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Comments on Absorption in Takagi–Taupin Equations

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Abstract

The Takagi–Taupin theory of X-ray diffraction leaves an ambiguity in the choice of the wave vector inside the crystal. This holds also for its imaginary part which describes absorption. One consequence of this ambiguity is that the wave vector $\mathbf{k}_0$ inside the crystal need not always satisfy the continuity conditions for the tangential component of the wave vector at the entrance surface. But if the direction of the imaginary part is once fixed then it determines the particular manner of solution of the Takagi–Taupin equations. Thus a direction of the imaginary part of the wave vector in the crystal parallel to the reflecting net planes will in general ensure that the continuity condition is not satisfied; only a wave vector with an imaginary part perpendicular to the crystal surface can satisfy this condition.

1. Introduction

Dynamical X-ray diffraction in perfect and distorted crystals may be described by the Takagi–Taupin equations (Takagi, 1962, 1969; Taupin, 1964). An important feature of this theory is that the 'amplitudes' $D_h(\mathbf{r})$ of the generalized Bloch waves are not constant but are slowly varying functions of position. This fact leaves an ambiguity in the choice of the wave vector $\mathbf{k}_0$ inside the crystal. After Takagi a convenient choice is that the magnitude of $\mathbf{k}_0$ is given by

$$|\mathbf{k}_0| = k = nK, \qquad (1a)$$

where n is the mean refractive index and $K = 1/\lambda$, the wave number in vacuum, and sometimes that

$\mathbf{k}_0$ may satisfy the continuity condition for the tangential component of the wave vector at the entrance surface. (1*b*)

The Takagi–Taupin equations are also applicable in the case of an absorbing crystal, provided that all relevant quantities now assume complex values. The real and imaginary parts of $\mathbf{k}_0$ are called $\mathbf{k}_{0r}$ and $\mathbf{k}_i$, respectively, so that

$$\mathbf{k}_0 = \mathbf{k}_{0r} + i\mathbf{k}_i. \qquad (2)$$

The direction of $\mathbf{k}_i$ may be chosen arbitrarily. However, this choice influences the particular manner of solution of the equations. Hence it determines whether or not $\mathbf{k}_0$ satisfies the continuity condition (1*b*) and whether or not the parameter β_h [(4)] may be chosen equal to zero. In principle these problems are solved in the literature but they have never been pointed out explicitly.

2. Takagi–Taupin equations and boundary conditions

Let us recall the well known equations, for simplicity in the case of a perfect crystal and for the two-beam case:

$$\begin{aligned} \frac{\partial}{\partial s_0} D_0(\mathbf{r}) &= -i\pi K \chi_{\bar{h}} C D_h(\mathbf{r}) \\ \frac{\partial}{\partial s_h} D_h(\mathbf{r}) &= -i\pi K \chi_h C D_0(\mathbf{r}) + 2\pi i K \beta_h D_h(\mathbf{r}) \end{aligned} \qquad (3)$$

where χ_h and $\chi_{\bar{h}}$ are the Fourier coefficients of the dielectric susceptibility; C is the polarization factor; s_0 and s_h are unit vectors along the refracted and reflected directions;

$$\beta_h = (\mathbf{k}_h^2 - \mathbf{k}_0^2)/2\mathbf{K}^2 \qquad (4)$$

where $\mathbf{k}_0$ and $\mathbf{k}_h$ are vectors inside the crystal and $\mathbf{K}$ is the wave vector in vacuum. The extremities of both wave vectors $\mathbf{k}_0$ and $\mathbf{k}_h$ are matched through the value of β_h.

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The boundary conditions along the entrance surface are, in the Laue case,

$$\begin{aligned} D_0(\mathbf{r}_e) &= D_0^{(a)}(\mathbf{r}_e)\exp[-2\pi i(\mathbf{K}_0-\mathbf{k}_0)\mathbf{r}_e] \\ D_h(\mathbf{r}_e) &= 0 \\ \frac{\partial D_0}{\partial s_0}(\mathbf{r}_e) &= 0 \\ \frac{\partial D_h}{\partial s_h}(\mathbf{r}_e) &= -i\pi K\chi_h C D_0^{(a)}(\mathbf{r}_e)\exp[-2\pi i(\mathbf{K}_0-\mathbf{k}_0)\mathbf{r}_e], \end{aligned} \tag{5}$$

and in the Bragg case,

$$\begin{aligned} D_0(\mathbf{r}_e) &= D_0^{(a)}(\mathbf{r}_e)\exp[-2\pi i(\mathbf{K}_0-\mathbf{k}_0)\mathbf{r}_e] \\ \frac{\partial D_h}{\partial s_h}(\mathbf{r}_e) &= -i\pi K\chi_h C D_0^{(a)}(\mathbf{r}_e)\exp[-2\pi i(\mathbf{K}_0-\mathbf{k}_0)\mathbf{r}_e] \\ &\quad +2\pi i K\beta_h D_h(\mathbf{r}_e), \end{aligned} \tag{6}$$

where $\mathbf{r}_e$ is the position vector of a point on the entrance surface and $D_0^{(a)}(\mathbf{r}_e)$ is the amplitude of the incident wave in vacuum at the point $\mathbf{r}_e$. The real wave vector of this wave is $\mathbf{K}_0$.

3. Introduction of absorption

The complex wave vectors have the form

$$\mathbf{k}_0 = \mathbf{k}_{0r} + i\mathbf{k}_i \tag{2}$$

$$\mathbf{k}_h = \mathbf{k}_{hr} + i\mathbf{k}_i. \tag{7}$$

From (1a) it follows that

$$k_0^2 = K^2(1+\chi_0) = K^2(1+\chi_{0r}+i\chi_{0i}).$$

After separating real and imaginary parts and neglecting quadratic terms of χ_{0r} and χ_{0i} one gets

$$k_{0i} = K\chi_{0i}/2\cos\eta_0 = -\mu/4\pi\cos\eta_0 \tag{8}$$

$$k_{0r} = K(1+\chi_{0r}) \tag{9}$$

where η_0 is the angle between $\mathbf{k}_{0r}$ and $\mathbf{k}_i$ and μ is the linear photoelectric absorption coefficient. The real and imaginary parts of β_h are

$$\beta_{hr} = (k_{hr}^2 - k_{0r}^2)/2K^2 \simeq (k_{hr}-k_{0r})/K \tag{10}$$

$$\beta_{hi} = (k_i/K^2)(k_{hr}\cos\eta_h - k_{0r}\cos\eta_0) \tag{11}$$

where η_h is the angle between $\mathbf{k}_{hr}$ and $\mathbf{k}_i$.

4. Solution by the method of Riemann

Authier & Simon (1968) presented analytical solutions of (3) for a perfect crystal in the Laue case. If one converts (3) into two second-order differential equations of hyperbolic form,

$$\begin{aligned} \frac{\partial^2 D_0}{\partial s_0\,\partial s_n} - 2\pi i\beta_h\frac{\partial D_0}{\partial s_0} + \pi^2K^2C^2\chi_h\chi_{\bar{h}}D_0 &= 0 \\ \frac{\partial^2 D_h}{\partial s_0\,\partial s_h} - 2\pi i\beta_h\frac{\partial D_h}{\partial s_h} + \pi^2K^2C^2\chi_h\chi_{\bar{h}}D_h &= 0, \end{aligned} \tag{12}$$

they can be solved by the method of Riemann.

Let us first assume the Laue case and let P be a point on the exit surface and A and B two points on the entrance surface [see Authier & Simon (1968), Fig. 2]. Then the solution at P for D_h can be expressed in the form

$$D_h(P) = -\frac{i\pi KC\chi_h\gamma_0}{\sin 2\Theta_B}\int_{BA} D_0^{(a)}(\xi)v(\xi)\,d\xi \tag{13}$$

where

$$\gamma_0 = \cos\Psi_0, \quad \Psi_0 = \angle(\mathbf{s}_0, \mathbf{n})$$

$$\gamma_h = \cos\Psi_h, \quad \Psi_h = \angle(\mathbf{s}_h, \mathbf{n})$$

and $\mathbf{n}$ is a unit vector perpendicular to the entrance surface and directed within the crystal. In the Bragg case equation (13) has the same form except for the definition of v. The function v in (13) is the Riemann function given by the following conditions:

$$\begin{aligned} \frac{\partial^2 v}{\partial s_0\,\partial s_h} + 2\pi iK\frac{\partial(\beta_h v)}{\partial s_0} + \pi^2K^2C^2\chi_h\chi_{\bar{h}}v &= 0 \\ \partial v/\partial s_h &= -2\pi iK\beta_h v \quad \text{on } BP \\ \partial v/\partial s_0 &= 0 \quad \text{on } AP \\ v(P) &= 1. \end{aligned} \tag{14}$$

Now there are two ways in the literature of solving (14):

(1) Because of the ambiguity in the choice of $\mathbf{k}_0$ (and $\mathbf{k}_h$) we can assume that

$$\beta_h = 0, \tag{15}$$

which leads to a specially simple form of (12).

But this choice has the following consequences:

(a) From (11) it follows that

$$\eta_h = \eta_0 = \Theta_B.$$

This means that $\mathbf{k}_i$ must be directed parallel to the reflecting net planes. So the choice (15) *fixes* also the direction of $\mathbf{k}_i$.

(b) When $\mathbf{k}_i$ is fixed parallel to the reflecting net planes, then condition (1b) cannot be fulfilled in all cases and the boundary conditions remain in the form (5).

(c) The solution of (14) for $\beta_h = 0$ is

$$v = J_0(\zeta) \tag{16a}$$

with

$$\zeta = 2\pi K(\chi_h\chi_{\bar{h}})^{1/2}\{[s_0 - s_0(P)][s_h - s_h(P)]\}^{1/2}$$

or

$$v(x) = J_0[B(l^2-x^2)^{1/2}] \tag{16b}$$

with

$$B = 2\pi K|C|(\chi_h\chi_{\bar{h}})^{1/2}(\gamma_0\gamma_h)^{1/2}/\sin 2\Theta_B$$

where x is a coordinate along $BA = 2l$ with respect

to the middle of BA (Authier & Simon, 1968) and J_0 is the Bessel function of order 0. The solution for D_h becomes

$$D_h(P) = -(i\pi KC\chi_h\gamma_0/\sin 2\Theta_B) \times \int_{-l}^{l} D_0^{(a)}(x) \exp(-2\pi iTx) \times J_0[B(l^2-x^2)^{1/2}]\,dx \quad (17)$$

where

$$T = (\mathbf{K}_0 - \mathbf{k}_0)\mathbf{r}_e = T_r + iT_i. \quad (18)$$

The value T of the projection of the difference between the vectors $\mathbf{K}_0$ and $\mathbf{k}_0$ on the entrance surface is complex because of the complexity of $\mathbf{k}_0$.

(2) We can also assume that

$$T = 0. \quad (19)$$

This has the following consequences:

(a) Condition (19) is equivalent to condition (1b). Both can be fulfilled only if $\mathbf{k}_i$ is perpendicular to the entrance surface. So the choice (19) also *fixes* the direction of $\mathbf{k}_i$, but in a different way from before. The boundary condition (5) takes on a simpler form.

(b) From (11) it now follows that

$$\beta_h \neq 0.$$

(c) The solution of (14) in the case $\beta_h \neq 0$ is

$$v = J_0(\zeta) \exp\{-2\pi iK\beta_h[s_h - s_h(P)]\} \quad (20)$$

or

$$v(x) = J_0[B(l^2-x^2)^{1/2}] \times \exp[-2\pi iK\beta_h(-t/2\gamma_h + \gamma_0 x/\sin 2\Theta_B)]$$

where t is the distance of P from the surface.

The solution for D_h becomes

$$D_h(P) = -(i\pi KC\chi_h\gamma_0/\sin 2\Theta_B)\exp(i\pi K\beta_h t/\gamma_h) \times \int_{-l}^{l} D_0^{(a)}(x) \exp\left(\frac{-2\pi iK\beta_h\gamma_0 x}{\sin 2\Theta_B}\right) \times J_0[B(l^2-x^2)^{1/2}]\,dx. \quad (21)$$

If one takes into account the phase factor for the entire amplitude (that is the boundary conditions on the exit surface)

$$D_h^G(\mathbf{r}) = D_h(\mathbf{r}) \exp(-2\pi i\mathbf{k}_h\mathbf{r}) \quad (22)$$

and neglects complex factors not contributing to the intensity, (17) and (21) can be written in the form

$$D_h^G(P) = -(i\pi KC\chi_h\gamma_0/\sin 2\Theta_B) \times \int_{-l}^{l} D_0^{(a)}(x)A(x)J_0[B(l^2-x^2)^{1/2}]\,dx \quad (23)$$

with

$$A(x) = \exp\left\{-\frac{\mu t}{4}\left[\left(\frac{1}{\gamma_0}+\frac{1}{\gamma_h}\right)+\frac{x}{l}\left(\frac{1}{\gamma_0}-\frac{1}{\gamma_h}\right)\right]\right\}. \quad (24)$$

Thus, independent of the method chosen and of the fact that the direction of $\mathbf{k}_i$ is not arbitrarily chosen, the result for D_h^G is the same.

The same considerations can be applied to the Bragg case. Here Uragami (1969) presented an analytical solution of (3). For a perfect crystal the amplitude at the point x^Q on the entrance surface is equal to

$$D_h(x^Q) = -(\chi_h\gamma_0/\chi_{\bar{h}}|\gamma_h|)^{1/2} \times \int_{BA} \frac{J_1[B(x^Q-x)]}{x^Q-x} D_0^{(a)}(x)A(x) \times \exp(-2\pi iT_r x)\,dx \quad (25)$$

with

$$A(x) = \exp\left(-\frac{\mu}{2}\frac{\gamma_0-\gamma_h}{\sin 2\Theta_B}x\right) \quad (26)$$

$$T_r = K[\gamma_0\,\Delta\Theta + \chi_{0r}(\gamma_0-\gamma_h)/2\sin 2\Theta_B], \quad (27)$$

where $\Delta\Theta$ is the deviation angle of $\mathbf{K}_0$ from the exact Bragg condition.

5. Concluding remarks

When solving the Takagi-Taupin equations, the direction of the imaginary part of the wave vector $\mathbf{k}_i$ is not of such great ambiguity as widely assumed. Once the direction of $\mathbf{k}_i$ is fixed the special form of the boundary conditions (T equal or not equal to zero) for the amplitudes is also fixed and the question of whether or not $\mathbf{k}_0$ satisfies the continuity condition for the tangential component of the wave vector at the entrance surface is answered. Two choices for $\mathbf{k}_i$, $\mathbf{k}_i$ perpendicular to the entrance surface of the crystal or $\mathbf{k}_i$ parallel to the reflecting net planes, are the most convenient ones. In other cases neither T nor β_h equals zero and absorption enters the final solution through the imaginary parts of T and β_h simultaneously. It must be emphasized that only a $\mathbf{k}_i$ (or the component of $\mathbf{k}_i$) perpendicular to the entrance surface has a *concrete physical meaning*. In the case of refraction and reflection of a wave at an absorbing medium (Born & Wolf, 1965) the imaginary part of the wave vector $\mathbf{k}_i$ is directed perpendicular to the surfaces of constant amplitude which are planes parallel to the boundary and is proportional to the absorption coefficient. This also holds in the case of dynamical X-ray diffraction.

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Investigation of Short-Range Order in Ni–10 at.% Al Single Crystals by Diffuse X-ray Scattering

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Abstract

The diffuse scattering from single crystals of nominally Ni-10 at.% Al, quenched after heat treatment at 973 K, has been measured with Mo $K\alpha$ radiation. The data were analysed for the contributions due to short-range order and static atomic displacements. Consistency of the structural parameters as determined by three different methods, the separation methods of Georgopoulos-Cohen and Borie-Sparks and the least-squares method of Williams, is demonstrated for the first time. The variation of the first few short-range-order parameters α_{lmn} with the coordination shell *lmn* strongly resembles that of the $L1_2$ superstructure. Employing these parameters to model a short-range-ordered computer crystal with 13 104 atoms, a preference for configurations of the $L1_2$ superstructure type is found, though no Ni_3Al embryos are observed. The values of α_{lmn} rapidly reach those for a random solid solution.

1. Introduction

Nickel base superalloys are technologically important because of their favourable mechanical properties at temperatures up to about 1200 K. In many of these alloys, ordered Ni_3Al-type precipitates (γ' phase with $L1_2$ superstructure), coherent with the γ matrix, are responsible for high mechanical strength by impeding dislocation motion. Ni-Al solid solutions may be seen as a prototype of other superalloys that can be obtained by a partial substitution of the Ni and Al atoms.

Ordering and decomposition have been studied in Ni-Al alloys as well as in technological alloys by various methods, such as small-angle neutron scattering (Beddoe, Haasen & Kostorz, 1984), transmission electron microscopy (Gröhlich, Haasen & Frommeyer, 1982) and atom-probe field-ion microscopy (Wendt & Haasen, 1983). Earlier work is found in the references just quoted.

High-angle scattering has been employed to study local atomic arrangements in various binary Ni-rich alloys, such as Ni-Cr, Ni-Mo, Ni-Fe; for a review see Kostorz (1983). One high-angle scattering measurement on Ni-12·7 at.% Al was recently performed by Epperson & Fürnrohr (1983). The short-range-ordered state quenched in from 1323 K was analysed by the method introduced by Borie & Sparks (1971).

In the present study, single crystals of Ni with nominally 10 at.% Al were used for an investigation of short-range order. As this concentration is still within the γ region, the formation of γ' particles is avoided, and the short-range-ordering properties of the γ phase may be investigated. The method of Georgopoulos & Cohen (1977), most appropriate for the diffuse scattering of X-rays, is employed to analyse the data. This method also allows individual atomic displacements to be determined. Two other methods, the one proposed by Borie & Sparks (1971) and the least-squares method suggested by Williams (1972), have also been applied in order to evaluate the influence of various methods on the resulting structural parameters.

2. Experimental

A single crystal of Ni with nominally 10 at.% Al, about 6 cm long and 12 mm in diameter, was grown under an argon (5N7) atmosphere in a high-purity Al_2O_3 crucible by the Bridgman technique; the starting alloy was prepared from 99·99 at.% Ni and 99·999 at.% Al. The single crystal was homogenized in argon for 24 h at 1373 K and water quenched. Subsequently two slices about 3 mm thick, with a surface normal near the [119] direction, were spark

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