Analytic Expressions for Displacement Fields of Dislocation Loops in Anisotropic Cubic Crystals

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Based on the analytic expression for the elastic Green’s function of anisotropic cubic crystals and on the description of a dislocation by a special distribution of dipole forces the displacement field of a dislocation loop is represented by a line integral. With the displacement field one can give analytic expressions for the dilation and the stress tensor in reciprocal space. From this, cross sections for Huang scattering and for nuclear and magnetic small-angle scattering at dislocations can be calculated. In the case of nuclear and magnetic scattering at straight dislocations, values were found which differ by a factor 0.5 to 5 from the values expected with the assumption of elastic isotropy.

Diffuse X-ray Scattering from the Displacement Field of Point Defects and Defect Clusters

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This review gives first a short introduction to the theoretical relations between experimentally observed diffuse scattering results and the desired information about defects. The situation is discussed under which diffuse scattered X-ray intensity from defects can be distinguished from other diffuse intensities (thermal diffuse scattering, Compton scattering). A typical experimental set-up is described to show what the requirements are in experimental resolution and how intensity distribution in reciprocal space can be measured most conveniently. A few typical experiments are discussed to demonstrate the physical potential of this method for studying impurities in metals and radiation-induced defects and defect clusters in ionic crystals. Special emphasis is given to the comparison of the experimental results with theoretical predictions: the intensity close to the Bragg peaks (Huang scattering) falls off as \(1/g^2\), \(g\) being the distance from a reciprocal-lattice point \(G\). The scattering intensity goes as \(G^2\). The intensity distribution in reciprocal space gives typical isointensity curves from which the symmetry of the double force tensor and its components can be deduced. Together with the measured shift of a Bragg peak, measurement of absolute scattering intensity gives a unique way of determining the defect concentration. Further away from the Bragg peaks (asymptotic scattering) the intensity is proportional to \(1/g^4\) and shows characteristic oscillations.

I. Introduction

Diffuse X-ray scattering measurements are a powerful tool for studying lattice distortions by defects. Although the technique was theoretically predicted long ago, only recently have experiments shown its real power. A review of the literature has been given by Krivoglaz (1969), Schmatz (1970), Dederichs (1973) and Peisl & Trinkaus (1973).

This review gives first a short introduction to the theoretical relations between experimental results and the desired information (§ 2). In § 3 we discuss the situations under which diffuse scattered X-ray intensity from defects can be distinguished from other diffuse intensities. § 4 describes a typical experimental set-up in order to show the requirements in experimental resolution and to show how intensity distributions in reciprocal space can be obtained most conveniently. In § 5 a few typical experiments will be discussed to demonstrate the physical potential of this method for studying impurities in metals, and radiation-induced defects and defect clusters in metals and ionic crystals. Special emphasis will be given to the comparison of the experimental results with theoretical predictions.
2. Huang scattering

In the kinematic approximation the elastic X-ray scattering cross section per unit-cell is obtained by summing the scattered amplitudes from the individual atoms or unit-cells:

$$\frac{d\sigma}{d\Omega} = \frac{1}{N} \sum_m |F_m| \exp[iK(r_m + u_m)]^2.$$ \hspace{1cm} (1)

The sum is taken over all \(N\) unit-cells, each having the structure amplitude \(F_m\). \(K\) is the scattering vector and \(r_m\) are the positions of the cells in an average lattice. \(u_m\) are deviations from this average lattice caused by the defects.

In the long-range limit the correlations of the lattice atoms define the positions and the intensities of the Bragg reflexions. For a finite but still large correlation length the deviations from this limit, due to the displacement field of the defects, determine the scattering intensities close to the Bragg reflexions. Here the phase factor can be expanded, since \(K \cdot u_m \ll 1\) and one gets equation (2).

$$\left(\frac{d\sigma}{d\Omega}\right)_{\text{HDS}} = |\tilde{F}|^2 |\tilde{u}|^2,$$

$$\tilde{u} = \frac{1}{\sqrt{N}} \sum_m u_m \exp(iKr_m). \hspace{1cm} (2)$$

The scattering cross section close to the Bragg peaks \((K \approx G)\) is determined by the Fourier transform \(\tilde{u}\) of the displacement field \(u_m\). The scattering by the defects themselves has been neglected and \(F_m\) has been replaced by its average \(\tilde{F}\). This part of the scattered intensity has been called Huang diffuse scattering (HDS). It corresponds to the one-phonon scattering from dynamical distortions. For a defect concentration \(c\) per unit cell the contributions from all defects have to be summed properly.

2.1. Huang scattering due to an isotropic defect in an isotropic lattice

In order to gain a rough idea of what one expects experimentally, let us consider the most simple case: an isotropically distorting defect in an isotropic medium. This shows all the important features.

The Fourier transform of the displacement field is usually described by a force array causing the same displacements as the actual defect. The dipole part of this force distribution is called the elastic-dipole tensor or double force tensor \(P_{ij}\). It describes the displacement field far away from the defect (Fernfeld) sufficiently well and enters the Huang scattering cross section. For an isotropic defect the elastic-dipole tensor has only the three equal diagonal components \(P\). In the Huang case we may also use continuum elasticity theory to describe the displacement field:

$$u(r) = \frac{P}{4\pi C_{11}} \cdot \frac{r}{r^4}. \hspace{1cm} (3)$$

\(C_{11}\) is an elastic constant, \(r\) denotes the distance of a lattice atom from the defect. In this case the Fourier transform of equation (3) is

$$\tilde{u}(q) = \frac{iP}{\Omega C_{11}} \cdot \frac{q}{q^2}. \hspace{1cm} (4)$$

\(\Omega\) is the atomic volume and \(q\) is a vector from any point in reciprocal space to the next-nearest reciprocal-lattice point \(g = K - G\).
Equations (2) and (4) lead to an expected X-ray scattering cross section,
\[
\left( \frac{d\sigma}{d\Omega} \right)_{\text{HDS}} = c \frac{|F|^2}{\Omega_c} \cdot \frac{P^2}{C_{11}} \cdot \frac{K \cdot \mathbf{g}^2}{g^2} = c \frac{|F|^2}{\Omega_c} \cdot \frac{P^2}{C_{11}} \cdot \frac{K^2}{g^2} \cdot \cos^2 (K, \mathbf{g}),
\]
where \( \Omega_c \) is the volume of the unit-cell. When we ask for surfaces of equal scattering intensity we expect from equation (5) spheres touching the reciprocal-lattice point (Huang spheres). Maximum diffuse scattering intensity is to be expected in the direction of \( \mathbf{G} \) and no intensity perpendicular to \( \mathbf{G} \).

3. Background scattering

Typical defect concentrations \( c \) and defect strengths \( P \) are of such an order of magnitude that the scattering cross section is quite small and has to compete with various types of background scattering. Close to a Bragg peak the most important contribution comes from thermal diffuse scattering (TDS). As the distortions by defects and the lattice vibration amplitudes may be of the same order of magnitude, one has to compare HDS with TDS in order to see whether there is any possibility of discriminating between the two types of scattering.

Fig. 1 shows the expected scattering intensity for aluminum close to the 200 reciprocal-lattice point. A defect concentration \( c = 10^{-4} \) and a dipole tensor \( P = 16 \) eV, which gives rise to a volume change \( \Delta V = 3P/(C_{11} + 2C_{12}) = 2\Omega \) per defect (interstitial atom in Al), were assumed. Normalized HDS intensity is plotted versus \( g/G \) the relative distance from the reciprocal-lattice point. For comparison, TDS intensity has been calculated (Warren, 1969) for two temperatures. At 300 K TDS intensity is about ten times the HDS intensity and it would be hard to detect the latter, at least with normal X-ray intensities. Cooling the sample to 5 K changes the situation; TDS greatly decreases and HDS is bigger and should be detectable.

There is another diffuse background scattering: Compton scattering. Close to the Bragg peak it is completely negligible. At some distance from the Bragg reflexion it may be of the same order as HDS. In this region TDS also may again become bigger than HDS. Here the high-temperature approximation for TDS is no longer valid and TDS no longer falls off as \( 1/g^2 \) but as \( 1/g \).

Other types of background scattering such as fluorescence of the sample, sample holder or cryostat windows, and stray radiation from slits and the air can in most cases be kept low by suitable experimental conditions.

4. Experimental arrangement

A typical experimental set-up which is used to measure HDS is shown schematically in Fig. 2. The X-ray source should most preferably be a high-power, rotating-anode tube. However, the first experiments and also later ones were quite successfully performed with lower-power, sealed-off tubes.

A bent quartz monochromator selects \( K\alpha_1 \) radiation. The X-rays hit the sample crystal and are reflected at a certain angle. Low-noise proportional counters are used as detector and monitor. Measurements are made point by point and with suitable electronics the results are produced on a teletype.
The ideal scattering geometry is given in Fig. 3. X-rays with a wave vector \( k_0 \) fall on the crystal at an angle \( \theta_1 \) and are detected at \( \theta_2 \). In the reciprocal lattice this means that we look for scattered intensity from a point at a distance \( g \) from the reciprocal-lattice point \( G \). A modified Bragg condition must be satisfied. By properly varying \( \theta_1 \) and \( \theta_2 \) we can detect scattering from any point in reciprocal space. The \( \phi-2\phi \) scan make it possible to scan along straight lines through the origin, the \( \omega \) scan on spheres around the origin.

Unfortunately we cannot use this ideal scattering geometry with infinitely good resolution. To gain intensity we have to allow some divergence of the incoming and scattered beams. We get scattered intensity from a whole volume element in reciprocal space.

Fig. 4 shows the extended volume element in reciprocal space due to finite divergence of the beams for two different scattering geometries. The arrangement in real space is shown on the left-hand side and the corresponding geometric situation in reciprocal space on the right-hand side. The divergence of the incoming beam, after the monochromator, is denoted by \( e_1 \) and the divergence of the scattered beam seen by the detector by \( e_2 \). Differently shaped volume elements in reciprocal space are observed for the so-called focusing and semi-focusing geometries.

Table 1. Information from diffuse X-ray scattering on the displacement field of point defects
Taken from Spalt, Lohstöter & Peisl (1973).

<table>
<thead>
<tr>
<th>Reciprocal-lattice point</th>
<th>( H00 )</th>
<th>( HH0 )</th>
<th>( HHH )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Direction of ( g ) in reciprocal lattice</td>
<td>( [100] )</td>
<td>( [110] )</td>
<td>( [111] )</td>
</tr>
<tr>
<td>( I_H (HKL), [hk1] )</td>
<td>( \frac{\alpha \gamma}{C_{11}} )</td>
<td>( \frac{2(\alpha - \beta)}{C_{14}} )</td>
<td>( \frac{\alpha + 2\beta + 4\gamma}{C_{14}} )</td>
</tr>
<tr>
<td>( I_0 )</td>
<td>( \frac{3F^2}{\Omega^2} \left( \frac{K^*}{g} \right)^2 I_e )</td>
<td>( \alpha = \sum_i P_{ii} )</td>
<td>( \beta = \sum_{i&gt;j} P_{ij} )</td>
</tr>
<tr>
<td>( a )</td>
<td>( \frac{2\alpha + 3\gamma}{C_{14}} )</td>
<td>( \gamma = \sum_{i&gt;j} P_{ij} )</td>
<td></td>
</tr>
</tbody>
</table>

Fig. 5. Huang diffuse scattering from oxygen-doped niobium (after Wombacher, 1973).
5. Experimental results

From diffuse X-ray scattering measurements close to Bragg peaks one wants to obtain information about the dipole tensor \( P_{ij} \), which describes the distortion field. For the isotropic case we have considered above we would have not needed HDS. Measuring the lattice-parameter change gives the full information \( 3\Delta a/ a \propto \text{Trace} \, P_{ij} \). For the general case of an anisotropic defect in an anisotropic cubic medium oriented at random Table 1 shows a recipe given by Trinkaus, by which one can obtain all possible information. One can obtain from HDS the three independent quadratic-tensor parameters \( \alpha, \beta \) and \( \gamma \).

Full information is obtained from measurements of HDS at reflexions and in directions for which \( \mathbf{G} \) and \( \mathbf{g} \) are parallel or perpendicular to each other and coincide with symmetry directions. For point defects no more information can be obtained by measuring the full iso-intensity contours. If one finds \( \gamma = 0 \) by measuring at a \( H00 \)-type reciprocal-lattice point in the [010] or [001] direction or at a \( (HH0) \) point in the [001] direction, it means that the dipole tensor has no off-diagonal elements and the defect main axes coincide with the lattice axis. \( \alpha - \beta = 0 \), which could be observed close to \( HHO \) in the [\{100\}] direction, means that the defect has trigonal symmetry.

HDS experiments have successfully been used to study point defects and their clusters (Baldwin, Sher-
Scattering from Frenkel pairs

When one measures $I_{\text{HDS}}$ as a function of the defect concentration $c$, one can easily also obtain the relative lattice-parameter change as a function of $c$. The quantities depend in different ways on the dipole tensor:

$$I_{\text{HDS}} \propto c(P_1^2 + P_v^2)$$
$$\Delta a/a \propto c(P_1 + P_v).$$

(6)

$P_1$ and $P_v$ are the elastic-dipole tensors for interstitials and vacancies respectively. One has two equations with two unknowns and one can determine $P_1$ and $P_v$ separately.

In the case of Frenkel pairs in metals, where no real reliable method exists for determining the absolute defect concentration, these relations [equation (6)] have been used to determine the defect concentration and then to calibrate other physical-property changes. An assumption about the magnitude of the vacancy contribution has to be made.

Effect of clustering

HDS is very sensitive to cluster formation. The reason for this can be understood in the following way: For statistically distributed point defects $I_{\text{HDS}} \propto c$. In the HDS approximation the same is true for clusters: $I_{\text{HDS}}' \propto n_c P_{1c}$. If $c$ defects form clusters of an average size $z$ the cluster concentration is $n_c = c/z$. If linear superposition of the defect strength is assumed, $P_{1c} = zP_{1}$. $I_{\text{HDS}}$ is therefore enhanced by a factor of $z$ if defects form clusters.

Asymptotic distortion scattering

In order to demonstrate all the possible information one can get from measuring diffuse scattering close to Bragg peaks, we have to consider the case where the phase factor in equation (1) can no longer be expanded. This has also been treated by Trinkaus (1971). He showed that a generalized form of the Stokes-Wilson approximation can be applied. That means that one only has to sum the reflexions from the heavily distorted areas. The intensity distribution of the asymptotic scattering shows some characteristic different features. The scattering cross section can be approximated by (Trinkaus, 1971, 1972, 1973, 1975)

$$\left( \frac{d\sigma}{d\Omega} \right)_{\text{as}} \simeq 1.2\pi^2 \cdot c \frac{|P|^2}{\Omega^2} \cdot \frac{P_1}{C_{11}} \cdot g^{-4}.$$

(7)

The asymptotic scattering cross section is proportional to $P$ and $K$, and it falls off more rapidly, as $1/g^4$. Trinkaus furthermore predicted oscillations of the scattered intensity (Trinkaus, Spalt & Peisl, 1970). The phase $\varphi$ of these oscillations is

$$\varphi - \varphi_0 \propto (|P| |K| g^{-2})^{1/3}.$$

(8)

Asymptotic scattering, in addition to HDS, has not been observed in the case of point defects. Only close to clusters are the distortions big enough to destroy the phase. Here both types of scattering have been observed.

Fig. 8 shows all the effects expected for diffuse scattering from clusters in LiF $\gamma$-irradiated at room temperature (Spalt, 1970). Diffuse scattered intensity is plotted on a log log scale. We see both the $1/g^2$ and the $1/g^4$ regions. The defect strength and concentration were known in this system. The asymptotic and the HDS intensities were therefore calculated under the assumption of a random distribution of defects. There is good agreement in the case of asymptotic scattering, but complete disagreement, of two orders of magnitude, for HDS. The experimental findings clearly show the enhancement of HDS due to clustering. After 200 h $\gamma$-irradiation, for which these are the results, an average number of $z = 200-300$ atoms in a cluster was determined. Here, where both types of scattering are observed, the concentration of the clusters can be determined from the different $P$ dependences. From the $g$ value where the $1/g^2$ dependence goes over to a $1/g^4$ dependence an average cluster size of about $7a$ could be determined.

These results also show, for the first time, the intensity oscillations predicted by Trinkaus et al. (1970) and mentioned earlier. The $P$ or $z$ value was determined from the phase of these oscillations and is quite consistent with the values mentioned above.

Clustering, cluster growth and change of the defect symmetry due to clustering have been successfully
studied on a variety of systems during irradiation and during thermal annealing. Some of the results will be given in a contributed paper (von Guérard & Peisl, 1975).

Asymmetry of the scattering distribution

We have omitted to mention one thing, the information one can in principle obtain from the asymmetry of the scattering distribution with respect to the Bragg peak. The asymmetry comes from the interference of the diffuse scattering amplitudes with X-rays scattered by the defect itself and by the heavily distorted immediate neighbourhood of the defect. From this asymmetry one could obtain information on the exact location of the defect in the unit-cell. Really conclusive results have however not been obtained from this.

Fig. 8. Diffuse scattering from γ-irradiated LiF. A: Asymptotic distortion scattering in [100]-direction the $1/g^2$ decrease is modulated as predicted by theory. H: Huang scattering expected for a random distribution of defects. The experimental results clearly show an enhancement due to clustering of defects. (after Spalt, 1970).

Conclusion

A series of experiments, partly reviewed in this paper, partly presented as original papers at this conference, have demonstrated that diffuse X-ray scattering experiments are very well suited to the study of point defects and their clusters and that this method gives some results which could not otherwise have been obtained.

Further work, both theoretical and experimental, will be necessary. Methods and sensitivity should be improved in order to apply this method to less favourable systems as well.

References