

nucleus of bromine, and it is not obvious that this method should give exact agreement with the nuclear coordinate. We hope to make a more precise determination by observation of high-index reflections both at $\psi = 0$ and 90° .

5. Discussion

The angular dependence displayed in Fig. 1 is a verification that the optical model based on kinematic diffraction and dipole scattering is valid for these experiments. It also demonstrates, except for a minority of the points, that the signals are not obscured by the Renninger reflections or by allowed reflections at other wavelengths. The detailed numerical results show that one can measure the magnitude of the polarization anisotropy of scattering in a new way. In the present case it indicates that the birefringence is even larger than was demonstrated in earlier experiments. This new method does not require polarized radiation and may have other advantages for some materials.

This new technique is a method of selective diffraction. The signal comes only from atoms of one element, and of that element only those atoms which are in a chemical state which has significant anisotropy of susceptibility. Thus it has potential for differentiation of oxidation states or bonding geometries. It is shown to be capable of rather precise determination of atomic coordinates. Perhaps its most obvious utility in contemporary science is to help locate a few heavy atoms in an otherwise very compli-

cated crystal structure, a frequent objective in macromolecular crystallography. All of these things can be accomplished with other techniques if the data are accurate enough. The advantage of the new method is that the signal is observed directly, rather than derived from the sometimes small and inaccurate differences of large quantities.

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On the Wavelength Dependence of the Reflectivity of One-Dimensionally Distorted Crystals

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Abstract

Scaling properties of the integrated reflectivity of non-absorbing perfect or ideally imperfect crystals as a function of wavelength, in the symmetrical Laue and Bragg cases, are shown also to be valid for distorted crystals where the gradient of the lattice phase factor is perpendicular to the crystal surfaces. This result is obtained by an analysis of the Taupin-Takagi equations. Some previous experiments (test of a proposal for extinction-free measurements of F_M/F_N in polarized neutron scattering by magnetic crystals, and neutron diffraction from curved and non-curved crystals) are discussed from this point of view.

1. Introduction

The integrated reflectivities of perfect crystals ρ_D (dynamical theory) and of ideally imperfect crystals ρ_K (kinematical theory) have common properties as functions of the wavelength λ , in symmetrical Bragg or Laue geometries. It will be shown that these properties are also verified for distorted crystals where the gradient of the lattice phase factor, which occurs in the Taupin-Takagi equations and is the scalar product of the local displacement field by the diffraction vector, is everywhere perpendicular to the crystal surfaces; this is actually a one-dimensional distortion, because the phase factor then depends only on the z coordinate of Fig. 1. The present analysis is related

to neutron rather than X-ray diffraction because absorption effects are supposed to be negligible. The dimensions of the beam must be smaller than the dimensions of the crystal surfaces in order to avoid edge effects.

In the symmetrical Laue case for non-absorbing crystals, ρ_D and ρ_K depend on the scattering length per unit cell F , on the crystal thickness t and on λ through the reduced parameter

$$A = \lambda Ft / V_c \cos \theta_B$$

where V_c is the volume of the unit cell and θ_B the Bragg angle (Zachariasen, 1945). One application of this property is the proposal for an extinction-free determination of the ratio of the magnetic and nuclear scattering lengths in polarized neutron scattering by magnetic crystals (Guigay, Schlenker & Baruchel, 1982).

In the symmetrical Bragg case for non-absorbing crystals, ρ_K and ρ_D are given by (Zachariasen, 1945)

$$\rho_K = \tan \theta_B A^2 \lambda / 2t \sin \theta_B$$

and

$$\rho_D = \tan \theta_B A \tanh(A) \lambda / 2t \sin \theta_B$$

with A now equal to $(\lambda t F / V_c \sin \theta_B)$; as $\lambda / \sin \theta_B$ is a constant, ρ_K and ρ_D are both proportional to $\tan \theta_B$ as λ is varied.

The Taupin-Takagi equations have their simplest form in the oblique coordinate system (s_o/s_g) defined by the directions of the incident and diffracted beams:

$$\Lambda \partial D_o / \partial s_o = i \exp [i\varphi(s_o, s_g)] D_g(s_o, s_g),$$

$$\Lambda \partial D_g / \partial s_g = i \exp [-i\varphi(s_o, s_g)] D_o(s_o, s_g);$$

but it will be convenient for our purpose to use the rectangular (x, z) coordinates of Fig. 1. We shall consider the solutions of these equations for an incident plane wave, with an angular deviation $\varepsilon = \theta - \theta_B$ from the exact Bragg incidence; a simple phase factor related to ε is to be added to the phase factor corresponding to the crystal distortion, since a crystal rotation around the Bragg position can be considered as a displacement field, as shown in Figs. 2(a) and (b). This leads to a reflection coefficient which is to be integrated over ε in order to obtain the integrated reflectivity.

2. Discussion of the symmetrical Laue case

The Taupin-Takagi equations are then (see Fig. 1a)

$$\Lambda \tan \theta_B \partial D_o / \partial x + \Lambda \partial D_o / \partial z$$

$$= i \exp [i\varphi(x, z) + i\epsilon g z] D_g(x, z),$$

$$- \Lambda \tan \theta_B \partial D_g / \partial x + \Lambda \partial D_g / \partial z$$

$$= i \exp [-i\varphi(x, z) - i\epsilon g z] D_o(x, z),$$

with boundary conditions independent of x : $D_o = 1$ and $D_g = 0$ on the front surface ($z = 0$) of the crystal. Λ is the *Pendellösung* length ($\Lambda = V_c \cos \theta_B / \lambda F$) and g is the magnitude of the scattering vector ($g = 4\pi \sin \theta_B / \lambda$).

If the crystal distortion is such that the corresponding phase factor $\varphi(x, z)$ does not depend on x , the solutions of these equations are clearly also independent of x . Setting $Z = z/\Lambda$ we get

$$dD_o/dZ = i \exp [i\varphi(Z) + i\epsilon g Z \Lambda] D_g(Z),$$

$$dD_g/dZ = i \exp [-i\varphi(Z) - i\epsilon g Z \Lambda] D_o(Z).$$

The intensity of the diffracted wave, outside the crystal of thickness t , is then a function of $\epsilon g \Lambda$ and t/Λ . The integrated reflectivity, obtained by integrating over ϵ , is then just a function of Λ only when λ is varied.

3. Discussion of the symmetrical Bragg case

The Taupin-Takagi equations are then (see Fig. 1b)

$$\frac{\Lambda}{\tan \theta_B} \frac{\partial D_o}{\partial x} + \Lambda \frac{\partial D_o}{\partial z} = i \exp [i\varphi(x, z) + i\epsilon g x] D_g(x, z)$$

$$\frac{\Lambda}{\tan \theta_B} \frac{\partial D_g}{\partial x} - \Lambda \frac{\partial D_g}{\partial z} = i \exp [-\varphi(x, z) - i\epsilon g x] D_o(x, z).$$

Λ is the *Pendellösung* length, $\Lambda = V_c \sin \theta_B / \lambda F$. Introducing $d_g(x, z)$ such that

$$d_g(x, z) = D_g(x, z) \exp [i\epsilon g x]$$

we get

$$\frac{\Lambda}{\tan \theta_B} \frac{\partial D_o}{\partial x} + \Lambda \frac{\partial D_o}{\partial z} = i \exp [i\varphi(x, z)] d_g(x, z)$$

$$\frac{\Lambda}{\tan \theta_B} \left\{ \frac{\partial D_g}{\partial x} - i\epsilon g d_g \right\} - \Lambda \frac{\partial D_g}{\partial z} = i \exp [-i\varphi(x, z)] D_o(x, z).$$

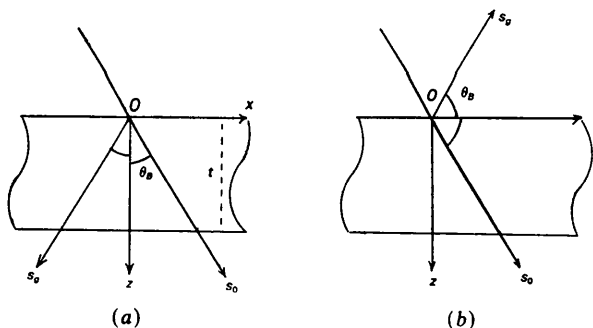


Fig. 1. Scattering geometry in: (a) the symmetrical Laue case: $x = (s_o - s_g) \sin \theta_B$; $z = (s_o + s_g) \cos \theta_B$; (b) the symmetrical Bragg case: $x = (s_o + s_g) \cos \theta_B$; $z = (s_o - s_g) \sin \theta_B$.

The boundary conditions are independent of x : $D_o = 1$ in the $z = 0$ plane (front surface) and $d_g = 0$ in the $z = t$ plane (back surface of the crystal).

If the phase factor $\varphi(x, z)$ does not depend on x , the solutions of these equations are clearly also independent of x . We then get

$$\begin{aligned} \Lambda \, dD_o/dz &= i \exp[i\varphi(z)] d_g(z) \\ -i\Lambda/2 \tan \theta_B \, \varepsilon g d_g(z) - \Lambda \, d(d_g)/dz \\ &= i \exp[-i\varphi(z)] D_o(z). \end{aligned}$$

We choose the parameter

$$y = (\Lambda/2 \tan \theta_B) \varepsilon g = \pi \sin(2\theta_B) (\Lambda/\lambda) \varepsilon$$

as the measure of the deviation from the exact Bragg position. The diffracted wave intensity outside the crystal is then a function of y and of t/Λ , which is independent of λ and of y . To obtain the integrated reflectivity we can integrate over y , and multiply the result ρ_y by the value of the ratio ε/y :

$$\rho = [\lambda/\pi\Lambda \sin(2\theta_B)] \rho_y (\Lambda \sin \theta_B, t/\Lambda \sin \theta_B).$$

It is then easy to verify that ρ is proportional to $\tan \theta_B$ when the wavelength λ is varied.

4. Polarized neutron scattering experiments (Laue case)

For a collinear magnetic crystal, if the magnetization is perpendicular to the scattering vector, the scattering length is equal to the sum or difference of the nuclear and magnetic scattering lengths per unit cell, F_N and F_M , depending on the incident neutron polarization direction. Our question is whether the reflectivities $\rho_{\pm}(u)$ as functions of $u = \tan \theta_B$ verify a scaling relation of the type

$$\rho_+(u) = \rho_-(Ku)$$

with K defined as $K = |(F_N + F_M)/(F_N - F_M)|$.

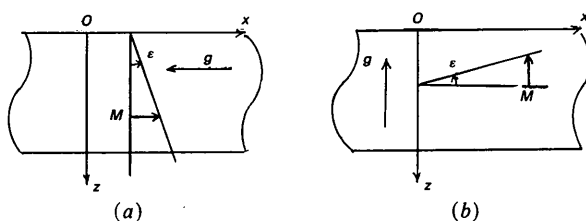


Fig. 2. Displacement field at a point $M(x, z)$ associated with a rotation $\varepsilon = \theta - \theta_B$ of the crystal from the exact Bragg position. (a) Laue case: displacement $z\varepsilon$ along x axis. (b) Bragg case: displacement $-x\varepsilon$ along z axis.

Some experiments have been performed (Baruchel, Guigay, Mazuré-Espejo, Schlenker & Schweizer, 1982) on the 220 reflection from a flux-grown crystal of yttrium iron garnet, whose main defects are growth bands resulting in a displacement field along the local growth direction. The scaling relation was reasonably satisfied when using a sample with a single growth direction parallel to the z axis of Fig. 1, leading to a value of K in close agreement with previous determinations based on usual extinction treatments (Bonnet, Delapalme, Becker & Fuess, 1976). Another set of measurements, using a sample having a different growth direction inclined to the z axis, did not agree with the scaling relation. This difference is in agreement with the present analysis because, in the second case, the phase factor is not only a function of z , but also of x .

5. Bragg-case experiments

A simple test of the present analysis can be obtained by measuring the ratio of the intensities diffracted by two crystals, for different values of the wavelength: if these intensities are both proportional to $\tan \theta_B$, their ratio should obviously be constant as λ is varied, even if the investigated crystals do not behave kinematically.

This was indeed observed when comparing the nuclear and magnetic diffracted intensities from a crystal of manganese phosphide, MnP, in its helimagnetic phase (Baruchel, Patterson & Guigay, 1986), the one-dimensionally distorted structure being in this case a sequence of helimagnetic domains with opposite chiralities.

Opposite results were obtained in measurements on curved and non-curved crystals by Boeuf & Rustichelli (1974). This is easily understood because the phase factor is then a function of x instead of being a function of z .

Further experiments on the Bragg case may provide a method to study the anisotropy of the displacement field in deformed crystals.

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