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A Quantum Approach to X-ray Multiple Diffraction

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Abstract

Quantum theory is applied to X-ray multiple diffraction. Expressions for three-, four-, five- and general N-beam diffraction intensities are derived using the Green-function technique. The asymmetry effect on the diffraction intensity profiles resulting from the reflection phases is revealed in the derived expressions. A schematic representation of the Green function to simplify the multiple-beam intensity problem is also presented.

1. Introduction

X-ray multiple diffraction is a consequence of X-ray scattering from the three-dimensional periodic arrangement of atoms in crystals. This particular diffraction technique has versatile applications in many crystallography-related researches (*e.g.* Hildebrandt, 1982; Chang, 1984). One vital application is the determination of the X-ray reflection phase from the intensity asymmetry of a multiple diffraction line profile. A fairly complete list of references on this subject can be found in the recent review article of Chang (1987).

Multi-beam interaction has been described successfully in terms of kinematical theory (*e.g.* Moon & Shull, 1964; Zachariasen, 1967; Caticha-Ellis, 1969; Chang, 1982*a*; Shen, 1986) and dynamical theory (Saccocio & Zajac, 1965*a*, *b*; Ewald & Héno, 1968; Uebach & Hildebrandt, 1969; Afanas'ev & Kohn, 1977; Juretschke, 1982*a*, *b*; Høier & Marthinsen, 1983; Hummer & Billy, 1986). In terms of quantum mechanics, although the theoretical work of Kohler (1933), Molière (1939), Ashkin & Kuriyama (1966), Kuriyama (1967, 1968), Ohtsuki & Yanagawa (1966) and many others dealt mostly with two-beam interaction, their treatments could be extended to multi-beam cases. In this article, in order to give a deep insight into the connection between the multi-beam interaction and the X-ray reflection phases in a more fundamental fashion, we apply the quantum-field-theoretical approach of Ohtsuki & Yanagawa (1966) to derive expressions for multiple diffracted intensities for three-, four-, five- and general N-beam interactions. The Green functions usually used in many-body problems are employed. The influence of reflected phases on the diffraction processes is revealed *via* the intensity expressions and the schematic of the Green function.

2. Theoretical background

In order to derive in later sections the expressions for multiply diffracted intensities the theoretical approach of Ohtsuki & Yanagawa (1966) is outlined here as the theoretical background. With the notation of Ohtsuki & Yanagawa (1966) the interaction of X-rays and the crystal can be described by the Hamiltonian

$$\begin{aligned}\mathscr{H} = {} & \sum_{i=1}^{N} [(P_i^2/2m) + V(\mathbf{r}_i)] \\ & + \sum_{i=1}^{N} (e/mc)\,[\mathbf{A}(\mathbf{r}_i)\,.\,\mathbf{P}_i] \\ & + \sum_{i=1}^{N} (e^2/2mc^2)A^2(\mathbf{r}_i) \\ & + (1/8\pi)\int (E^2+H^2)\,\mathrm{d}\mathbf{r}. \qquad (1)\end{aligned}$$

The crystal is assumed to contain N electrons. The first and the last terms are the Hamiltonians of the crystal and the X-ray field, respectively. The second and the third terms are due to the crystal-field and field-field interactions. $V(\mathbf{r}_i)$ is the potential energy binding the electrons. $\mathbf{A}(\mathbf{r}_i)$ is the vector potential of the field at $\mathbf{r}_i$. The terms e, m and c have their usual meanings. $\mathbf{E}$ and $\mathbf{H}$ are the electric and magnetic fields. $\mathbf{P}_i$ and $\mathbf{r}_i$ are the momentum and coordinate for the ith electron. The vector potential $\mathbf{A}(\mathbf{r})$ can be expressed as a Fourier series,

$$\mathbf{A}(\mathbf{r}) = (4\pi)^{1/2}C\sum_{k,h} q_{k,h}\,e^{i\mathbf{k}_h\cdot\mathbf{r}}\,\hat{\boldsymbol{\eta}}_{k,h} \qquad (2)$$

where $\hat{\boldsymbol{\eta}}_{k,h}$ and $q_{k,h}$ are the polarization unit vector and the second quantized field variable, respectively. The wave vectors $\mathbf{k}$ and $\mathbf{k}_h$ of the incident and the scattered waves satisfy Bragg's law, $\mathbf{k}_h = \mathbf{k} + 2\pi\mathbf{h}$, where $\mathbf{h}$ is the reciprocal-lattice vector of the h reflection. As usual, denoting $P_{k,h}$ as a field variable conjugate to $q_{k,h}$ and introducing the creation and annihila-

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tion operators b^+ and b of X-ray photons, we obtain

$$\begin{aligned} q_{k,h} &= (\hbar/2W_{k,h})^{1/2}(b_{k,h}+b^+_{-k,-h}) \\ &\equiv (\hbar/2W_{k,h})^{1/2}B_{k,h} \end{aligned} \tag{3}$$

$$P_{k,h} = i(\hbar W_{k,h}/2)^{1/2}(-b_{k,h}+b^+_{-k,-h}) \tag{4}$$

with

$$W_{k,h} = ck_h. \tag{5}$$

In the following, we concentrate only on the elastic scattering of X-rays by electrons. The scattering can be treated as the transition from $|0\rangle$ state with a photon of k_g to $|0\rangle$ state with a k_h photon. $|0\rangle$ satisfies the relation

$$\left[\sum_i P_i^2/2m + V(r_i)\right]|0\rangle = E|0\rangle.$$

E is the eigen energy. If we consider the perturbation due to the second and third terms of (1), and assume $k_h \sim k_g$, the Hamiltonian takes the form (Ohtsuki & Yanagawa, 1966)

$$\begin{aligned} \mathscr{H} &= \sum_g \hbar W_{k,g}B_{-k,-g}B_{k,g} \\ &\quad -\tfrac{1}{2}\sum_{h,g} \hbar\Omega(\varphi_{h-g}+C_{h,g})B_{-k,-h}B_{k,g} \\ &= \sum_g \hbar W_{k,g}B_{-k,-g}B_{k,g} \\ &\quad -\tfrac{1}{2}\sum_{h,g} \hbar\Omega\phi_{h,g}B_{-k,-h}B_{k,g} \\ &= \sum_g \hbar(W_{k,g}-\tfrac{1}{2}\Omega\phi_{h,g})B_{-k,-g}B_{k,g} \\ &\quad + \sum_{\substack{h,g \\ h\neq g\neq 0}} -\tfrac{1}{2}\hbar\Omega\phi_{h,g}B_{-k,-h}B_{k,g} \\ &\equiv H_0 + H', \end{aligned} \tag{6}$$

where $\Omega = cK$,

$$\begin{aligned} C_{h,g} &= \frac{4\pi e^2}{\hbar m^2\Omega^2} \\ &\quad \times \sum_{n=0} \Bigg\{ \Bigg[\langle 0|\sum_i (\mathbf{P}_i \,.\, \hat{\boldsymbol{\eta}}_{k,h})e^{-i\mathbf{k}_h\,.\,\mathbf{r}_i}|n\rangle \\ &\quad \times \langle n|\sum_i (\mathbf{P}_i \,.\, \hat{\boldsymbol{\eta}}_{k,g})e^{i\mathbf{k}_g\,.\,\mathbf{r}_i}|0\rangle \Bigg] \\ &\quad \times \{[(E_n-E_0)/\hbar]-\Omega-i\delta\}^{-1} \\ &\quad + \Bigg[\Bigg\langle 0|\sum_i (\mathbf{P}_i \,.\, \hat{\boldsymbol{\eta}}_{k,g})e^{i\mathbf{k}_g\,.\,\mathbf{r}_i}|n \Bigg\rangle \\ &\quad \times \langle n|\sum_i (\mathbf{P}_i \,.\, \hat{\boldsymbol{\eta}}_{k,h})e^{-i\mathbf{k}_h\,.\,\mathbf{r}_i}|0\rangle \Bigg] \\ &\quad \times \{[(E_n-E_0)/\hbar]+\Omega-i\delta\}^{-1} \Bigg\}. \end{aligned} \tag{7}$$

$$\begin{aligned} \varphi_{h-g} &= (-4\pi e^2/m\Omega^2)(\hat{\boldsymbol{\eta}}_{k,h} \,.\, \hat{\boldsymbol{\eta}}_{k,g}) \\ &\quad \times \int \exp[-2\pi i(\mathbf{h}-\mathbf{g})\,.\,\mathbf{r}]\rho(\mathbf{r})\,\mathrm{d}\mathbf{r} \end{aligned} \tag{8}$$

$$\rho(\mathbf{r}) = \langle n(\mathbf{r})\rangle = \langle 0|\sum_i \delta(\mathbf{r}-\mathbf{r}_i)|0\rangle. \tag{9}$$

The quantity φ_{h-g} can be expressed in terms of the polarization factor P and the electric susceptibility χ_{h-g}:

$$\varphi_{h-g} = P\chi_{h-g}, \tag{10}$$

where

$$P = \hat{\boldsymbol{\eta}}_{k,h} \,.\, \hat{\boldsymbol{\eta}}_{k,g}. \tag{11}$$

χ_{h-g} is linearly proportional to the structure factor F_{h-g} of the $h-g$ reflection in the dipole approximation. The quantity $C_{h,g}$ represents the Hönl correction for the photoelectric processes. For wavelengths far from the absorption edges of the constituent atoms of the crystal, $C_{h,g}$ approaches zero, and $\phi_{h,g} \simeq \varphi_{h-g}$. Hereafter we will only consider situations in which $\phi_{h,g} \simeq \varphi_{h-g} = \varphi_{hg}$. Next, we introduce the Green function G as

$$G(k_h, t; k_g, t') \equiv -i\langle 0|T\mathscr{B}_{k,h}(t)\mathscr{B}_{-k,-g}(t')|0\rangle, \tag{12}$$

where T is a time order operator. $\mathscr{B}_{k,h}(t')$ is a Heisenberg operator defined as

$$\mathscr{B}_{k,h}(t') = e^{(i/\hbar)Ht'}B_{k,h}\,e^{-(i/\hbar)Ht'}. \tag{13}$$

When H' equals zero, only the incident wave exists in the crystal. The corresponding Hamiltonian is

$$\mathscr{H} = H_0 = \sum_g [\hbar W_{kg} - (\hbar/2)\Omega\phi_{00}]B_{-k,-g}B_{k,g}.$$

The Green function G_0 for this one-beam case is

$$\begin{aligned} &G_0(k_g, t-t') \\ &= \begin{cases} -i\exp\{-i[W_{k,g}-(\Omega/2)\phi_{00}](t-t')\}, \\ \qquad\qquad\qquad\qquad (t-t')>0 \\ 0, \qquad\qquad\qquad\quad (t-t')<0. \end{cases} \end{aligned} \tag{14}$$

The Fourier transformation for the time-dependent part of $G_0(K_g, t-t')$ is

$$G_0(k_g, \Omega) = \int_{-\infty}^{\infty} G_0(k_g, \tau)\exp(i\Omega\tau - \delta|\tau|)\,\mathrm{d}\tau \tag{15}$$

$$= \{\Omega - [W_{k,g}-(\Omega/2)\phi_{00}] + i\delta\}^{-1} \tag{16}$$

where $\tau = t - t'$. δ is an infinitesimal positive quantity. $G_0(k_g, \Omega)$ is the free Green function in which no X-ray scattering is involved. When interaction turns on, *i.e.* $H' \neq 0$, the Green function $G(k_h, t; k_g, t')$ is usually calculated by the formula (*e.g.* Fetter & Walecka, 1971)

$$\begin{aligned} &G(k_h, t; k_g, t') \\ &= \sum_{n=0}^{\infty} \frac{1}{n!}\left(\frac{1}{i\hbar}\right)^n \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\dots\int_{-\infty}^{\infty} \mathrm{d}t_1\,\mathrm{d}t_2\dots\mathrm{d}t_n \\ &\quad \times \langle 0|T\{\mathscr{B}_{k,h}(t)\mathscr{B}_{-k,-g}(t')H'(t_1) \\ &\quad \times H'(t_2)\dots H'(t_n)\}|0\rangle. \end{aligned} \tag{17}$$

The Fourier transform of $G(k_h, t; k_g, t')$ for the time-dependent part can be derived in the same way as G_0. For example, up to second order, $G(k_g, \Omega)$ takes the form

$$\begin{aligned} G(k, k, \Omega) &= G_0(k, \Omega) + G_0(k, \Omega)(\Omega^2/4)\phi_{h0}\phi_{0h} \\ &\quad + G_0(k_h, \Omega)G(k, k, \Omega) \qquad (18) \\ G(k_h, k, \Omega) &= G_0(k, \Omega)(\Omega/2)\phi_{h0}G_0(k_h, \Omega) \\ &\quad + G_0(k, \Omega)(\Omega^2/4)\phi_{0h}\phi_{h0} \\ &\quad + G_0(k_h, \Omega)G(k_h, k, \Omega). \qquad (19) \end{aligned}$$

The diagrams representing (18) and (19) are shown in Figs. 1(*a*) and (*b*).

The intensity of X-ray diffraction can be obtained, in a first-order approximation, as (Inkson, 1984)

$$I = |E(k_g)/E(k)|^2 = |G(k_g, \Omega)/G(k, \Omega)|^2, \qquad (20)$$

where $E(k)$ is the incident-wave-field amplitude and $E(k_g)$ the diffracted-wave-field amplitude. In the following we apply the above approach to two-, three-, four- and general N-beam diffraction intensities.

3. Derivation of the expressions for multiple-diffraction intensity

(*a*) *Two-beam case*

In this case, only the incident wave k and the diffracted wave k_h are involved. The Hamiltonian, according to (6), is

$$\begin{aligned} \mathscr{H} &= \hbar W_{k,h}B_{-k,-h}B_{k,h} + \hbar W'_k B_{-k}B_k \\ &\quad - \tfrac{1}{2}\hbar\Omega\phi_{h0}B_{-k,-h}B_k - \tfrac{1}{2}\hbar\Omega\phi_{0h}B_{-k}B_{k,h}. \qquad (21) \end{aligned}$$

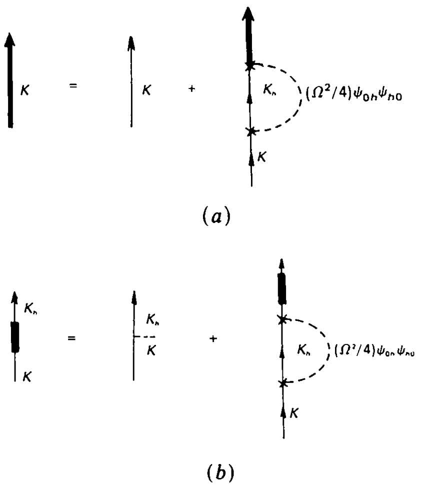

Fig. 1. Schematical representations of the Green functions of (*a*) equation (18) and (*b*) equation (19).

The Green function $G(k_h, k, \Omega)$ which has been expressed in (6) can be rewritten as

$$G(k_h, k, \Omega) = \tfrac{1}{2}\Omega\phi_{h0}G_0(k_h, \Omega)G(k, k, \Omega). \qquad (22)$$

Accordingly, the intensity I_h of the diffracted wave is

$$\begin{aligned} I_h &= |G(k_h, k, \Omega)/G(k, k, \Omega)|^2 \\ &= |\tfrac{1}{2}\Omega\phi_{h0}G_0(k_h, \Omega)|^2 \\ &= \frac{k^2}{4}\left(\frac{\chi_{h0}}{k-k'_h}\right)^2 |\hat{\sigma} + \cos 2\theta\hat{\pi}|^2 \\ &= \frac{k^2}{4}\left(\frac{\chi_{h0}}{k-k'_h}\right)^2 (1+\cos^2 2\theta), \qquad (23) \end{aligned}$$

where $k = \Omega/C$, $k'_h = (1/C)\{2_{kh} - \frac{1}{2}\Omega\phi_{00}\}$, and $\chi_{h-0} = \chi_{h0}$. This is exactly the expression of two-beam peak intensity that we usually find in the kinematical theory of X-ray diffraction.

(*b*) *Three-beam case*

In this case, three wave vectors, $\mathbf{k}$, $\mathbf{k}_h$ and $\mathbf{k}_g$ are involved. The Green function is expressed as

$$\begin{aligned} G(k_h, k_g, k, \Omega) &= G(k, k, k, \Omega)[d(\Omega/2)\varphi_{h0}G_0(k_h, \Omega) \\ &\quad + (\Omega^2/4)\varphi_{hg}\varphi_{g0}G_0(k_g, \Omega)G_0(k_h, \Omega)]. \qquad (24) \end{aligned}$$

The corresponding diagram is given in Fig. 2. The intensity of the h diffraction is

$$\begin{aligned} I &= |(\Omega/2)\varphi_{h0}G_0(k_h, \Omega) \\ &\quad + (\Omega^2/4)\varphi_{hg}\varphi_{g0}G_0(k_g, \Omega)G_0(k_h, \Omega)|^2 \\ &= \left|\frac{\chi_{h0}k}{2(k-k'_h)} + \frac{\chi_{hg}\chi_{g0}k^2}{4(k-k'_g)(k-k'_h)}\right|^2 \\ &= \frac{1}{4}\left|\frac{\chi_{h0}k}{k-k'_h}\right|^2 + \frac{1}{16}\left|\frac{\chi_{hg}\chi_{g0}k^2}{(k-k'_g)(k-k'_h)}\right|^2 \\ &\quad + \frac{|\chi_{0h}\chi_{hg}\chi_{g0}k^3|}{4(k-k'_h)^2(k-k'_g)}\cos(\delta_{hg}), \qquad (25) \end{aligned}$$

where $\delta_{hg} = \alpha_{hg} + \alpha_{g0} + \alpha_{0h}$ is the phase invariant of the structure-factor triplet $F_{h0}F_{hg}F_{g0}$ involved in the three-beam diffraction. α_{hg} is the phase of the structure factor F_{h-g}.

Clearly, the asymmetry of the diffraction intensity profile *versus* $k - k'_g$ arises because of the last term in (25). As proposed by Chang (1981, 1982*b*) for centrosymmetric crystals, the sign defined by the profile asymmetry is determined by the sign of $\cos \delta_{hg}$ and the sign of $(k - k'_g)$. This sign relation is consistent with (25).

(*c*) *Four-beam case*

An additional wave k_l is involved in the diffraction (k, k_h, k_g). There are two equally possible diffraction processes: one involves the interaction $k \rightarrow k_g \rightarrow k_l \rightarrow k_h$, the other involves $k \rightarrow k_l \rightarrow k_g \rightarrow k_h$. This is depicted in Fig. 3.

The last term in Fig. 3 represents the higher-order-contribution terms in the Green function. The diffraction intensity, therefore, can be calculated as

$$\begin{aligned} I_h = |&(\Omega/2)\varphi_{h0}G_0(k_h, \Omega) \\ &+(\Omega^2/4)\varphi_{g0}\varphi_{hg}G_0(k_g, \Omega)G_0(k_h, \Omega) \\ &+(\Omega^2/4)\varphi_{l0}\varphi_{hl}G_0(k_l, \Omega)G_0(k_h, \Omega) \\ &+(\Omega^3/8)\varphi_{g0}\varphi_{lg}\varphi_{hl}G_0(k_g, \Omega)G_0(k_h, \Omega)G_0(k_l, \Omega) \\ &+(\Omega^3/8)\varphi_{l0}\varphi_{gl}\varphi_{hg}G_0(k_l, \Omega) \\ &+G_0(k_g, \Omega)G_0(k_h, \Omega)|^2. \end{aligned} \tag{26}$$

The expansion of (26) is very lengthy and cumbersome. In order to reveal the effect of the interactions among the diffracted beams on the intensity I_h, the interaction terms involving the product of χ's are represented as the polygons shown in Fig. 4. For simplicity, only centrosymmetric crystals are considered, for which $\chi_{g-h} = \chi^*_{h-g}$. Among the terms $(a) \rightarrow (i)$ of Fig. 4, (c)–(f) involve self interactions, denoted as $g \rightleftarrows h$. Their interaction strength is proportional to $|\chi_{gh}|^2$, which is phase insensitive. Therefore, the diffraction intensity I_h depends only on four triplet phases, $\alpha_{hg} + \alpha_{g0} + \alpha_{0h}$, $\alpha_{hl} + \alpha_{l0} + \alpha_{0h}$, $\alpha_{hg} + \alpha_{gl} + \alpha_{lh}$ and $\alpha_{gl} + \alpha_{l0} + \alpha_{0g}$, and three quartet phases, $\alpha_{hl} + \alpha_{lg} + \alpha_{g0} + \alpha_{0h}$, $\alpha_{hg} + \alpha_{gl} + \alpha_{l0} + \alpha_{0h}$ and $\alpha_{0l} + \alpha_{lh} + \alpha_{hg} + \alpha_{g0}$. By definition, $|\varphi| < 1$. If the intensity near the exact four-beam position is concerned, the triplets $\varphi_{hg}\varphi_{g0}\varphi_{0h}$ and $\varphi_{hl}\varphi_{l0}\varphi_{0h}$ have a first-order effect and the quartets $\varphi_{hl}\varphi_{lg}\varphi_{g0}\varphi_{0h}$, $\varphi_{hg}\varphi_{gl}\varphi_{l0}\varphi_{0h}$ and $\varphi_{0l}\varphi_{lh}\varphi_{hg}\varphi_{g0}$ have a second-order effect on the diffraction intensity I_h. These results are in agreement with those obtained from the dynamical theory (Chang, Hong, Luh, Pan, Tang & Sasaki, 1988).

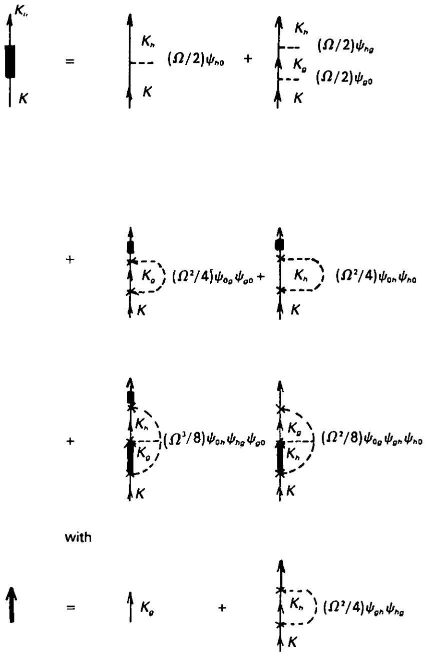

Fig. 2. Schematic of the Green function $G(k_h, k_g, k, \Omega)$ for three-beam interaction [equation (24)].

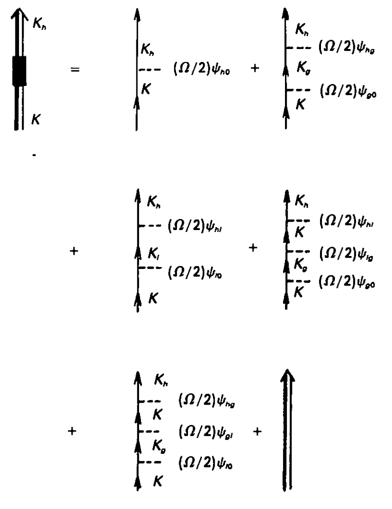

Fig. 3. Simplified schematical representation of the Green function for four-beam interaction.

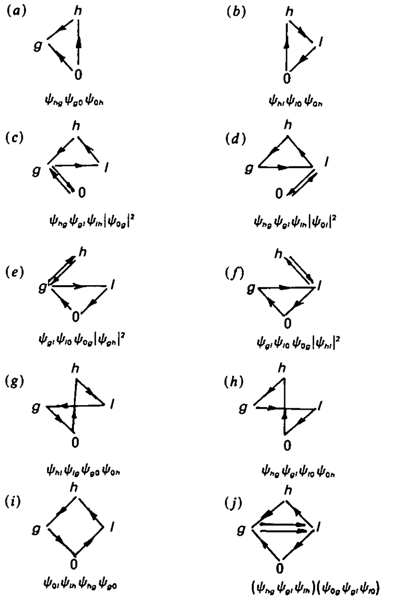

Fig. 4. Four-beam interactions (structure-factor multiplets) involved in equation (26).

(*d*) *N-beam case*

For a general N-beam case, there are N waves, k, k_h, k_1, $k_2, \ldots, k_{N-2}$, involved. The Green-function diagram is given in Fig. 5. The notation $P_{ijk}(N-2)$ means the permutation of (ijk) with i, j and k taken from the $N-2$ wave vectors k_1, k_2, $\ldots$, k_{N-2}. For simplicity, let us define $P_i(N-2) = P_1(N-2, 1)$, $P_{ij}(N-2) = P(N-2, 2)$, $P_{ijk}(N-2) = P(N-2, 3)$ *etc.* There are $n!^{N-2}_{\;n}C$ terms of $P(N-2, n)$, where n is the number of the subscripts of $P_{ijk\ldots z}(N-2)$. C is the usual notation of combination, *i.e.* ${}^{N}_{n}C = N!/[n!(N-n)!]$.

The corresponding Green function takes the form

$$\begin{aligned} G_N = {} & \overbrace{G(k, k, \ldots, k, \Omega)}^{N \text{ terms}} \Big\{ (\Omega/2)\varphi_{h0} G_0(k_h, \Omega) \\ & + \sum_{n=1}^{N-2} P_n^{(N-2)} [(\Omega/2)^{n+1} \varphi_{hs_1} \varphi_{s_1 s_2} \ldots \varphi_{s_{n-1} s_n} \\ & \times G_0(k_h, \Omega) G_0(k_{s_1}, \Omega) \\ & \times G_0(k_{s_2}, \Omega) \ldots G_0(k_{s_n}, \Omega)] \Big\} \\ = {} & G(k, k, \ldots, k, \Omega) A_G(N). \end{aligned} \tag{27}$$

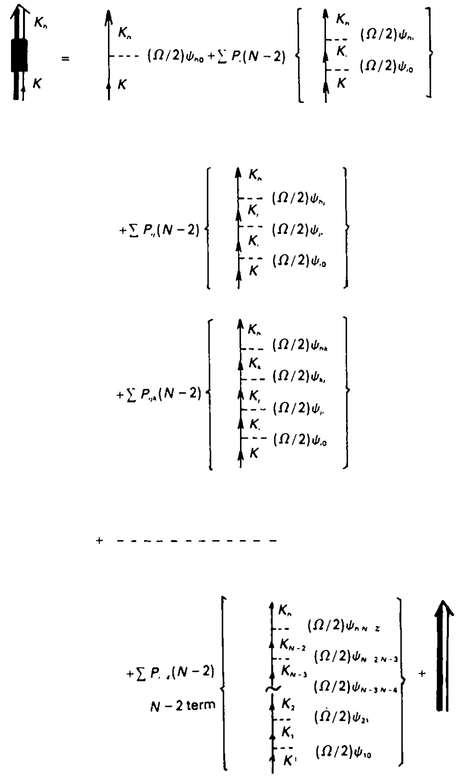

Fig. 5. Simplified schematical representation of the Green function for a general N-beam interaction.

The intensity $I_h(N)$ of the N-beam diffraction can then be expressed as

$$I_h(N) = |A_G(N)|^2. \tag{28}$$

4. Discussion and concluding remarks

In the course of deriving the intensity expression for the h reflection in a general N-beam case, the higher-order φ multiplets, which involve φ^{2N+1} or higher-order φ's, are neglected. Since the magnitude of $|\varphi|$ is much smaller than unity for X-rays, this omission of the $(2N+1)$th and higher-order terms has a negligible effect on $I_h(N)$.

The expression for $I_h(N)$ can be simplified in cases where very weak or space-group-forbidden reflections are involved. As examples, the following two cases are presented:

(*a*) *Modified four-beam case* $(0, h, g, l)$ *with* $\varphi_{gl} = 0$

The h diffracted intensity $I_h(4)$ can be written as

$$\begin{aligned} I_h(4) = {} & |(\Omega/2)\varphi_{h0} G_0(k_h, \Omega) \\ & + (\Omega^2/4)\varphi_{g0}\varphi_{hg} G_0(k_g, \Omega) G_0(k_h, \Omega) \\ & + (\Omega^2/4)\varphi_{l0}\varphi_{hl} G_0(k_l, \Omega) G_0(k_h, \Omega)|^2. \end{aligned} \tag{29}$$

Two triplets, $\varphi_{0h}\varphi_{hg}\varphi_{g0}$ and $\varphi_{0h}\varphi_{hl}\varphi_{l0}$ and one quartet, $\varphi_{hl}\varphi_{l0}\varphi_{0g}\varphi_{gh}$, are involved in (29).

(*b*) *Modified five-beam case* $(0, h, g, l, m)$ *with* $\varphi_{m0} = \varphi_{mh} = \varphi_{gl} = 0$

The h diffracted intensity is

$$\begin{aligned} I_h(5) = {} & |(\Omega/2)\varphi_{h0} G_0(k_h, \Omega) \\ & + (\Omega^2/4)\varphi_{g0}\varphi_{hg} G_0(k_g, \Omega) G_0(k_h, \Omega) \\ & + (\Omega^4/4)\varphi_{l0}\varphi_{hl} G_0(k_l, \Omega) G_0(k_g, \Omega) \\ & + (\Omega/2)^4 \varphi_{g0}\varphi_{mg}\varphi_{lm}\varphi_{hl} G_0(k_g, \Omega) \\ & \times G_0(k_m, \Omega) G_0(k_l, \Omega) G_0(k_h, \Omega) \\ & + (\Omega/2)^4 \varphi_{l0}\varphi_{ml}\varphi_{gm}\varphi_{hg} G_0(k_l, \Omega) \\ & \times G_0(k_m, \Omega) G_0(k_g, \Omega) G_0(k_h, \Omega)|^2. \end{aligned} \tag{30}$$

Equation (30) involves two triplets, $\varphi_{0h}\varphi_{hg}\varphi_{g0}$ and $\varphi_{oh}\varphi_{hl}\varphi_{lo}$, three quartets, $\varphi_{hl}\varphi_{l0}\varphi_{0g}\varphi_{gh}$, $\varphi_{0g}\varphi_{gm}\varphi_{ml}\varphi_{l0}$ and $\varphi_{gh}\varphi_{hl}\varphi_{lm}\varphi_{mg}$, and two quintets, $\varphi_{0h}\varphi_{hg}\varphi_{gm}\varphi_{ml}\varphi_{l0}$ and $\varphi_{0h}\varphi_{hl}\varphi_{lm}\varphi_{mg}\varphi_{g0}$.

In determining the sign of cos δ_{hg} from a multiple diffraction profile, the tail of the profile is important, since the intensities off the exact N-beam point provide most information about cos δ_{hg}. In this case the two-beam intensity I_h, proportional to χ_{0h}, is dominant in the diffracted intensity. The interaction φ_{0h} should play the major role in affecting the $I_h(N)$. If one takes this fact into account, the dominant terms in $I_h(N)$ are those triplets involving φ_{0h}. This agrees with the experimental results on phase determination from high-order multiple diffraction (Chang, Hong, Luh, Pan, Tang & Sasaki, 1988).

It should be noted that the derivation of $I_h(N)$ is a kinematical treatment. However, the use of Green functions is a dynamical approach. For example, the pole of the Green function gives the dispersion equation (Ohtsuki & Yanagawa, 1966). The real part of the equation describes the relation between the wave vectors inside the crystal and the angular deviation of the incident beam from the Bragg angle. The imaginary part of the dispersion relation defines the absorption coefficient. By solving the dispersion equation and introducing appropriate boundary conditions, the diffraction intensity $I_h(N)$ can be calculated dynamically.

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Quasiperiodic Tilings: a Generalized Grid-Projection Method

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Abstract

We generalize the grid-projection method for the construction of quasiperiodic tilings. A rather general fundamental domain of the associated higher-dimensional lattice is used for the construction of the acceptance region. The arbitrariness of the fundamental domain allows for a choice which obeys all the symmetries of the lattice, which is important for the construction of tilings with a given non-trivial point-group symmetry in Fourier space. As an illustration, the construction of a two-dimensional quasiperiodic tiling with 12-fold orientational symmetry is described.

0. Introduction

Interest in non-periodic tilings first arose from problems in mathematical logic (Wang, 1965; Robinson, 1971). However, since Penrose's invention of his well known non-periodic tilings (Penrose, 1974, 1979; Gardner, 1977), the motivation has changed to the study of geometrical questions related to such tilings. J. Conway (see Gardner, 1977) and N. G. de Bruijn (1981) have played a dominant role in this field.

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