Theory of non-collinear second harmonic generation in a non-steady state regime

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We consider non-collinear second harmonic generation from two ultrashort pulses intersecting in a nonlinear medium in spectral and time domains. We derive analytical expressions for the second harmonic amplitude in crystals of finite thickness and obtain a refined phase-matching condition. The contribution from characteristics of the fundamental radiation and interaction geometry to the process is analyzed. 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 shown that the fundamental pulse duration can be readily characterized with single pulses by means of measuring the second harmonic beam profile. The approach developed can potentially be used to calculate parametric interactions in one- and two-dimensional nonlinear photonic crystals.

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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 non-collinear 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 down conversion and optical parametrical amplification [4]. The non-collinear interaction is preferable for auto- and crosscorrelation measurements of femtosecond pulses because of the background free auto- and crosscorrelation traces [5–10]. Recently disordered nonlinear media were successfully used for these purposes [11–14]. Despite the numerous studies on non-collinear SHG [5, 11, 15], the theory though lacks consistency.

In the present work, we systematically study non-collinear SHG from two ultrashort pulses intersecting in a nonlinear medium. We elaborate the theoretical model for crystals of finite thickness and derive an analytical expression describing the process.

2. THEORETICAL MODEL

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)

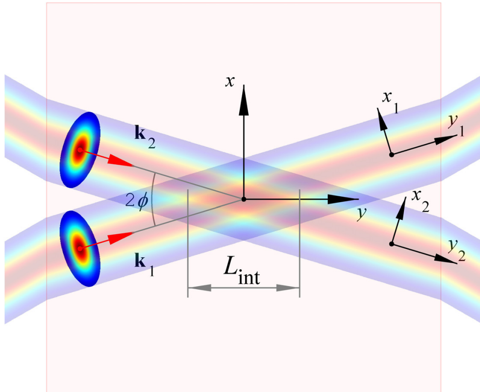

Fig. 1. Interaction of two intersecting fundamental beams in a homogeneous nonlinear medium.

to the coordinates (x,y) is given by formulas

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

Then in the negligible depletion 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 $\chi^{(2)}$ ($g(x,y)$ = 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 $\Delta(x,z) = \left(\partial^2/\partial x^2 + \partial^2/\partial z^2\right)$ is the transverse Laplacian.

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

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

here 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, the sign "+" refers to $j=1$ and "−" to $j=2$.

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 $A_{1j}(t,x,z) = A_{1j}\left(x\cos\phi, t\mp\frac{x\sin\phi}{u_1}\right)$.

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

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

Eq. (1) can be expressed in the form

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

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

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

where $\nu = \cos\phi/u_1 - 1/u_2$ is the group velocity mismatch (GVM). 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 spatio-temporal characteristics of the fundamental radiation.

Our analysis shows the following relations between spatial and spectral components

$$\Omega=\Omega_1+\Omega_2, \nu\Omega=(K_{x1}-K_{x2})\sin\phi \\ K_x=(K_{x1}+K_{x2})\cos\phi-\Omega\sin\phi/\ u_1 \\ K_z=K_{z1}+K_{z2}. \quad (7)$$

These relations represent phase-matching conditions for noncollinear SHG in a non-steady state regime.

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 second harmonic 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

$$p^2 = u_1^2\tau_0^2/\left(a^2 sin^2\phi + u_1^2\tau_0^2\cos^2\phi\right). \quad (9)$$

The spectral intensity of SH can be expressed as follows

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

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 $L_{int} = 2a/\sin\phi$. Consider the case of an infinite nonlinear medium, i.e. when the medium thickness satisfies the condition $L \gg L_{int}$. In this case, integration of Eq. (5) from $-\infty$ to ∞ results in the following expression

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

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

Fourier transform of Eq. (11) yields

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

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

By integrating Eq. (11) over the spatial frequencies, we obtain spectral intensity of the second harmonic

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

The spatial distribution of the second harmonic intensity defined as $I(t, x, z) = (c n_2/8\pi)|A(t, x, z)|^2$ has the form

$$I(t, x, z) = \frac{c n_2 \beta^2 g^2 \tau_0^2 a^2 I_1^2}{16\mu^2\sin^2\phi} \exp\left(-\frac{4x^2}{a^2p^2} - \frac{4z^2}{a^2}\right) \times \exp\left(-\frac{a^2\Delta k^2}{4\sin^2\varphi}\right) \exp\left(-\frac{4}{\mu^2}\left[t^2 - \left(\frac{\Delta k \nu a^2}{4\sin^2\phi}\right)^2\right]\right) \quad \textbf{(14)}$$

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 $\mu = \tau_0\sqrt{(1 + L_{int}^2/(4L_{gr}^2))}$, and, therefore, it is a function of both L_{int} and $L_{gr} = \tau_0/\nu$, the latter being the length of the group velocity mismatch. 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 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 non-collinear 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. [16].

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

From Eq. (13) , the SH spectral bandwidth (full width at half maximum) is

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

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

$$\delta\Omega = -\frac{\Delta k \nu a^2}{\mu^2\sin^2\phi} \quad \textbf{(16)}$$

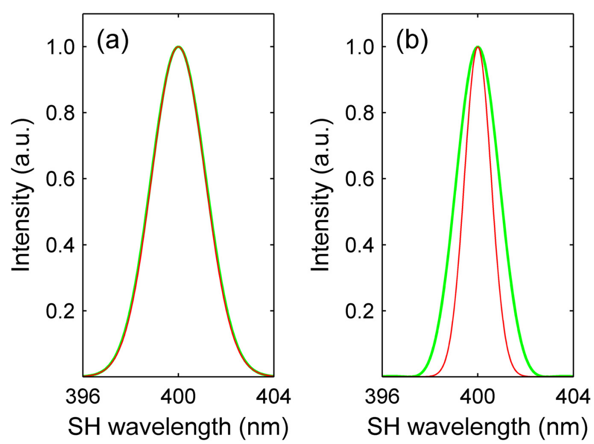

Fig. 2. Non-collinear SH spectral intensity calculated using Eq. (10) (green) and Eq. (13) (red) for intersection angles 5 (a) and 2 deg (b). The respective effective interaction lengths are 0.74 and 1.9 mm.

Fig. 3 shows the calculated spectral intensities for collinear and non-collinear interaction in a 2-mm-thick medium. In the case of collinear SHG, the spectral intensity derived from [17] has a sinc^2-shaped profile. The curves corresponding to the non-collinear 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. 4 illustrates in more detail the behaviour 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 behaviour is also accompanied by a remarkable reduction in the SH pulse duration.

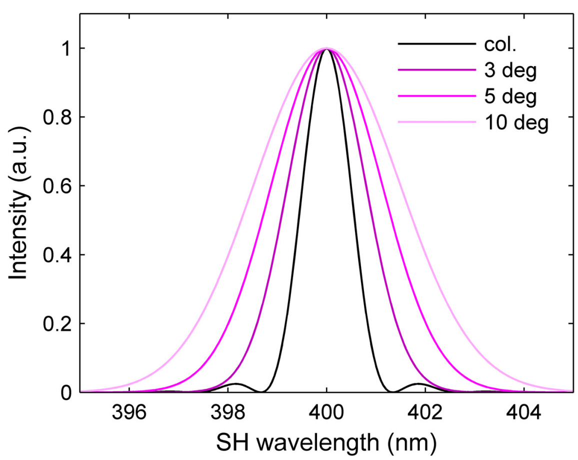

Fig. 3. Calculated spectral intensity for collinear (black) and non-collinear SHG (colored).

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

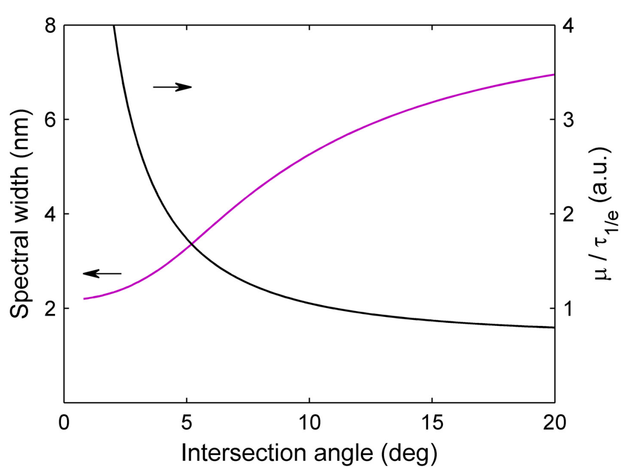

Fig. 4. Spectral width (left axis) and relative pulse broadening (right axis) versus intersection angle.

The approach proposed can be used for 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 second harmonic beam profile by a spatial detector. The extracted fundamental pulse duration is given by

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

The use of Eq. (17) requires exact knowledge of the size of fundamental beams (the ellipticity factor p) and their intersection angle. Fig. 5 illustrates the behaviour 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 the 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 ($p = 0.92$, $a = 34\ \mu m$, $2\phi = 20$ deg), the pulse width is equal to 85 fs (FWHM).

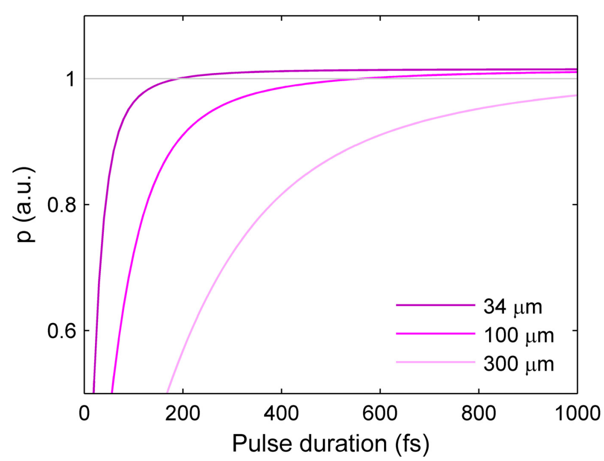

Fig. 5. Dependence of the parameter p on the FF pulse duration.

The second harmonic pulse energy at the exit from the medium is

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

As seen, the SH pulse energy significantly goes down with the growing intersection angle. For instance, there is a ten-fold energy drop, when the intersection angle changes from 1 to 3 deg. This results from the group velocity mismatch and fractional overlapping of the fundamental pulses inside the medium. To compensate for the latter factor, dispersion prisms can be used for tailoring fundamental pulses prior to entering the crystal [15, 18].

4. CONCLUSION

We have developed a consistent theory of non-collinear second harmonic generation by ultrashort laser pulses in homogeneous nonlinear media in spectral and time domains. We have derived analytical expressions for the second harmonic amplitude in crystals of finite thickness and obtained a refined phase-matching condition. A contribution from the characteristics of the fundamental radiation and interaction geometry to the process has been analyzed. 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 has been shown that the fundamental pulse duration can be readily characterized with single pulses by means of measuring the second harmonic beam profile by a spatial detector. The approach proposed can be used to calculate parametric interactions in one- and two-dimensional nonlinear photonic crystals [19, 20].

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