THE USE OF THE BORMANN EFFECT TO PRODUCE MONOENERGETIC X-RAYS

Charles T. Prevo
(Ph.D. Thesis)

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THE USE OF THE BORRMANN EFFECT TO PRODUCE

NEARLY MONOENERGETIC X-RAYS

ABSTRACT

Anomalous transmission due to diffraction from the (220) planes of germanium has been measured for 1.65 mm, 8.58 mm, and 19.6 mm thick single crystals at energies ranging from 15 to 55 keV. A method of producing a continuously variable, in energy, source of nearly monoenergetic x-rays based on anomalous transmission and a bremsstrahlung input spectrum is described. Collimator design, crystal selection, crystal preparation, peak shapes, full-widths at half-maximum, and intensities for fixed crystals are discussed and compared to theory. The anomalous transmission factors ranged from 1×10^{-7} to 5×10^{-6} resulting in usable intensities of 2.5 to 37 photons per sec for tungsten target bremsstrahlung input spectra.

CHAPTER 1

INTRODUCTION

Sources of monoenergetic low energy photons ($h\nu < 100$ keV) are useful in many research areas. Such situations are common when excitation of some physical process is necessary such as in Compton scattering, Raman scattering, and x-ray spectrometry studies. For example, it is frequently desirable to know the response of a detector or dosimeter to monoenergetic x- or gamma-rays. In x-ray fluorescence it is very desirable to have nearly monoenergetic exciting sources with energy just above the K-absorption edge of the element being analyzed. The determination of the composition of multielement substances such as the human body by x- and gamma-ray absorption requires energies lower than 100 keV in order to take advantage of the greater differences in the absorption coefficients of different elements at these energies.

There are a number of different types of monoenergetic sources such as gamma emitting radionuclides, radionuclides that decay by electron capture or internal conversion to ground states and emit x-rays, monochromators using a bremsstrahlung source and Bragg diffraction, filtered bremsstrahlung spectra, transmission anode x-ray tubes, and characteristic x-rays from a secondary target which has been excited by an x-ray tube.

There are only a very limited number of radionuclides that decay by internal conversion or electron capture with essentially monoenergetic emissions. Monochromators, although very useful at low energies, become less useful at higher energies because of interference from Compton scattering. Filtered bremsstrahlung and transmission anode spectra are

not even nearly monoenergetic. Filtered x-rays from a secondary target offer a good variety of high intensity low energy x-rays from about 7 keV to about 100 keV.

An alternative method, and the subject of this paper, is the use of the Borrmann effect to provide nearly monoenergetic beams of x-rays. The Borrmann effect is really a diffraction phenomena in nearly perfect crystals. Normally one would expect the intensity transmitted through a crystal to be given by $e^{-\mu x}$. However, for nearly perfect single crystals, it is found that when μx is in the range from 20 to 80, and the Bragg equation is satisfied for a given set of diffracted planes, something unusual occurs. If a parallel monochromatic beam were incident at the Bragg angle onto a single crystal cut in the form of a parallel-sided slab with the planes to be used in the diffraction perpendicular to the slab faces as shown in Fig. 1.1, there would be three beams exiting the crystal. One would be the transmitted beam which suffers normal attenuation. The other two would exit from the crystal directly opposite the point of incidence. The one parallel to the incident beam is called the forward diffracted beam and the one which makes an angle of $2\theta_B$ with respect to the incident beam is called the diffracted beam. The existence of the diffracted and forward diffracted beams is referred to as the Borrmann effect or anomalous transmission. An anomalous transmission experiment is shown in Fig. 1.2. If the crystal is rotated through an angle θ about an axis perpendicular to the plane of the figure, then when the crystal is well off the diffraction setting; i.e., when $\theta \neq \theta_B$ in Bragg's law, $\lambda = 2d \sin \theta_B$ the transmitted intensity is given by the usual expression for attenuation: $I_0 e^{-\mu x}$, while the diffracted beam intensity is zero. However,

Fig. 1.1 Illustration of the Borrmann Effect

Fig. 1.2 The Anomalous Transmission Experiment

as the crystal is rotated through $2\theta = 2\theta_B$, a peak is observed in the intensity of the diffracted beam as shown in Fig. 1.3. The diffracted and forward diffracted beams are approximately of equal intensity for sufficiently thick crystals [Okkerse 1963]. This condition holds in the present study. Thus the diffracted beam was chosen in order to minimize interference effects from the transmitted beam and fluorescent x-rays from the crystal. The remarkable thing about the Borrmann effect is that any energy at all gets through a crystal for which the attenuation factor is something like e^{-20} . The effective absorption coefficient for anomalous transmission is given by $\mu_{\text{eff}} = \mu(1 - \epsilon P)$ where ϵ is the ratio of the imaginary part of the structure factor for the diffracted beam to the imaginary part of the structure factor for the forward diffracted beam and P is the polarization factor. The polarization factor equals 1 for the component of the electric field perpendicular to the plane of incidence and equals $\cos(2\theta_B)$ for the component of the electric field parallel to the plane of incidence, where θ_B is the Bragg angle.

A monochromatic beam has been assumed in order to aid the discussion although it is not necessary in order to observe the effect. In fact, the whole point of this thesis is to demonstrate the usefulness of the Borrmann effect for producing nearly monoenergetic x-rays from a bremsstrahlung source. For a parallel incident beam only the wavelength which satisfies the Bragg equation is anomalously transmitted. Thus the crystal acts as a selective filter and by rotating the crystal one can provide a continuously variable, in energy, source of x-rays. In practice the incident beam is not parallel and results in the anomalous transmission of a band of wavelengths. The bandwidth can be controlled by collimation of

Fig. 1.3 Variation of Intensity as the Angle of Incidence is Varied

the incident beam.

Anomalous transmission has been observed in single crystals of quartz [Borrmann 1941], silicon, germanium [Batterman 1961], and silver using the K x-rays from copper and molybdenum as input spectra. The integrated anomalous transmission was determined by integrating the rocking curve, which is obtained by measuring the transmitted intensity as the crystal is rotated through the Bragg angle while a monochromatic beam is incident onto the crystal. The reason the measurements were made in this manner is that the integrated intensity can easily be compared to theory. However, for the present study the interest is in the anomalous transmitted intensity for a fixed crystal and a bremsstrahlung input spectrum. This is the configuration necessary for a practical utilization of the Borrmann effect as a monoenergetic x-ray source. The present study is the first investigation of this configuration and extends to higher energies than previously investigated. The theory has been adapted to calculate the anomalous transmission spectra for the fixed crystal and collimated bremsstrahlung input spectrum. In this study anomalous transmission spectra for the (220) diffraction planes have been measured for single crystals of germanium 1.65 mm, 8.58 mm, and 19.6 mm thick at energies from 15 to 55 keV. The measured anomalous transmission spectral shapes, full widths at half maximum, and intensities have been measured with an intrinsic germanium detector and compared to theory. The anomalous transmission factors ranged from 1×10^{-7} to 5×10^{-6} resulting in usable intensities of 2.5 to 37 photons per sec for tungsten target bremsstrahlung input spectra.

CHAPTER 2

THEORY

2.1 Introduction

There are two general theories which may be used to account for the intensities in x-ray diffraction studies. The kinematical theory treats the scattering from each volume element in the sample as being independent of that of other volume elements, except for incoherent power losses in reaching and leaving that particular volume element. The dynamical theory takes into account all wave interactions within the crystalline media, and must generally be used when diffraction from large perfect crystals is being considered. The original theoretical work done by Darwin [1914], Ewald [1962], and Von Laue [1931] has been summarized and extended in treatments by Zachariasen [1945], James [1950], and Hirsch [1952]. However, only a small fraction of their work deals directly with anomalous transmission, so this chapter will attempt to present a condensed but consistent theory which will enable one to make the calculations of interest in Chapters 5 and 6.

The theory starts with the classical scattering from an electron and then goes on to scattering from an atom, scattering from a unit cell, and then the atomic scattering factor. After these basic concepts have been introduced we will be mostly concerned with a problem in classical electromagnetic theory. The crystal will be represented by a periodic, anisotropic, complex, dielectric function which depends explicitly on time. Being periodic in three dimensions, the dielectric function can be represented by a Fourier series over the reciprocal lattice in exactly

the same way that the periodic charge density is so represented in crystallographic work. In fact, we associate the Fourier coefficients of the dielectric with those of the density. A complex dielectric function is used to handle the question of absorption, and an anisotropic one is required because waves propagating in different directions must have different indices of refraction. This difference becomes the important parameter in the whole problem.

The fundamental problem is to solve Maxwell's equations, use a complex dielectric function to describe the medium, assume wave solutions consistent with Bragg's law, obtain a set of homogeneous linear equations for the ratio of the field amplitudes, and write down a determinant whose value must be zero for nontrivial solutions of the equations to exist. The latter imposes certain restrictions on the wave vectors. The solutions are then used to develop expressions for the intensity of the anomalous transmission.

2.2 Classical Scattering From an Electron

First let us consider the scattering of radiation by a free electron. Let the incident radiation be propagating in the x-direction in a rectangular coordinate system as shown in Fig. 2.1, with the electron at the origin. We consider unpolarized radiation for which there is no preferred orientation in the y-z plane and choose the y axis in the plane determined by the incident and scattered waves. In general, the electric field produced at the point P due to the acceleration a of a charge e is

$$\text{given by } E_a = \frac{e \sin \alpha}{rc^2} a = \frac{e \sin \alpha}{rc^2} \cdot \frac{Ee}{m_e} = \frac{Ee \sin \alpha}{m_e rc^2}$$

Fig. 2.1 Scattering from an Electron

where

e = the charge of an electron - 4.8×10^{-10} esu

m_e = the mass of an electron - 9.1×10^{-28} grams

c = the velocity of light - 2.997×10^{10} cm/sec.

α is the angle between the direction of acceleration and the direction of interest, and E is the electric field producing the acceleration. Resolving the incident field into its components, E_y and E_z we see that $E^2 = E_y^2 + E_z^2$. Since the intensity, I , is proportional to the square of the electric field then

$$I_o = I_y + I_z$$

where I_y and I_z are the intensities of the y and z components of the incident electric field. Also, since there is no preferred orientation of the incident field then $I_y = I_z = I_o/2$.

The intensity I_1 of the scattered beam at p due to the y component of the incident wave is

$$I_1 = I_y \frac{e^4 \sin^2 \phi_1}{r^2 m_e^2 c^4} = \frac{I_o e^4 \cos^2 \theta}{2r^2 m_e^2 c^4}$$

and the intensity I_2 of the scattered beam at p due to the z component of the incident wave is

$$I_2 = \frac{I_z e^4 \sin^2 \phi_2}{r^2 m_e^2 c^4} = \frac{I_o e^4}{2r^2 m_e^2 c^4}$$

since $\phi_2 = \frac{\pi}{2}$.

The total intensity at P is

$$I_e = I_1 + I_2 = \frac{I_o e^4}{r_m^2 c^4} \left(\frac{1 + \cos^2 \theta}{2} \right)$$

$$= \frac{I_o r_e^2}{r^2} \left(\frac{1 + \cos^2 \theta}{2} \right)$$

where

$$r_e = \frac{e^2}{m_e c^2} \quad - \text{the classical electron radius}$$

and the factor $\frac{1 + \cos^2 \theta}{2}$ is referred to as the polarization factor.

This terminology of polarization factor is somewhat misleading since it came about as a result of assuming that the incident electric field is not polarized.

The Thomson scattering cross section can readily be derived from the above result. The Thomson cross section, σ_{Th} , is defined by

$$\sigma_{Th} = \frac{P}{I_o}$$

where P is the power radiated by the accelerated charge

$$P = \int I_e dA = \int_0^\pi \frac{I_o r_e^2}{r^2} \frac{(1 + \cos^2 \theta)}{2} \cdot 2\pi r^2 \sin \theta d\theta$$

$$= \pi r_e^2 I_o \int_0^\pi (1 + \cos^2 \theta) \sin \theta d\theta$$

$$= \frac{8}{3} \pi r_e^2 I_o$$

Thus,

$$\sigma_{Th} = \frac{P}{I_0} = \frac{8}{3} \pi r_e^2$$

which is a familiar result.

What we have been discussing is coherent scattering. An electron can also Compton scatter from an electron bound with energies small compared to $h\nu$. The electron must be free or nearly free. Its phase has no fixed relation to the phase of the incident beam and is therefore incoherent. It cannot take part in diffraction because its phase is only randomly related to that of the incident beam and cannot therefore produce any interference effects. Since we are discussing the classical theory it is important to realize that it can only account for coherent scattering. Quantum theory can account for both coherent and incoherent scattering.

2.3 Scattering From An Atom

Is the wave scattered by an atom simply the sum of the waves scattered by its component electrons? The answer is yes if the scattering is in the forward direction, since the waves can be algebraically added. However, at any other angle the phases are different and cannot simply be added. It is convenient at this point to introduce a quantity, f , called the atomic scatter factor which is defined as the ratio of the amplitude of the wave scattered by an atom to the amplitude of the wave scattered by a free electron. For any atom scattering in the forward direction, the atomic scattering factor is equal to the atomic number Z which is equal to the number of electrons in the atom. As the scattering

angle θ increases from zero the waves scattered by individual electrons become more and more out of phase and f decreases. We will see that the atomic scattering factor also depends on the wavelength of the incident beam. The net result is that f decreases as $\frac{\sin\theta}{\lambda}$ increases. It is important to remember that we are discussing coherent scattering which is the only kind capable of being diffracted. However, incoherent or Compton modified scattering is occurring at the same time. Since the modified scattering is due to collision of quanta with electrons, bound with energies small compared with $h\nu$, its intensity relative to that of the unmodified radiation increases as the proportion of loosely bound electrons increases; (i.e., as Z decreases). Also the intensity of the modified radiation increases as $\frac{\sin\theta}{\lambda}$ increases. Thus I_{mod} and I_{unmod} vary in opposite ways with Z and $\frac{\sin\theta}{\lambda}$.

At this time we shall just assume f in a formal way and proceed to demonstrate that the scattering factor plays a key role in diffraction theory. Later we shall return to a detailed discussion of the scattering factor.

2.4 Scattering By A Unit Cell

The next question to be treated is "How does the intensity of the diffracted beam depend on the position of the atom in the unit cell". This is essentially a matter of determining how the phase difference depends on the arrangement of the atoms. Let's consider the phase difference between waves scattered by an atom at the origin and another at position l^x as shown in Fig. 2.2. Atoms A and C are participating in Bragg diffraction from the $(h00)$ planes and we wish to determine the effect

Fig. 2.2 The Effect of Atom Position on the Phase Difference Between Diffracted Rays

of atom B. For the rays $1'$ and $2'$ the phase difference $\delta_{2'1'}$ is given by

$$\delta_{2'1'} = 2 d_{h00} \sin\theta = \lambda \quad (1)$$

where, d_{h00} , the perpendicular distance between (h00) planes is given by

$$d_{h00} = AC = \frac{a}{h} \quad (2)$$

where a is the lattice parameter. The x rays scattered in the same direction by atom B have a phase difference, $\delta_{3'1'}$, given by

$$\delta_{3'1'} = \frac{AB\lambda}{AC} \quad (3)$$

The phase difference can be expressed in terms of the path difference or an angular measure ϕ which is related to the path difference δ by

$$\phi = \delta \frac{2\pi}{\lambda} = \vec{k} \cdot \vec{\delta} \quad (4)$$

where $\vec{k}$ is the wave vector. Thus

$$\phi_{3'1'} = \delta_{3'1'} \frac{2\pi}{\lambda} = \frac{2\pi hx}{a} \quad (5)$$

Defining the fraction coordinate, u as $\frac{x}{a}$ we have

$$\phi_{3'1'} = 2\pi hu \quad (6)$$

Extending this to three dimensions

$$\begin{aligned} \phi &= 2\pi(hu + kv + \ell w) \\ &= 2\pi \vec{B} \cdot \vec{r} \end{aligned} \quad (7)$$

If the atom at B is a different kind of atom than is at A then the two waves can differ in amplitude as well as phase. In this case the amplitudes of these waves relative to the amplitude of the wave scattered by a single electron are given by the appropriate value of the atomic scattering factor, f . Thus the problem of scattering from a unit cell resolves itself into one of adding waves of different phase and amplitude. It is very convenient at

this point to use a complex representation for the wave. The wave scattered by an atom can be represented by

$$f e^{2\pi i (hu + kv + lw)} = f e^{2\pi i \vec{B} \cdot \vec{r}} \quad (8)$$

We now define a quantity called the structure factor, F_{hkl} , as the amplitude of the wave scattered by all the atoms of a unit cell compared to the amplitude of the wave scattered by one electron. The structure factor is given by

$$F_{hkl} = \sum_1^N f_n e^{2\pi i \vec{B} \cdot \vec{r}_n} \quad (9)$$

where the sum is over the N atoms in the unit cell. Thus we see that while f_n accounts for the charge distribution within an atom, F_{hkl} accounts for the distribution throughout the unit cell. To calculate F_{hkl} we need only know the positions of the atoms within the unit cell and the scattering factor for each atom. The imaginary part of the structure factor plays a key role in the existence of the Borrmann effect. Later we will show the effective absorption coefficient for anomalous transmission is given by

$$\mu_{\text{eff}} \approx \mu_a \left(1 - \frac{F''_{hkl}}{F'_{000}} P \right) \quad (10)$$

where F''_{hkl} denotes the imaginary part of F_{hkl} and P is the polarization factor.

2.5 The Atomic Scattering Factor

Conceptually it is easy to see how one obtains the atomic scattering factor. All one needs to do is add all the contributions to the scattering from all parts of the atom taking into account the difference in phase of each contribution. This is quite analogous to what we did in calculating the expression for the structure factor. The scattering factor is given by

[James 1945]

$$f_o = \int \psi^2 e^{-ik\vec{S} \cdot \vec{r}} dv \quad (11)$$

where $\psi^2 dv$ is the probability of finding an electron in the volume dv , r is the position of the volume element dv with the origin of the coordinate system at the nucleus of the atom, and $\vec{S} = \vec{s} - \vec{s}_o$ where $\vec{s}_o$ is a unit vector in the direction of scatter as shown in Fig. 2.3. In this expression for the scattering factor is assumed that the frequency of the incident radiation is large compared to any of the atomic transition frequencies. Later we will discuss the corrections which must be made to the scattering factor when this assumption is not valid.

Several extensive calculations have been made of the scattering factor. Hanson, Herman, Lea, and Skillman [Hanson 1964] have made calculations for $Z = 1$ to 100 for neutral elements based on self-consistent field Hartree-Fock-Slater wave functions calculated by Herman and Skillman [Herman 1962]. Cromer and Weber [Cromer 1965] have made calculations for a total of 208 neutral atoms, positive ions and a few negative ions using the self-consistent Dirac-Slater wave functions. These are the relativistic equivalent of the HFS wave functions.

2.6 The Anomalous Scattering and Dispersion of X-Ray's

In the previous discussion of the scattering factor it was assumed that the frequency of the incident radiation is large by comparison to the transition frequencies of the atom. When this assumption is not met it is found that the scattering factor can vary markedly with frequency. There will be an anomalous scattering which is closely connected with the anomalous dispersion of the radiation in the material.

Fig. 2.3 **Relation of the Volume Element and the Point of Interest**

A classical approach will be used to show that corrections of interest to the scattering factor can be related to the absorption cross sections of the electrons in various energy states. We start first by showing the relation between the atomic scattering factor, the index of refraction, and absorption in a medium. The classical theory of dispersion assumes that the atoms scatter as if they contained dipole oscillators having definite natural frequencies associated with the absorption frequencies of the atom. Consider an electron of charge e and mass m_e on which an electric field $\vec{E} = \vec{E}_0 e^{i\omega t}$ is incident. The equation of motion of this electron under the influence of the electric field is

$$\ddot{\vec{X}} + \gamma \dot{\vec{X}} + \omega_s^2 \vec{X} = \frac{e\vec{E}}{m_e} e^{i\omega t} \quad (12)$$

Where ω is the frequency of the incident field, γ is a damping factor and ω_s is the natural frequency of the dipole in the absence of any damping or applied field. The solution of this equation leads to a dipole moment, $\vec{M}$, given by

$$\vec{M} = e\vec{x} = \frac{e^2}{m} \frac{E_0 e^{i\omega t}}{\omega^2 - \omega_s^2 - i\gamma\omega} \quad (13)$$

This oscillating dipole gives rise to an electromagnetic wave of the same frequency. The amplitude of the scattered wave at a distance r in the equatorial plane is

$$E_s = \frac{e^2}{mc^2 r} \frac{\omega^2 E_0}{\omega_s^2 - \omega^2 + i\gamma\omega} \quad (14)$$

The scattering by a free electron is obtained by setting γ and ω_s equal to zero. Thus we obtain for the scattering factor

$$f = \frac{\omega^2}{\omega^2 - \omega_s^2 - i\gamma\omega} \quad (15)$$

For the case discussed previously where $\gamma = 0$ and $\omega \gg \omega_s$ we get $f = 1$ which is what one would expect. Also note that when ω is approximately equal to ω_s then the damping term becomes very important. If the damping is due purely to the classical radiation damping, then $\gamma = \frac{2e^2\omega^3}{3mc^3}$.

For x-rays, the index of refraction n is given in terms of the scattering factor f by

$$n = 1 - \frac{ne\lambda^2 e^2}{2\pi mc^2} f = 1 - \frac{2\pi n e^2}{m\omega^2} f \quad (16)$$

One should note that since $E_s = \frac{-e^2}{mc^2 r} E$ for a free electron, the scattered wave is π out of phase with the incident wave and thus the scattered wave is π out of phase with the primary wave when f is positive. When f is negative, corresponding to $\omega \ll \omega_s$, the refractive index of the medium will be greater than unity, and the phase velocity of the wave less than c . When $\omega \gg \omega_s$, so that f is positive, the index is less than unity and the phase velocity of the transmitted wave is greater than c . For both of these cases the main effect is to change the phase velocity of the transmitted wave with little absorption. However, if ω is comparable to ω_s then f will be complex, resulting in a complex index of refraction. This means there will be absorption of the radiation.

Let

$$f = f' + i f'' \quad (17)$$

$$n = 1 - \alpha - i\beta \quad (18)$$

$$\text{so that } \alpha = \frac{2\pi n e^2}{m\omega^2} f' \quad \beta = \frac{2\pi n e^2}{m\omega^2} f'' \quad (19)$$

For a wave traveling in the x direction in a medium with refractive index n the displacement must be multiplied by a phase factor $e^{-ikx} = e^{-ink_0 x} e^{-ik_0(1-\alpha-i\beta)x} = e^{-ik_0(1-\alpha)x} e^{-k_0\beta x}$ where $k_0 = \frac{2\pi}{\lambda}$. The real part of the refractive index is associated with the phase-lag while the imaginary part β corresponds to absorption, that is, a diminution of the amplitude of the wave with increasing x. Thus the intensity is reduced by a factor $e^{-k_0\beta x}$ and the absorption coefficient is

$$\mu = \frac{4\pi}{\lambda} \quad \beta = \frac{4\pi n'' e^2}{mc} f'' \quad (20)$$

where f'' is the imaginary part of f .

$$\text{Since } f'' = \frac{\gamma \omega^3}{(\omega^2 - \omega_s^2)^2 + \gamma^2 \omega^2} \text{ then}$$

$$\mu(\omega) = \frac{4\pi^2 \gamma}{mc} \frac{\omega^3}{(\omega^2 - \omega_s^2)^2 + \gamma^2 \omega^2} \quad (21)$$

Since f'' is always positive, then the imaginary part of f , if'', denotes a component of the scattered wave which is $\frac{\pi}{2}$ out of phase with the primary wave. We know that the dipoles lying in a thin sheet parallel to the primary wave front produce a resultant wave whose phase is retarded $\frac{\pi}{2}$ behind that of the waves scattered by the individual dipoles in the sheet [Jackson 1962]. Since this phase already lags $\frac{\pi}{2}$ behind that of the primary wave, the retardation of the resultant scattered wave will be π . The effect of the imaginary part of f will be to add to the resultant scattered wave a component that at each point of the medium is out of phase with the primary wave. This results in a progressive diminution in the amplitude of the transmitted wave, that is, absorption.

2.7 The Periodic, Complex, Dielectric Constant

The electron density at any point in the crystal, $\rho(\vec{r})$, can be

expressed as a Fourier sum over the lattice. That is:

$$\rho(\vec{r}) = \frac{1}{V} \sum_{\mathbf{H}} F_{\mathbf{H}} e^{-2\pi i \vec{B} \cdot \vec{r}} \quad (22)$$

where V is the volume of the unit cell and $\vec{b}$ is the reciprocal lattice vector. If $\vec{b}_1$, $\vec{b}_2$, and $\vec{b}_3$ are the reciprocal lattice vectors defining the unit cell in reciprocal space, then $\vec{B} = h\vec{b}_1 + k\vec{b}_2 + l\vec{b}_3$ where h , k , l are the Miller indices of the reflection described by the reciprocal lattice point. The sum over $\mathbf{H}$ signifies all possible values of h , k , l and $F_{\mathbf{H}}$ may be identified as the structure factor for the h , k , l reflection. Conversely:

$$F_{\mathbf{H}} = F(hkl) = \int_V \rho(\vec{r}) e^{2\pi i \vec{B} \cdot \vec{r}} dV \quad (23)$$

Assuming that the atoms behave as rigid spheres with respect to their charge densities and are not vibrating thermally, $F_{\mathbf{H}}$ can be written as

$$F_{\mathbf{H}} = \sum_n f_n e^{2\pi i \vec{B} \cdot \vec{r}_n} \quad (24)$$

where the sum over n is over the atoms in the unit cell. The atomic scattering factor of the n^{th} atom is represented by f_n .

The connection between the Fourier Series which describes the electron density, Eq. 22, and a representation of the periodic dielectric function can be made using the electronic polarizability. The electric displacement vector $\vec{D}$ can be written in terms of the electric field $\vec{E}$, and polarization $\vec{P}$, as

$$\vec{D} = \epsilon \vec{E} = \vec{E} + 4\pi \vec{P} \quad (25)$$

where ϵ is the dielectric function

$$\epsilon = 1 + 4\pi \chi_e \quad (26)$$

and χ_e is the electric susceptibility.

If we consider a sinusoidal field of amplitude E_0 , acting on a

collection of electrons held by restoring forces such that they have a natural frequency of oscillation ω_s , then the amplitude of the induced electron motion is given by

$$x = \frac{e}{m} \frac{E_0}{\omega_s^2 - \omega^2} \quad (27)$$

If ω is much greater than ω_s , the polarization amplitude is $P = pex$ and ϵ is now:

$$\epsilon(\vec{r}) = 1 - \frac{4\pi e \rho(\vec{r})}{m\omega^2} = 1 - \frac{r_e \lambda^2 \rho(\vec{r})}{\pi} \quad (28)$$

where the quantity (e^2/mc^2) is the classical electron radius r_e , and is equal to 2.818×10^{-13} cm. If we define Γ :

$$\Gamma = \frac{r_e \lambda^2}{v} \quad (29)$$

then from Eq (22) we have

$$\epsilon(r) = 1 - \Gamma \sum_H F_H e^{-2\pi i \vec{B} \cdot \vec{r}} \quad (30)$$

If the x-ray frequency is near a resonant frequency and if absorption is present, the polarization is more complicated than given above. The dielectric function, is then taken to be a complex quantity and generally has a complicated frequency dependence under such conditions. This can formally be handled by considering the atomic scattering factors as complex and wavelength dependent.

$$F_H = \sum_n (f + \Delta f' + i \Delta f'')_n e^{2\pi i \vec{B} \cdot \vec{r}_n} \quad (31)$$

The corrections to the atomic scattering factor f due to the resonance, $\Delta f'$, and absorption, $\Delta f''$, are called the Honl corrections. All the detailed physics of the scattering and absorption are included in these terms. For convenience, we separate the contributions to F_H of the real parts of the

atomic scattering factors ($f + \Delta f'$) and the imaginary parts ($\Delta f''$) and write F_H as

$$F_H = F_H' + i F_H'' \quad (32)$$

If we consider the $hk\ell = 000$ term in the Fourier Series for the dielectric we see that

$$\begin{aligned} \epsilon_0 &= 1 - \Gamma[F_0' + F_0''] \\ &= 1 - \sum_n (f_0 + \Delta f_0')_n - i\Gamma \sum_n (\Delta f_0'')_n \end{aligned} \quad (33)$$

Since these terms are zero order, they are approximate expressions for the average value of the dielectric constant, i.e. $\epsilon_0 = \langle \epsilon \rangle$. Thus the linear absorption coefficient is related to the imaginary part of the average dielectric constant.

$$\mu_0 = \frac{2\pi}{\lambda} \Gamma F_0'' \quad (34)$$

2.8 Solution of Maxwell's Equations in the Material with Braggs' Law as a Constraint

As mentioned in the introduction, the fundamental problem is to solve Maxwells' equations with Braggs' law as a constraint. This has been done by Batterman [Batterman 1958] but is included here for consistency. These solutions will then be used to develop expressions for the intensity of the anomalous transmission.

Maxwells' Equations for the crystal are

$$\nabla \times \vec{E} = \frac{1}{c} \frac{\partial \vec{B}}{\partial t} = - \frac{1}{c} \frac{\partial \vec{H}}{\partial t} \quad (35)$$

$$\nabla \times \vec{H} = \frac{1}{c} \frac{\partial \vec{D}}{\partial t} \quad (36)$$

We now assume that the fields $\vec{E}$, $\vec{D}$, and $\vec{H}$ can be expressed as sum of plane waves. If we assume there is a diffracted wave then the wave vectors must

satisfy the Bragg equation

$$\vec{K} = \vec{K}_0 + \vec{B} \quad (37)$$

where $\vec{K}_0 = 1/\lambda$.

Although any wave may propagate in the crystal, it is only the waves connected by Eq. 37 which are strongly coupled and of interest in diffraction.

The fields $\vec{D}$, $\vec{E}$, and $\vec{H}$ can be written in the form

$$\vec{A} = e^{i\omega t} \sum_H \vec{A}_H e^{-2\pi i \vec{K}_H \cdot \vec{r}} \quad (38)$$

Using Eq. 37, we have

$$\vec{A} = e^{i\omega t} e^{-2\pi i \vec{K}_0 \cdot \vec{r}} \vec{A}_H e^{-2\pi i \vec{B} \cdot \vec{r}} \quad (39)$$

Remembering that for plane waves the curl operator, ∇ , is equivalent to $-2\pi i \vec{K}$ and the operator $\frac{\partial}{\partial t}$ is equivalent to i , we have for the curl of the electric field

$$\nabla \times \vec{E} = -2\pi i e^{i\omega t} \sum_H (\vec{K}_H \times \vec{E}_H) e^{-2\pi i \vec{K}_H \cdot \vec{r}} \quad (40)$$

and for the time rate of change of the magnetic field

$$\frac{\partial \vec{H}}{\partial t} = i \omega e^{i\omega t} \sum_H \vec{H}_H e^{-2\pi i \vec{K}_H \cdot \vec{r}} \quad (41)$$

Substituting Eq. 40 and 41 into Maxwells' equations results in an equality which must hold true for each Fourier component at all points and all times. Thus from Faradays' law we get

$$2\pi (\vec{K}_H \times \vec{E}_H) = \frac{\omega}{c} \vec{H}_H \quad (42)$$

and from Amperes' law we get

$$2\pi (\vec{K}_H \times \vec{H}_H) = -\frac{\omega}{c} \vec{D}_H \quad (43)$$

Now using $\vec{D} = \epsilon \vec{E}$ and Eq. 30

$$D = \sum_H D_H e^{-2\pi i \vec{K}_H \cdot \vec{r}} = \left(1 - \Gamma \sum_H F_H e^{-2\pi i \vec{B} \cdot \vec{r}}\right) \left(\sum_H E_H e^{-2\pi i \vec{K}_H \cdot \vec{r}}\right) \quad (44)$$

After appropriate changes in the indices of summation and using

$\bar{K}_H = \bar{K}_O + \bar{B}$ and $\bar{K}_H + \bar{B} = \bar{K}_H + H$, Eq. 45 can be rewritten as

$$\sum_H \bar{D}_H e^{-2\pi i \bar{K}_H \cdot \bar{r}} = \sum_H \bar{E}_H e^{-2\pi i \bar{K}_H \cdot \bar{r}} - \Gamma \sum_H \left(\sum_P F_{H-P} \bar{E}_P \right) e^{-2\pi i \bar{K}_H \cdot \bar{r}} \quad (45)$$

$$D_H = E_H - \Gamma \sum_P F_{H-P} D_P = (1 - \Gamma F_O) E_H - \sum_{P \neq H} F_{H-P} E_P \quad (46)$$

$\bar{D}_H$ is thus predominately $(1 - \Gamma F_O) \bar{E}_H = \langle \epsilon \rangle \bar{E}_H$ but modified slightly by small contributions from the other Fourier components of the electric field.

By taking the cross product of $\bar{K}_H$ with Eq. 42 and substituting into 43

$$2\pi \bar{K}_H \times (\bar{K}_H \times \bar{E}_H) = \frac{\omega}{c} \bar{K}_H \times H_H$$

$$\bar{K}_H \times (\bar{K}_H \times \bar{E}_H) = -\frac{\omega^2}{4\pi^2 c^2} \bar{D}_H \quad (47)$$

$$\bar{K}_H \times (\bar{K}_H \times \bar{E}_H) = \frac{-\omega^2}{4\pi^2 c^2} (\bar{E}_H - \Gamma \sum_P F_{H-P} E_P) \quad (48)$$

$$= k_o^2 (E_H - \Gamma \sum_P F_{H-P} E_P) \quad (49)$$

$$\bar{K}_H \times (\bar{K}_H \times \bar{E}_H) - k_o^2 \bar{E}_H - k_o^2 \Gamma \sum_P F_{H-P} E_P = 0 \quad (50)$$

The vector identity for the triple cross product gives

$$\bar{K}_H \times (\bar{K}_H \times \bar{E}_H) = -(\bar{K}_H \cdot \bar{K}_H) \bar{E}_H + (\bar{K}_H \cdot \bar{E}_H) \bar{K}_H \quad (51)$$

so Eq. 48 becomes

$$-(\bar{K}_H \cdot \bar{K}_H) \bar{E}_H + (\bar{K}_H \cdot \bar{E}_H) \bar{K}_H + k_o^2 \bar{E}_H - k_o^2 \Gamma \sum_P F_{H-P} E_P = 0 \quad (52)$$

$$[k_o^2 (1 - \Gamma F_O) - (\bar{K}_H \cdot \bar{K}_H)] \bar{E}_H - k_o^2 \Gamma \sum_{P \neq H} F_{H-P} \bar{E}_P + (\bar{K}_H \cdot \bar{E}_H) \bar{K}_H = 0 \quad (53)$$

This is the fundamental set of equations describing the field inside the crystal. However, for anomalous transmission all of the field amplitudes

can be neglected in comparison to $\bar{E}_O$ and $\bar{E}_H$. Physically all waves exist in the crystal, but we assume only two have appreciable amplitude. The wave vectors $\bar{K}_O$ and $\bar{K}_H$ define the plane of incidence.

The waves can be considered as having two components, one perpendicular to the plane of incidence, referred to as the σ component, and one in the plane of incidence, referred to as the π component. For the σ component, Eq. 53 becomes

$$[k_O^2(1 - \Gamma F_O) - (\bar{K}_O \cdot \bar{K}_O)]E_O^\sigma - k_O^2 \Gamma F_H E_H^\sigma = 0 \quad (54)$$

$$-k_O^2 \Gamma F_H \bar{E}_O^\sigma + [k_O^2(1 - \Gamma F_O) - \bar{K}_H \cdot \bar{K}_H] \bar{E}_H^\sigma = 0 \quad (55)$$

where $F_H = F(-h-l-k)$.

This pair of equations has a nontrivial solution only if the determinant of the coefficients equals zero.

For the π component, E_O and E_H each have two components of the plane of incidence. One being a longitudinal component along the respective wave vectors and the other a transverse component. The longitudinal components can be shown to be negligible. If $\bar{K}_H$ is dotted into Eq. 52 we have

$$\bar{K}_H \cdot \bar{E}_H(1 - \Gamma F_O) = \Gamma \sum_{P \neq H} F_{H-P} (\bar{K}_H \cdot \bar{E}_P) \quad (56)$$

which since $\Gamma F \sim 10^{-5}$ implies that the component of $\bar{E}_H$ along $\bar{K}_H$ may be neglected.

Since the angle between $\bar{K}_O$ and $\bar{K}_H$ is 2θ we can write for the π components

$$[k_O^2(1 - \Gamma F_O) - \bar{K}_O \cdot \bar{K}_O]E_O^\pi - k_O^2 \cos(2\theta) F_H E_H^\pi \approx 0 \quad (57)$$

$$-k_O^2 \cos(2\theta) \Gamma F_H E_O^\pi + [k_O^2(1 - \Gamma F_O) - (\bar{K}_H \cdot \bar{K}_H)]E_H^\pi \approx 0 \quad (58)$$

If we let $P = 1$ for σ polarization and $P = \cos 2\theta$ for the π polarization then the determinant of Eqs. 57 and 58 can be written

$$\begin{vmatrix} k_o^2(1 - \Gamma F_o) - \bar{k}_o \cdot \bar{k}_o & -k_o^2 \Gamma P F_H \\ -k_o^2 \Gamma P F_H & k_o^2(1 - \Gamma F_o) - \bar{k}_H \cdot \bar{k}_H \end{vmatrix} \quad (59)$$

Factoring our k_o^2 , this can be written in the form

$$\begin{vmatrix} < \epsilon > - \epsilon_o & - \Gamma P F_H \\ - \Gamma P F_H & < \epsilon > - \epsilon_H \end{vmatrix} \quad (60)$$

which, when set equal to zero, gives the permitted solutions. If either the dielectric constant in the forward direction, ϵ_o , or the diffracted direction, ϵ_H , do not differ from the average dielectric constant then there is no solution. k_o is the vacuum value of the wave vector while $\bar{k}_o$ and $\bar{k}_H$ are the wave vectors inside the crystal. For crystal having unit cells with a center of symmetry, $F_H = F_H$. Thus the non trivial solution is

$$(< \epsilon > - \epsilon_o) (< \epsilon > - \epsilon_H) = P^2 \Gamma^2 F_H^2 \quad (61)$$

which is our dispersion relation. From Eq. 57 we find the allowed ratio of field amplitudes is given by

$$Z = \frac{E_H}{E_o} = \frac{\epsilon_o - < \epsilon >}{\Gamma P F_H} \quad (62)$$

At the boundary $\hat{n} \cdot \hat{r} = 0$ the incident wave must match with the wave in the crystal where there is no change in amplitude or frequency in crossing the surface. This places a restriction on the wave vectors. Since

$$\bar{k}_o \cdot \bar{r} = \bar{k}_o \cdot \bar{r} \quad (63)$$

and at the boundary r is in the surface, then $\bar{k}_o$ can differ from $\bar{k}_o$ by at

most a vector normal to the surface. So

$$\vec{K} = \vec{k}_o + a \hat{n} \quad (64)$$

where $\hat{n}$ is a unit vector normal to the surface and a is yet to be determined. The dielectric function differs only slightly from one, therefore, we can set the index of refraction, $\sqrt{\epsilon}$, equal to $1 + \delta_o$ for the forward diffracted wave and hence

$$K_o^2 = k_o^2 (1 + \delta_o)^2 \cong k_o^2 (1 + 2\delta_o) \quad (65)$$

But from the above boundary condition

$$K_o^2 = k_o^2 (1 + 2\delta_o) = (\vec{k}_o + \Delta \hat{n})^2 \quad (66)$$

$$\text{which implies } \frac{k_o^2 \delta_o}{\vec{k} \cdot \hat{n}} = \frac{k_o \delta_o}{\gamma_o} \quad (67)$$

where γ_o is the direction cosine of the forward diffracted wave vector.

Similarly for the diffracted wave we have

$$K_H^2 = k_o^2 (1 + \delta_H)^2 \cong k_o^2 (1 + 2\delta_H) \quad (68)$$

Combining this equation with the Bragg equation $K_H = \vec{k}_o + \vec{B}$ we have

$$K_H = k + \frac{k_o \delta_o}{\gamma_o} \hat{n} + \vec{B} \quad (69)$$

squaring both sides and setting it equal to Eq. 68 we obtain

$$\delta_H = \delta_o \left(1 + \frac{\hat{n} \cdot \vec{B}_H}{k_o \gamma_o} \right) + \frac{\vec{k}_o \cdot \vec{B}_H}{k_o^2} + \frac{B_H^2}{2k_o^2} \quad (70)$$

If we restrict ourselves to crystals with the diffracting planes perpendicular to the surface $\hat{n} \cdot \vec{r} = 0$, then Eq. 70 reduces to

$$\delta_H = \delta_o + \frac{\alpha}{a} \quad (71)$$

$$\text{where } \alpha = \frac{1}{k_o^2} \left(B_H^2 + 2\vec{k}_o \cdot \vec{B}_H \right) \quad (72)$$

Taking the dispersion relation

$$(\epsilon_o - \langle \epsilon \rangle) (\epsilon_H - \langle \epsilon \rangle) = p^2 \Gamma^2 F_H^2$$

and using $\langle \epsilon \rangle = 1 - \Gamma F_o$ and

$$\epsilon_o = 1 + 2\delta_o$$

$$\epsilon_H = 1 + 2\delta_H$$

we can rewrite the dispersion relation as

$$(2\delta_o + \Gamma F_o)(2\delta_H + \Gamma F_o) = p^2 \Gamma^2 F_H^2 \quad (73)$$

$$(2\delta_o + \Gamma F_o)(2\delta_o + \Gamma F_o + \alpha) = p^2 \Gamma^2 F_H^2 \quad (74)$$

Eq. 62 for the ratio of E_H to E_o can be substituted to yield

$$Z^2 + \frac{\alpha}{\Gamma p F_H} Z - 1 = 0 \quad (75)$$

whose solution is

$$Z = \left\{ \begin{matrix} Z_1 \\ Z_2 \end{matrix} \right\} = Y \pm \sqrt{Y^2 + 1} \quad (76)$$

$$\text{where } Y^2 = \alpha^2 / 4 \Gamma^2 p^2 F_H^2 \quad (77)$$

Solving Eq. 74 for δ_o we get

$$\delta_o = \left\{ \begin{matrix} \delta_o' \\ \delta_o'' \end{matrix} \right\} = -\frac{1}{2} (\Gamma F_o + \frac{\alpha}{2}) \pm \frac{p \Gamma F_H}{2} \sqrt{Y^2 + 1} \quad (78)$$

Since there are two possible values of δ_o and Z there are two forward diffracted beams and two diffracted waves in the general case. Each of these have two states of polarization. The general form of the forward diffracted wave is

$$e^{i\omega t - 2\pi i \vec{k}_o \cdot \vec{r}} \left[E_o' e^{-i\psi_1 x} + E_o'' e^{-i\psi_2 x} \right] \quad (79)$$

and for the diffracted beam

$$e^{i\omega t - 2\pi i(\vec{k}_o + \vec{B} \cdot \vec{r})} \left[z_1 E_o' e^{-i\psi_1 x} + z_2 E_o'' e^{-i\psi_2 x} \right] \quad (80)$$

where

$$\psi_1 = 2\pi k_o \delta_o' \quad (81)$$

$$\psi_2 = 2\pi k_o \delta_o'' \quad (82)$$

$$x = \hat{n} \cdot \vec{r} \quad (83)$$

Now we can use the boundary conditions to remove the arbitrariness in the ratio $E_o'' : E_o'$. At the surface $\hat{n} \cdot \vec{r} = 0$ the boundary condition is given by

$$E_o' + E_o'' = E_o^e \quad (84)$$

Since no diffracted wave enters the crystal through $\hat{n} \cdot \vec{r} = 0$, then the diffracted wave must vanish at this surface

$$z_1 E_o' + z_2 E_o'' = 0 \quad (85)$$

Thus, we have

$$E_o' = \frac{z_2}{z_2 - z_1} E_o^e \quad (86)$$

$$E_o'' = \frac{-z_2}{z_2 - z_1} E_o^e \quad (87)$$

and now the internal waves are completely determined. Letting I_o^e be the incident beam intensity we can obtain the forward diffracted and diffracted beam intensities, I_o , and I_H , respectively by setting $x = x_o$ in Eq. (79) and (80) and using Eqs. (86) and (92)

$$\frac{I_H}{I_o^e} = \left| \frac{z_1 z_2}{z_2 - z_1} \left(e^{-i\psi_1 x_o} - e^{-i\psi_2 x_o} \right) \right|^2 \quad (88)$$

$$\frac{I_o}{I_o^e} = \left| \frac{Z_2 e^{-\psi_1 x_o} - Z_1 e^{-i\psi_2 x_o}}{Z_2 - Z_1} \right|^2 \quad (89)$$

We can simplify Eq. 88 by using Eqs. 76, 81, 82 and recalling that F_H is a complex quantity, $F_H = F_H' + iF_H''$.

$$v + iw = \sqrt{\frac{a^2}{4} + p^2 \Gamma^2 F_H F_H^*} \quad (90)$$

$$= P \Gamma F_H \sqrt{Y^2 + 1} \quad (91)$$

where

$$v = P \Gamma F_H' \sqrt{Y^2 + 1} \quad (92)$$

$$w = P \Gamma F_H'' \sqrt{Y^2 + 1} \quad (93)$$

and let

$$C_1 = e^{-\frac{i\psi_1 x}{Y_o}} \quad (94)$$

$$C_2 = e^{-\frac{i\psi_2 x}{Y_o}} \quad (95)$$

we find

$$|C_1 - C_2|^2 = 2e^{-\frac{2\pi k x_o \Gamma F_o''}{Y_o}} \{ \cosh 2aw - \cos 2av \} \quad (96)$$

$$\text{where } a = \frac{\pi k x_o}{Y_o} \quad (97)$$

Using the two identities

$$\cosh 2\theta = 1 + 2 \sinh^2 \theta \quad (98)$$

$$\cos 2\theta = 1 - 2 \sin^2 \theta \quad (99)$$

we obtain

$$\begin{aligned} \left| c_1 - c_2 \right|^2 &= 4e^{-2a\Gamma_F^2} \left\{ \sinh^2 a_w + \sin^2 a_v \right\} \\ &= 4e^{-\frac{\mu_o x_o}{\gamma_o}} \left\{ \sinh^2 a_w + \sin^2 a_v \right\} \end{aligned} \quad (100)$$

The expression $\frac{z_1 z_2}{z_2 - z_1}$ can be evaluated using Eq. 76. The result is

$$\frac{z_1 z_2}{z_2 - z_1} = -\frac{1}{2 \gamma^2 + 1} \quad (101)$$

and thus

$$\frac{I_H}{I_o^e} = \frac{e}{\gamma^2 + 1} - \frac{\mu_o x_o}{\gamma_o} \left\{ \sinh^2 a_w + \sin^2 a_v \right\} \quad (102)$$

It is interesting at this point to look at the form of this expression for the diffracted beam for the case of zero absorption. No absorption means the imaginary part of the structure factor is zero or since the structure factor is directly proportional to the polarizability per unit volume another way to look at it is that the polarizability is real. This means $w = 0$ and thus for no absorption

$$\frac{I_H}{I_o^e} = \frac{\sin^2 a_v}{\gamma^2 + 1} \quad (103)$$

Even though we will stay with γ as the independent variable, one must remember that γ can be expressed as a function of either θ or λ . For now it is convenient to show that γ is approximately a linear function of λ . From the definition of α , Eq. 72, we have

$$\alpha = \frac{1}{k_o^2} \left\{ \overline{B}_H^2 + 2 \overline{k}_o \cdot \overline{B}_H \right\} \quad (72)$$

$$\begin{aligned}
&= \lambda_o^2 \left\{ \frac{1}{d^2} + \frac{2 \cos(\pi/2+\theta)}{\lambda_o d} \right\} \\
&= \lambda_o^2 \left\{ \frac{4 \sin^2 \theta_\beta}{2} - \frac{4 \sin \theta_\beta \sin \theta}{\lambda_o \lambda_\beta} \right\} \quad (104)
\end{aligned}$$

$$\begin{aligned}
&= 4 \lambda_o^2 \sin^2 \theta_\beta \left(\frac{1}{\lambda_\beta^2} - \frac{1}{\lambda_o \lambda_\beta} \right) \\
&= 4 \frac{(\lambda_o - \lambda_\beta)}{\lambda_\beta} \sin^2 \theta_\beta \quad (105)
\end{aligned}$$

Thus α is approximately a linear function of the incident wave length λ . We assumed that $\lambda_o \cong \lambda_\beta$ and we will see that this is a valid assumption.

Returning now to Eq. 103, we see that even if there were no absorption, since the diffracted beam is wavelength dependent, we must integrate the intensity expression with respect to Y (or λ_o) to obtain the total intensity. Before doing this, however, it is interesting to note that Eq. 103 predicts that the diffraction pattern will exhibit interference fringes. This is easier to see if Eq. 103 is rewritten in the form

$$\frac{I_H}{I_e} = \frac{\sin^2 (A \sqrt{1+Y^2})}{Y^2 + 1} \quad (106)$$

where

$$A = \frac{\pi k x_o}{Y_o} \text{ PTF}_H' \quad (107)$$

This raises the question of how does the variation in thickness of the crystal compare with argument $\sin^2 (A \sqrt{1+Y^2})$. Because if the argument varies by π while x_o varies by a small amount then we need to replace $\sin^2 (A \sqrt{1+Y^2})$ by its average value of $1/2$. For a Ag K x-ray and a 1 cm thick Ge crystal, $A \sqrt{1+Y^2} \cong 1.5 \times 10^3$ and since the variation in x_o could be π parts in 1.5×10^3 then

$$\frac{I_H}{I_o^e} = \frac{1}{2(1 + Y^2)} \quad (108)$$

and the integrated intensity becomes

$$R_Y^H = \int_{-\infty}^{\infty} \frac{1}{2} \frac{dY}{1 + Y^2} = \frac{\pi}{2} \quad (109)$$

This value of the integrated intensity without absorption will be used next when we calculate an effective absorption coefficient for the diffracted beam.

2.9 Absorbing Crystals

For absorbing crystals the fundamental equation for the diffracted intensity is Eq. 107

$$\frac{I_H}{I_o^e} = \frac{1}{2} \frac{e^{-\frac{\mu_o x_o}{Y_o}}}{Y^2 + 1} \cosh 2aw \quad (110)$$

where we have replaced $\sin^2 aw$ by its average value of $1/2$. First lets evaluate w from its defining equation

$$v + iw = \sqrt{\frac{a^2}{4} + p^2 \Gamma^2 F_H F_H^*} \quad (111)$$

For crystals with an inversion center $F_H = F_H^*$ and since $F_H^{''} \ll F_H'$, then

$|F_H|^2 \cong |F_H'|^2$. Now

$$\begin{aligned} F_H &= F_H' + iF_H^{''} \\ &= F_H' \left(1 + \frac{F_H^{''}}{F_H'} i\right) \\ &= F_H' (1 + is) \end{aligned} \quad (112)$$

where $s = \frac{F_H^{''}}{F_H'} \ll 1$.

$$|F_H|^2 = |F_H'|^2 (1 + is)^2 \quad (113)$$

$$F_H'^2 (1 + 2is)$$

so recalling that $z = \alpha/2P\Gamma F_H'$ we have

$$v + iw = \sqrt{\frac{\alpha}{4} + P^2 \Gamma^2 F_H'^2 (1 + 2is)} \quad (114)$$

$$= P\Gamma |F_H'| \sqrt{z^2 + 1 + 2is} \quad (115)$$

Expanding the square root we obtain

$$w = P\Gamma F_H' \frac{s}{\sqrt{1 + Y^2}} \quad (116)$$

$$aw = \frac{\pi k_o x_o}{Y_o} \Gamma P |F_H'| \frac{s}{\sqrt{1 + Y^2}} \quad (117)$$

and Eq. 110 becomes

$$\begin{aligned} \frac{I_H}{I_o e} &= \frac{1}{2} \frac{e^{-\frac{\mu_o x_o}{Y_o}}}{Y^2 + 1} \cosh \frac{2\pi k_o x_o P\Gamma |F_H'| s}{Y_o \sqrt{1 + Y^2}} \\ &= \frac{1}{2} \frac{e^{-\frac{\mu_o x_o}{Y_o}}}{(1 + Y^2)} \cosh \left(\frac{2As}{\sqrt{1 + Y^2}} \right) \end{aligned} \quad (118)$$

where

$$A = \frac{\pi k_o x_o P |F_H'|}{Y_o}$$

Using $s = \frac{F_H''}{F_H'}$

$$\mu_o = 2\pi k_o \Gamma F_o'' = 2\pi k_o \frac{P\Gamma F_H''}{\epsilon}$$

$$x = \frac{x_o}{Y_o}$$

$$\epsilon = P F_H''/F_o''$$

Equation (123) can be expressed in terms of more familiar quantities

$$\frac{I_H}{I_e} = \frac{1}{2} \frac{e^{-\mu_o x}}{1 + Y^2} \cosh \left(\frac{\mu_o x \epsilon}{\sqrt{1 + Y^2}} \right) \quad (119)$$

For thick crystals $\mu_o x$ is large and varies much more rapidly than $1 + Y^2$.

Thus to a first approximation we can write

$$\frac{I_H}{I_e} = \frac{1}{4} e^{-\mu_o x} e^{\mu_o x \epsilon} e^{-\frac{1}{2} \mu_o x \epsilon Y^2}$$

where

$$\frac{1}{\sqrt{1 + Y^2}} \approx \frac{1}{1 + \frac{1}{2} Y^2} \approx 1 - \frac{1}{2} Y^2$$

The integration of this expression gives the integrated intensity, R_H^Y

$$R_H^Y = \int_{-\infty}^{\infty} \frac{I_H}{I_e} dY = \sqrt{\frac{\pi}{8\epsilon\mu_o x}} e^{-\mu_o x(1-\epsilon)} \quad (120)$$

for either the diffracted or forward diffracted beam. Equation (120) really represents two equations, one for the parallel polarization component of the incident x-rays and one for the perpendicular component. Thus we have:

$$\parallel_{R_H}^Y = \left(\frac{\pi}{8\mu_o x \cos 2\theta_\beta} \frac{F_o'''}{F_H'''} \right)^{1/2} \exp \left[-\mu_o x \left(1 - \frac{F_H'''}{F_o'''} \cos(2\theta_\beta) \right) \right] \quad (121)$$

$$\perp_{R_H}^Y = \left(\frac{\pi}{8\mu_o x} \frac{F_o'''}{F_H'''} \right)^{1/2} \exp \left[-\mu_o x \left(1 - \frac{F_H'''}{F_o'''} \right) \right] \quad (122)$$

In an experiment the average of these are measured, that is

$$R_H^Y = \frac{1}{2} (\parallel_{R_H}^Y + \perp_{R_H}^Y) \quad (123)$$

For thick crystals and low energies this reduces to

$$R_H^Y = \frac{1}{2} R_T^Y$$

$$= \frac{1}{2} \left(\frac{\pi}{8\mu_0 x} \frac{F_o'''}{F_H'''} \right)^{1/2} \exp \left[-\mu_0 x \left(1 - \frac{F_H'''}{F_o'''} \right) \right] \quad (124)$$

It is important to remember that this represents the relative intensity for a parallel beam of x-rays. Also, it is expressed in terms of the parameter Y but can be expressed in terms of the wavelength by using equations (105) and (77)

$$\begin{aligned} R_H^\lambda &= \int_{-\infty}^{\infty} \frac{I_H}{I_o} d\lambda = \frac{d\lambda}{dY} \int_{-\infty}^{\infty} \frac{I_H}{I_o} dY \\ R_H^\lambda &= \frac{PF\lambda_\beta}{2\sin\theta_\beta} R_H^Y \end{aligned} \quad (125)$$

If the crystal is not rotated but remains fixed during the measurement then R_H^λ must be related to what is experimentally measured. At an angle θ the intensity ratio is given by

$$\begin{aligned} \frac{I(\theta)}{I_o} &= \int_{\lambda_1}^{\lambda_2} I(\lambda) d\lambda \bigg/ \int_{\lambda_1}^{\lambda_2} I_o(\lambda) d\lambda \\ &= \int_{\lambda_1}^{\lambda_2} I_o(\lambda) \left[\frac{I(\lambda)}{I_o(\lambda)} \right] d\lambda \bigg/ \int_{\lambda_1}^{\lambda_2} I_o(\lambda) d\lambda \end{aligned} \quad (126)$$

If $I(\lambda)$ varies rapidly with λ within a range less than $(\lambda_2 - \lambda_1)$, so that $I_o(\lambda)$ can be considered constant during the integration, then

$$\begin{aligned} \frac{I(\theta)}{I_o} &= \int_{\lambda_1}^{\lambda_2} \frac{I(\lambda)}{I_o(\lambda)} d\lambda \bigg/ \int_{\lambda_1}^{\lambda_2} d\lambda \\ &= \frac{R_H^\lambda}{\Delta\lambda} \end{aligned} \quad (127)$$

The wavelength range of the diffracted beam is given by

$$\Delta\lambda = \lambda \cot\theta \Delta\theta \quad (128)$$

where the angular width $\Delta\theta$ is determined by the experimental conditions. It is the average value of $I(\theta)/I_o^e$ which is determined experimentally. The average value is given by

$$\frac{I(\theta)}{I_o^e} = \frac{\int_{\Delta\lambda}^{R_H} i(\theta) d\theta}{\int i(\theta) d\theta} \quad (129)$$

where the weighting factor $i(\theta)$ is the number of photons incident at an angle θ . This angular distribution will be discussed in section 5.4.

2.10 The Parameter Epsilon

In the previous section we found that the intensity is controlled by the parameter ϵ where

$$\epsilon = P F_H''/F_o'' \quad (130)$$

but the dynamical theory did not tell us how to evaluate the imaginary parts of the structure factor for the diffracted and forward diffracted beams. For diffraction studies one needs F_H , but since the imaginary part is very small compared to F_H there is no real need to know the imaginary part. However, for anomalous transmission knowledge of the imaginary part is vital. Semi-classical calculations of the imaginary part has been made by Batterman [1962] but it does not give the correct dependence on energy. Wagenfeld [1966] has shown using a quantum mechanical calculation that for diamond like structures ϵ is given by

$$\epsilon = a_H (1 - 2\sin^2\theta_\beta \tau^Q/\tau) e^{-M} \quad (131)$$

where a_H is a geometrical term relating the structure factor to the atomic scattering factor, $\tau = \tau^D + \tau^Q$ is the linear photoelectric absorption coefficient, τ^D is the dipole term, τ^Q is the quadrupole term, and e^{-M} is the Debye-Waller factor. Ohtsuki [1964] has shown theoretically that the Debye-

Waller factor gives the correct temperature dependence of ϵ . This has been verified experimentally by Batterman [1962], Okerse [1962], and Ling [1965] for the $K\alpha$ x-rays of Cu and Mo. However, this is in the region where absorption can be accounted for by photoelectric absorption only. At high energies the Compton cross section must be included. Giardian [1973] has shown that contribution of Compton scattering to the effective absorption coefficient is given by

$$\mu_c = \frac{8\pi r_e^2}{3} N \frac{\lambda^2}{(\lambda + \lambda_c)^2} (Z - f_H e^{-M}) \quad (132)$$

where N is the number of atoms per cm^3 , and λ_c is the Compton wavelength $= 2.4263 \times 10^{-10} \text{ cm}$.

The Debye-Waller factor will be discussed in detail in the next section.

2.11 The Effect of Temperature on the Intensity

So far we have assumed the atoms to be fixed in position. The probability of absorption would be zero if all the electrons were concentrated in the nodes of the electric field. The thermal vibrations of the atoms and the finite volume of their electron cloud prevent the absorption from disappearing. It is found experimentally that the intensity of the anomalous transmission of x-rays through a nearly perfect crystal varies with the temperature of the crystal. The intensity decreases as the thermal vibration of the atoms increases. We wish to account for these temperature effect in our theory.

We have shown that if all the atoms of the unit cell scatter in phase, ϵ is given by

$$\epsilon = \frac{f_H'''}{f_0'''} P$$

where f_H'' and f_O'' are the imaginary parts of the atomic scattering factor in the direction of the diffracted and forward diffracted beams respectively. Ohtsuki [1964] has shown theoretically that the temperature dependence of the imaginary part of the atomic scattering factor should be equal to that of the real part as expected from previous experiments. This temperature dependence is given by the Debye-Waller factor, e^{-M} , where M is given by

$$M = 8\pi^2 \langle u_s^2 \rangle \frac{\sin^2 \theta}{\lambda^2} \quad (133)$$

where $\langle u_s^2 \rangle$ is the mean square of the component of displacement along the $s^n - s_o^n$ direction, that is, perpendicular to the diffraction planes. The mean square displacement $\langle u_s^2 \rangle$ is given by

$$u_s^2 = \frac{3}{4} \frac{h^2 T}{mk\pi^2 \theta_m} \left[\Phi(x) + \frac{x}{4} \right] \quad (134)$$

where

T - temperature in degrees Kelvin

h - Planks constant

θ_m - Debye temperature

k - Boltzman constant

m - mass of the atom

$$x = \frac{\theta_m}{T} \quad (135)$$

and

$$\Phi(x) = \frac{1}{x} \int_0^x \frac{\eta d\epsilon}{e^\eta - 1} \quad (136)$$

where

$$\eta = \frac{h\nu}{kT}$$

The Debye function for amplitudes $[\tilde{Q}(x) + \frac{x}{4}]$ is a very slowly varying function of temperature as will be seen in the section on calculations.

Using this temperature factor, $D_H(T) = e^{-M}$ in our dynamic theory, the minimum absorption coefficient becomes

$$\mu_{\min} = \nu(1 - D_H(T)) \quad (137)$$

Thus the ratio of the intensities at temperatures T_1 and T_2 becomes

$$\frac{I_1}{I_2} = e^{[D_H(T_1) - D_H(T_2)]} \frac{\mu x_o}{\gamma_o} \quad (138)$$

This equation explains the main features of the temperature dependence. Since $D_H(T)$ is usually only slightly less than one, then μx must be quite large to obtain much enhancement of the intensity by cooling. At the same time, large values of μx result in large absorption factors and play a much more important role than the temperature. This temperature dependence has been verified by Okerse [1962], Batterman [1964], and Wagenfeld [1965].

CHAPTER 3

CRYSTALS USED IN THE EXPERIMENT

3.1 Introduction

Although there are many natural and man-made crystals, those that are of interest in anomalous transmission are limited by such considerations as extreme purity, low dislocation density, high atomic number, and availability in sizes large enough to be of practical value. Anomalous transmission has been observed in single crystals of quartz Borrmann [1941], silicon Batterman [1961], and silver using the $K\alpha$ x-rays from copper and molybdenum. However, quartz and silicon were not considered because of their low atomic number and the single crystal of silver was 1 cm in length but only 1 mm in diameter. Unless a major effort were undertaken to try and grow large single crystals specifically for anomalous transmission one is limited to crystals which have been experimentally studied for possible application as solid-state detectors. One of these crystals, HgI, unfortunately must be handled with extreme care since the order is easily destroyed. Another high atomic number crystal is CdTe. Several samples of CdTe were obtained from researchers, but they were either not single crystals or too small to be of practical value. One material which is readily available is germanium. Samples of dislocated and dislocation free germanium which had been grown in a nitrogen or hydrogen atmosphere were obtained.

Before the crystal is cut one has to decide on the set of planes to be used for anomalous transmission. Since intensity is a prime consideration, the planes having the large structure factor are selected. In this chapter the structure factor for germanium is calculated. In addition, the preparation of the germanium crystals is described.

3.2 Structure Factor for Germanium

Germanium has a diamond structure with eight atoms at the positions:
 000; 0 1/2 1/2; 1/2 0 1/2; 1/2 1/2 0; 1/4 1/4 1/4; 1/4 3/4 1/4; 3/4 1/4 3/4;
 3/4 3/4 1/4. Fig. 3.2.1 show the diamond structure. Thus the structure
 factor becomes

$$\begin{aligned}
 F &= f_{\text{Ge}} \left[1 + e^{2\pi i \left(\frac{k}{2} + \frac{l}{2} \right)} + e^{2\pi i \left(\frac{h}{2} + \frac{l}{2} \right)} + e^{2\pi i \left(\frac{h}{2} + \frac{k}{2} \right)} \right] \\
 &+ f_{\text{Ge}} \left[e^{2\pi i \left(\frac{h}{4} + \frac{k}{4} + \frac{l}{4} \right)} + e^{2\pi i \left(\frac{h}{2} + \frac{3k}{4} + \frac{3l}{4} \right)} \right. \\
 &\quad \left. + e^{2\pi i \left(\frac{3h}{4} + \frac{k}{4} + \frac{3l}{4} \right)} + e^{2\pi i \left(\frac{3h}{4} + \frac{3k}{4} + \frac{l}{4} \right)} \right] \\
 &= f_{\text{Ge}} \left(1 + e^{\frac{\pi i}{2}(h+k+l)} \right) \left(1 + e^{\pi i(k+l)} + e^{\pi i(h+l)} + e^{\pi i(h+k)} \right) \\
 F^2 &= f_{\text{Ge}}^2 \cdot 2 \left(1 + \cos(h+k+l) \frac{\pi}{2} \right) \\
 &\quad (4 + 2(\cos \pi(k+l) + \cos \pi(k-l)) + 2(\cos \pi(h+l) + \cos \pi(h-l)) \\
 &\quad + 2(\cos \pi(h+k) + \cos \pi(h-k))
 \end{aligned}$$

There are four cases to consider

Case I h, k, l are all even

$$F^2 = f_{\text{Ge}}^2 \cdot 4 \cdot 16 = 64 f_{\text{Ge}}^2$$

$$F = 8 f_{\text{Ge}}$$

Case II one is even and 2 are odd

$$F^2 = f_{\text{Ge}}^2 \cdot 2 \left(1 + \cos(h+k+l) \frac{\pi}{2} \right) = 0$$

$$F = 0$$

Fig. 3.2.1 The Diamond Structure

Case III one odd and two are even

$$F^2 = f_{\text{Ge}}^2 \cdot 2(1 + \cos(h+k+l)\frac{\pi}{2}) \cdot 0$$

$$F = 0$$

Case IV h, k, l are all odd

$$F^2 = f_{\text{Ge}}^2 \cdot 2 \cdot 16 \quad \text{when } h+k+l = 4n \text{ where } n \text{ is a positive integer.}$$

$$F = \frac{8}{\sqrt{2}} f_{\text{Ge}}$$

In summary, we have

$$F = 8a_H f_{\text{Ge}}$$

$$\text{where } a_H = 1$$

$$h, k, l, \text{ all even and } h+k+l = 4n$$

$$= 1/\sqrt{2}$$

$$n, k, l, \text{ all odd}$$

$$= 0$$

$$\text{all other } h, k, l$$

3.3 Crystal Plane Selection

In 3.2 it was shown that $F = 8f_{\text{Ge}}$ for h, k, l all even and $h + k + l = 4n$; $F = \frac{8}{\sqrt{2}} f_{\text{Ge}}$ for h, k, l all odd; $F = 0$ for all other cases. $F = 0$ means that there will not be an anomalous transmission. Thus, anomalous transmission will occur for such planes as (220) and (400) but will not for (100), (111), and (110) planes.

In 2.9 it was shown that the parameter ϵ controls the anomalous transmission intensity for various sets of planes with the transmission increasing with increasing values of ϵ . We can see from the relation

$$\epsilon = \frac{e^{-m} a_{\mathbf{j}} \mu_{\mathbf{j}}(\omega) f_{\mathbf{j}}(hkl)}{\mu(\omega)}$$

that the intensity increases with an increasing scattering factor and a decreasing m . In 2.3 it was shown that the scattering factor decreases

with increasing

$$\frac{\sin}{\lambda} = \frac{1}{2d} = \frac{h^2 + k^2 + \ell^2}{2a}$$

In addition, m is proportional to $\sin^2\theta/\lambda^2$ and thus the greater intensities are associated with the smaller values of h, k, ℓ . For this reason the (220) planes were chosen for the germanium crystals.

3.4 Crystal Preparation

The back-reflection Laue method was used to verify the [100] direction of growth and to orient the crystals for cutting with a diamond saw. Then they were lapped with a 15 micron lapping compound to remove the surface damage caused by the diamond saw and chemically polished with fuming nitric acid to remove surface damage left by the lapping process. The crystals were then ready for anomalous transmission measurements. All of the crystals used in the experiment are shown in Figs. 3.4.1 and 3.4.2.

Fig. 3.4.1 Germanium Crystals Used in the Borrmann Experiment

Fig. 3.4.2 Germanium Crystals Used in the Borrmann Experiment

CHAPTER 4

EXPERIMENTAL APPARATUS

4.1 Introduction

An anomalous transmission measurement consists of a source of x-rays, a single nearly perfect crystal, a pre-crystal collimator to limit the angular divergence of the x-rays incident onto the crystal, an x-ray detector to measure the anomalous transmission, an apparatus to rotate and align the detector, and the associated counting electronics. The experimental setup used is depicted in Fig. 4.1.1. The diffracted beam rather than the forward diffracted beam was selected for measurement to avoid interference from the $e^{-\mu x}$ component and to minimize fluorescent x-rays in the measured spectra. In this chapter the experimental apparatus is described in detail. The description starts with the x-ray tube used to produce the bremsstrahlung input spectrum. Next the design of the pre-crystal collimator is described since it determines the full width at half maximum of the anomalous transmission spectra. Also the effects of crystal thickness, Bragg angle setting, crystal-to-detector distance, and pre-crystal collimator on the cross sectional area of the diffracted x-ray beam is discussed. The goniometer for rotating the germanium crystal and the rotation and translation tables for detector alignment will be described. Finally the detector and counting electronics used to make all input bremsstrahlung and anomalous transmission spectral measurements are discussed.

4.2 The X-Ray Tube

A Picker x-ray tube with a tungsten target and a .0762 mm Be window provided a source of x-rays. It has a maximum output rating of 160 kV at a 12 ma with 0.3 percent voltage and current regulation provided by a Universal

Fig. 4.1.1 Experimental Setup

Voltronics high voltage supply. The output voltage can be varied from 1 ma to 10 ma in .01 ma steps in the automatic control mode. In the manual mode the current can be varied continuously from zero to ten ma. The control module voltage settings were corrected for the voltage drop across a load resistor. It was found that the output of the tube was only approximately a linear function of the current. These measurements were made using a Radocon II integrating ratemeter designed to measure high and low-level radiation over a wide range of energies. The results are shown in Figs. 4.2.1 through 4.2.5 with the intensity normalized to 1.0 at 1.0 ma for various voltages ranging from 20 to 60 kV. From 60 kV on up the output intensity varied linearly with current.

4.3 Collimator Design

The collimators play a very important role in the experiment. They are the major factor in determining the shape of the peak and also the full width at half maximum.

In Appendix A it is shown that the full width of the peak is given by

$$\Delta E = E_f - E_i = \frac{12.396 \times 10^{-8}}{2d} \left(\frac{1}{\sin \theta_f} - \frac{1}{\sin \theta_i} \right)$$

where d is the innerplanar spacing in cm, and θ_f and θ_i are given by

$$\theta_f = \theta_\beta - \text{Atan}(W/L)$$

$$\theta_i = \theta_\beta + \text{Atan}(W/L)$$

where θ_β is the Bragg angle at which the crystal is set, W is the width of the collimator and L is the length of the collimator. This is for a rectangular collimator and it is independent of the height of the collimator. Also, in Appendix A, it is shown that the angular distribution of photons

Fig. 4.2.2 Relative Output Intensity of X-ray Tube at 20 kV

Fig. 4.2.3 Relative Output Intensity of X-ray Tube at 30 kV

Fig. 4.2.4 Relative Output Intensity of X-ray Tube at 40 kV

Fig. 4.2.5 Relative Output Intensity of X-ray Tube at 50 kV

Fig. 4.2.6 Relative Output Intensity of X-ray Tube at 60 kV

passing through the collimator is given by

$$\eta_n = \frac{\cos \theta \cos \phi}{\cos (\theta + \phi)} - \frac{L \sin |\theta| \cos \phi}{W \cos (\theta + \phi)} - \frac{\sin^2 \theta \cos \phi}{\cos (\theta + \phi)}$$

where θ is measured with reference to the Bragg angle setting of the crystal and ϕ is the angle the target makes with reference to a target which would be perpendicular to the axis of the collimator.

Because the full width of the peaks depends only on the angular divergence of the beam incident onto the crystal a soller slit design was chosen for the collimation of the incident beam. One of the pre-crystal collimator assemblies is shown in Fig. 4.3.1.

The collimator assembly consists of a brass housing which holds the lead collimator inserts and couples them to the x-ray tube. The inserts consist of five parallel slots each 0.508 mm wide and .686 mm high with .254 mm separations. The other collimator housings and various inserts are shown in Fig. 4.3.2. The single slit collimators were used for making measurements which were to be compared to theory and the five slot collimators are useful for increasing the transmitted intensity.

The cross sectional area of the diffracted x-ray beam at the position of the detector is defined by the geometry of the pre-crystal collimator, thickness of crystal, Bragg angle setting, and crystal to detector distance. The analytical form of the maximum dimension of the cross section is derived in Appendix B. Knowing this maximum dimension enables one to collimate the detector and still allow the detector to intercept the whole beam. Collimation of the detector reduces the fluorescent and scattered radiation seen by the detector. Both cylindrical and soller slit collimators were used on the detector.

Fig. 4.3.1 Entrance View of Collimator Assembly

Fig. 4.3.2 Collimator Housings and Collimators

4.4 Crystal Rotation

The crystal was positioned using a Huber model DH4310HL goniometer which was designed to position large specimens. Fig. 4.4.1 shows the goniometer with the 1 mm germanium crystal mounted. All degrees of freedom are motor driven so they can be remotely controlled.

4.5 Detector Rotation and Translation

When the crystal is rotated through an angle θ the detector must be rotated through an angle of 2θ and translated in both x and y directions in order for the detector to remain aligned and at the same crystal-to-detector separation. Expressions for the x and y translations are derived in Appendix C. The apparatus used to accomplish this rotation and translation is shown in Fig. 4.5.1. It consists of x and y translation tables which can be simultaneously rotated about the center axis of the goniometer. A control panel exists both at the apparatus and at a remote location with the multichannel analyzer.

4.6 The Detector

The x-ray detector used was a side-looking intrinsic germanium detector 1 cm in diameter and 7 mm thick. It has a 10 mm Be window and exhibited a resolution of ~ 800 eV for the 59.6 keV x-ray from Am when operated at liquid nitrogen temperature with a 1000 volt detector bias. The efficiency of this detector is shown in Fig. 4.6.1.

4.7 The Electronics System

The counting electronics consisted of a Lawrence Berkeley Laboratory designed optical feedback preamplifier, a Tennelec 205A amplifier, and a Nuclear Data 100 multichannel analyzer. The analyzer has a 1028 channel

Fig. 4.4.1 Goniometer with Germanium Crystals

Fig. 4.5.1 Detector Rotation and Translation Apparatus

Fig. 4.6.1 Intrinsic Germanium Detector Efficiency

memory with teletype and cassette tape output. A schematic giving the relation of the counting electrons to the experimental apparatus is shown in Fig. 4.1.1.

CHAPTER 5

THEORETICAL AND EXPERIMENTAL RESULTS

5.1 Introduction

The results of theoretical calculations and experimental measurements are given in this chapter. Included are calculations of the parameter ϵ for germanium in the energy range from 15 to 100 keV. The response function for a parallel monochromatic input beam is calculated for 20, 30, 40, and 50 keV. Particular emphasis is given to the angular distribution of the input spectra and its effect on the peak shapes and full widths at half maximum of the anomalous transmission spectra. Also a comparison is made between theoretical and experimental peak shapes and transmitted intensities. The computer code used to make the theoretical calculations is described in Appendix D and is available upon request. In summary, the conclusions and recommendations for future work are discussed.

5.2 Epsilon

As is obvious from equation (120) the parameter ϵ plays a very important role in anomalous transmission. From equations (131), (133), and (134) we have $\epsilon = \epsilon_0 e^{-M}$ where

$$M = \frac{6h^2 T}{mk_0^2} \phi(x) + \frac{x}{4} \frac{\sin^2 \theta}{\lambda^2}$$

Using Borrmann and Ludwig's [1969] Debye temperature of 290°K for germanium and a room temperature of 294.2°K

$$\phi(x) = \frac{x}{4} = 1 + \frac{x^2}{36} - \frac{x^2}{3600} = 1.0269$$

Now

$$\frac{\sin^2 \theta}{\lambda^2} = \frac{1}{4d_{hkl}^2} = \frac{1}{4} \frac{(h^2 + k^2 + l^2)}{a^2}$$

$$= \frac{1}{4} \frac{(2^2 + 2^2 + 0^2)}{(5.846)^2} = .05852$$

where a is the lattice constant for germanium.

From this we get $M = .0356$ and a Debye-Waller factor of .9650 for germanium. With this Debye-Waller factor the code can calculate ϵ . The variation of ϵ with energy for the (220) diffraction planes of germanium is given in Table 5.2.1 and is shown in Fig. 5.2.1. The calculations were restricted to energies above the K-edge. Since Wagenfeld calculations are based on a nonrelativistic approximation and hydrogen-like wavefunctions the calculations are restricted to energies not just above the K-edge.

5.3 Crystal Response Function

Even if you had a beam of mono-energetic x-rays which had an angle of incidence θ they would not exit the crystal completely monoenergetic. The distribution of exit photons in this case are shown in Figs. 5.3.1 through 5.3.4 for 20, 30, 40, and 50 keV x-rays for germanium. As can be seen, these distributions are extremely narrow, having full widths at half maximum (FWHM) ranging from 0.02 to 0.1 eV. Thus, they can certainly be considered mono-energetic.

5.4 The Angular Distribution of the Input Spectra

In any real experiment the incident x-rays will have a finite angular divergence and this must be considered when calculating the measured transmission spectra. The angular divergence will be defined by the pre-crystal

Table 5.2.1 The Parameter Epsilon as a Function of Energy

<u>E</u>	<u>ϵ_{Ge}</u>
15	.9635
20	.9635
25	.9636
30	.9638
35	.9639
40	.9640
45	.9641
50	.9642
55	.9643
60	.9643
65	.9644
70	.9644
75	.9645
80	.9645
85	.9646
90	.9646
95	.9646
100	.9646

Fig. 5.2.1 Epsilon as a Function of Energy for Germanium

Fig. 5.3.1 Crystal Response Function for Germanium at 20 KeV

Fig. 5.3.2 Crystal Response Function for Germanium at 30 KeV

Fig. 5.3.3 Crystal Response Function for Germanium at 40 KeV

Fig. 5.3.4 Crystal Response Function for Germanium at 50 KeV

collimator. Also, the angular distribution of incident x-rays will be defined by the collimation. This angular distribution is derived in Appendix A. The angular distribution at 20 keV is shown in Fig. 5.4.1 for a collimator with a 0.143 width to length ratio. Figs. 5.4.2 and 5.4.4 are angular distributions at 30, 40, and 50 keV for a 0.00833 width to length ratio. They have been converted to an energy basis in these figures and thus need some interpretation. If the crystal is set for anomalous transmission at an energy E_0 there will still be incident x-rays with an energy $E \neq E_0$ which can be anomalously transmitted because of the angular divergence of the incident beam about θ_0 . Thus, the distributions shown in Figs. 5.4.2 through 5.4.4 are for those photons available for anomalous transmission.

5.5 Anomalous Transmission Spectra

Measured anomalous transmission spectra for germanium are shown in Figs. 5.5.1 through 5.5.4 for 20, 30, 40, and 50 keV. These spectra have the background subtracted. The small peak shown at approximately 10 keV below the transmission peak is not real, but is due to the escape of K x-rays from the germanium detector. Figs. 5.5.5 through 5.5.8 show a comparison between the theoretical peak shape and the measured peak shape for 20, 30, 40 and 50 keV. In each case the theoretical curve was multiplied by the ratio of the measured intensity to theoretical intensity. Table 5.5.1 gives the measured and calculated FWHM for germanium. The calculated degree of polarization for the transmitted beam is shown in Table 5.5.2.

5.6 Anomalous Transmission Intensity

Anomalous transmission spectra were measured in 5 keV increments from

Fig. 5.4.1 Angular Distribution of Incident Photons when Crystal is set for 20 KeV Anomalous Transmission

Fig. 5.4.2 Angular Distribution of Incident Photons when Crystal is set for 30 KeV Anomalous Transmission

Fig. 5.4.3 Angular Distribution of Incident Photons when Crystal is set for 40 KeV Anomalous Transmission

Fig. 5.4.4 Angular Distribution of Incident Photons when Crystal is set for 50 KeV Anomalous Transmission

Fig. 5.5.1 Anomalous Transmission at 20 KeV

Fig. 5.5.2 Anomalous Transmission at 30 KeV

Fig. 5.5.3 Anomalous Transmission at 40 KeV

Fig. 5.5.4 Anomalous Transmission at 50 KeV

Fig. 5.5 Comparison of Theoretical and Measured Peak Shapes at 20 KeV

Fig. 5.5.6 Comparison of Theoretical and Measured Peak Shapes at 30 KeV

Fig. 5.5.7 Comparison of Theoretical and Measured Peak Shapes at 40 KeV

Fig. 5.5.8 Comparison of Theoretical and Measured Peak Shapes at 50 KeV

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Table 5.5.1

Measured and Calculated FWHM for Germanium

<u>E</u>	<u>B</u>	<u>FWHM</u>	
		<u>Calculated</u>	<u>Measured</u>
15	11.946 ^o	1.07	1.08 $\pm$.03
20	8.931 ^o	1.94	1.99 $\pm$.05
25	7.134 ^o	3.11	3.03 $\pm$.09
30	5.941 ^o	2.59	2.73 $\pm$ 0.15
35	5.090 ^o	3.80	3.6 $\pm$ 0.2
40	4.452 ^o	4.45	4.5 $\pm$ 0.2
45	3.956 ^o	5.73	5.8 $\pm$ 0.2
50	3.560 ^o	6.92	6.7 $\pm$ 0.3
55	3.236 ^o	9.54	9.4 $\pm$ 0.3

Table 5.5.2 Polarization of the Anomalous Transmission

<u>E</u>	<u>$I_{RH}^{\lambda} \quad (I_{RH}^{\lambda} + \parallel_{RH}^{\lambda})$</u>
	<u>Ge</u>
15	1.00
20	0.86
25	0.65
30	0.80
35	0.67
40	0.60
45	0.63
50	0.58
55	0.56

15 to 55 keV. A comparison between these measured and the calculated intensities is given in Table 5.6.1. I_m is the measured intensity, I_e the calculated intensity, R the ratio of measured to calculated intensities, and XFACT is the theoretical ratio of input to output intensity where the input intensity includes only those x-rays with energies that fall within the range of anomalous transmission. A 1.65 mm thick germanium crystal was used for the 15, 20, and 25 keV measurements, a 8.58 mm crystal for the 30, 35, and 40 keV measurements, and a 19.48 mm crystal for the 45, 50, and 55 keV measurements. The $\frac{I_x}{Y}$ ranged from 19.7 to 80.4. A 0.508 mm wide by 35.56 mm long slit collimator was used in the 15 to 25 keV measurements while the others were with a 0.508 mm wide by 60.96 mm long collimator.

The input spectra used for these measurements are shown in Figs. 5.6.1 and 5.6.2, and 5.6.3. All these spectra were measured with the same intrinsic germanium detector that was used to measure the anomalous transmission spectra. They were all measured through a 3 meter evacuated pipe with mylar windows and were corrected by the ratio of intensity at the exit of the collimator to intensity at the exit of the 3 meter evacuated pipe. This intensity ratio was determined using thermoluminescent dosimeters. The 30 kV and 50 kV bremsstrahlung spectra had a 0.0254 mm thick nickel filter preceding the pre-crystal collimator and the 70 kV bremsstrahlung spectrum had a 0.0762 mm nickel filter to reduce the L x-ray intensity from the tungsten target.

5.7 Discussion of Results and Conclusions

Although experimental values [Hunter 1959] and theoretical values [Batterman 1961 and Hountas 1973] of ϵ in Ge have been published for Cu K_α x-rays, no experimental values above the K-edge have been measured. Hountas [1973]

Tables 5.6.1

Anomalous Transmission Intensities

E-KeV	I_m -photons/sec	I_e -photons/sec	R	XFACT	$\frac{x}{y}$
15	1.65	2.62	.63	1.07×10^{-7}	80.4
20	13.0	21.4	.61	1.20×10^{-6}	36.7
25	26.4	45.5	.58	4.86×10^{-6}	19.7
30	2.48	4.63	.54	5.06×10^{-7}	60.8
35	13.3	15.5	.86	1.89×10^{-6}	39.0
40	25.8	27.2	.95	4.38×10^{-6}	26.5
45	14.5	18.3	.79	1.50×10^{-6}	42.8
50	23.5	29.6	.79	2.95×10^{-6}	31.6
55	35.0	36.9	.95	4.87×10^{-6}	24.0

Fig. 5.6.1 Input Bremsstrahlung Spectrum for 30 kV and 5 ma

Fig. 5.6.2 Input Bremsstrahlung Spectrum for 50 kV and 5 ma

Fig. 5.6.3 Input Bremsstrahlung Spectrum for 70 kV and 5 ma

reports a theoretical value of 0.969 for Mo K_{α} x-rays compared to a value of 0.964 reported here. Using the data given by Wagenfeld [1964], a value of 0.9984 for ϵ_0 can be calculated which, when used with a Debye-Waller factor of 0.9650 calculated in Section 3.2, gives $\epsilon = .9635$, which is in agreement with the value calculated by the author as shown in Table 5.2.1. The parameter epsilon has been calculated for germanium for energies above the K-edges up to 100 keV.

There is excellent agreement between the theoretical and measured peak shapes over the entire energy range. As can be seen by the data in Table 5.6.1 the theoretical and measured FWHM are in excellent agreement. We also can see that for a given collimator and crystal thickness the FWHM increase more rapidly than the energy. Thus, in order to achieve the same resolution on the anomalous transmission spectra the angular divergence of the incident beam must be reduced by better collimation. Here we use resolution in the sense of the ratio of the FWHM to the energy.

Since the crystals were not perfect, it is not surprising that the measured intensities are less than what is theoretically possible. This is of little practical consequence since any given crystal would have to be calibrated before it could be used in an experimental setup.

It has been suggested [Howell 1974] that anomalous transmission might be a useful method for providing polarized x-rays for x-ray fluorescence experiments. Although this is true at low energies, at higher energies the degree of polarization diminishes as shown in Table 5.5.2. This is not at all surprising as can be seen from the effective absorption cross section $\mu_{\text{eff}} = \mu(1-P\epsilon)$, where the polarization P equals 1 for the σ -polarization component and equals $\cos(2\theta_B)$ for the π -polarization component of the electric field. At higher energies,

$\cos(2\theta_\beta)$ approaches 1 since θ_β approaches zero and thus the effective absorption cross sections become equal for the two polarization cases.

In summary, anomalous transmission through nearly perfect single crystals of germanium has been measured for energies from 15 to 55 keV. Measured anomalous transmission spectral peak shape and FWHM have been accurately predicted by theory. The measured intensities of anomalous transmission through a fixed single crystal of germanium ranged from 54 to 95 per cent of the theoretical values. It is concluded that anomalous transmission is a viable method of producing a continuously variable source of nearly mono-energetic x-rays.

5.8 Recommendations for Future Work

The most important thing that could be done would be to increase the intensity of anomalous transmission so that it would have broader application. One way to increase the intensity is to cool the crystal. Ludewig [1969] has shown that the intensity for the (220) planes using Mo K_α radiation is increased by a factor of 13 by cooling to 5°K. Another way is to increase the output of the x-ray tube. The production of x-rays is a very inefficient process. At 70 kV and 5 ma this efficiency is about 0.5 percent. The rest of the energy goes into heating up the target. The rating of an x-ray tube is limited by its ability to dissipate this heat. The problem is compounded in most tubes by very small focal spot sizes. For anomalous transmission there is no need for a small focal spot. If a rotating target x-ray tube with a large focal spot size were used with a soller slit collimator, the intensity could be significantly increased. A factor of 1000 would be very easy to achieve and this coupled with cooling of the crystal would provide intensities large enough to do low level

dosimeter studies. If large single crystals of cadmium telluride or other high atomic crystals become available, it would be interesting to extend the energy range to 1000 keV.

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We can see from Fig. A.1 that the angular divergence of the x-rays transmitted by the collimator is given by $\text{Atan}(W/L)$ where W is the width of the collimator and L is the length. Thus, θ can range from θ_f to θ_i where

$$\theta_f = \theta_\beta - \text{Atan}(W/L)$$

$$\theta_i = \theta_\beta + \text{Atan}(W/L)$$

and θ_β is the Bragg angle at which the crystal is set. Using Bragg's law $2d \sin \theta = \lambda$ and $E = 12.396 \times 10^8 / \lambda$ where d is the interplanar separation for the planes of interest and E and λ are expressed in keV and cm respectively, we obtain for the full width of the peak

$$\Delta E = E_f - E_i = \frac{12.396 \times 10^8}{2d} \left(\frac{1}{\sin \theta_f} - \frac{1}{\sin \theta_i} \right)$$

The interatomic distance, d , for a cubic crystal is given by

$$d = \frac{a}{\sqrt{h^2 + k^2 + \ell^2}}$$

where a is the lattice constant and h , k , and ℓ are the Miller indices.

In Fig. A.2 it can be seen that the number of photons which exit the collimator at an angle θ is directly proportional to the length of target subtended by the A and B which are drawn at an angle θ relative to the axis of the collimator. For a target perpendicular to the collimator axis the length η' is given by

$$\eta' = \xi / \cos \theta_c$$

Fig. A.1 Angular Divergence of Collimator

Fig. A.2 Photon Angular Distribution

where

$$\xi = \frac{W}{\cos\theta} - L \sin |\theta| - W \sin^2\theta.$$

If the target makes an angle of ϕ relative to the perpendicular target then the length of target subtended, η , is given by

$$\eta = \xi / \cos(\theta+\phi)$$

Now normalizing η so that $\eta = 1$ when $\theta = 0$ we obtain

$$\eta_n = \frac{\cos\theta \cos\phi}{\cos(\theta+\phi)} - \frac{L \sin |\theta| \cos\phi}{W \cos(\theta+\phi)} - \frac{\sin^2\theta \cos\phi}{\cos(\theta+\phi)}$$

η_n can be used to account for the angular distribution of photons exiting the crystal by imposing the constraining $\eta_n \geq 1$.

APPENDIX B THE EFFECT OF ANGULAR DISPERSION ON THE ANOMALOUS TRANSMITTED BEAM SIZE AT THE DETECTOR

In designing a collimator for a measurement in which the transmission factor for the crystal is to be determined, it is very important that the whole beam be intercepted by the detector. In order to insure this it is necessary to follow the x-ray beam through the collimator and crystal with and without angular dispersion and calculate the maximum dispersion that can result.

Consider an x-ray beam incident on to the crystal at an angle θ_p with respect to the planes of interest which we will assume are perpendicular to the surface of the crystal since this is the case for the experiments performed in this study. Also assume the beam is parallel to the axis of the collimator. This represents the case of zero dispersion. Now consider a beam in which there is an angular dispersion $\theta = A \tan(W/L)$ induced by the width of the collimator and an angular dispersion $\phi = A \tan(H/L)$ induced by the height H of the collimator. In the crystal we see that θ causes a displacement of the beam by $t_o \tan \theta$ and ϕ causes a displacement of $t_o \tan \phi$ where t_o is the thickness of the crystal.

We can calculate the displacements after leaving the crystal by considering the rectangular coordinate system shown in Fig. B.1 where the origin is at the point the beam would have exited the crystal had the displacements in the crystal been ignored. In this coordinate system z is parallel to the direction the beam would go after exiting the crystal if $\phi = 0$ and x and y represent the displacements caused by θ and ϕ not being equal to zero relative to the case where $\theta \neq 0$ but $\phi = 0$. These same displacements relative to the case where θ and ϕ both equal zero are given by $x + h$ and y respectively. The vector $\vec{r}_2$ is the direction

the beam goes with angular dispersions of θ and ϕ .

We have now reduced the problem to where we need to find x and y . From the geometry we see

$$y^2 + r_1^2 + x^2 = r_2^2 \quad (\text{B.1})$$

$$y^2 + r_1^2 + h^2 = D^2 \quad (\text{B.2})$$

$$h = r_1 \tan \theta \quad (\text{B.3})$$

$$y^2 = D^2 \sin^2 \phi \quad (\text{B.4})$$

Substituting the value of D^2 from Eq. B.2 into B.4 we obtain

$$y^2 = \frac{(r_1^2 + h^2) \sin^2 \theta}{(1 - \sin^2 \phi)} = (r_1^2 + h^2) \tan^2 \phi \quad (\text{B.5})$$

Again from the geometry

$$(x + h)^2 = r_2^2 + D^2 - 2r_2 D \cos \theta \quad (\text{B.6})$$

and using $r_2^2 = r_1^2 + x^2 + y^2$

$$2hx + h^2 - r_1^2 - y^2 - D^2 = -2 \sqrt{r_1^2 + x^2 + y^2} D \cos \theta$$

Squaring both sides and collecting coefficients of like powers of x we obtain

$$ax^2 + bx + c = 0 \quad (\text{B.7})$$

where

$$a = 4h^2 + 4D^2 \cos^2 \theta.$$

$$b = 4h^3 - 4hr_1^2 - 4hy^2 - 4hD^2$$

$$c = h^4 - 2h^2r_1^2 - 2h^2y^2 - 2h^2D^2 + r_1^4 + 2r_1^2y^2 + 2r_1^2D^2 \\ + y^4 + 2y^2D^2 + D^4 + 4r_1^2D^2\cos^2\theta + 4y^2D^2\cos^2\theta$$

Combining these displacements we find the total displacement due to the angular dispersion of the incident photons is given by

$$z = \sqrt{(x + t_o \tan \theta)^2 + (y + t_o \tan \phi)^2}$$

where

$$x = \frac{-b + \sqrt{b^2 - 4ac}}{2a} \quad (B.7)$$

$$y = r_1 \sec^2 \theta \tan^2 \phi \quad (B.5)$$

Thus for given angular dispersions θ and ϕ and crystal-to-crystal detector distance r_1 we can calculate the maximum dimension of a cross section of the beam.

Fig. B.1 Geometry of Angular Divergence Displacements

When the crystal is rotated through an angle θ to a new position so as to change the energy of the anomalous transmission the detector must be rotated through 2θ so as to be at the correct angle to see the diffracted photons unless the forward diffracted beam is to be measured. Since the crystal has a finite thickness the detector must also be translated so as to remain the same distance from the crystal and to be in line with the emerging beam. This can be seen in Fig. C.1 where t_o is the crystal thickness, x_c is the perpendicular distance from the center of the axis of rotation of the goniometer to the entrance face of the crystal, θ is the angle of incidence that the incident x-ray beam makes with the diffracting planes, and x, y are the coordinates of the center of the beryllium window on the germanium detector. The origin of this x-y coordinate system rotates with the detector in such a manner that the x-axis always remains parallel to the axis of the germanium detector.

The values of x and y are how far the detector must be translated after rotation relative to its position when θ is zero. The changes are given by

$$\begin{aligned}\Delta y &= \left(\frac{x_o - t_o}{\cos \theta} + \frac{t_o}{2 \cos \theta} \right) \sin 2\theta \\ &= \left(x_c - \frac{t_o}{2} \right) \frac{\sin 2\theta}{\cos \theta} \\ \Delta x &= \left(\frac{(x_c - t_o)}{\cos \theta} + \frac{t_o}{2 \cos \theta} \right) \cos 2\theta - \frac{t_o}{2 \cos \theta} \\ &= x_c - t_o - \left(x_c - \frac{t_o}{2} \right) \frac{\cos 2\theta}{\cos \theta} + \frac{t_o}{2 \cos \theta}\end{aligned}$$

Thus for a given crystal thickness and angle of incidence we can readily calculate the translations required.

Fig. C.1 Rotation and Translation of Detector

APPENDIX D COMPUTER CODE FOR CALCULATING ANOMALOUS
TRANSMISSION SPECTRA AND INTENSITIES

So as to provide a basis for comparison with experiment a computer code was written to calculate the anomalous transmission parameters of interest. The code starts with an input spectra which is specified by the operator of the code. It follows the photons through the pre-crystal collimator, through the anomalously transmitting crystal and into the detector. Effects of collimation, air attenuation, the crystal, and detector efficiency are all included. Included in the output of the code are the spectral shape as measured by the detector, the full width of the peak and the predicted count rate.

The code consists of a main program plus 20 subroutines.

The first thing the code does is to read in information from a file IN. This information describes the input spectrum, the pre-crystal collimators, the crystal, and detector. Table D.1 briefly describes each of the input parameters. The subroutines PART and EPSILON are used to calculate and plot the parameter as a function of energy. The response of the crystal to a monoenergetic source of x-rays incident to an angle θ_p is given by equation (119). The subroutine RESPONSE with the assistance of RHINT calculates, plots and writes out the anomalously transmitted intensity as a function of energy into an output file OP.

Most of the variables needed for the calculation of anomalous transmission are dependent on energy. INT1 calculates these variables and writes them into OP. The integration over all angles of incidence as defined by the collimators is carried out in INT2. The resulting output spectrum, the ratio of output to input intensity, and the output intensity are written into OP.

The various output parameters are listed in Table D.2 along with a brief description. Other routines include CDTEX and GEXSECT for calculation of cadmium

telluride and germanium cross sections; SIMP for numerical integration;
WRITE for writing and output data into OP. All cross sections were taken
from McMaster [1969].

Table D.1 Input Parameters for the Code CRY

<u>Parameter</u>	<u>Description</u>
IFLAG1	If 1 the code plots the parameter ϵ as a function of energy. If 0 the code does not plot ϵ .
IFLAG2	If 1 the crystal is germanium. If 2 the crystal is cadmium telluride.
XINTOCON	Normalization constant for input spectrum.
XL	The length of the pre-crystal collimator - cm.
W	The width of collimator - cm.
XC	The perpendicular distance from the entrance face of the crystal to the axis of rotation of the goniometer - cm.
XORDER	The diffraction order. That is, n in Bragg's equation $2d\sin\theta_{\beta} = n\lambda$.
D	The perpendicular distance between the diffraction planes being considered - cm.
VOL	The volume of a unit cell - cm^3 .
ZGE	The atomic number of germanium.
ZCD	The atomic number of cadmium.
ZTE	The atomic number of telluride.
AH	The geometric term relating the structure factor to the atomic scattering factor.
FHGE	Structure factor for germanium.
FHCDTE	Structure factor for cadmium telluride.

<u>Parameter</u>	<u>Description</u>
A0,A1,A2,A3	Constants used for the polynomial description of the input spectrum where the number of photons per keV per sec is given by $I = A0 + A1 \times E + A2 \times E^2 + A3 \times E^3$.
SM	The parameter m in the Debye-Waller factor e^{-m} .
XXTAL	The thickness of the crystal - cm.
E(1),E(2)	The energies just below and above the K-edge of the crystal keV.
E(3)	The energy at which to start the calculation of the parameter ϵ - keV.
XI	the x-ray tube current - amps.
VO	The x-ray tube operating voltage - volts.
ZT	The atomic number of the x-ray tube target.
LMAX	The number of points in the calculated output spectrum.

Table D.2 Output Parameters for the Code CRY

<u>Parameter</u>	<u>Description</u>
EO	Energy at which the crystal is set for anomalous transmission keV.
XLAMB	The x-ray wavelength corresponding to EO - cm.
GAMMA	$r_e \lambda^2 / (\pi V)$ where r_e is the electron radius - cm; r is the wavelength XLAMB - cm; V is the volume of a unit cell.
TAB	The total absorption coefficient without coherent scattering for the crystal of interest - cm^{-1} .
GAMMAO	$\cos(\theta_\beta)$
RHL	The numerical integration evaluation of the perpendicular component of equation (125).
EPS	The value of ϵ at the energy EO.
TQ	The quadrapole term of the total absorption coefficient - cm^{-1} .
TTOT	The total absorption coefficient as calculated by EPSILON - cm^{-1} .
EPSO	The value EPS would have if the Debye-Waller factor were equal to 1.
RHL125PER	The perpendicular polarized component of equation (125) where the approximate equation (120) has been used for the integrated intensity R_H^Y .
RHL125PAR	The parallel polarized component of equation (125) where the approximate equation (120) has been used for the integrated intensity R_H^Y .

<u>Parameter</u>	<u>Description</u>
RHINT	$R_H^\lambda / \Delta\lambda$
TABEAPA	TAB(1 - EPS)
ETABPER	The sum of the effective photoelectric, compton, and thermal diffuse scattering cross sections for the perpendicular polarized component - cm^{-1} .
ETABPAR	The sum of the effective photoelectric compton, and thermal diffuse scattering cross sections for the parallel polarized component - cm^{-1} .
EPEX	The effective photoelectric absorption coefficient for the perpendicular component - cm^{-1} .
EINCX	The effective incoherent scattering coefficient for the perpendicular component - cm^{-1} .
ETDS	The effective thermal diffuse scattering coefficient - cm^{-1} .
EINT	The lower limit of integration - keV.
EFIN	The upper limit of integration - keV.
AIRATTEN	The attenuation due to air.
EFFICIENCY	The detector efficiency.
ETAA	The collimator shaping function.
SPECT	Photons/keV/sec anomalously transmitted through the crystal with an energy EO.
SPECTD	The calculated photons/keV/sec with an energy EO which should be recorded by the detector.
XTALFACT	The ratio of transmitted to incident photons with energy between EINT and EFIN.

BIBLIOGRAPHY

1. A. M. Afanas'ev and Y. Kagan, "The Role of Lattice Vibrations in Dynamical Theory of X-rays", Acta Cryst. A24, 163 (1967).
2. S. Balter, R. Feldman, and B. Post, "Multiple Borrmann Diffraction", Phys. Rev. Letters 27, 307 (1971).
3. B. W. Batterman, "Imaginary Part of X-ray Scattering Factor for Germanium", J. Appl. Phys 32, 998 (1961).
4. B. W. Batterman, "Effect of Thermal Vibrations on Diffraction from Perfect Crystals. I. Anomalous Transmission", Phys. Rev. 126, 1461 (1962).
5. B. W. Batterman and D. R. Chipman, "Effect of Thermal Vibrations on Diffraction from Perfect Crystals II. The Bragg Case of Reflection", Phys. Rev. 127, 690 (1962).
6. B. W. Batterman, "Redetermination of the Temperature Dependence of X-rays Anomalous Transmitted Through Germanium", Phys. Rev. 134, A1354 (1964).
7. B. W. Batterman and H. Cole, "Dynamic Diffraction of X-Rays by Perfect Crystals", Rev. Mod. Phys. 36, 681 (1964).
8. V. G. Borrmann, "Über Extinktionsdiagramme von Quarz", Z. Physik 42, 157 (1941).
9. G. Borrmann, "Die Absorption von Röntgenstrahlen im Fall der Interferenz", Z. Physik 127, 297 (1950).
10. V. G. Borrmann and W. Gerisch, "Ein Röntgenstrahl-Absorptionsdiagramm von Kalspat und Seine Indizierung", Z. Kristallogr. 106, 99 (1954).
11. V. G. Borrmann, "Der kleinste Absorptionskoeffizient Interferenzender Röntgenstrahlung", Z. Kristallogr. 106, 109 (1954).
12. V. G. Borrmann and W. Hartwig, "Die Absorption der Röntgenstrahlen im Dreistrahlfall der Interferenz", Z. Kristallogr. 121, 401 (1965).

13. B. Buras and L. Gerward, "Relations Between Integrated Intensities in Crystal Diffraction Methods of X-rays and Neutrons", Acta Cryst. A31, 372 (1975).
14. J. R. Cameron and J. Sorenson, Science, 142, 230 (1963).
15. H. N. Campbell, "X-ray Absorption in a Crystal Set at the Bragg Angle, J. Appl. Phys. 22, 1130 (1951).
16. J. L. Chartier, "Free-air Ionization Chamber for the Energy Range 12 keV to 100 keV", Nucl. Instr. Meth. 128, 251 (1975).
17. G. L. Clark, Applied X-rays, McGraw-Hill, New York (1955).
18. G. L. Clark, The Encyclopedia of X-rays and Gamma Rays, Reinhold Publishing Company, New York (1963).
19. M. J. Cohen and A. J. Gilson, Progress in Development of Methods in Bone Densitometry, NASA Sp-64, 103 (1966).
20. E. U. Condon and H. Odishaw, Handbook of Physics, McGraw-Hill, New York, (1958).
21. H. Cole, F. W. Chambers, and C. G. Wood, "X-ray Polarizer", J. Appl. Phys. 32, 1942 (1974).
22. D. T. Cromer, "Anomalous Dispersion Corrections Computed from Self-Consistent Field Relativistic Dirac-Slater Wave Functions", Acta Cryst. 18, 17 (1965).
23. D. T. Cromer and J. T. Waber, "Scattering Factors Computed from Relativistic Dirac-Slater Wave Functions", Acta Cryst. 18, 104 (1965).
24. C. G. Darwin, Phil Mag. 27, 315 (1914).
25. L. I. Datsenko and A. N. Gureev, "Thermal Diffuse Energy Loss in the Borrmann Effect in Germanium", Sov. Phys. Sol. State, 17, 2049 (1975).
26. J. J. DeMarco and R. J. Weiss, "Absolute X-ray Scattering Factors of Silicon and Germanium", Phys. Rev. 137, 1869 (1965).

27. P. A. Doyle and P. S. Turner, "Relativistic Hartree-Fock X-ray and Electron Scattering Factors", Acta Cryst. A24, 390 (1968).
28. T. G. Dzubay, B. V. Jarvett, and J. M. Jaklevic, "Background Reduction in X-ray Fluorescence Spectra Using Polarization", Nucl. Instr. Meth. 115, 297 (1974).
29. P. J. Ebert, J. L. Gaines and G. R. Leipelt, "Production of Monoenergetic X-ray Sources of Known Absolute Intensity", Nucl. Instr. Meth. 99, 29 (1972).
30. O. N. Efimov, "Contributions of Thermal Vibrations to the Anomalous Transmission of X-rays", Phys. Stat. Sol. 22, 297 (1967).
31. P. P. Ewald, Fifty Years of X-ray Diffraction, A. Oosthoek Publishers, Utrecht (1962).
32. P. P. Ewald, "Crystal Optics for Visible Light and X-rays", Rev. Mod. Phys. 37, 46 (1965).
33. A. J. Freeman, "X-ray Incoherent Scattering Functions", Phys. Rev. 113, 169 (1959).
34. A. J. Freeman, "X-ray Incoherent Scattering Functions", Acta Cryst. 12, 929 (1959).
35. A. J. Freeman, "A Study of the Compton Scattering of X-rays", Acta Cryst. 13, 190 (1960).
36. A. J. Freeman and R. E. Watson, "Compton Incoherent Scattering Functions for Ions of the First Transition Series", Acta Cryst. 15, 682 (1962).
37. J. L. Gaines, "A Generator for Producing Monoenergetic, High Intensity, Soft X-rays", Nucl. Instr. Meth. 102, 7 (1972).
38. P. Gay, P. B. Hirsch and J. N. Kellar, "Asymmetric X-ray Reflections from Abraded Crystals", Acta Cryst. 5, 7 (1952).
39. C. Ghezzi, A. Merlini, and S. Pace, "Effect of Temperature on the Anomalous Transmission of X-rays in Copper and Zinc", Phys. Rev. B 4, 1833 (1971).

40. M. D. Giardina and A. Merlini, "Effect of Compton Scattering on the Borrmann Effect of X-rays in Silicon Crystals", Z. Naturforsch. A28, 1360 (1973).
41. H. P. Hanson, F. Herman, J. D. Lea, and S. Skillman, "HFS Atomic Scattering Factors", Acta Cryst. 17, 1040 (1964).
42. P. B. Hirsch and G. N. Ramachandran, "Intensity of X-ray Reflexion from Perfect and Mosaic Absorbing Crystals", Acta Cryst. 3, 187 (1950).
43. P. B. Hirsch, "The Reflexion and Transmission of X-rays in Perfect Absorbing Crystals", Acta Cryst. 5, 176 (1952).
44. P. H. Howell and W. L. Pickles, "Possible Sources of Polarized X-rays for Fluorescence Spectra with Reduced Background", Nucl. Instr. and Meth. 120, 187 (1974).
45. L. P. Hunter, "X-ray Measurement of Microstrain in Germanium Single Crystals", J. Appl. Phys. 30, 874 (1959).
46. L. P. Hunter, "Direct Measurement of the Angular Dependence of the Imaginary Part of the Atomic Scattering Factor of Germanium", IBM Journal 3, 106 (1959).
47. A. Hountas, S. E. Filippakis, C. Papathanassopoulos, and T. Taskalakos, "Experimental and Theoretical Determination of the Imaginary Parts of the Atomic Scattering Factors by Means of the Borrmann Effect in Nearly Perfect Si Crystals", J. Phys. C 6, 1693 (1973).
48. R. H. Howell, W. L. Pickles, J. L. Cate, X-ray Fluorescence Experiments with Polarized X-rays, Lawrence Livermore Laboratory, Rept. UCRL-75623 (1974).
49. J. D. Jackson, Classical Electrodynamics, John Wiley and Sons, Inc. New York, (1962).
50. R. W. James, The Optical Principles of the Diffraction of X-rays, G. Bell and Sons, LTD. (1962).

51. N. Kato, "Dynamic Theory of Electron Diffraction for a Finite Polyhedral Crystal", J. Phys. Soc. Jap. 7, 397 (1952).
52. N. Kato, "Integrated Intensities of the Diffracted and Transmitted X-rays due to Ideally Perfect Crystal", J. Phys. Soc. Jap. 10, 46 (1954).
53. H. P. Klug and L. E. Alexander, X-ray Diffraction Procedures, John Wiley and Sons, Inc., New York, (1954).
54. M. V. Laue, Ergeb. Exakt. Naturw. 10, 133 (1931).
55. M. V. Laue, "Die Absorption der Röntgenstrahlen in Kristallen im Interferenzfall", Acta Cryst. 2, 106 (1949).
56. M. V. Laue, "Die Energiestromung bei Röntgenstrahl-Interferenzen in Kristallen", Acta Cryst. 5, 619 (1952).
57. D. Liberman, J. T. Waber, and D. T. Cromer, "Self-Consistent-Field Dirac-Slater Wave Functions for Atoms and Ions", Phys. Rev. 137, A27 (1965).
58. D. Ling and H. Wagenfeld, "Anomalous Transmission of X-rays in Perfect Single Germanium Crystals at Liquid Nitrogen Temperature", Phys. Letters 15, 8 (1965).
59. W. H. McMaster, "Compilation of X-ray Cross Sections", Lawrence Livermore Laboratory, Rept. UCRL-50174 (1969).
60. Y. H. Ohtsuki, "Temperature Dependence of X-ray Absorption by Crystals", J. Phys. Soc. Jap. 19, 2285 (1964).
61. B. Okkerse, "Anomalous Transmission of X-rays in Germanium", Phillips Res. Repts. 17, 464 (1962).
62. B. Okkerse and P. Penning, "Anomalous Transmission of X-rays in Germanium", Phillips Res. Repts. 18, 82 (1963).
63. P. Penning and D. Polder, "Dynamic Theory for Simultaneous X-ray Diffraction. Part I. Theorems Concerning the n-Beam Case", Phillips Res. Repts. 23, 1, (1968).

64. P. Penning, "Dynamic Theory for Simultaneous X-ray Diffraction. Part II. Application to the Three-Beam Case", Phillips Res. Repts. 23, 12 (1968).
65. E. Persson and O. N. Efimov, "The Anomalous Transmission of X-rays", Phys. Stat. Sol. 3, 757 (1970).
66. W. L. Pickles and J. H. Elliott, Lawrence Livermore Laboratory Rept. UCRL-51422 (1973).
67. H. Sana and Y. H. Ohtsuki, "Normal and Abnormal Absorption Coefficients of X-rays", J. Phys. Soc. Jap. 27, 1254 (1969).
68. R. C. Schroeder, On the Anomalous Diffraction of X-rays Through Perfect Crystals, Lawrence Livermore Laboratory, Rept. UCRL-50439 (1968).
69. J. C. Slater, "Interaction of Waves in Crystals", Rev. Mod. Phys. 30, 197 (1958).
70. J. D. Stephenson, "A Note Concerning the Temperature Dependence of the Linear Absorption Coefficient of X-rays in a Perfect Single Crystal of Silicon", Phys. Status Solid: A, 13, K71 (1972).
71. B. K. Tanner, "Dislocation-free Silver Single Crystals Grown by the Czochralski Method", Z. Naturforsch 28A, 676 (1973).
72. J. F. Vetelino, S. P. Gaur, and S. S. Mitra, "Debye-Waller Factor for Zinc-Blende Type Crystals", Phys. Rev. B 5, 2360 (1972).
73. H. Wagenfeld, "Comments on Anomalous Absorption of X-rays", J. Appl. Phys. 33, 2907 (1962).
74. H. Wagenfeld, "Normal and Anomalous Photoelectric Absorption of X-rays in Crystals", Phys. Rev. 144, 216 (1966).
75. B. E. Warren, X-ray Diffraction, Addison-Wesley Publishing Co., 1969.
76. R. E. Watson and A. J. Freeman, "Hartree-Fock Wave Functions for the 4-p-shell Atoms", Phys. Rev. 123, 521 (1961).

77. R. J. Weiss, X-ray Determination of Electron Distributions, John Wiley & Sons, Inc., 1966.
78. W. H. Zachariasen, "Theory of X-ray Diffraction in Crystals", Dover Publications, Inc., New York (1945).
79. W. H. Zachariasen, "On the Anomalous Transparency of Thick Crystals to X-rays", Proc. Nat. Acad. Sci. (USA) 38, 378 (1952).
80. C. A. Ziegler, Applications of Low Energy X- and Gamma Rays, Gordon and Breach Science Publishers, New York, 1971.