Solution to Problem Set 9
Nonlinear Optics (NLO)

Due: 4. July 2018

1) Second-harmonic generation in a BBO crystal

In this tutorial, second harmonic generation (SHG) using femtosecond laser pulses will be discussed. A mathematical program (for example MATLAB or Mathematica) is required for evaluation and visualization of the equations. For KIT students, MATLAB can be downloaded for free. The SCC provides a guide for the installation that can be accessed here: https://www.scc.kit.edu/downloads/sca/Matlab-Aktivierung-Studierende-v1.1.pdf. The guide is available in German only, but it has instructive pictures.

A titanium sapphire (Ti:Sa) laser creates 30 fs pulses with an average power of 2 W at a repetition rate of 100 MHz. Although the average power seems to be deceptively low, the peak power level that is reached by this laser amounts to 0.6 MW. The wavelength can be tuned in the range between 700 nm and 1000 nm. The laser is focussed on a birefringent crystal for an efficient generation of SHG pulses that have various applications in chemistry, semiconductor physics, and life sciences.

Beta Barium Borate (BBO), β-BaB_2O_4, is a uniaxial crystal that is often used for frequency doubling applications. For wavelengths λ emitted by the Ti:Sa laser, the ordinary refractive index n_o as well as the extraordinary refractive index n_e of BBO are given by the following empirical equations (for λ expressed in µm, valid in the range from 0.22 µm to 1.06 µm):

$$\begin{aligned} n_o^2(\lambda) &= 2.7359 + \frac{0.01878}{\lambda^2 - 0.01822} - 0.01354\lambda^2 \\ n_e^2(\lambda) &= 2.3753 + \frac{0.01224}{\lambda^2 - 0.01667} - 0.01516\lambda^2 \end{aligned} \tag{1}$$

1. Plot n_o and n_e as functions of wavelength in the range between 0.3 µm and 1 µm, and comment whether BBO is a positive or a negative uniaxial crystal.

Solution

The plots of n_o and n_e as functions of wavelength are given in Fig. 1. We can see that n_e is smaller than n_o, therefore, the crystal is negative uniaxial.

2. What is the phase matching condition required for an efficient second-harmonic generation? Is SHG in the given wavelength range possible without using critical phase matching or thermal tuning?

Solution

The phase matching condition required for an efficient SHG is that the wave vector mismatch is 0. In the following equations, the subscript 1 denotes the fundamental (pump) wave, and the subscript 2 denotes the generated 2nd harmonic wave. We can write:

$$\Delta k = k_1 + k_1 - k_2 = 2k_1 - k_2 = 0$$
$$2\frac{2\pi}{\lambda_1} = \frac{2\pi}{\lambda_2} \Leftrightarrow 2\frac{2\pi n(\lambda_1) f_1}{c} = \frac{2\pi n(\lambda_2) f_2}{c}$$
$$2n(\lambda_1)\omega_1 = 2n(\lambda_2)\omega_2$$

SHG means that $2\omega_1 = \omega_2$, so the phase matching condition is finally:

$$n(\lambda_1) = n(\lambda_2) \text{ or } n(\lambda_1) = n(\lambda_1/2).$$

The non-critical phase matching means that all waves are polarized along principal axes of the crystal, therefore, the waves can experience the indices of refraction n_e or n_o only. From Fig. 1 it can be easily understood that the condition $n(\lambda_1) = n(\lambda_1/2)$ cannot be fulfilled within the given wavelength range, because even the minimum value of n_o is larger than the maximum value of n_e.

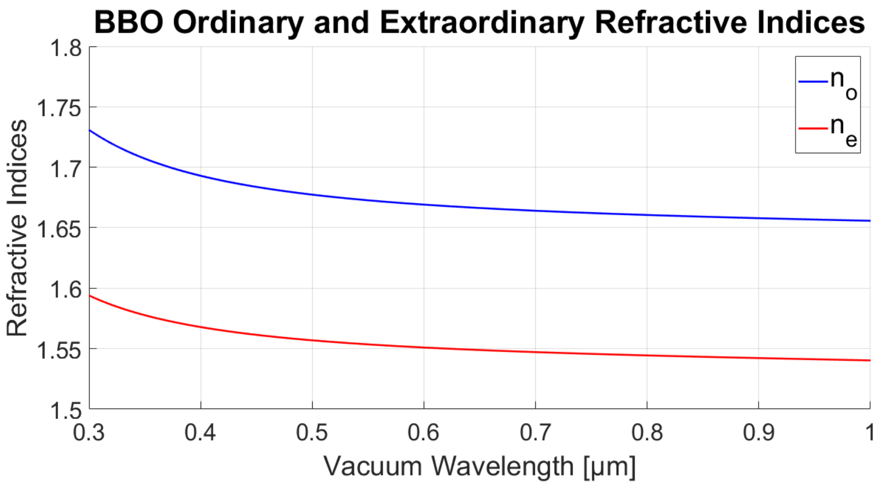

Fig.1: Ordinary and extraordinary refractive indices of Beta Barium Borate (BBO). It can be easily seen that the non-critical phase matching for SHG is not possible within the given vacuum wavelength range. This is because within this range there is no single refractive index value that is accessible by n_e and n_o at the same time (the maximum value of n_e is smaller than the minimum value of n_o).

3. Assuming critical phase matching of type-1, is SHG possible in the whole wavelength range? Calculate and plot the phase matching angle for all wavelengths that are accessible by type-1 phase matching.

Solution

In order to get phase matching, we must employ the angle tuning technique (also referred to as critical phase matching). The fundamental wave is polarized along the ordinary axis and experiences n_o, while the second harmonic wave experiences an angle-dependent extraordinary refractive index $n_e(2\omega_1,\theta)$, where θ is the angle between the k-vector and the optical axis of the crystal. By adjusting the θ, we can get the phase matching condition: $n_o(\omega_1) = n_e(2\omega_1,\theta)$ or $n_o(\lambda_1) = n_e(\lambda_1/2,\theta)$.

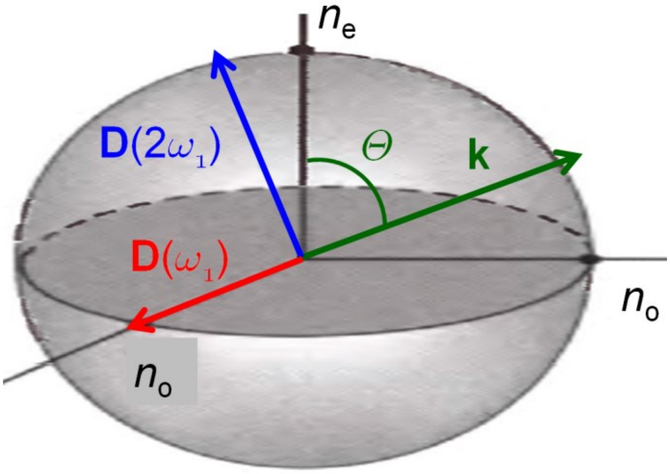

Fig.2: The fundamental wave (red) propagates as an ordinary mode under influence of a refractive index $n_o(\omega_1)$, while the second harmonic (blue) experiences an angle-dependent extraordinary refractive index $n_e(2\omega_1,\theta)$. (Figure adapted from: B. E. A. Saleh and M. C. Teich, *Fundamentals of Photonics.* John Wiley and Sons, 2007.

It can be shown that the phase matching condition and the corresponding angle θ_p can be expressed in the following way (see lecture notes):

$$\frac{1}{n_e^2(2\omega_1,\theta)} = \frac{\sin^2\theta}{n_e^2(2\omega_1)} + \frac{\cos^2\theta}{n_o^2(2\omega_1)} = \frac{1}{n_o^2(\omega_1)},$$

$$\tan\theta_p = \frac{n_e(2\omega_1)}{n_o(2\omega_1)}\sqrt{\frac{n_o^2(\omega_1)-n_o^2(2\omega_1)}{n_e^2(2\omega_1)-n_o^2(\omega_1)}}.$$

The wavelength range of the laser is between 700 nm and 1000 nm, meaning that the wavelength range of the generated SHG waves lies between 350 nm and 500 nm. The plot of the phase matching angle within the wavelengths accessible by the laser is shown in Fig. 3a.

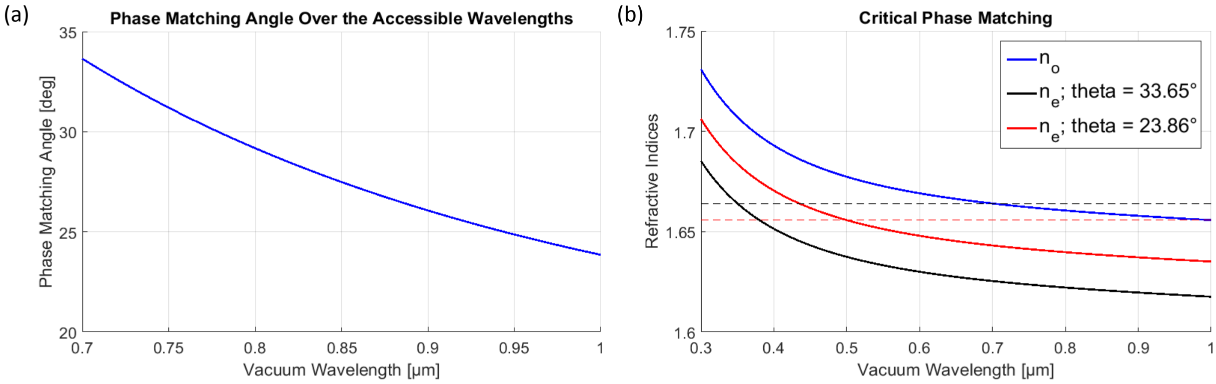

Fig 3. (a) Phase matching angle for SHG over the accessible wavelengths. (b) An illustration of the phase matching condition for $\lambda_1 = 1000\text{nm}$ and $\lambda_1 = 700\text{nm}$. The dashed red line indicates that the n_o at $\lambda_1 = 1000\text{nm}$ is equal to the n_e at $\lambda_2 = 500\text{nm}$ for $\theta = 23.86°$, therefore the SHG phase matching is fulfilled. The dashed blacked line indicates the same for $\lambda_1 = 700\text{nm}$ and $\lambda_2 = 350\text{nm}$ for $\theta = 33.65°$.

4. Plot the wavelength dependence of the walk-off angle between the *k*-vector and the Poynting vector of the SHG wave.

Solution

For critical phase matching using angle tuning, the fundamental wave is an ordinary wave, and therefore its Poynting vector (power flux) is parallel to its *k*-vector (propagation direction of the wave front). This is not the case for the generated wave at the second harmonic frequency, which has an ordinary and an extraordinary component. In this case the *k*-vector and the Poynting vector are not parallel; there is a “walk-off” angle ρ between them.

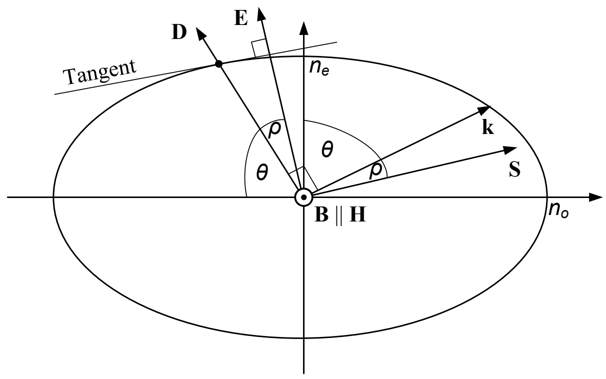

Fig 4. There are two sets of orthogonal vectors: **k**, **D**, **B** and **S**, **E**, **H**. The “walk-off” angle ρ can be found by first finding the point on the ellipse that the vector **D** intercepts (depending on θ). The vector **E** is perpendicular to the tangent to the ellipse constructed at this point. The angle ρ can be calculated from the dot product of vectors **D** and **E**. Since the dot product of the two vectors is equal to the product of their magnitudes and the cosine of the angle between them, then $\cos(\rho) = \dfrac{\mathbf{E} \cdot \mathbf{D}}{|\mathbf{E}||\mathbf{D}|}$.

We get: $\cos(\rho) = \frac{\mathbf{E}\cdot\mathbf{D}}{|\mathbf{E}||\mathbf{D}|} = \frac{n_e^2(\lambda_1/2)\cos^2(\theta_p) + n_o^2(\lambda_1/2)\sin^2(\theta_p)}{\sqrt{n_e^4(\lambda_1/2)\cos^2(\theta_p) + n_o^4(\lambda_1/2)\sin^2(\theta_p)}}$, or equivalently:

$\rho = \mp \quad \mathrm{an}\left(\frac{n_o^2}{n_e^2}\tan(\theta_p)\right)$. In the last equation, the upper signs are for negative, and the lower ones are for positive uniaxial crystals. The walk-off angle plot for the wavelengths accessible by the laser is displayed in Fig. 5.

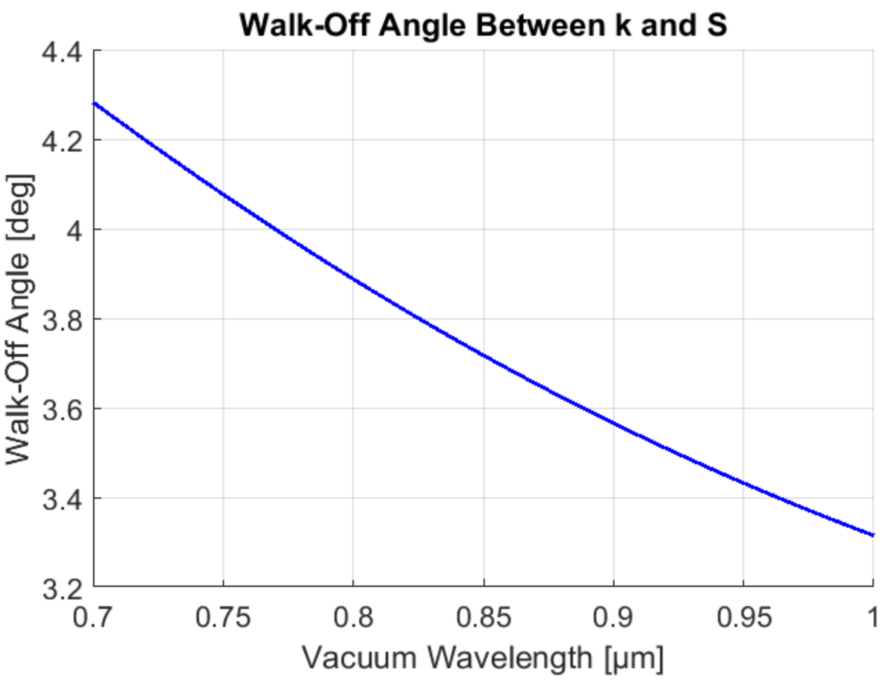

Fig .5 Walk-off angle between the wave vector and the Poynting vector for the second harmonic wave, at the wavelengths of the fundamental wave that are accessible by the laser.

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