + J ) ) )
DO 1040 I = 1, 4
RF( I ) = EXPMS2 *
+ ( F( I0 + I, J0 + J, ELNUM ) * RMLT - RADD )
1040 CONTINUE
FATM(J) = REAL(CINT(GM(I0 + 1), M, RF))

CONTINUE

FA = FE / BETA * CINT(GMS2(J0 + 1), MS2, FATM)

ELSE

C ----- INTERPOLATE ABSORPTIVE PART USING QUADRATIC ----- C

COUNT = 0
BFLAG = 1
LFLAG = 1
RFLAG = 1
DO 2010 J = 1, 5
   DO 2000 I = 1, 4
      QF(I, J) = 0.0
2000  CONTINUE
2010  CONTINUE
C

J0 = 1
J0 = J0 + 1
IF (MS2.GE.GMS2(J0).AND.J0.LT.JN) GOTO 1110
C

IO = IO - 3
J0 = J0 - 3
DO 1150 J = 1, 4
   J1 = J0 + J
   IF (J1.EQ.0) THEN
      BFLAG = 0
   ELSE
      EXPMS2 = EXP(REAL(GMS2(J1)))
      DO 1140 I = 1, 4
         II = IO + I
         IF (II.EQ.0) THEN
            LFLAG = 0
         ELSEIF (II.EQ.(IN + 1)) THEN
            RFLAG = 0
         ELSE
            QF(I, J) = EXPMS2 * (F(II, J1, ENUM) * RMLT - RADD)
         ENDIF
      1140    CONTINUE
      ENDIF
1150  CONTINUE
C

IF (BFLAG.EQ.1.AND.LFLAG.EQ.1) THEN
   FA = FA + BIQINT(M, MS2, GM(I0 + 1),
                   GMS2(J0 + 1), QF(1, 1))
   COUNT = COUNT + 1
ENDIF

IF (BFLAG.EQ.1.AND.RFLAG.EQ.1) THEN
FA = FA + BIQINT( M, MS2, GM( I0 + 2 ),
               GMS2( J0 + 1 ), QF( 2, 1 ) )
COUNT = COUNT + 1
ENDIF
IF ( LFLAG.EQ.1 ) THEN
FA = FA + BIQINT( M, MS2, GM( I0 + 1 ),
               GMS2( J0 + 2 ), QF( 1, 2 ) )
COUNT = COUNT + 1
ENDIF
IF ( RFLAG.EQ.1 ) THEN
FA = FA + BIQINT( M, MS2, GM( I0 + 2 ),
               GMS2( J0 + 2 ), QF( 2, 2 ) )
COUNT = COUNT + 1
ENDIF
C
FA = FA * FE / ( BETA * DBLE( COUNT ) )
C
ENDIF
C
RETURN
END
C
FUNCTION BIQINT( M, MS2, GM, GMS2, F )

DOUBLE PRECISION BIQINT, M, MS2, GM( 3 ), GMS2( 3 ), QQINT
REAL F( 4, 3 ), FATM( 3 )

FATM( 1 ) = REAL( QQINT( GM, M, F( 1, 1 ) ) )
FATM( 2 ) = REAL( QQINT( GM, M, F( 1, 2 ) ) )
FATM( 3 ) = REAL( QQINT( GM, M, F( 1, 3 ) ) )

BIQINT = QQINT( GMS2, MS2, FATM )
END
C
FUNCTION QQINT( X, XX, F )

DOUBLE PRECISION QQINT, X( 3 ), XX
DOUBLE PRECISION X1, X2, X3, X12, X13, X23
REAL F( 3 )

C ----- USE LAGRANGE INTERPOLATING QUADRATIC -----
C
X1 = XX - X( 1 )
X2 = XX - X( 2 )
X3 = XX - X( 3 )
X12 = X( 1 ) - X( 2 )
X13 = X( 1 ) - X( 3 )
X23 = X( 2 ) - X( 3 )

C
QQINT = DBLE( F( 1 ) ) * X2 * X3 / ( X12 * X13 ) -
+    DBLE( F( 2 ) ) * X1 * X3 / ( X12 * X23 ) +
+    DBLE( F( 3 ) ) * X1 * X2 / ( X13 * X23 )

C
END

C
FUNCTION CINT( X, XX, F )

C
DOUBLE PRECISION CINT, X( 4 ), XX
DOUBLE PRECISION X1, X2, X3, X4, X12, X13, X14, X23, X24, X34
REAL F( 4 )

C
----- USE LAGRANGE INTERPOLATING CUBIC ----- 

C
X1 = XX - X( 1 )
X2 = XX - X( 2 )
X3 = XX - X( 3 )
X4 = XX - X( 4 )
X12 = X( 1 ) - X( 2 )
X13 = X( 1 ) - X( 3 )
X14 = X( 1 ) - X( 4 )
X23 = X( 2 ) - X( 3 )
X24 = X( 2 ) - X( 4 )
X34 = X( 3 ) - X( 4 )

C
CINT = DBLE( F( 1 ) ) * X2 * X3 * X4 / ( X12 * X13 * X14 ) -
+    DBLE( F( 2 ) ) * X1 * X3 * X4 / ( X12 * X23 * X24 ) +
+    DBLE( F( 3 ) ) * X1 * X2 * X4 / ( X13 * X23 * X34 ) -
+    DBLE( F( 4 ) ) * X1 * X2 * X3 / ( X14 * X24 * X34 )

C
END

C
END OF FILE ATOM.P
SUBROUTINE FOR CALCULATION OF THE EXTINCTION DISTANCE FROM FREE ATOM
SCATTERING FACTOR (DOYLE & TURNER)

SUBROUTINE EXTN1 (ATOMNAME, NAME, P1, P2, P3, DW, IP, BEAM, ALAT, CLAT, V0
$ , XSI)
CHARACTER*2 ATOMNAME(10), NAME(3)
REAL P1(10), P2(10), P3(10), BEAM(3), FAtomic(10)
REAL FHKLREAL, XSI, DW(10)
COMMON/BLK5/IZ(2), AX(2,4), BX(2,4), CC(2), NATOMS
PI =3.141592654

FUNCTION FROM ZUO & SPENCE CHAPTER1 (WE ARE IN ANGSTROMS HERE)

WAVE = 12.2643/SQRT(V*(1.0+0.97845E-06*V))
GAMMA = (1+1.956934E+06*V)
A = ALAT**10.
C = CLAT**10.
VOLUM = A*A*C

CALCULATE THE STRUCTURE FACTORS

SSQUAR = 0.25*((BEAM(1)**2+BEAM(2)**2)/(A*A)+ $ (BEAM(3)**2)/(C*C))

CALCULATE THE ATOMIC SCATTERING FACTORS

DO 15 KJ = 1, NATOMS
FAtomic(KJ) = CC(KJ)
DO 16 KK = 1, 4
FAtomic(KJ) = FAtomic(KJ) + (AX(KJ, KK)*EXP(-BX(KJ, KK)*SSQUAR))
16 CONTINUE
15 CONTINUE
FHKLREAL = 0.0
ZRE = 0.0
VG = 0.0

DO 17 KM = 1, IP
PHAS = 2.0*PI*(BEAM(1)*P1(KM)+BEAM(2)*P2(KM)+BEAM(3)*P3(KM))
DO 18 JJ = 1, NATOMS
IF (ATOMNAME(KM) .EQ. NAME(JJ)) GOTO 19
18 CONTINUE
19 VG = VG+1.145896*((IZ(JJ)-FAtomic(JJ))/SSQUAR)* $ EXP(-DW(KM)*SSQUAR)*COS(PHAS)/VOLUM
ZRE = ZRE-IZ(JJ)*EXP(-DW(KM)*SSQUAR)*COS(PHAS)
17 FHKLREAL = FHKLREAL-FAtomic(JJ)*COS(PHAS)* $ EXP(-DW(KM)*SSQUAR)
C
C CALCULATION OF THEORETICAL EXTINCTION DISTANCES
  UG = 0.006648352*GAMMA*VG
C
C CONVERT TO NANO-METER
  IF(ABS(UG).LE.1.E-10) THEN
    XSI = 1.E+10
  ELSE
    XSI = 0.1/(UG*WAVEL)
  ENDF
  RETURN
END
C
C SUBROUTINE EXTN2 IS CALCULATING XSI'S
C FROM EXPERIMENTAL STRUCTURE FACTORS: By S. ALTYNOV
C
C VALID ONLY FOR ********SILICON********
C
C SUBROUTINE EXTN2(ATOMNAME, NAME, P1, P2, P3, DW, IP, BEAM, ALAT, CLAT, Y0
C $, XSI)
C
C CHARACTER*2 ATOMNAME(10), NAME(3)
C REAL P1(10), P2(10), P3(10), BEAM(3), FATOMIC(10)
C REAL FHKLREAL, XSI, DW(10)
C COMMON/BLX5/I2(2), AX(2,4), BX(2,4), CC(2), NATOMS
C
C CCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCC
C    DIMENSION IREFL(16), Q(16), ROE(16)
C DATA NREFL / 16 /,
1  IREFL / 3, 8, 11, 12,
2  16, 19, 24, 27,
3  27, 32, 48, 56,
4  72, 75, 96, 128 /
C DATA Q / 5.656854249, 8.000000000, 5.656854249, 8.000000000,
2  8.000000000, 5.656854249, 8.000000000, 5.656854249,
3  5.656854249, 8.000000000, 8.000000000, 8.000000000,
4  8.000000000, 5.656854249, 8.000000000, 8.000000000 /
C DATA ROE
1  / 10.728, 8.656, 8.020, 0.191,
2  7.449, 7.247, 6.716, 6.438,
3  6.427, 6.046, 4.979, 4.555,
4  3.866, 3.760, 3.135, 2.533 /
C
C DATA ABOVE IS TAKEN FROM:
CQ=>TABLE 1, ALDRED AND HART, PROC. R. SOC. LONDON, A, V332, PP. 223 (1973)
CROE=>TABLE IV, LU AND ZUNGER, PHYS. REV. B, APRIL 1993, P. 9385
C
C COMBINED TABLE:
C
C HKL ISUM Q ROE
C 1 111 3 5.656854249 10.723
C 2 220 6 8.000000000 8.656
C 3 311 11 5.656854249 8.020
C 4 222 12 8.000000000 0.191
C 5  400  16  8.000000000  7.449
C 6  331  19  5.656854249  7.247
C 7  422  24  8.000000000  6.716
C 8  511  27  5.656854249  6.438
C 9  333  27  5.656854249  6.427
C 10  440  32  8.000000000  6.046
C 11  444  48  8.000000000  4.979
C 12  642  56  8.000000000  4.555
C 13  660  72  8.000000000  3.866
C 14  555  75  5.656854249  3.760
C 15  844  96  8.000000000  3.135
C 16  880  128  8.000000000  2.533

PI =3.141592654
V=V0*1000.0

C FORMULA FROM ZUO & SPENCE CHAPTER 1 (WE ARE IN ANGSTROMS HERE)

WAVEL=12.2643/SGRT(V*(1.0+0.97845E-06*V))
GAMMA=(1+1.956934E-06*V)
A=ALAT*10.
C=CLAT*10.
VOLUM=A*A*C

C CALCULATE THE STRUCTURE FACTORS
SSQUAR=0.25*((BEAM(1)**2+BEAM(2)**2)/(A*A)+
$(BEAM(3)**2)/(C*C))

C C IF REFLECTION BEAM(3) IS FOUND IN THE TABLE,
C C THEN IFIND WILL BE CHANGED TO 1 AND EXPERIMENTAL VALUE FOR
FHKLREAL
C C WILL BE USED. IF THE GIVEN REFLECTION IS NOT IN THE TABLE (SAY,
C C THEN IFIND STAYS 0 AND FHKLREAL WILL BE CALCULATED FROM
C C THE FREE ATOM MODEL.
C

IFIND=0
ISUM=INT( BEAM(1)**2+BEAM(2)**2+BEAM(3)**2 )
DO 200 JJ=1,NREFL
IF ( ISUM.NE.IREFL(JJ) ) GO TO 200
IF ( ISUM.EQ.27) GO TO 160
FHKLREAL = ROE(JJ)*Q(JJ)
IFIND=1
GO TO 300

C HERE ISUM=27, SO HKL=333 OR 511:
160 IFIND=1
IF ( INT(ABS(BEAM(1))) .EQ. 3 ) FHKLREAL = ROE(9)*Q(9)
IF ( INT(ABS(BEAM(1))) .NE. 3 ) FHKLREAL = ROE(8)*Q(8)
GO TO 300
200 CONTINUE
IFIND=0
C CALCULATE THE ATOMIC SCATTERING FACTORS
DO15KJ =1,NATOMS
   FATOMIC(KJ) =CC(KJ)
DO16KK =1,K
   FATOMIC(KJ)=FATOMIC(KJ)+(AX(KJ,KK)*(EXP(- BX(KJ,KK)*SSQUAR)))
16 CONTINUE
15 CONTINUE
C
FHKLREAL=0.0
300 CONTINUE
ZRE=0.0
C
DO 17 KM =1,IP
   PHAS=2.0*PI*(BEAM(1)*P1(KM)+BEAM(2)*P2(KM)+BEAM(3)*P3(KM))
DO 18 JJ =1,NATOMS
   IF (ATOMNAME(KM),EQ.,NAME(JJ)) GOTO 19
18 CONTINUE
19 ZRE=ZRE-IZ(JJ)*EXP(-DW(KM)*SSQUAR)*COS(PHAS)
   IF (IFIND.EQ.1) GO TO 17
   FHKLREAL=FHKLREAL-FATOMIC(JJ)*COS(PHAS)
$   *EXP(-DW(KM)*SSQUAR)
17 CONTINUE
   IF (IFIND.EQ.1) FHKLREAL=FHKLREAL*EXP(-DW(1)*SSQUAR)
   IF(ZRE .LT. 0.0 .AND. IFIND .EQ.1) FHKLREAL=-1.*FHKLREAL
C
C CALCULATION OF THEORETICAL EXTINCTION DISTANCES
   UG = 7.618323E-03*GAMMA*(FHKLREAL-ZRE)/(SSQUAR*VOLUM)
   VG =150.41320*UG/GAMMA
C GET OUT IN NANGRAMETER
   IF (ABS(UG).LE.1.E-10) THEN
      XSI=1.E+10
   ELSE
      XSI= 0.1/(UG*WAVEL)
   ENDIF
RETURN
END
C MODIFIED BEAM SELECTION PROCEDURE:
C WRITTEN BY S. ALTNYOV AND MODIFIED BY S. SWAMINATHAN TO INCORPORATE
C MODIFIED BEAM SELECTION CRITERIA
C SUBROUTINE BEAMS DETERMINES REFLECTION INCLUDED IN
C DYNAMICAL MATRIX FOR CALCULATIONS OF CBED ROCKING CURVE.
C INPUT DATA ARE:
C BD(3) - BEAM DIRECTION (VECTOR IN K DIRECTION OF LENGTH AK)
C V - VOLTAGE (KV)
C WMAX - MAXIMUM DYNAMICAL INTERACTION PARAMETER.
C NLAUE - MAXIMUM NO. OF LAUE ZONES
C ONLY BEAMS WITH w < WMAX AND LAUE ZONE < NLAUE
C ARE INCLUDED: SEE FIG 4.24 AND 4.25 OF SWAMINATHAN'S THESIS
C FOR DETAILS
C P1(10), P2(10), P3(10) - THE LIST OF ATOMIC POSITIONS
C OUTPUT DATA : 
C
C XSI(300) - EXTINCTION DISTANCES FOR THOSE BEAMS
C IB(3) - CURRENT G-VECTOR IN UNITS OF REC. LAT (HKL, INTEGER)
C B(3) - CURRENT G-VECTOR IN UNITS OF REC. LAT (HKL, REAL)
C BEAM(3) - CURRENT G-VECTOR IN NM**(-1)
C
C SUBROUTINE BEAMS(WMAX)
C
C CHARACTER*2 ATOMNAME(10),NAME(3)
C REAL KSI,PSIDSH
C DIMENSION BEAM(3),B(3),SCATK(3),ZAXIS(3),BD(3),IB(3)
A,P1(10),P2(10),P3(10),DW1(10)

COMMON /BLK1/ XSI(200),XSIDSH(200),W(200),ALATT,CLATT,T,V,
A 1MGBM(5),NIMGBM,EXPTL(300,5),DEVRRG,
B CALC(300,5),MAXEXP,NORMBM,IVAR(35),
C ILOR(5,5),IHIR(5,5),BKG(11),IFLAGEXTN,IAPR
COMMON /BLK3/ BMDIR(3),IG(3),IUCEY(3),IBEAM(200,3),
A IBRAGG,NBEAMS,IGPIXEL,IDPIXEL,IZAXIS(3)
COMMON /BLK4/ P1,P2,P3,P4,DW1,IPOS
COMMON /BLK7/ATOMNAME,NAME
COMMON /H/NLAUE

C WAVE VECTOR (CONSTANTS FROM EADES ET AL.)
C
V1=V*1000.
AK=SQR(T(V1*(1.+.97848E-6*V1)))/1.2264
GIBRAGG=SQR((((IG(1)*IBRAGG)**2+IG(2)*IBRAGG)**2)/(ALATT**2)+
A((IG(3)*IBRAGG)**2)/(CLATT**2))

C CALCULATE THE BRAGG ANGLE
C
BRAGG = ASIN(GIBRAGG/2 AK)
C CALCULATION OF ALPHAG
ALPHA = BRAGG * FLOAT(IDPIXEL) / FLOAT(IBRAGG * IGPIXEL)
ALPHAG = TAN(ALPHA) * GIBRAGG
WRITE(*,*) 'BRAGG = ', BRAGG, 'ALPHA = ', ALPHA, 'ALPHAG = ', ALPHAG
COSBRAGG = COS(BRAGG) * AK
SINBRAGG = -I * SIN(BRAGG) * AK
CALL DIRNM(IZAXIS, ZAXIS)
CALL NORM(ZAXIS)
CALL RECNM(IG, GV)
CALL NORM(GV)
CALL SCALEVEC(ZAXIS, COSBRAGG)
CALL SCALEVEC(GV, SINBRAGG)
CALL ADDVEC(ZAXIS, GV, BD)
CALL NORM(BD)
CALL SCALEVEC(BD, AK)
C
CRUN LOOP ON ALL POSSIBLE G-VECTORS. CALCULATE EXTIN. DISTANCE FOR THEM
C AND DYN. PAR. AND COMPARE IT WITH "WMAX".
C
N = 61
MINUS = 31
NUMBER = 0
C
C SEARCH 3-D GRID OF G-VECTORS
C
DO1I = 1, N
IB(1) = I_MINUS
BEAM(1) = REAL(IB(1))
C
DO1J = 1, N
IB(2) = J_MINUS
BEAM(2) = REAL(IB(2))
C
DO1K = 1, N
IB(3) = K_MINUS
BEAM(3) = REAL(IB(3))
C
C ELIMINATE HIGHER ORDER LAUE ZONES
C
IZONE = ABS(IB(1) * IZAXIS(1) + IB(2) * IZAXIS(2) + IB(3) * IZAXIS(3))
C
25 B(1) = BEAM(1) / ALATT
B(2) = BEAM(2) / ALATT
B(3) = BEAM(3) / CLATT
CALL ADDVEC(BD, B, SCATK)
C
NOW SCATK(3) IS A WAVEVECTOR FROM THE CENTER OF EWALD SPHERE TO
C THE CURRENT G VECTOR BEAM(3) MEASURED IN NM**(-1)
C
C IF IFLAGEXTN = 0, THEN GO TO EXTN (FREE ATOM MODEL)
C IF IFLAGEXTN=1, THEN GO TO EXTN1 (FREE ATOM MODEL);
C IF IFLAGEXTN=2, THEN GO TO EXTN2 (EXP. STR. FACTORS);

SG= (AK**2 - SCALAR(SCATK,SCATK) ) / (2. * AK)

IF((IZONE.GT.NLAE).AND.(ABS(SG).GT.ABS(ALPHAG))) GO TO 1

CALL EXTN(ATOMNAME,P1,P2,P3,DW1,IPOS,BEAM,ALATT,CLATT,V,KSI,
AKSIDSH)
IF(BEAM(1).EQ.0.0.AND.BEAM(2).EQ.0.0.AND.BEAM(3).EQ.0.0)GOTO 26
IF (IFLAGEXTN .EQ. 1 ) THEN
CALL EXTN1(ATOMNAME,NAME,P1,P2,P3,DW1,IPOS,BEAM,ALATT,CLATT.V,
AKSI)
ELSEIF (IFLAGEXTN .EQ. 2 ) THEN
CALL EXTN2(ATOMNAME,NAME,P1,P2,P3,DW1,IPOS,BEAM,ALATT,CLATT.V,
AKSI)
ENDIF

26 IF (ABS(KSI).GE.100000.0 )GO TO 1

WCUR = SG*KSI
IF (IZONE.LT.NLAE) THEN
IF ( ABS(WCUR) .GT. WMAX ) GO TO 1
ELSE
IF((ABS(SG).GT.ABS(ALPHAG)).OR.(ABS(WCUR).GT.WMAX))GO TO 1
ENDIF

WRITE(*,1000)IB,KSI,SG,WCUR

C FOR MAIN BEAM DISC, CHECK IF NEW BEAM ALREADY IN DARK FIELD DISC LIST.
C
NUMBER=NUMBER+1
IBEAM(NUMBER,1)=IB(1)
IBEAM(NUMBER,2)=IB(2)
IBEAM(NUMBER,3)=IB(3)
XSI(NUMBER)=KSI
XSIDSH(NUMBER)=KSIDSH
W(NUMBER)=WCUR
1 CONTINUE

C
NBEAMS=NUMBER

C RE-ORDER BEAMS IN THE ORDER OF INCREASING W:
C
C CHANGE THE ORDER OF BEAMS SOTHAT 1ST BEAM BECOMES 000,
C 2ND BEAM BECOMES G, THIRD IS 2*G, FOURTH IS 3*G AND SO ON UPTO 2:
C POSITIVE G'S
KOUNT=0
DO35J=1,3
DO35I=1,NBEAMS
IF(IBEAM(I,1).EQ.IG(1)*(J-1).AND.IBEAM(I,2).EQ.IG(2)*(J-1)
A. AND.IBEAM(I,3).EQ.IG(3)*(J-1) THEN
    KOUNT=KOUNT+1
    DO40 K=1,3
    IBEAM(I,K)=IBEAM(J,K)
40    IBEAM(J,K)=IG(K)*(J-1)
    CALL EXCH(XSI(KOUNT),XSI(I))
    CALL EXCH(XSIDSH(KOUNT),XSIDSH(I))
    CALL EXCH(W(KOUNT),W(I))
ENDIF
35 CONTINUE

DO 50 I=4,NBEAMS
DO 50 J=5,NBEAMS-I+1
    IF( ABS(W(J-1)).LT.ABS(W(J)) ) GO TO 50
    CALL EXVECI( IBEAM(J,1), IBEAM(J,2), IBEAM(J,3),
                  IBEAM(J-1,1), IBEAM(J-1,2), IBEAM(J-1,3) )
    CALL EXCH( W(J), W(J-1) )
    CALL EXCH( XSI(J), XSI(J-1) )
    CALL EXCH( XSIDSH(J), XSIDSH(J-1) )
50 CONTINUE

C
DO 75 I=1,NBEAMS
75 WRITE(*,1000)(IBEAM(I,J), J=1,3),XSI(I),XSIDSH(I),W(I),I
C
1000 FORMAT ( ' BEAM = ', 3I5, 3X, 2F12.4, 2X, F12.5, I5)
RETURN
END

C
SUBROUTINE EXCH(A,B)
    C=A
    A=B
    B=C
RETURN
END

C
SUBROUTINE EXVECI(K1,K2,K3,L1,L2,L3)
    IEX=K1
    K1=L1
    L1=IEX
    IEX=K2
    K2=L2
    L2=IEX
    IEX=K3
    K3=L3
    L3=IEX
C
RETURN
END
LIST OF REFERENCES


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