Ultrafast Optical Physics II (SoSe 2021)
Lecture 19, June 10

1) Second-order nonlinear optical effects

2) Phase matching

3) Difference frequency generation and introduction to optical parametric amplification (CW)

[5] Largely follows the review paper of Cerullo et al., "Ultrafast Optical Parametric Amplifiers" Rev. Sci. Instr. 74, pp 1-17 (2003)

Second-order nonlinear optical effects

Wave equation: $$\left(\Delta - \frac{1}{c_0^2}\frac{\partial^2}{\partial t^2}\right)\vec{E} = \underline{\mu_0 \frac{\partial^2}{\partial t^2}\vec{P}}.$$

Source term describing light-matter interaction

$$P = \varepsilon_0[\chi^{(1)}E + \chi^{(2)}E^2]$$

$\chi^{(2)} \longrightarrow$ 2nd order susceptibility

Example: Pockels Effect

$$E = A_0 + A_1\cos(\omega t)$$

$\longrightarrow$ Input electric field

The total polarization at frequency ω is:

$$P^{(\omega)} = \varepsilon_0[\chi^{(1)} + 2\chi^{(2)}A_0]A_1\cos(\omega t)$$

New refractive index:

$$n = \sqrt{1 + \chi^{(1)} + 2\chi^{(2)}A_0}$$

The Pockels effect is used to make optical switch (or modulator) using an electrical field to control the interaction between an optical crystal and the optical field propagating in it.

Mixing of two sine waves

$$E = \frac{1}{2}\left[\tilde{E}(\omega_1)e^{j\omega_1 t} + \tilde{E}(\omega_2)e^{j\omega_2 t} + \text{c.c.}\right] \longrightarrow$$ Input electric field

$$P_{\mathrm{NL}}^{(2)} = \frac{\varepsilon_0}{4}\{\underline{\chi^{(2)}(2\omega_1 : \omega_1, \omega_1)\tilde{E}^2(\omega_1)e^{j2\omega_1 t}} + \underline{\chi^{(2)}(2\omega_2 : \omega_2, \omega_2)\tilde{E}^2(\omega_2)e^{j2\omega_2 t}}$$

Second-harmonic generation (SHG)

$$+\underline{2\chi^{(2)}(\omega_1 + \omega_2 : \omega_1, \omega_2)\tilde{E}(\omega_1)\tilde{E}(\omega_2)e^{j(\omega_1+\omega_2)t}}$$

Sum-frequency generation (SFG)

$$+\underline{2\chi^{(2)}(\omega_1 - \omega_2 : \omega_1, -\omega_2)\tilde{E}(\omega_1)\tilde{E}^*(\omega_2)e^{j(\omega_1-\omega_2)t}}$$

Difference-frequency generation (DFG)

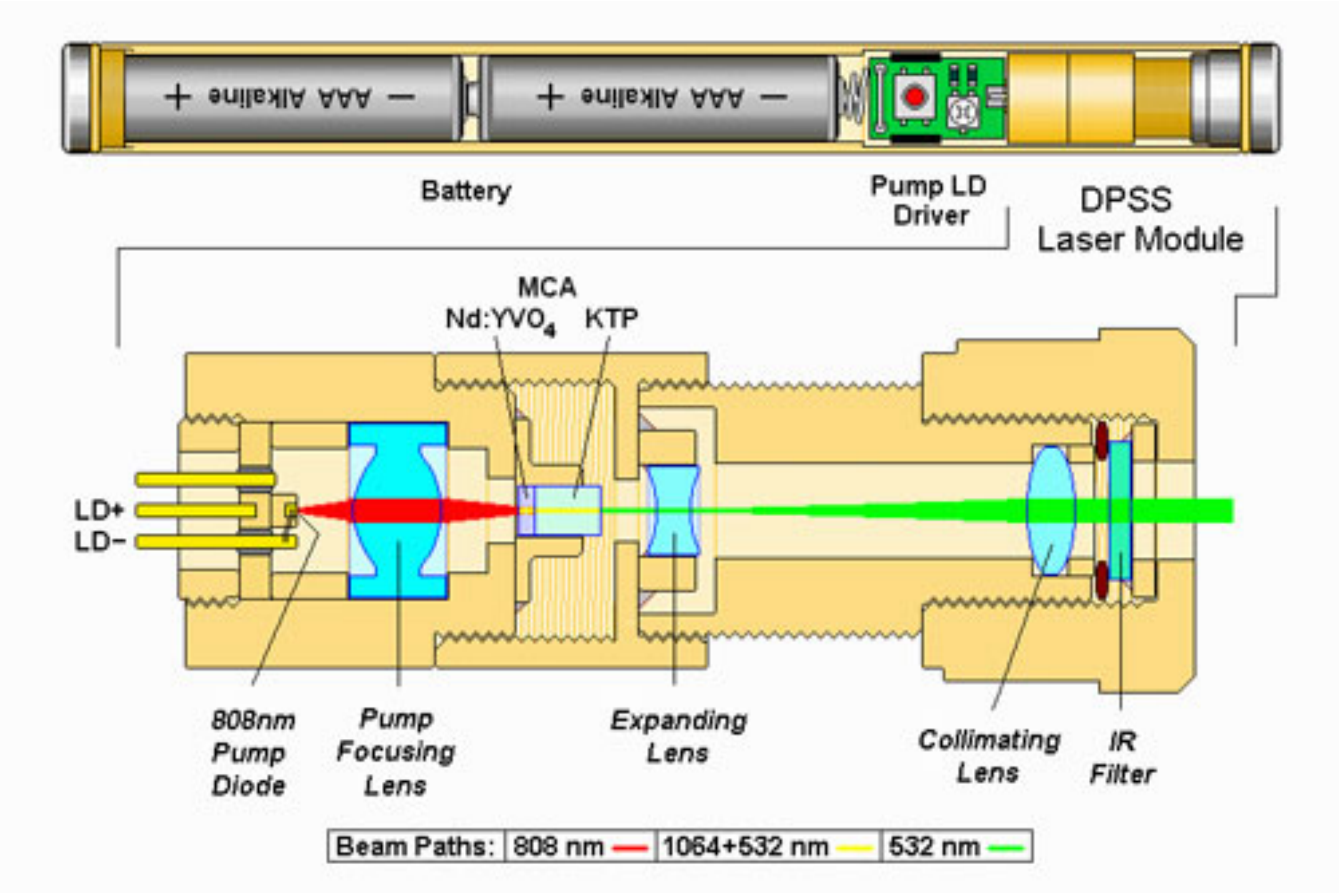
$$+\underline{\chi^{(2)}(0 : \omega_1, -\omega_1)\tilde{E}(\omega_1)\tilde{E}^*(\omega_1) + \chi^{(2)}(0 : \omega_2, -\omega_2)\tilde{E}(\omega_2)\tilde{E}^*(\omega_2)}\}$$

Optical rectification

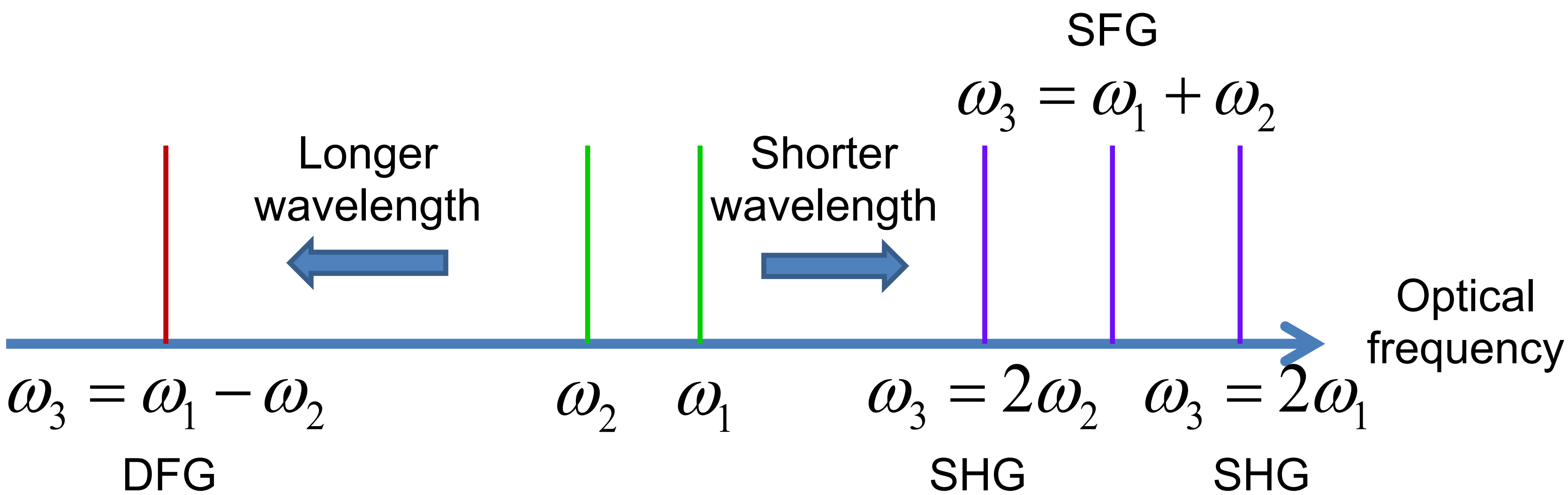
$\chi^{(2)}$ may be both complex and frequency dependent.

$\chi^{(2)}(\omega_a + \omega_b : \omega_a, \omega_b)$ keeps track of the input and output frequencies involved in a particular interaction.

SHG in daily life: green laser pointer

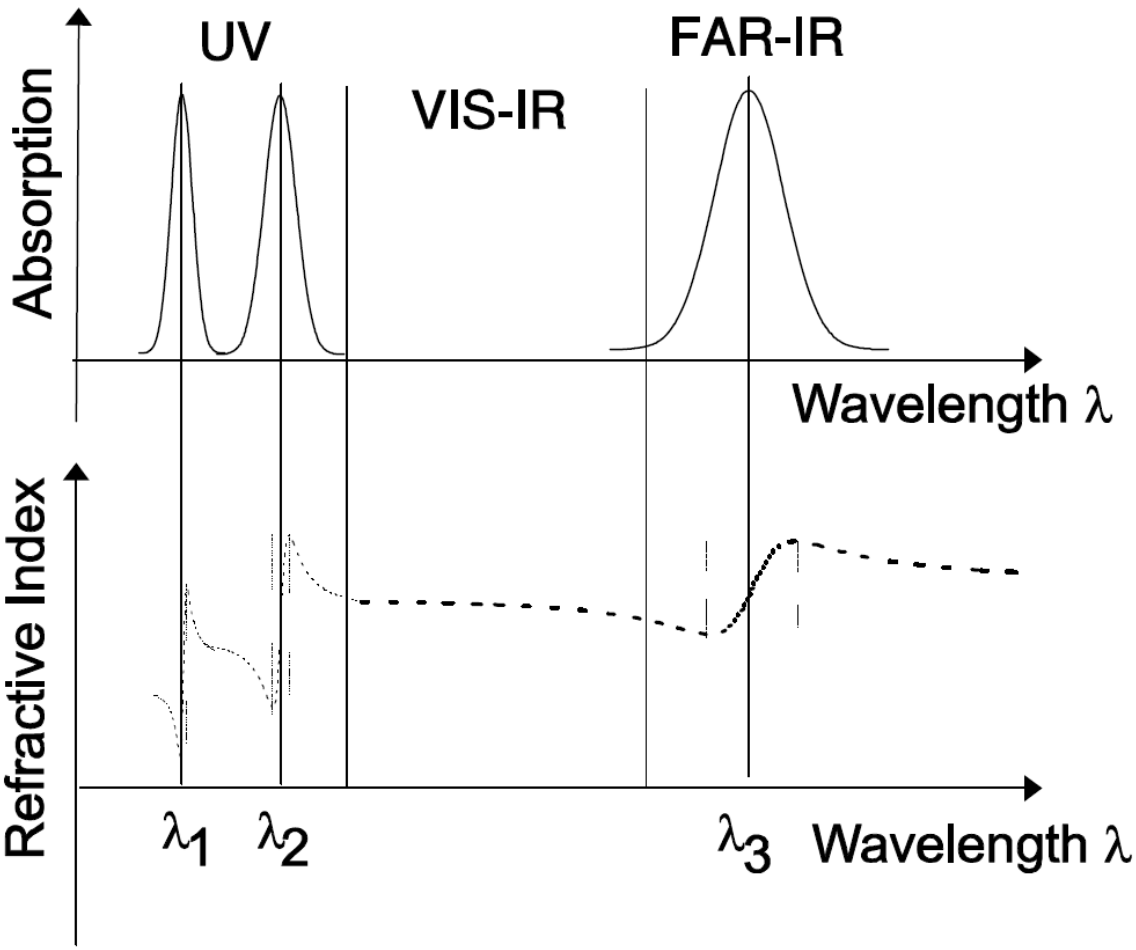


Wavelength conversion using 2nd order nonlinear optics



	Energy conservation	Momentum conservation	Phase matching condition
SHG	$\omega_3 = 2\omega_1$	$k_3 = 2k_1$	$\frac{\omega_3}{c} n_3 = 2\frac{\omega_1}{c} n_1$ $\quad n_3 = n_1$
SFG	$\omega_3 = \omega_1 + \omega_2$	$k_3 = k_1 + k_2$	$\omega_3 n_3 = \omega_1 n_1 + \omega_2 n_2$
DFG	$\omega_3 = \omega_1 - \omega_2$	$k_3 = k_1 - k_2$	$\omega_3 n_3 = \omega_1 n_1 - \omega_2 n_2$

How to achieve phase matching?

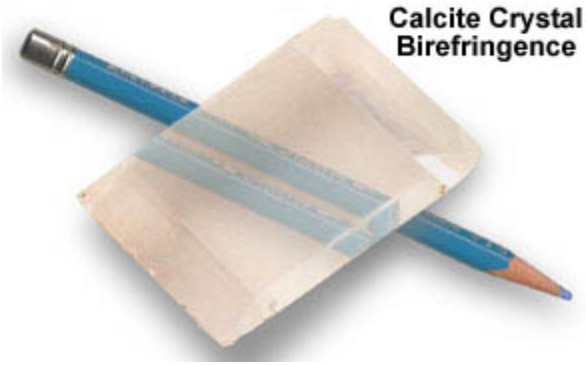


For the frequency (wavelength) far away from absorption resonance, refractive index increases with increasing frequency, which leads to

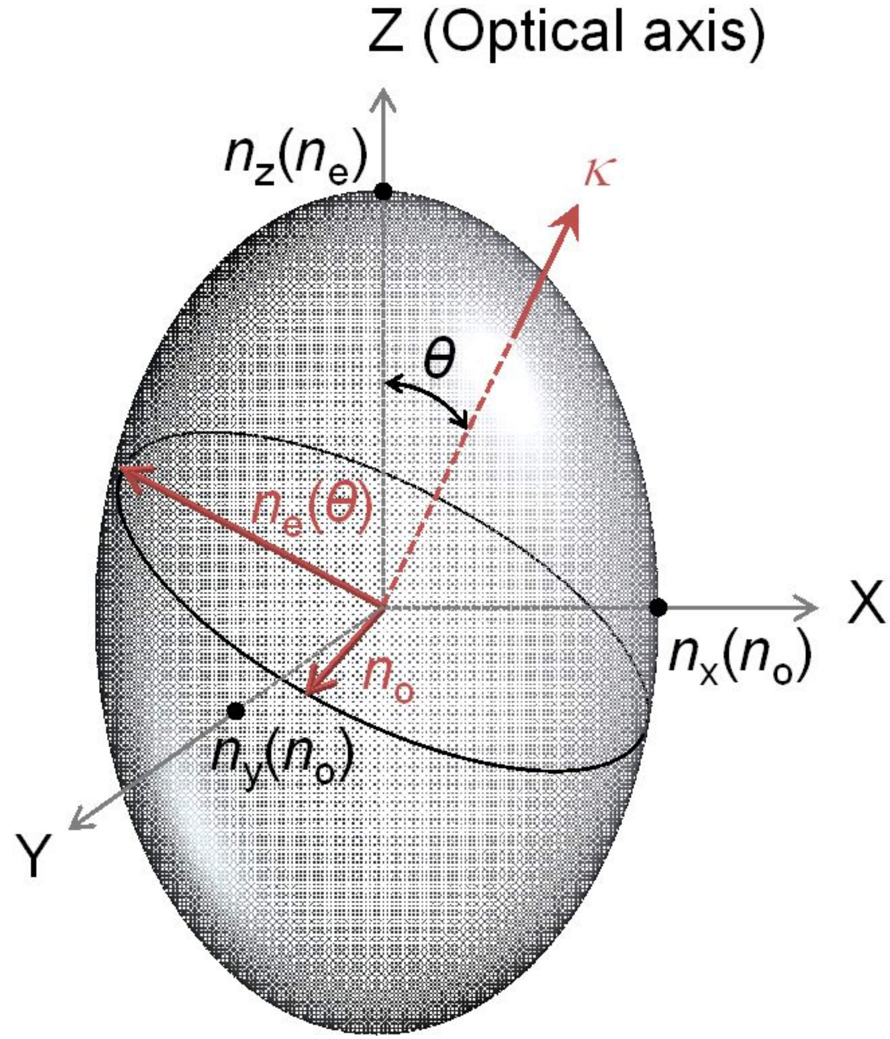
$$\omega_3 n_3 > \omega_1 n_1 + \omega_2 n_2$$

Dispersion prevents phase matching.

Phase matching in birefringent media



Birefringent materials have different refractive indices for different polarizations. **Ordinary** (o-wave) light has its polarization perpendicular to the optical axis and its refractive index, n_o , does not depend on propagation direction, θ. **Extraordinary** (e-wave) light has its polarization in the plane containing optical axis and propagation vector, and its refractive index, n_o , depends on propagation direction, θ.

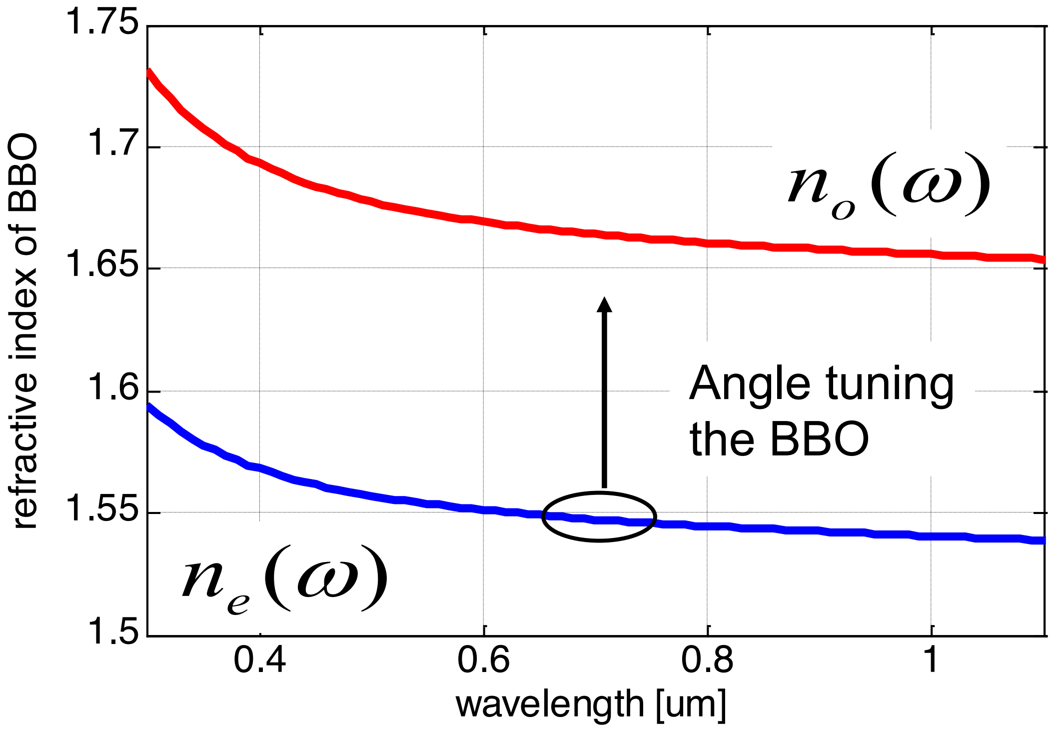


Phase matching in birefringent media

- In an isotropic medium, normal dispersion always results in

$$n(\omega) < n(2\omega)$$

- In birefringent uniaxial crystal there are ordinary wave and extraordinary wave.



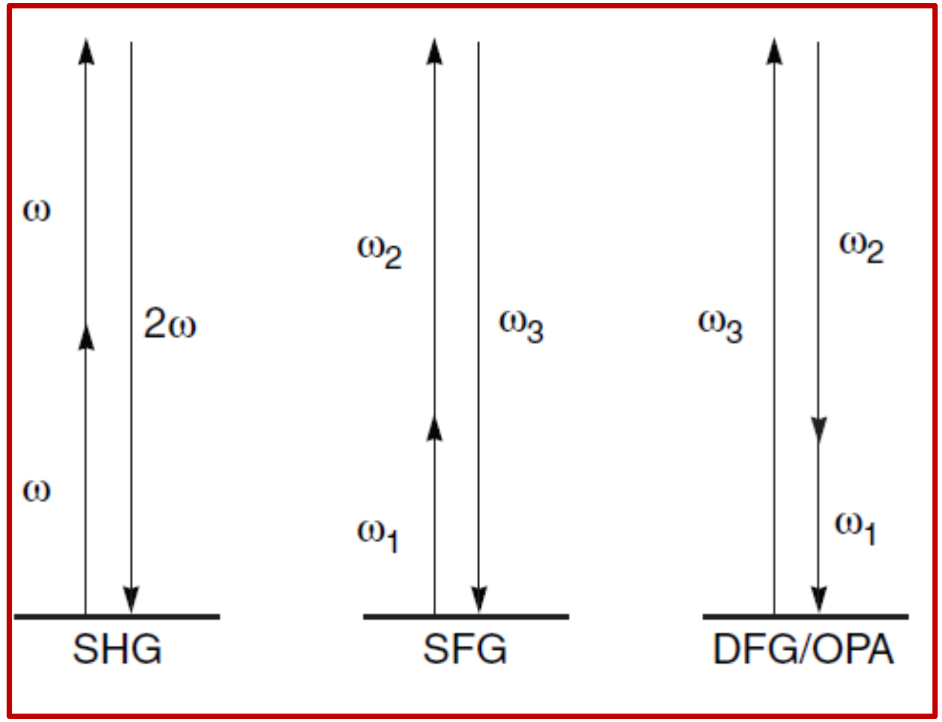
BBO crystal is a typical negative uniaxial crystal with $n_o > n_e$. If red light is set as the ordinary beam and the SHG the extraordinary one, angle tuning the BBO crystal permits achieving phase matching condition.

Phase matching: type I Vs. type II

In general, second-order nonlinear effects involve three waves with frequencies linked by the equation

$$\omega_1 + \omega_2 = \omega_3$$

Here ω_3 is the highest frequency of the three.



<u>Type I phase matching:</u>

ω_1 wave and ω_2 wave have the same polarization; that is, they are both ordinary waves or extraordinary waves: $o + o \rightarrow e$ or $e + e \rightarrow o$

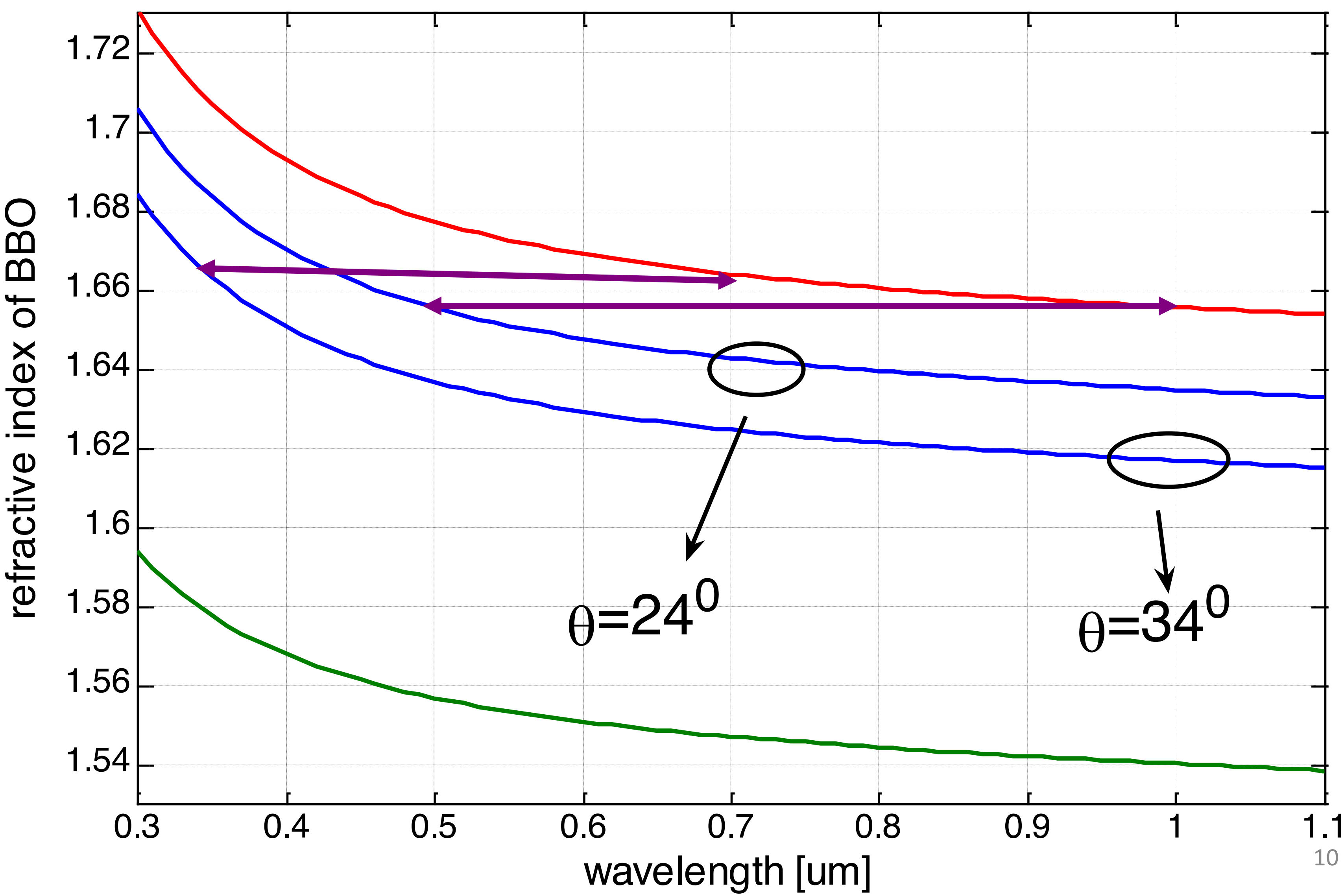
<u>Type II phase matching:</u>

ω_1 wave and ω_2 wave have different polarization: $o + e \rightarrow e$

$$o + e \rightarrow o \qquad e + o \rightarrow e \qquad e + o \rightarrow o$$

Type I phase matching SHG

$$n_e(34^0, 350nm) = n_o(700nm) \qquad n_e(24^0, 500nm) = n_o(1000nm)$$



Linear susceptibility is a matrix for optically anisotropic media

$$P = \varepsilon_0 \chi^{(1)} E \quad \begin{cases} P_x = \varepsilon_0 \chi^{(1)} E_x \\ P_y = \varepsilon_0 \chi^{(1)} E_y \\ P_z = \varepsilon_0 \chi^{(1)} E_z \end{cases}$$

Only true for optically isotropic media

For optically anisotropic media, linear susceptibility is a 3X3 matrix (a second-rank tensor):

$$\left.\begin{aligned} P_x &= \varepsilon_0 [\chi_{xx}^{(1)} E_x + \chi_{xy}^{(1)} E_y + \chi_{xz}^{(1)} E_z] \\ P_y &= \varepsilon_0 [\chi_{yx}^{(1)} E_x + \chi_{yy}^{(1)} E_y + \chi_{yz}^{(1)} E_z] \\ P_z &= \varepsilon_0 [\chi_{zx}^{(1)} E_x + \chi_{zy}^{(1)} E_y + \chi_{zz}^{(1)} E_z] \end{aligned}\right\}$$

$$P_i = \varepsilon_0 \sum_j \chi_{ij}^{(1)} E_j$$

$$(i, j) = (x, y, z)$$

2nd-order susceptibility is a 3rd-rank tensor

Take sum frequency generation(SFG) $\omega_1 + \omega_2 = \omega_3$ as an example:

$$P_x(\omega_3) = \varepsilon_0[\chi^{(2)}_{xxx}E_x(\omega_1)E_x(\omega_2) + \chi^{(2)}_{xxy}E_x(\omega_1)E_y(\omega_2) + \chi^{(2)}_{xxz}E_x(\omega_1)E_z(\omega_2)$$
$$+ \chi^{(2)}_{xyx}E_y(\omega_1)E_x(\omega_2) + \chi^{(2)}_{xyy}E_y(\omega_1)E_y(\omega_2) + \chi^{(2)}_{xyz}E_y(\omega_1)E_z(\omega_2)$$
$$+ \chi^{(2)}_{xzx}E_z(\omega_1)E_x(\omega_2) + \chi^{(2)}_{xzy}E_z(\omega_1)E_y(\omega_2) + \chi^{(2)}_{xzz}E_z(\omega_1)E_z(\omega_2)]$$

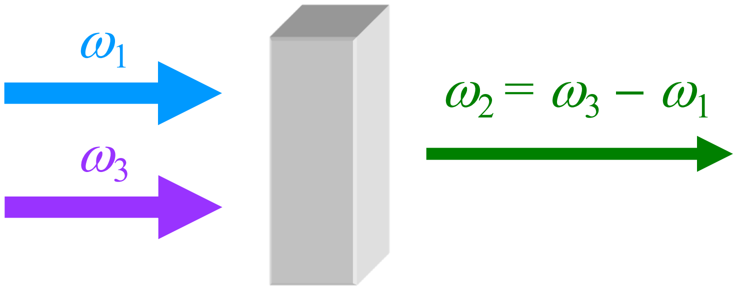
We can represent the lengthy expression using tensor notation:

$$P_i^{(2)}(\omega_3) = \varepsilon_0 \sum_{j,k} \chi^{(2)}_{ijk}(\omega_3 : \omega_1, \omega_2)E_j(\omega_1)E_k(\omega_2) \qquad (i,j,k) = (x,y,z)$$

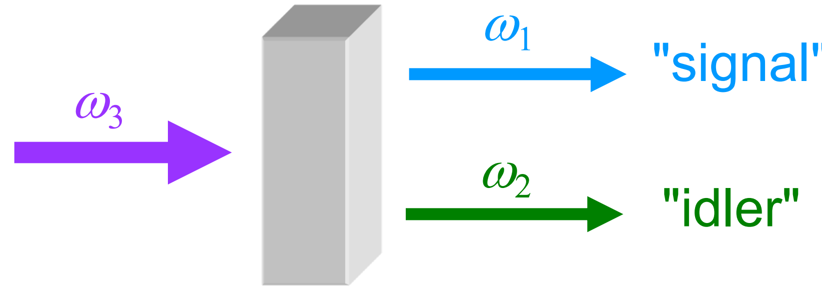
$\chi^{(2)}_{ijk}(\omega_3 : \omega_1, \omega_2)$ is a 3rd-order tensor with 27 (3X3X3) elements. According to the crystal symmetry, most of them are zeros.

Difference-frequency generation: optical parametric generation, amplification, oscillation

Difference-frequency generation takes many useful forms.

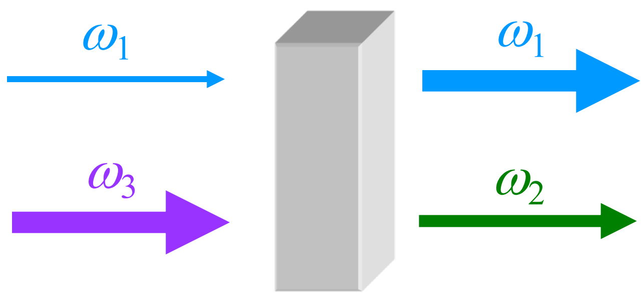


Parametric Down-Conversion
(Difference-frequency generation)

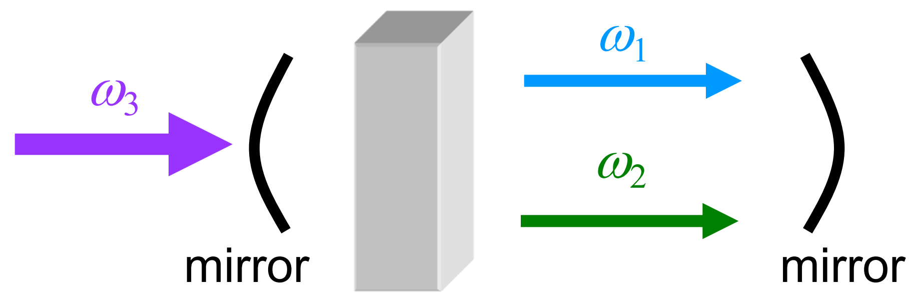


Optical Parametric
Generation (OPG)

By convention:
$\omega_{signal} > \omega_{idler}$



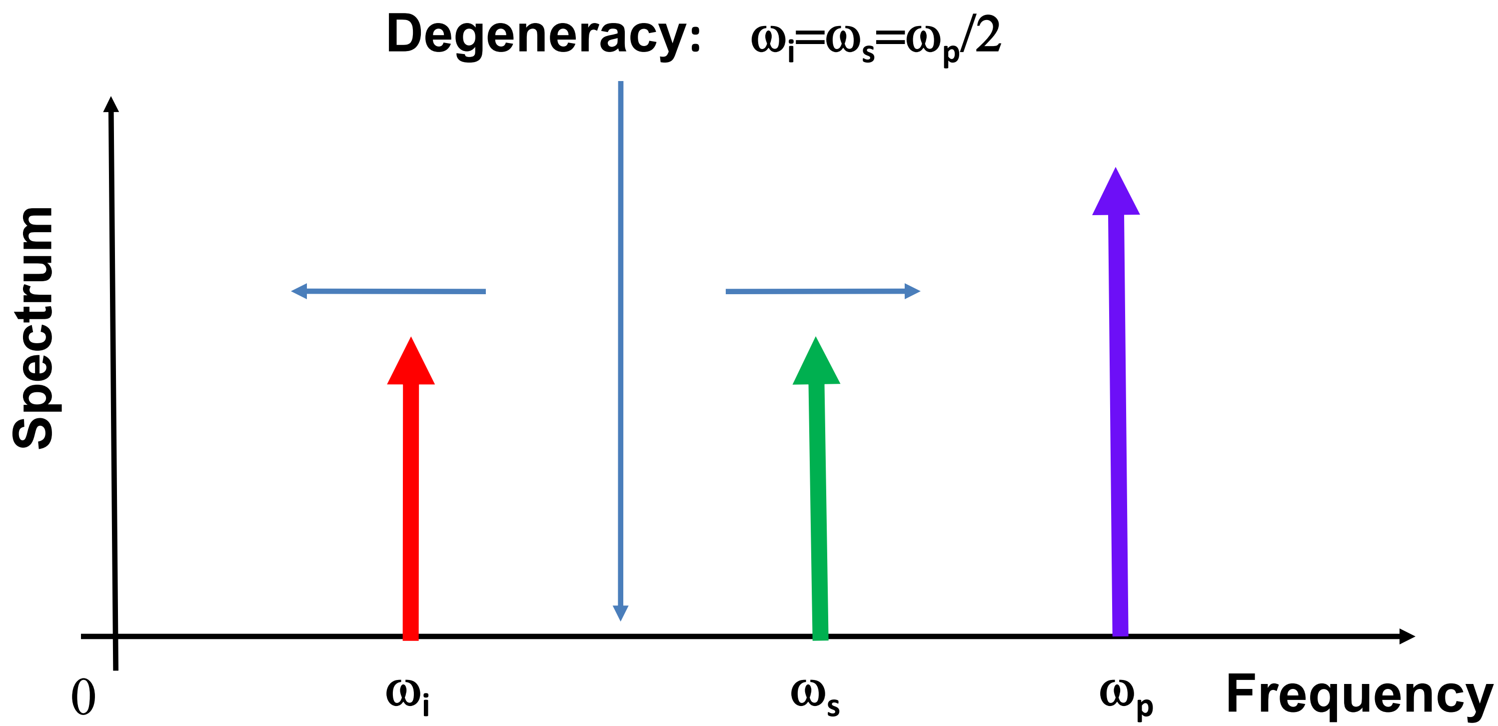
Optical Parametric
Amplification (OPA)



Optical Parametric
Oscillation (OPO)

Optical Parametric Amplifiers and Oscillators

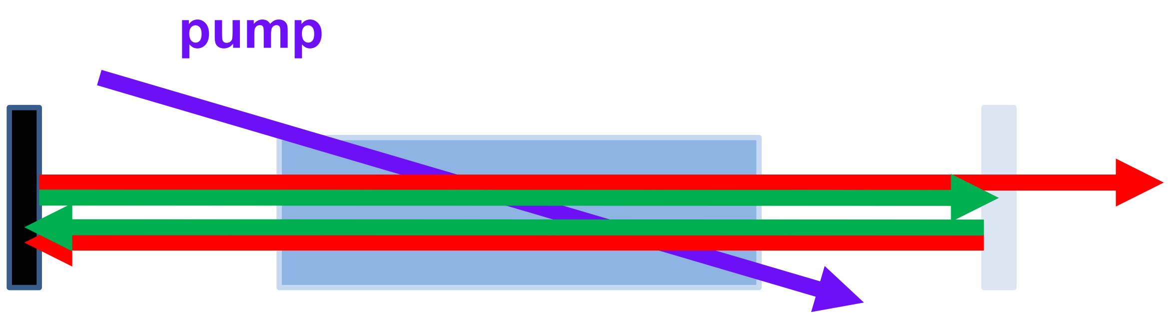
Optical Parametric Generation (OPG) or parametric fluorescence



Energy Conservation: $\hbar\omega_p = \hbar\omega_s + \hbar\omega_i.$

Momentum Conservation: $\hbar\vec{k}_p = \hbar\vec{k}_s + \hbar\vec{k}_i$

Optical Parametric Oscillator (OPO)



Double resonant: Signal and idler resonant

Single resonant: Only Signal resonant

Advantage: Widely tunable, both signal and idler can be used!

For OPO to operate, less gain is necessary in contrast to an OPA.

Nonlinear Optical Susceptibilities

Total field: Pump, signal and idler:

$$\vec{E}(\vec{r},t) = \sum_{\omega_a>0} \sum_{i=1}^{3} \frac{1}{2} \left\{ \hat{E}_i(\omega_a) e^{j(\omega_a t - \vec{k}_a \vec{r})} + c.c. \right\} \vec{e}_i.$$

Drives polarization in medium:

$$\vec{P}(\vec{r},t) = \sum \vec{P}^{(n)}(\vec{r},t)$$

$$\vec{P}^{(n)}(\vec{r},t) = \sum_{\omega_b>0} \sum_{i=1}^{3} \frac{1}{2} \left\{ P_i^{(n)}(\omega_b) e^{j(\omega_b t - \vec{k}'_b \vec{r})} + c.c. \right\} \vec{e}_i.$$

Polarization can be expanded in power series of the electric field:

$$P_i^{(n)}(\omega_b) = \frac{\varepsilon_0}{2^{m-1}} \sum_{P} \sum_{j...k} \chi_{ij...k}^{(n)}(\omega_b : \omega_1, ..., \omega_n) E_j(\omega_1) \cdots E_k(\omega_n)$$

$$\omega_b = \sum_{i=1}^{n} \omega_i \text{ and } \mathbf{k}'_b = \sum_{i=1}^{n} \mathbf{k}_i$$

Special Cases

$$\begin{aligned}\hat{P}_i^{(2)}(\omega_3) &= \varepsilon_0 \sum_{jk} \chi_{ijk}^{(2)}(\omega_3 : \omega_1, -\omega_2)\hat{E}_j(\omega_1)\hat{E}_k^*(\omega_2),\\ \omega_3 &= \omega_1 - \omega_2 \text{ und } \mathbf{k}_3' = \mathbf{k}_1 - \mathbf{k}_2.\end{aligned}$$

($\longrightarrow$ Difference Frequency Generation (DFG))

$$\begin{aligned}\hat{P}_i^{(2)}(\omega_2) &= \varepsilon_0 \sum_{jk} \chi_{ijk}^{(2)}(\omega_2 : \omega_3, -\omega_1)\hat{E}_j(\omega_3)\hat{E}_k^*(\omega_1),\\ \omega_2 &= \omega_3 - \omega_1 \text{ und } \mathbf{k}_2' = \mathbf{k}_3 - \mathbf{k}_1.\end{aligned}$$

($\longrightarrow$ Parametric Generation (OPG))

$$\begin{aligned}\hat{P}_i^{(3)}(\omega_4) &= \frac{6\varepsilon_0}{4} \sum_{jkl} \chi_{ijkl}^{(3)}(\omega_4 : \omega_1, \omega_2, -\omega_3)\hat{E}_j(\omega_1)\hat{E}_k(\omega_2)\hat{E}_l^*(\omega_3)\\ \omega_4 &= \omega_1 + \omega_2 - \omega_3 \text{ und } \mathbf{k}_4' = \mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_3.\end{aligned}$$

($\longrightarrow$ Four Wave Mixing (FWM))

Continuous Wave OPA

Wave equation (2.7) : $$\left(\Delta - \frac{1}{c_0^2}\frac{\partial^2}{\partial t^2}\right)\vec{E} = \mu_0 \frac{\partial^2}{\partial t^2}\vec{P}$$

Include linear and second order terms:

$$\left(\Delta - \frac{1}{c_0^2}\frac{\partial^2}{\partial t^2}\right)\vec{E} = \mu_0 \frac{\partial^2}{\partial t^2}\left(\vec{P}^{(l)}(\vec{r},t) + \vec{P}^{(2)}(\vec{r},t)\right)$$

Changes group and phase velocities of waves

$k(\omega)$

Nonlinear interaction of waves

z-propagation only:

$$\vec{E}_{p,s,i}(z,t) = \mathrm{Re}\left\{E_{p,s,i}(z)\; e^{j(\omega_{p,s,i}t - k_{p,s,i}\; z)}\vec{e}_{p,s,i}\right\}$$

Wave amplitudes

$$\vec{P}^{(2)}_{p,s,i}(z,t) = \mathrm{Re}\left\{ P^{(2)}_{p,s,i}(z)\ e^{j\left(\omega_{p,s,i}t - k'_{p,s,i}z\right)} \vec{e}_{p,s,i} \right\}$$

Separate into three equations for each frequency component:

Slowly varying amplitude approximation:

$$d^2_{p,s,i}E(z)\ /dz^2 << \ 2k\ dE_{p,s,i}(z)\ /dz,$$

$$\frac{\partial E_{p,s,i}(z)}{\partial z} = -\frac{jc_0^2\omega_{p,s,i}}{2n(\omega_{p,s,i})} P^{(2)}_{p,s,i}(z)\ e^{-j\left(k'_{p,s,i} - k_{p,s,i}\right)z}$$

Introduce phase mismatch: $\Delta k = k(\omega_p) - k(\omega_s) - k(\omega_i)$

and eff. nonlinearity and coupling coefficients:

$$d_{eff} = \frac{1}{2}\chi^{(2)}_{ijk}(\omega_p : \omega_s, \omega_i), \quad \kappa_{p,s,i} = \omega_{p,s,i}\ d_{eff}/(n_{p,s,i}c_0)$$

Coupled wave equations:

$$\begin{aligned} \frac{\partial E_p(z)}{\partial z} &= -j\kappa_p \, E_s(z)E_i(z) \, e^{j\Delta kz} \, , \\ \frac{\partial E_s(z)}{\partial z} &= -j\kappa_s \, E_p(z)E_i^*(z) \, e^{-j\Delta kz} , \\ \frac{\partial E_i(z)}{\partial z} &= -j\kappa_i \, E_p(z)E_s^*(z) \, e^{-j\Delta kz} . \end{aligned}$$

X $n_{p,s,i} c_0 \varepsilon_0 E^*_{p,s,i}/2$

Intensity of waves: $I_{p,s,i} = \frac{n_{p,s,i}}{2Z_{F_0}} \left| E_{p,s,i} \right|^2$

Manley-Rowe Relations: $-\frac{1}{\omega_p}\frac{dI_p}{dz} = \frac{1}{\omega_s}\frac{dI_s}{dz} = \frac{1}{\omega_i}\frac{dI_i}{dz}$