Exercise 1, 2021: Kleinman symmetry, wavelength dependence of $\chi^{(2)}$, coherence length, Sellmeier equation

1. Kleinman Symmetry and the $\chi^{(2)}$ tensor

a. Show that if Kleinman symmetry holds, the second order nonlinear susceptibility tensor can be written as:

$$\boldsymbol{d} = \begin{pmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{16} & d_{22} & d_{23} & d_{24} & d_{14} & d_{12} \\ d_{15} & d_{24} & d_{33} & d_{23} & d_{13} & d_{14} \end{pmatrix}$$

b. The second order susceptibility tensor for $LiNbO_3$ is given by:

$$\boldsymbol{d} = \begin{pmatrix} 0 & 0 & 0 & 0 & d_{31} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{31} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{pmatrix}$$

Write the generated nonlinear polarizations in $\hat{x}$, $\hat{y}$ and $\hat{\boldsymbol{z}}$, at 2ω, for the following electric fields:

$$\begin{pmatrix} E_x \\ 0 \\ 0 \end{pmatrix} e^{i\omega t} + c.c; \quad \begin{pmatrix} 0 \\ E_y \\ 0 \end{pmatrix} e^{i\omega t} + c.c; \quad \begin{pmatrix} 0 \\ 0 \\ E_z \end{pmatrix} e^{i\omega t} + c.c; \quad \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix} e^{i\omega t} + c.c;$$

$$\begin{pmatrix} E_0 \\ -E_0 \\ 0 \end{pmatrix} e^{i\omega t} + c.c$$

2. Calculating phase mismatch as a function of wavelength

Calculate and draw a figure of the phase mismatch $\boldsymbol{\Delta k}$ (in $\frac{\boldsymbol{1}}{\boldsymbol{\mu m}}$) as a function of pump wavelengths (in the range $\boldsymbol{0.8 - 5.0 \mu m}$) for second harmonic generation in $\boldsymbol{KTiOPO_4}$. The following equation describes the extraordinary refractive index dependence on wavelength (Sellmeier equation). The wavelength $\boldsymbol{\lambda}$ is in $\boldsymbol{\mu m}$. It can be assumed that both the fundamental and SH waves are polarized in the $\hat{z}$ direction.

$$n_Z^2 = 4.59423 + \frac{0.06206}{\lambda^2 - 0.04763} + \frac{110.80672}{\lambda^2 - 86.12171}$$

3. Wavelength dependence of nonlinear coefficient and of SHG, Miller's rule

a. Wavelength dependence of $\chi^{(2)}$: The nonlinear coefficient $d_{33} = \frac{1}{2}\chi^{(2)}_{zzz}$, as measured by second harmonic generation of a $1.064\mu m$ pump (i.e. 1.064 microns converted to 0.532 microns) in $KTiOPO_4$ is given as $d_{33}(1.064\mu m)$. Using the expression derived in class for the second order susceptibility (modified Lorentz model), evaluate the nonlinear coefficient in case of a second harmonic generation with a pump wavelength of 4 microns (4 microns converted to 2 microns).

Hint: The wavelength dependent part comes from the linear susceptibility $\chi^{(1)}$. Assuming there are negligible losses, it is related to the refractive index by:

$\varepsilon = 1 + \chi^{(1)} = n_\lambda^2$. You can assume the refractive index of $KTiOPO_4$ is given at any wavelength.

b. Miller's rule: Miller found empirically that the quantity:

$$\frac{\chi^{(2)}(2\omega;\omega,\omega)}{\chi^{(1)}(2\omega)[\chi^{(1)}(\omega)]^2}$$

is nearly constant for all non-centrosymmetric materials. Explain why.

c. Wavelength dependence of SHG: Assume that a 1.064 micron pump is frequency doubled in a $KTiOPO_4$ crystal, and that the efficiency of the process is given as:

$$\eta_{1.046\mu m} \triangleq \frac{I_{0.532\mu m}}{I_{1.046\mu m}} = 1\%$$

What will be the intensity of the second harmonic beam for frequency doubling of a 4 micron pump, having the same intensity? Assume phase matching is satisfied in both cases (**no need to get an actual number for $\boldsymbol{\eta_{4\mu m}}$ – an expression will suffice**).

4. Sellmeier equation and the Lorentz model

The equation that describes the dependence of the refractive index on the wavelength is called Sellmeier equation. Explain, using the Lorentz model, why the Sellmeier equation of $\boldsymbol{KTiOPO_4}$, written in question 2, has this shape.

Hints:

a. Assume that there are **two** resonances in the material (instead of the single resonance we found in Lorentz model). Actually there are three resonances but the frequency of the third is much smaller than that of the optical signal (and, therefore, this resonance can be neglected).

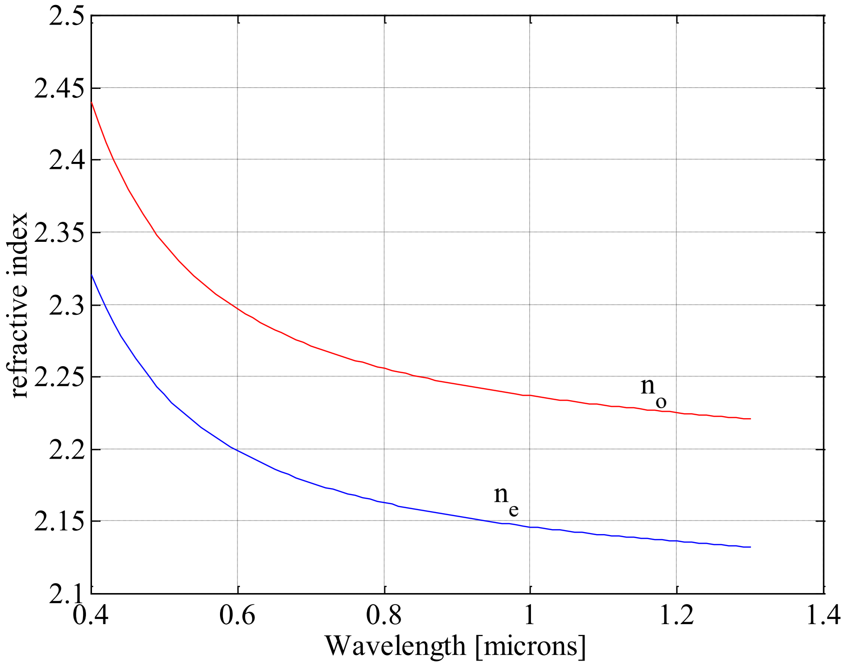
b. Assume that the resonance width is very narrow compared to the resonance frequency (i.e. damping can be neglected).

c. Another useful relation (prove it, and find relations between A, B and D) is:

$$\frac{A}{\omega_0^2 - \omega^2} = \frac{B}{\lambda^2 - \lambda_0^2} + D, \quad \omega_0 = \frac{2\pi c}{\lambda_0}$$

5. Dispersion and phase matching

In a uniaxial crystal, the refractive indices in the $\hat{x}$ and $\hat{y}$ directions are identical (this is called the *ordinary* refractive index n_o), and the refractive index in the $\hat{z}$ direction is different (the *extra-ordinary* refractive index n_e). The following figure shows the dispersion curves of a uniaxial crystal.



We want to use this crystal for frequency doubling of a laser source operating in the vicinity of λ=1μm. We are limiting this study only to the case of non-critical phase matching, in which the electric fields are polarized along the crystal principal axes ($\hat{x}$, $\hat{y}$ or $\hat{z}$).

a. Can this be done by birefringent phase matching? If so, what should be the polarization of the pump? What is the polarization of the second harmonic beam? Which coefficient d_{ij} should be used for this process?

b. What is the wavelength range (if any) that cannot be birefringently phase matched?

c. Assume now that we decide to double the 1 micron pump by first order quasi-phase-matching (QPM). We want both the fundamental and second harmonic beams to be polarized along the extraordinary (Z) direction. What should be the required QPM period? Which coefficient d_{ij} should be used for this process? Assume $n_e(\lambda) \cong 2.145,\ \ n_e\left(\frac{\lambda}{2}\right) = 2.2365$.