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Disclaimer

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Abstract

In order to utilize hard x-ray free electron lasers (XFEL's) when they are extended to attosecond pulse lengths, it is necessary to choose optical elements with minimal response time. Specular grazing incidence optics made of low-Z materials are popular candidates for reflectors since they are likely to withstand x-ray damage and provide sufficiently large reflectivities. Using linear-optics reflection theory, we calculated the transient reflectivity of a delta-function electric pulse from a homogenous semi-infinite medium as a function of angle of incidence for s- and p-polarized light. We specifically considered the pulse response of Be, diamond, silicon carbide, and silicon, all of which are of relevance to the XFEL's that are currently being built. We found that the media emit energy in a damped oscillatory way, and that the impulse-response times are shorter than 0.3 fs for normal incidence. For grazing incidence, the impulse-response time is substantially shorter, making grazing-incidence mirrors a good choice for deep-sub-femtosecond reflective optics.

Introduction

X-ray free-electron lasers (FEL's) can provide photon pulses that are intense, short in duration, and of small wavelength; these pulses are ideal for investigating ultrafast dynamic properties of matter at atomic resolution. At the recently-built soft x-ray free electron laser FLASH [1], pulse durations of 25 fs are now routinely achievable. The hard x-ray FEL's that will become available in a few years will produce photon pulses with pulse lengths of about 100 fs initially [2-3]. Plans for obtaining attosecond x-ray pulses through pulse compression schemes and slicing techniques are already under consideration [405]. The availability of such sources will have an enormous impact on time-resolved studies, x-ray imaging, and non-linear x-ray matter interaction.

With the unprecedented brightness of the FEL sources, one encounters severe requirements for the optical components. The optics have to be designed to withstand exposure to the extremely high-fluence x-ray pulses that may lead to damage of the material. It has been suggested [6-9] to use low-Z materials, such as beryllium, silicon carbide, silicon, or diamond. These materials have a relatively large penetration depth, so that the deposited energy density is small, and the risk for potential damage is reduced. It has further been suggested [9] that the deposited energy density is even smaller when *grazing-incidence* optics are used since they (i) reflect a large portion of the light and (ii) have an increased optical footprint; the decreased penetration depth of the photons under grazing incidence is offset by the mean-free path of the photo-electrons that are ejected primarily perpendicularly to the photon beam.

Whereas the detailed time response of optics is less important for longer pulses, pulses of a few femtoseconds or shorter require an understanding of the effect of the optics on the temporal structure of the FEL pulses. Previous work on the time-dependent diffraction of x-ray pulses from semi-infinite Si crystals has shown that the (111) reflection has a transient response that decays within a few femtoseconds for a photon energy of 8 keV. It was also found that this decay is slower for higher order reflections [10-11]. Such a diffractive optic is based on the volumetric (and thereby slow) response of the material and may be not suitable for pulses that are shorter than a femtosecond. For these shorter pulses, it is preferable to use optical elements that are based on the response of a single interface and a small volume, such as grazing-incidence mirrors. In this paper, we will explore the pulse response of these kinds of mirrors and determine for which pulse lengths they are adequate.

Description of model

Bragg diffraction from a crystal is best described using an atomistic model, such as dynamical diffraction theory [12]. Since we are primarily interested (i) in the materials response far away from the Bragg peaks or (ii) in non-crystalline materials, a continuum model is sufficient. We calculated the response of a material to a plane-wave packet by using linear-optics reflection theory [13]. We assume that the electric field strengths are sufficiently small so that non-linear effects are negligible. As sketched in Figure 1, we consider a semi-infinite medium occupying the half-space $z > 0$, with $z \leq 0$ being vacuum. When a linearly-polarized monochromatic plane wave with electric field $E_I(z = 0, t) = E_I(t)$ falls onto the half-space at an off-normal angle of incidence θ, it is reflected; the reflected beam has the same frequency, and the amplitude at $z = 0$ is given by

$$E_R(t) = r(\omega)E_I(t), \qquad (1)$$

with

$$r(\omega) = \frac{\cos(\theta) - n\cos(\theta')}{\cos(\theta) + n\cos(\theta')} \qquad (2)$$

for light with the electric field vector perpendicular to the plane of incidence (s-polarization), and

$$r(\omega) = \frac{\cos(\theta') - n\cos(\theta)}{\cos(\theta') + n\cos(\theta)} \qquad (3)$$

for light with the electric field vector parallel to the plane of incidence (p-polarization) [14]. $n(\omega)$ is the frequency-dependent complex index of refraction of the medium, and the complex angle θ' is given by Snell's law: $\sin(\theta) = n\sin(\theta')$ [14].

For finite wave packets $E_I(t)$, Eqn. (1) can be generalized by integrating over all frequencies, so that

$$\begin{aligned} E_R(t) &= \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \tilde{E}_R(\omega) e^{-i\omega t} d\omega & (4) \\ &= \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} r(\omega) \tilde{E}_I(\omega) e^{-i\omega t} d\omega . & (5) \end{aligned}$$

$\tilde{E}_I(\omega)$ and $\tilde{E}_R(\omega)$ are the Fourier transforms of the incoming and outgoing pulses, defined by

$$\tilde{E}(\omega) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} E(t) e^{-i\omega t} dt . \qquad (6)$$

Note that $r(-\omega) = r^*(\omega)$. An important special case is the crystal response to the delta function, $E_I(t) = \delta(t)$. In this case, $\tilde{E}_I(\omega) = 1/\sqrt{2\pi}$, and the impulse response $\tilde{E}_R(\omega)$ is given by the Green's function

$$G_R(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} r(\omega) e^{-i\omega t} d\omega . \qquad (7)$$

Eqn. (5) can then be rewritten in the form of a convolution integral,

$$E_R(t) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} G_R(\tau) E_I(t-\tau) d\tau . \qquad (8)$$

Results

We have calculated the transient response $G_R(t)$ of different low-Z materials to a delta-function electric field pulse. Values for the optical constants over the whole electromagnetic spectrum were taken from Ref. [15]. Figure 2 shows the transient intensity response $|G_R(t)|^2$ for silicon for different off-normal angles of incidence θ for p-polarized light for $t \geq 0$. Causality requires that $G_R(t) = 0$ for $t < 0$. For light under normal incidence, $|G_R(t)|^2$ quickly reaches its maximum after 0.15 fs and then decays in an oscillatory way. For larger incident angles, the maximum in $|G_R(t)|^2$ is reached earlier in time. To define an impulse response time $\tau_{0.5}$, we consider the total energy of the reflected light of a delta-function electric field pulse at time t, which is proportional to

$$I_R(t) = \int_0^t \left|G_R(\tau)\right|^2 d\tau \,. \qquad (9)$$

We define $\tau_{0.5}$ as the time at which half of the energy is reflected, that means

$$I_R(\tau_{0.5}) = \frac{1}{2} I_R(\infty) \,. \qquad (10)$$

Figures 2 (a) and (b) show $\tau_{0.5}$ as a function of grazing incident angle φ for silicon, berylium, diamond, and silicon carbide, for s- and p-polarized light. Note that $\varphi = 90^o - \theta$. Note also that these calculated properties are a function (i) of the material properties through the strengths of all excited oscillators and (ii) of the polarization of the light. These curves are universal through the electromagnetic spectrum since the driving impulse contains all frequencies.

Discussion and Conclusions

We numerically calculated the transient reflectivity of a delta-function electric pulse from a homogenous half-space of different low-Z materials. Since the frequency spectrum of a delta function is constant, these results are independent of frequency; frequency-dependent results are obtained when the pulse response is convolved with the incident electric field using Equation (8). We found that the materials continue to emit energy after the pulse has ended, as shown in Figure 2 for the case of silicon. Whereas Bragg reflections take a few femtoseconds or longer to decay [10-11], we found the impulse response time for normal surface reflections away from the Bragg peaks andfor non-crystalline materials to be shorter than 0.3 fs, see Figure 3. The impulse response time is substantially shorter (less than 0.3 as for $\varphi < 0.1^o$) for grazing incidence reflection which is in part related to the shorter penetration depth. The dependence of the impulse response time to polarization is especially pronounced for incident angles between 40^o and 60^o which is a consequence of the different angular dependence of the Fresnel coefficients.

Due to their damage resistance, low-Z materials are especially suitable for optics for XFEL's. We found the different materials to have generally similarly-shaped impulse responses, but quantitatively the impulse response times differ: For all incident angles and polarizations, we found that the impulse response time increases from diamond, silicon carbide, beryllium, to silicon, making diamond the best choice material for ultrafast sub-fs optics.

Attosecond XFEL pulses will become achievable in a few years, and the optical components have to be tailored to withstand the damage requirements without spoiling the temporal characteristics of the photon pulses. Optics based on grazing incidence reflection are preferable over Bragg-reflection-based optics since their impulse response time is substantially shorter.

Acknowledgements

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Figure Captions

Figure 1: Sketch of geometry.

Figure 2: Transient intensity response $|G_R(t)|^2$ for silicon for different off-normal angles of incidence θ for p-polarized light.

Figure 3: Impulse response time $\tau_{0.5}$ as a function of grazing angle of incidence φ for different materials for p-polarized light on a linear plot (a) and a log-log plot (b), and for s-polarized light on a linear plot (c) and a log-log plot (d).

Figures

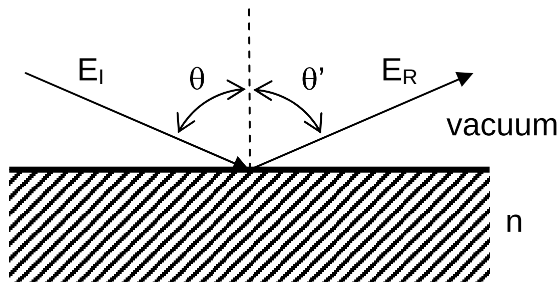

Figure 1

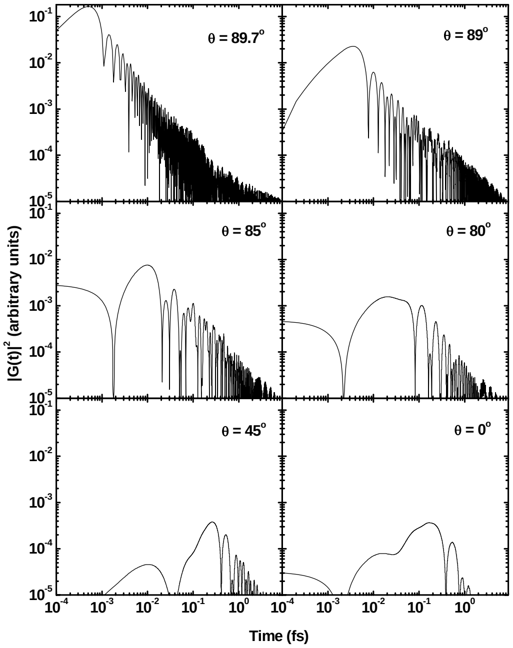

Figure 2

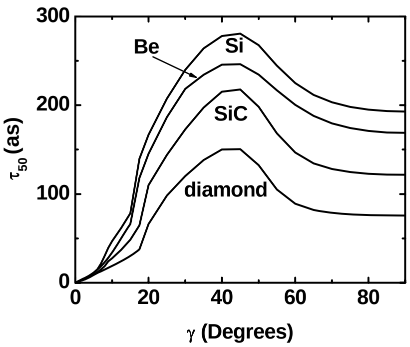

Figure 3 (a)

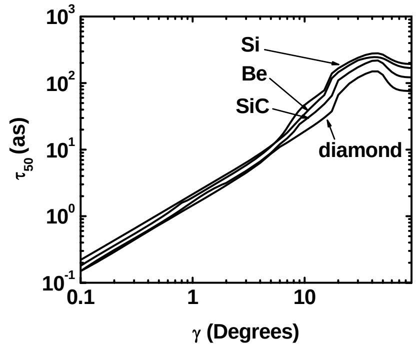

Figure 3 (b)

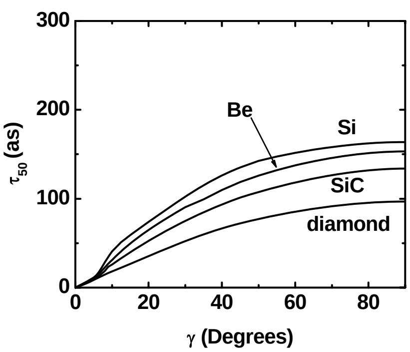

Figure 3 (c)

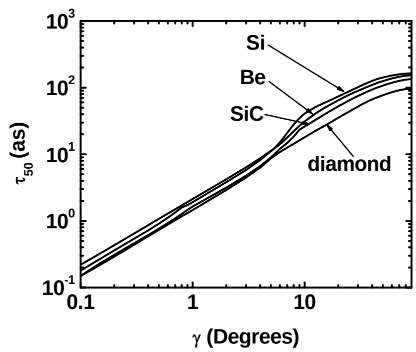

Figure 3 (d)