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Time-Dependent X-ray Bragg Diffraction

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Abstract

The theory of time-dependent X-ray Bragg diffraction by crystals is developed on the basis of the Green-function (point-source) formalism. A general case of incident radiation partially coherent in time and space is considered. The time-delay effect of the diffracted radiation is described when the ultrashort time duration incident pulse strikes the crystal surface. The problem in question is closely connected with the effect of time delay in the resonance scattering of synchrotron radiation by nuclei in a crystal. It is found that, for the case where the incident wave is plane (or is an incoherent superposition of plane waves) and the time-dependent pulse is a pseudo δ function in time, the instantaneous crystal reflectivity is a smooth temporal function and tends to the value corresponding to the integrated reflectivity calculated by means of the conventional dynamical–kinematical X-ray diffraction theory. If the incident X-ray pulse profile is a pseudo δ function in both time and space, the temporal crystal response has the same functional dependence as the spatial distribution of the diffracted intensity under the condition of conventional Bragg diffraction of the X-ray beam with lateral width $< t_oc$, where the time delay t_o is equal to $\Lambda/2\pi c$ and $(\mu_o c)^{-1}$ in the cases of dynamical and kinematical X-ray scattering within a crystal, respectively (Λ is the X-ray extinction length, μ_o is the linear absorption coefficient and c is the velocity of light in vacuum).

1. Introduction

The production of ultrashort X-ray pulse sources by the interaction of intense laser pulses with solid targets is an important current topic in plasma and X-ray physics (Uschmann *et al.*, 1995). From a physical viewpoint, it is of great interest to investigate how the Bragg reflectivity of a crystal depends on the X-ray-source time duration (or, in the case of partial time coherence of the incident radiation, the corresponding correlation length). This problem is briefly discussed by He & Wark (1993) for the case where the input pulse is a δ function in time (the input-pulse time duration is very short in comparison with the time parameter t_o characteristic to the diffraction phenomenon) and several examples of the time dependence of the output X-ray pulse were given by He & Wark (1993) but without any details of calculations of the time-dependent dynamical or kinematical Bragg diffraction.

On the other hand, the problem in question is closely connected with the effect of time delay in the resonance scattering of synchrotron radiation by nuclei in a crystal. The theory of the latter was first elaborated by Kagan, Afanasev & Kohn (1978, 1979). After several attempts, the successful observation of the decay of resonant nuclei following excitation by the short pulse of synchrotron radiation was done by van Bürck, Siddons, Hastings, Bergmann & Hollatz (1992).

Thermal neutrons may also be used in place of X-rays and offer another opportunity to observe the time-delay effect since in this case the value of the characteristic time t_o increases.

The aim of the present paper is to present the theory of the time-dependent X-ray Bragg diffraction by a thick (semi-infinite) crystal, expressed in the Green-function (point-source) formalism. The time-dependent Takagi–Taupin equations allow the general solution of this problem to be derived in a form suitable for physical analysis, *i.e.* where the crystal reflectivity is expressed as a function of time and the spatial distribution of the incident radiation.

2. Formulation and general solution of the problem

Within a perfect crystal oriented close to a single Bragg position with the diffraction vector $\mathbf{h}$, the X-ray Bloch-wave field is given by a coherent superposition of the transmitted wave $D_o(\mathbf{r},t)\exp[i\mathbf{k}_o\mathbf{r}-i(2\pi/\lambda)ct]$ and the diffracted wave $D_h(\mathbf{r},t)\exp[i\mathbf{k}_h\mathbf{r}-i(2\pi/\lambda)ct]$ and is governed by the time-dependent Takagi–Taupin equations [*cf.* equation (2.1) in the paper by Chukhovskii, Gabrielyan & Petrashen' (1978)]:

$$\begin{aligned} &i(\lambda/\pi)\gamma_o(\partial D_o/\partial s_o)+\chi_o D_o+\chi_{-h}CD_h \\ &\quad +i(\lambda/\pi)(\partial D_o/\partial T)=0, \\ &i(\lambda/\pi)|\gamma_h|(\partial D_h/\partial s_o)+\chi_o D_h+\chi_h CD_o \\ &\quad +i(\lambda/\pi)(\partial D_h/\partial T)=0. \end{aligned} \tag{1}$$

Here, χ_o and χ_h (χ_{-h}) are the zero and $\mathbf{h}$ $(-\mathbf{h}\ )$ Fourier components of the electric susceptibility of a crystal; $k_o=k_h=2\pi/\lambda$, λ is the vacuum wavelength; γ_o and γ_h are the direction cosines, $\gamma_o=\cos\varphi_o\equiv\cos(\mathbf{k}_o,\mathbf{n}), \gamma_h=\cos(\varphi_h)\equiv\cos(\mathbf{k}_h,\mathbf{n})$ and $\mathbf{n}$ is the inward normal to the entrance surface of a crystal; C is the polarization factor, where $C = 1$ for σ-polarized radiation and $C = \cos(2\vartheta)$ for π-polarized radiation, respectively. ϑ is the Bragg angle and the variable $T = ct$ is introduced.

The relationship between the oblique-angled coordinate system $(s_o 0 s_h)$ and the Cartesian one $(x0z)$, with the x axis along the crystal surface (the z axis is along n), is given by

$$\begin{aligned} z &= s_o - s_h, \\ x &= \tan(\varphi_o)s_o + |\tan(\varphi_h)|s_h. \end{aligned} \tag{2}$$

In our case, the boundary conditions are

$$\begin{aligned} D_o(x,z,T)|_{z=0} &= D_{\rm inc}(x,0,T), \\ D_h(x,z,T)|_{z=\infty} &= 0. \end{aligned} \tag{3}$$

Here, the amplitude of the X-ray incident wave on the entrance surface $D_{\rm inc}$ takes the general form

$$D_{\rm inc}(x,0,T) = D(s_h)F(T - s_o/\gamma_o)|_{s_o=s_h}, \tag{4}$$

where the functions $D(s_h)$ and $F(T - s_o/\gamma_o)$ describe the space and time dependence of the incident wave packet propagating in vacuum assuming $\mathbf{k}_0\mathbf{k}_h = 0$.

For example, the function $D(s_h)$ for incident plane and spherical waves, respectively, is

$$D(s_h) = \exp[i2\pi \sin(2\vartheta)\Delta\varphi_o s_h/\lambda|\gamma_h|], \tag{5}$$

$$D(s_h) = \delta(s_h), \tag{6}$$

where $\Delta\varphi_o$ is the angular deviation from the exact Bragg angle and $\delta(s_h)$ is a δ function.

If, furthermore, the input X-ray pulse is a δ function in time (the pulse duration length is much smaller than the characteristic length $T_o = t_o c$ of the diffraction phenomenon), the F function can be written simply as

$$F(T - s_o/\gamma_o) = \delta(T - s_o/\gamma_o). \tag{7}$$

Here, the phrase 'pulse is a δ function in time' refers to a mathematical model where a monochromatic wave train exists for an infinitesimally short time. For this, the associated spatial wave distribution $D(s_h)$ adopted can be arbitrary.

In a general case, any F function can be expressed as the Fourier integral

$$\begin{aligned} F(T - s_o/\gamma_o) = (1/2\pi) \int\limits_{-\infty}^{\infty} {\rm d}\omega\, F(\omega) \\ \times \exp(i\omega s_o/\gamma_o - i\omega T) \end{aligned} \tag{8}$$

and by the standard definition the Fourier transform $F(\omega)$ is

$$F(\omega) = \int\limits_{-\infty}^{\infty} {\rm d}T\, F(T)\exp(i\omega T).$$

It is clearly seen by performing the Fourier transformation of (1) over the variable T that this problem may be reduced to the solution of the ordinary Takagi–Taupin equations. As is well known, this solution may be obtained with the Green-function formalism (Chukhovskii, 1981). The solution finally obtained for the Bragg diffracted wave on the entrance surface is

$$\begin{aligned} D_h(s_h, s_h, T) = i\chi_h(c/2\lambda|\gamma_h|) \int\limits_{-\infty}^{\infty} {\rm d}\omega\, \exp(-i\omega T) \\ \times \int\limits_{-\infty}^{\infty} {\rm d}s'_h\, G_{ho}(s_h - s'_h, \omega) \\ \times D(s'_h)F(\omega)\exp(i\omega s'_h/\gamma_o), \end{aligned} \tag{9}$$

where the Green function G_{ho} has the form (Chukhovskii, 1981)

$$\begin{aligned} G_{ho}(s_h - s'_h, \omega) = 2\frac{J_1(2\sigma(s_h - s'_h))}{2\sigma(s_h - s'_h)} \exp[i\pi(\chi_o/\lambda) \\ \times (1/\gamma_o + 1/|\gamma_h|)(s_h - s'_h) \\ + i\omega(1/\gamma_0 + 1/|\gamma_h|)(s_h - s'_h)] \\ \times \theta(s_h - s'_h). \end{aligned} \tag{10}$$

Here, $J_1(...)$ is the Bessel function of first order; $\theta(...)$ is the step function; the complex dynamical coefficient is

$$\sigma = (\pi/\Lambda)(1 + ik); \tag{11}$$

Λ is the X-ray extinction length, $\Lambda = \lambda(\gamma_o|\gamma_h|)^{1/2} \times [|C|{\rm Re}^{1/2}(\chi_{-h}\chi_h)]^{-1}$ and k is the dynamical absorption coefficient, $|k| \ll 1$.

With (10), the integration over the variable ω in (9) can be carried out analytically and finally gives

$$\begin{aligned} D_h(s_h, s_h, T) = i\chi_h(c\pi/\lambda|\gamma_h|) \int\limits_{-\infty}^{\infty} {\rm d}s'_h\, D(s'_h) \\ \times F(T - s_h/\gamma_o - (s_h - s'_h)/|\gamma_h|) \\ \times 2\frac{J_1(2\sigma(s_h - s'_h))}{2\sigma(s_h - s'_h)} \exp[i\pi(\chi_o/\lambda) \\ \times (1/\gamma_o + 1/|\gamma_h|)(s_h - s'_h)] \\ \times \theta(s_h - s'_h). \end{aligned} \tag{12}$$

The analysis of the problem based on (12) will be described in §3. Notice that in the case of kinematical Bragg diffraction the basic formula (12) is simplified by setting $\sigma = 0$ throughout.

3. Ultrashort input X-ray pulse

In the case when the X-ray duration length is much shorter than the characteristic length of all the other terms within the integrand of (12), an input X-ray pulse may be approximated as a δ function in time, *i.e.*

$$\begin{aligned} F(T - s_h/\gamma_o - (s_h - s'_h)/|\gamma_h|) \\ = \delta(T - s_h/\gamma_o - (s_h - s'_h)/|\gamma_h|). \end{aligned} \tag{13}$$

Hence, one obtains directly

$$\begin{aligned} D_h(s_h, s_h, T) = {}& i\chi_h c\pi/\lambda D(s_h - (T - s_h/\gamma_o)|\gamma_h|) \\ & \times \theta(T - s_h/\gamma_o) \\ & \times \theta(s_h - (T - s_h/\gamma_o)|\gamma_h|) \\ & \times 2\frac{J_1(2\sigma(T - s_h/\gamma_o)|\gamma_h|)}{2\sigma(T - s_h/\gamma_o)|\gamma_h|} \\ & \times \exp[i\pi(\chi_o/\lambda)(1/\gamma_o + 1/|\gamma_h|) \\ & \times (T - s_h/\gamma_o)|\gamma_h|]. \end{aligned} \quad (14)$$

The temporal dependence of instantaneous crystal reflectivity is given by

$$\begin{aligned} I_h^t \equiv {}& (|\gamma_h|/\gamma_o)(\textstyle\int ds_h\, |D_h|^2)/(\int ds_h |DF|^2) \\ = {}& 4(|\gamma_h|/\gamma_o)2\pi\tau(1/\gamma_o)|\chi_h c\pi/\lambda|^2 \\ & \times \int\limits_{T\gamma_o/(1+\gamma_o/|\gamma_h|)}^{T\gamma_o} \left| \frac{J_1(2\sigma(T - s_h/\gamma_o)|\gamma_h|)}{2\sigma(T - s_h/\gamma_o)|\gamma_h|} \right|^2 \\ & \times \exp[-2\pi(\operatorname{Im}\chi_o/\lambda)(1/\gamma_o + 1/|\gamma_h|) \\ & \times (T - s_h/\gamma_o)|\gamma_h|]. \end{aligned} \quad (15)$$

The derivation of (15) assumes for simplicity that the wave incident on the crystal surface is plane, *i.e.* as described by (5), and τ is the duration length. Notice that the lower and upper limits of integration in (15) are set by the finite velocity c of the X-ray pulse propagation within a crystal diffractor.

The formula (15) can be rewritten in a more suitable form for practical analysis, *i.e.*

$$\begin{aligned} I_h^t = {}& (2\pi\tau/\gamma_o)|\chi_h c\pi/\lambda|^2 \\ & \times \int\limits_0^{\tilde{T}} du\, 4|J_1(2\sigma u)/2\sigma u|^2 \\ & \times \exp[-2\pi(\operatorname{Im}\chi_o/\lambda)(1/\gamma_o + 1/|\gamma_h|)u], \end{aligned} \quad (16)$$

where the limit $\tilde{T}$ in the integrand is given by $\tilde{T}^{-1} = T^{-1}(1/\gamma_o + 1/|\gamma_h|)$.

According to (16), it follows that, in the case of the ultrashort input pulse when its duration length is much smaller than the characteristic length of the diffraction scattering {recall that for the dynamical [kinematical] Bragg diffraction it is the extinction [absorption] length $\Lambda/2\pi$ $[\mu_o^{-1}]$ $\{\mu_o = [2\pi(\operatorname{Im}\chi_o/\lambda)(1/\gamma_o + 1/|\gamma_h|)]\}$}, the instantaneous crystal response has the shape of the saturation phenomenon. Further, if the upper limit of a time is such that $T^{-1}(1/\Lambda_o + 1/|\gamma_h|) \ll (\lambda/2\pi)^{-1}$ as well, the instantaneous crystal response tends to the constant value corresponding to the integrated Bragg reflectivity in the conventional dynamical–kinematical theory [CDKT, see Pinsker (1978)].

In the dynamical diffraction case, the typical behavior of I_h^t is shown in Fig. 1. For this, the saturation time of the I_h^t curve is defined by the extinction length $\Lambda/2\pi$. It is worth saying that the reflectivity I_h^t does not depend on the angular deviation $\Delta\varphi_o$ of the incident plane wave and, hence, keeps the same shape after integration over the whole angular range $\delta\Delta\varphi_o$ of an input pulse.

4. Partially coherent incident wave packet

From (12), the instantaneous amplitude of the diffracted wave on the entrance surface is

$$\begin{aligned} D_h(s_h, s_h, T) = {}& i\chi_h(c\pi/\lambda|\gamma_h|) \int\limits_{-\infty}^{\infty} ds_h'\, D(s_h') \\ & \times F(T - s_h/\gamma_o - (s_h - s_h')/|\gamma_h|) \\ & \times G_{ho}(s_h - s_h'), \end{aligned} \quad (17)$$

where the Green function G_{ho} is independent of time and is equal to

$$\begin{aligned} G_{ho}(s_h - s_h') = {}& 2\frac{J_1(2\sigma(T - s_h/\gamma_o)|\gamma_h|)}{2\sigma(T - s_h/\gamma_o)|\gamma_h|} \\ & \times \exp[i\pi(\chi_o/\lambda)(1/\gamma_o + 1/|\gamma_h|) \\ & \times (s_h - s_h')]\theta(s_h - s_h'). \end{aligned} \quad (18)$$

In the general case, the instantaneous intensity distribution $I_h(s_h, s_h, T)$ along the crystal surface is given by

$$\begin{aligned} I_h(s_h, s_h, T) \equiv {}& \langle |D_h(s_h, s_h, T)|^2 \rangle \\ = {}& |\chi_h c\pi/\lambda\gamma_h|^2 \int\limits_{-\infty}^{\infty} ds_h' \int\limits_{-\infty}^{\infty} ds_h'' \\ & \times \langle D(s_h')D^*(s_h'') \rangle \\ & \times \langle F(T - s_h/\gamma_o - (s_h - s_h')/|\gamma_h|) \\ & \times F^*(T - s_h/\gamma_o - (s_h - s_h'')/|\gamma_h|) \rangle \\ & \times G_{ho}(s_h - s_h')G_{ho}^*(s_h - s_h''), \end{aligned} \quad (19)$$

where $\langle \ldots \rangle$ means the average over the statistical ensemble describing the features of the partial space and time coherence of the incident radiation.

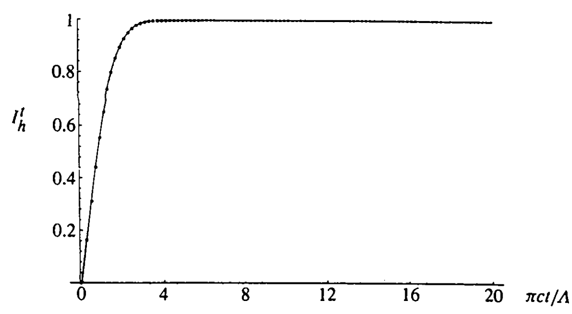

Fig. 1. Instantaneous crystal reflectivity [normalized by the value I_h^t $(t = \infty)$]. The incident wave is plane in space and a δ function in time [λ = 3.151 Å, Bragg angle 74.18° and the numerical coefficients corresponding to the Si 311 reflection are taken from Uschmann *et al.* (1995)].

As a further example, the case of the incident spherical wave (6) is considered. Then, (19) reduces to

$$\begin{aligned} I_h(s_h, s_h, T) = {} & |\chi_h c\pi/\lambda\gamma_h|^2 G_{ho}(s_h) G^*_{ho}(s_h) \\ & \times \langle F(T - s_h(1/\gamma_o + 1/|\gamma_h|)) \\ & \times F^*(T - s_h(1/\gamma_o + 1/|\gamma_h|))\rangle. \quad (20) \end{aligned}$$

In the case under consideration, the intensity of the incident radiation is given by

$$I_o(s_h, s_h, T) = |\delta(s_h)|^2 \langle F(T - s_h/\lambda_o) F^*(T - s_h/\gamma_o)\rangle. \quad (21)$$

Comparison of (20) with (21) shows that the intensity of the diffracted radiation is defined by the same time correlation function but with the single difference that there is a shift in the argument owing to the X-ray propagation inside a crystal.

With the Fourier transformation, the statistically averaged time-dependent functions in (20) and (21) can be written as

$$\begin{aligned} & \langle F(T - s_h/\gamma_o) F^*(T - s_h/\gamma_o)\rangle \\ & \quad = (1/2\pi)^2 \textstyle\int \mathrm{d}\omega' \int \mathrm{d}\omega'' \exp[-i\omega'(T - s_h/\gamma_o)] \\ & \qquad \times \exp[i\omega''(T - s_h/\gamma_o)] \\ & \qquad \times \Gamma(\omega' - \omega'')|F((\omega' + \omega'')/2)|^2, \quad (22a) \end{aligned}$$

$$\begin{aligned} & \langle F(T - s_h(1/\gamma_o + 1/|\gamma_h|)) \\ & \quad \times F^*(T - s_h(1/\gamma_o + 1/|\gamma_h|))\rangle \\ & \quad = (1/2\pi)^2 \textstyle\int \mathrm{d}\omega' \int \mathrm{d}\omega'' \\ & \qquad \times \exp\{-i\omega'[T - s_h(1/\gamma_o + 1/|\gamma_h|)]\} \\ & \qquad \times \exp\{i\omega''[T - s_h(1/\gamma_o + 1/|\gamma_h|)]\} \\ & \qquad \times \Gamma(|\omega' - \omega''|)|F((\omega' + \omega'')/2)|^2. \quad (22b) \end{aligned}$$

In (22), we have introduced the spectral autocorrelation function $\Gamma(|\omega' - \omega''|)$, which describes the partial coherence phase of incident radiation.

It is clear that, in the case when the phase correlation frequency ω_o [$\omega_o = \int \mathrm{d}\omega\ \Gamma(\omega)$] goes to zero, completely incoherent in the time incident wave, from (22) one obtains the conventional Bragg diffraction solution for the incident X-ray spherical wave [*cf.* (18)].

In the opposite case where the phase correlation frequency ω_o tends to infinity (Γ function assumed to be constant), *i.e.* the case where the incident radiation is fully phase coherent, and if the Fourier components $F(\omega) \simeq$ constant (this is equivalent to the assumption for the ultrashort input pulse) the direct calculations using (20)–(22) yield

$$\begin{aligned} I^t_h = {} & \frac{|\gamma_h| \int \mathrm{d}s_h\ I_h(s_h, s_h, T)}{\gamma_o \int \mathrm{d}s_h\ I_o(s_h, s_h, T)|_{T \Rightarrow 0}} \\ = {} & |\chi_h c\pi^2 \Delta s_h/\lambda|^2 (\gamma_o)^{-1} (\gamma_o + |\gamma_h|)^{-1} \\ & \times 4\left|\frac{J_1(2\sigma T(1/\gamma_o + 1/|\gamma_h|)^{-1})}{2\sigma T(1/\gamma_o + 1/|\gamma_h|)^{-1}}\right|^2 \\ & \times \exp[-2\pi(\mathrm{Im}\,\chi_o/\lambda)T]. \quad (23) \end{aligned}$$

{The derivation of (23) assumes that the incident-wave amplitude along the crystal surface $D(s_h)$ depends on the coordinate s_h according to a Lorentzian profile of width Δs_h [*cf.* (6)].}

It is interesting that now the instantaneous reflectivity I^t_h has the same functional dependence in time as does the conventional Bragg diffracted intensity distribution along the crystal surface for the incident spherical wave (Pinsker, 1978) with corresponding substitution of variables.

Also notice that the above formula (23) describes the time delay in the resonance scattering of synchrotron radiation by nuclei in a crystal (Kagan, Afanasev & Kohn, 1978, 1979) with the substitution of the Fourier coefficients χ_o, χ_h and χ_{-h} corresponding to the excitation of isomeric nuclear levels.

An example of a calculation using (23) for the same numerical parameters as in Fig. 1 (the dynamical Bragg diffraction) is shown in Fig. 2.

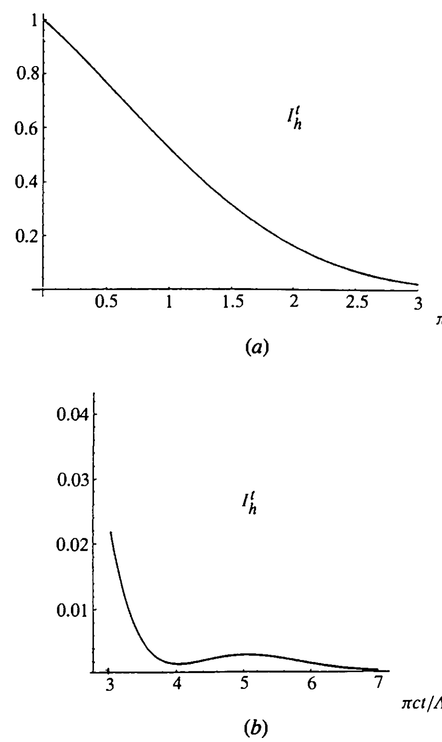

Fig. 2. Instantaneous crystal reflectivity [normalized by the value I^t_h ($t = 0$)]. The incident wave is a δ function in space and in time [$\lambda = 3.151$ Å, Bragg angle 74.18° and the numerical coefficients corresponding to the Si 311 reflection are taken from Uschmann *et al.* (1995)]. Values of I^t_h *vs* $\pi ct/\Lambda$ in regions of $\pi ct/\Lambda$: (*a*) between 0 and 3 and (*b*) between 3 and 7. Note differences in the scales.

5. Concluding remarks

The theory of time-dependent X-ray Bragg diffraction by a crystal is developed allowing for the effects of the partial time and space coherence of the incident beam. It is found that, for the case where the input wave is plane (or is the incoherent superposition of plane waves) and the amplitude is a δ function in time (the ultrashort time pulse approximation), the instantaneous crystal reflectivity is a smooth temporal function. Furthermore, in the limit where the observation time t is much longer than the characteristic value t_o [$t_o = \Lambda/2\pi c$ and $t_o = (\mu_o c)^{-1}$, respectively, for the dynamical and kinematical Bragg diffraction], the crystal reflectivity tends to the value for the integrated Bragg reflectivity calculated by CDKT. If the input X-ray pulse profile is a pseudo δ function in both time and space, the temporal crystal response has a functional dependence identical with that of the spatial distribution of the diffracted intensity under the conventional Bragg diffraction of the X-ray beam with the lateral width $t_o c$.

In the general case, where the input X-ray pulse is partially coherent in time and space, calculations of practical interest can be carried out with the formulae (20)–(22). The important conclusion following from this study is that the temporal crystal response is determined by the characteristic length when the input duration length is much smaller than the latter. So, taking into account that for the X-rays the typical values are $\Lambda/2\pi \simeq 3\,\mu m$ and $\mu_o \simeq 60\,\mu m$, $t_o \simeq 10\,fs$ and $t_o \simeq 200\,fs$ for the dynamical and kinematical Bragg diffraction, respectively. The last value of t_o for the kinematical Bragg diffraction is comparable with the time duration of ultrashort X-ray pulse sources by the interaction of intense laser pulses with solid targets (Uschmann *et al.*, 1995). The present calculations will have potential applications in time-dependent X-ray optics when X-ray pulse sources with a comparable time duration come to be used in practice.

Also notice that thermal neutrons may also be used in place of X-rays and offer another opportunity to observe the time-delay effect since in this case the value of the characteristic time is increased.

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The Enumeration and Symmetry-Significant Properties of Derivative Lattices. III. Periodic Colourings of a Lattice

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Abstract

In the triclinic case, structures that can be described in terms of arrangements of a set number of possible subunits occupying the unit cells of an underlying lattice may be enumerated by their derivative lattice index n and stoichiometry, *e.g.* $X_m Y_{(n-m)}$ for two types of subunits. This process involves counting the number, $H(n, m)$, of such patterns possible on the frame of the colour lattice group, followed by the elimination of any patterns that belong to a derivative lattice of lower index. The resulting numbers, $K(n, m)$, then have the property

$$K(n, m) \leq (1/n)\left[{n \atop m}\right] \leq H(n, m)$$

where $\left[{n \atop m}\right]$ is the binomial coefficient. These expressions are equalities if n and m are mutually prime. $H(n, m)$ and

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