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Diffuse x-ray scattering near Bragg peaks from point defects in a general lattice

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Abstract. Using the continuum theory of linear elasticity, diffuse x-ray scattering has been calculated in the immediate neighbourhood of Bragg peaks from point defects in a lattice containing more than one atom in the unit cell. General expressions are obtained for the Debye-Waller factor, Huang diffuse scattering and the asymmetric scattering due to the defect. For lattices with one atom per unit cell, these expressions reduce to the well-known formulae of diffuse scattering.

Keywords. Interstitials; general lattice; Debye-Waller factor; Huang diffuse scattering; asymmetric scattering.

1. Introduction

Over the past few years, diffuse x-ray scattering has become increasingly important for investigating the relaxation behaviour and the configuration of point defects in metals (Dederichs 1973; Haubold 1975). While the diffuse scattering from point defects in cubic metals has been investigated extensively both theoretically and experimentally, the point defects in other lattices have not received much attention. Recently diffuse scattering measurements were reported on self-interstitials in zinc, an *hcp* lattice (Ehrhart and Schonfeld 1979). But the existing theoreies of diffuse scattering from cubic lattices (Krivoglaz 1969) are inadequate for analyzing the diffuse scattering data from point defects in a lattice containing more than one atom in the unit cell and require modification. In an earlier paper, a general theory of diffuse x-ray scattering was developed (Khanna and Haubold 1982) from self-interstitials in a crystal with a multi-atomic basis. This theory is based on lattice dynamical methods and is valid over the entire region of q space. In this paper a theory of diffuse x-ray scattering for some special regions of q space is presented using the continuum theory of linear elasticity. While retaining the basic features and approximations of the already existing theories, we calculate diffuse scattering in the neighbourhood of Bragg peaks. Expressions are also obtained for the Huang diffuse scattering, asymmetric scattering and the Debye-Waller factor of the defect. For lattices with one atom per unit cell, these expressions reduce to the well-known formulae of the diffuse scattering theory.

2. Theory

For a concentration C (number per unit volume) of point defects distributed at random, we assume a linear superposition of the strain fields around the point defects and neglect the interference between the scattering contributions from different defects (the 'single defect approximation'). The diffuse x-ray scattering cross-section can be written as

$$I_D = C|F(\mathbf{Q})|^2, \tag{1}$$

where

$$F(\mathbf{Q}) = f_Q^D + f\sum_n [\exp(i\,\mathbf{Q}\cdot\mathbf{t}_n) - 1]\exp(i\,\mathbf{Q}\cdot\bar{\mathbf{R}}_n) \tag{2}$$

$F(\mathbf{Q})$ is the defect structure factor; f_Q^D represents the Laue scattering at the defect and f is the atomic scattering factor of the atoms in the perfect lattice. $\bar{\mathbf{R}}_n$ is the position vector of an atom n in the average lattice, *i.e.*, in a lattice homogeneously relaxed by all the defects and $\mathbf{t}_n$ is the static displacement of the atom n due to the defect singled out. The scattering vector $\mathbf{Q}$ satisfies the relation, $\mathbf{Q} = \mathbf{h} + \mathbf{q}$, where $\mathbf{h}$ is a reciprocal lattice vector and $\mathbf{q}$ a vector within the first Brillouin zone.

Close to Bragg peaks, $\mathbf{q} \ll \mathbf{h}$ and $\mathbf{Q} \approx \mathbf{h}\cdot\exp(i\,\mathbf{Q}\cdot\mathbf{t}_n)$ can be approximated by $\exp(i\mathbf{h}\cdot\mathbf{t}_n)$, we have then for the defect structure factor

$$F(\mathbf{Q}) = f_Q^D + f\sum_n \exp(i\,\mathbf{Q}\cdot\bar{\mathbf{R}}_n)(\cos\mathbf{h}\cdot\mathbf{t}_n - 1)$$

$$+ if\sum_n \sin\mathbf{h}\cdot\mathbf{t}_n \exp(i\,\mathbf{Q}\cdot\bar{\mathbf{R}}_n) \tag{3}$$

We now look into the evaluation of each of the three terms in (3). The differences arising due to the lattice containing more than one atom in the unit cell will be pointed out as we go along.

2.1 *Term I:* f_Q^D

f_Q^D represents the Laue scattering at the defect. For an interstitial having an atomic form factor f_I (f for self-interstitials) at a position R_I

$$f_Q^D = f_I \exp(i\,\mathbf{Q}\cdot\mathbf{R}_I). \tag{4}$$

For a dumb-bell interstitial

$$f_Q^D = f[2\cos\mathbf{Q}\cdot\mathbf{R}_I - 1], \tag{5}$$

and $-f$ for a vacancy.

Since f_Q^D contains only the scattering at the defect, its evaluation does not depend on the number of atoms in the unit cell of the perfect lattice.

2.2 *Term II:* $f \sum_n \exp(i\mathbf{Q}\cdot\bar{\mathbf{R}}_n)(\cos \mathbf{h}\cdot\mathbf{t}_n - 1)$

As the displacements around the point defects decrease rapidly within a few atomic distances (Seeger *et al* 1962), $\cos \mathbf{h}\cdot\mathbf{t}_n$ can be approximated to one for the more distant neighbours. Thus term II will have contributions mainly from the strongly distorted immediate neighbourhood of the point defect.

The lattice displacements $\mathbf{t}_n$ can be calculated from the methods of lattice statics (Kanzaki 1957, Tewary 1973). Using the Kanzaki method, we illustrate in figure 1, a typical example of a distorted lattice in the presence of a point defect. Figure 1a shows the positions of atoms in a perfect lattice and the arrows point towards the probable direction of atomic relaxation due to the presence of the defect. Figure 1b shows the relaxed lattice. The exact magnitude and direction of the atomic displacement is immaterial for the present discussion. If the Kanzaki forces $\mathbf{K}$ extend to M atoms in the neighbourhood of the defect, then the displacement $\mathbf{t}_n$ of an atom n can be written as

$$\mathbf{t}_n = \sum_{m=1}^{M} G_{mn}\,\mathbf{K}_m. \tag{6}$$

where G_{mn} is the perfect lattice Green's function. The displacements of atoms in the strongly distorted neighbourhood of the defect have to be calculated explicitly from (6). In this method, $\mathbf{t}_n$ contains the explicit sum over the individual contributions from the defect forces at all the affected atoms and its calculation involves no assumption regarding the nature of the lattice.

Thus for the strongly distorted immediate neighbourhood of the defect, the method of evaluating t_n and hence term II will not depend on the number of atoms in the unit cell of the perfect lattice.

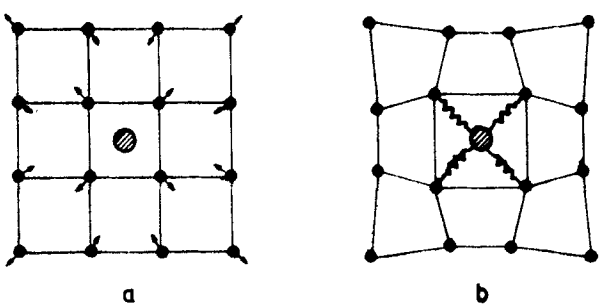

Figure 1 a. Atoms at their perfect lattice positions. Arrows point towards the probable direction of atomic relaxation due to the presence of the defect. **b.** Distorted lattice.

2.3 *Term III:* $if \sum_n \sin \mathbf{h} \cdot \mathbf{t}_n \exp(i\mathbf{Q} \cdot \bar{\mathbf{R}}_n) \simeq if \mathbf{h} \cdot \sum_n \mathbf{t}_n \exp(i\mathbf{Q} \cdot \bar{\mathbf{R}}_n)$ (7)

This term has contributions from all the atoms of the lattice affected by the presence of the defect. In regions very close to Bragg peaks, it is mainly governed by the long range displacement field of the point defect which can be calculated using the continuum theory of linear elasticity. The summation n in (7) can be divided into two sums: one over the atoms k in the unit cell and other over all the unit cells l of the lattice. Term III then becomes

$$if \mathbf{h} \cdot \sum_l \sum_k \mathbf{t}(lk) \exp\{i\mathbf{Q} \cdot [\mathbf{x}(l) + \mathbf{x}(k)]\} \tag{8}$$

Since all the atoms in a unit cell will be displaced equally in the elastic displacement field of the point detect, $\mathbf{t}(lk)$ is independent of the sublattice index k and may be replaced by $\mathbf{t}(l)$. (8) then reduces to

$$\begin{aligned} & i\left(f \sum_k \exp[i\mathbf{Q} \cdot \mathbf{x}(k)]\right) h \cdot \sum_l \mathbf{t}(l) \exp[i\,\mathbf{Q} \cdot \mathbf{x}(l)] \\ & \simeq i\left(f \sum_k \exp[i\mathbf{h} \cdot \mathbf{x}(k)]\right) h \cdot \sum_l \mathbf{t}(l) \exp[i\mathbf{q} \cdot \mathbf{x}(l)] \quad (q \ll h) \\ & \simeq i\left(f \sum_k \exp[i\mathbf{h} \cdot \mathbf{x}(k)]\right) \frac{\mathbf{h} \cdot \mathbf{t}(\mathbf{q})}{V_c}, \end{aligned} \tag{9}$$

where $\mathbf{t}(\mathbf{q}) = \int d\mathbf{R} \exp(i\mathbf{qR})\, \mathbf{t}(R)$ as the Fourier transform of the elastic displacement field of the point defect and V_c denotes the volume of the unit cell. $\mathbf{t}(\mathbf{q})$ can be calculated using the elastic stiffness tensor $\mathbf{C}_{ijkl}$ and the force dipole tensor of the defect (Trinkaus 1972)

$$t_k(\mathbf{q}) = iD_{ki}^{-1} P_{ij}\, q_j \tag{10}$$

where $D_{ik} = \mathbf{C}_{ijkl}\, q_j\, q_l$ and $P_{ij} = \sum_n K_i^n\, r_j^n$. P_{ij} is the first moment of the Kanzaki forces $\mathbf{K}$ due to the defect. The summation convention for the repeated indices is implied. Term III can then be written as

$$-\left(\frac{f \sum_k \exp[i\mathbf{h} \cdot \mathbf{x}(k)]}{V_c}\right)(h_i\, D_{ik}^{-1}\, P_{kj}\, q_j) \tag{11}$$

Combining (3) and (11) the general form of the diffuse scattering cross-section close to Bragg peaks can be written as

$$I_D = C\left|f_Q^D + f\sum_n \exp\left[i\,\mathbf{Q}\cdot\bar{R}_n\right](\cos\mathbf{h}\cdot\mathbf{t}_n - 1)\right.$$

$$\left.\left(\frac{f\sum_k \exp\left[i\,\mathbf{h}\cdot\mathbf{x}(k)\right]}{V_c}\right)(h_i\, D_{ik}^{-1}\, P_{kj}\, q_i)\right|^2 \tag{12}$$

(12) readily reduces to the well-known formulas of diffuse scattering for a lattice containing one atom in the unit cell.

3. Debye-Waller factor

Static distortions associated with point defects also give rise to a weakening of the intensity of the x-ray reflections apart from the diffuse scattering in the region between Bragg peaks. For a distorted lattice, x-ray scattering cross-section at the Bragg peak ($\mathbf{Q} = \mathbf{h}$) can be written as

$$I(\mathbf{h}) = \left|f\sum_l \sum_k \exp\left[i\,\mathbf{h}\cdot\mathbf{x}(l\,k)\right]\exp\left[i\,\mathbf{h}\cdot\boldsymbol{\delta}\,\mathbf{x}(l\,k)\right]\right|^2 \tag{13}$$

where $\mathbf{x}(lk)$ is the position vector of the kth atom in the lth until cell and $\boldsymbol{\delta}\,\mathbf{x}(lk)$ is the corresponding shift in its position caused by the presence of the defects.

For a low concentration of randomly distributed defects, each defect can be considered isolated in a supercell in the average lattice (the 'single defect approximation'). The scattering cross-section for the defective crystal can then be obtained by incoherently adding the individual contributions from the supercells containing defects and from the regions unaffected by the presence of the defects. If the supercell around a defect consists of M unit cells (a variable number, depending on the strength of the defect), there will be $N(1 - MC)$ unit cells unaffected by the presence of the defects, N being the total number of unit cells. For atoms in these unit cells, $\boldsymbol{\delta}\mathbf{x}(lk)$ will be zero. The scattering cross-section can then be written as

$$I(\mathbf{h}) = \left|f\sum_k \exp\left[i\mathbf{h}\cdot\mathbf{x}(k)\right]\left[CN\sum_{l=1}^{M}\exp\left[i\mathbf{h}\cdot\boldsymbol{\delta}\mathbf{x}(lk)\right] + N(1 - MC)\right]\right|^2$$

$$= \left|f\sum_k \exp\left[i\mathbf{h}\cdot\mathbf{x}(k)\right]\left\{N\left[1 - C\sum_{l=1}^{M}(1 - \exp\left[i\mathbf{h}\cdot\boldsymbol{\delta}\,\mathbf{x}(lk)\right])\right]\right\}\right|^2 \tag{14}$$

$$= N^2\left|f\sum_k \exp\left[i\,\mathbf{h}\cdot\mathbf{x}(k)\right]\exp(-M_k)\right|^2 \tag{15}$$

where $$M_k = C\sum_{l=1}^{M}\{1 - \exp\left[i\,\mathbf{h}\cdot\boldsymbol{\delta}\,\mathbf{x}(lk)\right]\}, \tag{16}$$

is the Debye-Waller factor of the atoms in the kth sub-lattice of the unit cell. For defects having the inversion symmetry of the displacement field, we have

$$M_k = C \sum_{l=1}^{M} [1 - \cos \mathbf{h} \cdot \boldsymbol{\delta}\, \mathbf{x}\,(l\,k)]. \tag{17}$$

Rewriting the second term in (3) in terms of Debye-Waller factor

$$\sum_n \exp\,(i\ \mathbf{Q}\cdot\bar{\mathbf{R}}_n)\,(\cos\ \mathbf{h}\cdot\mathbf{t}_n - 1) = \sum_l \sum_k \exp\,[i\,\mathbf{h}\cdot\mathbf{x}\,(k)]\,(\cos\,\mathbf{h}\cdot\mathbf{t}\,(lk) - 1)$$

$$= -\frac{1}{C}\sum_k \exp\,[i\,\mathbf{h}\cdot\mathbf{x}(k)]\,M_k. \tag{18}$$

We have then for the diffuse scattering cross-section

$$I_D = C\Bigg| f_Q^D - \frac{f}{C}\sum_k \exp\,[i\,\mathbf{h}\cdot\mathbf{x}(k)]\,M_k$$

$$-\frac{f\sum_k \exp\,[i\,\mathbf{h}\cdot\mathbf{x}(k)]}{V_c}\,(h_t\ D_{ik}^{-1}\,P_{kj}\,q_j)\Bigg|^2 \tag{19}$$

4. Special features of diffuse x-ray scattering

4.1 *Huang diffuse scattering*

Close to Bragg peaks ($\mathbf{q} \ll \mathbf{h}$, $\mathbf{h} \approx \mathbf{Q}$), the leading term (third) of (19) varies as $1/q^2$ and the contribution from this term (known as Huang diffuse scattering (HDS)) is

$$I_H = C\left|\frac{f\sum_k \exp\,[i\,\mathbf{h}\cdot\mathbf{x}(k)]}{V_c}\,(h_t\ D_{ik}^{-1}\,P_{kj}\,q_j)\right|^2 \tag{20}$$

The only difference in the HDS cross-section due to the presence of more than one atom in the unit cell is the appearance of a phase factor $\sum_k \exp\,[i\,\mathbf{h}\cdot\mathbf{x}(k)]$. This phase factor equals unity for lattices with one atom per unit cell. Equation (20) is the contribution to HDS from a given orientation of a defect configuration. If the crystal contains many equivalent orientations of the defect configuration, then their contribution to HDS must be appropriately averaged over. We require an average of the product $P_{ij}\,P_{kl}$ of the dipole force tensors over the possible equivalent orientations. All equivalent orientations of an anisotropic defect configuration can be generated by the appropriate symmetry operations of the perfect lattice and the corresponding values of the dipole tensor components can be calculated using standard formulae. The method of computing the required average contribution to HDS is a general one

and requires no modification for lattices containing more than one atom in the unit cell.

4.2 *Asymmetry of diffuse scattering*

Next order contribution to I_D, varying as $1/q$, comes from the interference of the Huang scattering amplitude with the first two terms in (19)

$$I_{\text{as}}(\mathbf{Q}) = -2C\,|F|^2\left(\frac{hi\,D_{ik}^{-1}P_{kj}q_j}{V_c}\right)\times$$

$$\left[\text{Re}\left(\frac{f_Q^D}{F}\right) - \text{Re}\left(\frac{\sum_k \exp\,[i\mathbf{h}\cdot\mathbf{x}\,(k)]\;M_k}{CF}\right)\right], \tag{21}$$

where $$F = f\sum_k \exp\,[i\mathbf{h}\cdot\mathbf{x}\,(k)]. \tag{22}$$

Since (21) is linear in $\mathbf{t}(\mathbf{q})$, it is asymmetric about the Bragg reflection:

$$I_{\text{as}}(\mathbf{h}+\mathbf{q}) = -I_{\text{as}}(\mathbf{h}-\mathbf{q}). \tag{23}$$

This asymmetry arises from the interference of the scattering at the atoms in the elastic displacement field of the defect with the Laue scattering at the defect itself and the scattering contribution from the strongly distorted immediate neighbourhood of the defect.

The sign of the asymmetric term depends sensitively on the strength of the displacement field and changes characteristically depending on the value of the Debye-Waller factor

$$\frac{M_k}{C} = \sum_l (1 - \cos\,\mathbf{h}\cdot\mathbf{t}\,(lk)) \ll 1 \text{ or } \gg 1. \tag{24}$$

For very small displacement fields ($M_k/C \ll 1$), the contribution from the atoms in the immediate neighbourhood of the defect can be neglected in (21) and one gets

$$I_{\text{as}}(\mathbf{Q}) = -2C\,|F|^2\left(\frac{h_i\,D_{ik}^{-1}P_{kj}q_j}{V_c}\right)\left[\text{Re}\left(\frac{f_Q^D}{F}\right)\right]. \tag{25}$$

For small displacement fields of the defects, I_{as} directly yields information about the sign of the force dipole tensor P_{kj}.

For strong distortion fields of the point defect the Laue scattering at the defect can be neglected in comparison and one gets

$$I_{\text{as}}(\mathbf{Q}) = 2C\,|F|^2\left(\frac{h_i\,D_{ik}^{-1}P_{kj}q_j}{V_c}\right)\left[\text{Re}\left(\frac{\sum_k \exp\,[i\,\mathbf{h}\cdot\mathbf{x}\,(k)]\;M_k}{CF}\right)\right] \tag{26}$$

For strong distortion fields of the point defect, I_{as} yields direct information about the Debye-Waller factor. I_{as}, though relatively weak as compared to HDS (due to the cancellation between the two terms in (21) and $1/q$ dependence), gives important information about some defect parameters.

5. Conclusions

We have shown how the existing theories of diffuse scattering get modified when the lattice contains more than one atom in the unit cell. The present theory, based on the continuum theory of linear elasticity, is valid only in the region very close to Bragg peaks (where one gets particularly strong intensities) and has the advantage of being relatively simple. The general theory of diffuse x-ray scattering, based on lattice dynamical methods, is valid over the entire region of q space and can also be used to study the diffuse scattering close to Bragg peaks. But it has the disadvantage of being rather complicated and requires prior information about the atomic force constants and the lattice Green functions. Nonetheless both the theories must give identical results close to Bragg peaks. Since the atomic force constants and the elastic constants are related, a comparison of HDS (main contribution close to Bragg peaks) as calculated by these two different methods will provide a good check on the theory developed in previous sections.

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