Nonlinear Optics (WiSe 2017/18)

Lecture 5: November 2, 2016

4 Frequency doubling

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4.4 Phase matching

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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

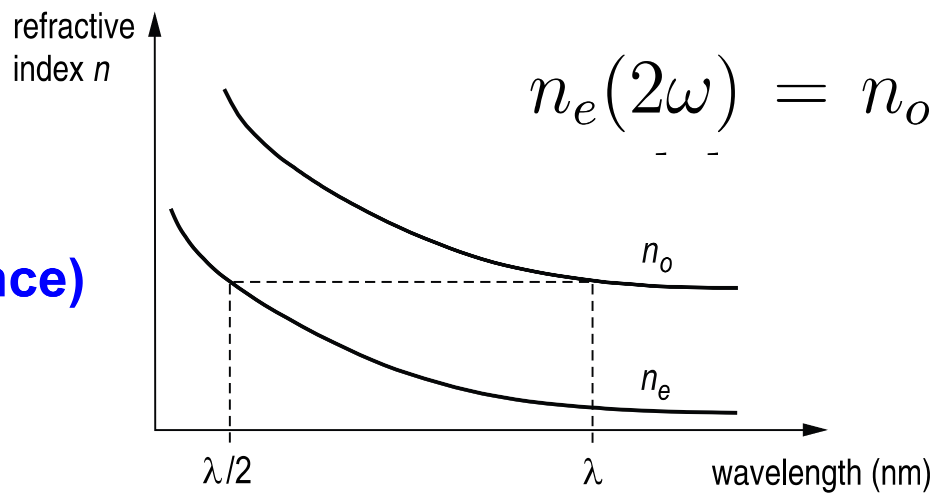


Figure 4.6: Non-critical phase matching

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.

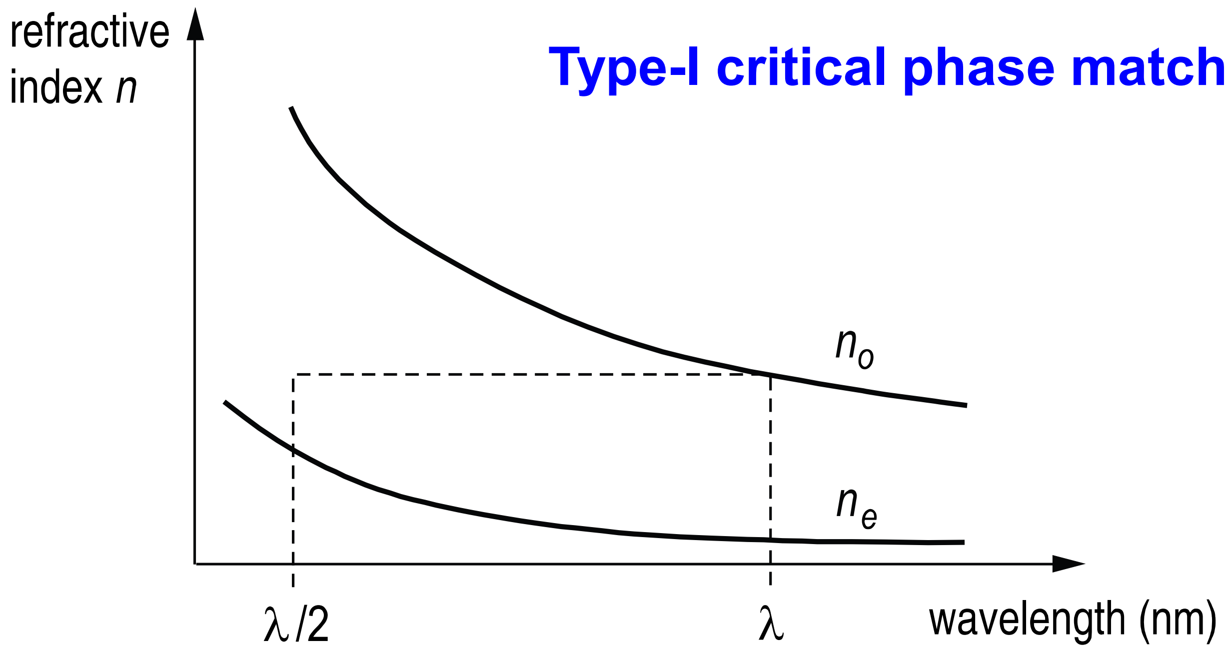


Figure 4.7: Type-I critical phase matching.

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

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

walk-off angle

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

only valid for small birefringence

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

Walk-off

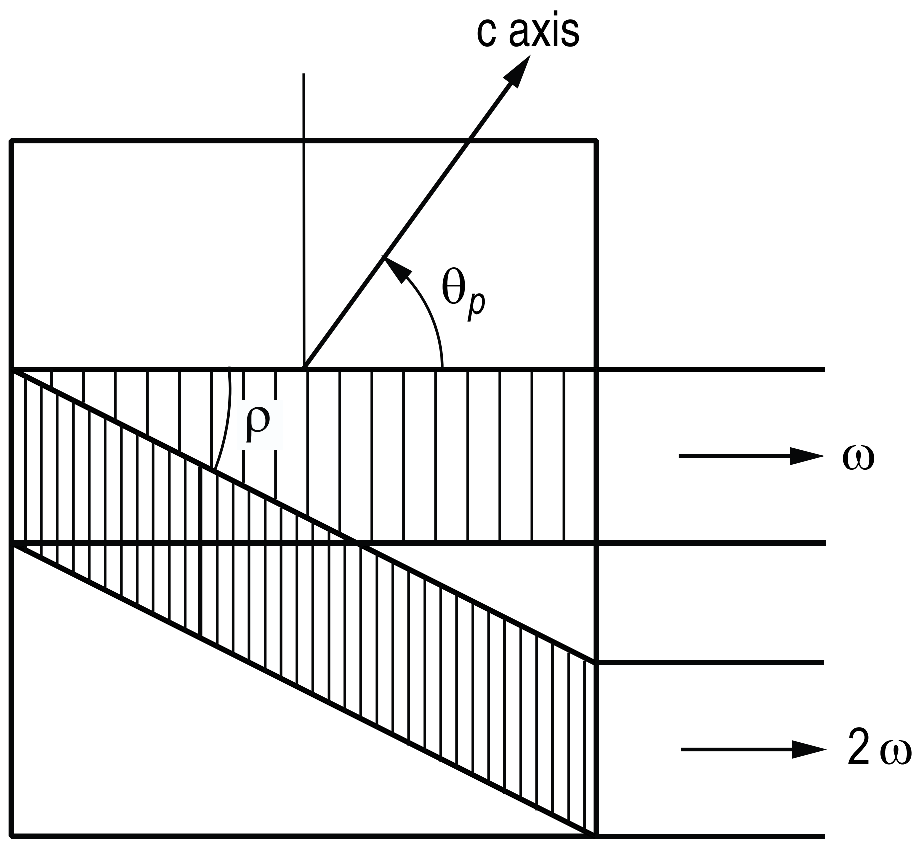
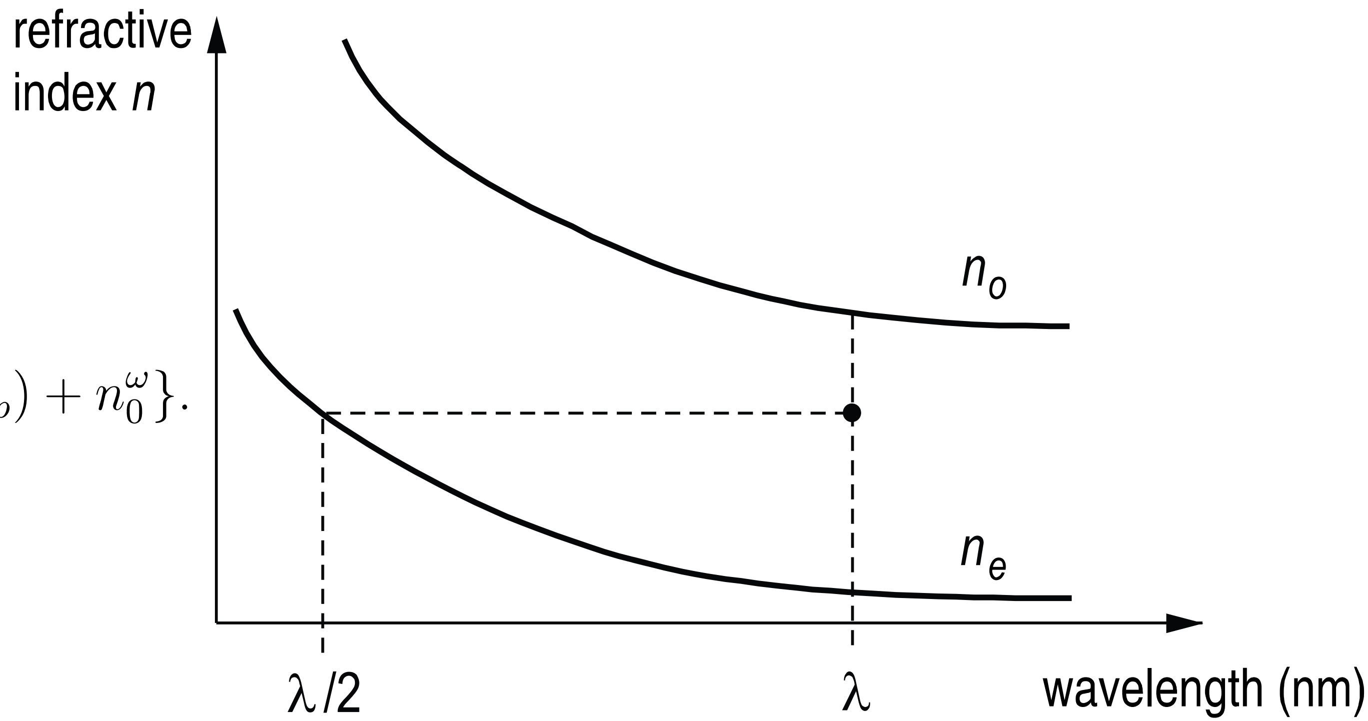


Figure 4.8: Walk-off between ordinary and extraordinary wave.

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

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) :	$oo \rightarrow e$	$oe \rightarrow e$
$n_e > n_o$ (pos. uniaxial) :	$ee \rightarrow o$	$oe \rightarrow o$

Table 4.2: Phase-matching configurations

Acceptance angle

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

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 $\lambda = 1.064\ \mu\mathrm{m}$, $n_e^{\omega} = 1.466$, $n_o^{\omega} = 1.506$, $n_e^{2\omega} = 1.487$, $n_o^{2\omega} = 1.534$. For this case, the phase-matching angle is $\theta_p = 49.9°$ 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

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

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 λ =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

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

the doubled pulse will be longer and the efficiency will be reduced. This can also be understood as temporal walk-off between the fundamental pulse and 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

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

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

Guoy phase shift

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

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

A. E. Siegman, *Lasers* (University Science Books)

Gaussian beam

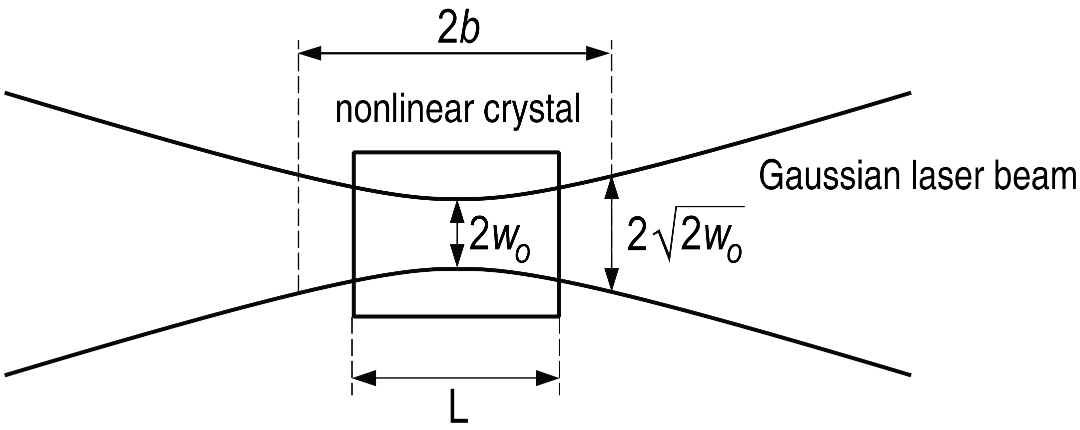


Figure 4.10: Intensity distribution of a Gaussian beam.

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.

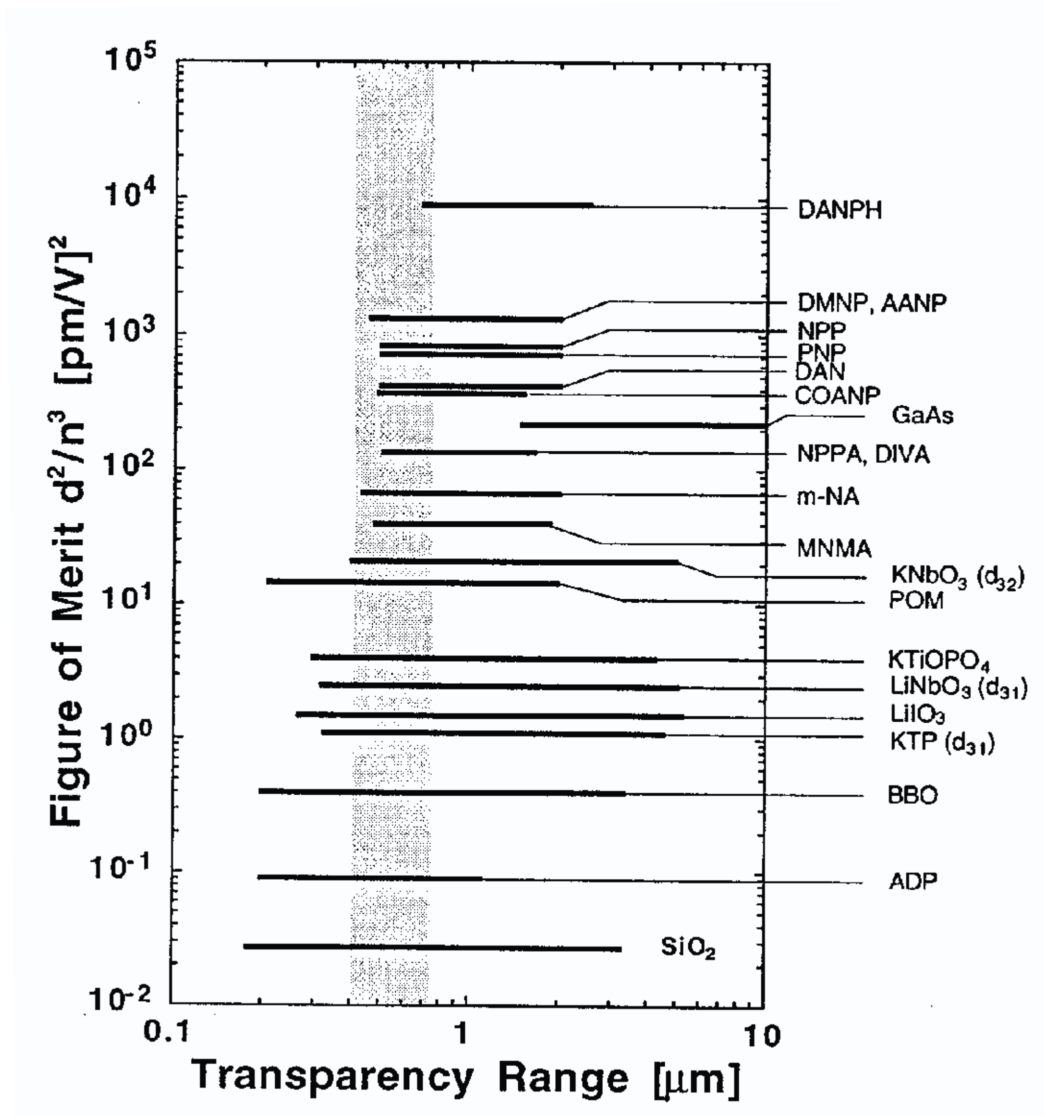


Figure 4.11: Figure of merit (FOM) for different nonlinear optical materials.

4.4.3 Frequency doubling of pulses

$$P(z,\omega) = \epsilon_0 d_{eff} \int_{-\infty}^{\infty} E_1(\omega-\omega_1) E_1(\omega_1) e^{-j(k(\omega-\omega_1)+k(\omega_1))z} d\omega_1.$$

$$k(\omega-\omega_1) = k_0 + \left(\frac{\partial k}{\partial \omega}\right)_{\omega_0} (\omega-\omega_1-\omega_0), \tag{4.63}$$

$$k(\omega_1) = k_0 + \left(\frac{\partial k}{\partial \omega}\right)_{\omega_0} (\omega_1-\omega_0). \tag{4.64}$$

With Eqs. (4.63) and (4.64)

$$\Rightarrow k(\omega-\omega_1) + k(\omega_1) = 2k_0 + \frac{1}{\upsilon_{g1}}(\omega-2\omega_0) \tag{4.65}$$

where

$$\frac{1}{\upsilon_{g1}} = \left.\frac{1}{\upsilon_g}\right|_{\omega_0} = \left(\frac{\partial k}{\partial \omega}\right)_{\omega_0} \tag{4.66}$$

is the inverse group velocity. Then the polarization at the sum-frequency is

$$P(z,\omega) = \epsilon_0 d_{eff} e^{-j\left(2k_0 + \frac{1}{\upsilon_{g1}}(\omega - 2\omega_0)\right)z} \int_{-\infty}^{\infty} E_1(\omega - \omega_1) E_1(\omega_1) d\omega_1. \tag{4.67}$$

The electric field at frequency ω grows according to Eq. (3.8)

$$\frac{\partial E_2(z,\omega)}{\partial z} = -\frac{j\omega_0 d_{eff}}{nc_0} e^{-j\left(2k_0 + \frac{1}{\upsilon_{g1}}(\omega - 2\omega_0) - k(\omega)\right)z} \times \int_{-\infty}^{\infty} E_1(\omega - \omega_1) E_1(\omega_1) d\omega_{1.} \tag{4.68}$$

If $E_1(\omega)$ is the spectrum of the pulse centered around ω_0, then the integral will only be non-zero around $\omega \approx 2\omega_0$ The wave number $k(\omega)$ around $2\omega_0$ is

$$k(\omega) = k_2 + \frac{1}{\upsilon_{g2}}(\omega - 2\omega_0), \tag{4.69}$$

with

$$\frac{1}{\upsilon_{g2}} = \left.\frac{1}{\upsilon_g}\right|_{2\omega_0} = \left(\frac{\partial k}{\partial \omega}\right)_{2\omega_0}. \tag{4.70}$$

For the case of phase matching ($k_2 = 2k_0$) and low conversion

$$E_2(\ell,\omega) = G(\ell,\omega) \cdot F(\omega) \tag{4.71}$$

where

$$G(\ell,\omega) = -\frac{j\omega_0 d_{eff}}{nc_0} e^{j(\Delta k\ell/2)} \cdot \ell \cdot \left\{ \frac{\sin\frac{\Delta k\ell}{2}}{\Delta k\ell/2} \right\}, \tag{4.72}$$

$$\Delta k = \left(\frac{1}{v_{g1}} - \frac{1}{v_{g2}} \right) (\omega - 2\omega_0), \tag{4.73}$$

and

$$F(\omega) = \int_{-\infty}^{\infty} E_1(\omega - \omega_1) E_1(\omega_1)\, d\omega_1. \tag{4.74}$$

The electric field at the second harmonic can then be written as a Fourier transform. In the time domain we obtain with the convolution theorem

$$\frac{1}{2\pi} \int_{-\infty}^{\infty} G(\omega) F(\omega) e^{j\omega t} d\omega = \int_{-\infty}^{\infty} g(t') f(t-t')\, dt' \tag{4.75}$$

where

$$\begin{aligned} g(t) &= \frac{1}{2\pi} \int_{-\infty}^{\infty} G(\omega) e^{j\omega t} d\omega \qquad \color{red}{f(t)=E_1(t)^2} \\ &= \left\{ \begin{array}{ll} \underbrace{e^{j2\omega_0 t}}_{\text{phase}} \frac{\omega_0 d_{eff}}{4nc_0} \frac{1}{\left(\frac{1}{v_{g2}} - \frac{1}{v_{g1}}\right)}, & 0 < t < \left(\frac{1}{v_{g2}} - \frac{1}{v_{g1}}\right) \ell \\ 0, & \text{elsewhere} \end{array} \right\} \end{aligned} \tag{4.76}$$

For a fundamental wave $E_1(t) = A_1(t)\cos(\omega_0 t - k_0 z)$ we obtain a second harmonic wave $E_2(\ell, t) = A_2(\ell, t)\cos(2\omega_0 t - 2k_0 z)$

$$A_2(\ell, t) = \frac{\omega_0 d_{eff}}{4nc_0} \frac{1}{\left(\frac{1}{v_{g2}} - \frac{1}{v_{g1}}\right)} \int_0^{\ell/v_{g2} - \ell/v_{g1}} A_1^2(t - t')\, dt', \tag{4.77}$$

where $A_2(\ell, t)$ is the envelope of the generated second-harmonic pulse obtained by a convolution of a squared input field and a rectangularly shaped pulse of duration $\left(\frac{1}{v_{g2}} - \frac{1}{v_{g1}}\right)\ell$. In the limit $\left(\frac{1}{v_{g2}} - \frac{1}{v_{g1}}\right)\ell \to 0$, we obtain

large doubling bandwidth

$$A_2(\ell, t) = \frac{\omega_0 d_{eff}}{4nc_0} \cdot \ell \cdot A_1^2(t). \tag{4.78}$$

In the case of $\left(\frac{1}{v_{g2}} - \frac{1}{v_{g1}}\right)\ell \gg t_p =$ pulse length, we obtain from Eq. (4.77) a rectangularly shaped pulse with duration

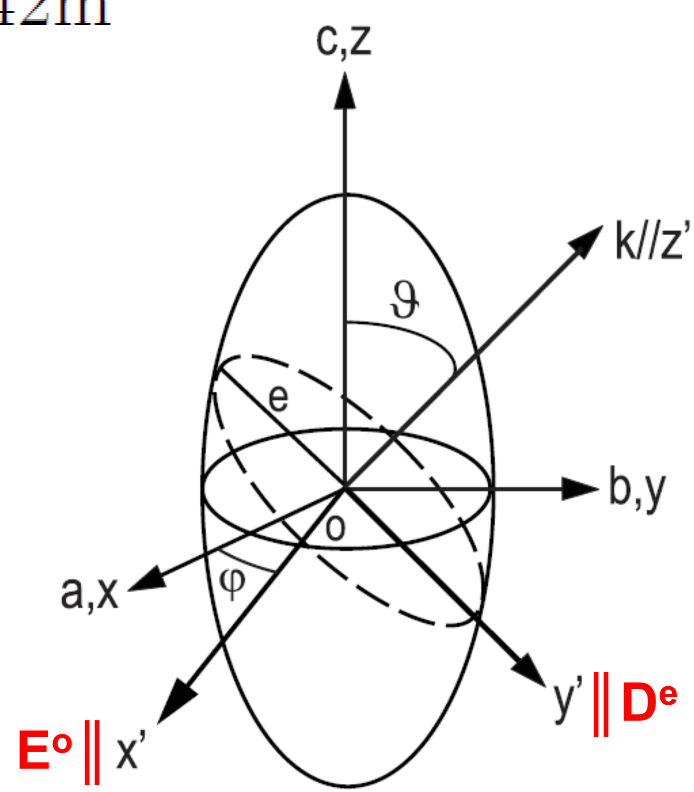
very small doubling bandwidth

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

4.4.3 Effective nonlinear coefficients

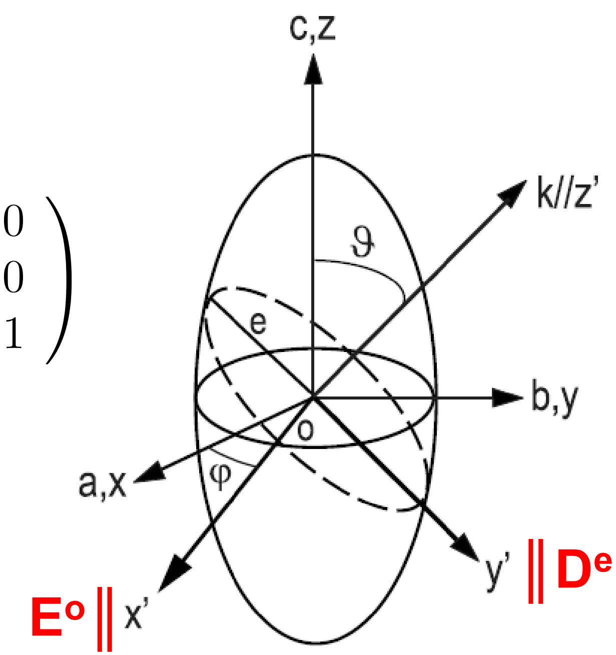
Fig. 4.12. The d tensor of the crystal in a coordinate system $(\mathbf{x}, \mathbf{y}, \mathbf{z})$ aligned with the main axis $(\mathbf{a}, \mathbf{b}, \mathbf{c})$ of the index ellipsoid is in diagonal form. For the purpose of phase matching the crystal is rotated such that the beams propagate in direction $\mathbf{z}'$ of a new coordinate system $(\mathbf{x}', \mathbf{y}', \mathbf{z}')$. The new coordinate system follows from the old one by two transformations, a rotation around the $\mathbf{z}$-axis by an angle φ and another rotation around the $\mathbf{x}'$-axis by an angle $-\vartheta$. The transformation of a vector $\mathbf{u}$ from the old to the new coordinate system

point group $\bar{4}2m$
KDP



4.4.3 Effective nonlinear coefficients

$$\begin{pmatrix} u_{x'} \\ u_{y'} \\ u_{z'} \end{pmatrix} = \mathbf{T} \cdot \begin{pmatrix} u_x \\ u_y \\ u_z \end{pmatrix} \qquad (4.79)$$



with the transformation matrix $\mathbf{T}$

$$\begin{aligned} \mathbf{T} &= \begin{pmatrix} 1 & 0 & 0 \\ 0 & \cos\vartheta & -\sin\vartheta \\ 0 & \sin\vartheta & \cos\vartheta \end{pmatrix} \begin{pmatrix} \cos\varphi & \sin\varphi & 0 \\ -\sin\varphi & \cos\varphi & 0 \\ 0 & 0 & 1 \end{pmatrix} \\ &= \begin{pmatrix} \cos\varphi & \sin\varphi & 0 \\ -\sin\varphi\cos\vartheta & \cos\varphi\cos\vartheta & -\sin\vartheta \\ -\sin\varphi\sin\vartheta & \cos\varphi\sin\vartheta & \cos\vartheta \end{pmatrix} . \end{aligned}$$

The inverse is

$$\mathbf{T}^{-1} = \mathbf{T}^{T} = \begin{pmatrix} \cos\varphi & -\sin\varphi\cos\vartheta & -\sin\varphi\sin\vartheta \\ \sin\varphi & \cos\varphi\cos\vartheta & \cos\varphi\sin\vartheta \\ 0 & -\sin\vartheta & \cos\vartheta \end{pmatrix} . \qquad (4.81)$$

The fundamental and second-harmonic waves are ordinary or extraordinary waves. The ordinary wave, $(\mathbf{E}\|\mathbf{D})$, is polarized along the x'-axis

$$\mathbf{E}^o = \hat{E}^o \cdot \mathbf{x}' = \hat{E}^o \left(\cos\varphi \cdot \mathbf{x} + \sin\varphi \cdot \mathbf{y}\right) \tag{4.82}$$

The dielectric displacement of the extraordinary beam ($\mathbf{E} \nparallel \mathbf{D}$), is polarized along the y'-axis

$$\mathbf{D}^e = D^e \cdot \mathbf{y}' = D^e \left(-\sin\varphi\cos\vartheta \cdot \mathbf{x} + \cos\varphi\cos\vartheta \cdot \mathbf{y} - \sin\vartheta \cdot \mathbf{z}\right). \tag{4.83}$$

There are two possible ways to determine the effective nonlinear coefficient. One way is by transforming the d tensor to a new coordinate system or by substitution of the fundamental and second-harmonic waves in the old coordinate system and decomposing the second-harmonic fields. For example, for frequency doubling with KDP, which is a negative uniaxial crystal belonging to the point group $\bar{4}2\mathrm{m}$, with type-I phase matching:

$$\begin{aligned} \text{fundamental} &\;:\; \mathbf{E}(\omega) = \mathbf{E}^o \| \mathbf{D}^o \\ \text{second harmonic} &\;:\; \mathbf{D}(2\omega) = \mathbf{D}^e \end{aligned}$$

$$
\begin{bmatrix} P_x^{(2)}(2\omega) \\ P_y^{(2)}(2\omega) \\ P_z^{(2)}(2\omega) \end{bmatrix} = \varepsilon_0 \begin{bmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{bmatrix} \cdot \begin{bmatrix} E_x(\omega)^2 \\ E_y(\omega)^2 \\ E_z(\omega)^2 \\ 2E_y(\omega)E_z(\omega) \\ 2E_x(\omega)E_z(\omega) \\ 2E_x(\omega)E_y(\omega) \end{bmatrix}
$$

type-I PM

type-II PM

$$
= \varepsilon_0 \begin{bmatrix} 0 & 0 & 0 & d_{14} & 0 & 0 \\ 0 & 0 & 0 & 0 & d_{14} & 0 \\ 0 & 0 & 0 & 0 & 0 & d_{36} \end{bmatrix} \cdot \begin{bmatrix} \cos^2\varphi \\ \sin^2\varphi \\ 0 \\ 0 \\ 0 \\ \sin(2\varphi) \end{bmatrix} \hat{E}^{o2}
$$

2cosφsinφ

$$
\begin{bmatrix} P_x^{(2)}(2\omega) \\ P_y^{(2)}(2\omega) \\ P_z^{(2)}(2\omega) \end{bmatrix} = \varepsilon_0 \begin{bmatrix} 0 \\ 0 \\ d_{36}\sin(2\varphi) \end{bmatrix} \hat{E}^{o2}
$$

In the new system this corresponds to the polarization multiplication with **T**

$$
\begin{bmatrix} P_{x'}^{(2)}(2\omega) \\ P_{y'}^{(2)}(2\omega) \\ P_{z'}^{(2)}(2\omega) \end{bmatrix} = \varepsilon_0 d_{36}\sin(2\varphi) \begin{bmatrix} 0 \\ -\sin\vartheta \\ \cos\vartheta \end{bmatrix} \hat{E}^{o2} \qquad (4.84)
$$

since the polarization $P^{(2)}_{y'}(2\omega)$ is related to the dielectric displacement of the extraordinary beam. To see that, we would need to rederive Eq. (3.8) in non-isotropic media for the dielectric displacement, instead of the electric fields

$$d_{eff} = -d_{36} \sin(2\varphi) \sin\vartheta. \tag{4.85}$$

Because of Kleinman symmetry $d_{36} = d_{14}$. The effective nonlinear coefficients for type-I phase matching for the different point groups are given in Table 4.3.

crystal class	$2e \to o$	$2o \to e$
6,4	0	$d_{15} \sin\vartheta$
622,422	0	0
6mm,4mm	0	$d_{15} \sin\vartheta$
$\bar{6}$m2	$d_{22} \cos^2\vartheta \cos 3\varphi$	-$d_{22} \cos\vartheta \sin 3\varphi$
3m	$d_{22} \cos^2\vartheta \cos 3\varphi$	$d_{15} \sin\vartheta - d_{22} \cos\vartheta \sin 3\varphi$
$\bar{6}$	$(d_{11} \sin 3\varphi + d_{22} \cos 3\varphi) \cos^2\vartheta$	$(d_{11} \cos 3\varphi - d_{22} \sin 3\varphi) \cos\vartheta$
3	$(d_{11} \sin 3\varphi + d_{22} \cos 3\varphi) \cos^2\vartheta$	$d_{15} \sin\vartheta + (d_{11} \cos 3\varphi - d_{22} \sin 3\varphi) \cos\vartheta$
32	$d_{11} \sin 3\varphi \cos^2\vartheta$	$d_{11} \cos 3\varphi \cos\vartheta$
$\bar{4}$	$(d_{14} \cos 2\varphi - d_{15} \sin 2\varphi) \sin 2\vartheta$	$-(d_{14} \cos 2\varphi + d_{15} \cos 2\varphi) \sin\vartheta$
$\bar{4}$2m	$d_{14} \cos 2\varphi \sin 2\vartheta$	$-d_{14} \sin 2\varphi \sin\vartheta$

Table 4.3: Effective conversion coefficient d_{eff}, if Kleinman symmetry is valid.

4.4.5 Quasi-phase matching (QPM)

Sometimes to achieve phase matching of a nonlinear process in the desired wavelength range is not possible by birefringence only. In that case, or for achieving a collinear interaction of waves, one can use quasi-phase matching (QPM), a technique introduced by N. Bloembergen, Nobel Prize in Physics 1981 (J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, "Interactions between Light Waves in a Nonlinear Dielectric," Phys. Rev. **127**, 6

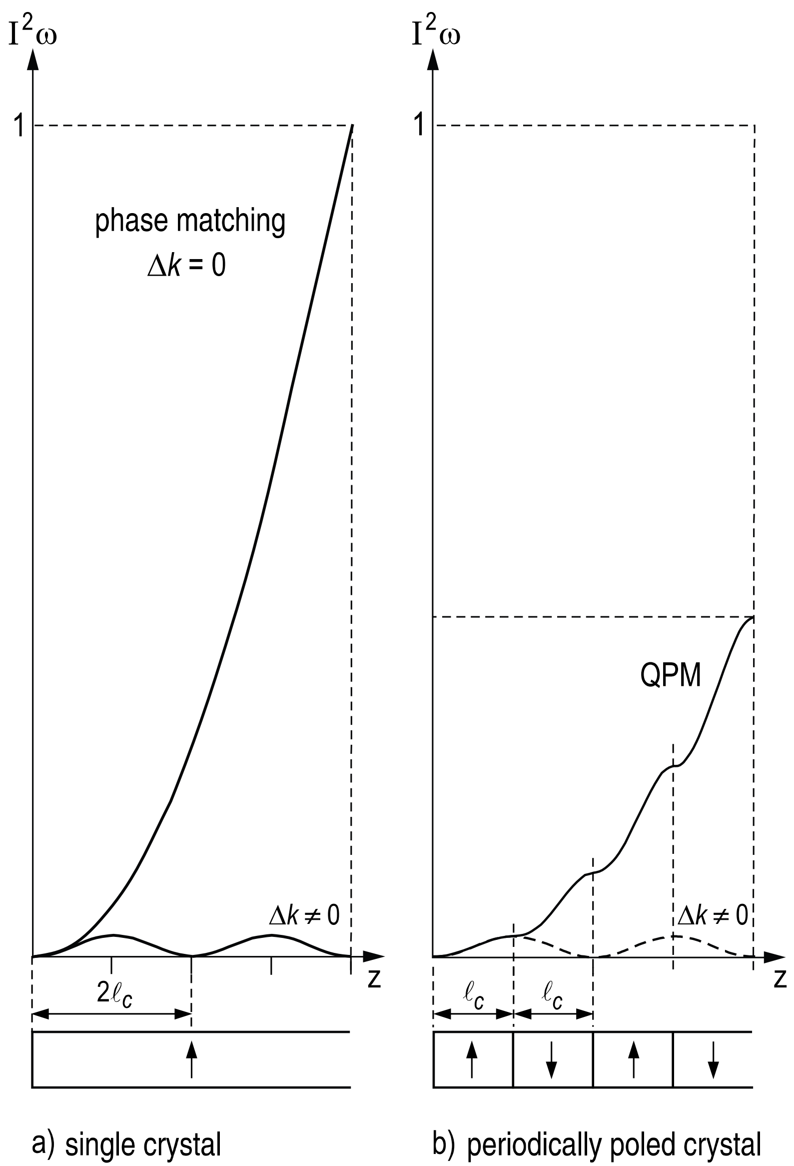
high technological relevance!

custom-engineer phase matching
e.g., mid-IR, THz generation

fan-out QPM gratings

chirped QPM gratings

waveguide QPM devices etc.



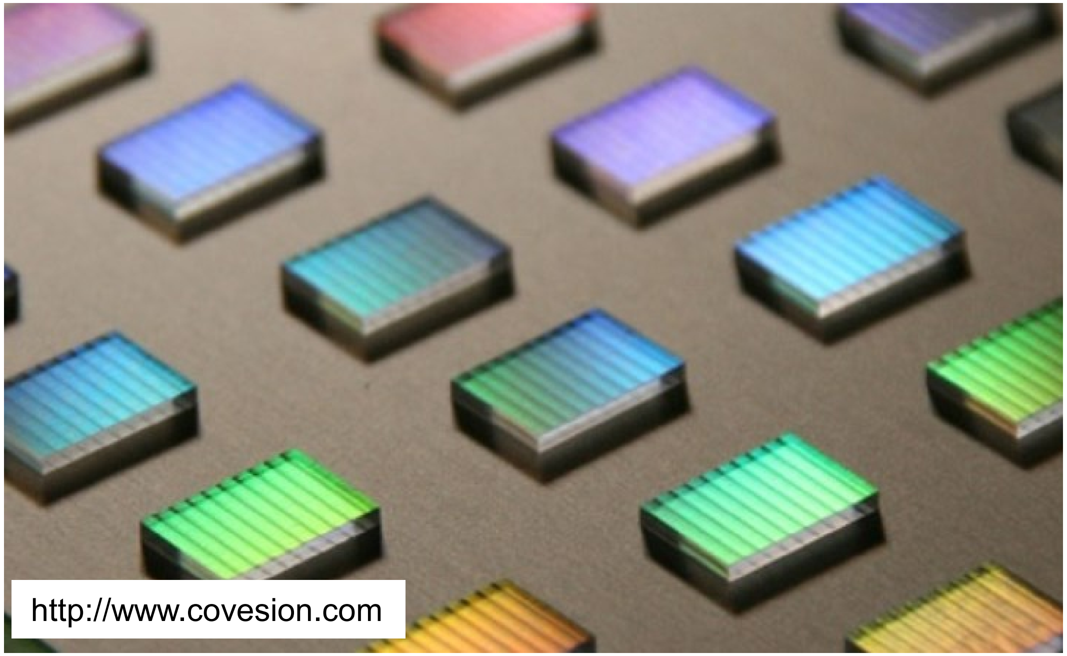


Figure 4.13: Growth of second harmonic as a function of distance z in a crystal for different cases: a) homogeneous crystal and b) periodically poled crystal.

occurs. Due to phase mismatch the second harmonic runs out of phase with the driving wave and therefore the generating polarization. If the sign of the nonlinearity is switched in the second layer, a phase advance by π is introduced in the driving polarization, which rephases it with the already present second harmonic and the process continues with maximum efficiency, see Fig. 4.13.

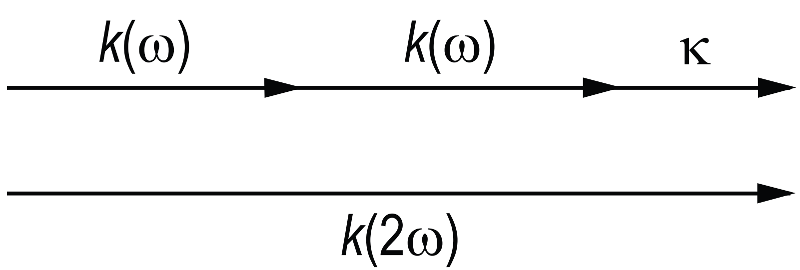
$$\frac{\partial \hat{E}(2\omega)}{\partial z} = -\frac{j\omega}{n_{2\omega}c} d_{eff}(z)\hat{E}(\omega)\hat{E}(\omega)\mathrm{e}^{j(k(2\omega)-2k(\omega))z}. \tag{4.86}$$

Since the spatial modulation is periodic, we can represent it as a Fourier series

$$d_{eff}(z) = \sum_{m=-\infty}^{+\infty} d_m e^{jm\kappa z}. \tag{4.87}$$

If the period of the nonlinear coefficient corresponds to twice the coherence length at a given frequency, i.e., $\kappa = k(2\omega) - 2k(\omega)$, then SHG is rephased and grows over multiple periods on average like

$$\frac{\partial \hat{E}(2\omega)}{\partial z} = -\frac{j\omega}{n_{2\omega}c} d_{-1}\hat{E}(\omega)\hat{E}(\omega) \tag{4.88}$$



$\Delta k_{total} = \Delta k_{process} + 2\pi/\Lambda(z)$
$\Lambda(z)$ grating period

no walk-off