A Comparison of Diffuse Scattering by Defects Measured in Anomalous Transmission and Near Bragg Reflections

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Introduction

One of the consequences of the discovery of the anomalously (Borrmann) transmitted X-rays through nearly perfect crystals has been a renewed interest in the use of X-ray scattering for the investigation of defects in crystals. The decrease in anomalous transmission resulting from defects has been studied for impurities in silicon, and for interstitials in copper. The decrease in transmission results from the lattice strains introduced by the defects and the theory for describing this effect has been developed by Dedereichs, and we have shown the theory to be phenomenologically correct for neutron irradiated copper. The decrease in anomalous transmission is described in terms of an effective absorption coefficient \( \mu_{\text{eff}} \) which is the sum of two components, \( \mu_{\text{eff}} = \mu_{\text{abs}} + \mu_{\text{scat}} \), due to enhanced photoelectric absorption.

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\[ k + H + q \text{ and } k + H + q' \text{ beams may not. In the terminology of the local reciprocal lattice, this statement implies that the two-beams, } k \text{ and } k + H(r), \] are considered to be under dynamical diffraction (see Paper II).

Here we apply the two-wave approximation with the understanding described above. The canonical transformation of interest then functions only to diagonalize the matrix \( K \) with respect to \( q \) and \( q' \) defined in Section 2, not with respect to \( I \) and \( J \).

From this point on, the subscripts \( q \) on \( W \) in (3.2) really become identical to the \( q \)'s defined in Section 2. Equation (3.3) then gives a set of equations for the \( O \) and \( H \) beams just as for a perfect crystal.

The Hamilton-Jacobi Eq. (3.5 b) now takes the form of the well-known dispersion equation for the two-wave case. Using the Hamiltonian defined by this equation, we can derive Hamilton's canonical equation in the two-wave approximation. This equation gives the "relativistic" equation for the rays as Kato and Kambe derived previously.

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tion at the atoms which are distorted from lattice positions, and $\mu_{DS}$ due to the waves which are diffusely scattered from the dynamical wave and hence absorbed in the crystal. Also, there is diffuse scattering near reflections in the Bragg geometry which is caused by the defects. From the theories this diffuse scattering and $\mu_{DS}$ of anomalous transmission are related. For this paper we have measured both anomalous transmission and diffuse scattering on copper crystals which had been neutron irradiated and compared these values through the theory.

**Theory**

In the theoretical treatment of the effect of clustered defects on anomalous transmission given by Dederichs the decrease in transmitted intensity

$$\mu_{DS} = \frac{r_0^2}{V_c} \left( 1 - M \frac{h}{\lambda} \right)^2 C_L \left( \frac{b \pi R^2}{V_c} \right)^2 \ln \left( \frac{q_c}{q_L} \right)$$

where $r_0$ is the classical electron radius, $V_c$ is an atomic volume, $e^{-M}$ is the atomic scattering factor with Debye-Waller factor $e^{-M}$, $k = 2 \pi/\lambda$ where $\lambda$ is the X-ray wavelength, $q_c = 4 \pi r_0 e^{-M}/V_c h$ is a reciprocal extinction length of the reflection, $q_L = 1/R$ and $\nu$ is Poisson’s ratio. $\mu_{PE}$ is given approximately by

$$\mu_{PE} = \mu_0 e^{-M} L = \mu_0 e^{-M} C_L R^3 (h b)^{1/2} V_c$$

where $\mu_0$ is the linear photoelectric absorption coefficient and $L$ is the static Debye-Waller factor associated with the strain fields of the defects.

The Huang diffuse scattering near Bragg reflections (also thin crystal Laue reflections) measured is represented in terms of an effective absorption coefficient $\mu^*$ such that the ratio of the integrated Borrmann intensities for crystals containing defects $R_{def}$ to that for a perfect crystal $R_{per}$ is given by

$$R_{def}/R_{per} = e^{-\mu^* t}$$

where $t$ is the effective thickness of the crystal. $\mu^* = \mu_{PE} + \mu_{DS}$ has two components, $\mu_{PE}$ related to the absorption caused by atoms displaced by the strain fields around defects and $\mu_{DS}$ arising from X-rays scattered out of the anomalously transmitted beam by Huang diffuse scattering generated by the strain fields.

Considering a concentration of dislocation loops $C_L$ (loops/atom) with radius $R$ and Burgers vector $b$, $\mu_{DS}$ for a reflection of reciprocal lattice vector $h$ is given by

$$\mu_{DS} = \frac{r_0^2}{V_c} \left( 1 - M \frac{h}{\lambda} \right)^2 C_L \left( \frac{b \pi R^2}{V_c} \right)^2 \ln \left( \frac{q_c}{q_L} \right)$$

with a wide open detector, as shown in Fig. 1, is closely related to that associated with $\mu_{DS}$ in the Borrmann case. Dederichs has considered this "integral" diffuse scattering which is characterized by a minimum $q$-vector $q_0 = h/(A \theta) \cos \Theta_B$ when the sample is set at an angle $A \theta$ off the Bragg angle $\Theta_B$. The diffuse intensity is calculated by considering contributions from all $q$’s having components $q_0$ perpendicular to the Ewald sphere (considered as a tangent plane here) and integrating over components $q'$ in the tangent plane such that $q = (q_0 + q')^{1/2}$. This intensity is asymmetrical; however, the symmetrical part of this intensity

$$I^* (q_0) = \frac{1}{2} \left( I (q_0) + I (-q_0) \right)$$

**Fig. 1.** Schematic drawing of the scattering geometry for “integral” diffuse scattering using a Bragg monochromator showing the incident beam $k_0$, and the scattered X-rays $k$. 

has been shown to be of the form

\[ I^*(q_0) \sim C_L (b \pi R^2/V_c)^2 \ln (q_L/q_0) \]  

(5) for \( q_0 \ll q_L \). The above expression and the expression for \( \mu_{DS} \) were obtained neglecting the contribution from \( q \geq q_L = 1/R \) which characterizes the end of the Huang scattering which has the well-known \( q^{-2} \) scattering law.

For \( q_0 \gg q_L \) and in the anomalous transmission case the scattering from \( q \geq q_L \) is not too significant; however, for \( q_0 \geq q_L \) this is not the case so we have considered the contribution from \( q \geq q_L \) by approximating the scattering with a \( q^{-4} \) dependence for \( q \geq q_L \) and joining to the \( q^{-2} \) dependence at \( q = q_L \). The integral over \( q' \) in the tangent plane now has the form

\[
\int \frac{q' \, dq'}{q_0^2 + q'^2} \left( 1 + \frac{q'^2}{q_0^2 + q'^2} \right)^2 \quad (q_0 \leq q_L); \quad \int \frac{q_L^2 \, q' \, dq'}{(q_0^2 + q'^2)^2} \quad (q_0 \geq q_L). \]

(6)

The upper limit \( q_M \) is rather indefinite, but since it is much larger than \( q_0 \) it has been considered infinite in evaluating the integrals. These integrations lead to the result that \( \ln(q_L/q_c) \) in Eq. (2) is replaced by \( \ln(q_L/e) \) and \( \ln(q_L/q_0) \) in Eq. (5) is replaced by \( \ln(q_L/e) \) if \( q_0 \leq q_L \) and \( q_L^2/2q_0^2 \) if \( q_0 > q_L \).

In analogy with Eq. (2) and as suggested by Dederichs it is possible to include both Eqs. (2) and (5) in a single expression if we write the integral diffuse scattering cross section in the form of a linear scattering cross section \( \mu_{DS}(q_0) \). That is

\[
\mu_{DS}(q_0) = \frac{I_0}{V_c} \left( \frac{b \pi R^2}{V_c} \right)^2 F(q_L/q_0) = \ln(q_L/e) \quad (q_0 \leq q_L),
\]

\[
F(q_L/q_0) = \frac{8\pi}{15} + \frac{\tau}{15} \left( -1 + 6 \tau + 3 \tau^2 \right) \cos^2 \Theta_B \quad (q_0 \geq q_L).
\]

(7)

with

\[
F(q_L/q_0) = \ln(q_L^2/q_0^2) \quad (q_0 \leq q_L),
\]

(8)

therefore leads to a straight line with intercept at \( q_0 = q_L e^{1/2} \) and for large \( q_0 \) a plot of \( \ln[I^*(q_0)] \) vs \( \ln(q_0) \) has a slope of \(-2\).

In this form \( \mu_{DS} \) obtained from anomalous transmission measurements can be directly compared with the measured diffuse scattering in Bragg or Laue cases given by

\[
\frac{I^*(q_0)}{I_0} = \mu_{DS}(q_0) \quad \text{symmetrical Bragg}
\]

\[
\frac{I^*(q_0)}{I_0} = \mu_{DS}(q_0) t e^{-\mu t} \quad \text{symmetrical Laue}
\]

(9)

where \( I_0 \) is the incident beam intensity and \( t = t_0/\cos \Theta_B \) (\( t_0 \) = perpendicular thickness of the sample).

When a distribution of sizes is to be considered rather than defects all of the same size, the interpretation of these formulae becomes somewhat more difficult and their application to experimental measurements must be considered in conjunction with the size distribution.

In particular for a distribution \( C_L(R_i) \), \( \mu_{DS}(q_0) \) has the form

\[
\mu_{DS}(q_0) \sim \sum_i C_L(R_i) R_i^4 \ln(e^{1/2}/q_0 R_i)
\]

(10)

Fig. 2. Integral diffuse scattering vs \( \ln(D\Theta) \) for a dose of \( 10.8 \times 10^{10} \text{n/cm}^2 \). The coarse dashed line drawn through the experimental points defines the intercept and the solid line is the computed curve for an exponential distribution with \( R_0 = 18.5 \text{Å} \) \( (q_0 = 9.88 \times 10^{-4} \text{Å}^{-1/\text{div}}) \).
for \( q_0 \) much less than those \( q_L \) associated with the largest \( R_i \), and

\[
\mu_{DS}(q_0) \sim \sum C_L(R_i) R_i^2/2 q_0^2
\]

(11)

for large \( q_0 \) (\( q_L \) has been replaced by \( 1/R_i \)). \( \mu_{DS}(q_0) \) for small \( q_0 \) weights the sizes similar to a fourth moment while for large \( q_0 \), \( \mu_{DS}(q_0) \) represents a second moment sampling of the size distribution.

For an exponential distribution \( C_L(R) = C_L(0) e^{-R/R_0} \) truncated at \( R = R_0 \), Eq. (10) can be written in integral form as

\[
\mu_{DS}(q_0) \sim (C_L(0)/R_0) \int_0^{R_0} e^{-R/R_0} R^4 \ln(e^{1/q_0 R}) dR.
\]

(12)

This expression is apparently not intergrable in closed form for finite \( R_0 \); however, for \( 1/q_0 \gg 5 R_0 \), it appears that \( R_0 \) can be taken as infinity without introducing serious error. The integral can then be performed \(^{10}\) giving

\[
\mu_{DS}(q_0) \sim 24 C_L(0) R_0^4 \left( \ln(e^{1/q_0 R_0}) + C_E - \frac{\pi^2}{6} \right).
\]

(13)

\( C_E = 0.5772 \) is Euler's constant. This can be written approximately as

\[
\mu_{DS}(q_0) \sim 24 C_L(0) R_0^4 \ln(1/q_0 R_0 e) \quad q_0 \ll 1/5 R_0
\]

(14)

which leads to an intercept at \( q_0 \approx 1/R_0 e \) in a plot of \( I^*(q_0) \) versus \( \ln(q_0) \).

### Results

The results of the anomalous transmission measurements on the 111 reflection of the irradiated crystals for CuKa and MoKa X-rays are tabulated in the form of \( \mu^* \) values in Table 1. The separation of \( \mu^* \) into \( \mu_{PE} \) and \( \mu_{DS} \) was carried out \(^6\) by satisfying the wavelength dependence of the two components as given in Eq. (2) and Eq. (3), with the constraint that the sum be the measured \( \mu^* \).

![Fig. 3. Integral diffuse scattering vs \( \Delta \theta \) \((q_0=9.88 \times 10^{-4} \text{ Å}^{-1} /\text{div}) \) for a neutron dose of \( 10.8 \times 10^{18} \text{ n/cm}^2 \) at 316 K. The points are experimental. The solid line was computed for an exponential distribution with \( R_0=18.5 \text{ Å} \) and the dashed line for defects of uniform size \( R=84 \text{ Å} \).](image-url)
The integral diffuse scattering measurements, made near the 111 Bragg reflection using CuKα radiation, are presented in terms of the ratio of the diffuse scattering intensity to the incident beam intensity in Figs. 2, 3, 4 and 5. The individual points are the measured values as obtained using a Borrmann monochromator; measurements made with a Bragg monochromator were about 20% smaller, as expected from polarization corrections, but the shapes of the curves were equivalent. Figure 2 contains a plot of the integral scattering intensity versus \( \ln q_0 \) for a neutron dose of \( 10.8 \times 10^{18} \) n/cm\(^2\), irradiation temperature 316 °K. Similar plots were made for the other doses, yielding measurements of the intercept in each case. This intercept is inversely related to the characteristic size of the defect clusters. Thus, it is possible to identify qualitatively a trend toward increasing defect cluster sizes for increasing irradiation doses and irradiation temperatures from the intercept values listed in Table 1.

<table>
<thead>
<tr>
<th>Dose (n/cm(^2))</th>
<th>( \lambda )</th>
<th>( \mu^* )</th>
<th>( \mu_{DS} )</th>
<th>( \mu_{PE} )</th>
<th>Intercept</th>
<th>( R_0 )</th>
</tr>
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<tr>
<td>( 1.3 \times 10^{18} )</td>
<td>CuKα</td>
<td>12.9 cm(^{-1})</td>
<td>3.6 cm(^{-1})</td>
<td>9.3 cm(^{-1})</td>
<td>( 6.9 \times 10^{-2} ) Å(^{-1})</td>
<td>5.3 Å</td>
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<td>1.0</td>
<td>MoKα</td>
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<td>1.0</td>
<td>9.1</td>
<td>2.50</td>
<td>14.5</td>
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<tr>
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<td>2.9</td>
<td>3.3</td>
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<tr>
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<td>25.6</td>
<td>23.4</td>
<td>1.97</td>
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<td>MoKα</td>
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<td>7.0</td>
<td>23.0</td>
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</table>
The values of $\mu_{DS}$ from anomalous transmission can be compared with the intensities measured in the Bragg geometry by utilizing the $q_0$ dependence of the diffuse scattering given in Eq. (7):

$$I^i(q_0) = \frac{\mu_{DS} F(q_L/q_0)}{I^0} \frac{\mu_0 F(q_L/q_c)}{2\mu_0 F(q_L/q_c)}. \quad (15)$$

**Thus the integral diffuse scattering is related to the $\mu_{DS}$ from the anomalous transmission multiplied by the “form factor”** $(F(q_L/q_0))/(F(q_L/q_c))$.

The dashed curve in Fig. 3 was calculated in this manner for an intercept of $1.97 \times 10^{-2}$ Å$^{-1}$ obtained from Fig. 2 and $\mu_{DS} = 39$ cm$^{-1}$ taken from Table 1. Of course this calculation assumes all of the loops to be of the same size, $R = e^{\gamma}/1.97 \times 10^{-2} = 84$ Å. A more reasonable assumption for neutron irradiated copper$^{11}$ is an exponential distribution defined by $C_L(R) = C_L(0) e^{-R/R_0}/R_0$ where $R_0$ characterizes the average size in the distribution. From Eqs. (10 – 12) the effect of this distribution of sizes can be taken into account by replacing Eq. (15) by

$$I^i(q_0) = \frac{\mu_{DS}}{I^0} \frac{\mu_0}{2\mu_0} \sum_{i} e^{-R_i/R_0} R_i^4 F(1/q_0 R_i) \frac{1}{F(1/q_0 R_i)}. \quad (16)$$

The intercept of a plot of this function versus $\ln(q_0)$ for small $q_0$ is given approximately by $1/R_0 e$ [see Eq. (14)]. Equation (16) has been evaluated numerically using an exponential size distribution with various $R_0$. Increments of $R_i$ in the summation were taken as 1 Å and a maximum $R_i$ of 400 Å was used for $R_0$ in the range 3 – 25 Å. It was then possible to choose the $R_0$ values required to obtain the measured intercepts, as demonstrated in Fig. 2, and these are listed in Table 1. $R_0$ values calculated using Eq. (14) are within 2% of those listed in Table 1. Using the values of $\mu_{DS}$ and $R_0$ from Table 1, the diffuse scattering intensities were calculated as a function of $q_0$ and the results are presented as the solid curves in Figures 2 – 5.

**Discussion**

The good agreement between the diffuse scattering as measured by anomalous transmission and that measured near Bragg reflections gives confidence that these processes are adequately accounted for by the theory. There are two aspects of this comparison, the magnitudes and the detailed shapes of the scattering curves. The magnitudes are derived from the experimental measurements of $\mu_{DS}$ and the shapes are determined by the “form factor” $(\mu_{DS}(q_0))/(\mu_{DS}(q_c))$. The agreement ($\sim 10\%$) between the magnitudes is very good since there are no adjustable parameters. The smallest $q_0$ for which intensities were measured in these experiments was about five times $q_c$ (the effective $q_0$ value for anomalous transmission) but for reasonable exponential distributions the “form factor” was insensitive to changes in $R_0$ for these $q_0$. For larger $q_0$ ($> 10^{-2}$) the form factor becomes quite sensitive to changes in $R_0$. Thus, although overall the fit between the computed and experimental scattering curves is good, the small discrepancies between them are considered to be significant. The assumptions used in the computation are: 1) that the scattering curve could be approximated as $q^{-2}$ for $q \leq 1/R$ and $q^{-4}$ for $q \geq 1/R$, 2) any changes in the anisotropy factor in Eq. (7) as a function of $q_0$ could be neglected, and 3) an exponential size distribution of dislocation loops. Assumptions 1) and 2) are oversimplifications made to facilitate computation, but the “form factor” ratio $(\mu_{DS}(q_0))/(\mu_{DS}(q_c))$ tends to minimize their effects, especially for small $q_0$. The effect of a more realistic approximation to the scattering curve is being investigated further. The shape of the computed curves for small $q_0$ is rather insensitive to the size distribution used, but at larger $q_0$ the size distribution was found to be important, and it should be noted that an exponential distribution (observed in electron microscopy studies) produces a much better fit than does a single size (Figure 3). The effect of small deviations from an exponential distribution is being investigated in further detail and will probably lead to improved fits.

Since the best fits were obtained for the low temperature and the $10.8 \times 10^{18}$ irradiations, those with the smallest and largest size distributions, it is clear that the experimental data can be fitted over this range of $R_0$. Therefore, the systematic variation in the fits with irradiation dose at $316 \, ^{\circ}\text{K}$ suggests a change in size distribution with dose. While such a change in size distribution has not been observed in electron microscopy studies, it is possible their experimental precision has not been sufficient to detect it.
It had been pointed out earlier \(^7\) that the value of the intercept in a plot \(I(q_0) \) vs \(q_0 \) was related to the size of the loops. In the more complete development presented in this paper it was shown that this intercept is at \(q_0 = e^{2}/R \) for defects of uniform radius \(R \) and at \(q_0 \approx 1/R_0 e \) for an exponential size distribution. Hence the integral diffuse scattering measurements can be used to obtain quantitative measurements of loop sizes. From the electron microscopy studies, which indicate an exponential distribution, values for \(R_0 \) were obtained which are in approximate agreement with the values deduced from the present studies, Table 1.

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Use of Asymmetric Dynamical Diffraction of X-rays for Multiple-Crystal Arrangements of the \((n_1, + n_2)\) Setting

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Dedicated to Prof. Dr. G. Borrmann on the occasion of his 65th birthday

Asymmetric dynamical diffractions of X-rays are successively used for multiple crystals arranged in the \((n_1, + n_2)\) setting. Double successive asymmetric diffractions of the \((n_1, + n_2)\) arrangement give a beam of peculiar spread in angle and wavelength. Triple successive asymmetric diffractions of the \((n_1, + n_2, - n)\) arrangement give a highly parallel and monochromatic beam for some appropriate combination of the asymmetry factors of the three crystals. Possible use of multiple asymmetric diffractions is discussed.

1. Introduction

Detailed information about dynamical diffraction phenomena of X-rays is obtained by using an incident beam of sufficiently narrow spread in angle and wavelength compared with the diffraction range of the crystal under investigation, which is of the order of seconds of arc. For such studies two kinds of arrangements are usually used: the double-crystal one of parallel or \((n, - n)\) setting and the triple-crystal one of \((n_1, + n_2, \pm n_3)\) setting.

In the former the spread of the beam used is narrow in angle only for each wavelength component, but, as a whole, not very narrow in angle nor wavelength. However, the dispersion effect due to the wavelength spread is eliminated owing to the parallel setting, in which the interplanar spacings of the diffracting planes of the first and second (specimen) crystals are equal to each other. In the parallel setting an extremely narrow angular spread of the incident beam is obtained by making use of the effects characteristic to dynamical diffraction.

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