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Extended X-ray interbranch resonance concept for crystals with a one-dimensional deformation

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The X-ray interbranch resonance concept is extended to crystals with a one-dimensional deformation. The original approach, which proceeds from the Lagrange formalism, is developed. The principle of local 'lattice homogeneity' is also incorporated in the resonance concept. It postulates the local translation symmetry by lattice spacings in the vicinity of any point of X-ray trajectories. In this connection, interbranch scattering is considered as the process that violates this principle. The new interbranch effect is predicted for a strongly distorted crystal with thickness of the order of the interbranch extinction length. In this case, the interbranch wavefield oscillations related to the resonance interbranch splitting are suggested for the diffraction profile.

1. Introduction

X-ray dynamical diffraction by continuously deformed crystals is one of the most intricate physical problems. In past years, a deep insight into many diffraction phenomena in distorted crystals has been obtained with the help of extensive theoretical studies, reviewed by Authier (2005). Nevertheless, new theoretical investigations that could facilitate solution of the diffraction problem are still in development for X-ray diffraction techniques. This is of great importance in the further study of the interbranch scattering phenomenon, predicted by Penning (1966) and suggested by Authier (1967) to explain empirically topographic contrast observed for a single dislocation (Hart, 1963). These processes increase with increasing deformation, such that solution of the diffraction problem runs into serious difficulties. It is worth remarking that this phenomenon is caused by long-range strain fields but, in spite of this fact, it occurs in a sufficiently small part of a crystal. Bearing this in mind, it would be possible to use the interbranch effects for X-ray studies on a small spatial scale, which is of practical interest to advanced X-ray diffraction applications.

As is well known, the solid analytical foundations of interbranch processes were formulated by Balibar *et al.* (1983), who used the influence function for a crystal with uniform strain gradient. It was pointed out that the appropriate Green function should be separated into four parts, two of which can be considered as 'normal' wavefields (*i.e.* those predicted by the eikonal theory) and the two others are the 'new' ones, which are due to interbranch scattering. Based on the adiabatic invariance approach, a new view of the interbranch problem was given by Shevchenko & Pobydaylo (2003). They showed that, in the case of non-adiabatic variation of the crystal lattice, interbranch scattering can be treated as a beating process similar to quantum beats. It should be noted that the same interpretation of interbranch processes follows also from the optic approach to the dynamical theory of X-ray diffraction with deformed crystals, proposed by Mana & Palmisano (2004).

If X-ray interbranch beating is considered as the diffraction effect caused by a 'new' wavefield (Authier & Balibar, 1970), the resonance concept of X-ray interbranch scattering was developed for crystals with one-dimensional homogeneous bending (Shevchenko & Pobydaylo, 2005). In the present paper, this concept is extended to crystals with one-dimensional continuous deformation. The fundamental equations are derived from the original analytical approach. The novel features of the X-ray dynamical diffraction by strained crystals are also deduced from the dispersion analysis of the 'new' wavefields. In this connection, implication of the given results to diffractometry of strongly distorted crystals is discussed.

2. Results

According to the dynamical Takagi theory, the X-ray coherent wavefield inside a crystal with one-dimensional continuous distortion is described by the following equations:

$$\begin{cases} \dfrac{\mathrm{d}D_0(z)}{\mathrm{d}z} = \dfrac{i\pi}{\xi_g} D_g(z) & (1) \\ \dfrac{\mathrm{d}D_g(z)}{\mathrm{d}z} = \dfrac{i\pi}{\xi_g} D_0(z) + i\left[s + \mathbf{g}\dfrac{\mathrm{d}\mathbf{u}(z)}{\mathrm{d}z}\right] D_g(z), & (2) \end{cases}$$

where $D_{0,g}$, ξ_g, $\mathbf{u}(z)$ and s stand for the amplitudes of the transmitted and diffracted waves, the X-ray extinction length, a continuous displacement field which depends on the depth in the crystal z and the departure from Bragg's law of an incident wave, respectively. Moreover, we assume transmission geometry and consider the symmetric case without loss of generality for the further presentation.

With the help of the substitutions $D_{0,g} = \exp\{i \int q(z)\,\mathrm{d}z\}\tilde{D}_{0,g}$, where $q(z) = [s + \mathbf{g}\,\mathrm{d}\mathbf{u}(z)/\mathrm{d}z]/2$, equations (1) and (2) can be rearranged to the form:

$$\frac{\mathrm{d}^2\tilde{D}_{0,g}(z)}{\mathrm{d}z^2} + \left\{\left[\frac{\pi^2}{\xi_g^2} + q^2(z)\right] \pm \frac{i}{2}\mathbf{g}\frac{\mathrm{d}^2\mathbf{u}(z)}{\mathrm{d}z^2}\right\}\tilde{D}_{0,g}(z) = 0. \quad (3)$$

For a slightly deformed crystal, the solutions of equations (3) make up the modified Bloch waves, which were introduced by Kato (1963, 1964*a*,*b*) within the eikonal approach. It is necessary to point out that these waves describe into-a-branch scattering only. This means that the 'tie point' of a modified Bloch wave on the corresponding dispersion branch moves on the branch according to the local lattice orientation. In the case of a strong distortion, amplitude and phase modulation of the eikonal solutions of equations (3) takes place owing to interbranch scattering, therefore one can write the amplitudes $\tilde{D}_{0,g}(z)$ as follows:

$$\tilde{D}_{0,g}(z) = \sum_{j=1}^{2} \frac{A^j_{0,g}(z)}{[p(z)]^{1/2}} \exp\left\{i\frac{\pi}{\xi_g}\int Q^j_{0,g}(z)\,\mathrm{d}z\right\}. \quad (4)$$

Here $A^j_{0,g}(z)$ are the modulation amplitudes for the transmitted and diffracted waves associated with the appropriate dispersion branches; $p(z) = [1 + \eta^2(z)]^{1/2}$, where the deviation $\eta(z) = \omega + (\mathbf{g}\,\mathrm{d}\mathbf{u}/\mathrm{d}z)\xi_g/(2\pi)$ and $\omega = s\xi_g/(2\pi)$. The exponents in (4) are the eikonal functions such that

$$Q^1_{0,g}(z) = p(z) \pm i\xi_g \frac{\mathrm{d}\eta(z)/\mathrm{d}z}{2\pi p(z)} \quad \text{and} \quad Q^1_{0,g}(z) = -Q^2_{0,g}(z).$$

It should be noted that the eikonal solutions, which are the basis of the expansions of (4), can be easily obtained from (3) by using the non-dimensional quantity $\varepsilon(z) = (\xi_g/\pi)\,\mathrm{d}\eta(z)/\mathrm{d}z$ as the small parameter of the singular perturbation theory.

Thus, expressions (4) introduce a new representation of the dynamical diffraction by a distorted crystal, which is called the 'eikonal' representation. By application of such a representation, the study of interbranch processes, specified by the excitation errors, which are functions of the local orientation of the lattice, is simplified. This is attained due to reformulation of the dynamical Takagi theory, such that we pass from the fundamental equations for $D_{0,g}(z)$ to differential equations for $A^j_{0,g}(z)$, which describes only interbranch contributions. For this purpose, we employ the original approach based on the variational Lagrange formalism. In doing so, we consider the amplitudes $A^j_{0,g}(z)$ as indeterminate Lagrange multipliers and impose the following conditions on them:

$$\sum_{j=1}^{2}\left\{\frac{\mathrm{d}A^j_{0,g}(z)}{\mathrm{d}z} - \frac{A^j_{0,g}(z)}{2p(z)}\left[\frac{\mathrm{d}p(z)}{\mathrm{d}z} \mp (-1)^j\frac{\mathrm{d}\eta(z)}{\mathrm{d}z}\right]\right\} \times \exp\left[i\frac{\pi}{\xi_g}\int Q^j_{0,g}(z)\,\mathrm{d}z\right] = 0. \quad (5)$$

It should be noted that such conditions will provide the complete elimination of the into-a-branch contribution from the fundamental equations. Then, exploiting the conditions (5), we obtain the derivatives $\mathrm{d}\tilde{D}_{0,g}(z)/\mathrm{d}z$ as

$$\frac{\mathrm{d}\tilde{D}_{0,g}(z)}{\mathrm{d}z} = i\frac{\pi}{\xi_g}[p(z)]^{1/2}\sum_{j=1}^{2}(-1)^{j+1}A^j_{0,g}(z) \times \exp\left[i\frac{\pi}{\xi_g}\int Q^j_{0,g}(z)\,\mathrm{d}z\right]. \quad (6)$$

Substituting the expansions (4) and (6) into Takagi's equations (3), it is easy to rearrange them as

$$\sum_{j=1}^{2}(-1)^{j+1}\left\{\frac{\mathrm{d}A^j_{0,g}(z)}{\mathrm{d}z} + \frac{A^j_{0,g}(z)}{2p(z)}\left[\frac{\mathrm{d}p(z)}{\mathrm{d}z} \mp (-1)^j\frac{\mathrm{d}\eta(z)}{\mathrm{d}z}\right]\right\} \times \exp\left[i\frac{\pi}{\xi_g}\int Q^j_{0,g}(z)\,\mathrm{d}z\right] = 0. \quad (7)$$

Using the expressions (5) and (7) related to the same reciprocal-lattice vector, we make up their sum and difference combinations that yield the differential expressions for $A^2_{0,g}(z)$ and $A^1_{0,g}(z)$, respectively. Thus, one can obtain

$$\frac{\mathrm{d}A_0^{1,2}(z)}{\mathrm{d}z} = \frac{A_0^{2,1}(z)}{2p(z)}\left[\frac{\mathrm{d}p(z)}{\mathrm{d}z} \mp \frac{\mathrm{d}\eta(z)}{\mathrm{d}z}\right]\exp\left[\mp i\frac{2\pi}{\xi_g}\int Q_0^1(z)\,\mathrm{d}z\right] \quad (8)$$

$$\frac{\mathrm{d}A_g^{1,2}(z)}{\mathrm{d}z} = \frac{A_g^{2,1}(z)}{2p(z)}\left[\frac{\mathrm{d}p(z)}{\mathrm{d}z} \pm \frac{\mathrm{d}\eta(z)}{\mathrm{d}z}\right]\exp\left[\mp i\frac{2\pi}{\xi_g}\int Q_g^1(z)\,\mathrm{d}z\right]. \quad (9)$$

As is seen from equations (8) and (9), owing to the correct choice of the additional conditions for $A^j_{0,g}(z)$, this set of equations describes interbranch scattering solely. Indeed, the first equations in (8) and (9), which correspond to the '−' sign in the exponentials, describe only the energy transfer from 'branch 2' to 'branch 1', and the second equations, which correspond to the '+' sign in the exponentials, an inverse interbranch process. From the relation

$$\xi_g/(2\pi)\int \mathrm{d}z[\mathbf{g}/p(z)]\mathrm{d}^2\mathbf{u}/\mathrm{d}z^2 = \ln|\eta(z) + p(z)|,$$

equations (8) and (9) can be finally reduced to the following form after straightforward manipulation:

$$\frac{\mathrm{d}A_0^{1,2}(z)}{\mathrm{d}z} = -\frac{A_0^{2,1}(z)\exp[\mp(2i\pi/\xi_g)\int_0^z p(z)\,\mathrm{d}z]}{2p^2(z)\{\eta(0) \pm [1 + \eta^2(0)]^{1/2}\}}\frac{\mathrm{d}\eta(z)}{\mathrm{d}z} \quad (10)$$

$$\frac{\mathrm{d}A_g^{1,2}(z)}{\mathrm{d}z} = \frac{A_g^{2,1}(z)\exp\left[\mp(2i\pi/\xi_g)\int_0^z p(z)\,\mathrm{d}z\right]}{2p^2(z)\{\eta(0) \mp [1 + \eta^2(0)]^{1/2}\}}\frac{\mathrm{d}\eta(z)}{\mathrm{d}z}. \quad (11)$$

It should also be observed that these equations are valid in the case of any kind and strength of continuous deformations, which can induce interbranch crossover in the transmitted channel. In this case, it was shown by Penning (1966) that the drastic changes in the intensities of the transmitted and diffracted waves mean that the intensive 'new' wavefield propagates in the transmitted direction.

If a homogeneously bent crystal is assumed, equations (10) and (11) will be identical to the appropriate equations derived by Shevchenko (2003) from the 'lamellar-crystal' approach.

Bearing this in mind, the X-ray interbranch resonance concept developed for a bent crystal can be extended to a crystal with a one-dimensional deformation. In this connection, we consider the transmitted wavefields, which permit the 'jump' of the tiepoint. Then, assuming strong deformation and $|\omega| \gg 1$, one can obtain the approximate solutions of equations (10) in the vicinity of the point z_0 such that $\eta(z_0) = 0$:

$$A_0^{1,2}(z) = C_{1,2}^{+}(z_0)\exp[i(\Delta Q_0/2 \mp W_0)z] + C_{1,2}^{-}(z_0)\exp[i(-\Delta Q_0/2 \mp W_0)z], \qquad (12)$$

where

$$C_1^{\pm}(z_0) = \frac{\Delta Q_0/2 \pm W_0}{\Delta Q_0} \quad \text{and} \quad C_2^{\pm}(z_0) = \pm\frac{i\gamma_0}{\Lambda_0 \Delta Q_0}\exp(i\pi/2). \qquad (13)$$

Here $\gamma_0 = \exp[(2i\pi/\xi_g)\int_0^{z_0} p(z)\,\mathrm{d}z]$; $\Lambda_0 = 2\pi/\eta'(z_0)$, $W_0 = \pi/\xi_g$ and $\Delta Q_0 = 2[W_0^2 + (\pi/\Lambda_0)^2]^{1/2}$. In expressions (12) and (13), the signs '+' and '−' denote the 'upper' and 'lower' new branches, respectively. It should be noted that the phase factor $\exp(i\pi/2)$ is included in the amplitudes $C_2^{\pm}(z_0)$ as well, owing to the energy transfer from branch 1 to branch 2. It follows from (13) that the sharp changes of the amplitudes of the 'new' wavefields $C_{1,2}^{\pm}(z_0)$ occur under the condition

$$\pi/\Lambda_0 \geq W_0. \qquad (14)$$

When the reduced deformation parameter $\varepsilon(z) \gg 1$ and denoting $\varepsilon_0 = \varepsilon(z_0)$, we have $|C_{1,2}^{\pm}(z_0)| \to 1/2$ such that the behavior of the interbranch amplitudes is similar to the dependence of the amplitudes of transmitted and diffracted waves on departure of the incident wave near the exact conditions. Clearly, in analogy with Bragg's resonance, the quantity W can be interpreted as the 'deviation' parameter, which smooths out the resonance variations of the 'new' wavefields. Moreover, the quantity ΔQ_0 should be considered as the interbranch splitting, which determines the distance between the 'new' branches near z_0. Such splitting is the result of interbranch interchange, which is effective in the range Δz_D that specifies the X-ray dynamical diffraction in the deformed crystal. It is evident that, with the approximation $\eta(z) \approx \eta(z_0) + \eta'(z_0)z$, the value Δz_D can be estimated as Λ_0. As is obvious from (14), the resonance condition, which corresponds to the beginning of the interbranch process, has the form $\pi/\Lambda(z_0) = W(z_0)$. It is interesting to observe that this condition coincides with the limit of validity of the eikonal approximation of the dynamical diffraction theory, established by Authier & Balibar (1970).

In the vicinity of any point z_s, solutions of equations (10) evidently have the form

$$A_0^{1,2}(z) = C_{1,2}^{+}(z_s)\exp\left[\frac{i(\xi_g \Delta Q_s \mp 2\pi p_s)z}{2\xi_g}\right] + C_{1,2}^{-}(z_s)\exp\left[\frac{i(-\xi_g \Delta Q_s \mp 2\pi p_s)z}{2\xi_g}\right]. \qquad (15)$$

As appears from (15), the interbranch splitting $\Delta Q_s = [(2\pi p_s/\xi_g)^2 + (\eta_s'/p_s^2)^2]^{1/2}$ near z_s, where $p_s = p(z_s)$ and $\eta_s' = \eta'(z_s)$. Combining this with equations (10), one can obtain the following ratios for the amplitudes $C_{1,2}^{\pm}(z_s)$:

$$\frac{C_2^{\pm}(z_s)}{C_1^{\pm}(z_s)} = -i\frac{\pi p_s^2}{\xi_g \eta_s'}\gamma_s(\xi_g \Delta Q_s \mp 2\pi p_s), \qquad (16)$$

where the phase factor

$$\gamma_s = \exp\left[(2i\pi/\xi_g)\int_0^{z_s} p(z)\,\mathrm{d}z\right]\{\eta(0) - [1 + \eta^2(0)]^{1/2}\}^{-1}.$$

Analyzing expressions (16) and the eikonal solutions of the dynamical diffraction theory at infinity, we find that only the 'new' wavefield corresponding to 'branch 2^-' will propagate in the transmitted direction far from z_0. Using the considerations developed for a bent crystal by Shevchenko & Pobydaylo (2005), it is easy to obtain the asymptotic expression I_N for the intensity of this wavefield:

$$I_N = \left\{1 - \frac{1}{[1 + (\varepsilon_0/2)^2]^{1/2}}\right\}^2. \qquad (17)$$

Assuming $\varepsilon_0 \gg 1$, one can find from (17) the intensity $I_N = 1 - 4/\varepsilon_0$ that corresponds to the Penning–Polder limit associated with the interbranch crossover. However, with decreasing deformation, intensity I_N tends to zero as $(\varepsilon_0/2)^4$, such that it can be neglected within the eikonal approximation. It is important to emphasize that expression (17) is valid for deviation $|\omega| \gg 1$. At the same time, when $|\omega| \approx 1$, the local structure of the 'new' wavefields may manifest itself near the exit surface of a crystal. In this case, by using (14) one can obtain the following estimation:

$$t \approx \Lambda_0, \qquad (18)$$

where t is the crystal thickness.

Based on general physical arguments, we suggest that, under the condition (18), X-ray intensities will show the oscillations determined by the interbranch extinction length. However, the interbranch oscillations are damped or cannot be established if $t \gg \Lambda_0$ or $t \ll \Lambda_0$, respectively. For the sake of simplicity, we assume a bent displacement field $u(z) = \alpha z^2/(2R)$ oriented along the surface, where R and α are the radius of curvature and a constant describing deformation, respectively. Then, estimation (18) is rewritten as follows:

$$t \approx 2\xi_g/\varepsilon, \qquad (19)$$

where $\varepsilon = \alpha g \xi_g^2/(2\pi^2 R)$. As is well known, the strong bending, which causes the interbranch scattering, can be specified by large $\varepsilon \gg 1$. Supposing such an ε in (19), we can predict the interbranch oscillations of the X-ray intensities for a crystal with thickness considerably less than the X-ray extinction length and satisfying the relation (19). It is natural to expect that these oscillations may manifest themselves in Bragg peaks as 'fine structure', which indicates strong continuous deformations.

It is clear that the above suggestions given for X-rays will remain valid for electron diffraction by distorted crystals too. In this connection, we pay attention to the 'fine-structure' effect in electron diffraction patterns from icosahedral silver nanoclusters, which was reported by Reinhard *et al.* (1997). Furthermore, it was established that a satisfactory fit to this structure could not be obtained with the help of the kinematical diffraction theory. Bearing in mind that strong strain can be inherent to such clusters (MacKay, 1962; Howie & Marks, 1984), it would be possible to explain this effect as interbranch wavefield oscillations.

It is worth noting that the interbranch splitting can also be regarded as a fundamental physical phenomenon connected with the symmetry of physical space. This symmetry turns out to depend on the strength of deformation, which dictates the translation symmetry properties for small distortions. Indeed, assuming weak deformations, the value of the 'lamellar' Poynting vector is conserved independent of the method of dividing the crystal into infinitesimal 'lamellae'. This means that the local translation symmetry by lattice spacings, related to conservation of the wavefield energies, can be postulated in the eikonal approximation of dynamical theory. Clearly, similar considerations can also be applied to waves propagating in isotropic media with small inhomogeneity. Then, the local translation symmetry related to local homogeneity of space takes place in the vicinity of any point of the eikonal trajectory. Bearing this in mind and following Penning & Polder's (1961) notation of 'lattice inhomogeneity', we formulate the principle of local 'lattice homogeneity', which reflects the conservation of the eikonal invariants. Then, interbranch scattering can be considered as the process that violates the local 'lattice homogeneity' by splitting of the local dispersion modes. Evidently, this is analogous to taking off degeneracy in quantum-mechanical phenomena. It is known to consist in the separation of energy levels that is also accompanied by violation of the initial symmetry of the space.

One can hope that the presented results are of interest to clarify the physical discussion of the diffraction problem for a crystal with a continuous deformation. Moreover, the predicted interbranch features might be helpful to increase the diagnostic capabilities of the diffraction techniques so that the studies of 'thick' crystals progress. This implies the further development of the interbranch resonance concept and carrying out the appropriate experimental investigations.

3. Conclusions

Here we sum up the main results obtained in this work.

1. The X-ray interbranch resonance concept is generalized for crystals with a one-dimensional continuous deformation. By applying the original approach based on the Lagrange formalism, the fundamental equations are derived from Takagi's equations for it.

2. The local translation symmetry by lattice spacings is postulated in the vicinity of any point of the eikonal trajectories. In this connection, the principle of local 'lattice homogeneity', which expresses conservation of the eikonal invariants, is formulated. This principle is violated in the case of the interbranch interchange, which leads to the splitting of the branches of the local dispersion surface. It is noted that this is similar to taking off a degeneracy phenomenon in quantum mechanics.

3. The new interbranch effect is predicted for strongly deformed crystals with thickness considerably less than the X-ray extinction length ξ_g. It is supposed that the diffraction profile may show interbranch wavefield oscillations associated with the resonance splitting of the local dispersion modes. In this case, the crystal thickness must be of the order of the interbranch extinction length Λ_0.

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