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OPTICAL PHYSICS

Theory of noncollinear frequency doubling of transform limited pulses in non-steady-state regime

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Noncollinear second-harmonic generation from two ultrashort pulses intersecting in a nonlinear medium is studied in spectral and time domains. We derive analytical expressions for the second-harmonic (SH) amplitude in crystals of finite thickness neglecting diffraction. The contribution from characteristics of the fundamental radiation and interaction geometry to the process is analyzed. In addition, refined phase-matching conditions are obtained. We find that the spectral bandwidth is determined by the intersection angle and can be enlarged. The SH pulse duration can be optimized by varying the fundamental beam size and the intersection angle. It is found that the SH beam excited by a Gaussian fundamental beam becomes elliptical. It is shown that the fundamental pulse duration can be readily characterized with single pulses by means of measuring the ellipticity of the SH beam profile. The approach developed can potentially be used to calculate parametric interactions of fundamental pulses with an arbitrary spectrum.

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1. INTRODUCTION

The second-harmonic generation (SHG) is a well-known nonlinear optical process where two photons at frequency ω are converted into a single photon at double frequency 2ω. The most efficient SHG takes place when the fundamental frequency (FF) and second-harmonic (SH) waves are phase matched in a quadratic nonlinear medium. This may be achieved by using birefringent nonlinear media [1] or periodically poled nonlinear crystals [2,3]. When interacting waves propagate in the same direction (the so-called collinear interaction) in a homogeneous medium, the phase-matching angle is determined by the fundamental wavelength. Unlike the above, the noncollinear interaction is of interest because the phase-matching angle is a function of both the FF wavelength and the intersection angle between two FF beams. This enables tunable spontaneous downconversion and optical parametrical amplification [4]. The noncollinear interaction is preferable for auto- and cross-correlation measurements of femtosecond pulses because of the background free auto- and cross-correlation traces [5–11]. Recently disordered nonlinear media were successfully used for these purposes [12–15]. Despite the numerous studies on noncollinear SHG [5,12,16], the theory lacks consistency. In [16], a noncollinear SHG has been studied in the special case of tilted fundamental pulses to compensate the mismatch of group velocities between the fundamental pulse and SH one. The scheme was considered when fundamental pulse fronts are orthogonal to the SH propagation direction in nonlinear crystal.

In the present work, we systematically study noncollinear SHG from ultrashort pulses in another statement of the problem; namely, the phase and pulse (amplitude) fronts of the fundamental radiations coincide, and therefore, amplitude fronts of the fundamental pulse and the SH one are inclined to each other. As a result, the SH beam profile differs from the fundamental beam one. We elaborate the theory of non-steady-state noncollinear SHG from ultrashort pulses; that is, we take into account the group velocity mismatch (GVM), in the traditional approximation of undepleted fundamental pulses for the case of not spreading pulses and not diffracting beams. Analytical expressions were derived for the SH frequency and spatial spectra and the spatial distribution of intensity.

2. THEORETICAL MODEL AND BASIC FORMULAS

We consider propagation of two intersecting fundamental beams through a homogeneous nonlinear medium in the plane XY as shown in Fig. 1. Each of the two FF beams propagates at angle ϕ to the y axis. Let us suppose for simplicity that the intersection angle between the FF beams 2ϕ is large enough to provide for their overlapping inside the medium of thickness L. In this representation, SH is generated along the bisector of the angle between the two FF beams (i.e., in the positive direction of the axis y). Each of the FF beams propagates along its respective axis y_j in the reference coordinate system (x_j, y_j), and its transverse coordinates are x_j $(j = 1, 2)$.

Transformation from the reference coordinate system (x_j, y_j) to the coordinates (x, y) is given by formulas

$$\begin{cases} x_j = x \cos\phi \mp y \sin\phi \\ y_j = \pm x \sin\phi + y \cos\phi \end{cases}.$$

Then, in the undepleted fundamental pulse approximation, the SHG process will be governed by the equation

$$\left(\frac{\partial}{\partial y} + \frac{1}{u_2}\frac{\partial}{\partial t} - \frac{1}{2k_2}\Delta(x, z)\right)A(t, x, z, y) = i\beta g(x, y)A_{11}(t, x, z, y)A_{12}(t, x, z, y)\exp(i\Delta k y), \quad (1)$$

where A is the SH amplitude, $\beta = 2\pi k_2\chi^{(2)}/n_2^2$, $g(x, y)$ is the function characterizing modulation of the nonlinear susceptibility $\chi^{(2)}$ $(g(x, y) = \text{const})$, n_2 is the refractive index at the 2ω frequency, $\Delta k = 2k_1 \cos\phi - k_2$ is the wave-vector mismatch, u_2 is the SH pulse group velocity, and $\Delta(x, z) = (\partial^2/\partial x^2 + \partial^2/\partial z^2)$ is the transverse Laplacian.

Propagation of FF beams A_{11} and A_{12} in the coordinate system (x, y) obeys the equation

$$\left(\frac{\partial}{\partial y} \pm \tan\phi\frac{\partial}{\partial x} + \frac{1}{u_1 \cos\phi}\frac{\partial}{\partial t}\right)A_{1j}(t, x, z, y) = 0, \quad (2)$$

where ϕ is the inner angle between the propagation direction of the fundamental beam and the y axis, u_1 is the FF pulse group velocity, and the sign "+" refers to $j = 1$ and "−" to $j = 2$. It should be noted that at oblique incidence of the fundamental pulse on a nonlinear crystal, there is the transverse group delay, caused by mismatch of phase and amplitude fronts, in the crystal. The angle between them is equal to $\psi = \arctan([u_1/v_1 - 1]\sin\phi)$ [11], where v_1 is the FF phase velocity. In Eq. (2), this effect is not taken into account as for the below presented calculations $\psi = -0.007$ that is $|\psi| \ll 1$.

By solving the Cauchy problem for the equation under consideration we obtain

$$A_{1j}(t, x, z, y) = A_{1j}\left(x\cos\phi \mp y\sin\phi, t - \frac{y\cos\phi \pm x\sin\phi}{u_1}\right), \quad (3)$$

where the condition at the input of the nonlinear crystal $(y = 0)$ has been taken into account $A_{1j}(t, x, z) = A_{1j}(x \cos\phi, t \mp \frac{x \sin\phi}{u_1})$. It follows from Eqs. (1) and (3) that in the case under consideration, the so-called GVM of the interacting FF and SH pulses along the y axis is equal to $\nu = \cos\phi/u_1 - 1/u_2$, and the condition of group velocity matching takes the form $u_2 \cos\phi = u_1$. This condition differs from the corresponding one [16] obtained for other SHG geometry.

Further, as in the case of FF beams, diffraction of the SH beam can be neglected. Using the Fourier transform

$$A(\Omega, K_x, K_z, y) = \frac{1}{(2\pi)^{3/2}}\iiint A(t, x, z, y) \times \exp(-i(\Omega t - K_x x - K_z z))\mathrm{d}t\mathrm{d}x\mathrm{d}z, \quad (4)$$

Eq. (1) can be expressed in the form

$$\frac{\partial}{\partial y}A(\Omega, K_x, K_z, y) = \frac{i\beta g}{(2\pi)^{3/2}}e^{i\Delta k y}\iiint A_{11}(t, x, z, y)A_{12}(t, x, z, y) \times \exp(-i\Omega t + iK_x x + iK_z z)\mathrm{d}t\mathrm{d}x\mathrm{d}z. \quad (5)$$

The frequency domain representation of fundamental pulses [Eq. (3)] allows us to write down Eq. (5) as follows:

$$\begin{aligned}\frac{\partial}{\partial y}A(\Omega, K_x, K_z, y) = {} & \frac{i\beta g}{(2\pi)^{3/2}}e^{i\left(\Delta k + \left(\frac{\cos 2\phi}{u_1 \cos\phi} - \frac{1}{u_2}\right)\Omega\right)y} \\ & \times \iiint \mathrm{d}\Omega_1 \mathrm{d}K_{x1}\mathrm{d}K_{z1}\tilde{A}_{11}(\Omega_1, K_{x1}, K_{z1}) \\ & \times \tilde{A}_{12}\left(\Omega - \Omega_1, \frac{K_x}{\cos\phi} - K_{x1} + \frac{\tan\phi}{u_1}\Omega, K_z - K_{z1}\right) \\ & \times \exp\left(-i\left(\frac{K_x}{\cos\phi} - 2K_{x1}\right)\sin\phi y\right). \quad (6)\end{aligned}$$

The analysis at derivation Eq. (6) showed the existence of the following relations between spatial and spectral components:

$$\begin{aligned}\Omega &= \Omega_1 + \Omega_1', \\ K_x &= (K_{x1} + K_{x1}')\cos\phi - \Omega\sin\phi/u_1, \\ K_z &= K_{z1} + K_{z1}', \quad (7)\end{aligned}$$

where Ω_1, Ω_1' are the FF spectral components, and K_{x1}, K_{x1}', K_{z1}, K_{z1}' are the spatial components of FF radiation. The relations in Eq. (7) represent the phase-matching conditions for frequency and transverse wave vector components. One can see from Eq. (6) that the SH spectrum will depend on the

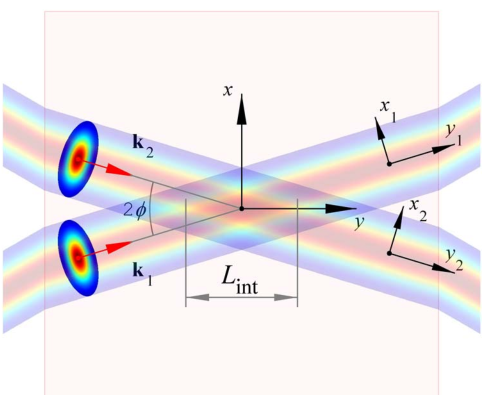

Fig. 1. Interaction of two intersecting fundamental beams in a homogeneous nonlinear medium.

transverse FF spectrum components distribution, whereas the transverse SH wave vector distribution will depend on the FF spectrum and transverse FF wave vector distribution. Eq. (6) can be used for FF pulses of an arbitrary spectral shape. On the contrary, the use of Eq. (5) requires the knowledge of spatiotemporal characteristics of the fundamental radiation.

Next we assume for definiteness that the two incident fundamental pulses are spatially identical. It is reasonable to consider interaction of spectrally limited Gaussian pulses with a Gaussian intensity distribution in transverse direction $A_{1j}(t, \mathbf{r}) = A_{1j}(0)\exp(-t^2/\tau_0^2)\exp(-\mathbf{r}^2/a^2)$; here index $j = 1, 2$ refers to the respective fundamental beam, $2\tau_0$ is the pulse duration, a is the beam radius, and $r^2 = x^2 + z^2$. Integration of Eq. (5) along the propagation coordinate over $[-L/2, L/2]$ yields the SH amplitude:

$$A(\Omega, K_x, K_z) = \frac{(-1)^{3/4}\sqrt{i}\sqrt{\pi}\beta g p \tau_0 a^3}{16\sqrt{2}\sin\phi}A_{11}(0)A_{12}(0) \times \exp\left(-\frac{\Omega^2\tau_0^2}{8}\right)\exp\left(-\frac{a^2p^2K_x^2}{8}-\frac{a^2K_z^2}{8}-\frac{a^2\Delta\tilde{k}^2}{8\sin^2\phi}\right) \times 2i\,\mathrm{Im}\,\mathrm{erfi}\left[\frac{a\sqrt{2}(\Delta\tilde{k}+2iL\sin^2\phi/a^2)}{4\sin\phi}\right], \quad (8)$$

where $\Delta\tilde{k} = \Delta k - \nu\Omega$ and

$$p = 1/(\cos^2\phi + [a/u_1\tau_0]^2\sin^2\phi)^{1/2}. \quad (9)$$

The spectral intensity of SH can be expressed as follows:

$$S(\Omega) = \frac{\pi c n_2 p \beta^2\tau_0^2 a^4}{256\sin^2\phi}g^2 I_1^2\exp\left(-\frac{a^2\Delta k^2}{4\sin^2\phi}\right) \times \exp\left(-\frac{(\tau_0^2 + a^2\nu^2/\sin^2\phi)}{4}\Omega^2 + \frac{\Delta k \nu a^2}{2\sin^2\phi}\Omega\right) \times \mathrm{Im}\left(\mathrm{erfi}\left[\frac{a\sqrt{2}(\Delta\tilde{k}+2iL\sin^2\phi/a^2)}{4\sin\phi}\right]\right)^2. \quad (10)$$

Since interaction between FF and SH waves takes place over the region where the fundamental beams overlap, we can introduce an effective interaction length. According to Fig. 1, the effective interaction length is $L_{\mathrm{int}} = 2a/\sin\phi$. Consider the case of an infinite nonlinear medium (i.e., when the medium thickness satisfies the condition $L \gg L_{\mathrm{int}}$). In this case, integration of Eq. (5) from $-\infty$ to ∞ results in the following expression:

$$A(\Omega, K_x, K_z) = \frac{i\sqrt{\pi}\beta g p \tau_0 a^3}{8\sqrt{2}\sin\phi}A_{11}(0)A_{12}(0) \times \exp\left(-\frac{\mu^2}{8}\Omega^2 + \frac{\Delta k \nu a^2}{4\sin^2\phi}\Omega\right)\exp\left(-\frac{a^2\Delta k^2}{8\sin^2\phi}\right) \times \exp\left(-\frac{a^2p^2K_x^2}{8}\right)\exp\left(-\frac{a^2K_z^2}{8}\right). \quad (11)$$

Here $\mu^2 = \tau_0^2 + a^2\nu^2/\sin^2\phi = \tau_0^2 + L_{\mathrm{int}}^2\nu^2/4$.

Fourier transform of Eq. (11) yields

$$A(t, x, z) = \frac{i\sqrt{\pi}\beta g\tau_0 a}{\sqrt{2}\mu\sin\phi}A_{11}(0)A_{12}(0)\exp\left(-\frac{a^2\Delta k^2}{8\sin^2\phi}\right) \times \exp\left(-\frac{2x^2}{a^2p^2}-\frac{2z^2}{a^2}\right)\exp\left(\frac{2}{\mu^2}\left(it-\frac{\Delta k\nu a^2}{4\sin^2\phi}\right)^2\right). \quad (12)$$

Equations (11) and (12) prove to satisfy the Parseval theorem.

By integrating a module of square of Eq. (11) over the spatial frequencies, we obtain spectral intensity of the SH:

$$S(\Omega) = \frac{\pi c n_2 p\beta^2\tau_0^2 a^4}{256\sin^2\phi}g^2 I_1^2\exp\left(-\frac{a^2\Delta k^2}{4\sin^2\phi}\right) \times \exp\left(-\frac{\mu^2}{4}\Omega^2 + \frac{\Delta k\nu a^2}{2\sin^2\phi}\Omega\right). \quad (13)$$

The spatial distribution of the SH intensity defined as $I(t, x, z) = (cn_2/8\pi)|A(t, x, z)|^2$ has the form

$$I(t, x, z) = \frac{cn_2\beta^2 g^2\tau_0^2 a^2 I_1^2}{16\mu^2\sin^2\phi}\exp\left(-\frac{4x^2}{a^2p^2}-\frac{4z^2}{a^2}\right) \times \exp\left(-\frac{a^2\Delta k^2}{4\sin^2\varphi}\right)\exp\left(-\frac{4}{\mu^2}\left[t^2-\left(\frac{\Delta k\nu a^2}{4\sin^2\phi}\right)^2\right]\right). \quad (14)$$

The SH pulse envelope represents a Gaussian function, and the parameter μ determines the SH pulse duration. The SH pulse duration can be represented as $\mu = \tau_0\sqrt{(1 + L_{\mathrm{int}}^2/(4L_{\mathrm{gr}}^2))}$, and, therefore, it is a function of both L_{int} and $L_{\mathrm{gr}} = \tau_0/\nu$, the latter being the length of the GVM. Conversely, the interaction length depends on the beam spot size and the intersection angle. By varying these parameters we can change the SH pulse duration.

3. RESULTS AND DISCUSSION

For the calculations, we choose a beta barium borate (BBO) crystal as a nonlinear medium, which is commonly used for SHG of a Ti:Sapphire oscillator. The fundamental spectrum has a Gaussian shape with the full width at half-maximum (FWHM) 10 nm at the central wavelength 800 nm. It is also preferable to consider the SHG process of the I-type ($oo-e$ interaction). Under noncollinear interaction, the calculated phase-matching angle equals 41.2 deg for the inner intersection angle 20 deg. The refractive indexes of BBO were approximated using the Sellmeier coefficients from Ref. [17].

The calculated SH spectra are shown in Fig. 2. The length of the medium is 1 mm. For the calculations, Eqs. (10) and (13) were used. As seen, Eq. (13) provides a good description of the noncollinear SHG when the actual thickness of the medium is larger than the effective interaction length $L > L_{\mathrm{int}}$ [Fig. 2(a)]. A different situation arises with $L \le L_{\mathrm{int}}$ [Fig. 2(b)]. In this case, Eq. (13) gives a spectrum profile with an underestimated width. The condition of use of Eq. (13) ($L > L_{\mathrm{int}}$) can be represented in the form $\phi' \ge \arcsin(2a/L)$.

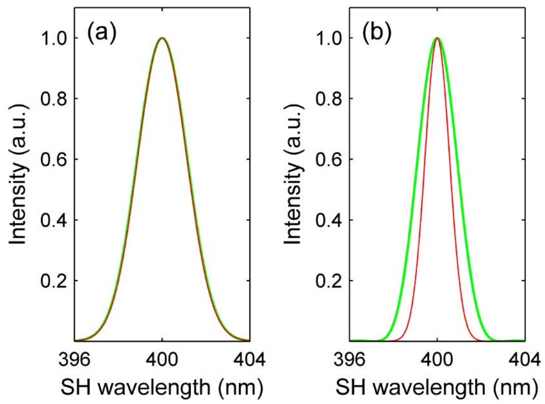

Fig. 2. Noncollinear SH spectral intensity calculated using Eq. (10) (green) and Eq. (13) (red) for intersection angles (a) 5 deg and (b) 2 deg. The respective effective interaction lengths are 0.74 mm and 1.9 mm.

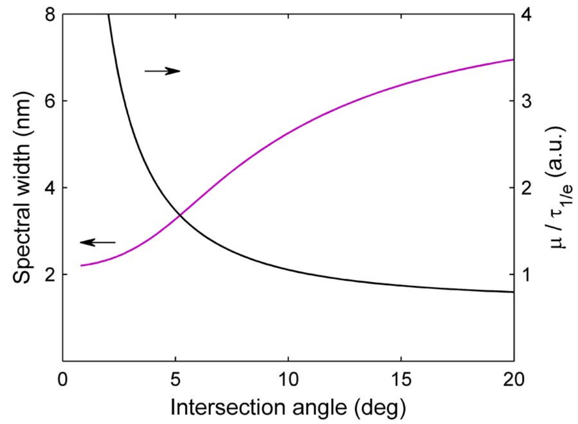

Fig. 4. Spectral width (left axis) and relative pulse broadening (right axis) versus intersection angle.

From Eq. (13), the SH spectral bandwidth (FWHM) is

$$\Delta\Omega(\phi) = \frac{\Delta k \nu a^2}{\mu^2 \sin^2 \phi} + \frac{2}{\mu}\sqrt{\ln(2)}. \tag{15}$$

Note that only the second term in Eq. (15) contributes to the spectral width if the phase-matching condition is fulfilled. The presence of the phase mismatch ($\Delta k \neq 0$) leads to modification of the spectral envelope. In particular, there appears a spectral shift if phase mismatch is introduced:

$$\delta\Omega = -\frac{\Delta k \nu a^2}{\mu^2 \sin^2 \phi}. \tag{16}$$

Figure 3 shows the calculated spectral intensities for collinear and noncollinear interaction in a 2-mm-thick medium. In the case of collinear SHG, the spectral intensity derived from [18] has a sinc^2-shaped profile. The curves corresponding to the noncollinear SHG are found from Eq. (10). As one can see, increasing the intersection angle results in widening of the spectral curve. This result can be accounted for by the short interaction length within the fundamental beam intersection, which implies a larger spectral range where the phase-matching condition is fulfilled.

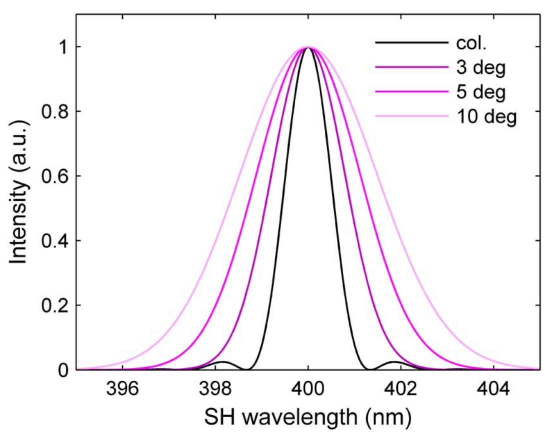

Fig. 3. Calculated spectral intensity for collinear (black) and noncollinear SHG (colored).

Figure 4 illustrates in more detail the behavior of the spectral width depending on the intersection angle. The spectral width was calculated using Eq. (15) and then scaled to the wavelength. In the extreme case, when the intersection angle goes to zero, the spectral width comes close to the spectral width of collinear SHG. This angular behavior is also accompanied by a remarkable reduction in the SH pulse duration.

Taking into account that $\mu^2 = \tau_0^2(1 + L_{\text{int}}^2/(4L_{\text{gr}}^2))$ in Eq. (14), the broadening of SH pulses can obviously be attributed to the GVM and depends on the relation between L_{int} and L_{gr}. For example, SH pulses are $\sqrt{2}$ times wider than FF pulses for $L_{\text{int}} = 2L_{\text{gr}}$. The choice of the interaction length must rely on GVM for a given material.

The approach proposed can be used to simulate intensity autocorrelation measurements by introducing time delay between the fundamental pulses. On the other hand, from Eq. (14) one can see that the SH beam cross section becomes elliptical with the ellipticity factor p. It was shown previously that the ellipticity factor is determined by the pulse duration of incident beams, their cross-section size, and the intersection angle. Hence, we can find the pulse duration with single pulses by means of measuring the SH beam profile by a spatial detector. The extracted fundamental pulse duration is given by

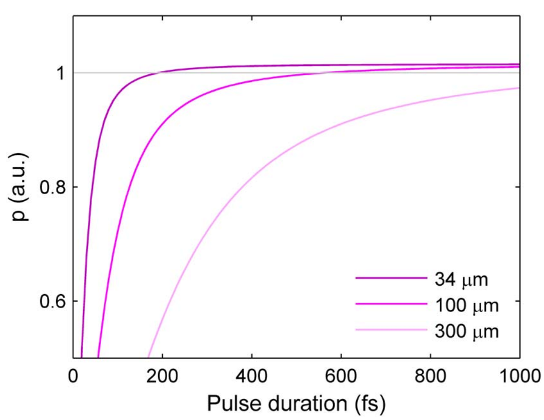

Fig. 5. Dependence of the parameter p on the FF pulse duration.

$$\tau_{\mathrm{ext}} = \frac{2ap \sin \phi}{u_1 \sqrt{1 - p^2 \cos^2 \phi}}. \quad (17)$$

The use of Eq. (17) requires exact knowledge of the size of fundamental beams (the ellipticity factor p) and their intersection angle. Figure 5 illustrates the behavior of the parameter p depending on the pulse duration for three different beam radii a. These curves are identical, except for the scaling factor, which is determined by the relation between the terms in the denominator of Eq. (9). If $a = u_1\tau_0$, there is no SH beam ellipticity, and the parameter $p = 1$. However, if $a \ll u_1\tau_0$, the curve in Fig. 5 is saturated. Our analysis shows that this technique can be applied for monitoring sub-100-fs pulses. In particular, in the situation under study ($p = 0.92$, $a = 34\ \mu\mathrm{m}$, $2\phi = 20\ \mathrm{deg}$), the pulse width is equal to 85 fs (FWHM).

The SH pulse energy at the exit from the medium is

$$E = \frac{\sqrt{\pi}\pi c n_2 p \beta^2 g^2 \tau_0^2 a^4 I_1^2}{128\mu \sin^2 \varphi} \exp\left(-\frac{a^2 \Delta k^2}{4 \sin^2 \varphi}\right) \times \exp\left(\frac{(\Delta k \nu a^2)^2}{\mu^2 (2 \sin^2 \phi)^2}\right). \quad (18)$$

The SH pulse energy goes down with the growing intersection angle. For instance, there is a tenfold energy drop when the intersection angle changes from 1 to 3 deg. This results from the GVM and fractional overlapping of the fundamental pulses inside the medium. To compensate for the GVM, dispersion prisms can be used for tailoring fundamental pulses prior to entering the crystal [16,19].

4. CONCLUSION

We have developed a consistent theory of noncollinear SHG by ultrashort laser pulses in homogeneous nonlinear media in spectral and time domains. We have derived analytical expressions for the SH amplitude in crystals of finite thickness, neglecting diffraction phenomena and fundamental pulse depletion. A contribution from the characteristics of the fundamental radiation and interaction geometry to the process has been analyzed. In addition, refined phase-matching conditions are obtained. We have found that the spectral bandwidth is determined by the intersection angle and can be enlarged. The SH pulse duration can be optimized by varying the fundamental beam size and the intersection angle. It is found that the SH beam at noncollinear interaction of ultrashort pulses becomes an elliptical one. It has been shown that the fundamental pulse duration can be readily characterized with single pulses by means of measuring the ellipticity of the SH beam profile. The approach developed can potentially be used to calculate parametric interactions of fundamental pulses with an arbitrary spectrum in homogeneous media as well as in one- and two-dimensional nonlinear photonic crystals [20–22].

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