Femtosecond x-ray dynamical diffraction by perfect crystals

S. D. Shastri, P. Zambianchi, D. M. Mills

Advanced Photon Source, Argonne National Laboratory,
Argonne, IL 60439, USA

ABSTRACT

X-ray free-electron lasers (XFELs) designed to operate at $\sim 1 \text{ \AA}$ wavelengths are currently being proposed by several laboratories as the basis for the next (4th) generation of synchrotron radiation sources. The unique radiation properties of these proposed sources, which include 200 fs pulse duration and peak beam brilliance in excess of $10^{33} \text{ photon/(s .1\%-bw mrad}^2 \text{ mm}^2)$, offer the possibility of ultrafast time-resolved experiments, perhaps down to 10 fs resolution levels using pulse compression or slicing techniques. Motivated by such potential applications, this paper addresses the relevant instrumentation issue of perfect crystal dynamical diffraction of ultrashort x-ray pulses when the pulse lengths become comparable to the extinction length scales. The basic calculations reported here show the transient time-dependent diffraction from perfect crystals excited by plane-wave delta-function electromagnetic impulses. Time responses have been calculated for 8 keV photon energy, for reflected and transmitted beams in both Bragg and Laue cases. Interesting diffraction effects arise, and their implications for XFEL optics are discussed.

Keywords: x-ray free-electron lasers, x-ray optics, dynamical diffraction

1. INTRODUCTION

The accelerator-based x-ray free-electron laser (XFEL) sources currently being proposed are the LCLS project¹ at SLAC (USA) and a component of the TESLA project² at DESY (Germany). Both projects aim to produce angstrom-wavelength radiation through the self-amplified spontaneous emission (SASE) process arising when a highly relativistic, dense, low-emittance electron beam passes through a long undulator. The SASE x-rays would be delivered in a $1 \text{ } \mu\text{rad}$ divergence beam of intense, 200 fs bursts having full transverse coherence. Such a source would likely have the greatest impact in time-resolved studies, coherence-relying imaging techniques, and nonlinear x-ray/matter interactions. This paper is motivated by the first type of application mentioned where one could attempt to conduct numerous x-ray diffraction and spectroscopy studies with 200 fs temporal resolution. Furthermore, there are techniques under consideration and development, involving bunch compression³ or slicing^{4,5} optics combined with specially prepared (e.g., chirped) electron beams, that could make it possible to produce pulse durations approaching 10 fs. Exploiting such ultrashort pulses for the pump-probe class of time-resolved studies requires specialized x-ray optics development to provide stable (jitter- and drift-free) components, such as controllable photon "delay lines", beam splitters, polarizers, lenses, etc. In this regard, perfect crystal optics will continue to serve as a valuable instrumentation basis to control x-ray beams. Given this, it is worthwhile to examine the physics of perfect crystal diffraction (i.e., dynamical diffraction theory) in the regime of ultrashort (femtosecond) pulses.

Treatments of dynamical diffraction⁶ are usually presented (like most signal or wave propagation phenomena) in the form of a plane wave theory. This approach, which provides a clear view of the steady-state response to a sinusoidal input, does not immediately present as clear a picture for an ultrashort input pulse. Nevertheless, the transient response to any finite pulse can be straightforwardly derived from the frequency-dependent steady-state behaviors using a Fourier transform method. This approach is used here to obtain

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, make any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

$$\mathbf{E}_{in}(\mathbf{r}, t) = E_{vac} \delta(t - \hat{\mathbf{u}} \cdot \mathbf{r}/c)$$

RECEIVED
NOV 08 2000
OSTI

Figure 1: Illustration of delta-function plane wave pulses incident on crystals in symmetric Bragg (left) and symmetric Laue (right) geometries.

the responses of perfect crystal Bragg reflections to the most instructive and shortest possible excitations, namely, delta-function electric field impulses. The transient responses in the various diffraction channels will be shown to be in the femtosecond temporal regime and therefore of relevance to XFEL optics for time-resolved experiments.

The subject of ultrashort pulse Bragg diffraction has been pursued by other researchers.⁷⁻¹⁰ The treatments did not employ the Fourier transform technique used here, but instead used the so-called time-dependent Tagaki-Taupin formalism, which in original form was developed to handle distorted crystals. In addition to reproducing some of their results (for the case of reflected beams from semi-infinite crystals in Bragg geometry), this paper discusses additional situations such as finite crystals in both Bragg and Laue geometries, looking at transmitted as well as reflected beams.

2. CALCULATION APPROACH

Figure 1 shows an incident delta-function plane wave with electric field $\mathbf{E}_{in}(\mathbf{r}, t) = E_{vac} \delta(t - \hat{\mathbf{u}} \cdot \mathbf{r}/c)$ propagating in the direction of unit vector $\hat{\mathbf{u}}$, which makes an angle θ_B with respect to the diffracting crystal planes associated with the reciprocal lattice vector $\mathbf{H} = (hkl)$. The polarization of the wave, i.e., of the vector $\mathbf{E}_{vac}$, is assumed to be either σ or π , referring to pointing either out of or within the scattering plane, respectively. Only a spectrally narrow range of the broadband delta-function in the vicinity of some photon energy $E_B = \hbar\omega_B$ will satisfy the Bragg condition at angle θ_B . Hence, the resulting transient wave trains in the reflected and transmitted beams will be quasi-monochromatic with mean frequency ω_B . The origin $\mathbf{r} = 0$ is defined as the point on the entrance surface at which the delta-function impinges at exactly time $t = 0$. In the Bragg case, one is interested in the time response of the beam reflected at the same point $\mathbf{r} = 0$ and in the time response of the transmitted beam emerging from the point $\mathbf{r} = \mathbf{r}_D$ positioned on the opposite face directly across from $\mathbf{r} = 0$. In the Laue case, one is interested in the time responses of both the reflected and transmitted beams at the point $\mathbf{r} = \mathbf{r}_D$ located opposite $\mathbf{r} = 0$. Due to the assumption of unlimited transverse spatial dimensions for the crystal surfaces and the delta-function, one can argue by translational invariance that the time dependences of the exit waves from any two points on the same surface are identical, except for a shift in time.

Let $\tilde{E}_{in}(\omega)$ be the temporal Fourier transform of $E_{in}(0, t)$, the time dependence of the incident wave packet $E_{in}(\mathbf{r}, t)$ at the entrance surface spot $\mathbf{r} = 0$, and be given by

$$E_{in}(0, t) = \int_{-\infty}^{+\infty} \tilde{E}_{in}(\omega) e^{i\omega t} d\omega = 2 \operatorname{Re} \int_0^{\infty} \tilde{E}_{in}(\omega) e^{i\omega t} d\omega \quad .$$

Then, for example, if one considers the diffracted wave packet emerging from the point $\mathbf{r}_D$ in the Laue

case, it has a time dependence

$$E_H(\mathbf{r}_D, t) = 2 \operatorname{Re} \int_0^\infty \tilde{E}_{in}(\omega) R(\omega) e^{i\omega t} d\omega = 2 \operatorname{Re} e^{i\omega_B t} \int_{-\infty}^{+\infty} \tilde{E}_{in}(\omega_B + u) R(\omega_B + u) e^{iut} du ,$$

where $u = \omega - \omega_B$ represents the frequency deviation from the Bragg frequency ω_B , and $R(\omega)$ is the frequency-dependent complex reflectivity amplitude

$$R(\omega) = \frac{E_H(\mathbf{r}_D)}{E_0(\mathbf{0})}$$

obtainable from ordinary steady-state, sinusoidal, plane wave dynamical diffraction theory. $R(\omega)$ relates the diffracted wave $E_H(\mathbf{r}_D)e^{i\omega t}$ at $\mathbf{r}_D$ to the incident wave $E_0(\mathbf{0})e^{i\omega t}$ at $\mathbf{r} = \mathbf{0}$, arriving via angle θ_B (i.e., along direction $\mathbf{u}$). The subscripts 0 and H are used to denote forward and diffracted beams, respectively. In determining $R(\omega)$, care must be taken not only to include both the so-called α and β Bloch waves, but also to keep track of the exact temporal phase shifts between $E_0(\mathbf{0})$ and $E_H(\mathbf{r}_D)$ by properly incorporating the wave vectors in the crystal medium. These wave vectors are normally decomposed into the "vacuum value" plus dynamical corrections. One must particularly note that in the Fourier integral, as ω varies about the Bragg reflection frequency ω_B , there is a change in the vacuum value of the wave vectors as well as in the dynamical corrections. The time-dependent diffracted intensity, defined as the square of the electric field amplitude, leaving point $\mathbf{r}_D$ is simply

$$I(t) = \left| 2 \int_{-\infty}^{+\infty} \tilde{E}_{in}(\omega_B + u) R(\omega_B + u) e^{iut} du \right|^2 .$$

For the specific case of the incident delta-function impulse described at the beginning of this section, one has $E_{in}(\mathbf{0}, t) = E_{vac} \delta(t)$ and hence $\tilde{E}_{in}(\omega) = E_{vac}/2\pi$. This results in the transient intensity response being

$$I(t) = \frac{E_{vac}^2}{\pi^2} \left| \int_{-\infty}^{+\infty} R(\omega_B + u) e^{iut} du \right|^2 .$$

If instead of the transient Laue reflected beam discussed so far, one is interested in generalizing to the other diffraction channels, such as the reflected beam of the Bragg case or the transmitted beam (of either the Bragg or Laue cases), the previous mathematical expressions are still correct provided one uses the complex amplitude ratio $R(\omega)$, from steady-state dynamical diffraction theory, appropriate to the exit beam of interest. For the reflected beam of the Bragg case one uses $R(\omega) = E_H(\mathbf{0})/E_0(\mathbf{0})$, and for the transmitted beam one uses $R(\omega) = E_0(\mathbf{r}_D)/E_0(\mathbf{0})$.

Having defined intensity as the square of the field, $I(t)$ has units of (electric field)². The magnitude of the response $I(t)$ will scale with the strength of the incident delta-function impulse, which is determined by E_{vac} . Hence, it makes sense to normalize $I(t)$ by dividing by E_{vac}^2 . So, the normalized time response functions $I_n(t)$ to be presented later are defined as

$$I_n(t) = (\hbar/E_{vac})^2 I(t) = \frac{\hbar^2}{\pi^2} \left| \int_{-\infty}^{+\infty} R(\omega_B + u) e^{iut} du \right|^2$$

and have units of (energy)², since E_{vac} has units of (electric field)×(time).

Since this study is justified by possible applications to XFEL optics, one should ask whether the extreme peak beam intensities (even after a premonochromator that will remove most of the heat load for subsequent crystal optics) result in nonlinear interactions that in any way invalidate the assumptions of the well-known Ewald-von Laue dynamical diffraction theory. Of particular concern is the correctness of the mathematical expressions for the susceptibilities that relate the fields in an atom's vicinity to the induced

Figure 2: The magnitude, real part, and imaginary part of the reflectivity amplitude ratio for the semi-infinite, symmetric Bragg Si(111) reflection at 8 keV.

electronic current density, which are derived using quantum mechanical perturbation theory in low order. It is possible that this first-stage derivation must be refined to include higher order (nonlinear) effects, before proceeding to the second stage of the dynamical diffraction exercise wherein Maxwell's equations are solved in the crystal medium possessing the previously obtained spatially periodic susceptibilities. The present study does not address this issue, but is based on the assumption that the standard theory is still valid.

3. CALCULATION RESULTS

All the time-dependent responses $I_n(t)$ shown in this section are for σ -polarized diffraction from perfect silicon at 8 keV, meaning that a σ -polarized delta-function impulse wave is incident with respect to the diffraction planes Si(hkl) of interest at the Bragg angle θ_B for $\hbar\omega_B = 8$ keV photons. With the exception of one calculation for the Si(444) reflection, this section focuses on Si(111) diffraction in various geometrical cases.

Consider a semi-infinitely thick, symmetric Bragg Si(111) crystal that is impulse-excited at the 8 keV Bragg angle ($\theta_B = 14.3^\circ$). The real and imaginary parts of the frequency response $R(\omega)$ associated with the reflected beam are shown in Fig. 2. Also plotted is $|R(\omega)|^2$, which is simply the well-known Darwin-Prins reflectivity curve (displayed here as a function of energy as opposed to the usually presented angle) having an energy acceptance width $\Delta E = 1.1$ eV. From the uncertainty relation $\Delta E \Delta t / (2.35)^2 > \hbar/2$, one expects the transient reflection to last a few femtoseconds. The exact calculation, based on Fourier transforming $R(\omega)$ as described in the previous section, gives the normalized transient response $I_n(t)$ which is seen in Fig. 3a to decay away in less than 5 fs. In agreement with causality, $I_n(t) = 0$ for $-\infty < t < 0$. At $t = 0$, $I_n(t)$ seems to jump discontinuously. A careful mathematical analysis of time $t = 0^+$ reveals that an instantaneous jump is indeed present due to scattering from the surface atomic layer—but is not up to the maximum intensity value, which is then rapidly attained in a time evolution of less than .05 fs. The semilogarithmic display in Fig. 3b shows a periodic temporal beating among the frequencies within the reflected wave packet's bandwidth.

It is worthwhile repeating the previous calculation, modified to examine a high-order reflection. For 8 keV and silicon, 13 reflections are allowed. Fig. 4 shows the angular acceptances of all these reflections (for

Figure 3: Transient reflected intensity at 8.0 keV from a semi-infinite, symmetric Bragg Si(111) crystal given in linear (a) and semilogarithmic (b) scales.

symmetric Bragg geometry), for both σ - and π -polarized diffraction. For each polarization, the acceptance values lie on two curves depending on whether the reflection is “even” ($h+k+l = 4n$) or “odd” ($h+k+l = 4n \pm 1$), thereby resulting in four curves. Note that with the exception of a couple of reflections in the π -polarization case having near-45° Bragg angles, all the angular acceptances are well above the 1–2 μrad divergence of the SASE radiation of an XFEL. The highest order allowed reflection is Si(444), having $\theta_B = 81.3^\circ$ and an energy acceptance width $\Delta E = 41.4$ meV. Again using $\Delta E \Delta t / (2.35)^2 > \hbar/2$, one estimates the transient reflected intensity to last a few tens of femtoseconds. The exact calculation (Fig. 5) shows a behavior qualitatively similar to the previous Si(111) case, but with the expected longer duration of 100 fs for $I_n(t)$ to subside almost completely.

Figure 4: Angular acceptance (Darwin width) versus Bragg angle for all silicon reflections allowed at 8 keV. Two reflection types (even and odd) and two polarizations (σ and π) result in four curves. The horizontal line represents the divergence of the SASE radiation from an XFEL.

Next, one returns to the Si(111) reflection, but examines it for a thin (thickness $l = 10 \mu\text{m}$) Bragg crystal. The reflected intensity evolution (Fig. 6) closely resembles that of the semi-infinite crystal (Fig. 3),

Figure 5: Transient diffracted intensity at 8.0 keV from a semi-infinite, symmetric Bragg Si(444) reflection.

except for a remarkable artifact at a delay time of $t = 16.5$ fs. This interesting “photon echo,” which becomes earlier and stronger with reduction in the thickness, is due to back-face diffraction and has a simple ray-trace interpretation sketched in the inset of Fig. 6a. A signal (dashed ray), intersecting the entrance surface earlier and displaced to the side of $r = 0$, propagates directly to the back face in such a way that it reflects back to the point $r = 0$ and appears at time $t = \frac{2l}{c}(1/\sin\theta_B - \cos\theta_B/\tan\theta_B) = 16.5$ fs. Such delayed echos will be seen later to also appear in the Laue geometry, and could be of relevance in the development of beam-splitter optics for ultrashort pulses.

Figure 6: Transient 8 keV reflected intensity from a $10\text{ }\mu\text{m}$ thick, symmetric Bragg Si(111) crystal given in linear (a) and (b) semilogarithmic scales. The inset sketch in (a) geometrically depicts the origin of the echo at 16.5 fs.

One might also be interested in the Bragg-transmitted beam leaving the crystal from the point r_D on the opposite face, directly across from the reference point $r = r_0$. For the same $10\text{ }\mu\text{m}$ thick crystal, the transmitted beam's normalized time-dependent intensity $I_n(t)$ is shown in Fig. 7. From the dashed ray path in the inset sketch (Fig. 7a), the earliest instant at which a signal can reach r_D is $\frac{d}{c}\sin\theta_B = 8.2$ fs. The calculated $I_n(t)$ is consistent with this causality argument. Unlike the Bragg-reflected case where far

Figure 7: Transient 8 keV transmitted intensity through a 10 μm thick, symmetric Bragg Si(111) crystal in linear (a) and semilogarithmic (b) scales. The inset sketch in (a) depicts the earliest transmitted radiation at r_D .

off-Bragg energies $\hbar\omega \neq \hbar\omega_B$ have a vanishing contribution to $R(\omega)$, in the Bragg-transmitted case far off-Bragg Fourier components are not completely suppressed but are transmitted in accordance with the ordinary total x-ray attenuation coefficient ($1/\mu_{\text{tot}} = 17.0 \mu\text{m}$). So the transmitted wave packet has an overall broadband spectral background, which in the time domain should result in $I_n(t)$ having a sharp delta-function “spike” superimposed at earliest emergence instant of $t = 8.2$ fs. This prompt spike is not seen in Fig. 7 because it was effectively removed in the frequency domain by subtracting out the nonzero asymptotic behavior of $R(\omega)$ prior to Fourier transformation.

Figure 8: Transient 8 keV intensity reflected by a 50 μm thick, symmetric Laue Si(111) crystal. The inset sketch illustrates the physical origins of the two main flashes.

Proceeding onward to the symmetric Si(111) Laue geometry, Fig. 8 presents $I_n(t)$ for the reflected beam emerging from the point r_D on the exit surface of a 50 μm thick crystal. The two prominent flashes, occurring at times $t = 161.5$ fs and $t = 182.5$ fs, have simple geometrical explanations sketched in the inset of Fig. 8. The dashed ray illustrates the path leading to the earliest signal arrival at the detection point

Figure 9: Transient 8 keV intensity transmitted through a 50 μm thick, symmetric Laue Si(111) crystal.

$\mathbf{r}_D$ at time $\frac{d}{c} \cos \theta_B = 161.5$ fs. The dotted ray path represents the echo that comes out after an additional delay of $\frac{2d}{c} \tan \theta_B \sin \theta_B = 21.0$ fs or at absolute time $t = 161.5$ fs + 21.0 fs = 182.5 fs. Unlike the case of the Bragg reflection echo (Fig. 6), the Laue reflection echo is as strong as its precursor since both signal propagation paths traverse the same distance in the material. The intensity modulation between the two main bursts is reminiscent of the spatial *pendellösung* pattern observed on the exit side of a Laue crystal illuminated with a small, divergent beam of monochromatic x-rays. Here, since the impulse illumination is instead collimated and polychromatic, the spatial *pendellösung* gets mapped onto the time domain.

Finally, the same symmetric Laue geometry is analyzed with regard to the transmitted beam (Fig. 9). The time-dependent transmitted intensity bears strong resemblance to that of the reflected beam. However, the echo at 182.5 fs is strongly suppressed as one would expect, since the echo should radiate primarily into the reflected channel, as implied by its intuitive geometrical path. The earlier discussion, pertaining to the non-vanishing asymptotic value of $R(\omega)$ for the Bragg transmission case, is also valid here, and similarly, the prompt spike has been removed from the Laue-transmitted wave packet.

4. CONCLUSION

Diffraction from perfect crystals has played a major role in x-ray optics for manipulating synchrotron radiation. The plausibility of femtosecond time-resolved experimental studies utilizing XFEL radiation from proposed 4th-generation sources lead to this basic theoretical study of dynamical diffraction of ultrashort x-ray pulses. Specifically, the Fourier transform technique was used to determine the transient diffracted intensities from silicon crystals illuminated by delta-function impulses. These impulse responses (i.e., Green's functions) describe well the intrinsic influence of the diffraction process in the time domain and how time resolution is preserved or degraded. In addition to demonstrating precisely how the pulse is stretched depending on the energy bandwidth of the reflection, the calculations show interesting delayed echos when reflecting from thin Bragg and Laue crystals. Understanding these effects could be of importance in the development of XFEL optical components such as beam splitters. Continued investigations in this subject require refinements of the treatment to include realistic pulses and crystals (in terms of duration, spectral content, divergence, beam size, surface roughness, and thermal strain), propagation through sequential arrangements of crystals, and the possible necessity of incorporating nonlinear x-ray/atom interactions in dynamical diffraction theory.

ACKNOWLEDGMENTS

Valuable insights were gained from discussions with W.-K. Lee and W. Sturhahn of Argonne National Laboratory. This work was supported by the U.S. Department of Energy, Basic Energy Sciences, Office of Science, under Contract No. W-31-109-Eng-38.

REFERENCES

1. *Linac Coherent Light Source (LCLS) Design Study Report*, Report SLAC-R-521, UC-414 (1998), Stanford Linear Accelerator Center, CA 94025, USA.
2. TESLA project information is available via the DESY internet website—<http://www-hasylab.desy.de> (home page) and <http://www-hasylab.desy.de/facility/fel/parameters.htm> (XFEL parameters).
3. R. O. Tatchyn, *Proc. SPIE—Int. Soc. Opt. Eng.* **4143**—this volume (2000). Title and page numbers not available at time of submission.
4. R. Bionta, "A transmissive optics approach for time-slicing the LCLS x-ray pulse," Technical Note LCLS-TN-00-7 (2000), Stanford Linear Accelerator Center, CA 94025, USA.
5. P. H. Bucksbaum and R. D. Merlin, "The phonon Bragg switch: A proposal to generate subpicosecond x-ray pulses," *Solid State Comm.* **111**, 535–539 (1999).
6. B. W. Batterman and H. Cole, "Dynamical diffraction of x-rays by perfect crystals," *Rev. Mod. Phys.* **36**, 681–717 (1964).
7. J. S. Wark and H. He, "Subpicosecond x-ray diffraction," *Laser and Particle Beams* **3**, 507–513 (1994).
8. F. N. Chukovskii and E. Förster, "Time-dependent x-ray Bragg diffraction," *Acta Cryst.* **A51**, 668–672 (1995).
9. T. Missalla, I. Uschmann, E. Förster, G. Jenke, and D. von der Linde, "Monochromatic focusing of subpicosecond x-ray pulses in the keV range," *Rev. Sci. Instrum.* **70**, 1288–1299 (1999).
10. J. S. Wark and R. W. Lee, "Simulations of femtosecond x-ray diffraction from unperturbed and rapidly heated single crystals," *J. Appl. Cryst.* **32**, 692–703 (1999).