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A Fourier optics approach to the dynamical theory of X-ray diffraction – perfect crystals

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A new formalism is presented concerning the dynamics of X-rays in crystals. It is based on Takagi's equations and Fourier optics; it also offers an alternative to the usual Ewald–von Laue approach. The article does not give new results but shows a new way to formulate the dynamical theory of X-ray diffraction. In addition, it proposes a novel description of X-ray propagation based on the analogy between the dynamics of X-rays in crystals and that of two-level quantum systems.

1. Introduction

Our article is based on an original and undeservedly neglected paper by Kato (1973), the reading of which will facilitate the understanding of our article. Following Kato, we represent the dynamics of X-rays in crystals in reciprocal space and describe X-ray propagation in terms of an optical transfer function. This is the usual approach to dynamical problems in quantum mechanics (Griffiths & Steinke, 2001) and it is known as Fourier optics in visible optics (Goodman, 1968). Additionally, we represent the crystal field as a two-component state vector to make a matrix formulation of the field dynamics possible (Kato, 1962). This simplifies the study of multicrystal systems, for instance of interferometers, the transfer function of which can be built step-by-step by assembling simpler elements (Davis, 1996; Mana & Vittone, 1997; Shvyd'ko *et al.*, 2003). We provide a unified treatment of the manifold aspects of X-ray diffraction (coplanar *versus* non-coplanar reflections, transmission *versus* reflection, plane *versus* spherical waves, perfect *versus* distorted crystals, coherent *versus* incoherent waves), further insight into the approximations adopted and additional methods to solve dynamical problems. From a didactic point of view, we particularly emphasize the cross talk between different specific mainstreams of physics.

In the free space, provided polarization is not considered, plane waves whose wavevectors belong to any point on the dispersion surface (a sphere with ω/c radius) are monochromatic wavefields, that is, they are a set of basis functions for Maxwell's equations. Beams are wavefield superpositions. Essentially, in this superposition wavevectors are grouped within a small region of the dispersion surface, so that the field distribution in the observation plane is a wavepacket belonging to the $\mathcal{L}^2(\mathbb{R}_2)$ Hilbert vector space of square-integrable two-variable functions. From this point of view, the specific set of basis functions selected to represent the field distribution depends on consideration of convenience: direct and reciprocal spaces are different frameworks to derive and study the same dynamics. They correspond to the time and frequency domains in electrical engineering. In optics, information is of a spatial nature, whereas in electrical engineering it is of a temporal nature; however, the mathematics necessary to describe its propagation is the same. Ever closer is the resemblance with quantum mechanics: when the paraxial approximation is adopted, the propagation equation (the parabolic approximation of the Helmoltz equation) is nothing else than the Schrödinger equation in quantum mechanics.

The dynamical theory of X-ray diffraction in crystals (Authier, 2001) takes account of the collective interaction of atoms with X-rays. As a result, wavevectors fall into continuous bands (dispersion surface branches) separated by forbidden gaps. Wavefields (Ewald waves) are superpositions of plane waves propagating in directions that are coupled by the reciprocal vectors of the crystal. In the simplest case, they are the superposition of two plane waves coupled by any reciprocal vector. As occurs in the free space, the wavefields identified by the same reciprocal vector and belonging to a small region of the dispersion surface form beams. A crystal field resembles a quantum system in which, among the many eigenstates, only two are considered, provided that all couplings are neglected, except those between the pair of interest. Two-level systems can be described as falling into the same mathematical form, irrespective of whether the two coupled states are spin states of an electron, energy levels of an atom, polarization states of a photon or branches of the dispersion surface of a crystal field (Cohen-Tannoudji *et al.*, 1977; Silverman, 1994). From a mathematical point of view, wavefields define the Hilbert space $V_2 \otimes \mathcal{L}^2(\mathbb{R}_2)$, where V_2 is a two-dimensional vector space (the space of the dispersion-surface branches) and $\mathcal{L}^2(\mathbb{R}_2)$ is the space of the square-integrable two-variable functions.

In §2, we anticipate our approach in the simplest case of a vacuum. We shall extend these concepts to crystal fields in the subsequent sections, presupposing a solid foundation in Fourier transform, linear operators in Hilbert spaces and the

representation theory. Throughout this paper, we confine our attention to perfect crystals. However, in §3, we derive the master equation for X-ray diffraction for application to the general case of a deformed crystal. In formulating the theory, we used vector algebra extensively and, since vector algebra makes no difference between two or three dimensions, we considered the most general case of a non-coplanar reflection. We emphasize that approximations reducing the master equation to Takagi's equations are nothing else than first-order approximations. After giving a canonical form to Takagi's equations (§5), in §6 we present a novel way to solve them. In our approach, transmission and reflection are treated simultaneously, our solution not making a distinction between them. In §7, we discuss a few examples, derive the amplitudes of reflected and refracted waves in Laue and Bragg geometries, and show how the symmetry between transmission and reflection is broken by boundary conditions. Table 1 in Appendix *A* gives a list of the main symbols.

2. Free-space propagation of X-rays

Before studying the dynamics of X-rays in crystals, we investigate the simplest case of propagation in a homogeneous medium. Therefore, we introduce the monochromatic wavepacket

$$\mathbf{D}(\mathbf{r};\, t) = \mathbf{D}(\mathbf{r}) \exp(-\mathrm{i}\omega t), \tag{1}$$

where $\mathbf{D}(\mathbf{r})$ is the electric displacement and ω the angular frequency. From Maxwell's equations we obtain

$$\begin{aligned} \Delta\mathbf{D} + K^2\mathbf{D} + \nabla \times (\nabla \times \chi\mathbf{D}) &= 0 \\ \nabla\mathbf{D} &= 0, \end{aligned} \tag{2}$$

where $\chi \ll 1$ is dielectric susceptibility and $K = \omega/c$. Let us introduce the Fourier transform (angular spectrum)

$$\tilde{\mathbf{D}}(\mathbf{k}) = \int_{-\infty}^{+\infty} \mathbf{D}(\mathbf{r}) \exp(\mathrm{i}\mathbf{k}\mathbf{r})\, \mathrm{d}\mathbf{r} \tag{3}$$

and rewrite (2) by using $\nabla\mathbf{D} \rightarrow -\mathrm{i}\mathbf{k}\tilde{\mathbf{D}}$, $\Delta\mathbf{D} \rightarrow -k^2\tilde{\mathbf{D}}$ and $\nabla \times \mathbf{D} \rightarrow -\mathrm{i}\mathbf{k} \times \tilde{\mathbf{D}}$. Hence, in the reciprocal space, (2) is

$$\begin{aligned} k^2\tilde{\mathbf{D}} - K^2\tilde{\mathbf{D}} + \mathbf{k} \times (\mathbf{k} \times \widetilde{\chi\mathbf{D}}) &= 0 \\ \mathbf{k}\tilde{\mathbf{D}} &= 0 \end{aligned} \tag{4}$$

and, by using $\chi = \chi_o = \text{constant}$ and $\mathbf{k} \times (\mathbf{k} \times \tilde{\mathbf{D}}) = \mathbf{k}(\mathbf{k}\tilde{\mathbf{D}}) - k^2\tilde{\mathbf{D}}$, we can also write

$$[(1 - \chi_o)k^2 - K^2]\tilde{\mathbf{D}}(\mathbf{k}) = 0. \tag{5}$$

Equation (5) has non-trivial solutions only if $(1 - \chi_o)k^2 - K^2 = 0$ (dispersion equation), which defines the dispersion surface for X-rays in the free space.

We confine our study to waves whose angular spectrum is limited within a small region of the dispersion surface centered around $\mathbf{k}_o$, where $k_o^2 = (1 + \chi_o)K^2$. If we set $\mathbf{k} = \mathbf{k}_o + \mathbf{p}$ and $\tilde{\mathbf{D}}(\mathbf{k}_o + \mathbf{p}) = \tilde{\mathbf{D}}_o(\mathbf{p})$, which is equivalent to shifting the reciprocal-space origin to $\mathbf{k}_o$ (Kato, 1973), the field dynamics is

$$[(1 - \chi_o)(\mathbf{k}_o + \mathbf{p})^2 - K^2]\tilde{\mathbf{D}}_o(\mathbf{p}) = 0, \tag{6}$$

where

$$\mathbf{D}(\mathbf{r}) = \mathbf{D}_o(\mathbf{r}) \exp(-\mathrm{i}\mathbf{k}_o\mathbf{r}), \tag{7}$$

as proved by calculating the inverse Fourier transform of $\mathbf{D}(\mathbf{k}_o + \mathbf{p})$. In order to approximate the wave equation (6), we remember that $|p| \ll K$. Subsequently, we expand the dispersion equation in series and neglect all terms but those of the first order,

$$(1 - \chi_o)(\mathbf{k}_o + \mathbf{p})^2 - K^2 \approx 2\mathbf{p}\mathbf{k}_o = 2k_o p_o, \tag{8}$$

where $p_o = \mathbf{p}\hat{\mathbf{k}}_o$. Consequently, the linear approximation of (6) is $p_o\tilde{\mathbf{D}}_o(\mathbf{p}) = 0$ and the dispersion surface is approximated by the plane $p_o = 0$, which is tangent, in $\mathbf{k} = \mathbf{k}_o$, to the sphere $k^2 = (1 + \chi_o)K^2$. In order to make the field dynamics explicit, we set $\mathrm{Im}(\mathbf{k}_o)$ along the z axis. Then, we introduce the wavevector

$$\mathbf{K}_o = K\widehat{\mathrm{Re}(\mathbf{k}_o)} = K(\gamma_o\hat{\mathbf{z}} + \sqrt{1 - \gamma_o^2}\hat{\mathbf{x}}_o), \tag{9}$$

where $\gamma_o = \hat{\mathbf{K}}_o\hat{\mathbf{z}} = \cos\psi_o$ is a direction cosine and the positive z direction is taken in such a way that $\gamma_o > 0$. Since $|\chi_o| \ll 1$, when $\gamma_o \gg 0$, the dispersion equation ensures that $\mathbf{k}_o \approx \mathbf{K}_o$. In fact,

$$k_o^2 = \mathrm{Re}^2(\mathbf{k}_o) - \mathrm{Im}^2(\mathbf{k}_o) + 2\mathrm{i}\mathrm{Re}(\mathbf{k}_o)\mathrm{Im}(\mathbf{k}_o), \tag{10}$$

so that, provided $|\widehat{\mathrm{Re}(\mathbf{k}_o)}\widehat{\mathrm{Im}(\mathbf{k}_o)}| = \gamma_o \gg 0$, $k_o^2 = (1 + \chi_o)K^2$ implies $|\mathrm{Im}(\mathbf{k}_o)| \ll K$. A γ_o value near to zero indicates that the z axis has been ill chosen. If we, additionally, set $\mathbf{p} = p_z\hat{\mathbf{z}} + \mathbf{q}$, p_o can be approximated by

$$p_o \approx \mathbf{p}\hat{\mathbf{K}}_o = \gamma_o p_z + q_o\sqrt{1 - \gamma_o^2}, \tag{11}$$

where $q_o = \mathbf{q}\hat{\mathbf{x}}_o$. Hence, (6) can be approximated as

$$\mathrm{i}\partial_z\tilde{\mathbf{D}}_o(\mathbf{q};\, z) = -q_o \tan\psi_o\tilde{\mathbf{D}}_o(\mathbf{q};\, z), \tag{12}$$

where $\mathrm{i}\partial_z$ is the direct-space representation of the p_z operator, $\tan\psi_o = \sqrt{1 - \gamma_o^2}/\gamma_o$,

$$\tilde{\mathbf{D}}_o(\mathbf{q};\, z) = \int_{-\infty}^{+\infty} \mathbf{D}_o(\mathbf{x};\, z) \exp(\mathrm{i}\mathbf{q}\mathbf{x})\, \mathrm{d}\mathbf{x} \tag{13}$$

is the two-dimensional Fourier transform of the field distribution over the observation plane and $\mathbf{r} = z\hat{\mathbf{z}} + \mathbf{x}$. The solution of (12) is

$$\tilde{\mathbf{D}}_o(\mathbf{q};\, z) = \tilde{U}(\mathbf{q};\, z)\tilde{\mathbf{D}}_o(\mathbf{q};\, 0), \tag{14}$$

where the reciprocal-space representation of the propagation operator, that is, the optical transfer function from $z = 0$ to z, is

$$\tilde{U}(\mathbf{q};\, z) = \exp(\mathrm{i}q_o z \tan\psi_o). \tag{15}$$

The propagation in the direct space is given by the inverse Fourier transform of (14). Hence, by orientation of the x_1 axis along $\mathbf{x}_o$,

$$\mathbf{D}_o(\mathbf{x};\, z) = \mathbf{D}_o(x_1 - z\tan\psi_o,\, y;\, 0). \tag{16}$$

The above equation shows that the linear approximation of the wave equation corresponds to geometric optics: the field distribution propagates without spreading in the $\hat{\mathbf{K}}_o$ direction.

In (12), free-space diffraction, *e.g.* the spreading of wavepackets, is left out. In order to recover diffraction, we must expand (8) up to the second order,

$$(1-\chi_o)(\mathbf{k}_o+\mathbf{p})^2-K^2\approx 2k_op_o+p^2, \tag{17}$$

so that the dispersion surface is approximated by $2k_op_o+p^2=0$. Since $|p|\ll K$, (17) implies $|p_o|\ll|p_\perp|$ (otherwise $|k_op_o|\gg|p|^2$ and the field dynamics is described by the first-order approximation), where $p_\perp$ is the component of $\mathbf{p}$ orthogonal to $\mathbf{k}_o$. Hence, $p^2\approx p_\perp^2$ and we can approximate the dispersion surface by the paraboloid $p_o=-p_\perp^2/(2k_o)$, which is tangent in $\mathbf{k}=\mathbf{k}_o$ to the sphere $k^2=(1+\chi_o)K^2$. Consequently, (6) is approximated by

$$-2k_op_o\tilde{\mathbf{D}}_o(\mathbf{p})=p_\perp^2\tilde{\mathbf{D}}_o(\mathbf{p}). \tag{18}$$

In order to make field dynamics explicit, we consider again (9) and, for the sake of simplicity, we set the z axis parallel to $\mathbf{K}_o$. Hence, $\gamma_o=1$, $\tan\psi_o=0$, $p_o\approx p_z$, $p_\perp\approx q$ and

$$2\mathrm{i}K\partial_z\tilde{\mathbf{D}}_o(\mathbf{q};z)=-q^2\tilde{\mathbf{D}}_o(\mathbf{q};z), \tag{19}$$

which is the parabolic approximation of the wave equation. In the direct space, (19) is

$$2\mathrm{i}K\partial_z\mathbf{D}_o(\mathbf{x};z)=\Delta\mathbf{D}_o(\mathbf{x};z), \tag{20}$$

where the operator $-\Delta$ is the direct-space representation of q^2. Finally, the optical transfer function from $\tilde{\mathbf{D}}_o(\mathbf{q};0)$ to $\tilde{\mathbf{D}}_o(\mathbf{q};z)$ is

$$\tilde{U}(\mathbf{q};z)=\exp(\mathrm{i}q^2z/2K). \tag{21}$$

2.1. Remarks on complex vectors

X-ray dynamics in crystals is described by complex wavevectors. We summarize here a few definitions, to clarify the notations we are using. A complex vector space is a vector space whose field of scalars is the set of complex numbers. Given any vector pair $\mathbf{a}$ and $\mathbf{b}$, in addition to the Hermitian inner product (scalar product) $\mathbf{ab}^*$ (the asterisk indicates complex conjugation), we introduce the non-Hermitian inner product $\mathbf{ab}$. When $\mathbf{b}=\mathbf{a}$, we indicate the product $\mathbf{aa}$ by a^2, which, in the general case, is a complex number, and retain the symbol $||\mathbf{a}||^2$ to indicate the squared norm $\mathbf{aa}^*$. In addition, while the real number $||a||=\sqrt{\mathbf{aa}^*}$ indicates the $\mathbf{a}$ norm, the complex number a indicates $\sqrt{\mathbf{aa}}$. If not otherwise specified, the unit vector $\hat{\mathbf{a}}$ is $\mathbf{a}/a$.

2.2. Remarks on the Fourier transform

In general, the wavevector $\mathbf{k}$ is complex. Therefore, the Fourier transform must be considered in a complex plane (Morse & Feshbach, 1953, p. 453) and we must prove that any solution of (2) can be expressed as a superposition of inhomogeneous plane waves $\exp(-\mathrm{i}\mathbf{kr})$. Up to now we have dealt with all the three directions of the direct space on an equal footing but it is not necessary to do so. We can single out one direction, say z, and treat it separately. The reason for this possibility is that we are interested in solving initial-value problems in which $\mathbf{D}(\mathbf{r})$ is given on the $z=0$ plane and the domain of $\tilde{\mathbf{D}}(\mathbf{q};z=0)$ is a set of real two-dimensional wavevectors. For these problems, $\mathrm{Im}(\mathbf{k})$ is along z, so that the inverse of (3) is (Morse & Feshbach, 1953, p. 461)

$$\mathbf{D}(\mathbf{r})=\frac{1}{(2\pi)^3}\int_{-\infty}^{+\infty}\tilde{\mathbf{D}}(\mathbf{k})\exp(-\mathrm{i}\mathbf{kr})\,\mathrm{d}\mathbf{k}, \tag{22}$$

where, as far as integration over k_z is concerned, the integration is performed along any horizontal contour that leaves all the singularities of $\tilde{\mathbf{D}}(k_z)$ below it and is closed by the infinitely remote lower semicircle, which makes no contribution. Equation (22) is the plane-wave superposition we have been looking for, the basis functions being the (infinite) set of inhomogeneous plane waves $\exp(-\mathrm{i}\mathbf{kr})$, where $\mathrm{Im}(k_z)$ is a constant. It is worth noting that the actual value of $\mathrm{Im}(k_z)$ is not essential, provided that all singularities of $\tilde{\mathbf{D}}(k_z)$ are below it.

2.3. Remarks on the delta distribution

In the present paper, we use the orthogonality and completeness (in the Dirac sense) of the $\exp(-\mathrm{i}\mathbf{kr})$ basis. Orthogonality and completeness are expressed by the integral representations of the delta distribution

$$\delta(\mathbf{r}-\mathbf{r}')=(2\pi)^{-3}\int_{-\infty}^{+\infty}\exp[\mathrm{i}\mathbf{k}(\mathbf{r}-\mathbf{r}')]\,\mathrm{d}\mathbf{k}, \tag{23}$$

where the integration contour is specified as in (22), and

$$\delta(\mathbf{k}-\mathbf{k}')=\int_{-\infty}^{+\infty}\exp[\mathrm{i}\mathbf{r}(\mathbf{k}-\mathbf{k}')]\,\mathrm{d}\mathbf{r}, \tag{24}$$

where $\mathrm{Im}(\mathbf{k})=\mathrm{Im}(\mathbf{k}')$.

3. X-ray propagation in a crystal

Our derivation of the master equations describing the dynamics of X-rays in a crystal, no matter whether perfect or deformed, follows Kato (1973). Owing to the intricacy and subtlety of Kato's derivation, we think it is advisable to summarize it here and focus on assumptions and steps. Our starting point is still (4), but now

$$\chi=\sum_h\chi_h^u(\mathbf{r})\exp(-\mathrm{i}\mathbf{hr}), \tag{25}$$

where $\chi_h^u(\mathbf{r})=\chi_h\exp(\mathrm{i}\mathbf{hu})$ and $\mathbf{u}$ is the crystal deformation. In (4), we need also the Fourier transform

$$\begin{aligned}[\widetilde{\chi\mathbf{D}}](\mathbf{k})&=\sum_h\int_{-\infty}^{+\infty}\tilde{\chi}_h^u(\mathbf{k}-\mathbf{h}-\mathbf{k}')\tilde{\mathbf{D}}(\mathbf{k}')\,\mathrm{d}\mathbf{k}'\\&=\sum_h[\tilde{\chi}_h^u*\tilde{\mathbf{D}}](\mathbf{k}-\mathbf{h}),\end{aligned} \tag{26}$$

where $[f_1*f_2](x)$ is the convolution integral between $f_1(x)$ and $f_2(x)$ and

$$\tilde{\chi}(\mathbf{k})=\sum_h\int_{-\infty}^{+\infty}\tilde{\chi}_h^u(\mathbf{k}-\mathbf{k}')\delta(\mathbf{k}'-\mathbf{h})\,\mathrm{d}\mathbf{k}'=\sum_h\tilde{\chi}_h^u(\mathbf{k}-\mathbf{h}) \tag{27}$$

is the Fourier transform of (25).

We resort now to two-beam approximation. Therefore, we set

$$\tilde{\mathbf{D}}(\mathbf{k}) = \begin{cases} \tilde{\mathbf{D}}_O(\mathbf{k}) & \text{if } \mathbf{k} \approx \mathbf{k}_o \\ \tilde{\mathbf{D}}_G(\mathbf{k}) & \text{if } \mathbf{k} \approx \mathbf{k}_g = \mathbf{k}_o + \mathbf{g} \\ 0 & \text{otherwise.} \end{cases} \tag{28}$$

Equation (28) implies that we are looking for a solution of (4), which should be the superposition of two quasi-plane waves propagating in the $\mathbf{k}_o$ and $\mathbf{k}_g$ directions. It must be noted that this trial solution is defined in the reciprocal space and has two well spaced peaks centered on $\mathbf{k}_o$ and $\mathbf{k}_g$ and has negligible widths with respect to both K and g, that is, $\tilde{\mathbf{D}}(\mathbf{k}) \approx \mathbf{D}_O\delta(\mathbf{k}-\mathbf{k}_o) + \mathbf{D}_G\delta(\mathbf{k}-\mathbf{k}_g)$. Additionally, $\tilde{\chi}_o^u(\mathbf{k}) = \chi_o\delta(\mathbf{k})$ and, if $\tilde{\chi}_h^u(\mathbf{k}) \approx \chi_h\delta(\mathbf{k})$, the convolution in (26) is the average (with $\tilde{\chi}_h^u$ weight) of $\tilde{\mathbf{D}}$ about $\mathbf{k}-\mathbf{h}$. Hence, we can write

$$[\widetilde{\chi\mathbf{D}}](\mathbf{k}) \approx \begin{cases} \chi_o\tilde{\mathbf{D}}_O(\mathbf{k}) + [\tilde{\chi}_g^u * \tilde{\mathbf{D}}_G](\mathbf{k}+\mathbf{g}) & \text{if } \mathbf{k} \approx \mathbf{k}_o \\ \chi_o\tilde{\mathbf{D}}_G(\mathbf{k}) + [\tilde{\chi}_g^u * \tilde{\mathbf{D}}_O](\mathbf{k}-\mathbf{g}) & \text{if } \mathbf{k} \approx \mathbf{k}_g \\ 0 & \text{otherwise.} \end{cases} \tag{29}$$

As we did in the case of free-space propagation, we now shift the reciprocal-space origin in $\mathbf{k}_o$ when $\mathbf{k} \approx \mathbf{k}_o$ and in $\mathbf{k}_g$ when $\mathbf{k} \approx \mathbf{k}_g$. In other words, we set $\mathbf{k} = \mathbf{k}_o + \mathbf{p}$ when $\mathbf{k} \approx \mathbf{k}_o$, $\mathbf{k} = \mathbf{k}_g + \mathbf{p}$ when $\mathbf{k} \approx \mathbf{k}_g$ and (Kato, 1973)

$$\tilde{\mathbf{D}}(\mathbf{k}) = \begin{cases} \tilde{\mathbf{D}}_O(\mathbf{k}_o+\mathbf{p}) = \tilde{\mathbf{D}}_o(\mathbf{p}) & \text{if } \mathbf{k} \approx \mathbf{k}_o \\ \tilde{\mathbf{D}}_G(\mathbf{k}_g+\mathbf{p}) = \tilde{\mathbf{D}}_g(\mathbf{p}) & \text{if } \mathbf{k} \approx \mathbf{k}_g \\ 0 & \text{otherwise,} \end{cases} \tag{30}$$

where the small vector $\mathbf{p}$ is termed resonance error. In the direct space, the above transformations correspond to

$$\mathbf{D}(\mathbf{r}) = \mathbf{D}_o(\mathbf{r})\exp(-\mathrm{i}\mathbf{k}_o\mathbf{r}) + \mathbf{D}_g(\mathbf{r})\exp(-\mathrm{i}\mathbf{k}_g\mathbf{r}), \tag{31}$$

where $\mathbf{D}_o(\mathbf{r})$ and $\mathbf{D}_g(\mathbf{r})$ are slowly varying functions of $\mathbf{r}$, as can be verified by calculating the inverse Fourier transform of (30). Our next step is to evaluate convolutions in (29). Since

$$\begin{aligned} [\tilde{\chi}_g^u * \tilde{\mathbf{D}}_G](\mathbf{k}+\mathbf{g}) &= \int_{-\infty}^{+\infty} \tilde{\chi}_g^u(\mathbf{k}+\mathbf{g}-\mathbf{k}')\tilde{\mathbf{D}}_G(\mathbf{k}')\,\mathrm{d}\mathbf{k}' \\ &= \int_{-\infty}^{+\infty} \tilde{\chi}_g^u(\mathbf{k}_o+\mathbf{p}+\mathbf{g}-\mathbf{k}_g-\mathbf{p}')\tilde{\mathbf{D}}_G(\mathbf{k}_g+\mathbf{p}')\,\mathrm{d}\mathbf{p}' \\ &= \int_{-\infty}^{+\infty} \tilde{\chi}_g^u(\mathbf{p}-\mathbf{p}')\tilde{\mathbf{D}}_g(\mathbf{p}')\,\mathrm{d}\mathbf{p}', \end{aligned} \tag{32}$$

because $[\tilde{\chi}_g^u * \tilde{\mathbf{D}}_G](\mathbf{k}+\mathbf{g}) \neq 0$ only if $\mathbf{k} \approx \mathbf{k}_o$ and $\tilde{\mathbf{D}}_G(\mathbf{k}') \neq 0$ only if $\mathbf{k}' \approx \mathbf{k}_g$, and since, similarly,

$$[\tilde{\chi}_g^u * \tilde{\mathbf{D}}_O](\mathbf{k}-\mathbf{g}) = \int_{-\infty}^{+\infty} \tilde{\chi}_g^u(\mathbf{p}-\mathbf{p}')\tilde{\mathbf{D}}_o(\mathbf{p}')\,\mathrm{d}\mathbf{p}', \tag{33}$$

(29) can be rewritten as

$$[\widetilde{\chi\mathbf{D}}](\mathbf{k}) \approx \begin{cases} \chi_o\tilde{\mathbf{D}}_o(\mathbf{p}) + [\tilde{\chi}_g^u * \tilde{\mathbf{D}}_g](\mathbf{p}) & \text{if } \mathbf{k} \approx \mathbf{k}_o \\ \chi_o\tilde{\mathbf{D}}_g(\mathbf{p}) + [\tilde{\chi}_g^u * \tilde{\mathbf{D}}_o](\mathbf{p}) & \text{if } \mathbf{k} \approx \mathbf{k}_g \\ 0 & \text{otherwise.} \end{cases} \tag{34}$$

By using (34) in the first equation of (4), together with $\mathbf{a}\times(\mathbf{a}\times\mathbf{b}) = -a^2\mathbf{b} + \mathbf{a}(\mathbf{ab})$ and by multiplying the resulting two equations by $\hat{\mathbf{D}}_o$ and $\hat{\mathbf{D}}_g$ (the third equation being a trivial identity), respectively, we obtain

$$\begin{aligned} [(1-\chi_o)(\mathbf{k}_o+\mathbf{p})^2 - K^2]\tilde{D}_o - (\mathbf{k}_o+\mathbf{p})^2(\tilde{\chi}_{\bar{g}}^u * \tilde{\mathbf{D}}_g)\hat{\mathbf{D}}_o = 0 \\ [(1-\chi_o)(\mathbf{k}_g+\mathbf{p})^2 - K^2]\tilde{D}_g - (\mathbf{k}_g+\mathbf{p})^2(\tilde{\chi}_g^u * \tilde{\mathbf{D}}_o)\hat{\mathbf{D}}_g = 0. \end{aligned} \tag{35}$$

The equation $(\mathbf{k}_n+\mathbf{p})\tilde{\mathbf{D}}_n = 0$, $n = o, g$, which is obtained by using (30) in the second equation of (4), indicates that only two components of $\tilde{\mathbf{D}}_o$ and $\tilde{\mathbf{D}}_g$ are independent. Let us set these components orthogonal and parallel to the plane defined by $\mathbf{k}_o$ and $\mathbf{k}_g$ (reflection plane) and denote them by σ and π. Hence, these components satisfy the master equations for X-ray dynamics

$$\begin{aligned} [(1-\chi_o)(\mathbf{k}_o+\mathbf{p})^2 - K^2]\tilde{D}_o - (\mathbf{k}_o+\mathbf{p})^2(\tilde{\chi}_{\bar{g}}^u * \tilde{D}_g) = 0 \\ [(1-\chi_o)(\mathbf{k}_g+\mathbf{p})^2 - K^2]\tilde{D}_g - (\mathbf{k}_g+\mathbf{p})^2(\tilde{\chi}_g^u * \tilde{D}_o) = 0, \end{aligned} \tag{36}$$

where the σ and π superscripts have been omitted to simplify notations, $\tilde{\chi}_{\pm g}^{u\sigma} = \tilde{\chi}_{\pm g}^u$ and $\tilde{\chi}_{\pm g}^{u\pi} = \tilde{\chi}_{\pm g}^u\hat{\mathbf{D}}_o^\pi\hat{\mathbf{D}}_g^\pi$. Equations (36) had already been obtained by Kato (1973), although in his derivation the critical formulae (29) and (34) are not explicitly stated. In (36), we stated all the needed assumptions and approximations in terms of the $\chi(\mathbf{r})$ and $\mathbf{D}(\mathbf{r})$ spectra, without resorting to vague hypotheses about smoothness and negligible derivatives of field amplitudes and crystal deformation. With a perfect crystal, $\tilde{\chi}_h^u(\mathbf{p}) = \chi_h\delta(\mathbf{p})$, so that, in (36), convolutions reduce to the simple $\chi_{\pm g}\tilde{D}_{og}$ products.

4. Infinite perfect crystals

In this section, we show how the usual dynamical equations for X-ray diffraction in infinite perfect crystals can be recovered from (36). Up to now, we did not make any specific choice for the $\mathbf{k}_o$ wavevector. Since, when the interaction with the crystal is switched off, (36) reduces to (6), on the basis of our study of free propagation in §2, we set

$$k_o^2 = k_g^2 = (1+\chi_o)K^2. \tag{37}$$

Since $k_g^2 = k_o^2 + g^2 + 2\mathbf{gk}_o$, (37) requires $g^2 + 2\mathbf{gk}_o = 0$, which expresses Bragg's law and implicitly defines $\mathbf{k}_o$. Furthermore, it is clear that $\mathrm{Im}(\mathbf{k}_o) = \mathrm{Im}(\mathbf{k}_g)$ and that $\mathrm{Im}(\mathbf{gk}_o) = 0$. Therefore, $\tilde{D}_o$ and $\tilde{D}_g$ are equally absorbed and damping occurs along a direction orthogonal to $\mathbf{g}$. According to what we have discussed in §2, we set, therefore, the z axis orthogonal to $\mathbf{g}$. This point will be further clarified in the next section.

In order to further simplify (36), let us introduce the following first-order approximations:

$$\begin{aligned} (1-\chi_o)(\mathbf{k}_n+\mathbf{p})^2 - K^2 &\approx 2Kp_n, \\ (\mathbf{k}_n+\mathbf{p})^2\chi_{\pm g} &\approx \chi_{\pm g}K^2, \end{aligned} \tag{38}$$

where n is either o or g, $p_n = \mathbf{p}\hat{\mathbf{K}}_n$, $\mathbf{K}_n = K\widehat{\mathrm{Re}(\mathbf{k}_n)}$, and $\gamma_n = \hat{\mathbf{K}}_n\hat{\mathbf{z}} = \cos\psi_n$. As already discussed in §2, in order to ensure that $\mathbf{pk}_n$ could be approximated by $\mathbf{pK}_n$, it is necessary that $|\gamma_n| \gg 0$. Additionally, if $\mathbf{g} \approx -2\mathbf{K}_o$, the above approximations fail because, if the Bragg angle is close to $\pi/2$, the projections p_o and p_g are insufficient to specify $\mathbf{p}$, but we shall not examine this case. By these approximations, (36) becomes

$$\begin{aligned} 2p_o\tilde{D}_o - \chi_{\bar{g}}K\tilde{D}_g = 0 \\ 2p_g\tilde{D}_g - \chi_g K\tilde{D}_o = 0 \end{aligned} \tag{39}$$

and the dispersion equation for X-rays in a crystal is approximated by

$$4p_o p_g = K^2\chi_g\chi_{\bar{g}}. \tag{40}$$

The above dispersion equation brings to light that $\mathbf{p}$ is, in the general case, complex. We must note that, if $\chi_{\pm g} = 0$, that is, if interaction with the crystal is switched off, we do not obtain the free-space dispersion equation $(1-\chi_o)k_n^2 - K^2 = 0$ but its approximation $p_o p_g = 0$. This is due to our having left out the second-order terms in (39) which, consequently, do not include diffraction as is understood in visible optics. Therefore, in the absence of any interaction with the crystal, the crystal field propagates according to geometric optics.

5. Semi-infinite perfect crystals

Let us consider a crystal occupying the semi-infinite region $z \geq 0$, while the region $z < 0$ is a vacuum. Let the z axis be normal to the crystal surface and be directed inwards and let the free-space field $\mathbf{D}^e(\mathbf{r})$ be defined on $-\infty < z \leq 0$. The crystal-surface normal has no special characteristics; in principle, the selection of the z coordinate is arbitrary. We shall presently show that our choice is the most convenient with boundary conditions given on the crystal surface. Let the free-space field be represented as

$$\mathbf{D}^e(\mathbf{r}) = D_o^e(\mathbf{r})\exp(-\mathrm{i}\mathbf{K}_o\mathbf{r})\hat{\mathbf{e}}_o + D_g^e(\mathbf{r})\exp(-\mathrm{i}\mathbf{K}_g\mathbf{r})\hat{\mathbf{e}}_g, \tag{41}$$

where $K_o = K$, $\mathbf{K}_g = \mathbf{K}_o + \mathbf{g}$, and $\hat{\mathbf{e}}_o$ and $\hat{\mathbf{e}}_g$ are unit vectors orthogonal (σ polarization) or parallel (π polarization) to the reflection plane, and let the free-space wavevector $\mathbf{K}_o$ satisfy the $2\hat{\mathbf{g}}\mathbf{K}_o = -g$ Bragg's law. The glancing angle of the plane wave $\exp(-\mathrm{i}\mathbf{K}_o\mathbf{r})$ to the diffracting planes (Bragg angle) is implicitly given by $\sin\Theta_\mathrm{B} = -\hat{\mathbf{K}}_o\hat{\mathbf{g}}$. Boundary conditions require that, if $z = 0$, (41) and (31) must be identical. Consequently, they require $D_n(\mathbf{x}, 0) = D_n^e(\mathbf{x}, 0)$ and $\mathbf{k}_o\mathbf{x} = \mathbf{K}_o\mathbf{x}$. It is now necessary to select the crystal wavevector $\mathbf{k}_o$. Our choice (37) is incompatible with the $\mathbf{k}_o\mathbf{x} = \mathbf{K}_o\mathbf{x}$ requirement, but we can still select $\mathbf{k}_o$ alone in such a way that $k_o^2 = (1+\chi_o)K^2$. This selection, together with $\mathbf{k}_o\mathbf{x} = \mathbf{K}_o\mathbf{x}$ and $|\chi_o|/\gamma_o \ll 1$, implies that

$$\mathbf{k}_o = \mathbf{K}_o + \chi_o K\hat{\mathbf{z}}/(2\gamma_o). \tag{42}$$

Furthermore, let $\mathbf{K}_o$ be directed inward, that is, $\cos\psi_o = \gamma_o = \hat{\mathbf{K}}_o\hat{\mathbf{z}} > 0$. The $|\chi_o|/\gamma_o \ll 1$ requirement indicates that a grazing incidence on the crystal surface is excluded. If it is not, the approximations we are going to use fail and it is convenient to take the z axis on the crystal surface and inside the reflection plane. However, we shall not examine this case. It must be pointed out that $\mathbf{k}_o$ and $\mathbf{k}_g$ have the same imaginary part, which is directed along z. We now calculate

$$k_g^2 = k_o^2 + g^2 + 2\mathbf{g}\mathbf{k}_o = (1+\chi_o)K^2 + g_z\chi_o K/\gamma_o, \tag{43}$$

where $g_z = \mathbf{g}\hat{\mathbf{z}}$, and observe that $\gamma_g K = \mathbf{K}_g\hat{\mathbf{z}} = \gamma_o K + g_z$, that $g_z = (\gamma_g - \gamma_o)K$ and that

$$k_g^2 = (1+\chi_o)K^2 - (1-\gamma)\chi_o K^2, \tag{44}$$

where $\gamma = \gamma_g/\gamma_o$ is the asymmetry. Our next step is to approximate the coefficients of $\tilde{D}_o$ and $\tilde{D}_g$ in (36) up to first order with respect to the resonance error and to the Fourier components of electric susceptibility. Hence,

$$\begin{aligned} (1-\chi_o)(\mathbf{k}_o+\mathbf{p})^2 - K^2 &\approx 2K(\gamma_o p_z + \sqrt{1-\gamma_o^2}q_o), \\ (1-\chi_o)(\mathbf{k}_g+\mathbf{p})^2 - K^2 &\approx 2K(\gamma_g p_z + \sqrt{1-\gamma_g^2}q_g) \\ &\quad - (1-\gamma)\chi_o K^2, \\ (\mathbf{k}_n+\mathbf{p})^2\chi_{\pm g} &\approx \chi_{\pm g}K^2, \end{aligned} \tag{45}$$

where $\mathbf{K}_n = K(\gamma_n\hat{\mathbf{z}} + \sqrt{1-\gamma_n^2}\hat{\mathbf{x}}_n)$, $\mathbf{p} = p_z\hat{\mathbf{z}} + \mathbf{q}$, $q_n = \mathbf{q}\hat{\mathbf{x}}_n$ and, for the same reasons discussed in §§2 and 4, we assume both γ_o and γ_g sufficiently different from zero. We also note that, since the wavevectors of the D_n^e spectra are real, the constraint $\tilde{D}_n(\mathbf{q}; 0) = \tilde{D}_n^e(\mathbf{q}; 0)$ implies that $\mathbf{q}$ is real. Therefore, with (42) also taken into consideration, the imaginary part of $\mathbf{p}$ is along the z direction, as we anticipated in §2. With these approximations, (36) becomes

$$\begin{aligned} 2(\gamma_o p_z + \sqrt{1-\gamma_o^2}q_o)\tilde{D}_o - \chi_{\bar{g}}K\tilde{D}_g = 0 \\ [2(\gamma_g p_z + \sqrt{1-\gamma_g^2}q_g) - (1-\gamma)\chi_o K]\tilde{D}_g - \chi_g K\tilde{D}_o = 0. \end{aligned} \tag{46}$$

If we take account of the correspondence $p_z \rightarrow \mathrm{i}\partial_z$ between the reciprocal- and direct-space representations of the impulse, we obtain the Takagi equations

$$\mathrm{i}\partial_z\begin{bmatrix}\tilde{D}_o \\ \tilde{D}_g\end{bmatrix} = \begin{bmatrix}-q_o\tan\psi_o & \nu_o \\ \nu_g & -q_g\tan\psi_g + \beta_g\end{bmatrix}\begin{bmatrix}\tilde{D}_o \\ \tilde{D}_g\end{bmatrix}, \tag{47}$$

where the crystal field is represented by

$$\begin{aligned} \mathbf{D}(\mathbf{r}) = [D_o(\mathbf{r})\exp(-\mathrm{i}\mathbf{K}_o\mathbf{r})\hat{\mathbf{e}}_o + D_g(\mathbf{r})\exp(-\mathrm{i}\mathbf{K}_g\mathbf{r})\hat{\mathbf{e}}_g] \\ \times \exp[-\mathrm{i}\chi_o Kz/(2\gamma_o)], \end{aligned} \tag{48}$$

$\tan\psi_n = \sqrt{1-\gamma_n^2}/\gamma_n$, $\nu_o = \chi_{\bar{g}}K/(2\gamma_o)$, $\nu_g = \chi_g K/(2\gamma_g)$ and $\beta_g = (1-\gamma)\chi_o K/(2\gamma_g)$. Note that the sign of $\tan\psi_n$ is determined by the sign of γ_n, the square root function taking its principal value.

If we go back to the direct space, Takagi's equations take the form

$$\mathrm{i}\partial_z\begin{bmatrix}D_o \\ D_g\end{bmatrix} = \begin{bmatrix}-\mathrm{i}\tan\psi_o\partial_{xo} & \nu_o \\ \nu_g & -\mathrm{i}\tan\psi_g\partial_{xg} + \beta_g\end{bmatrix}\begin{bmatrix}D_o \\ D_g\end{bmatrix}, \tag{49}$$

where ∂_{xn} are derivatives along the $\mathbf{K}_n$ components in the crystal surface. The approximations (45), retaining only the linear terms in the resonance error, are equivalent to the linearization (8) of the dispersion surface in a free space. However, in a crystal, (45) approximates a fourth-order dispersion surface with a second-order one and, therefore, X-ray dynamics imitates visible-optics diffraction. Additionally, by remembering the $p_i \rightarrow \mathrm{i}\partial_i$ and $p_i^2 \rightarrow -\partial_i^2$ operator correspondences between direct and reciprocal spaces, we can easily see that the approximations (45) are equivalent to neglecting, in (2), the second derivatives and the products of first derivatives (Takagi, 1962, 1969). The slowly varying field and deformation requirement has been specified precisely by (28) and (29) in terms of the relative widths of the field and

susceptibility angular spectra with respect to the reciprocal vector of diffracting planes. The approximations implied in constructing the several variants of the dynamical theory of X-ray propagation in crystals are further discussed by Härtwig (2001).

From a mathematical point of view, $[D_o(\mathbf{x}; z), D_g(\mathbf{x}; z)]^T$ is the direct-space representation $\langle \mathbf{x}|D(z)\rangle$ of the abstract state vector $|D(z)\rangle$ belonging to the Hilbert space $V_2 \otimes \mathcal{L}^2(\mathbb{R}_2)$, where V_2 is the space of the dispersion-surface branches (a two-dimensional vector space) and $\mathcal{L}^2(\mathbb{R}_2)$ is the space of the square-integrable two-variable functions. Throughout this paper, we use the 2×1 matrix representation of V_2. If the Hamiltonian operator $\mathbf{H}$ is so defined that the matrix elements of its reciprocal-space representation

$$\langle \mathbf{q}, n|\mathbf{H}|\mathbf{q}', m\rangle = \delta(\mathbf{q} - \mathbf{q}')\tilde{H}_{nm}(\mathbf{q}), \tag{50}$$

where n and m are the 'quantum' numbers o and g, are the elements of the 2×2 Hamiltonian matrix $\tilde{H}(\mathbf{q})$ on the right side of (47), then the Dirac-type equation

$$\mathrm{i}\partial_z|D(z)\rangle = \mathbf{H}|D(z)\rangle, \tag{51}$$

where

$$\langle \mathbf{q}, n|D(z)\rangle = \tilde{D}_n(\mathbf{q}; z) \tag{52}$$

and

$$\langle \mathbf{x}, n|D(z)\rangle = D_n(\mathbf{x}; z), \tag{53}$$

is equivalent to Takagi's equations. The representation (48) of the crystal field, which we term the O–G representation, uses the plane waves $\langle \mathbf{r}, n|n\rangle = \exp(-\mathrm{i}\mathbf{K}_n\mathbf{r})$ as the basis functions in the space of dispersion-surface branches. Readers familiar with quantum mechanics or physical optics will recognize (47) as an analog of the equation of the motion of two-level atoms or of polarized light (Silverman, 1994, p. 139). The analogy holds because X-rays in crystals, two-level atoms (Marte *et al.*, 1991; Bordé, 2002) and polarized light (Azzam & Bashara, 1977), as well as other physical systems, are all described in terms of two basis states and, from the basic mathematical point of view, are all two-state systems (Cohen-Tannoudji *et al.*, 1977, p. 424).

5.1. Canonical form of Takagi's equation

If the normal to the crystal surface is contained in the reflection plane and the crystal is cut symmetrically, that is, if $|\gamma_g| = \gamma_o$, then $q_o \tan\psi_o = -q_g \tan\psi_g$. If, additionally, the unit cell is symmetrical with respect to the reflection planes and the origin of the reference frame is on a reflection plane, then $\nu_o = \nu_g$, which we shall indicate with ν. In this case, the Hamiltonian matrix in (47) takes the simple form

$$\tilde{H}(\mathbf{q}) = \nu\begin{bmatrix} -\eta & 1 \\ 1 & \eta \end{bmatrix}, \tag{54}$$

where $\eta = q_o \tan\psi_o/\nu = -q_g \tan\psi_g/\nu$. In the present section, we shall prove that (54) is relevant also in a non-coplanar and asymmetric geometry.

The asymmetry of the Hamiltonian matrix in (47) is due to a non-optimal choice of crystal-field representation. In order to distill from (47) the basic mathematical structure of the Hamiltonian matrix, let us introduce the deviation parameter $q_- = \mathbf{q}\mathbf{v}_- + \beta_g/2$, where $2\mathbf{v}_- = \hat{\mathbf{x}}_o \tan\psi_o - \hat{\mathbf{x}}_g \tan\psi_g$, and write (Cohen-Tannoudji *et al.*, 1977, p. 420)

$$\tilde{H}(q_-, q_+) = \begin{bmatrix} -q_- & \nu_o \\ \nu_g & q_- \end{bmatrix} - \begin{bmatrix} q_+ & 0 \\ 0 & q_+ \end{bmatrix} + \begin{bmatrix} \beta_g/2 & 0 \\ 0 & \beta_g/2 \end{bmatrix}, \tag{55}$$

where $q_+ = \mathbf{q}\mathbf{v}_+$ and $2\mathbf{v}_+ = \hat{\mathbf{x}}_o \tan\psi_o + \hat{\mathbf{x}}_g \tan\psi_g$. We shall presently show that a series of transformations eliminates one term after the other and brings (55) to the form (54). Let us first anticipate that $2\mathbf{v}_\pm$ are the total and differential propagation velocities of the crystal-field components D_o and D_g in the fictitious time z. A detailed discussion of this $2\mathbf{v}_\pm$ interpretation will be given in §6.1, where we shall solve Takagi's equations *in vacuo*. Additionally, $2\mathbf{v}_-$ is the same vector as $\hat{\mathbf{K}}_o/\gamma_o - \hat{\mathbf{K}}_g/\gamma_g$. Since $2\mathbf{v}_-$ lies on the crystal surface and $\hat{\mathbf{K}}_o/\gamma_o - \hat{\mathbf{K}}_g/\gamma_g$ on the reflection plane, then $\mathbf{v}_-$ is situated along the intersection between the reflection plane and the crystal surface. When the normal to the crystal surface is contained inside the reflection plane (coplanar geometry), $\mathbf{v}_+$ is situated along the same intersection. In this case, if the crystal is, additionally, cut symmetrically, then $q_+ = 0$.

Since the solution of Takagi's equations is (Cohen-Tannoudji *et al.*, 1977, p. 308)

$$|D(z)\rangle = \exp(-\mathrm{i}\mathbf{H}z)|D(0)\rangle, \tag{56}$$

the last term of (55) originates a phase factor $\exp(-\mathrm{i}\beta_g z/2)$. Consequently, this term can be eliminated by using the crystal-field representation

$$\mathbf{D}(\mathbf{r}) = [D'_o(\mathbf{r})\exp(-\mathrm{i}\mathbf{K}_o\mathbf{r})\hat{\mathbf{e}}_o + D'_g(\mathbf{r})\exp(-\mathrm{i}\mathbf{K}_g\mathbf{r})\hat{\mathbf{e}}_g]\exp(-\mathrm{i}\kappa z), \tag{57}$$

where $\kappa = \chi_o K/(2\gamma_o) + \beta_g/2 = (1+\gamma)\chi_o K/(4\gamma_g)$.

Also the second term of (55), which originates the phase factor $\exp(\mathrm{i}q_+ z)$, can be eliminated by redefining the crystal field as

$$\tilde{D}'_n(\mathbf{q}; z) = \tilde{\varphi}'_n(\mathbf{q}; z)\exp(\mathrm{i}q_+ z). \tag{58}$$

This transformation,

$$D'_n(\mathbf{x}; z) = \int_{-\infty}^{+\infty} \delta(\mathbf{x} - \mathbf{v}_+ z - \mathbf{x}')\varphi'_n(\mathbf{x}')\,\mathrm{d}\mathbf{x}' = \varphi'_n(\mathbf{x} - \mathbf{v}_+ z; z), \tag{59}$$

is a translation along $\hat{\mathbf{v}}_+$. It makes X-ray dynamics depending only on z and x_1, a coordinate along the intersection between the reflection plane and the crystal surface. Consequently, since only two coordinates are involved, X-ray diffraction is two-dimensional, also in the case of non-coplanar geometry, where $\mathbf{v}_+$ has a component orthogonal to the x_1 axis. In fact, in this direction, X-rays propagate in a straight line. We have thus proved that X-ray diffraction occurs in a plane, which is defined by the surface normal and by the surface intersection with the reflection plane.

The first term of (55) is unsatisfactory because, in the case of non-absorbing crystals, it is not Hermitian, owing to the

geometrical factors γ_o and γ_g in ν_o and ν_g. In order to retrieve a Hermitian Hamiltonian (when crystals are non-absorbing), we redefine the crystal field as

$$\begin{aligned}\tilde{\varphi}'_o &= \tilde{\varphi}_o \\ \tilde{\varphi}'_g &= \sqrt{\nu_g/\nu_o}\tilde{\varphi}_g = \sqrt{\sigma/\gamma}\tilde{\varphi}_g,\end{aligned} \tag{60}$$

in which $\sigma = \chi_g/\chi_{\bar{g}}$. To summarize, by means of the crystal-field representation

$$\begin{aligned}\mathbf{D}(\mathbf{r}) = [&\varphi_o(\mathbf{x} - \mathbf{v}_+ z; z)\exp(-\mathrm{i}\mathbf{K}_o\mathbf{r})\hat{\mathbf{e}}_o \\ &+ \sqrt{\sigma/\gamma}\varphi_g(\mathbf{x} - \mathbf{v}_+ z; z)\exp(-\mathrm{i}\mathbf{K}_g\mathbf{r})\hat{\mathbf{e}}_g]\exp(-\mathrm{i}\kappa z),\end{aligned} \tag{61}$$

the reciprocal-space representation of Takagi's equations is

$$\mathrm{i}\partial_\zeta\begin{bmatrix}\tilde{\varphi}_o \\ \tilde{\varphi}_g\end{bmatrix} = \begin{bmatrix}-\eta & 1 \\ 1 & \eta\end{bmatrix}\begin{bmatrix}\tilde{\varphi}_o \\ \tilde{\varphi}_g\end{bmatrix}, \tag{62}$$

where

$$\eta = q_-\big/\sqrt{\nu_o\nu_g} = \Lambda_e q_-/\pi \tag{63}$$

is the reduced deviation parameter,

$$\Lambda_e = (2\pi\gamma_o/K)\sqrt{\gamma/\chi_g\chi_{\bar{g}}}, \tag{64}$$

is the *Pendellösung* (extinction) length and $\zeta = \pi z/\Lambda_e$ is a new time parameter. It must be noted that, contrary to Authier (2001, p. 85), in (64) we do not consider the absolute value of γ. Consequently, the real parts of Λ_e and η exchange with the imaginary ones when Laue's geometry, $\gamma > 0$, exchanges with Bragg's geometry, $\gamma < 0$.

Let us now examine the meaning of the deviation parameter q_-. Provided that $\gamma_o \gg 0$, the component in the $x_1 z$ plane of the glancing-angle deviation from Θ_B of the $\mathbf{K}_o + \mathbf{p}$ component of the free-space field is given by

$$\theta \approx \frac{\mathbf{p}\hat{\mathbf{x}}_1}{K_{oz}} \approx \frac{q_1}{\gamma_o K} = \frac{q_- - \beta_g/2}{\gamma_o K||\mathbf{v}_-||}. \tag{65}$$

Hence, when observing that

$$||\mathbf{v}_-||^2 = \frac{||\hat{\mathbf{K}}_o/\gamma_o - \hat{\mathbf{K}}_g/\gamma_g||^2}{4} = \frac{\gamma_o^2 + \gamma_g^2 - 2\gamma_o\gamma_g\cos 2\Theta_\mathrm{B}}{4\gamma_o^2\gamma_g^2}, \tag{66}$$

where we used $\hat{\mathbf{K}}_o\hat{\mathbf{K}}_g = \cos 2\Theta_\mathrm{B}$, we obtain

$$q_- = \frac{K\theta\sqrt{\gamma_o^2 + \gamma_g^2 - 2\gamma_o\gamma_g\cos 2\Theta_\mathrm{B}}}{2\gamma_g} + \tfrac{1}{2}\beta_g. \tag{67}$$

If z lies in the reflection plane (coplanar geometry), by noting that $\gamma_g = \cos(2\Theta_\mathrm{B} - \psi_o)$ and by using a few trigonometry relations (Wolfram, 1999), (67) reduces to

$$q_- = \frac{K\theta\sin 2\Theta_\mathrm{B}}{2\gamma_g} + \tfrac{1}{2}\beta_g. \tag{68}$$

In this case, account being taken of (63), η is nothing else than the reduced deviation parameter introduced by Authier (2001, p. 127). The q_- offset, $-\beta_g/2$, measures the glancing-angle deviation corresponding to the center of the reflection domain,

$$\begin{aligned}\theta_0 &= -\frac{\mathrm{Re}(\beta_g)\gamma_g}{K\sqrt{\gamma_o^2 + \gamma_g^2 - 2\gamma_o\gamma_g\cos 2\Theta_\mathrm{B}}} \\ &= -\frac{(1-\gamma)\mathrm{Re}(\chi_o)}{2\sqrt{\gamma_o^2 + \gamma_g^2 - 2\gamma_o\gamma_g\cos 2\Theta_\mathrm{B}}},\end{aligned} \tag{69}$$

which, for coplanar geometry, is (Authier, 2001, p. 85)

$$\theta_0 = -\frac{\mathrm{Re}(\beta_g)\gamma_g}{K\sin 2\Theta_\mathrm{B}} = -\frac{(1-\gamma)\mathrm{Re}(\chi_o)}{2\sin 2\Theta_\mathrm{B}}. \tag{70}$$

5.1.1. Remarks on the square root function. We use the symbol $\sqrt{w}$ to indicate the single-value square-root function $\sqrt{|w|}\exp(\mathrm{i}\phi/2)$ of the complex number $w = |w|\exp(\mathrm{i}\phi)$. In order to write $\sqrt{w}$ as $\mathrm{Re}(\sqrt{w}) + \mathrm{Im}(\sqrt{w})$ and ensure the correctness of our algebraic manipulations, for example, that $\sqrt{\nu_o}\sqrt{\nu_g} = \sqrt{\nu_o\nu_g}$, the following specifications are necessary. If $0 \le \phi < 2\pi$ (first Riemann sheet), then

$$\begin{aligned}\sqrt{2}\,\mathrm{Re}(\sqrt{w}) &= \pm\sqrt{|w| + \mathrm{Re}(w)} \\ \sqrt{2}\,\mathrm{Im}(\sqrt{w}) &= +\sqrt{|w| - \mathrm{Re}(w)},\end{aligned} \tag{71}$$

else if $2\pi \le \phi < 4\pi$ (second Riemann sheet), then

$$\begin{aligned}\sqrt{2}\,\mathrm{Re}(\sqrt{w}) &= \mp\sqrt{|w| + \mathrm{Re}(w)} \\ \sqrt{2}\,\mathrm{Im}(\sqrt{w}) &= -\sqrt{|w| - \mathrm{Re}(w)},\end{aligned} \tag{72}$$

where the plus/minus sign is set according (first branch) or counter-according (second branch) to the sign of $\mathrm{Im}(w)$.

5.1.2. Remarks on polarizability. The sign of $\mathrm{Im}(\chi)$ depends on the choice of the exponential functions representing the crystal field and on the orientation of the propagation axis z. The exponential functions can be either $\exp(+\mathrm{i}\mathbf{K}_n\mathbf{r})$ or $\exp(-\mathrm{i}\mathbf{K}_n\mathbf{r})$, the latter being the functions used here. Additionally, since the crystal field is absorbed when propagating towards the interior crystal (*i.e.* in the positive direction of the z axis), $\mathrm{Im}(\chi)$ must be negative, and so must $\mathrm{Im}(\chi_o)$. Both χ_g and $\chi_{\bar{g}}$ are complex numbers and their phases depend on the coordinate origin. A translation $\mathbf{u}$ shifts the $\chi_{\pm g}$ phases according to $\chi^u_{\pm g} = \chi_{\pm g}\exp(\mp\mathrm{i}\mathbf{gu})$. Since Takagi's equations depend on $\chi_{\pm\mathbf{g}}$ only through the $\chi_g\chi_{\bar{g}}$ product in (63) and the $\chi_g/\chi_{\bar{g}}$ ratio in (61), a translation of the origin affects the dynamics of X-rays only through the unimportant phase factor $\exp(\mathrm{i}\mathbf{gu})$ in the g component of the crystal field. Note, however, that this phase factor has tangible consequences when the two field components are parts of an interferometer.

6. A solution to Takagi's equations

We now propose a new way to solve Takagi's equations. Let us write the solution of Takagi's equations as $|D(z)\rangle = \mathbf{U}(z)|D(0)\rangle$, where $|D(0)\rangle$ is the initial state vector characterizing the crystal field at $z = 0$ (Cohen-Tannoudji *et al.*, 1977, p. 420). Clearly, $\mathbf{U}(z) = \exp(-\mathrm{i}\mathbf{H}z)$.

6.1. Free space

Let us begin from the solution of Takagi's equations *in vacuo*. In this case, since $\nu_o = \nu_g = 0$ and $\beta_g = 0$, the reciprocal-space representation of the Hamiltonian operator **H** is

$$\tilde{H}(\mathbf{q}) = \begin{bmatrix} -q_o \tan\psi_o & 0 \\ 0 & -q_g \tan\psi_g \end{bmatrix}. \tag{73}$$

It must be noted that, in the free space, Takagi's equations do not reduce to the parabolic approximation of the wave equation. As we have already pointed out in §§4 and 5, this reduction is prevented by the first-order approximations (45). The calculation of the reciprocal-space representation of the free-space propagation operator **U** is trivial; by remembering (56), it is

$$\tilde{U}(\mathbf{q}; z) = \exp[-\mathrm{i}\tilde{H}(\mathbf{q})z] = \begin{bmatrix} \exp(\mathrm{i}q_o \tan\psi_o z) & 0 \\ 0 & \exp(\mathrm{i}q_g \tan\psi_g z) \end{bmatrix}. \tag{74}$$

Propagation in the direct space,

$$\begin{bmatrix} D_o(\mathbf{x}; z) \\ D_g(\mathbf{x}; z) \end{bmatrix} = \begin{bmatrix} D_o(\mathbf{x} - \hat{\mathbf{x}}_o z \tan\psi_o; 0) \\ D_g(\mathbf{x} - \hat{\mathbf{x}}_g z \tan\psi_g; 0) \end{bmatrix}, \tag{75}$$

is given by the inverse Fourier transform of (56). As expected, it is a translation of D_o and D_g, with velocities $\tan\psi_o$ and $\tan\psi_g$, along the $\mathbf{K}_o$ and $\mathbf{K}_g$ components in the plane orthogonal to z.

6.2. Perfect crystals

In perfect crystals, the reciprocal-space canonical representation of the Hamiltonian operator is

$$\tilde{H}_0(\mathbf{q}) = \begin{bmatrix} -\eta & 1 \\ 1 & \eta \end{bmatrix}. \tag{76}$$

The standard way to obtain the the propagation operator in perfect crystals (Cohen-Tannoudji *et al.*, 1977, p. 421) is to diagonalize (76), because the exponential of a diagonal matrix takes the form of a diagonal matrix whose elements are the exponential of the Hamiltonian eigenvalues. The calculations can easily be performed with the aid of a program for symbolic algebra such as *Mathematica* (Wolfram, 1999). In the first place, we must solve the eigenvalue problem $\tilde{H}_0|\pm\rangle = \lambda_\pm|\pm\rangle$, which yields the two eigenvalues $\lambda_\pm = \pm\sqrt{\eta^2+1}$ (Kato, 1962). In the reciprocal space, with the use of the O–G representation of the crystal field, eigenvectors $|\pm\rangle$, in Ewald's terminology the wavefields, are

$$|\pm\rangle = \begin{bmatrix} \eta - \lambda_\pm \\ -1 \end{bmatrix}. \tag{77}$$

With the $|\pm\rangle$ states as a basis, the Hamiltonian

$$\tilde{H}_\pm = \begin{bmatrix} -\sqrt{\eta^2+1} & 0 \\ 0 & \sqrt{\eta^2+1} \end{bmatrix} \tag{78}$$

is diagonal. Our next step is to calculate the inverse of the matrix

$$A = \begin{bmatrix} \eta + \sqrt{\eta^2+1} & \eta - \sqrt{\eta^2+1} \\ -1 & -1 \end{bmatrix} \tag{79}$$

constructed by juxtaposition of the $\mathbf{H}_0$ eigenvectors by which the representation change $\tilde{H}_\pm = A^{-1}\tilde{H}_0 A$ is achieved. The needed inverse is

$$A^{-1} = \frac{1}{2\sqrt{\eta^2+1}} \begin{bmatrix} 1 & \eta - \sqrt{\eta^2+1} \\ -1 & -\eta - \sqrt{\eta^2+1} \end{bmatrix}. \tag{80}$$

Eventually, the optical transfer function is

$$\begin{aligned} \tilde{U}_0(\mathbf{q}; \zeta) &= A \exp(-\mathrm{i}\tilde{H}_\pm \zeta) A^{-1} \\ &= \begin{bmatrix} T(\eta, \zeta) & -\mathrm{i}R(\eta, \zeta) \\ -\mathrm{i}R(\eta, \zeta) & T(-\eta, \zeta) \end{bmatrix}, \end{aligned} \tag{81}$$

where (Bonse & Graeff, 1977, p. 101)

$$R(\eta, \zeta) = \frac{\sin(\zeta\sqrt{\eta^2+1})}{\sqrt{\eta^2+1}}, \tag{82}$$

$$T(\eta, \zeta) = \cos(\zeta\sqrt{\eta^2+1}) + \mathrm{i}\eta R(\eta, \zeta), \tag{83}$$

$\zeta = \pi z/\Lambda_e$, $\eta = \Lambda_e q_-/\pi$ and Λ_e is given by (64).

6.2.1. Properties of transmission and reflection coefficients. According to (82) and (83), reflection and transmission coefficients oscillate with respect to both the z and q_- coordinates. Let us remark again that the dynamical theory of X-ray diffraction does not include visible-optics diffraction, that is, the second-derivative contribution to field dynamics. However, in crystals, the coupling between transmitted and reflected waves originates interference effects which imitate diffraction closely. Although a detailed examination of these effects with influential quantities (X-ray energy, diffracting planes, crystal asymmetry and so on) can be found in the copious literature, we think it useful to recall to mind here some of the reflection and transmission symmetry properties and relationships. In the first place,

$$R(-\eta) = R(\eta) \tag{84}$$

$$T(\eta)T(-\eta) = 1 - R^2(\eta) \tag{85}$$

are immediate consequences of (82) and (83). Secondly, in the case of non-absorbing crystals, $|T|^2 + |R|^2 = 1$, $R = R^*$ and $T(-\eta) = T^*(\eta)$. Finally, we consider the case when ζ tends to infinity. The simplest way to obtain asymptotic expressions of the transmission and reflection coefficients is to evaluate the limit of $\exp(-\mathrm{i}\tilde{H}_\pm\zeta)$ with ζ tending to infinity and then repeat the $A\exp(-\mathrm{i}\tilde{H}_\pm\zeta)A^{-1}$ calculation. Since the diagonal elements of $\tilde{H}_\pm$ have opposite imaginary parts, when $z \to \infty$, one element of $\exp(-\mathrm{i}\tilde{H}_\pm\zeta)$ becomes negligibly small and can be set to zero. Hence,

$$\lim_{\zeta\to\infty} R(\eta, \zeta) = \pm\frac{\mathrm{i}\exp(\mp\mathrm{i}\zeta\sqrt{\eta^2+1})}{2\sqrt{\eta^2+1}} \tag{86}$$

$$\lim_{\zeta\to\infty} T(\eta, \zeta) = \mathrm{i}(\eta \mp \sqrt{\eta^2+1})R(\eta, \zeta) \tag{87}$$

and $T(\eta)T^*(-\eta) = |R(\eta)|^2$. The plus/minus sign in (86) and (87) is set according to the $\lambda_\pm$ sign in the $\exp(-\mathrm{i}\tilde{H}_\pm\zeta)$ element that does not vanish when ζ goes to infinity.

7. Solution analysis

When $z \to \infty$, the crystal field must vanish. However, since it is a superposition of the $|\pm\rangle_\zeta = \exp(-i\pi\lambda_\pm z/\Lambda_e)|\pm\rangle$ wavefields, where $\pi\lambda_\pm/\Lambda_e = \pm\sqrt{q_-^2 + \nu_o\nu_g}$ are the Hamiltonian eigenvalues, and since the eigenvalues have opposite imaginary parts, one of the two wavefields diverges (Borrmann effect). This does not necessarily imply also crystal field divergence because of the $\exp(-i\kappa z)$ factor in (61). In the following sections, we shall investigate the asymptotic behavior of the crystal field in order to verify its fulfilling the vanishing requirement. The possible diverging wavefield will be rejected.

7.1. Laue geometry

In transmission geometry (Laue), we have $\gamma_o, \gamma_g > 0$. If, additionally, the crystal is cut symmetrically, the z axis is orthogonal to $\mathbf{g}$. Hence, $\mathbf{g}\hat{\mathbf{z}} = 0$ and $K\gamma_g = \mathbf{K}_g\hat{\mathbf{z}} = K\gamma_o$. Therefore, $\gamma_g = \gamma_o$ and $\beta_g = 0$. Since $\mathbf{g}$ belongs to the reflection plane as well as to the crystal surface, it follows that $\mathbf{g} = -g\hat{\mathbf{x}}_1$. Hence,

$$K_{g2} = K_{o2} \tag{88}$$

$$K_{g1} = -\mathbf{K}_o\hat{\mathbf{g}} - g = \mathbf{K}_o\hat{\mathbf{g}} = -K_{o1} \tag{89}$$

because $\mathbf{g}\hat{\mathbf{x}}_2 = 0$ and $\mathbf{K}_o$ satisfies Bragg's law $\mathbf{K}_o\hat{\mathbf{g}} = -g/2$. If, in addition to the condition of a symmetrically cut crystal, the reflection plane coincides with the plane defined by $\mathbf{g}$ and $\hat{\mathbf{z}}$, then $\gamma_o = \gamma_g = \cos\Theta_B$. Therefore, in this case, the deviation parameter is $q_- = q_1\tan\Theta_B$.

Let us now study the imaginary part of Hamiltonian eigenvalues. Following Azaroff *et al.* (1974, p. 339) and remembering (71), let us calculate an upper bound to the imaginary part of the Hamiltonian eigenvalues. Our calculation starts from the following relations:

$$\begin{aligned} 2\,\mathrm{Im}^2(\pi\lambda_\pm/\Lambda_e) &= |q_-^2 + \nu^2| - \mathrm{Re}(q_-^2 + \nu^2) \\ &\le |q_-^2| + |\nu^2| - \mathrm{Re}(q_-^2 + \nu^2) \\ &= 2\,\mathrm{Im}^2(q_-) + 2\,\mathrm{Im}^2(\nu) \\ &\le \mathrm{Im}^2(\beta_g)/2 + K^2\,\mathrm{Im}^2(\chi_o)/(2\gamma_o\gamma_g) \\ &= \frac{(\gamma_o + \gamma_g)^2K^2\,\mathrm{Im}^2(\chi_o)}{8\gamma_o^2\gamma_g^2}, \end{aligned} \tag{90}$$

where $\nu^2 = \nu_o\nu_g$ and $\mathrm{Im}^2\sqrt{\chi_g\chi_{\bar{g}}} \le \mathrm{Im}^2(\chi_o)$ is assumed. Finally, the upper bound is

$$|\mathrm{Im}(\pi\lambda_\pm/\Lambda_e)| \le \frac{(\gamma_o + \gamma_g)K|\mathrm{Im}(\chi_o)|}{4\gamma_o\gamma_g}. \tag{91}$$

We must now evaluate the $\mathbf{D}(\mathbf{r})$ absorption, which, according to (57), includes the $\exp(-i\kappa z)$ factor. Since

$$\mathrm{Im}(\kappa) = \frac{(\gamma_o + \gamma_g)K\,\mathrm{Im}(\chi_o)}{4\gamma_o\gamma_g} < 0, \tag{92}$$

then $\mathrm{Im}(\pi\lambda_\pm/\Lambda_e) + \mathrm{Im}(\kappa) < 0$ and $\mathbf{D}(\mathbf{r})$ decays, no matter which $|\pm\rangle$ wavefield combination $\mathbf{D}(\mathbf{r})$ is.

7.2. Bragg geometry

In reflection geometry (Bragg), $\gamma_g < 0$. If, additionally, the crystal is cut symmetrically, in other words, if the crystal surface coincides with the diffraction planes, $\mathbf{g}\hat{\mathbf{z}} = -g$. Hence,

$$\mathbf{K}_g\hat{\mathbf{z}} = -\mathbf{K}_o\hat{\mathbf{g}} - g = \mathbf{K}_o\hat{\mathbf{g}} = -\mathbf{K}_o\hat{\mathbf{z}}, \tag{93}$$

so that $\gamma_o = -\gamma_g = \sin\Theta_B$, and $\beta_g = -\chi_o K/\sin\Theta_B$. In this case, $K_{g2} = K_{o2} = 0$ because the z axis belongs to the reflection plane and the x_1 axis is parallel to the intersection between the crystal surface and the reflection plane. Additionally, the deviation parameter is $q_- = q_1/\tan\Theta_B - \chi_o K/(2\sin\Theta_B)$. Finally, the absorption term, $\mathrm{Im}(\kappa)$, in (61) is zero.

To study the imaginary parts of the Hamiltonian eigenvalues, we need a lower bound to them. Again, our calculation starts from

$$\begin{aligned} 2\,\mathrm{Im}^2(\pi\lambda_\pm/\Lambda_e) &= |q_-^2 + \nu^2| - \mathrm{Re}(q_-^2 + \nu^2) \\ &\ge |q_-^2| - |\nu^2| - \mathrm{Re}(q_-^2 + \nu^2) \\ &= 2\,\mathrm{Im}^2(q_-) - 2\,\mathrm{Re}^2(\nu) \\ &\ge \mathrm{Im}^2(\beta_g)/2 - K^2\,\mathrm{Im}^2(\chi_o)/(2\gamma_o|\gamma_g|) \\ &= \frac{(\gamma_o - |\gamma_g|)^2K^2\,\mathrm{Im}^2(\chi_o)}{8\gamma_o^2\gamma_g^2} \end{aligned} \tag{94}$$

because, with $\gamma_g \le 0$,

$$\mathrm{Re}^2(\nu) = \frac{K^2\,\mathrm{Im}^2(\sqrt{\chi_g\chi_{\bar{g}}})}{4\gamma_o|\gamma_g|} \le \frac{K^2\,\mathrm{Im}^2(\chi_o)}{4\gamma_o|\gamma_g|}. \tag{95}$$

Finally, the required lower bound is

$$|\mathrm{Im}(\pi\lambda_\pm/\Lambda_e)| \ge \left|\frac{(\gamma_o - |\gamma_g|)K\,\mathrm{Im}(\chi_o)}{4\gamma_o\gamma_g}\right|. \tag{96}$$

Since the $\mathbf{D}(\mathbf{r})$ absorption is now dictated by

$$\mathrm{Im}(\kappa) = -\frac{(\gamma_o - |\gamma_g|)K\,\mathrm{Im}(\chi_o)}{4\gamma_o|\gamma_g|}, \tag{97}$$

the free-space field at $z = 0$ (the initial condition of Takagi's equations) must be orthogonal to the wavefield corresponding to $\mathrm{Im}(\pi\lambda_\pm/\Lambda_e) + \mathrm{Im}(\kappa) > 0$, which is forbidden. We can express this orthogonality requirement by

$$\begin{aligned} &\begin{bmatrix} \eta \pm \sqrt{\eta^2 + 1} & -1 \end{bmatrix}\begin{bmatrix} \tilde{D}_o^e(\mathbf{q}; 0) \\ \sqrt{\gamma/\sigma}\tilde{D}_g^e(\mathbf{q}; 0) \end{bmatrix}_0 \\ &\quad = (\eta \pm \sqrt{\eta^2 + 1})\tilde{D}_o^e(\mathbf{q}; 0) - \sqrt{\gamma/\sigma}\tilde{D}_g^e(\mathbf{q}; 0) \\ &\quad = 0. \end{aligned} \tag{98}$$

Consequently, the $\tilde{D}_o^e$ and $\tilde{D}_g^e$ components of the free-space field must satisfy the relationship $\tilde{D}_g^e(\eta; 0) = \mathcal{R}(\eta)\tilde{D}_o^e(\eta; 0)$, where (Authier, 2001, p. 173)

$$\mathcal{R}(\eta) = \sqrt{\sigma/\gamma}\,(\eta \pm \sqrt{\eta^2 + 1}), \tag{99}$$

and the plus/minus sign is set according to the sign that makes $\mathrm{Im}(\pi\lambda_\pm/\Lambda_e) + \mathrm{Im}(\kappa) > 0$. Accordingly, in Bragg's geometry, the free-space field is a superposition of the incoming field propagating in the $\mathbf{K}_o$ direction and of a reflected field

propagating in the $\mathbf{K}_g$ direction. The presence of the reflected field is a consequence of the crystal-field extinction, required for $z \to \infty$.

7.3. Bragg–Bragg geometry

We consider again the Bragg geometry, but now let the crystal be a plane-parallel infinite slab with thickness t. The absence of any free-space field propagating backwards and entering the crystal at $z = t$ presupposes that $\tilde{\varphi}_g(z = t) = 0$. Since $|\varphi(z = t)\rangle = \mathbf{U}_0(t)|\varphi(z = 0)\rangle$, the above condition implies

$$\begin{aligned} T(\eta; t)\tilde{\varphi}_o(\mathbf{q}; 0) - \mathrm{i}R(\eta; t)\tilde{\varphi}_g(\mathbf{q}; 0) &= \tilde{\varphi}_o(\mathbf{q}; t) \\ -\mathrm{i}R(\eta, t)\tilde{\varphi}_o(\mathbf{q}; 0) + T(-\eta, t)\tilde{\varphi}_g(\mathbf{q}; 0) &= 0, \end{aligned} \tag{100}$$

from which we obtain

$$\tilde{D}_g^e(\mathbf{q}; 0) = \frac{\mathrm{i}\sqrt{\sigma/\gamma}R(\eta, t)}{T(-\eta, t)}\tilde{D}_o^e(\mathbf{q}; 0) \tag{101}$$

and

$$\tilde{D}_o(\mathbf{q}; t) = \frac{\exp[-\mathrm{i}(\beta_g/2 - q_+)t]}{T(-\eta, t)}\tilde{D}_o^e(\mathbf{q}; 0), \tag{102}$$

in which we used (84). It should be observed that

$$\lim_{t\to\infty} \frac{\mathrm{i}R(\eta, t)}{T(-\eta, t)} = \frac{1}{\eta \mp \sqrt{\eta^2 + 1}} = \eta \pm \sqrt{\eta^2 + 1}, \tag{103}$$

where we used (87). Therefore, when crystal thickness tends to infinity, (101) reduces to (99), as expected.

8. Conclusions

As regards Laue's geometry, a description of coplanar X-ray diffraction in a symmetrically cut crystal was given in terms of an optical transfer matrix by Mana & Vittone (1997). We have extended here that description to Bragg's geometry, to asymmetrically cut crystals, and to non-coplanar geometry. We have also treated the Laue and Bragg cases on an equal footing. An advantage of our approach is to provide, through common mathematics, a treatment of the dynamics of X-rays similar to that of other classic and quantum systems. In such a way, the analogy between X-ray scattering by crystals and atom scattering by electromagnetic gratings (in which light and matter exchange their roles) is made explicit. An immediate outcome of our present approach is a new method to solve Takagi's equations, in which method we use their formal identity with the Schrödinger representation of Dirac's equation for a two-level quantum system. This quantum-mechanics formalism promises further benefits in the investigation into X-ray dynamics in deformed crystals. Likewise, the description of n-beam diffraction could benefit from such techniques. Another potential application concerns the study of X-ray diffraction when the Bragg angle is close to $\pi/2$ and the study of Fabry–Pérot resonators for X-rays. Last but not least is application to the study of partially coherent X-rays, which correspond to a quantum-mechanics mixture. In this case, our formalism leads to a description of the dynamics of incoherent X-rays in terms of a density matrix.

APPENDIX *A*
List of the main symbols

$\mathbf{k}$	Complex wavevector of plane waves in crystals
$\mathbf{k}_n$, $n = o, g$	Origin of the reciprocal space in crystals, complex wavevectors of basis plane waves, equation (42)
$\mathbf{p} = \mathbf{k} - \mathbf{k}_n$	Resonance error
$\mathbf{K}$	Real wavevector of plane waves *in vacuo*
$\mathbf{K}_n = K\,\widehat{\mathrm{Re}(\mathbf{k}_n)}$	Origin of the reciprocal space *in vacuo* ($K = 2\pi/\lambda$), real wavevectors of basis plane waves, equation (45)
$p_n = \mathbf{p}\hat{\mathbf{K}}_n$	Resonance-error component along $\mathbf{K}_n$, equation (38)
$\hat{\mathbf{z}}$	Normal to the crystal surface
$\mathbf{x}$	Two-dimensional position vector in the crystal surface
$\hat{\mathbf{x}}_1$	Intersection between crystal surface and reflection plane
$\hat{\mathbf{x}}_n$	Unit component of $\mathbf{K}_n$ in the crystal surface, equation (45)
ψ_n	Angle between $\hat{\mathbf{z}}$ and $\mathbf{K}_n$
$\gamma_n = \cos\psi_n$	Direction cosine, $\gamma_o > 0$, equations (9), (42) and (45)
$\gamma = \gamma_g/\gamma_o$	Asymmetry
$\mathbf{q} = \mathbf{p} - p_z\hat{\mathbf{z}}$	Resonance error in the crystal surface, equation (45)
$q_n = \mathbf{q}\hat{\mathbf{x}}_n$	Resonance error along $\hat{\mathbf{x}}_n$, equation (45)
$q_1 = \mathbf{q}\hat{\mathbf{x}}_1$	Resonance error along the x_1 axis
$2\mathbf{v}_\pm = \mathbf{x}_o \tan\psi_o \pm \mathbf{x}_g \tan\psi_g$	Total and differential velocities of field components, equation (55)
$q_- = \mathbf{q}\mathbf{v}_- + \beta_g/2$	Deviation parameter, equations (55) and (67)
$q_+ = \mathbf{q}\mathbf{v}_+$	Modified resonance-error component, equation (55)
Λ_e	*Pendellösung*/extinction length, equation (64)
$\eta = \Lambda_e q_-/\pi$	Reduced deviation parameter
$\zeta = \pi z/\Lambda_e$	Reduced propagation parameter
$\nu_n = \chi_{\mp g}K/(2\gamma_n)$	Coupling coefficients, equation (47)
$\nu = \sqrt{\nu_o\nu_g}$	Geometric mean of coupling coefficients, equation (90)
$\beta_g/2$	Deviation from Bragg incidence, equations (47) and (67)
$\sqrt{\sigma} = \sqrt{\chi_g/\chi_{\bar{g}}}$	Phase shift between the field components (canonical representation), equation (61)
κ	Modified z component of $\mathbf{k}_o$, equation (57)
D_n, $\tilde{D}_n$	Field components (O–G representation), equation (48)
φ_n, $\tilde{\varphi}_n$	Field components (canonical representation), equation (61)
$\mathbf{H}$, $\mathbf{H}_0$	Hamiltonian operator, equation (51)
$\tilde{H}$	Reciprocal-space O–G representation of $\mathbf{H}$, equation (55)
$\tilde{H}_0$	Reciprocal-space canonical representation of $\mathbf{H}$, equation (76)
$\tilde{H}_\pm$	Reciprocal-space diagonal representation of $\mathbf{H}$, equation (78)
$\lambda_\pm$	Hamiltonian eigenvalues, equation (78)
$\|\pm\rangle$	Hamiltonian eigenvectors, equation (77)
$\mathbf{U} = \exp(-\mathrm{i}\mathbf{H}z)$	Propagation operator, equation (56)
$\tilde{U}_0$	Reciprocal-space canonical representation of $\mathbf{U}$, equation (81)

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