Nonlinear Optics (WiSe 2018/19)

Lecture 3: November 2, 2018

4 Frequency doubling

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4.2 With depletion of fundamental wave

4.3 Wave propagation in linear non-isotropic media

4.3.1 Ordinary wave

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4.4 Phase matching

4.4.1 Birefringent phase matching

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4.4.3 Frequency doubling of pulses

4.4.4 Effective nonlinear coefficient d_{eff}

4.4.5 Quasi-phase matching (QPM)

Repetition: Nonlinear Wave Equation

Employing the dispersion relation $k^2 = \mu_0 \varepsilon_0 \varepsilon_r \omega^2$ the two leading terms cancel, and within the SVEA (3.6),(3.7) it follows

$$2jk\frac{\partial}{\partial z}E + 2j\frac{\omega}{c^2}\frac{\partial}{\partial t}E = -j\omega\mu_0\sigma E + \mu_0\omega^2 P_{NL}\left(\hat{\mathbf{e}}\cdot\hat{\mathbf{p}}\right)e^{j(k-k_p)z},$$

where we introduced the velocity of light in the linear medium as $c = \sqrt{\mu_0\varepsilon_0\varepsilon_r}^{-1}$. We divide this equation by $2jk$ and transform it into a comoving time frame using $t' = t - z/c$, $(z = z')$, and obtain

$$\frac{\partial}{\partial z}E(z,t') = -\alpha E(z,t') - \frac{1}{2}j\omega Z_\omega P_{NL}(z,t')\left(\hat{\mathbf{e}}\cdot\hat{\mathbf{p}}\right)e^{j(k-k_p)z}, \qquad (3.8)$$

with the damping constant $\alpha = \sigma Z_\omega/2$ and the impedance of the medium $Z_\omega = \frac{1}{\varepsilon_0\sqrt{\varepsilon_{r,\omega}}c_0} = \sqrt{\frac{\mu_0}{\varepsilon_0\varepsilon_{r,\omega}}}$.

Some remarks on

$$\frac{\partial}{\partial z}E(z,t') = -\alpha E(z,t') - \frac{1}{2} j\omega Z_\omega P_{NL}(z,t')\,(\hat{\mathbf{e}}\cdot\hat{\mathbf{p}})\,e^{j(k-k_p)z}, \qquad (3.8)$$

•The medium **conductivity σ** leads to **losses** and therefore **damping** of the propagating wave.

•The medium’s nonlinear polarization can lead to both **gain or damping**, depending on the **relative phase** between the electric field and the polarization (parametric amplification, frequency conversion, stimulated scattering processes as Raman and Brillouin scattering, multi-photon absorption).

•If the nonlinear polarization is **in phase or in opposite phase** of the electric field, it corresponds to a a nonlinear change of the refractive index, leading to a **phase shift of the electric field** (Pockels effect, Kerr effect).

•If the polarization is advancing the field by 90°, the polarization is supplying energy to the field. In the opposite case, the polarization is extracting energy from the field.

•**phase relation is changing during propagation**, if no phase matching of the process, i.e., $k = k_p$, is achieved.

4. Frequency doubling

VOLUME 7, NUMBER 4 PHYSICAL REVIEW LETTERS AUGUST 15, 1961

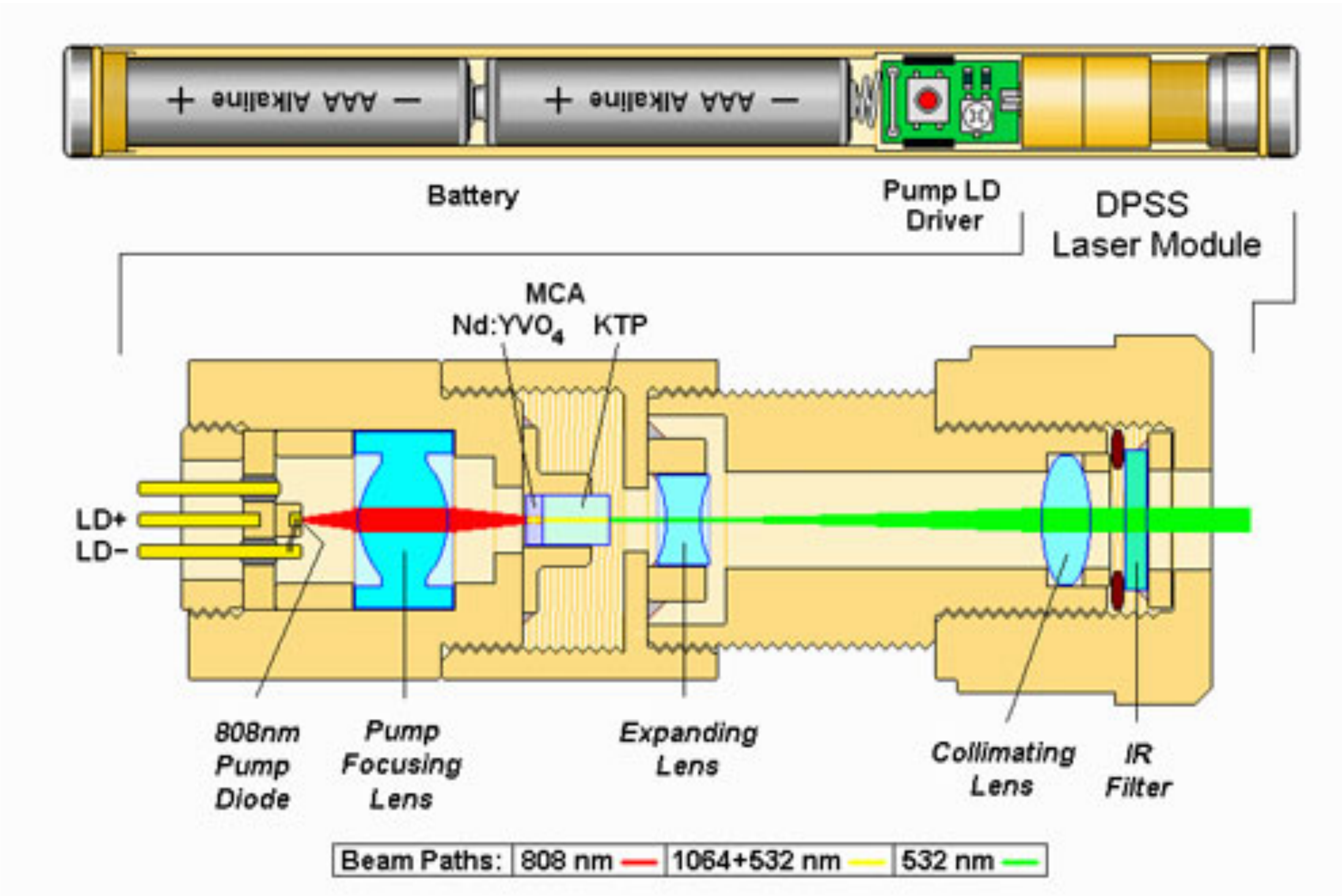
GENERATION OF OPTICAL HARMONICS*

P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich
The Harrison M. Randall Laboratory of Physics, The University of Michigan, Ann Arbor, Michigan
(Received July 21, 1961)

FIG. 1. A direct reproduction of the first plate in which there was an indication of second harmonic. The wavelength scale is in units of 100 A. The arrow at 3472 A indicates the small but dense image produced by the second harmonic. The image of the primary beam at 6943 A is very large due to halation.

The very weak spot due to the second harmonic is missing. It was removed by an overzealous Physical Review Letters editor, who thought it was a speck of dirt and didn't ask the authors anymore.

SHG in daily life: green laser pointer



Second harmonic generation (SHG)

$$\hat{P}(2\omega) = \varepsilon_0 d_{eff}(2\omega;\omega,\omega)\hat{E}(\omega,z)\hat{E}(\omega,z). \tag{4.1}$$

We neglect any losses for the moment ($\alpha = 0$), and $Z_\omega = \frac{1}{n_\omega}\sqrt{\frac{\mu_0}{\varepsilon_0}} = \frac{1}{n_\omega}\frac{1}{\varepsilon_0 c_0}$ from Eq..(3.8)

$$\frac{\partial \hat{E}(2\omega)}{\partial z} = -\frac{j\omega}{n_{2\omega} c_0} d_{eff}(2\omega;\omega,\omega)\hat{E}(\omega,z)\hat{E}(\omega,z)\mathrm{e}^{j(k(2\omega)-2k(\omega))z} \tag{4.2}$$

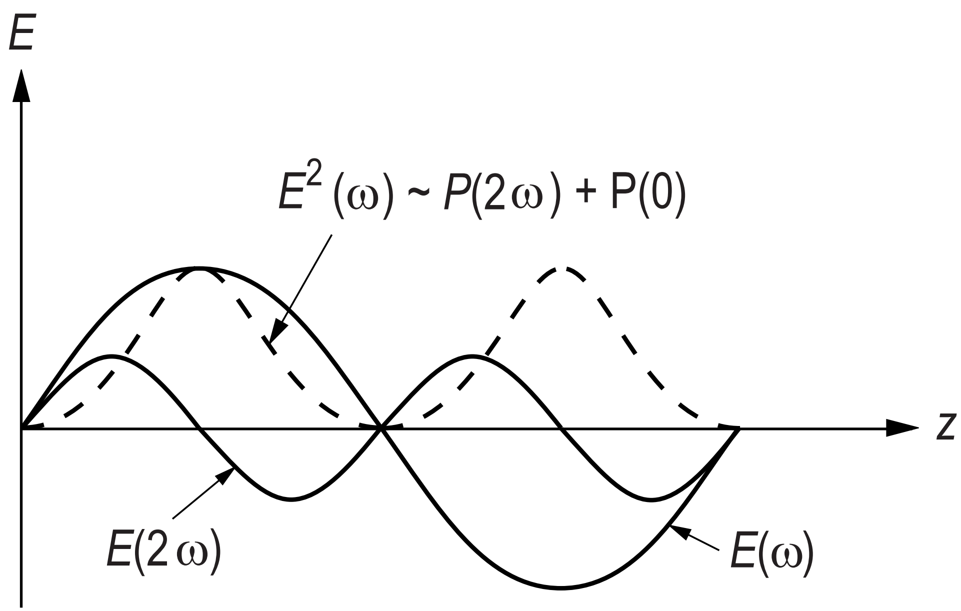


Fig. 1: Phase relationships between fundamental, second harmonic and nonlinear polarization.

4.1 Without depletion of fundamental wave

$$\hat{E}(2\omega, z=\ell) = -\frac{j\omega d_{eff}}{n_{2\omega} c_0} \hat{E}^2(\omega) \int_0^\ell \mathrm{e}^{j\Delta k z} dz$$

where $\Delta k = k(2\omega) - 2k(\omega)$ is the difference in wave number between the second harmonic light and twice the wavenumber of the fundamental light or the driving second order nonlinear Polarization.

Second-harmonic generation (SHG)

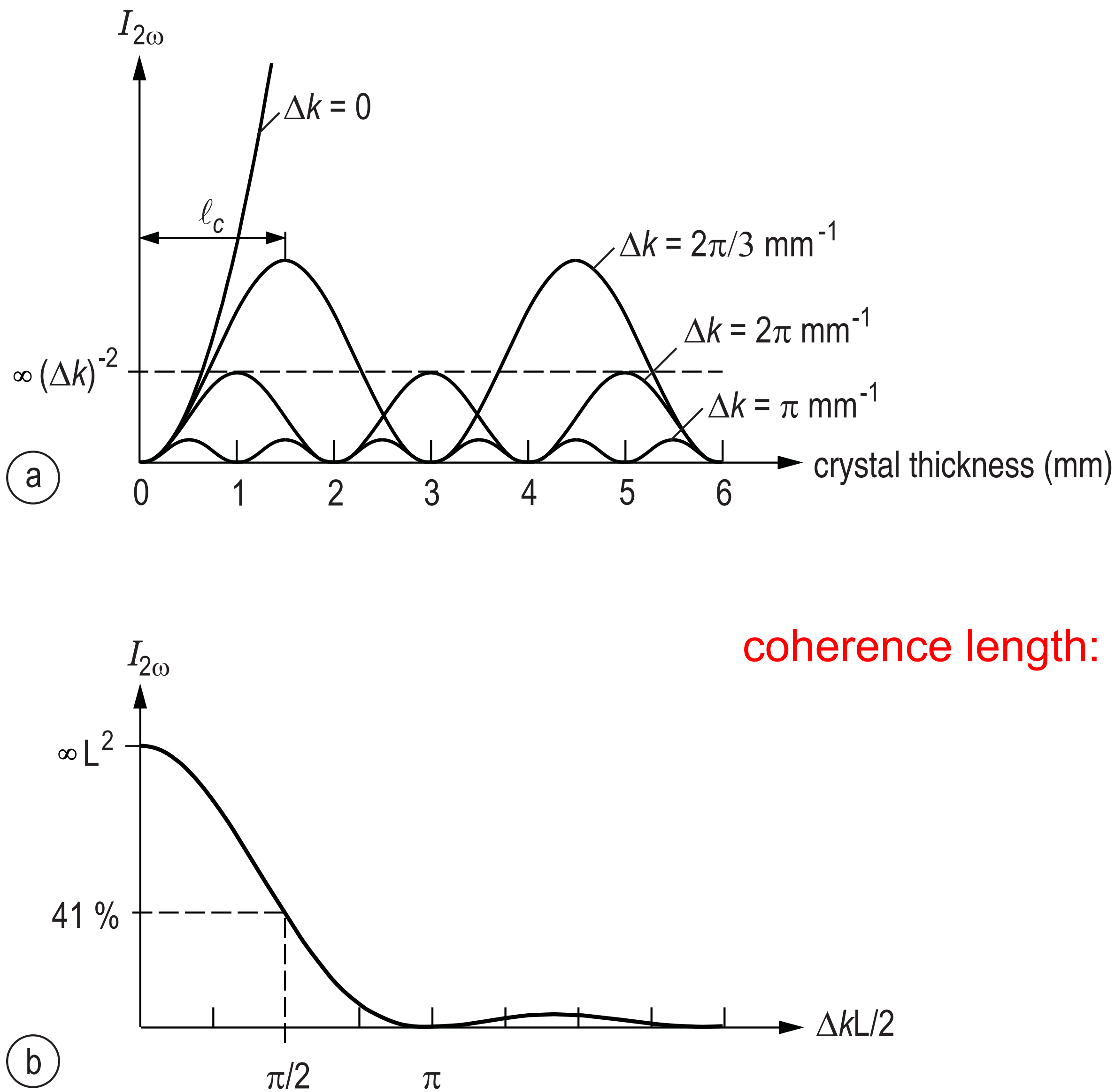
$$\hat{E}(2\omega, \ell) = -\frac{j\omega d_{eff}}{n_{2\omega} c_0} \hat{E}^2(\omega)\ell \cdot \left[\frac{\sin \Delta k\ell/2}{\Delta k\ell/2}\right] \mathrm{e}^{j\Delta k\ell/2}. \qquad (4.3)$$

Introducing the intensities of the fundamental and second harmonic waves

$$I_{\omega,2\omega} = \frac{n_{\omega,2\omega}}{2}\sqrt{\varepsilon_0/\mu_0}|\hat{E}_{\omega,2\omega}|^2$$

wie obtain

$$I(2\omega, \ell) = \frac{2\omega^2 d_{eff}^2}{n_{2\omega} n_\omega^2 c_0^3 \varepsilon_0} \ell^2 I^2(\omega) \left[\frac{\sin \Delta k\ell/2}{\Delta k\ell/2}\right]^2. \qquad (4.4)$$



coherence length: $\ell_c = \dfrac{\pi}{\Delta k}$

Figure 4.2: Second-harmonic generation as function of phase mismatch.

If phase matching can be achieved, one can use Eq. (4.4) to define an inverse conversion length Γ as

$$\Gamma = \frac{\omega d_{eff}}{nc} |\hat{E}(\omega)|, \text{ with } n = \sqrt{n_\omega n_{2\omega}}, \tag{4.6}$$

and

$$I(2\omega, \ell) = \Gamma^2 \ell^2 I(\omega). \tag{4.7}$$

If the medium length reaches the conversion length, i.e., $\Gamma\ell = 1$, then Eq. (4.7) would indicate, that all fundamental light is converted to the second harmonic, which contradicts the assumption of small conversion, and therefore we have to work a little more to correct for it.

4.2 With depletion of the fundamental wave

$$\hat{P}(\omega) = \varepsilon_0 d'_{eff}(\omega; 2\omega, -\omega)\hat{E}(2\omega)\hat{E}^*(\omega).$$

The coupled equations are

$$\frac{\partial \hat{E}(2\omega)}{\partial z} = -\frac{j\omega}{n_{2\omega}c_0} d_{eff}\hat{E}(\omega)\hat{E}(\omega)\mathrm{e}^{j\Delta kz} \tag{4.8}$$

and

$$\frac{\partial \hat{E}(\omega)}{\partial z} = -\frac{j\omega}{n_{\omega}c_0} d'_{eff}\hat{E}(2\omega)\hat{E}^*(\omega)\mathrm{e}^{-j\Delta kz}. \tag{4.9}$$

Both equations describe the energy exchange between fundamental and second-harmonic wave. The intensities are

$$I_\omega = \frac{n_\omega}{2Z_0}\left|\hat{E}(\omega)\right|^2 \text{ and } I_{2\omega} = \frac{n_{2\omega}}{2Z_0}\left|\hat{E}(2\omega)\right|^2 \tag{4.10}$$

lossless media, i.e., d'_{eff} and d_{eff} are real.

$$
\begin{aligned}
2Z_0 \frac{dI_{2\omega}}{dz} &= n_{2\omega}\left[\hat{E}^*(2\omega)\frac{\partial \hat{E}(2\omega)}{\partial z} + c.c.\right] = \\
&= -\frac{j\omega}{c_0} d_{eff} \hat{E}^*(2\omega)\hat{E}(\omega)\hat{E}(\omega)\mathrm{e}^{j\Delta k z} + c.c. \\
2Z_0 \frac{dI_{\omega}}{dz} &= n_{\omega}\left[\hat{E}(\omega)\frac{\partial \hat{E}^*(\omega)}{\partial z} + c.c.\right] = -2Z_0 \frac{dI_{2\omega}}{dz}, \text{ if } d'_{eff} = d^*_{eff}.
\end{aligned}
$$

Energy conservation demands permutation symmetry of the conversion coefficients

$$
n_{2\omega}|\hat{E}(2\omega)|^2 + n_{\omega}|\hat{E}(\omega)|^2 = \text{ const.} \equiv n_{\omega}\hat{E}_0^2 = \text{const.} \tag{4.11}
$$

Separating the wave amplitudes with respect to amplitude and phase

$$\hat{E}(\omega) = |\hat{E}(\omega)|\mathrm{e}^{j\Phi(\omega)}$$

$$\hat{E}(2\omega) = |\hat{E}(2\omega)|\mathrm{e}^{j\Phi(2\omega)}$$

$$\frac{d\hat{E}(2\omega)}{dz} = \frac{d|\hat{E}(2\omega)|}{dz}\mathrm{e}^{j\Phi(2\omega)} + j\frac{d\Phi(2\omega)}{dz}|\hat{E}(2\omega)|\mathrm{e}^{j\Phi(2\omega)} \tag{4.12}$$

$$\frac{d|\hat{E}(2\omega)|}{dz} = \mathrm{Re}\left\{-\frac{j\omega d_{eff}}{n_{2\omega}c_0}|\hat{E}(\omega)|^2\mathrm{e}^{2j\Phi(\omega)-j\Phi(2\omega)}\mathrm{e}^{j\Delta kz}\right\} \tag{4.13}$$

$$= \mathrm{Re}\left\{-\frac{j\omega d_{eff}}{n_{\omega}c_0}\{\hat{E}_0^2 - |\hat{E}(2\omega)|^2\}\mathrm{e}^{2j\Phi(\omega)-j\Phi(2\omega)}\mathrm{e}^{j\Delta kz}\right\}. \tag{4.14}$$

General solution: Jacobi elliptic function!

For Δk=0, second harmonic builds up such that

$$-j\mathrm{e}^{2j\Phi(\omega)-j\Phi(2\omega)} = 1.$$

Solution for Δk=0

$$\int_0^{|\hat{E}(2\omega)|_\ell} \frac{d|\hat{E}(2\omega)|}{\hat{E}_0^2 - |\hat{E}(2\omega)|^2} = -\int_0^\ell \frac{\omega d_{eff}}{n_\omega c_0} dz. \tag{4.15}$$

Using the integral

$$\int \frac{dx}{a^2 - x^2} = \frac{1}{a} \tanh^{-1}[x/a] \tag{4.16}$$

we obtain

$$|\hat{E}(2\omega)|_{z=\ell} = \hat{E}_0 \tanh\left\{\hat{E}_0 \left(\frac{\omega d_{eff}}{n_\omega c_0}\right) \ell\right\} \tag{4.17}$$

or for the intensity

$$I(2\omega, \ell) = I(\omega, 0) \tanh^2\left\{\frac{\hat{E}_0 \omega d_{eff}}{n_\omega c_0} \cdot \ell\right\} \tag{4.18}$$

With the conversion rate $\Gamma = \frac{\omega d_{eff}}{n_\omega c_0} \hat{E}_0$ introduced above, we obtain

$$I(2\omega, \ell) = I(\omega, 0) \tanh^2\{\Gamma\ell\} \tag{4.19}$$

With $1 - \tanh^2 = \cosh^{-2} = \mathrm{sech}^2$

$$I(\omega, \ell) = I(\omega, 0)\mathrm{sech}^2\{\Gamma\ell\}. \tag{4.20}$$

For perfect phase matching, 100% conversion possible for $\Gamma\ell$ >> 1

What to do if there is phase mismatch?

4.3 Wave propagation in linear non-isotropic media

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

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

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

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

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

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

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

From Faraday's law we have

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

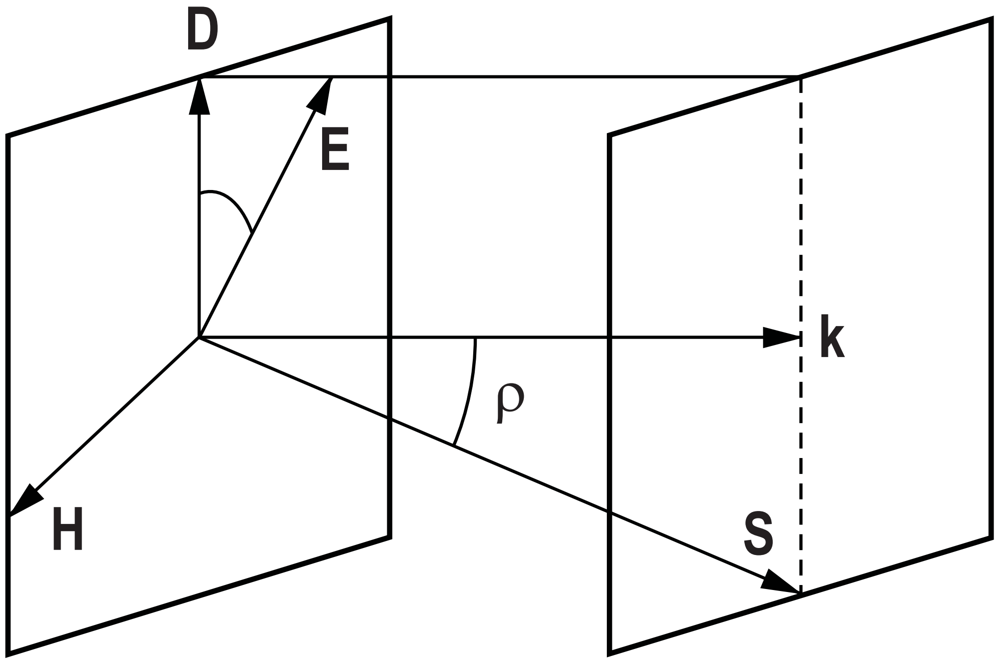
and therefore, as in the isotropic case, we have

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

$\hat{\mathbf{E}} \parallel \hat{\mathbf{D}}$: only when 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

isotropic	$\begin{bmatrix} xx & 0 & 0 \\ 0 & xx & 0 \\ 0 & 0 & xx \end{bmatrix}$	cubic
uniaxial	$\begin{bmatrix} xx & 0 & 0 \\ 0 & xx & 0 \\ 0 & 0 & zz \end{bmatrix}$	tetragonal trigonal hexagonal
biaxial	$\begin{bmatrix} xx & 0 & 0 \\ 0 & yy & 0 \\ 0 & 0 & zz \end{bmatrix}$	orthorhombic
	$\begin{bmatrix} xx & 0 & xz \\ 0 & yy & 0 \\ xz & 0 & zz \end{bmatrix}$	monoclinic
	$\begin{bmatrix} xx & xy & xz \\ xy & yy & yz \\ xz & yz & zz \end{bmatrix}$	triclinic

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 negativ uniaxial, $n_e < n_o$, crystals.

Propagation different from main axes

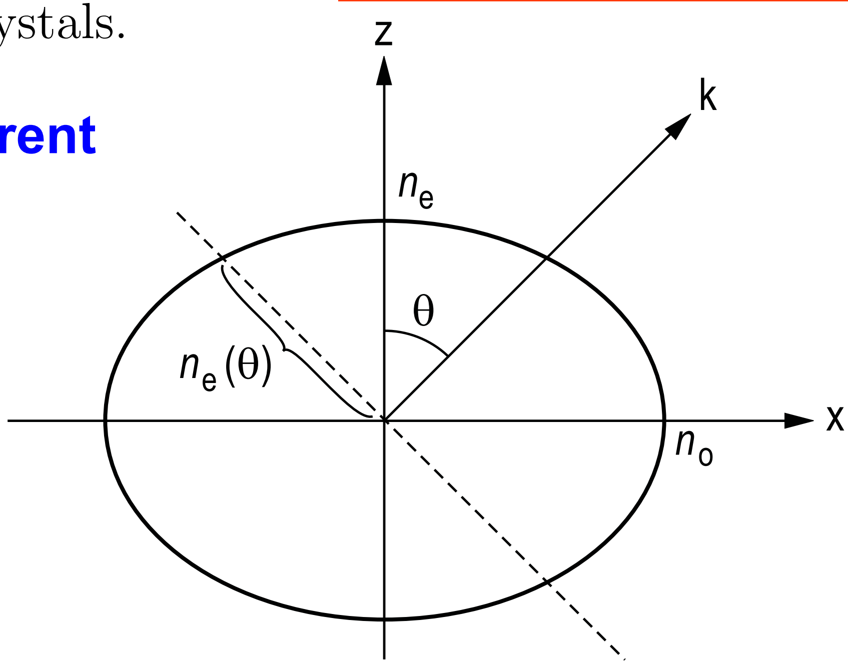


Figure 4.4: Index ellipsoid

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 $\mathbf{A} \times (\mathbf{B} \times \mathbf{C}) = (\mathbf{A} \cdot \mathbf{C})\,\mathbf{B} - (\mathbf{A} \cdot \mathbf{B})\,\mathbf{C}$

$$\left(\mathbf{k} \cdot \hat{\mathbf{E}}\right)\mathbf{k} - k^2\hat{\mathbf{E}} + \omega^2\mu_0\varepsilon\hat{\mathbf{E}} = \mathbf{0}. \tag{4.25}$$

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

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

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

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

or after some brief transformations

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

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

$$\frac{1}{n(\theta)^2} = \frac{\cos^2(\theta)}{n_o^2} + \frac{\sin^2(\theta)}{n_e^2}. \tag{4.28}$$

$$v_g = \nabla_k \omega(\mathbf{k}) \parallel \mathbf{S},$$

normal to index ellipsoid and parallel to Poynting vector

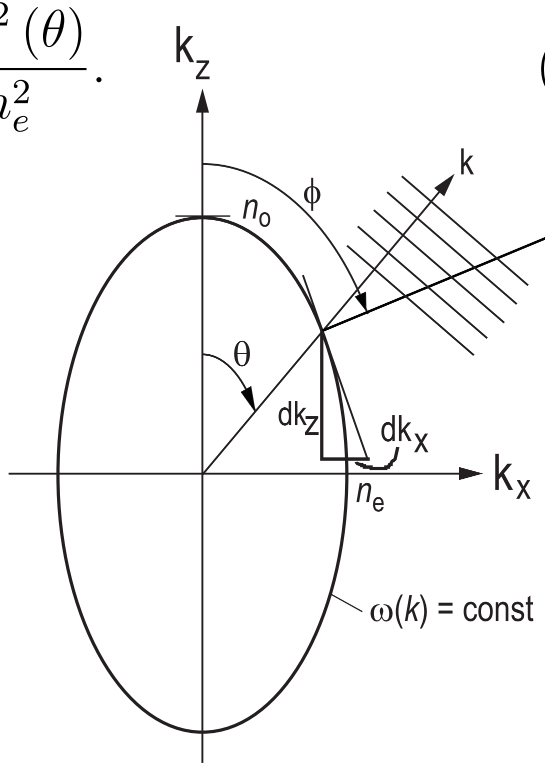


Figure 4.5: Cut through the surface of the index ellipsoid with constant free-space value $k_o(k_x, k_y, k_z)$ or frequencies.

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 Eq. (4.27) we find

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

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

$$\tan\varrho = \tan(\theta - \phi) = \frac{\tan\theta - \tan\phi}{1 + \tan\theta\tan\phi} \tag{4.30}$$

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

4.4 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.

$$n_e(2\omega) = n_o(\omega)$$

non-critical phase matching (for neg. birefringence)

similar for pos. birefringence

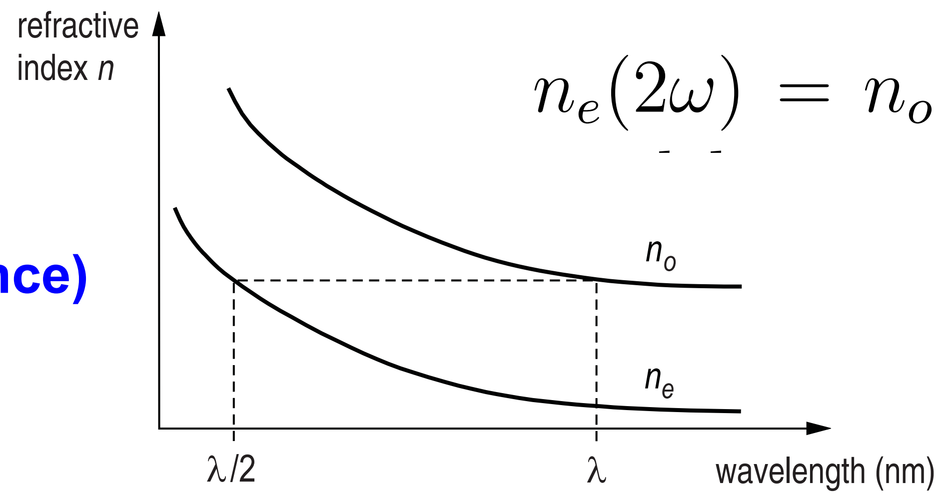


Figure 4.6: Non-critical phase matching

only approximately. Often this can be further matched by temperature tuning. Important examples for this technique is the frequency doubling of 1.06-μm radiation in $LiNbO_3$, CD*A and LBO or frequency doubling of 530-nm light in KDP.

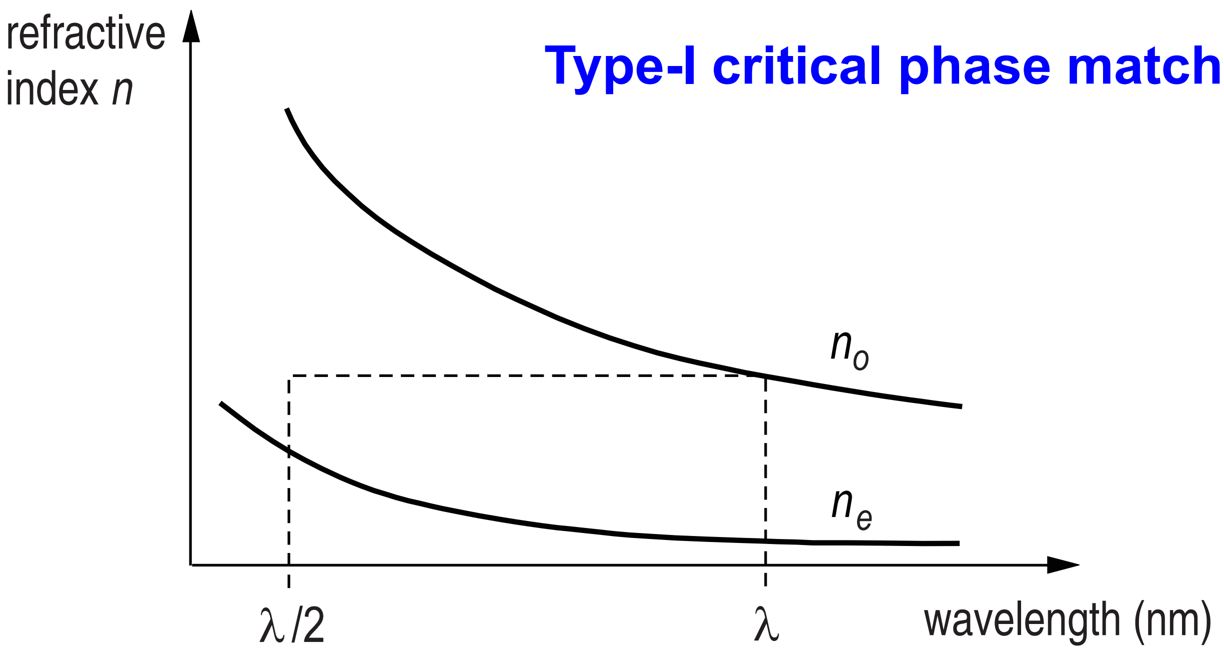


Figure 4.7: Type-I critical phase matching.

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

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

which leads to

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

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

only valid for small birefringence

Gaussian beam with w_0 → walk-off length $$\ell_a = \frac{\sqrt{\pi}}{\varrho} w_0.$$

Walk - Off

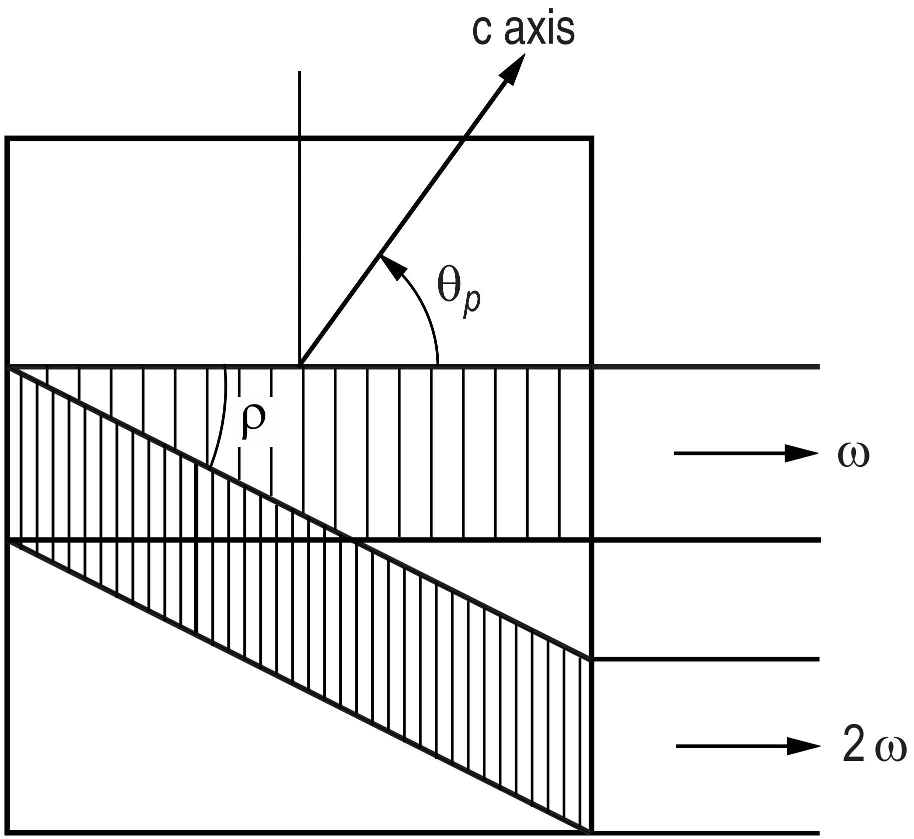
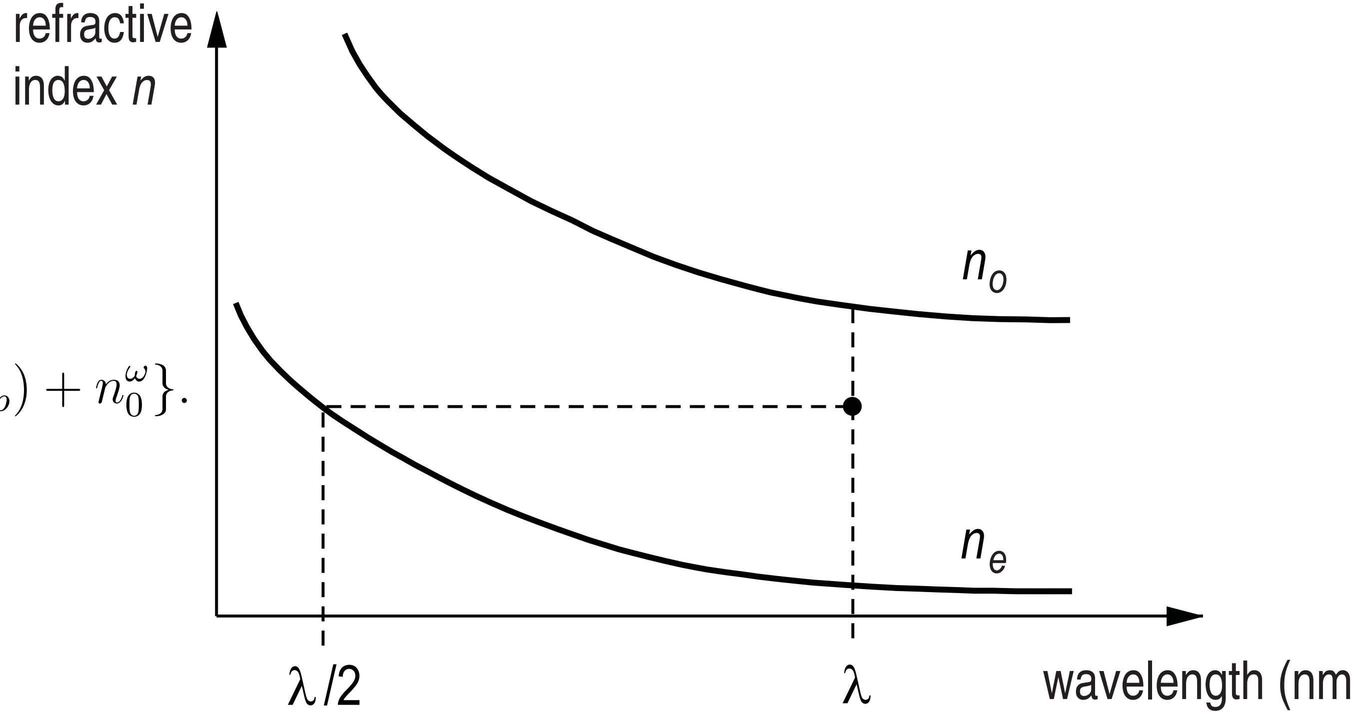


Figure 4.8: Walk-off between ordinary and extraordinary wave.

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

Type-II phase matching



$$n_e^{2\omega}(\theta_p) = \frac{1}{2}\{n_e^{\omega}(\theta_p) + n_0^{\omega}\}.$$

Figure 4.9: Type-II non-critical phase matching.

	Type I	Type II
$n_e < n_o$ (neg. uniaxial) :	$oo \to e$	$oe \to e$
$n_e > n_o$ (pos. uniaxial) :	$ee \to o$	$oe \to o$

Table 4.2: Phase-matching configurations

Acceptance angle

$$\begin{aligned}
\Delta k &= (k_{2\omega} - 2k_\omega)|_{\theta_p} + \frac{d}{d\theta}(k_{2\omega} - 2k_\omega)\Big|_{\theta_p} \Delta\theta + \ldots \\
&= \frac{4\pi\Delta\theta}{\lambda}\left\{\frac{dn_{2\omega}(\theta)}{d\theta} - \frac{dn_\omega}{d\theta}\right\}_{\theta_p}
\end{aligned}$$

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

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

For a given crystal length ℓ the phase mismatch should not be larger than the half-width at half-maximum (HWHM) of the sinc$^2-$ 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 sinc$^2-$ function, i.e., $\Delta k = \pi/\ell$,

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

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 KH_2PO_4 (KDP) at $\lambda = 1.064$ μm , $n_e^\omega = 1.466$, $n_o^\omega = 1.506$, $n_e^{2\omega} = 1.487$, $n_o^{2\omega} = 1.534$. For this case, the phase-matching angle is $\theta_p = 49.9°$ and for a 1-cm long crystal, there is $|\Delta\theta| = 0.001$.

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

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

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

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

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 μm, $\Delta n = 0.047$ 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

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

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

$$= 4\pi\frac{\Delta\lambda}{\lambda}\left\{\frac{1}{2}\frac{dn}{d\lambda}\bigg|_{2\omega} - \frac{dn}{d\lambda}\bigg|_{\omega}\right\} \tag{4.37}$$

The acceptance bandwidth follows again from the condition, that the phase mismatch over the propagation length must stay smaller than the HWHM of the sinc^2- function, i.e., $|\Delta k| < \pi/\ell$ or

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

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

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

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

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

where

$$\begin{aligned} \frac{d\lambda}{dk} &= \frac{d}{dk}\left(\frac{2\pi n}{k}\right) = -\left(\frac{2\pi n}{k^2}\right) + \frac{2\pi}{k}\frac{dn}{d\lambda}\frac{d\lambda}{dk} \\ \frac{d\lambda}{dk} &= \frac{-\left(2\pi n/k^2\right)}{1 - \frac{2\pi}{k}\frac{dn}{d\lambda}}, \end{aligned} \tag{4.41}$$

that is

$$v_g = \frac{c}{n}\left\{1 - \frac{\lambda}{n}\frac{dn}{d\lambda}\right\}^{-1}. \tag{4.42}$$

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

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

Acceptance bandwidth

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

$$\begin{aligned} \hat{E}(x,y,z) &= \hat{E}_0 \frac{w_0}{w(z)} \exp\{-j(kz-\phi)\} \times \\ & \exp\left\{-(x^2+y^2)\left[\frac{1}{w^2(z)}+\frac{jk}{2R(z)}\right]\right\} \end{aligned} \tag{4.44}$$

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

$$\phi = \tan^{-1}\left\{\frac{\lambda z}{\pi w_0^2}\right\} \tag{4.46}$$

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

Gaussian beam

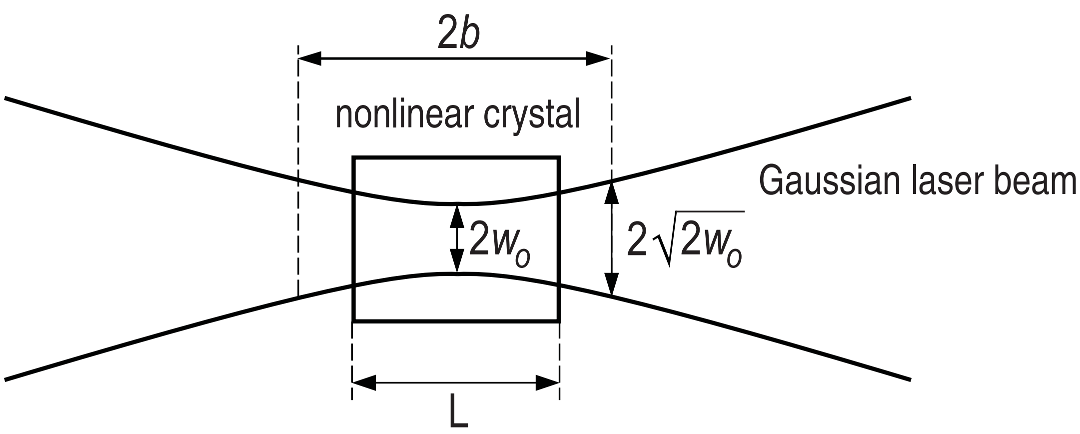


Figure 4.10: Intensity distribution of a Gaussian beam.

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

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

Gaussian beam continued

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

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

or

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

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

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

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

Estimate of conversion efficiency for Gaussian beam

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

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

Thus the conversion efficiency is proportional to $\left(d_{eff}^2/n^3\right)$. 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 n should 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

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

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.

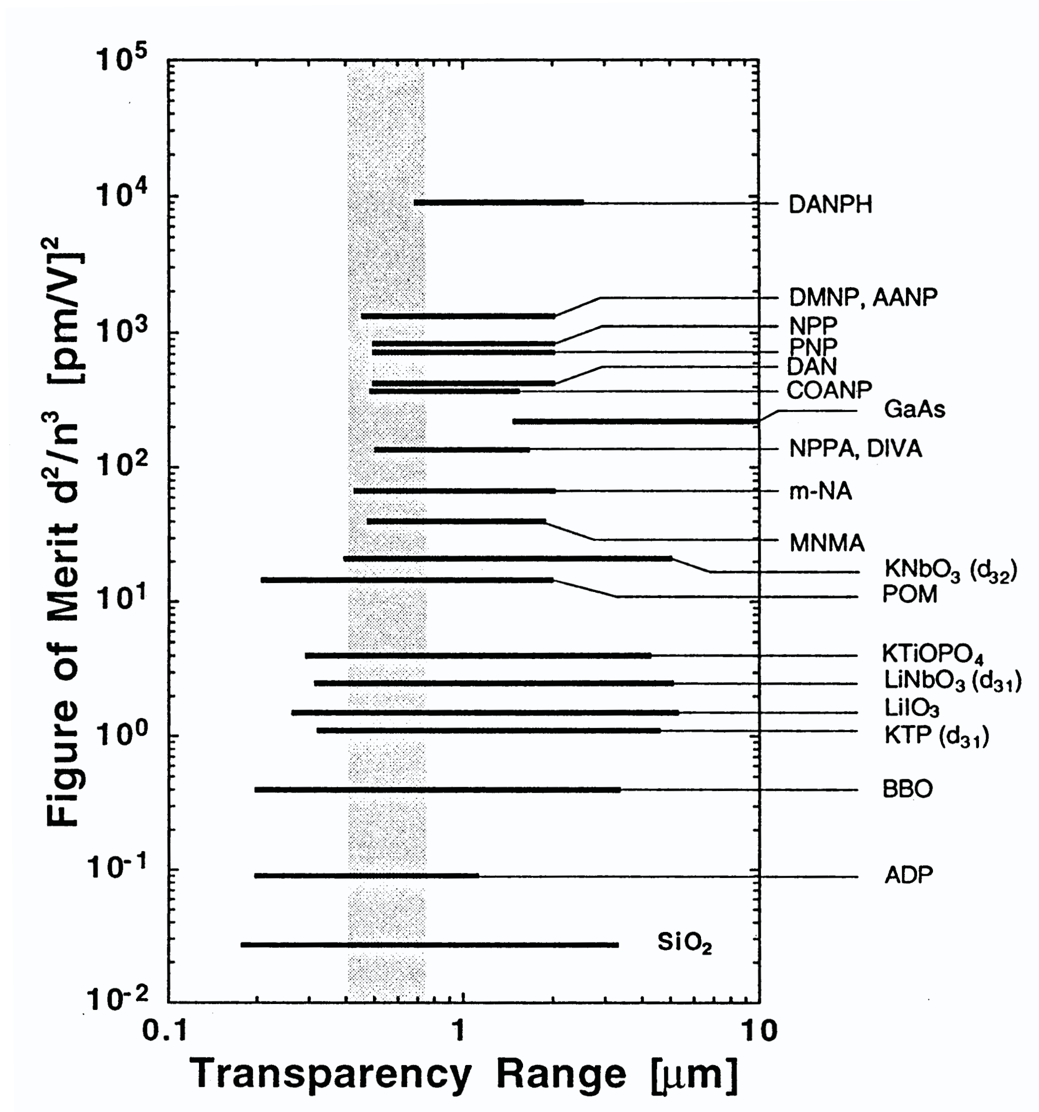


Figure 4.11: Figure of merit (FOM) for different nonlinear optical materials.