

OPTICAL PHYSICS

Second-harmonic generation of focused ultrashort x-ray pulses

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We study the process of x-ray second-harmonic generation (SHG) from focused ultrashort pulses. We numerically simulate and derive approximated analytical expressions for the efficiency and tolerances of the SHG process, considering the effects of temporal and spatial walk-off, and the coupling to the linear Bragg scattering. We show that for the recently observed x-ray SHG in diamond, the simultaneous Bragg diffraction of the generated second harmonic is negligible. However, spatiotemporal walk-off effects are crucial components regarding the wave propagation aspects of this process, due to the highly noncollinear phase-matching geometry. © 2015 Optical Society of America

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1. INTRODUCTION

The nonlinear response of matter to electromagnetic fields holds many scientific and technological applications. Commonly, nonlinear processes are weak and require high-intensity sources in order to be observed. Since far above resonances, nonlinearities decrease with frequency, and since highly brilliant sources in the x-ray portion of the electromagnetic spectrum have been absent until very recently, experimental observations of nonlinear phenomena, such as second-harmonic generation (SHG), have not been reported in the x-ray regime. Indeed, with earlier hard x-ray sources-x-ray tubes and third-generation synchrotrons, only spontaneous parametric downconversion (SPDC) has been observed [1-3]. Since SPDC relies on the vacuum field as one of the driving fields for nonlinear interaction, the large density of states of electromagnetic modes at x-ray wavelengths compensates for the low nonlinear coupling. However, the construction of hard x-ray free electron lasers (XFELs), producing femtosecond pulses of high brightness and peak intensity, led to the possibility of performing further experiments to explore nonlinear effects, such as x-ray and optical wave mixing [4], stimulated Raman scattering [5], and x-ray two-photon absorption [6]. X-ray nonlinear processes are different than conventional nonlinear processes in the optical regime in many aspects [3,7-9]. For example, the short wavelengths of x rays result in a local space-dependent optical response, which enables second-order nonlinear effects in centrosymmetric materials. A fundamental second-order nonlinear effect, SHG, which vanishes in homogeneous media, has a phase-matching scheme that imposes large angles between the pump and the generated second-harmonic (SH) beams. Nazarkin *et al.* theoretically studied the effect of x-ray SHG from a plane wave monochromatic pump in a perfect crystal [10]. They pointed out that the phasematching condition is only slightly different than the Bragg condition; thus, the generated SH can elastically scatter. They found the conditions for an optimal synchronous propagation of the two SH fields, which depended on both the linear and nonlinear susceptibilities. Recently, using the SPring-8 Angstrom Compact Free-Electron Laser (SACLA), Shwartz *et al.* [11] observed SHG from pump pulses at 7.3 keV with an average intensity of ~10¹⁶ W/cm², measuring a maximal conversion efficiency of 5.8×10^{-11} . That measurement agreed with expected properties of this process, such as phase-matching angles and the quadratic scaling of the SH counts with the pulse-energy.

In this paper we extend the work of Nazarkin *et al.* and analyze the process of SHG from short and focused pump beams in a perfectly ordered crystal. We solve the wave equations numerically using the undepleted-pump approximation and compare the results with the recently performed experiment [11]. By using the assumptions of strong walk-off between the pump and SH beams, and small scattering angles, we derive a closed-form solution and compare with the numerical results. We find that the elastic scattering of the SH beam is negligible in many practical experimental circumstances. We discuss the dependencies of the spatial and temporal overlap between the pump and the SH beams on the pulse duration and beam size of the pumping beam. As expected, we find that although the use of short and focused pulses, which leads to high peak intensities, is required to achieve reasonable SHG efficiency, it introduces spatial and temporal walk-off effects, which limit the efficiency enhancement. In addition, as in conventional SHG using focused pump beams, when the Rayleigh range is shorter than the propagation lengths of the pump or the SH beams in the nonlinear medium, the SHG process is dominated by diffraction, and tighter focusing leads to a reduction of the efficiency.

2. MODEL

A. Nonlinear Current Density and Envelope Equations

In this section we describe the envelope equations for the SH field and the induced nonlinear current density, which is the source for the radiation at the SH.

As pointed out by Nazarkin et al., since the indices of refraction at both the pump and SH frequencies are nearly equal, the phase-matching angles are very near the Bragg angle at the SH frequency (Fig. 1). Therefore, a fraction of the generated SH may be elastically Bragg scattered [10]. We analyze this effect by considering two SH fields: the SH that is generated from the pump, denoted as \tilde{E}_{sh} , and its Bragg scattered field, which we denote as \tilde{E}_{sc} . We write the fields as $\tilde{E}_i(\mathbf{r}, t) =$ $1/2E_i(\mathbf{r}, t) \exp(i\mathbf{k_i} \cdot \mathbf{r} - 2i\omega_0 t)\mathbf{e_i} + \text{c.c.}$, where $E_i(\mathbf{r}, t)$ is the envelope function over the $\exp(i\mathbf{k_i} \cdot \mathbf{r} - 2i\omega_0 t)$ carrier wave, and *i* denotes the generated and scattered SH modes. We use the slowly varying envelope approximation (SVEA) [12] $|\partial^2 \mathbf{E}_i / \partial z_i^2| \ll |\mathbf{k}_i \partial \mathbf{E}_i / \partial z_i|$ and $|\partial^2 \mathbf{E}_{\rm i}/\partial t^2| \ll |\omega_0 \partial \mathbf{E}_{\rm i}/\partial t|,$ where $z_i = x \sin(\theta_i) + z \cos(\theta_i)$ is the axis along the propagation direction of the generated SH or its elastically scattered mode. Using a similar separation for the induced current density for the nonlinear process $\mathbf{J}_{NL}(\mathbf{r}, t) = 1/2 \mathbf{J}_{NL}(\mathbf{r}, t)$ $\exp(i\mathbf{k}_{NL}\cdot\mathbf{r}-2i\omega_0t)\mathbf{e}_{NL}$ + c.c. and the two complementary linear Bragg processes $\mathbf{J}_{\mathbf{L},\pm}(\mathbf{r},t) = 1/2 \mathbf{J}_{L,\pm}(\mathbf{r},t) \times \exp(i\mathbf{k}_{\mathbf{L},\pm})$ $\mathbf{r} - 2i\omega_0 t$) $\mathbf{e}_{\mathbf{L},\pm}$ + c.c., we get the following envelope equations:

$$\frac{\partial \mathbf{E}_{\mathrm{sh}}}{\partial z_{\mathrm{sh}}} - \frac{i}{2k(2\omega_0)} \left(\frac{\partial^2}{\partial x_{\mathrm{sh}}^2} + \frac{\partial^2}{\partial y^2} \right) \mathbf{E}_{\mathrm{sh}} + \frac{1}{v_g(2\omega_0)} \frac{\partial \mathbf{E}_{\mathrm{sh}}}{\partial t}$$
$$= -\frac{1}{2} \eta(2\omega_0) (\mathbf{e}_{\mathrm{NL}} \cdot \mathbf{e}_{\mathrm{sh}}) \mathbf{J}_{\mathrm{NL}}(\mathbf{r}, t) \exp(i\mathbf{k}_{\mathrm{NL}} \cdot \mathbf{r} - i\mathbf{k}_{\mathrm{sh}} \cdot \mathbf{r})$$
$$-\frac{1}{2} \eta(2\omega_0) (\mathbf{e}_{\mathrm{L},+} \cdot \mathbf{e}_{\mathrm{sh}}) \mathbf{J}_{L,+}(\mathbf{r}, t) \exp(i\mathbf{k}_{\mathrm{L},+} \cdot \mathbf{r} - i\mathbf{k}_{\mathrm{sh}} \cdot \mathbf{r}), \quad (\mathbf{1})$$



Fig. 1. Phase-matching schemes. (a) SHG of the pump beam, using a specific reciprocal lattice vector **G** to phase match the nonlinear process. (b) Since the generated SH propagates in a direction near the Bragg condition of the SH frequency, it is partially elastically scattered. (c) The scattered SH is partially backscattered by the complementary Bragg process.

$$\frac{\partial \mathbf{E}_{sc}}{\partial z_{sc}} - \frac{i}{2k(2\omega_0)} \left(\frac{\partial^2}{\partial x_{sc}^2} + \frac{\partial^2}{\partial y^2} \right) \mathbf{E}_{sc} + \frac{1}{v_g(2\omega_0)} \frac{\partial \mathbf{E}_{sc}}{\partial t}$$
$$= -\frac{1}{2} \eta(2\omega_0) (\mathbf{e}_{\mathbf{L},-} \cdot \mathbf{e}_{sc}) \mathbf{J}_{\mathbf{L},-} (\mathbf{r}, t) \exp(i\mathbf{k}_{\mathbf{L},-} \cdot \mathbf{r} - i\mathbf{k}_{sc} \cdot \mathbf{r}), \quad (2)$$

where $x_i = x \cos(\theta_i) - z \sin(\theta_i)$ is the axis normal to the propagation direction of each beam. *k* and η are the wave vector and impedance inside the material, respectively, ω_0 is the pump central angular frequency, and v_{σ} is the group velocity.

In the hard x-ray regime, high above resonances, the linear interaction is described by Thomson scattering giving the susceptibility $\chi(\mathbf{r}) = -R\lambda^2 \rho(\mathbf{r})/\pi$ [13] where $\rho(\mathbf{r})$ is the charge distribution, λ is the wavelength of the driving field, and $R = q^2/4\pi\epsilon_0 m_e c^2$ is the classical radius of the electron, m_e and q being the electron mass and charge, respectively. Since the electron charge distribution $\rho(\mathbf{r})$ in ideal crystals is periodic, we may expand it in a Fourier series $\rho(\mathbf{r}) = \sum_{m} \rho_{m}$ $\exp(i\mathbf{G}_{\mathbf{m}}\cdot\mathbf{r})$, where $\rho_m = V^{-1}\int_V \rho(\mathbf{r})\exp(-i\mathbf{G}_{\mathbf{m}}\cdot\mathbf{r})d\overline{v}$ is its Fourier components, with G_m being the reciprocal vectors, and with the integration carried over the unit cell volume V. Similarly, we perform a Fourier expansion of the linear susceptibility $\chi_m = -R\lambda^2 \rho_m/\pi$. We assume that the phase-matching condition for the nonlinear process is near the Bragg condition at the SH for a specific reciprocal lattice vector G. Therefore, the related current densities describing the two complementary Bragg scattering processes are $\tilde{\mathbf{J}}_{\mathbf{L},+}(\mathbf{r},t) = -2i\omega_0\varepsilon_0\chi_G\tilde{\mathbf{E}}_{\mathbf{sc}}(\mathbf{r},t)$ $\exp(i\mathbf{G}\cdot\mathbf{r})$ and $\tilde{\mathbf{J}}_{\mathbf{L}_{\mathbf{r}}}(\mathbf{r},t) = -2i\omega_0\varepsilon_0\chi_{-G}\tilde{\mathbf{E}}_{\mathbf{sh}}(\mathbf{r},t)\exp(-i\mathbf{G}\cdot\mathbf{r}).$

To describe the interaction for the induced nonlinear current, we consider a classical model of cold collisionless plasma fluid [14]. Assuming a pump beam $1/2E_p(\mathbf{r}, t) \exp(i\mathbf{k_p} \cdot \mathbf{r} - i\omega_0 t)\mathbf{e_p} + \text{c.c}$ with central angular frequency ω_0 , which is much higher than any bound electronic state in the material, we use a perturbative approach and find the second-order nonlinear current oscillating at the SH frequency [10]:

$$\tilde{\mathbf{J}}_{\mathbf{NL}}(\mathbf{r},t) = -\frac{q^2 \rho_G \mathbf{E}_p^2(\mathbf{r},t)}{8m_e^2 \omega_0^3} [\mathbf{k}_{\mathbf{p}} + 2(\mathbf{G} \cdot \mathbf{e}_{\mathbf{p}})\mathbf{e}_{\mathbf{p}}] \\ \times \exp[i(2\mathbf{k}_{\mathbf{p}} + \mathbf{G}) \cdot \mathbf{r} - 2i\omega_0 t] + \text{c.c.}$$
(3)

Furthermore, we may use Eq. (3) to write the second-order nonlinear susceptibility $\chi^{(2)}(2\omega_0;\omega_0,\omega_0) = -iq^2\rho_G(4\varepsilon_0m_e^2\omega_0^4)^{-1}\times$ $[\mathbf{k}_{\mathbf{p}} + 2(\mathbf{G} \cdot \mathbf{e}_{\mathbf{p}})\mathbf{e}_{\mathbf{p}}] \cdot \mathbf{e}_{\mathbf{sh}}$ [10]. We note that in a uniform plasma (where only the ρ_0 component of the charge density is nonzero), the nonlinear current oscillates in the propagation direction of the pump beam; thus, since only the transverse component of the current density contributes to the electromagnetic radiation at the far field, SHG does not occur in uniform plasmas. For this reason, a medium with periodic charge distribution (a perfect crystal) is used as the nonlinear medium. In addition, it is also important to note that we consider the regime in which, on the one hand, the pump frequency is much higher than any resonance frequency in the system, so all of the electrons in an atom react the same and constitute the cold plasma. On the other hand, the photon energy is much less than the electron rest energy, so relativistic dynamics is neglected.

Here we expand the model suggested by Nazarkin *et al.* for pump pulses that are finite in time and space, introducing walkoff and diffraction effects. We use the SVEA, and assume an undepleted pump, which is valid because of the low efficiency of this process. Using Eqs. (1)–(3), we get the equations governing the electric field envelopes of the two SH modes:

$$\sin(\theta_{\rm sh})\frac{\partial E_{\rm sh}}{\partial x} + \cos(\theta_{\rm sh})\frac{\partial E_{\rm sh}}{\partial z} - \frac{i}{2k(2\omega_0)} \left[\cos^2(\theta_{\rm sh})\frac{\partial^2}{\partial x^2} + \sin^2(\theta_{\rm sh})\frac{\partial^2}{\partial z^2} - \sin(2\theta_{\rm sh})\frac{\partial^2}{\partial z\partial x} + \frac{\partial^2}{\partial y^2}\right] E_{\rm sh} + \frac{1}{v_g(2\omega_0)}\frac{\partial E_{\rm sh}}{\partial t}$$
$$= \frac{q^2\rho_G\eta(2\omega_0)E_{\rm p}^2(\mathbf{r},t)}{2k^2} \{\mathbf{k}_{\rm sh} + 2(\mathbf{G},\mathbf{r}_{\rm sh})\mathbf{e}_{\rm sh}\} \exp(i\Delta\mathbf{k}_{\rm sh},\mathbf{r})$$

$$=\frac{1+O(\mathbf{r}_{c}^{*})}{8m_{e}^{2}\omega_{0}^{3}}\{[\mathbf{k}_{\mathbf{p}}+2(\mathbf{G}\cdot\mathbf{e}_{\mathbf{p}})\mathbf{e}_{\mathbf{p}}]\cdot\mathbf{e}_{\mathbf{sh}}\}\exp(i\Delta\mathbf{k}_{\mathbf{NL}}\cdot\mathbf{r})$$

$$+i\varepsilon_0\eta(2\omega_0)\omega_0\chi_G E_{sc}(\mathbf{e_{sc}}\cdot\mathbf{e_{sh}})\exp(i\Delta\mathbf{k_B}\cdot\mathbf{r}),$$
(4)

$$\sin(\theta_{sc})\frac{\partial E_{sc}}{\partial x} + \cos(\theta_{sc})\frac{\partial E_{sc}}{\partial z} - \frac{i}{2k(2\omega_0)}\left[\cos^2(\theta_{sc})\frac{\partial^2}{\partial x^2} + \sin^2(\theta_{sc})\frac{\partial^2}{\partial z^2} - \sin(2\theta_{sc})\frac{\partial^2}{\partial z\partial x} + \frac{\partial^2}{\partial y^2}\right]E_{sc} + \frac{1}{v_g(2\omega_0)}\frac{\partial E_{sc}}{\partial t}$$

$$= i\varepsilon_0 \eta(2\omega_0)\omega_0 \chi_{-G} E_{\rm sh}(\mathbf{e_{sh}} \cdot \mathbf{e_{sc}}) \exp(-i\Delta \mathbf{k_B} \cdot \mathbf{r}).$$
(5)

In Eqs. (4) and (5) we neglect the absorption of the SH wave since for the case in which all pertinent frequencies are well above resonances, the absorption coefficient of the pump is much larger than the absorption coefficient at the SH. $\Delta k_{NL} = 2k_p + G - k_{sh}$ and $\Delta k_B = k_{sc} + G - k_{sh}$ are the phase mismatch vectors for the nonlinear and Bragg scattering processes, respectively. We note that the structure of Eqs. (4) and (5) is similar to the structure obtained by using the Takagi–Taupin formalism for deformed crystals [13]. Here, however, we assume that the medium is an ideal crystal, but the inhomogeneity is introduced by the pump beam since it is not a homogeneous monochromatic plane wave.

B. Modeling the XFEL Pump Pulse

Pulses emerging from self-amplified spontaneous emission (SASE)-based XFELs contain a large number of randomly distributed spikes. Unfortunately, due to the stochastic nature of the pulses and since complete measurements of the pulse structure are still out of reach, there is no deterministic model that describes the temporal structure of the pulses. It is well known that for nonlinear interactions, in general, the details of the pulse structures can be very important.

Here we consider three different models for the longitudinal pulse structure $f(z_p, t)$: transform-limited (TL) pulses, linearly chirped (LC) pulses, and temporally stochastic (TS) pulses. We compare between the models by keeping the same Gaussian power spectrum with full width at half-maximum (FWHM) bandwidth ΔE . To model the pulse structure of the pump, we first write the general envelope of a Gaussian beam [15]:

$$E_{p}(x_{p}, y, z_{p}, t) = \frac{E_{0} \exp(-\alpha_{p} z_{p})}{\sqrt{[1 - i\tau_{x}(z_{p})][1 - i\tau_{y}(z_{p})]}} \\ \times \exp\left\{-\frac{x_{p}^{2}}{w_{0,x}^{2}[1 - i\tau_{x}(z_{p})]} - \frac{y^{2}}{w_{0,y}^{2}[1 - i\tau_{y}(z_{p})]}\right\} \\ \times f(z_{p}, t).$$
(6)

Here $\tau_x(z_p) = 2(z_p - z_{0,x})/b_x$ and $\tau_y(z_p) = 2(z_p - z_{0,y})/b_y$. z_p is the propagation axis of the pump pulse, and x_p is the orthogonal axis parallel to the scattering plane. α_p is the field absorption coefficient. $w_{0,x}$ and $w_{0,y}$ determine the Gaussian pulse size, $z_{0,x}$ and $z_{0,y}$ set the focal points, and $b_x = k(\omega_0)w_{0,x}^2$, $b_y = k(\omega_0)w_{0,y}^2$ are the confocal parameters inside the material. We use a general complex amplitude of a LC Gaussian pulse to describe the LC pulses [15]:

$$f(z_{p},t) = \exp\left\{-\frac{(1+i\xi)}{\tau^{2}}\left[t - \frac{z_{p}}{v_{g}(\omega_{0})}\right]^{2}\right\},$$
 (7)

where τ determines the Gaussian pulse duration and ξ defines the linear chirp. We estimate the pulse duration τ by taking the linear chirp coefficient as $\xi = \sqrt{[\Delta E^2 \tau^2/(8\hbar^2 \ln 2)] - 1}$ (\hbar is the reduced Planck constant). We also use Eq. (7) to model the TL Gaussian pulse, by setting $\xi = 0$, having the minimal pulse duration $\tau_0 = 2\sqrt{2 \ln 2\hbar}/\Delta E$.

We describe the TS pulses, constructed by a closely packed and equally spaced train of coherent TL Gaussian spikes with random phases, as follows:

$$f(z_p, t) = \exp\left\{-\frac{1}{\tau^2} \left[t - \frac{z_p}{v_g(\omega_0)}\right]^2\right\} \sum_{m=-\infty}^{\infty} \exp(i\xi_m)$$
$$\times \exp\left\{-\frac{1}{\tau_0^2} \left[t - m\Delta\tau - \frac{z_p}{v_g(\omega_0)}\right]^2\right\}.$$
(8)

 $\Delta \tau$ is the duration between the spikes (taken to be $\Delta \tau = \tau_0/5$). For each pulse we generate a set $\{\xi_m\}$ of uniformly distributed [0, 2π] random numbers. Similar to Ref. [16], the average over a large number of these stochastic pulses corresponds to an assumed average Gaussian pulse structure of duration τ and FWHM bandwidth ΔE (see Fig. 2).

3. APPROXIMATED ANALYTICAL EXPRESSIONS

To describe the efficiency dependence upon the different parameters, analytical calculations of the overall SHG efficiency and tolerances are shown in the following section. As we will



Fig. 2. Input pump pulse. (a) and (b) show the intensity and power spectrum, respectively, of a single TS pulse realization, assuming an average FWHM bandwidth of 1 eV and an average FWHM duration of 20 fs. The spectrum is shown as a function of photon energy relative to the central frequency. (c) and (d) show the averaged intensity and power spectrum, respectively, over 1000 TS pulse realizations (black solid lines), plotted along the corresponding Gaussian functions (blue circles).

show below, in many practical cases it is possible to find a simple closed-form solution for the discussed problem.

Wang and Weiner [15] developed a theoretical framework for calculating the efficiency of Type I SHG by tightly focused and ultrashort pulses with simultaneous spatial and temporal walk-offs, generalizing previous work by Boyd and Kleinman [17]. We use their approach to describe the propagation aspects of SHG at x-ray wavelengths in the regime where the Bragg scattering of the generated SH is negligible. In essence, the problem is to solve the SVEA equation for the SH field where the pump and generated SH beams propagate in different directions, which are imposed by the phase-matching scheme. This is similar to noncollinear Type I SHG in uniaxial crystals, where a walk-off effect is introduced by the different directions of the Poynting and wave vectors related to the extraordinary polarization. Therefore, it is expected that the description of the propagation aspects of x-ray SHG will have the same form as the propagation of noncollinear SHG in uniaxial crystals.

We apply the results of Ref. [15], and assume a small walkoff angle ρ and no absorption of the pump $\alpha_p = 0$. Within these assumptions, we use Eq. (19) in Ref. [15], which shows that under the paraxial approximation, the overall SHG efficiency (which we define as the number of output SH photons per incident pump photon) from a LC Gaussian pump pulse may be evaluated by calculating the double integral:

$$\eta_{\rm sh} = \frac{\gamma U_p}{2\sqrt{\pi}w_{0,x}w_{0,y}\tau} \int_0^{\frac{L}{\cos(\theta_p)}} \int_0^{\frac{L}{\cos(\theta_p)}} \frac{\exp[i\Delta k(z_1 - z_2)]}{\sqrt{[1 - i\tau_x(z_1)][1 - i\tau_y(z_1)]}} \\ \times \frac{\exp\left[-\frac{(z_1 - z_2)^2}{L_{i-t}^2}\right] dz_1 dz_2}{\sqrt{[1 + i\tau_x(z_2)][1 + i\tau_y(z_2)]}},$$
(9)

where $\gamma = \omega_0^2 |\chi^{(2)}(2\omega_0; \omega_0, \omega_0)|^2 / (2\pi n_p^2 n_{\rm sh} \varepsilon_0 c^3)$, U_p is the pump pulse total energy, and *L* is the crystal length, assuming a symmetrical Laue propagation geometry (transmission geometry where the reciprocal lattice vector is parallel to the surface of the slab). Equation (9) is similar to Eq. (19) in Ref. [15], with the only differences being the assumed propagation geometry (hence the different integration limit), the presentation of the LC Gaussian pulse in Eqs. (6) and (7), and the definition of the conversion efficiency (which here is in terms of the number of photons). We choose to take the direction of the carrier wave vector of the SH field $\mathbf{k_{sh}}$ in which the SHG phase mismatch $\Delta k = |\Delta \mathbf{k_{NL}}|$ is normal to the boundary surface. The characteristic length within the pump and generated SH overlap, the spatiotemporal walk-off length, is defined as

$$L_{s-t} = \left[\frac{\tan^2(\rho)}{w_{0,x}^2} + \frac{\xi^2 + 1}{\tau^2} \,\mathrm{GVM^2}\right]^{-1/2}.$$
 (10)

In Eq. (10) GVM = $[1/(v_g(2\omega_0) \cos(\rho))] - [1/v_g(\omega_0)]$ is the group velocity mismatch. This expression is similar to Eq. (15) in Ref. [15]. L_{s-t} represents an effective walk-off length that considers both the spatial and temporal walk-off effects, causing the limited interaction length between the pump and generated SH waves. The value $(\xi^2 + 1)/\tau^2$ remains a constant for a given bandwidth ΔE . Therefore, observing Eq. (10), one notes that the interaction length L_{s-t} is limited by the coherence time of the pulse, which is inversely proportional to ΔE . In the case of weak focusing, where the confocal parameters are much

larger than the crystal length, and a long crystal, which contains many interaction lengths, we assume b_x , $b_y \gg L \gg L_{s-t}$ and $L \gg L_{s-t}^2 \Delta k$. Under these assumptions, Eq. (9) is reduced to a simplified expression:

$$\eta_{\rm sh} = \frac{\gamma U_p L_{s-t} L}{2w_{0,x} w_{0,y} \tau \cos(\theta_p)} \exp\left[-\left(\frac{\Delta k L_{s-t}}{2}\right)^2\right].$$
(11)

Similar to noncollinear phase-matching schemes in the optical regime, Eq. (11) shows that the efficiency grows linearly with the propagation length, as opposed to the quadratic dependence when walk-off effects are negligible [12]. From Eq. (11), we also note that when the pump waist size in the direction parallel to the scattering plane $w_{0,x}$ decreases, the efficiency approaches a constant value with respect to $w_{0,x}$. A similar argument may be used to describe the efficiency dependence on the pulse duration τ . We note that for further focusing, where b_{x} , $b_y \gg L$ is not satisfied, the efficiency decreases due to diffraction.

Next, we generalize Eq. (11) to account for the effect of linear absorption of the pump pulse throughout its propagation inside the nonlinear medium. Since the efficiency grows linearly with the pump-pulse energy and with the crystal length, we modify Eq. (11) by including the exponential decay of the pump field along its propagation path. Therefore, after an additional integration Eq. (11) becomes

$$\eta_{\rm sh} = \frac{\gamma U_p L_{s-t}}{4\alpha_p w_{0,x} w_{0,y} \tau} \left[1 - \exp\left(-\frac{2\alpha_p L}{\cos(\theta_p)}\right) \right] \exp\left[-\left(\frac{\Delta k L_{s-t}}{2}\right)^2\right].$$
(12)

We now discuss the rocking curve (the acceptance angle bandwidth) of the SHG process. As we elaborated in the previous paragraphs, the phase-matching scheme in this process is similar to the implementation of quasi-phase-matching (QPM) by periodically poled devices. In their study of tuning and tolerances of QPM, Fejer *et al.* [18] derived the tolerance of the SHG efficiency to any parameter the phase mismatch Δk depends on. From Eq. (11), we observe that the efficiency is reduced to half of its perfectly phased-matched value when $\exp[-(\Delta k L_{s-t}/2)^2] = 1/2$ is satisfied. Therefore, the mismatch Δk for which the SHG process will be half as efficient as the phase-matched case is

$$\Delta k = \frac{2\sqrt{\ln 2}}{L_{s-t}}.$$
 (13)

Following [18], we expand the mismatch Δk dependence on the pump angle θ_p to the first order $\Delta k(\theta_p) = (\theta_p - \theta_{p,0})$ $\partial \Delta k / \partial \theta_p |_{\theta_p = \theta_{p,0}}$, where $\theta_{p,0}$ is the angle of the pump beam at perfect phase-matching. Using Eq. (13), the FWHM of the rocking curve is therefore $\Delta \theta_{p,\text{FWHM}} = 4\sqrt{\ln 2L_{s-t}^{-1}}|\partial\Delta k/$ $\partial \theta_p|_{\theta_p=\theta_{p,0}}^{-1}$. Observing the geometry of the phase-matching scheme in Fig. 1(a), we find that $\partial \Delta k / \partial \theta_p |_{\theta_p=\theta_{p,0}} = 2k_p$ $\tan(\theta_{p,0} + \theta_{\text{sh},0})$, where $\theta_{\text{sh},0}$ is the SH angle at perfect phase matching. Since the dispersion in x-ray frequencies is relatively weak, we may assume that the perfect phase-matching angles of both the pump and SH beams are near the Bragg angle for the SH frequency θ_B . Therefore, we find that the FWHM of the rocking curve is

$$\Delta \theta_{p,\text{FWHM}} = \frac{2\sqrt{\ln 2}}{k_p L_{s-t} \tan(2\theta_B)}.$$
 (14)

The spectral bandwidth of the SHG process is derived in a similar manner. We expand the dependence of the mismatch Δk on the pump angular frequency ω_0 to the first order $\Delta k(\omega_0) = (\omega_0 - \omega_c)\partial\Delta k/\partial\omega_0|_{\omega_0=\omega_c}$, where ω_c is the central pump angular frequency at perfect phase matching. Assuming weak dispersion $\partial n(\omega_0)/\partial\omega_0 = 0$, we obtain $\partial\Delta k/\partial\omega_0|_{\omega_0=\omega_c} = (2/c)\{n(\omega_c) - [n(2\omega_c)/\cos(\theta_{p,0} + \theta_{sh,0})]\}$. Taking both perfect phase-matching angles $\theta_{p,0}$, $\theta_{sh,0}$ as the Bragg angle for the SH frequency θ_B , and, in addition, taking the indices of refraction at both the pump and SH frequencies as unity, we obtain the FWHM spectral bandwidth:

$$\Delta \omega_{\rm FWHM} = \frac{\sqrt{\ln 2 \cos(2\theta_B)c}}{L_{s-t} \sin^2(\theta_B)}.$$
 (15)

We note that the dispersion has very little influence on the spectral bandwidth, which is determined mainly by the noncollinear nature of the phase-matching scheme.

4. NUMERICAL SIMULATIONS

In this section we describe the numerical solutions of the SVEA equations, and compare them with the experimental observations and the approximated analytical expressions presented in the previous section. We numerically solve Eqs. (4) and (5) assuming the parameters of the recently performed experiment [11], and using the fast-Fourier-transform method with respect to variables x and t and an integration with respect to z. We assume that the wavelength is 1.7 Å (7.3 keV), the polarization is parallel to the scattering plane (π -polarized), the pulse total energy is 4.3 µJ, the FWHM bandwidth is 1 eV, and the symmetric Gaussian FWHM waist is 1.5 µm. The nonlinear medium is a 0.5 mm long diamond crystal, with the pump focused at half of its thickness. We choose to work in a symmetrical Laue geometry and phase match the nonlinear process with the (2 2 0) reciprocal vector. Figure 3 depicts the differences between the predictions of the pump models described in Section 2.B.

In Figs. 3(a) and 3(b) we plot the efficiency of the process as a function of the propagation distance. In Fig. 3(c) we plot the rocking curves. Comparing these calculations to the experimental results [11], we find that the two broadened structures (LC and TS), with an assumed 20 fs FWHM duration, are adequately in agreement with the measured values. From Fig. 3(a) we note that the SHG efficiency dependence on the propagation length, which is close to linear with a saturating effect due to the absorption of the pump, as expected by Eq. (12), has a similar form for the three pump models. The different scaling of these curves originates from the different pulse structures; the TL pulses have the highest peak intensity, and therefore the highest efficiency, and the two broadened pulse models lead to a similar efficiency, since they have a similar average pulse structure. Inserting these parameters into Eq. (14), we find that the rocking curve width is $\Delta \theta_{p,\text{FWHM}} =$ 49.5 μ rad, which is in a good agreement with Fig. 3(c). We note that the measured rocking curve in Ref. [11] (180 µrad FWHM) is broader than the rocking curve we calculated here.



Fig. 3. Numerical simulations of the SHG process for different pump models. (a) and (b) show the efficiencies of the generated SH and its elastically scattered mode, respectively, as a function of propagation distance. Using the parameters described in Section 4 for perfect phase matching, we consider three pump structures: a transform-limited (TL, black solid lines), linearly chirped (LC, red dashed lines), and temporally stochastic (TS, blue dotted line, averaged over 200 realizations) Gaussian pulses. The average FWHM duration is assumed to be 20 fs. The circles in (a) represent the efficiency estimations for the TS pulses were performed assuming negligible Bragg scattering. (c) The SHG efficiency dependence on the deviation of the pump beam from the phase-matched angle $\Delta\theta$. Using normalized units, the two rocking curves are nearly identical.

Possible origins for this broadening, which were not taken into account in the current work, could be the presence of spatiotemporal coupling in the short spikes emerging from the XFEL and deviations of the transverse structure of the pump field induced by the optical elements along the beam path. An additional cause might be imperfections of the nonlinear diamond crystal. Since both the TL and LC pump structures of the same bandwidth result in an equal walk-off length L_{s-t} , their rocking curves are similar, as is depicted in Fig. 3(c). In addition, Eq. (15) predicts a FWHM spectral bandwidth of 1.53×10^{15} Hz (1 eV) for the above parameters, which is confirmed with an additional simulation.

We note that the Bragg scattering process of the SHgenerated mode is negligible. This is because the effective interaction length between the two modes and the coherence length arising from the nonzero phase mismatch are much shorter than the extinction length of the elastic scattering process. From Eqs. (4) and (5) one notices that, for short propagation distances, where the second term on the right-hand side of Eq. (4) is much smaller than the first term, an optimal coupling between the pump beam and the scattered SH mode occurs when $\Delta \mathbf{k}_{NL} = \Delta \mathbf{k}_B$. However, since the refractive indices at the fundamental and the SH are different due to dispersion, this synchronization between the phase mismatch of the nonlinear and linear processes is impractical.

Next, we describe the dependencies of the efficiency on the beam size and pulse duration. As mentioned, due to the weak nonlinear susceptibility, it is necessary to focus the pump beam in order to achieve higher SH signal for a given pump-pulse energy. However, since the focused pump and the generated SH signal propagate in different directions where $\rho =$ $\theta_p + \theta_{sh}$ is the angle between them, they have a finite distance within which they overlap, causing a spatial walk-off. Furthermore, the group velocity mismatch (GVM) results in a finite distance within which short pulses overlap, which causes temporal walk-off after a finite time. We note that focusing in the direction normal to the scattering plane does not introduce walk-off effects. The higher peak intensity leads to enhancement of the SHG efficiency, which is inversely proportional to $w_{0,v}$, as long as the crystal length is shorter than the confocal parameter. However, reducing the beam size in the scattering plane results in a shorter interaction length, which leads to a reduction of the efficiency enhancement by the higher peak



Fig. 4. (a) Efficiency dependence on the focusing in the scattering plane (solid line). Focusing in a direction parallel to the scattering plane reduces the effective interaction length and therefore moderates the enhancement of the SHG efficiency owing to the higher peak intensities. Focusing further reduces the overall efficiency due to diffraction. (b) Efficiency dependence on the pulse duration (solid line). The numerical simulations were performed assuming the parameters described in Section 4 for a 50 μ m crystal, a TL pulse, and varying beam waists and durations of down to 35 nm and 85 as, respectively. The approximated efficiency estimations using Eq. (12) are plotted along both figures (dashed lines).



Fig. 5. Comparison between numerical simulations (solid line) and the approximated analytical expression (dashed line) for the efficiency as a function of the photon energy of the pump. The parameters are described in Section 4. The pump pulses are transform limited and the crystal length is L = 0.5 mm. The results of the analytical expression approach the numerical results for high photon energies, where the walk-off angle is small.

intensity. This effect is depicted in Fig. 4(a), which shows the dependence of the efficiency on the beam size of the pump in the scattering plane, using the parameters described at the beginning of the section. Figure 4(b) shows the efficiency dependence on the pulse duration, assuming a TL pump pulse. Both numerically calculated curves (solid lines) in Fig. 4 are plotted along the estimated efficiency dependence using the approximated analytical expression in Eq. (12) (dashed lines). As mentioned in the previous section, in the regime where the crystal length is much smaller than the confocal parameter, the SHG efficiency approaches a constant value with respect to both $w_{0,x}$ and τ as they decrease. This behavior is shown in Fig. 4(b), where as opposed to diffraction, dispersion is still negligible for the discussed parameters.

Finally, in Fig. 5 we compare the numerical calculations of the dependence of the efficiency on the photon energy of the pump (solid line) with calculations obtained by using Eq. (12) (dashed lines). As expected, the higher the pump photon energy (resulting in a smaller walk-off angle), the better the approximation holds. However, even with lower and more relevant photon energies, this simple analytical expression gives a decent estimation for the expected efficiency.

5. CONCLUSIONS AND OUTLOOK

To conclude, we analyzed the process of SHG at x-ray wavelengths from short and focused pulses. Modeling the input pump pulse using both deterministic and random spiky pulse structures, we simulated a recently performed experiment observing SHG using a pump pulse from an XFEL and showed good agreement with the experimental results. In addition, we studied several general aspects regarding the efficiency optimization and tolerances of the x-ray SHG process. Since the nature of the phase-matching geometry causes the pump and the generated SH to propagate in different directions, the finite interaction distance between them limits the efficiency enhancement when focusing in the direction parallel to the scattering plane. Therefore, walk-off considerations support small angles between the pump and the SH beams. However, the nature of the plasma-like nonlinearity we discussed in this paper results in a vanishing SH field as the angle between the pump and the SH approaches to zero. Hence, the optimization between the nonlinearity and the walk-off should be considered for any SHG experiment. This consideration does not affect the efficiency enhancement by focusing the pump beam in the direction normal to the scattering plane, which is efficient until the confocal parameter becomes comparable to the crystal length. In addition, in contrast to nonlinear optics in the optical regime, in the x-ray regime the nonlinear materials are not transparent and, thus, radiation damage could be critical. Recent studies on radiation damage with XFELs suggest that the important quantity for damage is the pulse energy per area [19]. The implication is that the improvement of the SHG efficiency by focusing the beam is rather limited. Since the size of the beam is limited by the radiation damage, it would be useful to enhance the peak intensity of the pump beam by optimizing its structure in the temporal domain. The most efficient approach to achieve this requirement is to use TL pulses. Our work suggests that it is possible to achieve much higher efficiencies than the efficiency observed in the SACLA experiment by using current and future XFELs. For example, x-ray pulses generated by using self-seeding techniques with intensities that are more than 100 times larger than the maximum intensity reported in the SACLA experiment are now available at the Linac Coherent Light Source (LCLS). With this source and with further optimization of the SHG process, as discussed in this paper, the expected efficiency would be about 10^{-7} . Finally, under the assumptions of negligible elastic Bragg scattering of the generated SH and small scattering angles, we showed simple analytical expressions for the total efficiency, rocking curve width, and spectral bandwidth of the SHG process. These simple expressions can be used to give estimations for many practical cases of experiments observing SHG in the x-ray regime.

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