Simulation Example

Second harmonic generation for ultrashort pulses in phase-matching conditions

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Released 08.12.2015

Abstract
Second harmonic generation is an important effect in nonlinear optics. It is well discussed for cw processes, and in this project we want to show its properties for ultrashort pulses by using the simulation software VirtualLab Fusion. [1]

Keywords: Nonlinear Optics; Second harmonic generation; Ultrashort pulses

Physics

Second harmonic generation appears during the interaction of electromagnetic fields with a nonlinear material. Inside the material the incoming field creates local dipoles, which themselves generate new fields. In general the group velocity of a pulse in z direction can be described as

$$v_{\text{group}} = v_{\text{phase}} + k\frac{\partial v_{\text{phase}}}{\partial k} \tag{1}$$

$$v_{\text{phase}} = \frac{c}{n(\omega)}, k = \frac{2\pi}{\lambda} \tag{2}$$

It is obvious that the incoming and the generated fields will move with the same group velocity when the corresponding refractive indices for all fields are the same. In this case we expect constructive interference. In every other case, the fields that are continuously generated as the incoming field moves through the media will interact and destructive interference will happen at certain lengths, depending on the material properties. The incoming field is called fundamental field and the overlay of all generated ones the second harmonic. An visualization of this procedure can be seen in Fig. 1.

From Maxwell equations for a nonmagnetic medium with no free charges, we can easily derive

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r},\omega) = \frac{i\omega}{c^2}\mathbf{E}(\mathbf{r},\omega) + \omega^2\mu\mathbf{p}(\mathbf{r},\omega) \tag{3}$$

with

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r},\omega) = \nabla(\nabla\mathbf{E}(\mathbf{r},\omega)) - \nabla^2\mathbf{E}(\mathbf{r},\omega) = \nabla^2\mathbf{E}(\mathbf{r},\omega) \tag{4}$$

without free charges, this results in

$$-\nabla^2\mathbf{E}(\mathbf{r},\omega) = \frac{\omega^2}{c^2}\mathbf{E} + \omega^2\mu_0(\mathbf{P}^L(\mathbf{r},\omega) + \mathbf{P}^{NL}(\mathbf{r},\omega) \tag{5}$$

where $c^2 = \frac{1}{\epsilon_0\mu_0}$ is the vacuum speed of light and E is the field strength in z direction. We now want to model second harmonic generation for a fundamental pulse with a central frequency of ω_1 and an second

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harmonic with a central Frequency of ω_3 in a nonlinear medium. Therefore, the nonlinear polarization can be written as [2]:

$$\mathbf{P}_{NL} = \epsilon_0 \int d_{eff} \mathbf{E}_1(\omega') \mathbf{E}_2(\omega - \omega') d\omega' \tag{6}$$

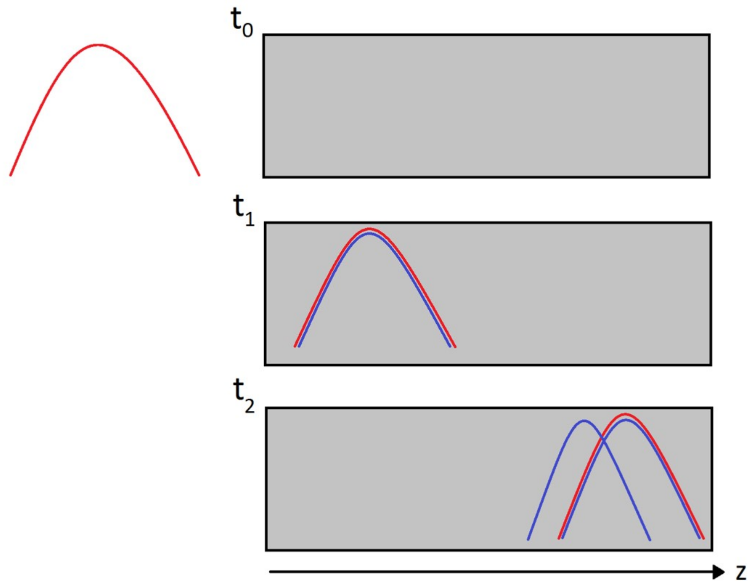

Figure 1: Principle of second harmonic generation in a nonlinear material. The fundamental pulse (red) generates a second harmonic (blue), which travels with an different group velocity. At time t_2 a new second harmonic is generated by the fundamental pulse, which interferes with the previous second harmonic. This happens at each infinitesimal time step.

Using the slowly varying Amplitudes and a plane pulse approach we can assume

$$\mathbf{E}_F(\mathbf{r}, \omega_1) = \hat{E}_F(z, \omega_1) e^{ik(\omega_1)z} \tag{7}$$

$$\mathbf{E}_{SH}(\mathbf{r}, \omega_1) = \hat{E}_{SH}(z, \omega_3) e^{ik(\omega_3)z} \tag{8}$$

.

Furthermore, for a plan pulse traveling along the z axis we can write

$$\nabla^2 \mathbf{E}(\mathbf{r}, \omega) = \left(\frac{\partial^2 E(z,\omega)}{\partial z^2} + 2ik \frac{\partial E(z,\omega)}{\partial z} - k^2 E(z,\omega)\right) e^{ikz} \tag{9}$$

Assuming $\frac{\partial^2 E}{\partial z^2} << k \frac{\partial E}{\partial z}$ we insert (9),(8),(7) and (6) in (5) and can write down the equations for the second harmonic generation process as

$$\partial_z \hat{E}_F(\omega_1, z) = -\alpha_1 \int d\omega_2 \hat{E}_F^*(\omega_2, z) \hat{E}_{SH}(\omega_2 + \omega_1, z) e^{+i\Delta k(\omega_1, \omega_2 + \omega_1)z} \tag{10}$$

$$\partial_z \hat{E}_{SH}(\omega_3, z) = -\alpha_3 \int d\omega_1 \hat{E}_F(\omega_1, z) \hat{E}_F(\omega_3 - \omega_1, z) e^{+i\Delta k(\omega_1, \omega_3 - \omega_1)z} \tag{11}$$

with

$$\hat{E}_i(\omega_i, z) = E_i(\omega_i, z) e^{ik_i L} \tag{12}$$

$$\Delta k(\omega_1, \omega_2) = (\omega_1 + \omega_2) n(\omega_1 + \omega_2) - \omega_2 \cdot n(\omega_2) - \omega_1 \cdot n(\omega_1) \tag{13}$$

$$\alpha_i = \frac{\omega_i d_{eff}}{c_0 n_i} \tag{14}$$

which is taking care of the linear dispersion.

In general we do not have an analytical solution, but if we assume that the second harmonic is rather small in comparison to the fundamental pulse, we can assume that the fundamental field will not be depleted. This results in $\Delta Q_{1/2} = 0$, so we have only one equation left to solve [3].

$$E_{SH}(\omega_2) = -\alpha_2 \int d\omega_1 E_F(\omega_1) E_F(\omega_3 - \omega_1) [\frac{e^{+i\Delta k(\omega_1, \omega_3 - \omega_1)L} - 1}{\Delta k}] \quad (15)$$

with

$$\Delta k(\omega_1, \omega_2) = (\omega_1 + \omega_2) n(\omega_1 + \omega_2) - \omega_2 \cdot n(\omega_2) - \omega_1 \cdot n(\omega_1)$$

(16)

This approximation is only for small crystal lengths good. For longer crystal the depletion of the fundamental pulse must be considered. To do that, we can combine this approach with a split-step-method. In this method we divide the material in small slices and calculate the corresponding second harmonic. With this information we can determine the depletion of the fundamental field which can then be used to calculate the Second Harmonic of the next slice, as shown in Fig. 2.

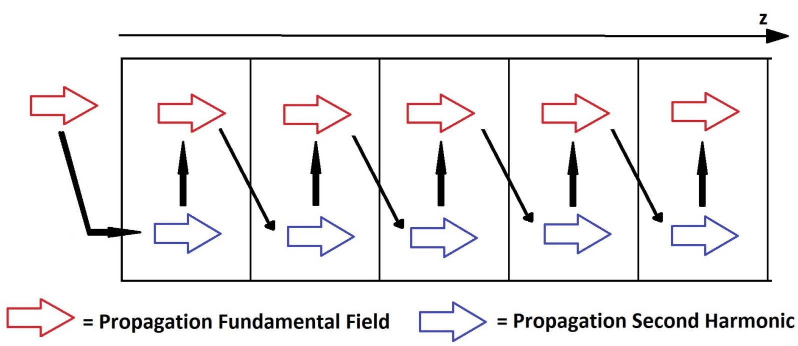

Figure 2: Combination of the small-size-approximation with a split-step-method. The original field is used to determine the second harmonic of the first slice, which is used to calculate the depletion of the original field and therefore the shape of the fundamental field at the end of the first slice. With this we can simulate the second harmonic of the second slice and the cycle continues.

Simulation Results

In our simulations we use on BBO crystal as nonlinear material. You can find detailed optical parameter of BBO in the work from Nikogosy [4].

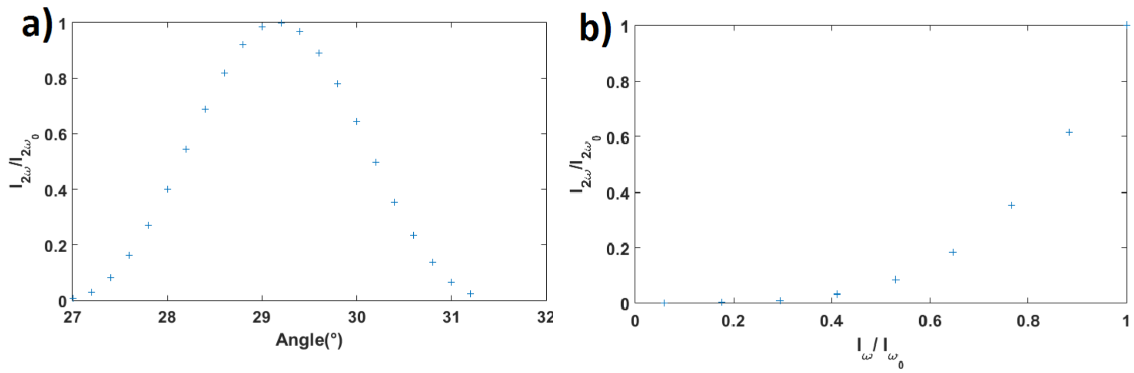

Figure 3: a) Angle tuning to find the area where phase-matching conditions are fulfilled. b) Dependence of the intensity of the second harmonic on the intensity of the fundamental pulse.

In our simulations we want to test the dependence of the peak intensity of the second harmonic from the crystal length, the angle between crystal and optical axis and the intensity of fundamental pulse. From the approximated equation for second harmonic generation (15) you can easily see, that metting the phase-matching conditions $\Delta k = 0$ means a maximal peak intensity of the second harmonic and that Δk is highly depended on the refractive Index of the crystal. It is also quite natural to assume (and derived more detailed in the work from Robert W. Boyd in [5]) that the angle between the crystal and the optical axis will influence the refractive index.

As shown in Fig. 3a) we could determine that the phase-matching conditions are met when the incoming pulse has a $29.17°$ - tilt with respect to the optical axis. This also matches with experimental data ([4]).

For this angle we observe an increase of the amplitude depending on the crystal length and the intensity of the fundamental pulse (Fig. 3b) and Fig. 4). This effect can be explained through constructive interference. The created second harmonics are all in phase and traveling with the same group velocity, therefore the effect accumulates. On the other hand, this match only occurs for the central frequency of the pulse, for all other frequencies the condition is not valid. This means, that the pulse should broaden while moving through the nonlinear crystal, which is also visible in Fig. 4. As also expected from the theory, the intensity of the second harmonic depends quadratically on the intensity of the fundamental pulse.

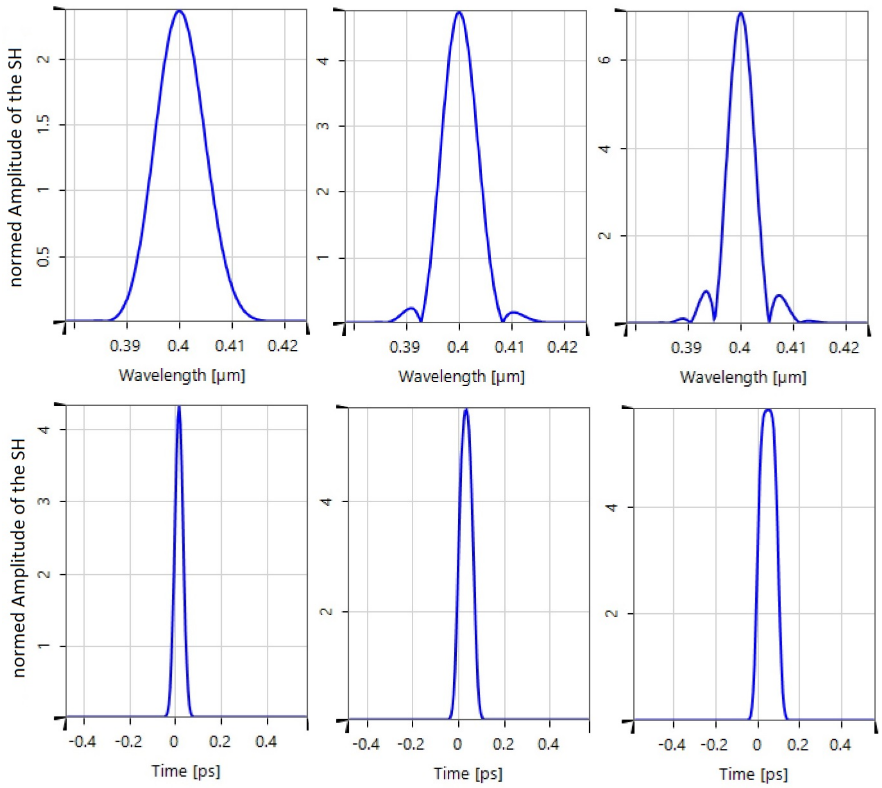

Figure 4: Second harmonic at various crystal lengths in the phase-matching case. The angle is 29.17° and the nonlinear material BBO. The costructive Interference accumulates, resulting in a constant growth of the amplitude of the second harmonic.

Bibliography

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