NLO Lecture 6: Phase Matching

4.3 Wave propagation in linear non-isotropic media

(repetition)

4.4 Phase matching

4.4.1 Birefringent phase matching

4.4.2 Frequency doubling of Gaussian beams

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Later:

Quasi-Phasematching in periodically poled crystals, fibers, waveguides, Bragg-structures

Reminder SHG



$$\ell_c = \pi |k(2\omega) - 2k(\omega)|^{-1} = \frac{\lambda(\omega)}{4(n(2\omega) - n(\omega))}.$$

4.3 Wave propagation in linear non-isotropic media

$$\dot{\mathbf{D}} = \varepsilon \dot{\mathbf{E}}$$

$$\varepsilon = \varepsilon_0 \begin{bmatrix} \varepsilon_x & 0 & 0 \\ 0 & \varepsilon_y & 0 \\ 0 & 0 & \varepsilon_z \end{bmatrix}$$

$$\nabla \times \nabla \times \hat{\mathbf{E}} = -\omega^2 \mu_0 \varepsilon \hat{\mathbf{E}}$$
(4.21)

Wave propagation in linear non-isotropic media

As in isotropic media, there are plane-wave solutions with

$$\hat{\mathbf{E}} = \hat{\mathbf{E}}_0 e^{-j\mathbf{k}\cdot\mathbf{r}} \tag{4.22}$$

that obey

$$\mathbf{k} \times \mathbf{k} \times \mathbf{\hat{E}} = -\omega^2 \mu_0 \varepsilon \mathbf{\hat{E}}$$
(4.23)

The wave vector is orthogonal to the displacement vector but in general not anymore to the electric field

$$\mathbf{k} \perp (\varepsilon \mathbf{\hat{E}} = \mathbf{\hat{D}}).$$

From Faraday's law we have

$$j\mathbf{k} \times \mathbf{\hat{E}} = -\omega \mathbf{\hat{B}} \tag{4.24}$$

and therefore, as in the isotropic case, we have

$$\mathbf{k} \perp \hat{\mathbf{B}} \parallel \hat{\mathbf{H}}.$$

$\hat{E} \parallel \hat{D}$: only when propagation parallel to a main axis

Poynting vector $\mathbf{S} = \mathbf{E} \times \mathbf{H}$, is always normal to \mathbf{E} and \mathbf{H}

not necessarily parallel to the wave vector



D parallel to phase fronts

E in general not parallel to phase fronts

S not necessarily parallel to **k**

Figure 4.3: Relationship between field vectors, wave vector and Poynting vector of a plane wave in birefringent media.

Form of dielectric susceptibility tensor



Table 4.1: Form of the dielectric susceptibility tensor for the different crystal systems.

In the following, we consider the uniaxial case

$$\varepsilon_{xx} = \varepsilon_{yy} = \varepsilon_1 \neq \varepsilon_{zz} = \varepsilon_3$$

The corresponding refractive indices are called ordinary and extraordinary indices.

$$n_1 = n_o \neq n_3 = n_e.$$

Further one distinguishes between positive uniaxial, $n_e > n_o$, and negative uniaxial, $n_e < n_o$, crystals.





Nonlinear optical susceptibilities

generality, we assume the wave vector lies in the x-z-plane. If we inspect Eq. (4.23) closer, we find with $\mathbf{A} \times (\mathbf{B} \times \mathbf{C}) = (\mathbf{A} \cdot \mathbf{C}) \mathbf{B} - (\mathbf{A} \cdot \mathbf{B}) \mathbf{C}$ $\mathbf{k} \times \mathbf{k} \times \hat{\mathbf{E}} = -\omega^2 \mu_0 \varepsilon \hat{\mathbf{E}}$ $(\mathbf{k} \cdot \hat{\mathbf{E}}) \mathbf{k} - k^2 \hat{\mathbf{E}} + \omega^2 \mu_0 \varepsilon \hat{\mathbf{E}} = \mathbf{0}.$ (Possible polarizations) (4.25)

$$\begin{pmatrix} k_0^2 n_o^2 + k_x^2 - k^2 & k_x k_z \\ k_0^2 n_o^2 - k^2 & k_0^2 n_e^2 - k^2 \\ k_z k_x & k_0^2 n_e^2 + k_z^2 - k^2 \end{pmatrix} \hat{\mathbf{E}} = \mathbf{0}$$
(4.26)

y-polarized wave decouples \rightarrow ordinary wave $k^2 = k_0^2 n_o^2$.

As the wave in an isotropic medium, it is purely transversal, $\mathbf{k} \perp \hat{\mathbf{E}} \perp \hat{\mathbf{H}}$.

Wave in the x-z plane with polarization in x-z plane: extraordinary wave

$$\det \begin{vmatrix} k_0^2 n_o^2 + k_x^2 - k^2 & k_x k_z \\ k_z k_x & k_0^2 n_e^2 + k_z^2 - k^2 \end{vmatrix} = 0$$

or after some brief transformations

$$\frac{k_z^2}{n_o^2} + \frac{k_x^2}{n_e^2} = k_0^2. aga{4.27}$$

With $k_x = k \sin(\theta)$, $k_z = k \cos(\theta)$ and $k = n(\theta) k_0$ we obtain for the refractive index of the extraordinary wave



Figure 4.5: Cut through the surface of the index ellipsoid with constant free-space value $k_o(k_x, k_y, k_z)$ or frequencies.

and is normal to the index ellipsoid. To determine the "walk-off" angle between the Poynting vector and the wave vector, we consider

$$\tan \theta = \frac{k_x}{k_z}$$
$$\tan \phi = -\frac{dk_z}{dk_x}.$$

From $\frac{k_z^2}{n_o^2} + \frac{k_x^2}{n_e^2} = k_0^2$ we find

$$\frac{2k_z dk_z}{n_o^2} + \frac{2k_x dk_x}{n_e^2} = 0, (4.29)$$

and

$$\tan \phi = \frac{n_o^2 k_x}{n_e^2 k_z} = \frac{n_o^2}{n_e^2} \tan \theta \,.$$

Therefore, we obtain for the walk-off angle between Poynting vector and wave number vector

$$\tan \rho = \tan \left(\theta - \phi\right) = \frac{\tan \theta - \tan \phi}{1 + \tan \theta \tan \phi}$$

$$\tan \rho = \frac{\left(1 - \frac{n_o^2}{n_e^2}\right) \tan \theta}{1 + \frac{n_0^2}{n_e^2} \tan^2 \theta}$$
(4.30)

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Intermediate Summary

- **k** not same direction as **S** ("walk-off")
- Refractive index depends on polarization and angle θ between k and optical axis (z): n₀, n_e(θ)

4.4 Phase matching

4.4.1 Birefringent phase matching

In SHG, we introduced the coherence length

$$\ell_c = \pi |k(2\omega) - 2k(\omega)|^{-1} = \frac{\lambda(\omega)}{4(n(2\omega) - n(\omega))}.$$

coherence length may be as short as a few microns, if fundamental and second harmonic have the same polarization.



Figure 4.6: Non-critical phase matching

only approximately. Often this can be further matched by temperature tuning. Important examples for this technique is the frequency doubling of $1.06-\mu m$ radiation in LiNbO₃, CD*A and LBO or frequency doubling of 530-nm light in KDP.



Figure 4.7: Type-I critical phase matching.

A more general situation is shown in Fig. 4.7. The birefringence is too strong for non-critical phase matching. However, by angle-tuning with respect to the optical axis every index value between $n_e(2\omega)$ and $n_o(2\omega)$ can be dialed in, especially $n_o(\omega)$. This phase matching angle, θ_p , is determined by

$$n_e^{2\omega}(\theta_p) = \left\{ \frac{\sin^2 \theta_p}{(n_e^{2\omega})^2} + \frac{\cos^2 \theta_p}{(n_0^{2\omega})^2} \right\}^{-1/2} = n_0^{\omega} \qquad \begin{array}{c} \text{for } \theta_p = 90 \text{deg} \\ \text{---> Non-critical phase} \\ \text{matching} \end{array}$$

which leads to

$$\tan \theta_p = \frac{n_e^{2\omega}}{n_0^{2\omega}} \sqrt{\frac{(n_0^{\omega})^2 - (n_0^{2\omega})^2}{(n_e^{2\omega})^2 - (n_0^{\omega})^2}}.$$

Walk-off angle (between fundamental and harmonic propagation direction)

$$\tan \rho = \frac{(n_0^{\omega})^2}{2} \left\{ \frac{1}{(n_e^{2\omega})^2} - \frac{1}{(n_0^{2\omega})^2} \right\} \sin 2\theta_p \approx \frac{\Delta n}{n} \sin 2\theta_p$$

only valid for small birefringence

Walk - Off



Figure 4.8: Walk-off between ordinary and extraordinary wave.

Gaussian beam with $w_0 \rightarrow walk$ -off length

$$\ell_a = \frac{\sqrt{\pi}}{\varrho} w_0.$$

Type-II phase matching



$$n_e < n_o \text{ (neg. uniaxial)}: \quad \begin{array}{l} \text{Type I} \quad \text{Type II} \\ oo \rightarrow e \quad oe \rightarrow e \\ ee \rightarrow o \quad oe \rightarrow o \end{array}$$

Table 4.2: Phase-matching configurations

Acceptance angle

$$\Delta k = (k_{2\omega} - 2k_{\omega})|_{\theta_p} + \frac{d}{d\theta} (k_{2\omega} - 2k_{\omega})|_{\theta_p} \Delta \theta + \dots$$
$$= \frac{4\pi\Delta\theta}{\lambda} \left\{ \frac{dn_{2\omega}(\theta)}{d\theta} - \frac{dn_{\omega}}{d\theta} \right\}_{\theta_p}$$

For type-I phase matching, there is $dn_{\omega}/d\theta = dn_{o}^{\omega}/d\theta = 0$ and

$$n_{2\omega}(\theta) = \left\{ \frac{\sin^2 \theta}{(n_e^{2\omega})^2} + \frac{\cos^2 \theta}{(n_0^{2\omega})^2} \right\}^{-1/2}$$

The angle-induced phase mismatch can then be rewritten as

$$\Delta k = -\frac{2\pi\Delta\theta}{\lambda} n_{2\omega}(\theta)^3 \left\{ \frac{2\sin\theta\cos\theta}{(n_e^{2\omega})^2} - \frac{2\sin\theta\cos\theta}{(n_0^{2\omega})^2} \right\}$$
$$= \frac{2\pi\Delta\theta}{\lambda} (n_o^{\omega})^3 \left\{ \frac{1}{(n_0^{2\omega})^2} - \frac{1}{(n_e^{2\omega})^2} \right\} \sin 2\theta_p.$$

For a given crystal length ℓ the phase mismatch should not be larger than the half-width at half-maximum (HWHM) of the sinc²- function, i.e., $\Delta k = \pi/\ell$,

For a given crystal length ℓ the phase mismatch should not be larger than the half-width at half-maximum (HWHM) of the sinc²- function, i.e., $\Delta k = \pi/\ell$,

$$\Delta \theta = \frac{\lambda}{2\ell \sin 2\theta_p} (n_o^{\omega})^{-3} \left\{ \frac{1}{(n_0^{2\omega})^2} - \frac{1}{(n_e^{2\omega})^2} \right\}^{-1}$$

With $\Delta n^{2\omega} = n_0^{2\omega} - n_e^{2\omega}$, $(n_0^{2\omega})^{-2} = (n_e^{2\omega})^{-2} - 2(n_e^{2\omega})^{-3}\Delta n^{2\omega}$ and $n_e^{2\omega} = n_o^{\omega}$, we obtain

$$\Delta \theta = -\frac{\lambda}{4\ell \sin 2\theta_p \Delta n^{2\omega}}.$$

For most cases $|\Delta\theta|$ is on the order of a few milliradians, e.g., for KH₂PO₄ (KDP) at $\lambda = 1.064 \ \mu m$, $n_e^{\omega} = 1.466$, $n_o^{\omega} = 1.506$, $n_e^{2\omega} = 1.487$, $n_o^{2\omega} = 1.534$. For this case, the phase-matching angle is $\theta_p = 49.9^{\circ}$ and for a 1-cm long crystal, there is $|\Delta\theta| = 0.001$.

For type-II phase matching under the condition $n_e^{2\omega}(\theta_p) = [n_e^{\omega} + n_o^{\omega}]/2$, we obtain

$$\Delta k = \frac{2\pi\Delta\theta}{\lambda} \left\{ 2\frac{dn_e^{2\omega}(\theta)}{d\theta} - \frac{dn_e^{\omega}(\theta)}{d\theta} \right\}_{\theta_p}$$
(4.32)

Weak birefringence

For weak birefringence and if the wavelength dependence of both indices is similar, than the acceptance angle is roughly twice as large as for type-I phase matching. For non-critical phase matching, that is 90°-phase matching, the above derivation can not be used, since the phase-matching error depends second order on the acceptance angle. One finds

$$\Delta k = \frac{2\pi}{\lambda} (n_o^{\omega})^3 \left\{ \frac{1}{(n_e^{2\omega})^2} - \frac{1}{(n_0^{2\omega})^2} \right\} (\Delta \theta)^2$$
(4.33)

which simplifies for small birefringence to

$$\Delta \theta \approx \left\{ \frac{\lambda}{2\ell \Delta n^{2\omega}} \right\}^{1/2} \tag{4.34}$$

For $\lambda = 1 \ \mu m$, $\Delta n = 0.047$ and $\ell = 1 \ cm$, we find $|\Delta \theta| = 0.02$, e.g., this acceptance angle is an order of magnitude higher than for cricital phase matching, which justifies the names critical and non-critical phase matching.

Acceptance bandwidth

$$\Delta k = \{k_{2\omega} - 2k_{\omega}\}|_{\lambda_p} + \left\{\frac{d}{d\lambda}(k_{2\omega} - 2k_{\omega})\right\}_{\lambda_p} \Delta \lambda + \dots \qquad (4.35)$$

$$\approx 4\pi\Delta\lambda \left\{ \frac{d}{d\lambda} \left(\frac{n_{2\omega}}{\lambda} - \frac{n_{\omega}}{\lambda} \right) \right\}_{\lambda_p} = 4\pi\frac{\Delta\lambda}{\lambda} \left\{ \frac{1}{2}\frac{dn_{2\omega}}{d(\lambda/2)} - \frac{dn_{\omega}}{d\lambda} \right\}_{\lambda_p}$$
(4.36)
$$= 4\pi\frac{\Delta\lambda}{\lambda} \left\{ \frac{1}{2}\frac{dn}{d\lambda} \Big|_{2\omega} - \frac{dn}{d\lambda} \Big|_{\omega} \right\}$$
(4.37)

The acceptance bandwidth follows again from the condition, that the phase mismatch over the propagation length must stay smaller than the HWHM of the sinc²- function, i.e., $|\Delta k| < \pi/\ell$ or

$$\Delta \lambda = \left| \frac{\lambda}{4\ell} \left\{ \frac{1}{2} \left. \frac{dn}{d\lambda} \right|_{2\omega} - \left. \frac{dn}{d\lambda} \right|_{\omega} \right\}^{-1} \right|,\tag{4.38}$$

where λ is the wavelength of the fundamental wave and ℓ the interaction length. The other way around, if a bandwidth $2\Delta\lambda$ needs to be frequency doubled, a phase matched crystal can only have the length ℓ

$$\ell = \frac{\lambda}{2\Delta\lambda} \left\{ \left. \frac{1}{2} \frac{dn}{d\lambda} \right|_{2\omega} - \left. \frac{dn}{d\lambda} \right|_{\omega} \right\}^{-1}$$
(4.39)

Acceptance bandwidth... when frequency doubling a pulse (temporal overlap)

its second harmonic. The group velocity of a pulse is given by

$$\upsilon_g = \frac{d\omega}{dk} = \frac{d}{dk} \left(\frac{c}{n}k\right) = \frac{c}{n} - \frac{ck}{n^2} \frac{dn}{d\lambda} \frac{d\lambda}{dk}$$
(4.40)

where

$$\frac{d\lambda}{dk} = \frac{d}{dk} \left(\frac{2\pi n}{k}\right) = -\left(\frac{2\pi n}{k^2}\right) + \frac{2\pi}{k} \frac{dn}{d\lambda} \frac{d\lambda}{dk}$$

$$\frac{d\lambda}{dk} = \frac{-(2\pi n/k^2)}{1 - \frac{2\pi}{k} \frac{dn}{d\lambda}},$$
(4.41)

that is

$$\upsilon_g = \frac{c}{n} \left\{ 1 - \frac{\lambda}{n} \frac{dn}{d\lambda} \right\}^{-1}.$$
(4.42)

Two pulses with duration t_p but with different group velocities will overlap over a length

$$\ell \approx \frac{t_p}{2} \left\{ \left. \frac{1}{\upsilon_g} \right|_{\omega} - \left. \frac{1}{\upsilon_g} \right|_{2\omega} \right\}^{-1}$$

Acceptance bandwidth... when frequency doubling a pulse

With Eq. (4.42) we obtain

$$\Rightarrow \ell \approx \frac{t_p c}{2\lambda} \left\{ \frac{1}{2} \left. \frac{dn}{d\lambda} \right|_{2\omega} - \left. \frac{dn}{d\lambda} \right|_{\omega} \right\}^{-1}.$$

Using the time-bandwidth relationship

$$t_p \approx \frac{1}{\Delta f} = \frac{\lambda^2}{c\Delta\lambda} \tag{4.43}$$

we find the maximum crystal length similar to the one derived from the phase matching condition (4.39)

$$\Rightarrow \ell \approx \frac{\lambda}{2\Delta\lambda} \left\{ \frac{1}{2} \left. \frac{dn}{d\lambda} \right|_{2\omega} - \left. \frac{dn}{d\lambda} \right|_{\omega} \right\}^{-1}.$$

4.4.2 Frequency doubling of Gaussian beams

A laser emits radiation in a TEM_{00} - mode, i.e., a Gaussian beam. The electric field of a Gaussian beam is described by

$$\hat{E}(x,y,z) = \hat{E}_0 \frac{w_0}{w(z)} \exp\{-j(kz-\phi)\} \times$$

$$\exp\left\{-(x^2+y^2)\left[\frac{1}{w^2(z)} + \frac{jk}{2R(z)}\right]\right\}$$
(4.44)

$$w(z) = w_0 \left\{ 1 + \left(\frac{\lambda z}{\pi w_0^2}\right)^2 \right\}^{1/2}$$
(4.45)
$$\phi = \tan^{-1} \left\{ \frac{\lambda z}{\pi w_0^2} \right\}$$
(4.46)

$$R(z) = z \left\{ 1 + \left(\frac{\pi w_0^2}{\lambda z}\right)^2 \right\}$$
(4.47)

Gaussian beam



Figure 4.10: Intensity distribution of a Gaussian beam.

The confocal parameter of the beam is twice the Rayleigh range and given by

$$b = \frac{2\pi w_0^2}{\lambda} \tag{4.48}$$

see Fig. 4.10. The Rayleigh range is the distance, over which the beam cross sectional area doubles, $\pi w^2(z) < 2\pi w_0^2$. The opening angle of the beam due to diffraction is

$$\Delta \theta \approx \frac{w(z)}{z} \approx \frac{\lambda}{\pi w_0}.$$
(4.49)

Gaussian beam continued

In the near field $(z \ll b)$, the beam is close to a plane wave

$$\hat{E}(x,y) = \hat{E}_0 \exp\left(-\frac{x^2 + y^2}{w_0^2}\right) \exp(-jkz)$$
(4.50)

or

$$\hat{E}(r) = \hat{E}_0 \exp\left(-\frac{r^2}{w_0^2}\right) \exp(-jkz)$$
(4.51)

$$P = \frac{nc\varepsilon_0}{2} \int_0^\infty \int_0^{2\pi} |\hat{E}_0|^2 \exp\left(-\frac{2r^2}{w_0^2}\right) r dr d\phi \qquad (4.52)$$
$$= \frac{nc\varepsilon_0}{2} |\hat{E}_0|^2 \left(\frac{\pi w_0^2}{2}\right) \Rightarrow P = I_0 \left(\frac{\pi w_0^2}{2}\right), \qquad (4.53)$$

with the peak intensity $I_0 = \frac{nc\varepsilon_0}{2} |\hat{E}_0|^2$ on beam axis. The effective area, A_{eff} , of a Gaussian beam is therefore

$$A_{eff} = \frac{P}{I_0} = \frac{\pi w_0^2}{2}.$$
(4.54)

Gaussian beam continued

Plane wave with radial beam profile:
$$\hat{E}(r) = \hat{E}_0 \exp\left(-\frac{r^2}{w_0^2}\right) \exp(-jkz)$$

$$\hat{E}_2(r,\ell) = -\frac{j\omega_1 d_{eff}}{n_{2\omega}c} \hat{E}_1^2(r)\ell = -j\kappa \hat{E}_1^2(r)\ell$$

where we introduced the interaction coefficient $\kappa = \frac{\omega_1 d_{eff}}{n_{2\omega}c} = \frac{\omega_2 d_{eff}}{2n_{2\omega}c}$. Gaussian shape for the second harmonic

$$\hat{E}_2(r,\ell) = -j\kappa \hat{E}_1^2 \ell \mathrm{e}^{-2r^2/w_1^2}.$$

The frequency-doubled beam shows only half the cross section compared to the fundamental beam $w_2 = w_1/\sqrt{2}$ or the confocal parameter $b_2 = \pi w_2^2/(\lambda/2) = \pi w_1^2/\lambda = b_1$. Thus the confocal parameters of both beams are the same. The total generated power at 2ω is

$$P_{2} = \frac{n_{2\omega}c\varepsilon_{0}}{2} \int_{0}^{2\pi} \int_{0}^{\infty} |\hat{E}_{2}(r)|^{2} r dr d\phi = \frac{n_{2\omega}c\varepsilon_{0}}{2} \kappa^{2} \hat{E}_{1}^{4} \ell^{2} \left(\frac{\pi w_{1}^{2}}{4}\right)$$
(4.57)
$$= \frac{n_{2\omega}}{n_{\omega}} P_{1} \kappa^{2} \hat{E}_{1}^{2} \ell^{2} / 2$$
(4.58)

Estimate of conversion efficiency for Gaussian beam

similar to the case of plane waves. From Eq. (4.59) we obtain for the conversion efficiency

$$\eta = \frac{P_2}{P_1} = \frac{2\omega^2}{\varepsilon_0 c^3} \left(\frac{d_{eff}^2}{n^3}\right) \left(\frac{P_1}{\pi w_1^2}\right) \cdot \ell^2.$$
(4.61)

Thus the conversion efficiency is proportional to (d_{eff}^2/n^3) . Thus for choosing a crystal for efficient frequency doubling, not only the effective nonlinearity d_{eff} should be as high as possible, but simultaneously, the refractive index nshould be small. Fig. 4.11 gives an overview over the figure of merit defined by FOM= d_{eff}^2/n^3 . From Fig. 4.10 we see that for $\ell > b$ the beam cross section increases and the conversion drops. A numerical optimization without any approximations results in the crystal length $\ell = 2.84 \cdot b$ for maximum conversion. With this result and $b = 2\pi w_1^2/\lambda$, we obtain for the maximum conversion efficiency

$$\eta_{opt} = \frac{P_2}{P_1} = \frac{2\omega^2}{\varepsilon_0 \lambda c^3} \left(\frac{d_{eff}^2}{n^3}\right) 5.68P_1 \cdot \ell.$$
(4.62)

The weaker the focus and the longer the crystal, the larger is the conversion in a $\chi^{(2)}$ -process, if phase matching is maintained over the full length.



Figure 4.11: Figure of merit (FOM) for different nonlinear optical materials.