# Problem Set 9
# Nonlinear Optics (NLO)

Due: 29. June 2016

## 1) Second-harmonic generation in a BBO crystal

In this tutorial, second harmonic generation (SHG) using femtosecond laser pulses will be discussed. A computational program (for example MATLAB or Mathematica) is required for the evaluation and visualization of the equations concerning the tutorial.

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), $\beta$-BaB$_2$O$_4$, is a uniaxial crystal that is often used for frequency doubling applications. In the wavelength range given by the Ti:Sa source laser, the ordinary (extraordinary) refractive index $n_o$ ($n_e$) of BBO is given by the following empirical equations (wavelength $\lambda$ in µm)

$$n_o^2(\lambda) = 2.7359 + \frac{0.01878}{\lambda^2 - 0.01822} - 0.01354\lambda^2$$

$$n_e^2(\lambda) = 2.3753 + \frac{0.01224}{\lambda^2 - 0.01667} - 0.01516\lambda^2$$

(1)

1. Plot $n_o$ and $n_e$ as a function of wavelength and comment whether BBO is a positive or a negative uniaxial crystal?

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?

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.

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

**Solution**

In the case of SHG the phase matching condition can be expressed directly in terms of refractive indices as

$$\Delta k = k(2\omega) - 2k(\omega) = \frac{2\omega}{c} n_{2\omega} - 2 \frac{\omega}{c} n_{\omega} = 2k_0 \left[ n(2\omega) - n(\omega) \right] \overset{!}{=} 0$$

$$n(2\omega) = n(\omega)$$

This relation is generally not fulfilled due to material dispersion. This however can be compensated for, e.g. by exploiting the birefringence of a uniaxial crystal.

1. By plotting Eq. (1) we see that the refractive index in the wavelength range between 300 and 1000 nm is lower for the extraordinary refractive index ($n_e < n_o$), so the crystal is negatively uniaxial.

2. Noncritical phase matching in BBO (wave vector orthogonal to extraordinary principal axis of the crystal):

    a. Fundamental (pump) wave is ordinary wave

    b. Second Harmonic is extraordinary wave, polarized orthogonally with respect to the fundamental

   The phase matching condition can be written as $n_o(\lambda) = n_e(\lambda/2)$, which cannot be fulfilled in the interesting wavelength region.
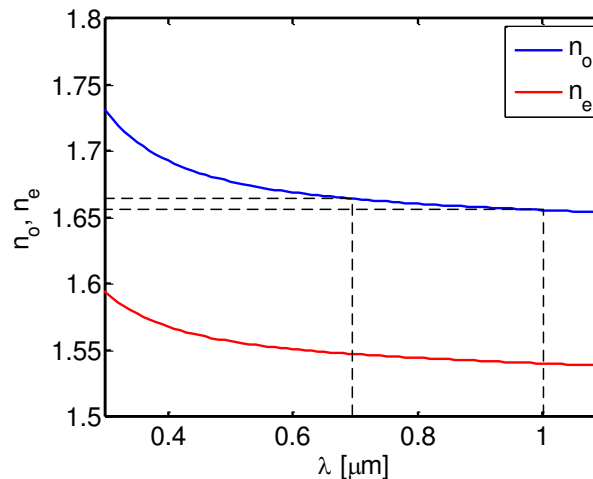


3. Critical phase matching (propagation vector not orthogonal to the extraordinary principal axis of the crystal)
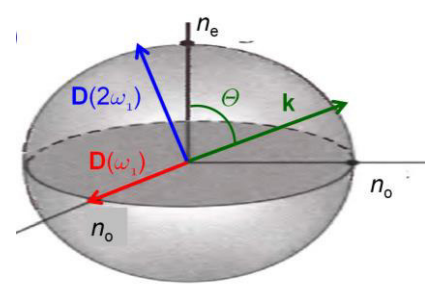
   Type 1:

    a. Fundamental (pump) wave is ordinary wave

    b. Second harmonic has ordinary and extraordinary components:

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

(4)

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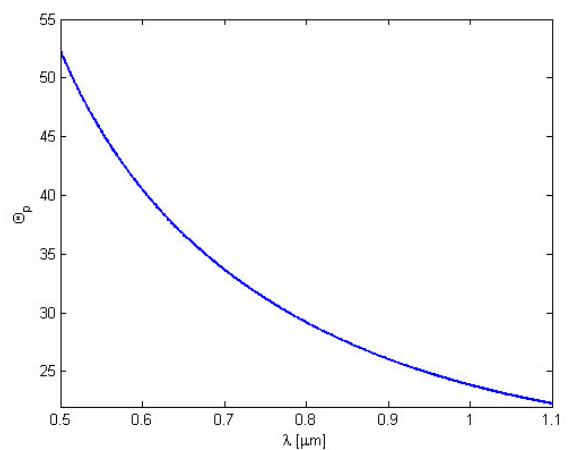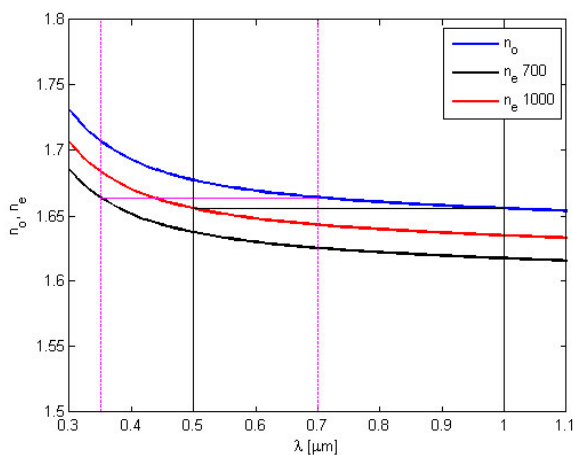


For 700 nm

$$\theta_p = 33.65°$$

For 1000 nm

$$\theta_p = 23.85°$$



Dispersion diagram indicating the phase matching angle for $\lambda = 700\,\text{nm}$ and $\lambda = 1000\,\text{nm}$ and phase matching angle over wavelength

4. For critical phase matching using angle tuning the *k*-vectors of the fundamental wave and the second harmonic wave are not parallel. The fundamental wave is an ordinary wave and the therefore its Poynting vector (propagation direction of the energy) 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 is not parallel anymore and there is a 'walk-off' between the pump and the second harmonic. This 'walk-off' limits the interaction length between the fundamental and the frequency doubled wave.

The walk-off angle can be found by looking at the index ellipsoid
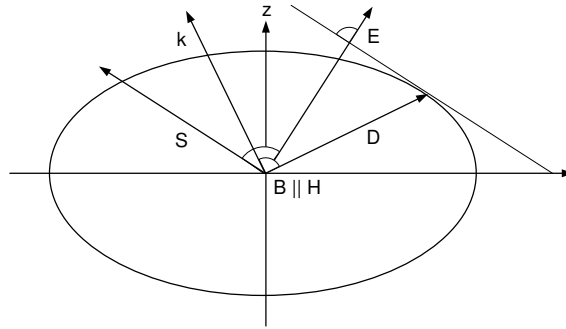
In an anisotropic medium the electric field vector and the displacement field are not parallel anymore:

$$\vec{D} = \underline{\varepsilon}\vec{E} \tag{5}$$

There are two sets of orthogonal vectors

$$
\begin{aligned}
\vec{k}, \vec{D}, \vec{B} \\
\vec{S}, \vec{E}, \vec{H}
\end{aligned}
\tag{6}
$$

It can be shown (see lecture notes) that the E-Field vector can be found by constructing the tangent to the ellipse in the intersection point of the displacement field vector D. The normal vector is the electric field.
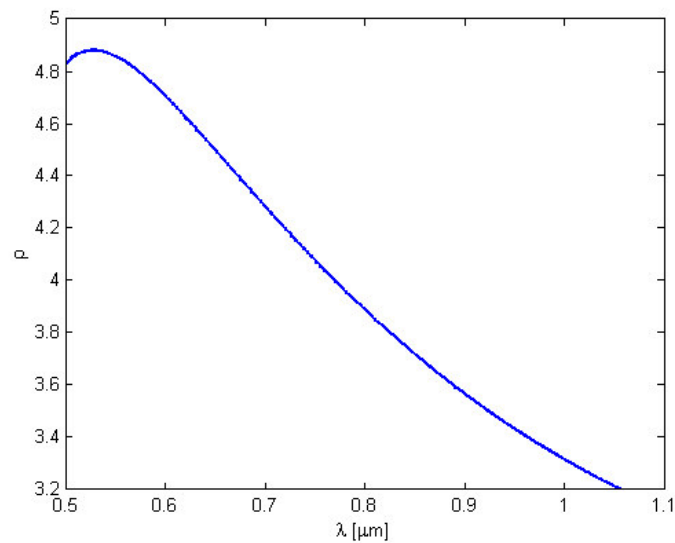
The walk-off angle for a negatively uniaxial crystal can be calculated from

$$\rho = -\theta + \arctan\left\{\frac{n_o^2}{n_e^2}\tan\theta\right\} \tag{7}$$
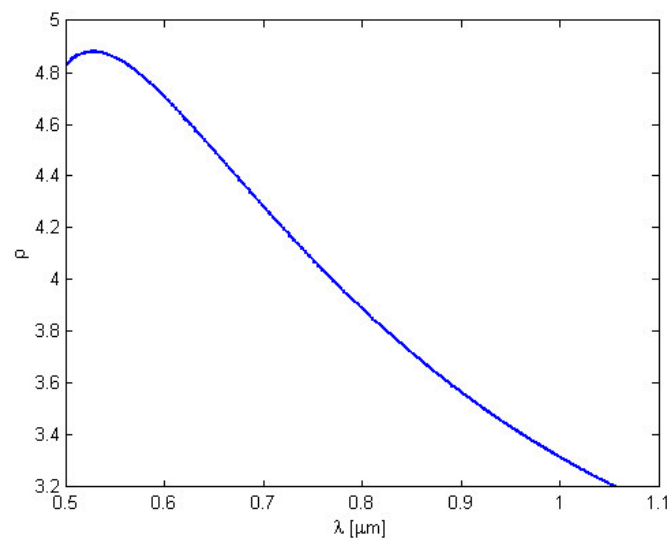
Note: The walk-off angle can also be calculated from the following equation (Eqn. 3.66 in the script)

$$\cos(\rho_k) = \frac{n_e^2(\lambda/2)\cos^2(\Theta_p) + n_e^2(\lambda/2)\sin^2(\Theta_p)}{\sqrt{n_e^4(\lambda/2)\cos^2(\Theta_p) + n_e^4(\lambda/2)\sin^2(\Theta_p)}}$$



## Questions and Comments:

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