PHYS2202 Nonlinear Optics
Problem Set 7 solutions

1. (20 points) **Nonlinear crystal orientation and length**
   Suppose that you have a pulsed Ti:sapphire laser system producing 100 fs pulses at 800 nm wavelength and you would like to generate terahertz pulses at ∼20 THz (15 μm wavelength). As a first step, you will convert the Ti:sapphire output to signal and idler beams in the infrared (frequencies ω_s and ω_i), which you will then mix in an appropriate nonlinear crystal (e.g., GaSe) to generate the terahertz radiation by difference frequency mixing $\omega_s - \omega_i = \omega_{\mathrm{THz}}$.

   You need to purchase a β-BBO (BBO=BaB_2O_4, barium borate, symmetry group: $3m$) crystal to convert the 800 nm light to the infrared. Suppose that your laser source produces pulses of 800 nm light in a TEM_{00} spatial mode with an energy of 20 μJ.

   Explain which type of phase matching (Type I or II) you should use to realize the most efficient conversion of the 800 nm light, and then explain the orientation, length, and cross section of the BBO crystal.

The following are some relevant parameters for BBO:

1. Wavelength dispersion of the refractive indices:

$$\begin{aligned} n_o^2 &= 2.7405 + \frac{0.0184}{\lambda^2 - 0.0179} - 0.0155\lambda^2 \\ n_E^2 &= 2.3730 + \frac{0.0128}{\lambda^2 - 0.0156} - 0.0044\lambda^2, \end{aligned} \tag{1}$$

   where λ is in μm.

2. Effective nonlinearity[1]:

$$\begin{aligned} d_{\mathrm{ooe}} &= d_{31}\sin\theta - d_{22}\cos\theta\sin 3\phi \\ d_{\mathrm{eoe}} &= d_{\mathrm{oee}} = d_{22}\cos^2\theta\cos 3\phi \\ d_{22} &= \pm(2.22 \pm 0.09)\,\mathrm{pm/V} \\ d_{31} &= \pm(0.16 \pm 0.08)\,\mathrm{pm/V} \end{aligned} \tag{2}$$

3. Angular acceptance at 800 nm ∼0.8 mrad cm.

4. Damage threshold for 1 ps pulses at 1064 nm ∼50 GW/cm^2. Suppose that the damage threshold for 100 fs, 800 nm pulses is the same.

To generate pulses of 20 THz frequency, we need to mix a pair of infrared beams of (angular) frequencies ω_s and ω_i, such that $\omega_s - \omega_i = 2\pi\,(20\,\mathrm{THz})$. The frequencies ω_s and ω_i are produced in a separate nonlinear process that must satisfy $\omega_{800\,\mathrm{nm}} = \omega_s + \omega_i$. Adding these two equations yields $2\omega_s = \omega_{800\,\mathrm{nm}} + \omega_{20\,\mathrm{THz}}$ or

$$\frac{1}{\lambda_s} = \frac{1}{2}\left(\frac{1}{800\,\mathrm{nm}} + \frac{1}{\lambda_s}\right) \tag{3}$$

$$= \frac{1}{2}\left(\frac{1}{800\,\mathrm{nm}} + \frac{20\,\mathrm{THz}}{c}\right) = \frac{1}{2}\left(\frac{1}{8\times 10^{-5}\,\mathrm{cm}} + \frac{1}{1.5\times 10^{-3}\mathrm{cm}}\right) = \frac{1}{1.519\,\mu\mathrm{m}}. \tag{4}$$

This in turn implies that the idler wavelength is

$$\frac{1}{\lambda_i} = \frac{1}{800\,\mathrm{nm}} - \frac{1}{\lambda_s} = \frac{1}{1.690\,\mu\mathrm{m}}. \tag{5}$$

We then need to phase match the process for converting 800 nm light to 1.519 μm and 1.690 μm light. Since the extraordinary index of BBO is smaller than the ordinary index, the highest frequency photons cannot be ordinarily polarized since, for $\omega_p > \omega_s > \omega_i$, dispersion means that

$$k_o(\omega_p) > k_o(\omega_s) > k_o(\omega_i) \tag{6}$$

and the *negative* uniaxial character means that $k_o(\omega) > k_e(\omega)$. Therefore, we have the following options for polarization of idler, signal, and pump (800 nm), respectively: ooe, eoe, and oee. We just need to solve the corresponding phase-matching equations

$$\vec{k}_p - \vec{k}_s - \vec{k}_i = 0 \tag{7}$$

or equivalently

$$\frac{n_e(\lambda_p,\theta)}{\lambda_p} - \frac{n_{o,e}(\lambda_s,\theta)}{\lambda_s} - \frac{n_{o,e}(\lambda_i,\theta)}{\lambda_i} = 0, \tag{8}$$

where $n_e(\lambda,\theta)$ is given by

$$\frac{1}{n_e^2(\lambda,\theta)} = \frac{\cos^2\theta}{n_o^2(\lambda)} + \frac{\sin^2\theta}{n_E^2(\lambda)}. \tag{9}$$

With the dispersion relations given, we find that we can phase match any of the three polarization combinations ooe, eoe, and oee (polarizations given in the order idler, signal, pump) if the beams travel at $\theta_{ooe} = 20.63^\circ$, $\theta_{eoe} = 28.09^\circ$, or $\theta_{oee} = 29.85^\circ$. Note how close the last two angles are; this is unsurprising given that the wavelengths of the signal and idler are very close to each other, so it should matter little which is ordinarily polarized and which is extraordinarily polarized.

The question now is which of these three polarization combinations is to be preferred. First, we need to know the magnitude of the effective nonlinear susceptibility in each case. Although the linear optical properties do not depend on the polarization angle ϕ relative to the crystal axes, the effective nonlinearities do. We see that for eoe and oee polarizations, the maximum effective nonlinearity is for $\phi = 0$, in which case

$d_{eoe} = d_{oee} = d_{22}\cos^2\theta$, while for ooe polarizations, the maximum effective nonlinearity is for $\phi = 90°$, in which case $d_{ooe} = d_{31}\sin\theta + d_{22}\cos\theta$. For the phase matching angles calculated, we then find $d_{ooe} = 2.13$ pm/V, $d_{eoe} = 1.73$ pm/V, and $d_{oee} = 1.67$ pm/V. Given that the nonlinear response is quadratic in the effective susceptibility (until significant pump depletion occurs), it looks like the ooe polarization combination is to be preferred. However, we should make sure that the interaction length as limited by group velocity mismatch is not so different between the polarization combinations to make us reconsider.

The group velocity is given by

$$v_g(\omega) = \left(\frac{dk}{d\omega}\right)^{-1} = \left(\frac{d}{d\omega}\frac{n(\omega)\omega}{c}\right)^{-1} = c\left(n(\omega) + \omega\frac{dn(\omega)}{d\omega}\right)^{-1}. \tag{10}$$

We can express this as a function of wavelength:

$$v_g(\lambda) = c\left(n(\lambda) + \frac{2\pi c}{\lambda}\frac{d\lambda}{d\omega}\frac{dn(\omega)}{d\lambda}\right)^{-1} = c\left(n(\lambda) - \lambda\frac{dn(\lambda)}{d\lambda}\right)^{-1}. \tag{11}$$

For the ordinary waves, the calculation is straightforward:

$$\frac{dn_o}{d\lambda} = \frac{d}{d\lambda}\sqrt{2.7405 + \frac{0.0184}{\lambda^2 - 0.0179} - 0.0155\lambda^2} = -\frac{1}{2n_o(\lambda)}\left[\frac{0.0184}{(\lambda^2 - 0.0179)^2}\lambda + 0.0155\lambda\right].$$

The group velocity is then

$$v_{go}(\lambda) = c\left\{n_o(\lambda) - \frac{1}{2n_o(\lambda)}\left[-\frac{0.0184}{(\lambda^2 - 0.0179)^2}2\lambda - 2(0.0155)\lambda\right]\right\}^{-1}. \tag{12}$$

For extraordinary waves there is just a little more tedium involved.

$$\frac{dn_e}{d\lambda} = \frac{d}{d\lambda}\left[\frac{\cos^2\theta}{n_o^2(\lambda)} + \frac{\sin^2\theta}{n_E^2(\lambda)}\right]^{-1/2} = -\frac{1}{2}n_e^3(\lambda,\theta)\frac{d}{d\lambda}\left[\frac{\cos^2\theta}{n_o^2(\lambda)} + \frac{\sin^2\theta}{n_E^2(\lambda)}\right] \tag{13}$$

$$= n_e^3(\lambda,\theta)\left[\frac{\cos^2\theta}{n_o^3(\lambda)}\frac{dn_o}{d\lambda} + \frac{\sin^2\theta}{n_E^3(\lambda)}\frac{dn_E}{d\lambda}\right], \tag{14}$$

where

$$\frac{dn_E}{d\lambda} = \frac{d}{d\lambda}\sqrt{2.3730 + \frac{0.0128}{\lambda^2 - 0.0156} - 0.0044\lambda^2} = -\frac{1}{2n_E(\lambda)}\left[\frac{0.0128}{(\lambda^2 - 0.0156)^2}\lambda + 0.0044\lambda\right]$$

The group velocity of an extraordinary wave is then given by

$$v_{ge}(\lambda,\theta) = c\Big\{n_e(\lambda,\theta) - \lambda n_e^3(\lambda,\theta)\Big(\frac{\cos^2\theta}{n_o^3(\lambda)}\frac{dn_o}{d\lambda} + \frac{\sin^2\theta}{n_E^4(\lambda)}\Big[-\frac{0.0128}{(\lambda^2 - 0.0156)^2}\lambda - 0.0044\lambda\Big]\Big)\Big\}^{-1}. \tag{15}$$

Putting everything together, we find that the group velocities of the pump, signal, and idler for the different polarization configurations and crystal angles are

λ	$\theta = 20.63°$	$\theta = 28.09°$	$\theta = 29.85°$
$v_{ge}(0.800\,\mu\text{m})$	$0.5993c$	$0.6040c$	$0.6053c$
$v_{go}(1.519\,\mu\text{m})$	$0.5975c$	$0.5975c$	$0.5975c$
$v_{ge}(1.519\,\mu\text{m})$	$0.6036c$	$0.6084c$	$0.6097c$
$v_{go}(1.690\,\mu\text{m})$	$0.5971c$	$0.5971c$	$0.5971c$
$v_{ge}(1.690\,\mu\text{m})$	$0.6033c$	$0.6081c$	$0.6094c$

For a pair of pulses of given group velocities, the delay that develops between the two pulses over a crystal length L is

$$\Delta t = L\left(v_{g,\min}^{-1} - v_{g,\max}^{-1}\right). \tag{16}$$

We see that the group velocity mismatch (GVM) between the fastest and slowest pulses in the three polarization configuration configurations we are considering is as in the following table

$v_{g,\max} - v_{g,\min}$	$\theta_{ooe} = 20.63°$	$\theta_{eoe} = 28.09°$	$\theta_{oee} = 29.85°$	$\Delta t/L$
$v_{ge}(0.800\,\mu\text{m}) - v_o(1.690\,\mu\text{m})$	$0.0022c$			20 fs/mm
$v_e(1.690\,\mu\text{m}) - v_{go}(1.519\,\mu\text{m})$		$0.0106c$		97 fs/mm
$v_e(1.519\,\mu\text{m}) - v_{go}(1.690\,\mu\text{m})$			$0.0126c$	115 fs/mm

Not only is the nonlinearity larger for the type I case (ooe), but the group velocity mismatch is smaller, so we definitely want to use this configuration. The question now is how long a crystal we need.

Given that our laser pulses have a temporal length of 100 fs and that the temporal separation of pump and idler pulses is 20 fs/mm, there is not much point in using a crystal longer than 5 mm. However, in principle we may be able to use a shorter crystal if the intensity of our pump is sufficiently large. We will aim for the largest intensity that is below the damage threshold and allows us to still be within the acceptance angle of the crystal.

Supposing a Gaussian TEM_{00} spatial mode with a Gaussian temporal profile, the spatio-temporal profile is

$$I(x,y,t) = I_0 e^{-(x^2+y^2)/(w/\sqrt{2})^2 - t^2/(\tau/\sqrt{2})^2}. \tag{17}$$

For either direction normal to the direction of propagation or either direction in time, the intensity reaches half its maximum at $t_{1/2} = \pm\sqrt{\frac{1}{2}\ln 2}\tau = \pm 0.59\tau$ or $x_{1/2} = \pm\sqrt{\frac{1}{2}\ln 2}w = \pm 0.59w$. For a given beam width and length, we can determine I_0 by simply integrating over x, y, and t to obtain the total pulse energy:

$$\begin{aligned} U_{\text{pulse}} &= \int_{x=-\infty}^{\infty} dx \int_{y=-\infty}^{\infty} dy \int_{t=-\infty}^{\infty} dt I(x,y,t) \\ &= I_0 \int_{x=-\infty}^{\infty} dx \int_{y=-\infty}^{\infty} dy \int_{t=-\infty}^{\infty} dt\; e^{-(x^2+y^2)/(w/\sqrt{2})^2 - t^2/(\tau/\sqrt{2})^2} \\ &= I_0 \left(\sqrt{\frac{\pi}{2}} w\right)^2 \sqrt{\frac{\pi}{2}} \tau. \end{aligned} \tag{18}$$

For a pulse energy of $U_{\text{pulse}} = 20\,\mu\text{J}$, a damage threshold of 50 GW/cm^2, taking $\tau = 85\text{fs}$ (this would yield a pulse with a full width at half-maximum of $2*0.59(85\,\text{fs}) = 100\,\text{fs}$; the difference between a choice of 85 fs and 100 fs is an unimportant detail for the purposes of this problem), we find that the w is given by

$$w = \sqrt{\frac{U_{\text{pulse}}}{I_{\text{damage}}} \left(\frac{2}{\pi}\right)^{3/2} \frac{1}{\tau}} = \sqrt{\frac{2 \times 10^{-5}\,\text{J}}{5 \times 10^{10}\,\text{J}\,\text{s}^{-1}\text{cm}^{-2}} \left(\frac{2}{\pi}\right)^{3/2} \frac{1}{8.5 \times 10^{-14}\text{s}}} = 0.049\,\text{cm}. \tag{19}$$

This is so much larger than our wavelengths that if we were to focus the beam to such a size, the Rayleigh range would be much longer than any length scales of concern in this problems; the Rayleigh range for the longest wavelength in our problem (the wavelength for which the Rayleigh range would be shortest) is

$$z_{R,\max} = \frac{\pi w^2}{\lambda_{\min}} = \frac{\pi(0.049\,\text{cm})}{1.690 \times 10^{-4}\,\text{cm}} = 44\,\text{cm}. \tag{20}$$

How does the range of angles involved in such focusing compare to the angular acceptance, though? For a beam waist corresponding to $w = 0.049\,\text{cm}$, the beam radius a distance z from the waist is characterized by

$$w(z) = w_0 \sqrt{1 + \left(\frac{\lambda z}{\pi w_0^2}\right)^2}. \tag{21}$$

For large z, this is $w(z) \approx \dfrac{\lambda z}{\pi w_0}$. This corresponds to an angle of the half-cone of the beam of

$$\theta_{1/2} = \arctan\left(\frac{w(z)}{z}\right) \approx \arctan\left(\frac{\lambda}{\pi w_0}\right). \tag{22}$$

In our case, this yields

$$\theta_{1/2} = \arctan\left(\frac{1.690 \times 10^{-4}\,\text{cm}}{\pi\,(0.049\,\text{cm})}\right) = 0.063^\circ. \tag{23}$$

The specified angular acceptance of the crystal is 0.8 mrad cm or 0.045°cm. We already have seen that there is no point in a crystal longer than about 0.5 cm, which would correspond to an acceptance angle of $0.045^{\circ}\mathrm{cm}/(0.5\,\mathrm{cm}) = 0.9^{\circ}$, which is larger than what we would have for the smallest beam radius permitted by the damage threshold. Therefore, we can use such a small beam.

We just need to check for such a beam radius as permitted by the damage threshold whether walk-off of the beams occurs in less than 5 mm. The walk-off angle ρ is given by

$$\tan\rho(\lambda) = \frac{1}{2}n_e^2(\lambda,\theta)\left[\frac{1}{n_E^2(\lambda)} - \frac{1}{n_o^2(\lambda)}\right]\sin(2\theta), \tag{24}$$

where θ is here our phase-matching angle. We obtain a walk-off angle of $\rho = 2.9^{\circ}$. For a beam with a full-width at half maximum diameter of $d = 2(0.59\,w_0) = 2(0.59)(0.049\,\mathrm{cm}) = 0.058\,\mathrm{cm}$, spatial walk off will happen over a length

$$L_{\mathrm{walk-off}} \sim \frac{d}{\tan\rho} = 1.14\,\mathrm{cm} \tag{25}$$

This is longer than the 5 mm crystal length determined by group velocity mismatch.

In total, we will order a crystal cut at $\theta = 20.63^{\circ}$ and $\phi = 90^{\circ}$ with a length of 5 mm and width of about cross section of about 1 mm×1 mm. In reality we would probably order a crystal with larger cross section to avoid clipping the wings of the spatial mode.