

# **Second Order Nonlinear Processes**

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# Nonlinear Effects

$$P_{tot} = \epsilon_0 \chi^{(1)} E + \epsilon_0 \chi^{(2)} E^2 + \epsilon_0 \chi^{(3)} E^3 + \dots$$

## Second order effects

Linear + 2<sup>nd</sup> + 3<sup>rd</sup> + Higher

Second harmonic generation (SHG)

Optical parametric oscillation (OPO)

Optical parametric amplification (OPA).

Pockel's effect.

Electro-optical beam deflection.

Optical rectification.

## Third order effects

Third harmonic generation (THG).

Raman effect.

Kerr effect - induced birefringence.

Four- wave difference frequency mixing (FWM).

Self- diffraction

Self-phase modulation

Stimulated Brillouin scattering

## Higher Order effects

High harmonic generation

## 2<sup>nd</sup> Order Effects

Two different electric fields  $E_1$ ,  $E_2$  are superimposed and generate the NL polarization  $P^{(2)} = \epsilon_0 \chi^{(2)} E_1 E_2$

The incident optical field is:

$$\tilde{E}(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + c.c.$$

Nonlinear polarization contains the following terms:

$$P(2\omega_1) = \chi^{(2)} E_1^2$$

2<sup>nd</sup> harmonic generation (SHG)

$$P(2\omega_2) = \chi^{(2)} E_2^2$$

2<sup>nd</sup> harmonic generation (SHG)

$$P(\omega_1 + \omega_2) = 2\chi^{(2)} E_1 E_2$$

Sum freq. generation (SFG)

$$P(\omega_1 - \omega_2) = 2\chi^{(2)} E_1 E_2^*$$

Difference freq. generation (DFG)

$$P(0) = 2\chi^{(2)} (E_1 E_1^* + E_2 E_2^*)$$

Optical rectification (OR)

# Second Order Susceptibility

The second-order nonlinear susceptibility  $\chi^{(2)}$  of used crystals reaches values of up to about  $10^{-10}$  cm/V and thus demands intensities of up to 0.1–1 GW cm<sup>-2</sup>. These values are close to the damage threshold of optical surfaces of the commonly applied materials and thus new materials are developed

$$\chi^{(2)} \simeq 1.94 \times 10^{-12} \text{ m/V}$$

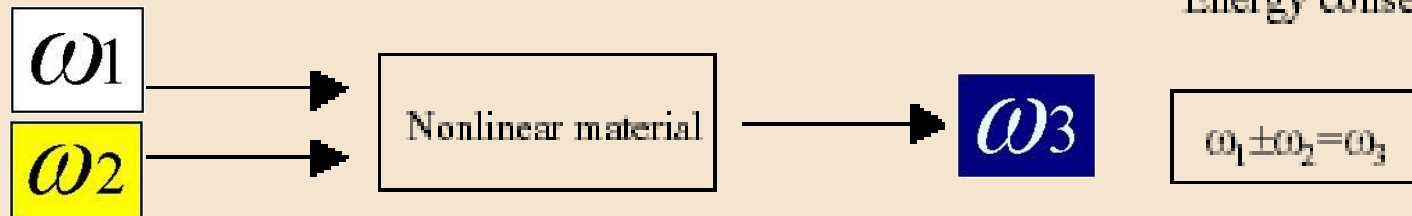
In comparison with the 3<sup>rd</sup> order

$$\chi^{(3)} \simeq 3.78 \times 10^{-24} \text{ m}^2/\text{V}^2$$

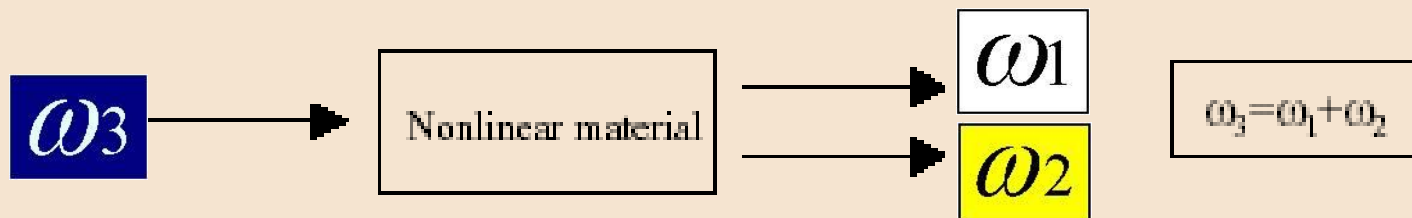
# Frequency Conversion Processes

Several electromagnetic waves of different  $\omega$ 's can interact with each other in the material under the condition of energy conservation.

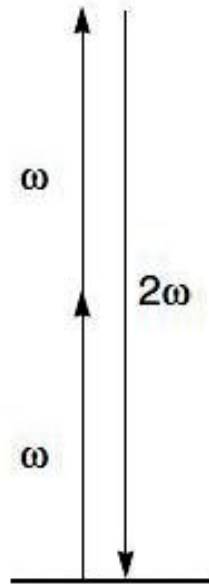
1. SHG, SFG, DFG



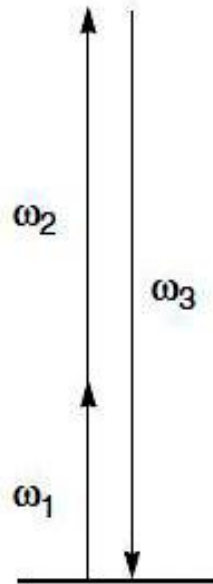
2. Parametric down conversion



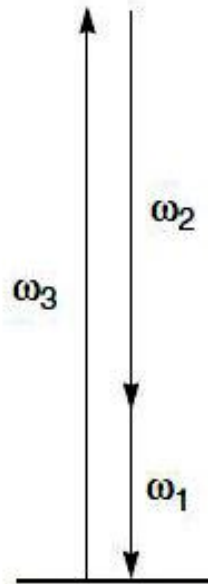
# Diagram of some NL processes



**SHG**



**SFG**



**DFG/OPA**

Diagrammatic representation of various nonlinear processes including second harmonic generation (SHG), sum frequency generation (SFG), difference frequency generation (DFG), optical parametric amplification (OPA)

## ***Frequency Conversion Processes***

- **Second harmonic generation (SHG) or *frequency doubling*:**  
Generation of light with a doubled frequency (half the wavelength).
- **Sum frequency generation (SFG):**  
Generation of light with a frequency that is the sum of two other frequencies (SHG is a special case).
- **Difference frequency generation (DFG):**  
Generation of light with a frequency that is the difference between two other frequencies.
- **Optical parametric amplification (OPA):**  
Amplification of a signal input in the presence of a higher-frequency pump wave, at the same time generating an *idler* wave.

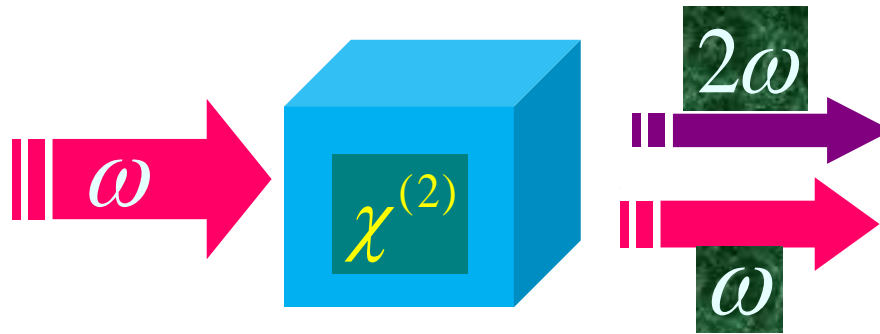
## *Frequency Conversion Processes, Cont.*

- **Optical parametric oscillation (OPO):**  
Generation of a signal and idler wave using a parametric amplifier in a resonator (with no signal input).
- **Optical parametric generation (OPG):**  
Like parametric oscillation but without a resonator, using a very high gain instead.
- **Optical rectification (OR):**  
Generation of quasi-static electric fields.



# Second Harmonic Generation (SHG)

- **Second harmonic generation (SHG, also called frequency doubling) is a nonlinear optical process, in which photons interacting with a nonlinear material are effectively "combined" to form new photons with twice the energy, and therefore twice the frequency and half the wavelength of the initial photons.**



- **In the framework of classical physics, light propagation is described in terms of coherent emission by harmonic electronic dipoles, which have been set to oscillate by the light itself. This picture can be extended to the nonlinear regime by assuming that at high excitation intensities (meaning large oscillation amplitude of the electronic oscillators), the emitting dipoles can radiate energy at frequencies which are integer multiples of the original frequency.**

# SHG



$$I_{532} = \text{constant} \cdot (I_{1064})^2$$

In second-order nonlinear optical effects two different light waves with their electric field vectors  $E_1$  and  $E_2$  can superimpose and generate the nonlinear polarization  $P^{(2)}$ :

$$P^{(2)} = \varepsilon_0 \chi^{(2)} E_1 E_2. \quad (4.16)$$

In Cartesian coordinates  $x, y, z$  described by the indices  $m, p, q$  the components  $P_x, P_y$  and  $P_z$  of the vector  $P^{(2)}$  can be calculated from:

$$P_m^{(2)} = \varepsilon_0 \sum_{p,q} \chi_{m qp}^{(2)} E_p E_q \quad (4.17)$$

with the components  $\chi_{mpq}$  of the third-order tensor with 27 components and the components of the two electric field vectors. The  $\chi$  components are not independent. In any case  $\chi_{mpq} = \chi_{mqp}$  is valid as a consequence of the interchangeability of the two electric fields, resulting in 18 independent components.

For nonabsorbing materials these components are real and the equation  $\chi_{mpq} = \chi_{pmq}$  also has to be fulfilled. In this case only 10 elements of the  $\chi$  tensor are different

## Generation of the Second Harmonic

In the simplest possible case of two equal monochromatic light waves with the same polarization, frequency  $\nu_{\text{inc}}$  and direction  $\mathbf{k}_{\text{inc}}$ , which can be two shares of the same wave, the second-order nonlinear polarization is determined by the product of the two electric fields. This second-order nonlinear polarization shows terms with frequencies  $\nu = 0$  and  $2\nu_{\text{inc}}$ .

$$\begin{aligned} P^{(2)} &= \varepsilon_0 \chi^{(2)} \mathbf{E}_1 \mathbf{E}_2 \\ &= \varepsilon_0 \chi^{(2)} \mathbf{E}^2 \\ &= \varepsilon_0 \chi^{(2)} \{E_0(\mathbf{k}, \varphi) \cos(2\pi\nu_{\text{inc}}t)\}^2 \\ &= \frac{1}{2} \varepsilon_0 \chi^{(2)} E_0^2(\mathbf{k}, \varphi) + \frac{1}{2} \varepsilon_0 \chi^{(2)} E_0^2(\mathbf{k}, \varphi) \cos(4\pi\nu_{\text{inc}}t) \\ &= P^{(2)}(0) + P^{(2)}(2\nu_{\text{inc}}). \end{aligned} \tag{4.18}$$

The second term describes the material polarization which oscillates with twice the frequency of the incident wave. This polarization will emit a new light wave with twice the frequency of the incident wave. This wave is called the *second harmonic* produced by Second Harmonic Generation (SHG). The first term describes the generation of a “rectified” field, resulting in a macroscopic charge separation in the material.

Thus second-order nonlinearity can be used in photonic applications for the generation of light with twice the photon energy of the incident light with,

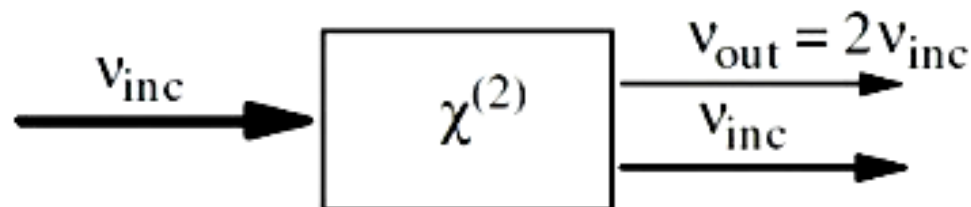


Fig. 4.4. Schematic of the generation of frequency-doubled light via second-order nonlinear susceptibility, e.g. in a suitable crystal

frequency  $\nu_{\text{inc}}$  (see Fig. 4.4) which is, e.g., blue light from red or green light from infrared laser radiation

The energy efficiency of the second harmonic generation (SHG) is smaller than 1 and some nonconverted light will be observed behind the nonlinear medium.

The calculation of the nonlinear polarization has to include all real second-order products of the electric field vectors:

$$\begin{pmatrix} P_x^{(2)}(2\nu_{\text{inc}}) \\ P_y^{(2)}(2\nu_{\text{inc}}) \\ P_z^{(2)}(2\nu_{\text{inc}}) \end{pmatrix} = \varepsilon_0 \begin{pmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{pmatrix} \cdot \begin{pmatrix} E_x^2 \\ E_y^2 \\ E_z^2 \\ 2E_y E_z \\ 2E_x E_z \\ 2E_x E_y \end{pmatrix} \quad (4.19)$$

leading to 18  $d$  coefficients of the nonlinear material instead of the 27  $\chi_{mpq}$  values. For nonabsorbing materials the following relations are valid:

$$\begin{aligned}
 d_{14} &= d_{25} = d_{36} \\
 d_{12} &= d_{26} \quad d_{13} = d_{35} \quad d_{15} = d_{31} \quad d_{16} = d_{21} \\
 d_{23} &= d_{34} \quad d_{24} = d_{32}
 \end{aligned}
 \tag{4.20}$$

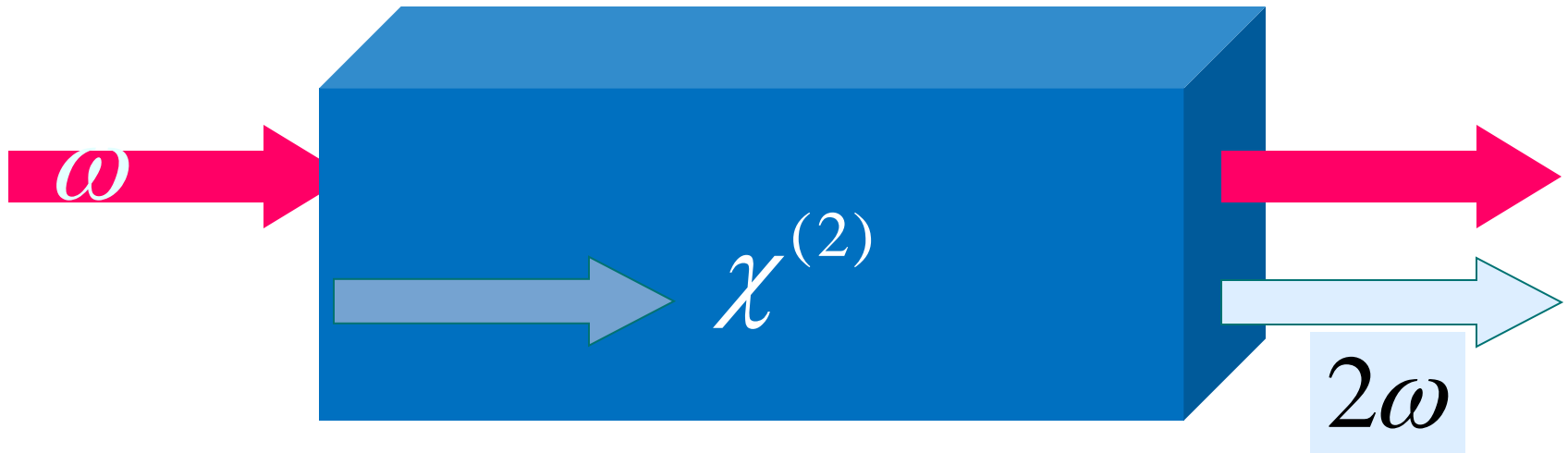
finally resulting in 10 different components. The assignment of the  $d_{rs}$  to  $\chi_{mpq}$  can be obtained by comparing (4.19) and (4.17). It should be mentioned again that these matter parameters  $d_{rs}$  are functions of the applied light frequencies.

Generation of new frequencies via NL polarization in matter is more efficient the better the incident light waves, and the new generated waves are in suitable phase over the interaction length.

→ Achieved by choosing a suitable orientation of the crystal with respect to the light beam and is called **phase matching**

→ Birefringence in anisotropic materials like crystals is used.

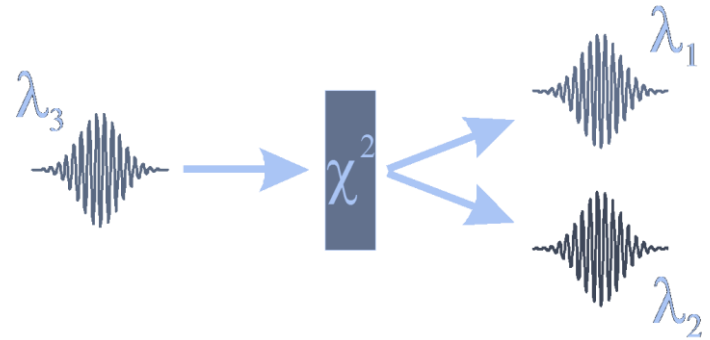
# Phase Matching



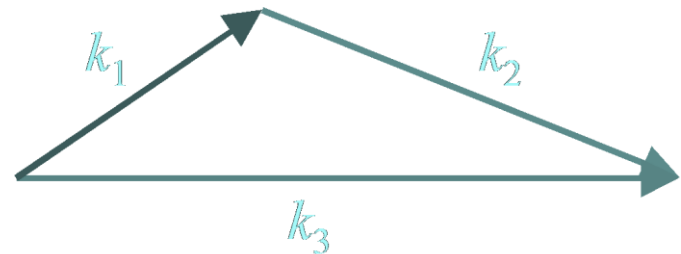
- Since the optical NL media are dispersive, the fundamental and the harmonic signals have different propagation speeds inside the media.
- The harmonic signals generated at different points interfere destructively with each other.



- **Energy Conservation**



- **Momentum conservation**



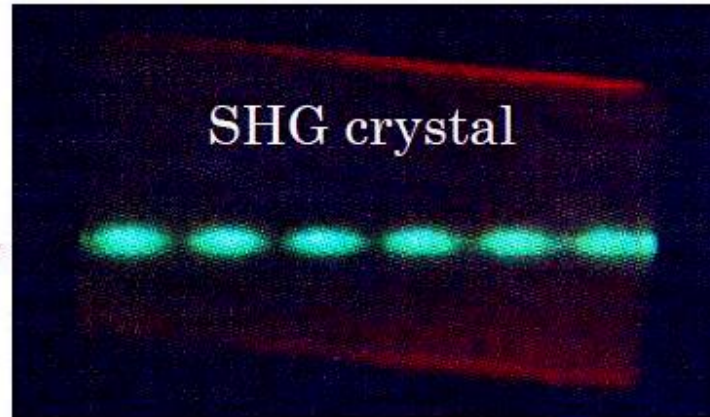
- **Phase condition**

$$\Delta\phi = \phi_3 - \phi_1 - \phi_2 = -\frac{\pi}{2} \Rightarrow \text{parametric amplification}$$

# SHG in real crystals

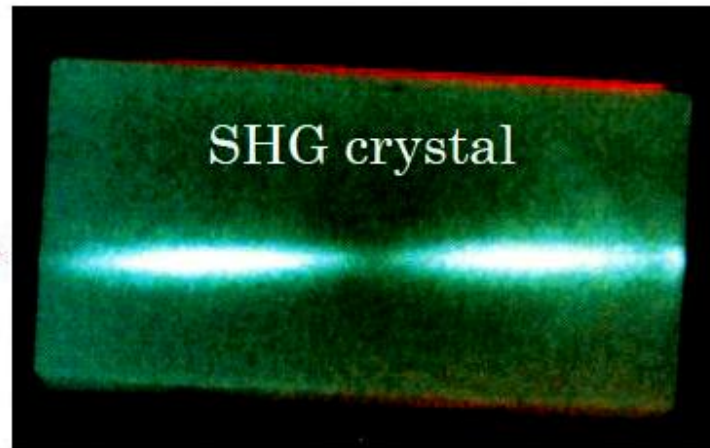
Far from  
phase-matching:

Large  $\Delta k$   
Input beam



Closer to  
phase-matching:

Small  $\Delta k$   
Input beam



Note that SH beam is brighter as phase-matching is achieved.  
The SHG intensity is sharply maximized if  $\Delta k = 0$ .

## Efficient parametric conversion requires:

- \* Appropriate nonlinear medium (*birefringence*).
- \* Interacting waves maintain temporal overlap.
- \* Interacting waves remain phase matched ( $\Delta\mathbf{k} \approx \mathbf{0}$ ).

Birefringence is not a perfect solution, alternative is the concept of **quasi-phase matching (QPM)**

### *Phase Matching for Second Harmonic Generation*

Some details will be discussed here for the example of the generation of second harmonic light but the mechanisms are valid analogously for other frequency conversion setups, too.

The increasing amplitude of the SHG wave can be calculated from (4.10). The nonlinear polarization for this wave is:

$$P_{\text{nl}}^{(2)}(2\nu_{\text{inc}}) = \frac{1}{2}\epsilon_0\chi^{(2)}E_{\text{inc},0}^2(z)e^{i2(2\pi\nu_{\text{inc}}t - k_{\text{inc}}z)}. \quad (4.21)$$

With this nonlinear polarization (4.10) results for the SHG wave in:

$$\frac{\partial E_{\text{SHG}}(z)}{\partial z} = -i \frac{k_{\text{SHG}} \chi^{(2)}}{4n_{\text{SHG}}^2} E_{\text{inc}}^2(z) e^{-i\Delta k z} \quad (4.22)$$

which cannot easily be solved. Assuming an undepleted incident wave, it describes an oscillation of the amplitude of the electric field of the generated second harmonic light with  $z$  as a function of  $\Delta k = |k_{\text{SHG}} - k_{\text{inc}}|$ . This oscillation of the intensity of the SHG light can be calculated as:

$$I_{\text{SHG}}(z) = I_{\text{inc}}^2 \frac{2\pi^2 d^2}{\varepsilon_0 c_0 \lambda_{\text{inc}}^2 n_{\text{inc}}^2 n_{\text{SHG}}} \left[ \frac{\sin(\Delta k z / 2)}{\Delta k / 2} \right]^2 \quad (4.23)$$

with the different refractive indices  $n_{\text{inc}}$  and  $n_{\text{SHG}}$  for the wavelengths  $\lambda_m$  of the incident wave and the SHG wave in the material and  $d$  as the relevant matrix element of the  $d$  matrix of the material.

This oscillation results from the phase mismatch of the incident and the SHG wave which are in phase at the entrance surface and at distances  $z_{\text{ip}}$

$$z_{\text{ip}} = \frac{\pi}{\Delta k} m = \frac{\lambda_{\text{inc}}}{4(n_{\text{SHG}} - n_{\text{inc}})} m \quad (4.24)$$

and out of phase after a path length  $z_{\text{oop}}$  of:

$$z_{\text{oop}} = \frac{2\pi}{\Delta k} m = \frac{\lambda_{\text{inc}}}{2(n_{\text{SHG}} - n_{\text{inc}})} m \quad (4.25)$$

and so on with the integer  $m$ . For energy conservation the amplitude of the incident wave will oscillate, too.

This problem can be avoided by choosing

$$\text{phase matching } \Delta k = |k_{\text{SHG}} - k_{\text{inc}}| \stackrel{!}{=} 0 \quad (4.26)$$

applying a suitable crystal orientation. In this case (4.22) can be solved analytically and the intensity of the SHG  $I_{\text{SHG}}$  follows from the pump or incident light intensity  $I_{\text{inc}}$  as:

$$\begin{aligned} I_{\text{SHG}}(z) &= I_{\text{inc}}(z=0) \tanh^2 \left( z \sqrt{\frac{8\pi^2 d^2 I_{\text{inc}}}{\varepsilon_0 c_0 n_{\text{inc}}^2 n_{\text{SHG}} \lambda_{\text{inc}}^2}} \right) \\ &= I_{\text{inc}}(z=0) \tanh^2 \left( 0.88 \frac{z}{z_{\text{HM}}} \right) \end{aligned} \quad (4.27)$$

again with  $d$  as the relevant matrix element of the  $d$  matrix of the material and a characteristic length  $z_{\text{HM}}$  at which the intensity of the SHG reaches 50% of the original incident intensity  $I_{\text{inc}}(z=0)$ :

$$z_{\text{HM}} = 0.31 \sqrt{\frac{\varepsilon_0 c_0 n_{\text{inc}}^2 n_{\text{SHG}} \lambda_{\text{inc}}^2}{\pi^2 d^2 I_{\text{inc}}}} \quad (4.28)$$

The incident intensity  $I_{\text{inc}}$  will decrease with  $z$  in non-absorbing materials by:

$$I_{\text{inc}}(z) = I_{\text{inc}}(z = 0) - I_{\text{SHG}}(z) \quad (4.30)$$

and thus the intensities of the incident and the new generated SHG wave can be plotted as in Fig. 4.5.

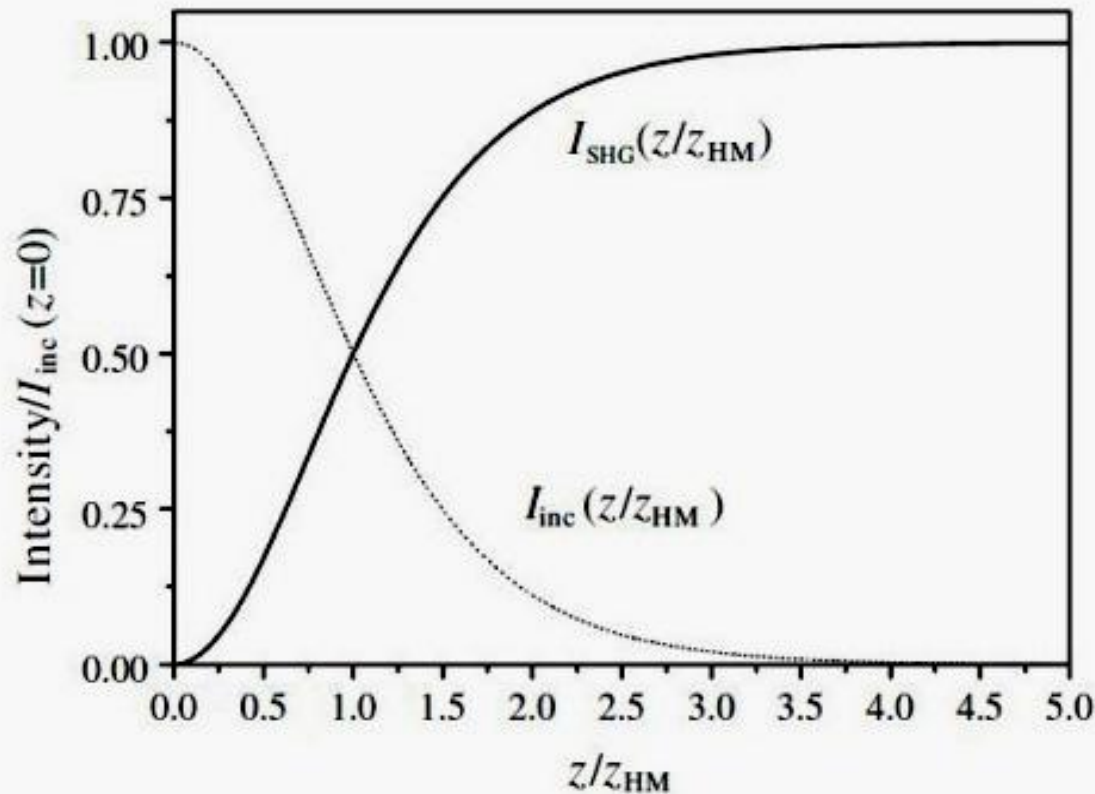
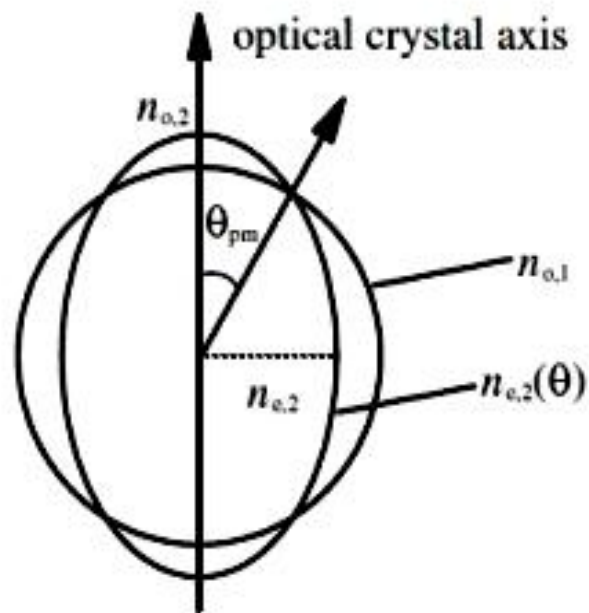


Fig. 4.5. Intensities of incident and new generated light waves in second harmonic generation SHG as a function of the interaction length in material with phase matching

Phase matching can be achieved with anisotropic materials like crystals and is then based on the birefringence in these materials. Therefore the orientation and the temperature of the crystal have to be chosen for equal refractive indices for the incident and the newly generated waves. Often the polarization of the incident and the newly generated waves are perpendicular as a result of the nonlinear polarization in the crystal and thus different refractive index ellipsoids are available for phase matching.

For example in the case of second harmonic generation (SHG) it is possible to match the refractive index for the incident fundamental wave as an ordinary beam with the second harmonic wave as an extraordinary beam even in a uniaxial crystal





**Fig. 4.6.** Phase matching angle for SHG in a uniaxial crystal with refractive index sphere for an ordinary incident beam  $n_{o,1}$  and refractive index ellipse for an extraordinary SHG beam  $n_{e,2}$ . The refractive index sphere for an ordinary SHG beam  $n_{o,2}$  is not shown. It has the radius of the extraordinary index at the optical axis

If an intersection of the two refractive index surfaces for the two waves exists then phase matching can be achieved. In this example the phase matching angle  $\theta_{pm}$  follows with the definitions of Fig. 4.6 to:

phase matching angle

$$\sin^2 \theta_{pm} = \frac{\frac{1}{n_{o,1}^2} - \frac{1}{n_{o,2}^2}}{\frac{1}{n_{e,2}^2} - \frac{1}{n_{o,2}^2}} \quad (4.31)$$

and reaches values of 40–60° in crystals such as, e.g. KDP or KTP. The two refractive indices for an incident light wavelength of 1064 nm as a function of the angle of the incident light beam with respect to the optical axis of the crystal for KDP at room temperature are shown in Fig. 4.7. Phase matching is realized at the angle where the two refractive index curves cross in this figure. Crystals can be ordered in this cut to have a perpendicular entrance surface which can be anti-reflection coated for a higher damage threshold.

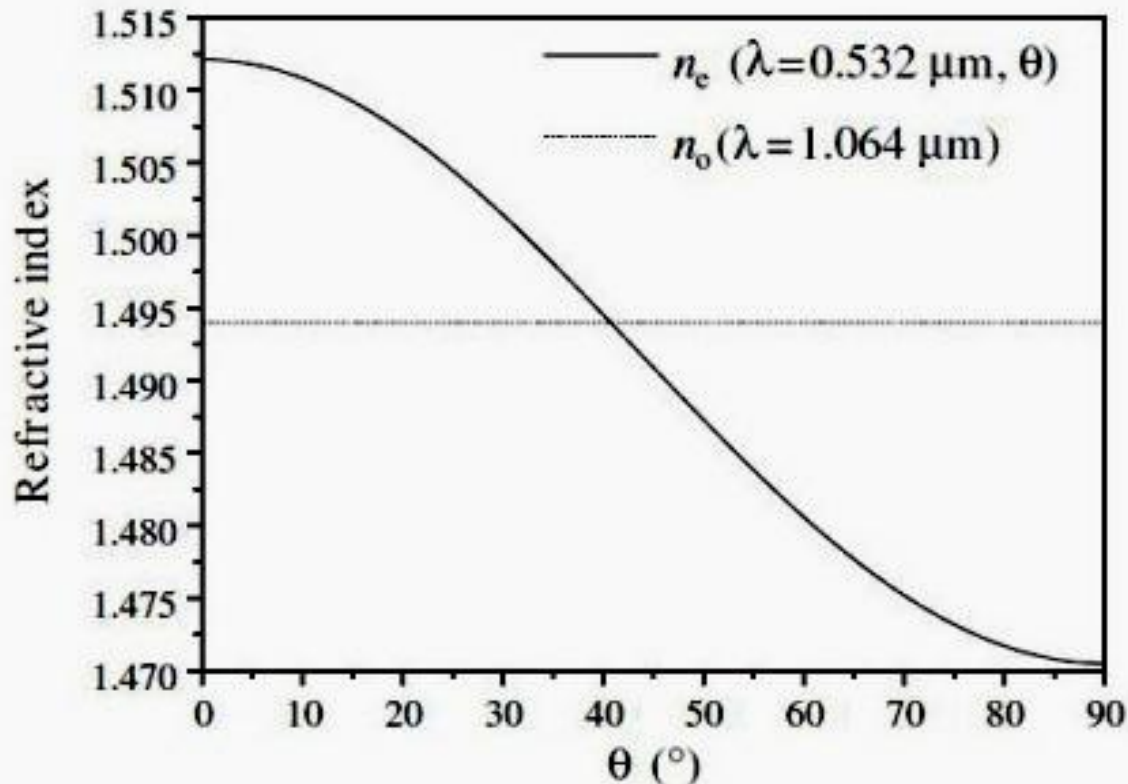
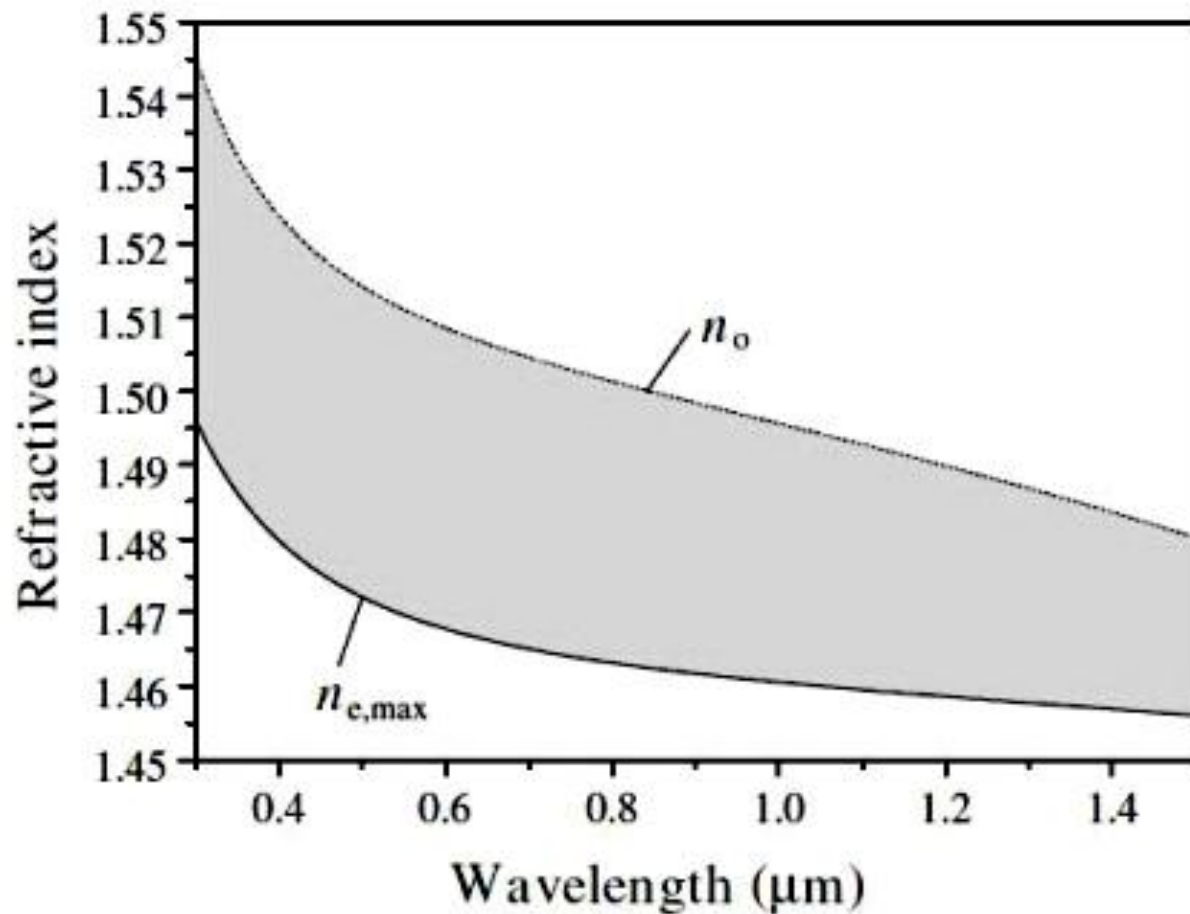


Fig. 4.7. Refractive indices for the ordinary beam of the fundamental and of the extraordinary beam of the second harmonic wave for KDP as a function of angle

The refractive index  $n_e$  is the value of the index ellipsoid perpendicular to the optical axis and thus the extreme value. As an example the refractive indices are given for some typical crystals for the wavelengths of the Nd lasers and their harmonics in Table 4.4.

**Table 4.4.** Refractive indices for the ordinary and extraordinary beams for some typical crystals for the wavelengths of Nd lasers and their harmonics. The value  $n_o$  is given for the direction perpendicular to the optical axis

		KDP	KD*P	ADP	CDA	CD*A
1064 nm	$n_o$	1.4942	1.4931	1.5071	1.5515	1.5499
	$n_e$	1.4603	1.4583	1.4685	1.5356	1.5341
532 nm	$n_o$	1.5129	1.5074	1.5280	1.5732	1.5692
	$n_e$	1.4709	1.4683	1.4819	1.5516	1.5496
355 nm	$n_o$	1.5317	1.5257	1.5487	1.6026	1.5974
	$n_e$	1.463	1.4833	1.4994	1.5788	1.5759



**Fig. 4.8.** Refractive indices for the ordinary and extraordinary beams for KDP as a function of the wavelength. By choosing the appropriate phase matching angle the refractive index for the extraordinary beam can be varied between  $n_{e,max}$  and  $n_o$  to get the same value but for different wavelengths

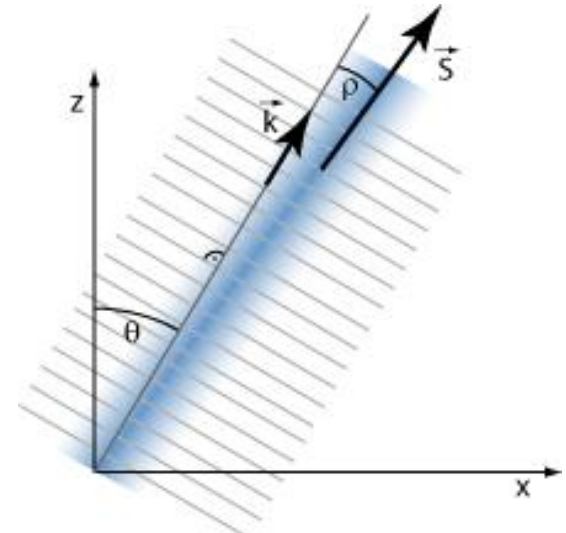
But not all materials allow phase matching for the required combination of wavelengths. Therefore the user should check with the suppliers of nonlinear materials for frequency conversion for current information. A few examples are given in [4.71–4.77]. Because of the large economical importance new materials are under investigation. Larger nonlinear parameters, higher damage threshold and other useful spectral ranges are the main goals. Especially for applications in the UV and mid-IR are new materials demanded. In addition new concepts such as periodical poling (see below) and waveguide structures are developed.

For some materials phase matching is reached by exact angle tuning of the crystal, which is called critical phase matching. Examples are KTP and KDP. KTP is useful for low powers in the Watt range because of its high nonlinear coefficients. At higher powers a color center can occur in KTP which may be taken out by heating the crystal. KDP allows very large sizes and can be used for large beams. However its acceptance angle is small.  $\text{LiNbO}_3$  is temperature tuned in noncritical phase matching and is widely spread today.

# Spatial Walk-off

**Definition:** the phenomenon that the intensity distribution of a beam in an anisotropic crystal drifts away from the direction of the wave vector

**Figure 1:** Spatial walk-off: the intensity distribution of a beam in an anisotropic crystal propagates in a direction which is slightly different to that of the wave vector.



For an **isotropic** medium, the transverse intensity distribution propagates along the beam axis as defined by the medium  $k$  vector. In anisotropic crystals it can occur that the intensity distribution drifts away from the direction defined by the  $k$  vector, as illustrated in Fig. 1, where the gray lines indicate wavefronts and the blue color the region with significant optical intensity.

Spatial walk-off occurs only for a beam with extraordinary polarization, propagating at some angle  $\theta$  against the optical axes, so that the refractive index  $n_e$  and the phase velocity become dependent on that angle.

The walk-off angle can then be calculated from:

Walk-off angle  $\rho = -\frac{1}{n_e} \frac{\partial n_e}{\partial \theta}$

The  $-ve$  sign indicates that the walk-off occurs in the direction where the refractive index would decrease.

A beam with ordinary polarization ( $n$  is independent of the propagation angle) does not experience walk-off.

### *Walk-Off Angle*

Even in case of perfect phase matching the directions of the beam propagation of the new and old frequencies and the propagation direction of their energy (Poynting vector) can be different because the electrical displacement  $\mathbf{D}$  and the electric field  $\mathbf{E}$  are not parallel in case of extraordinary beams. In this type of critical phase matching the interaction length is limited by the “walk-off” between the beam and energy propagation directions [e.g. 4.78–4.80]. In this example the walk-off angle  $\varphi_{\text{wo}}$  results from:

$$\text{walk-off angle} \quad \tan \varphi_{\text{wo}} = \frac{(n_{e,1}^2 - n_{e,2}^2) \tan \theta_{\text{pm}}}{n_{e,2}^2 + n_{e,1}^2 \tan \theta_{\text{pm}}} \quad (4.34)$$

with, e.g., a value of  $1.4^\circ$  for KDP and a wavelength of 1064 nm. By contrast, in *noncritical phase matching* with  $\theta_{\text{pm}} = 90^\circ$  no walk-off occurs.



For high efficiency of frequency conversion the intensity should be as high as possible (but safely below damage threshold of the material), but stronger focusing reduces the interaction length. Secondly it decreases the phase matching because of the higher divergence of the incident light. The acceptance angle  $\Delta\theta_{\text{pm}}$  depends on the dimensions of the ellipsoids and is given for the above example by:

$$\Delta\theta_{\text{pm}} = \frac{0.442\lambda_{\text{inc}}n_{\text{o},1}}{n_{\text{o}}^2(n_{\text{o},2} - n_{\text{e},2})L_{\text{crystal}} \sin 2\theta_{\text{pm}}} \quad (4.35)$$

as the full angle around  $\theta_{\text{pm}}$  for half intensity of SHG generation. It results in values of, e.g. a few  $0.1^\circ$  for KTP and  $25^\circ$  for KDP.

From both equations the critical wavelength range for the incident beam can be estimated by the dispersion of the crystal  $\partial n_i / \partial \lambda_{inc}$ . Typical values are 11 nm·cm for KDP and 0.6 nm·cm for KTP.

The temperature has to be constant in the range 25 K·cm for KTP and 4 K·cm in the case of LBO. On the other hand the phase matching can also be tuned by temperature variation for certain crystals. In high-power applications this cannot easily be achieved, because of residual absorption in the material and the resulting heating. Thus for high powers temperature insensitive crystals are suitable.

As with wide angles, a wide spectrum of the incident wave will not be phase matched because of dispersion. Therefore correct material selection as a function of the parameters of the application is most important for high efficiencies.

### *Focusing and Crystal Length*

Optimal focusing has to be chosen as a function of the material and its phase matching acceptance angle, its damage threshold and its temperature sensitivity, which changes the refractive index ellipsoids. The optimal crystal length  $L_{\text{crystal}}$  should be nearly three times the Rayleigh length in the crystal [4.81] which is:

optimal crystal length  $L_{\text{crystal}} = 2.84 \frac{\pi w_0^2}{\lambda_{\text{inc}}}$  (4.37)

for a Gaussian beam neglecting the walk-off. The optimal length can be shorter for short pulses with durations of fs or ps to avoid pulse lengthening from dispersion in the crystal [e.g. 4.82]. In the case of 10 to 100 fs pulses, lengths of 1 to 3 mm may be optimal whereas for ps pulses a few more mm are usually best. For ns pulses or cw radiation several cm may be useful.

### *Type I and Type II Phase Matching*

Because of the large number of material structures there are many different types of phase matching principles. In the given example the incident wave was used as the ordinary beam for the quadratic nonlinear effect and the second harmonic as the extraordinary beam in the material. This type of phase matching is called type I:

**type I phase matching**  $k_{\text{SHG}}(e) = k_{\text{inc}}(o) + k_{\text{inc}}(o).$  (4.38)

The polarization of the incident pump light and of the second harmonic are perpendicular and thus the two wavelengths can be separated using a polarizer instead of the spectrally designed dielectric mirror.

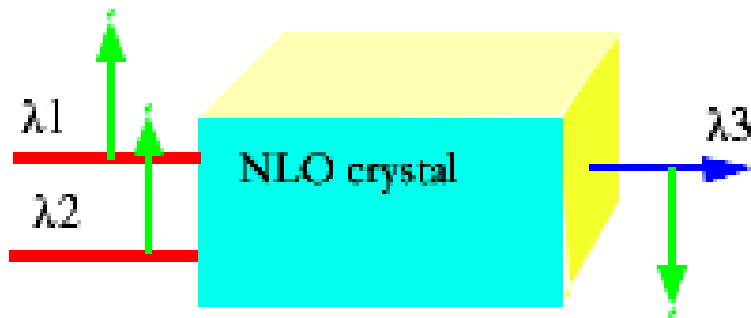
If the incident wave is used as a mixture of an ordinary and an extraordinary beam producing an extraordinary SHG the phase matching is of type II:

**type II phase matching**  $k_{\text{SHG}}(e) = k_{\text{inc}}(o) + k_{\text{inc}}(e)$  (4.39)

resulting, in suitable materials, in about twice the acceptance angle compared to type I.

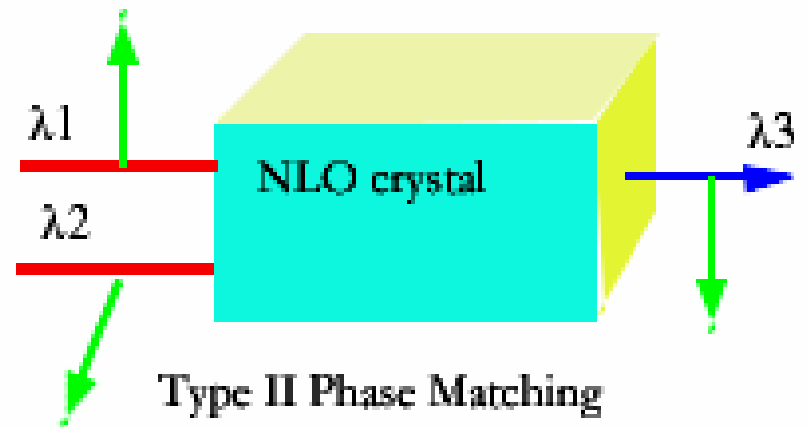
In high-power applications other nonlinear effects and thermal problems may disturb the phase matching and finally limit the conversion efficiency. In particular, the thermally induced birefringence will disturb the process and destroy the good beam quality of the beams. Nevertheless, conversion efficiencies of 80% have been reported

# Type I and Type II Phase Matching



Type I Phase Matching

**Type I**



Type II Phase Matching

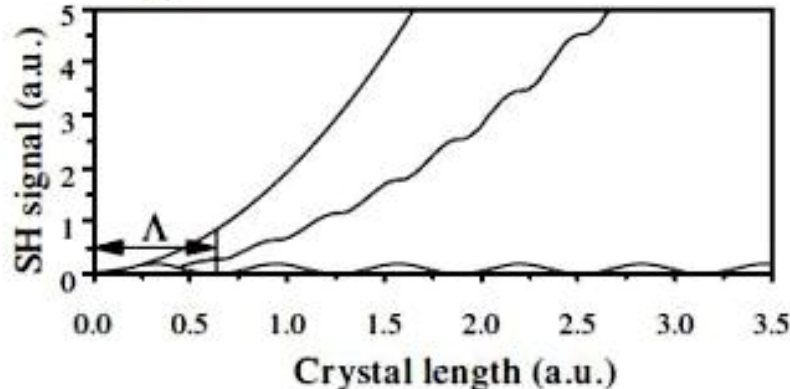
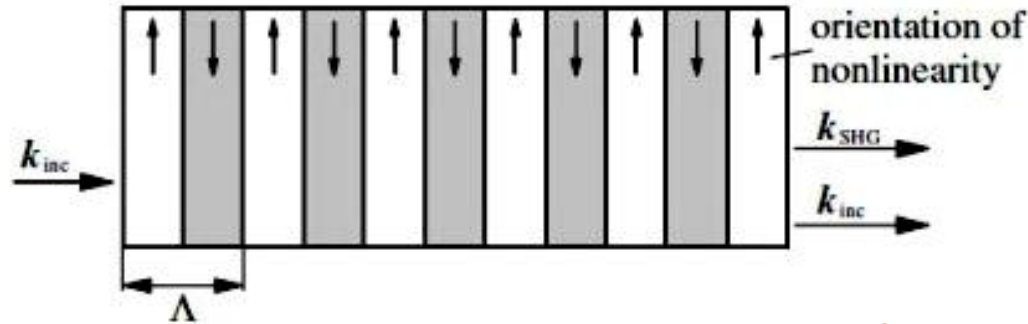
**Type II**

# Quasi-Phase Matching (QPM)

## A way to control phase mismatch

The phase mismatch between the original light wave and the newly generated waves as a result of the different refractive indices for the different wavelengths can also be compensated very nicely by grating structures of the orientation of the nonlinearity of the material . With these structures the phase error can be periodically reset and thus long interaction lengths of several cm can be realized. Especially for low power cw-laser radiation, high efficiencies are reached.

The grating is typically produced by periodic poling of ferroelectrical crystals as shown in Fig. 4.9.



**Common materials include:**

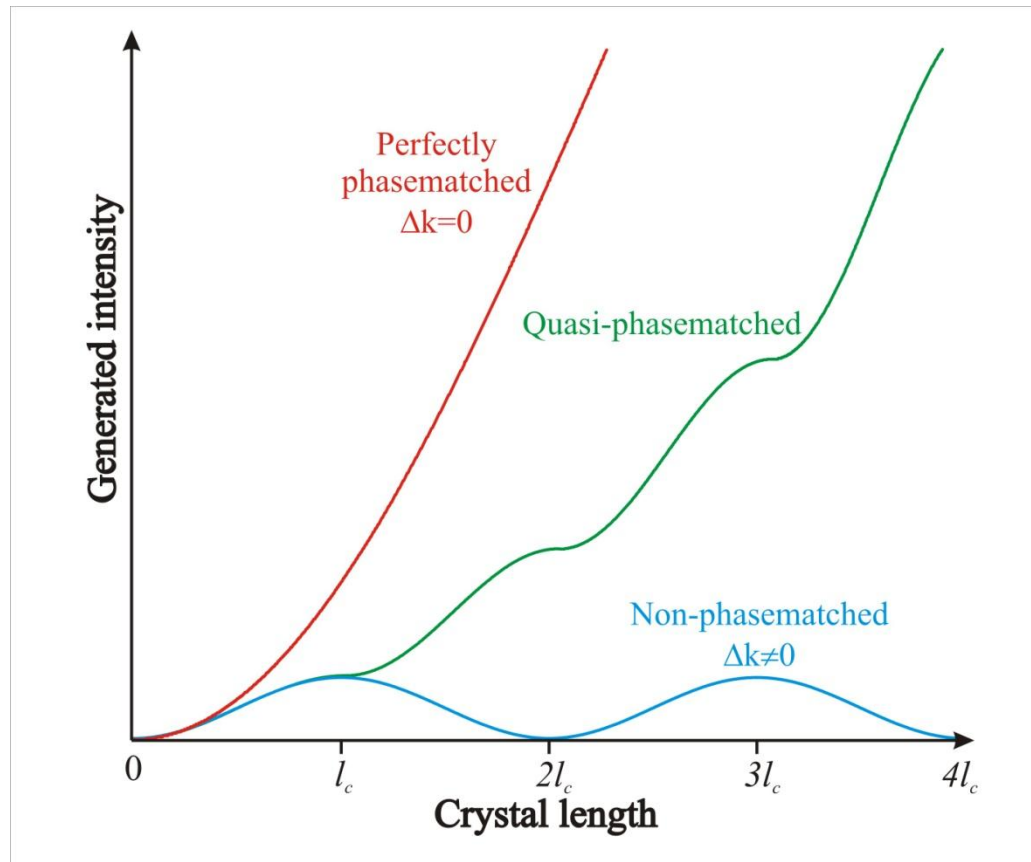
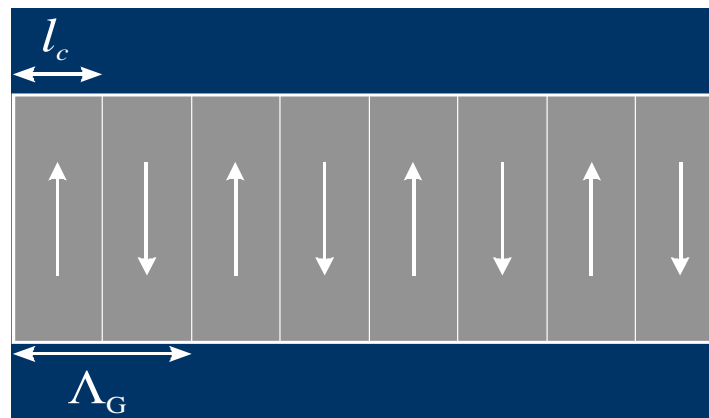
- Lithium Niobate (LN)
- Potassium Titanyl Phosphate (KTP)

# QPM

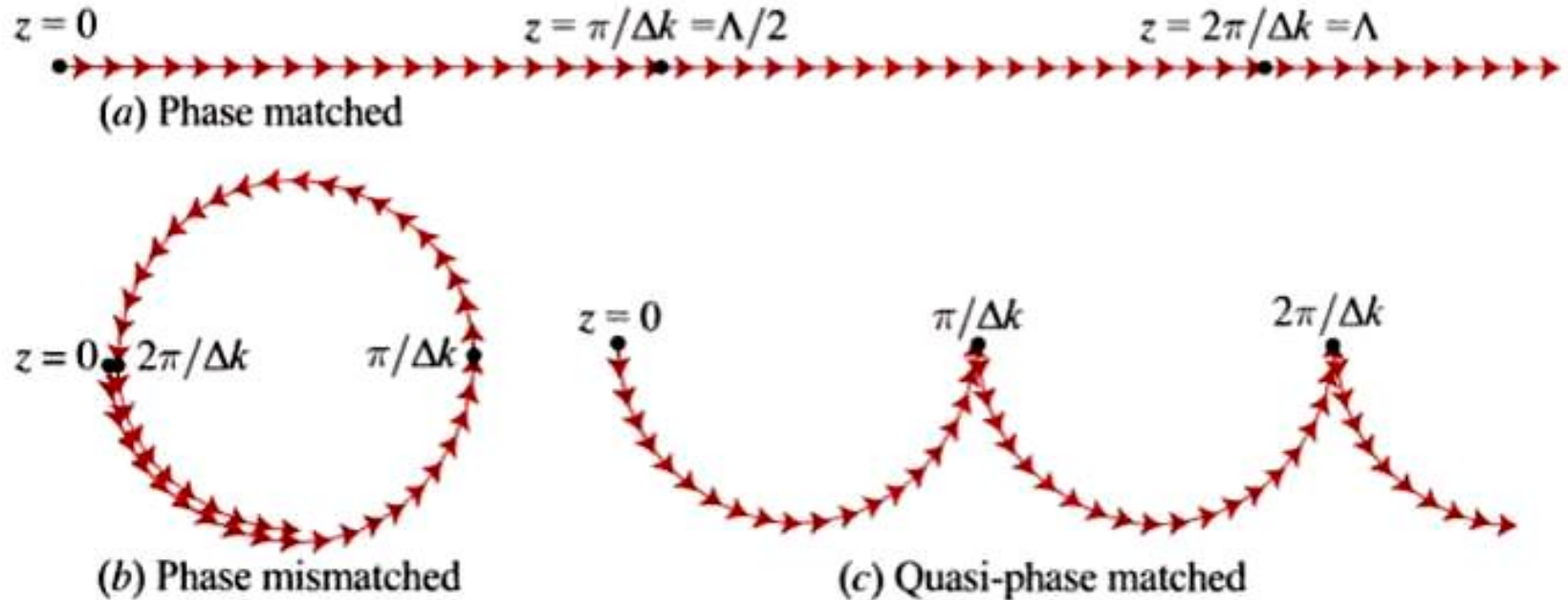
Periodic reversal  
of electric field.

Regular domain  
structure with period

$$\Lambda_G = 2l_c$$



## Quasi-phase matching



**Figure** Phasors of the waves radiated by incremental elements at different positions  $z$  in the nonlinear medium. (a) In the phase-matched case ( $\Delta k = 0$ ) the phasors are all aligned and maximum conversion efficiency is attained. (b) In the presence of a phase mismatch  $\Delta k$ , the phasors are misaligned and the efficiency is significantly reduced. (c) In the quasi-phase matched case, the misaligned phasors are periodically reversed by reversing the sign of the nonlinear coefficient at  $\Lambda/2$  intervals. The conversion efficiency is partially restored.



The grating period  $\Lambda_{\text{qpm}}$  depends on the mismatch of the refractive indices of the incident and the second harmonic waves:

$$\text{qpm period } \Lambda_{\text{qpm}} = m \frac{2\pi}{|k_{\text{inc}} - k_{\text{SHG}}|} = m \frac{2\pi}{|n_{\text{inc}} - n_{\text{SHG}}|} \quad (4.40)$$

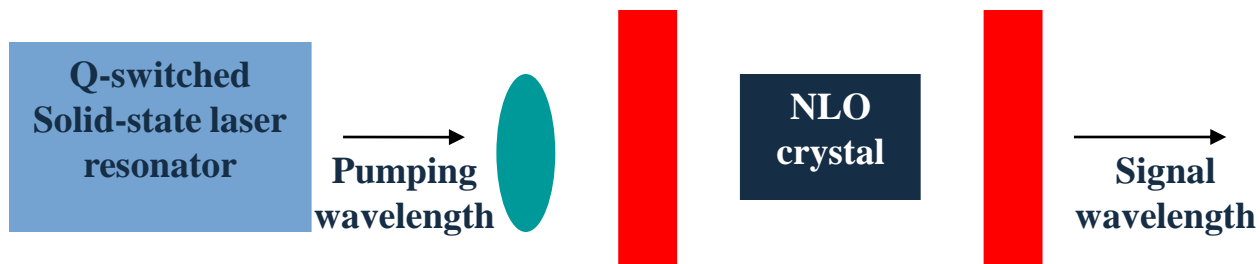
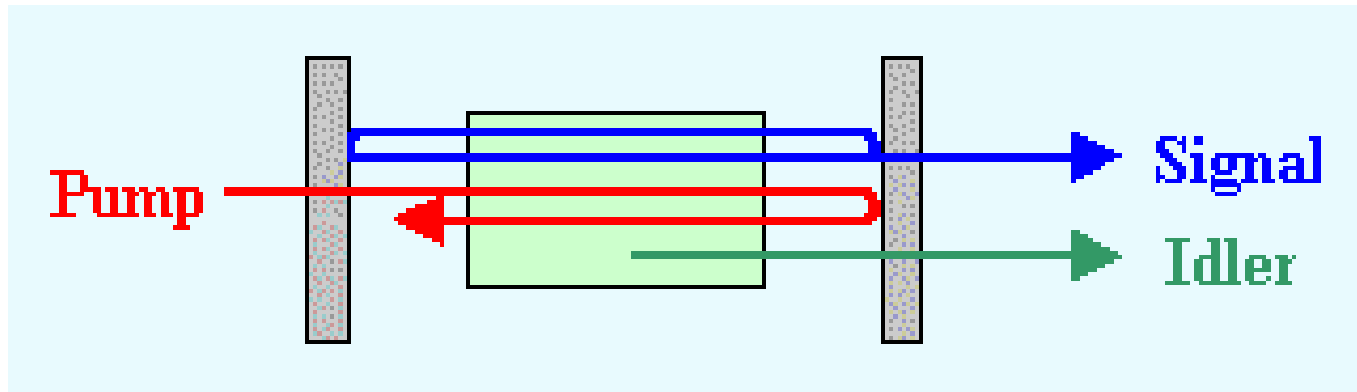
with  $m$  as the order of the periodic poling. A first-order grating ( $m = 1$ ) means that the phase mismatch between the two waves after half the period is  $\pi$ . The optimum case is obtained if the sign of the nonlinearity is reversed every  $\pi/|k_{\text{inc}} - k_{\text{SHG}}|$  second. Typical poling periods are in the range of tens of  $\mu\text{m}$  for conversion of light with wavelengths in the range around are  $\mu\text{m}$ .

The advantage of this quasi-phase matching compared to conventional phase matching is the higher efficiency from crystals such as  $\text{LiNbO}_3$ ,  $\text{LiTaO}_3$  and KTP. They can be applied in optimal directions showing maximum nonlinearity, which cannot be achieved with conventional phase matching. With a 53 mm long KTP crystal conversion efficiencies above 40% were achieved with an input power of 6.4 W from a cw Nd:YAG laser

Periodically poled Mg-doped lithium niobate crystals are currently available in sizes of 0.5 mm thickness, 5 mm width and up to 50 mm length.

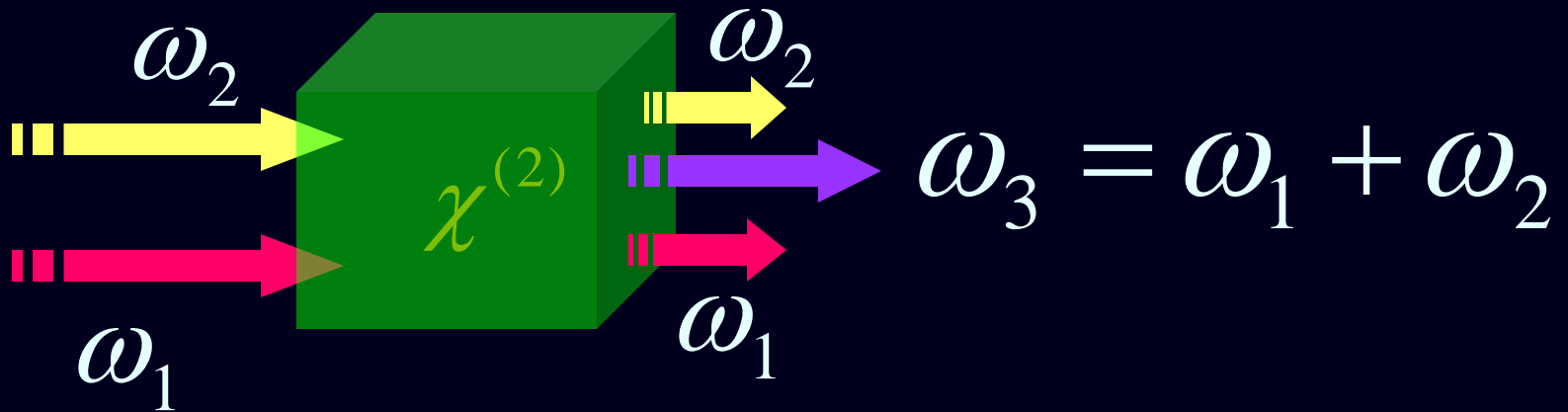
# Optical Parametric Oscillator (OPO)

- “Pump” is converted into two other frequencies
  - Example: 355 nm creates 500 nm and 1224 nm light
- Tuneable laser beam over Visible, IR
  - Change angle of the crystal to tune
- Squeezing

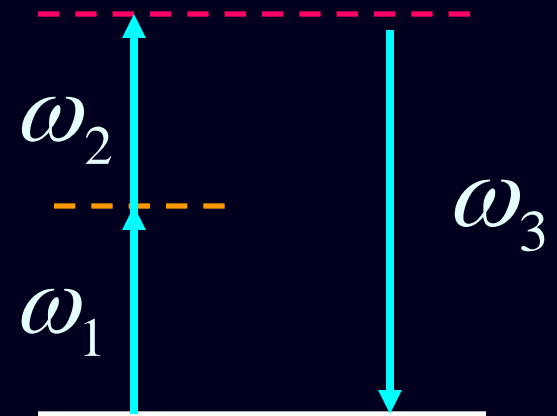


**An OPO with return pump beam**

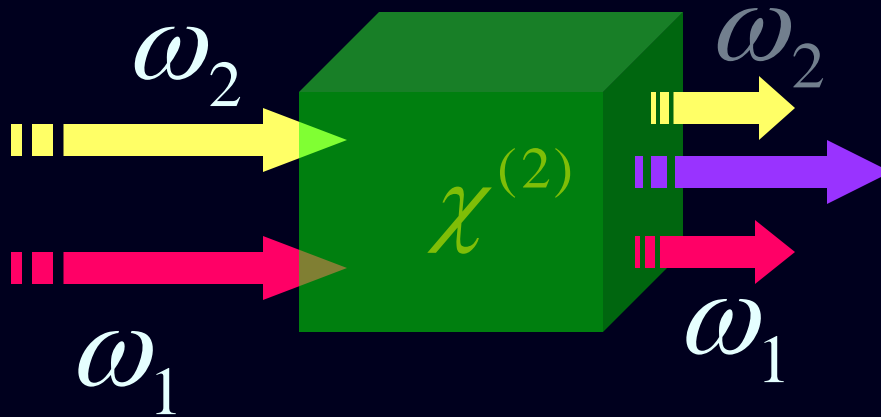
# Sum Frequency Generation SFG



Application:  
**Tunable radiation in the  
UV Spectral region.**



# Difference Frequency Generation DFG

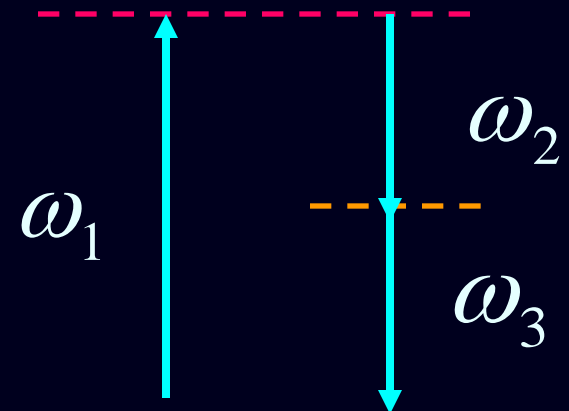


$$\omega_3 = \omega_1 - \omega_2$$

Application:

**The low frequency photon,  $\omega_2$  amplifies in the presence of high frequency beam  $\omega_1$ .**

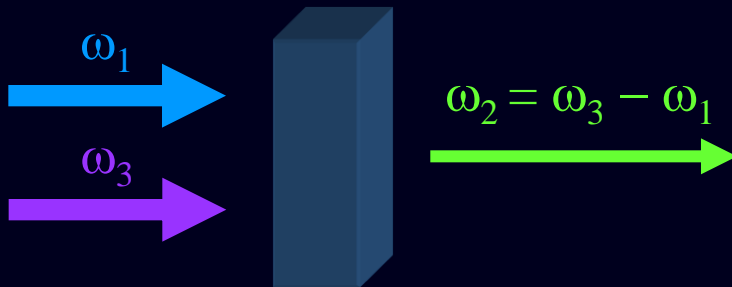
This is known as parametric amplification.



# DFG

## Optical Parametric Generation, Amplification, Oscillation

Difference-frequency generation takes many useful forms



Parametric Down-Conversion  
(Difference-frequency generation)

:

## Frequency Mixing of Two Monochromatic Fields

Via the second-order nonlinear polarization of the material two light waves with different frequencies  $\nu_1$  and  $\nu_2$  can be mixed and new frequencies generated [1]. In addition to the frequencies  $2\nu_1$  and  $2\nu_2$  and  $\nu = 0$  two new waves occur with frequencies:

$$\text{sum frequency } \nu_{\text{sum}} = \nu_1 + \nu_2 \quad (4.41)$$

and

$$\text{difference frequency } \nu_{\text{diff}} = |\nu_1 - \nu_2|. \quad (4.42)$$

These frequencies follow from the electric fields  $E_1$  and  $E_2$ , e.g. of the two collinear waves with parallel polarization combining to give the total field  $E$ :

$$E = E_1 + E_2 = E_{0,1} \cos(2\pi\nu_1 t - k_1 z) + E_{0,2} \cos(2\pi\nu_2 t - k_2 z). \quad (4.43)$$

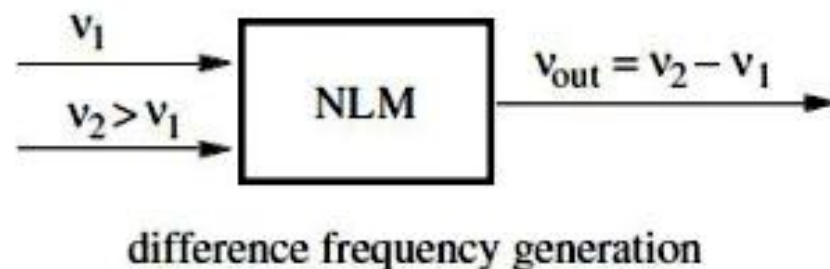
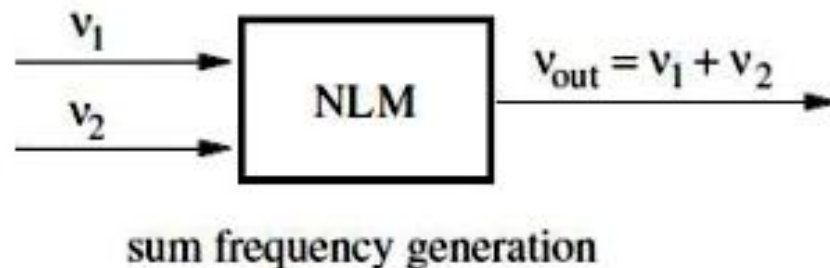
Under the assumption of parallel beams the frequency terms of  $E^2$  can be calculated and the product terms with mixed factors from both fields yield to new frequencies in the polarization formula analog to Eq. (4.18). As a result the above new frequencies are obtained.

The nonlinear polarization in the general form results from:

$$\begin{pmatrix} P_x^{(2)} \\ P_y^{(2)} \\ P_z^{(2)} \end{pmatrix} = 2\varepsilon_0[d] \cdot \begin{pmatrix} E_x(\nu_1)E_x(\nu_2) \\ E_y(\nu_1)E_y(\nu_2) \\ E_z(\nu_1)E_z(\nu_2) \\ E_y(\nu_1)E_z(\nu_2) + E_y(\nu_2)E_z(\nu_1) \\ E_x(\nu_1)E_z(\nu_2) + E_x(\nu_2)E_z(\nu_1) \\ E_x(\nu_1)E_y(\nu_2) + E_x(\nu_2)E_y(\nu_1) \end{pmatrix} \quad (4.44)$$

## Parametric Amplifiers and Oscillators

If two beams with frequencies  $\nu_1$  and  $\nu_2$  are superimposed in a suitable nonlinear material (NLM) with a total intensity reaching the nonlinear regime the additional sum frequency  $\nu_{\text{sum}} = \nu_1 + \nu_2$  or difference frequency  $\nu_{\text{diff}} = |\nu_1 - \nu_2|$  (or both) occur with intensities  $I_{\text{sum}}$  and  $I_{\text{diff}}$



**Fig. 4.10.** Sum and difference frequency generation in a nonlinear material (NLM) with suitable phase matching for the new light



Depending on the phase matching conditions in the nonlinear material, we can select which of these beams with the new or old frequency will be strong after the nonlinear interaction.

In particular, if one of the two incident beams, e.g. a *signal beam* with frequency  $\nu_{\text{signal}}$  is originally weak it can be amplified at the expense of the other strong incident pump light beam with frequency  $\nu_{\text{pump}}$ . Additionally, a new light frequency  $\nu_I$  will occur in a so-called *idler beam* for photon energy conservation (see Fig. 4.11).

If the signal beam frequency is smaller than the pump beam frequency the idler frequency will appear as difference frequency with:

$$\nu_{\text{idler}} = \nu_{\text{pump}} - \nu_{\text{signal}}. \quad (4.46)$$

This arrangement is called an optical parametric amplifier (OPA) and is increasingly being used in photonic applications to generate wavelengths not

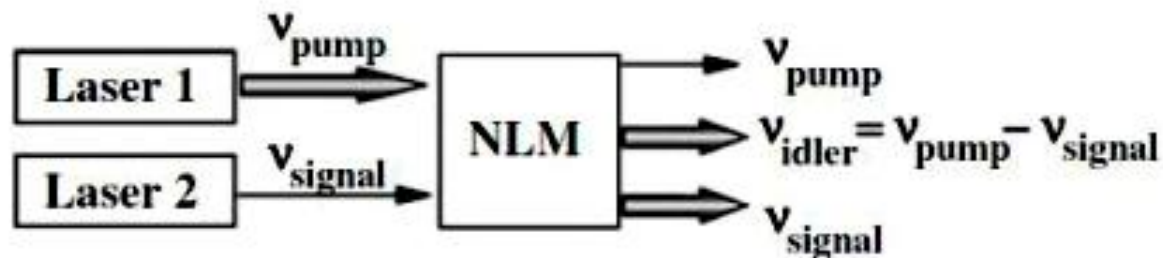


Fig. 4.11. Optical parametric amplifier (OPA): amplification of signal beam at the expense of pump beam and additional generation of idler beam in a nonlinear material (NLM) with suitable phase matching

available from the lasers directly. This process can be applied, e.g. in lithium niobate ( $\text{LiNbO}_3$ ) crystals with laser pump light in the visible range and signal and idler frequencies in the infrared spectral region up to wavelengths of  $7 \mu\text{m}$ . Conversion efficiencies can reach values larger than 50% and thus this method has become quite popular in the generation of new frequencies especially in fs lasers. With these ultra-short pulses very high intensities can be achieved without large thermal loads in the nonlinear material and the damage threshold is increased for these short pulses, too. The seeding of the OPA has the advantage of possible preselection of the idler wavelength and thus better stability. The disadvantage is the necessity of producing this wavelength.

If only one beam with one frequency  $\nu_{\text{pump}}$  is used as incident light in suitable nonlinear materials these photons can be divided into two photons with the same total energy as the pump photon:

$$\nu_{\text{signal}} + \nu_{\text{idler}} = \nu_{\text{pump}} \quad \text{with} \quad \nu_{\text{signal}} > \nu_{\text{idler}} \quad (4.47)$$

with the given convention.

In addition the phase matching condition giving momentum conservation has to be fulfilled. Again we have type I and II behavior as given in the previous chapter, and for a negative uniaxial crystal:

$$\text{type I} \quad k_{\text{pump}}^e = k_{\text{signal}}^o + k_{\text{idler}}^o \quad (4.48)$$

and

$$\text{type II} \quad \left. \begin{aligned} k_{\text{pump}}^e &= k_{\text{signal}}^o + k_{\text{idler}}^e \\ k_{\text{pump}}^e &= k_{\text{signal}}^e + k_{\text{idler}}^o \end{aligned} \right\} \quad (4.49)$$

This arrangement, shown in Fig. 4.12, is called an optical parametric oscillator (OPO).

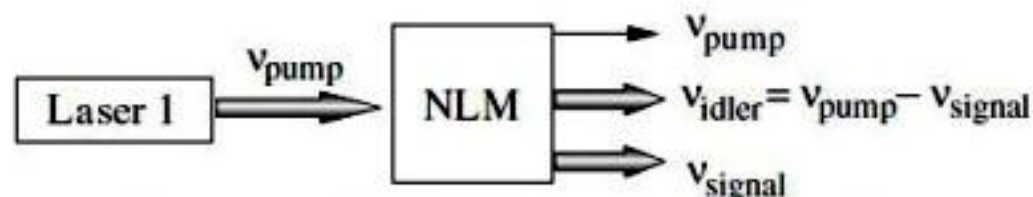


Fig. 4.12. Optical parametric oscillator (OPO): generation of signal and idler beams at the expense of pump beam in a nonlinear material (NLM) with suitable phase matching

Considering energy and momentum conservation leads to spatial distributions of the emission as a function of the crystal, its orientation and the pump parameters.

This nonlinear OPO scheme is increasingly being used in photonics because of the simplicity of the generation of new coherent light beams with new frequencies which are otherwise difficult to generate. Several crystals are useful for OPA and OPO applications as given in Table 4.5.

**Table 4.5.** Tuning ranges for the signal and idler wavelengths  $\lambda$  as a function of the pump wavelength for different useful OPA and OPO crystals [4.156]

$\lambda_{\text{pump}}$ (nm)	laser	crystal	$\lambda_{\text{signal}}, \lambda_{\text{idler}}$ (nm)
1064	Nd:YAG	LiNbO <sub>3</sub>	1400–4400
694	Ruby	LiIO <sub>3</sub>	770–4000
532	Nd:YAG-SHG	KDP	957–1117
355	Nd:YAG-THG	KDP	480–580 960–1160
266	Nd:YAG-FHG	ADP	420–730

Again critical and non-critical phase matching can be realized by angle or temperature tuning of the crystal. As an example the possible wavelengths for the new beam are given as a function of the angle of BBO pumped with 800 nm in Fig. 4.13.

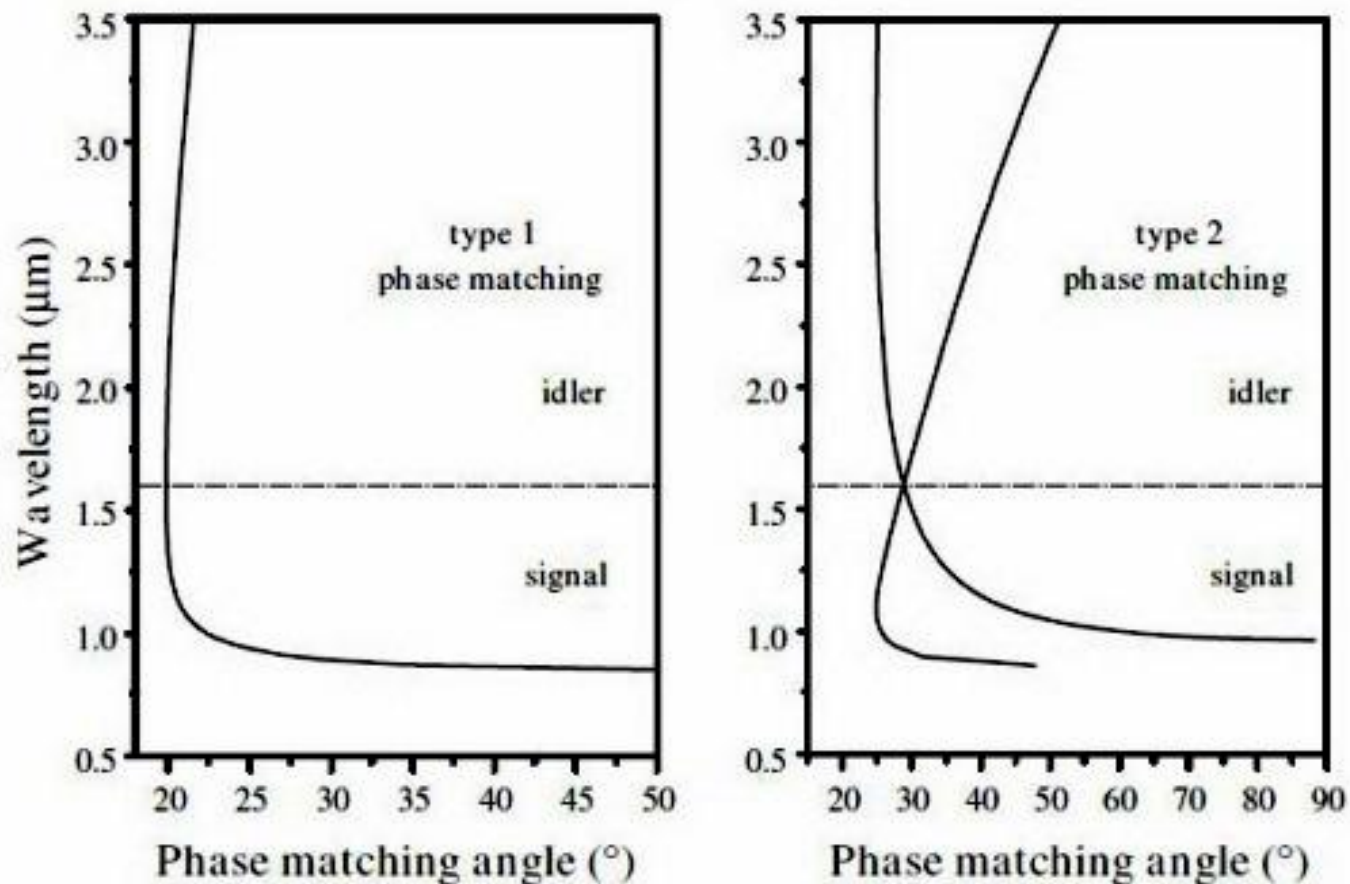


Fig. 4.13. Tuning curve of OPO pumped at 800 nm in a BBO crystal

# Pockel's Effect

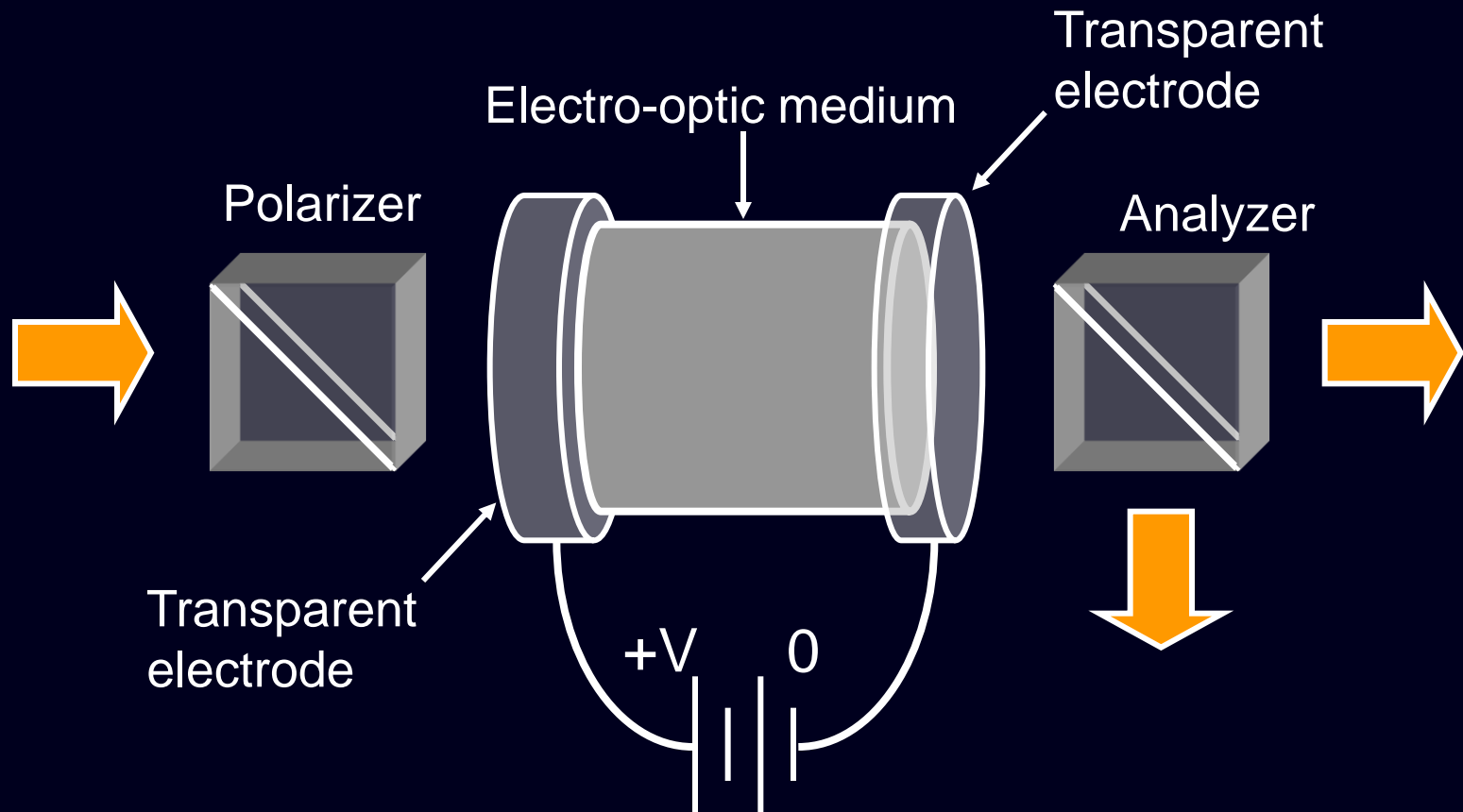
In a NL material, the electric field of the light wave is superimposed on the externally applied field and resulting NL polarization. Hence **it's a *field – field interaction.***

The Pockel's effect rotates the polarization of the incident light as a function of the externally applied electric field.

**$n \propto E$  linear electro-optic effect or Pockels effect**

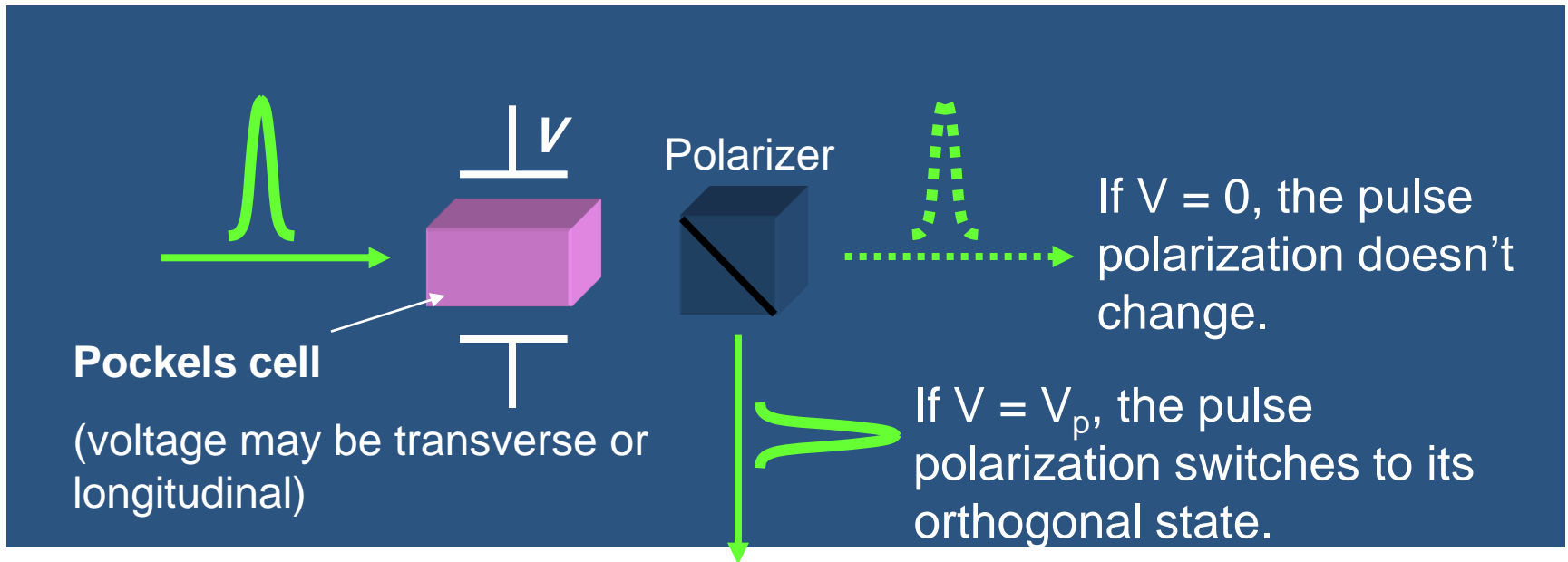
# Pockels' Effect

An electric field can induce birefringence.



The Pockels' effect allows control over the polarization rotation.

Applying a voltage to a crystal changes its refractive indices and introduces birefringence. In a sense, this is sum-frequency generation with a beam of zero frequency (but not zero field). A few kV can turn a crystal into a half- or quarter-wave plate.



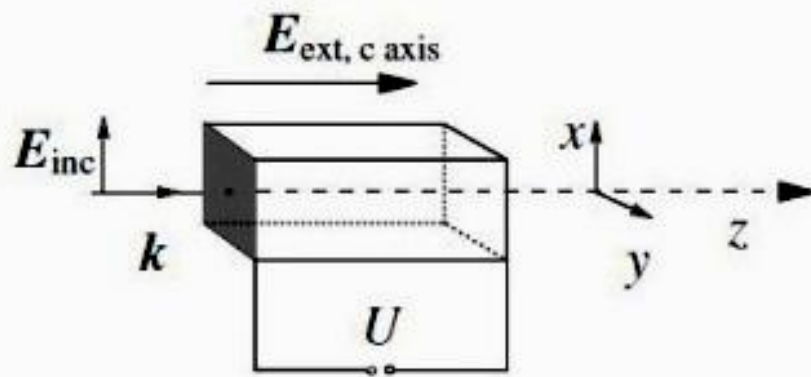
Rapidly switching a Pockels cell allows us to switch a pulse into or out of a laser.



## Pockels' Effect

Besides the opto-optical second-order nonlinear effects, some electro-optical second-order effects are important in photonics [see e.g. 4.231, 4.233, 4.234]. In the nonlinear material the electric field of the light wave is superimposed on the externally applied electric field and will be influenced by the resulting nonlinear polarization.

The Pockels effect rotates the polarization of the incident light as a function of the externally applied electrical field. This can be acquired longitudinally as in Fig. 4.16 or transversally with respect to the wave vector of the light beam. In both cases the anisotropic refractive index ellipsoid (see Sect. 3.12) will couple the two electric fields.



**Fig. 4.16.** Superposition of the electrical light field with the external field via the nonlinear polarization in the  $x$  and  $y$  directions in the Pockels effect

The electric light field of the incident monochromatic planar wave has components  $E_x$  and  $E_y$  as shown in Fig. 4.16 which are given by:

$$E_{\text{inc},x} = E_{\text{inc},x,0} e^{i(2\pi\nu_{\text{inc}}t - k_{\text{inc}}z)} \quad (4.66)$$

and

$$E_{\text{inc},y} = E_{\text{inc},y,0} e^{i(2\pi\nu_{\text{inc}}t - k_{\text{inc}}z)}. \quad (4.67)$$

If a uniaxial crystal is used and its optical axis has the same direction as the external electric field  $\mathbf{E}_{\text{ext}}$  as in Fig. 4.16 the resulting nonlinear polarization can be calculated from a formula similar to (4.19). For this geometry only the two terms with  $E_y \cdot E_z$  and  $E_x \cdot E_z$  will be nonzero. Thus the quadratic nonlinear polarization with the frequency  $\nu_{\text{inc}}$  has the components:

$$P_x(\nu) = 2\varepsilon_0 d_{14}^{(0)} E_{\text{inc},y,0} E_{\text{ext}} \cos(2\pi\nu_{\text{inc}}t - k_{\text{inc}}z) \quad (4.68)$$

and

$$P_y(\nu) = 2\varepsilon_0 d_{25}^{(0)} E_{\text{inc},x,0} E_{\text{ext}} \cos(2\pi\nu_{\text{inc}}t - k_{\text{inc}}z) \quad (4.69)$$

with the condition  $d_{14} = d_{25}$  for, e.g. KDP. These coefficients have different values because of the dispersion for different light frequencies. If this nonlinear

polarization is applied to (4.10) the amplitudes of the electric field of the light wave  $E_{\text{light},x}(z)$  and  $E_{\text{light},y}(z)$  can be calculated to:

$$\frac{\partial E_{\text{light},x}(z)}{\partial z} = \frac{-ik_{\text{inc}}d_{14}^{(0)} E_{\text{inc},y,0} E_{\text{ext}}}{n^2} \quad (4.70)$$

and

$$\frac{\partial E_{\text{light},y}(z)}{\partial z} = \frac{-ik_{\text{inc}}d_{14}^{(0)} E_{\text{inc},x,0} E_{\text{ext}}}{n^2} \quad (4.71)$$

with the ordinary refractive index  $n$  of the crystal for the incident wavelength. These equations can be solved in the case of  $E_{\text{inc},y,0} = 0$  to give the result:

$$E_{\text{light},x}(z) = E_{\text{inc},x,0} \cos \phi_{\text{Pockels}} \quad (4.72)$$

and

$$E_{\text{light},y}(z) = -i E_{\text{inc},x,0} \sin \phi_{\text{Pockels}} \quad (4.73)$$

where the  $-i$  indicates a phase shift by  $90^\circ$  of the fast oscillating light wave. The angle  $\phi_{\text{Pockels}}$  results from:

$$\phi_{\text{Pockels}}(z) = \frac{k_{\text{inc}}d_{14}^{(0)}}{n^2} E_{\text{ext}}z. \quad (4.74)$$

Thus in the Pockels effect the incident linearly polarized light is converted to circular polarization, to linear polarization rotated by 90°, to circular polarization, to linear polarization rotated by 180° and so on (see Fig. 4.17).

Thus for a certain external electric field the crystal works as a quarter-wave plate, producing circular polarized light, and for twice this field as a half-wave plate, producing linear but 90° rotated light. The necessary voltages  $U_i$  are:

$$E_{\text{light},x}(z) = E_{\text{inc},x,0} \cos \Phi_{\text{Pockels}}$$

$$E_{\text{light},y}(z) = -iE_{\text{inc},x,0} \sin \Phi_{\text{Pockels}}$$

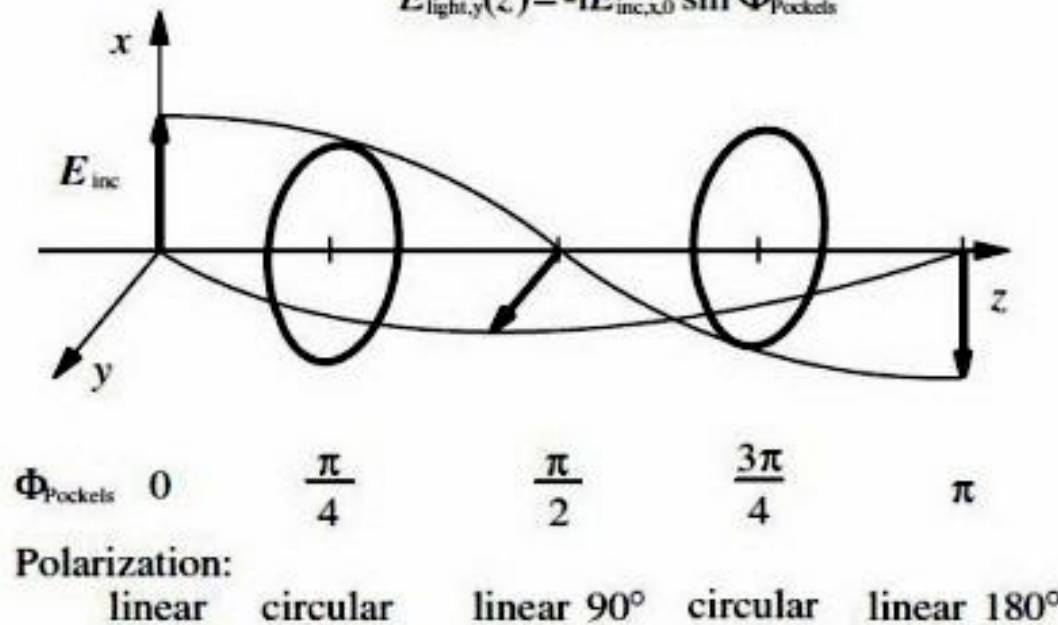


Fig. 4.17. Light polarization in the Pockels effect as a function of the optical path in an optically uniaxial crystal

$$\text{quarter-wave voltage } U_{\lambda/4} = \frac{n\lambda_{\text{inc}}}{4d_{14}^{(0)}} \quad (4.75)$$

and

$$\text{half-wave voltage } U_{\lambda/2} = \frac{n\lambda_{\text{inc}}}{2d_{14}^{(0)}} \quad (4.76)$$

with  $\lambda_{\text{inc}}$  as the wavelength of the incident light outside the crystal.

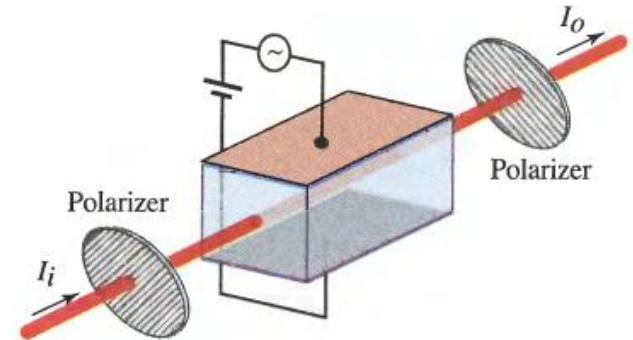
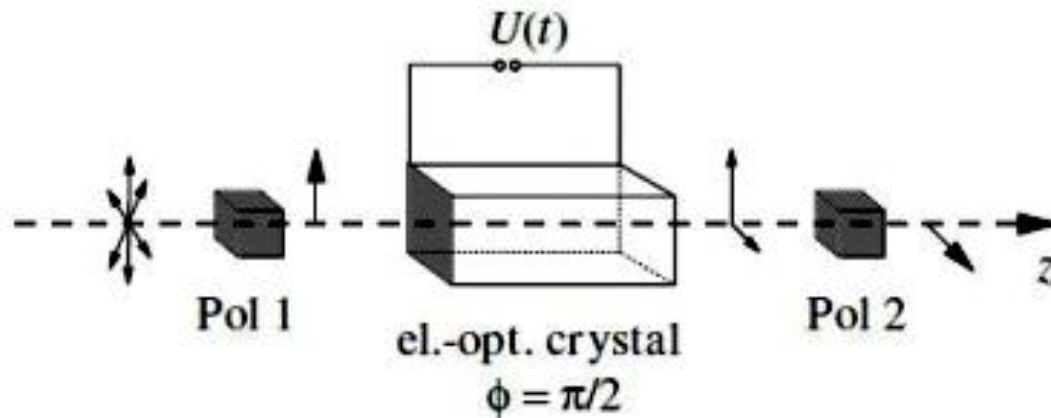
The electro-optical Pockels effect can also be applied with uniaxial crystals which are not arranged along the optical axis as in Fig. 4.16 (p. 200) or with a transversal external electric field. Crystals with less symmetry such as, e.g. a two-axial material, can also be used. In any case the electro-optical effect is based on the deformation of the refractive index ellipsoid in the matter by the electric field. The theoretical description of this second-order nonlinear effect can be given in these more complicated cases similar to the example given above.

The nonlinear coefficients  $d_{ij}^{(0)}$  for electro-optical applications are given for several commonly used crystals in Table 4.6.

**Table 4.6.** Coefficients for electro-optical applications of some widely applied non-linear crystals from [4.233]. Refractive indices are given for the light wavelength and permittivities for temporal constant fields

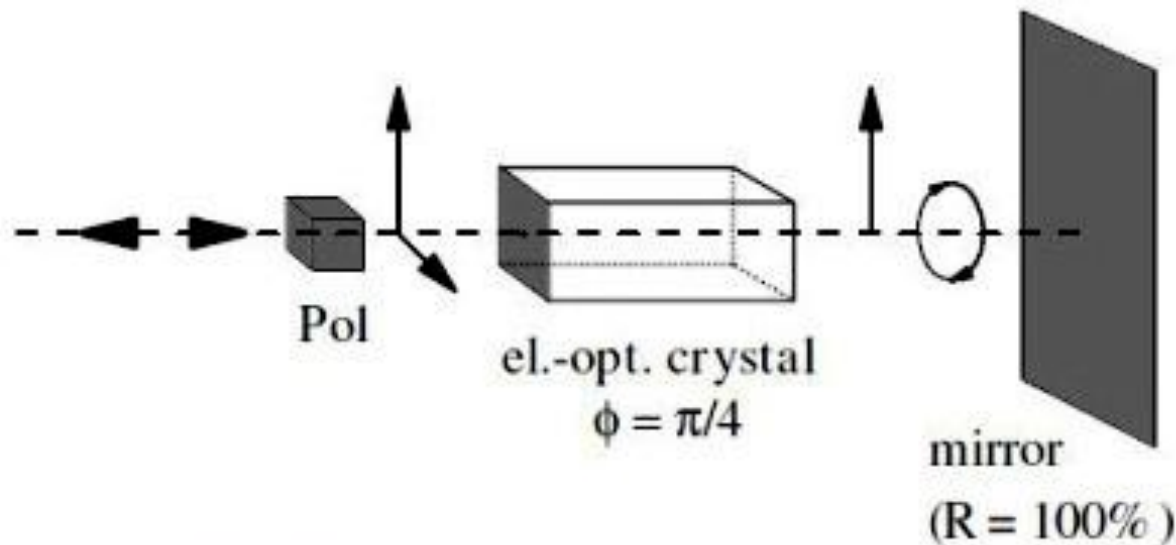
crystal	$\lambda_{\text{inc}}$	$n_o$	$n_{eo}$	$\varepsilon_1 = \varepsilon_2$	$\varepsilon_{eo}$	$d_{ij}$ [ $10^{-12}$ m/V]	$U_{\lambda_{\text{inc}}/4}$
KDP	550 nm	1.51	1.47	42	50	$d_{14} = d_{25} = -8.8$ $d_{36} = -10$	2400 V
ADP	630 nm	1.52	1.48	56	15	$d_{14} = d_{25} = -120$ $d_{36} = -40$	1000 V
LiNbO <sub>3</sub>	1064 nm	2.232	2.156	85	29.5	$d_{15} = d_{24} = -830$ $d_{16} = -d_{21} = d_{22} = -170$ $d_{31} = d_{32} = -242$ $d_{33} = -780$	1500 V

The Pockels effect can be applied for light modulation and optical switching if the Pockels crystal is combined with conventional polarizers as shown in Fig. 4.18.



**Fig. 4.18.** Using the Pockels effect in combination with polarizers as an electro-optical modulator or switch

The incident unpolarized light will be vertically linearly polarized behind the polarizer Pol 1. If no voltage is applied the light polarization will stay vertical and cannot pass the crossed polarizer Pol 2. If the half-wave voltage  $U_{\lambda/2}$  is used the light polarization will be changed to horizontal and the beam can pass Pol 2 undisturbed. Any voltage between 0 and  $U_{\lambda/2}$  will let part of the light intensity transmit. If the incident light beam is well enough linearly polarized Pol 1 is of course not necessary.



**Fig. 4.19.** Pockels effect for Q switching in optical resonators with quarter-wave operation

In this case a 100% reflecting mirror is placed at the position of the second polarizer Pol 2 of Fig. 4.18. If no voltage is applied the whole system will reflect linearly polarized light completely, but if the quarter-wave voltage  $U_{\lambda/4}$  is used the light beam will be circularly polarized behind the nonlinear crystal. After reflection and a second pass through the crystal the light will be horizontally linearly polarized. Then it cannot pass the polarizer Pol 1. Thus the setup works as a mirror with electrically tunable reflectivity between 0



and 100% as a function of the applied voltage at the crystal. This arrangement can be used in laser resonators for modulation of the output. In particular, it is applied for the generation of pulses with ns duration in solid-state lasers via Q switching as already mentioned above. The Pockels cell switch is also applied for the single pulse selection in ps-lasers and in regenerative amplifiers for the selection of the single pulses out of a pulse sequence.

The reaction time of the useful crystals is faster than  $10^{-10}$  s and therefore the switching time is limited by the electric transient times for the necessary voltages. Transient times of less than 1 ns are possible. Because of the isolating properties of the crystals the necessary electrical energy is quite low. It is mostly determined by the required time constant which demands sufficiently low impedance of the electric circuit for recharging of the crystal capacity. This is typically a few pF, e.g. 5 pF for KD\*P which has a quarter-wave voltage of 3.4 kV for 1.06  $\mu\text{m}$  radiation. Present systems allow for ns gates and the repetition rates can be as high as several 100 kHz.

## Electro-Optical Beam Deflection

Another possibility of switching or modulating light based on the electro-optical effect [4.232, 4.233] uses a refractive index step at the border of two differently oriented nonlinear crystals (see Fig. 4.20).

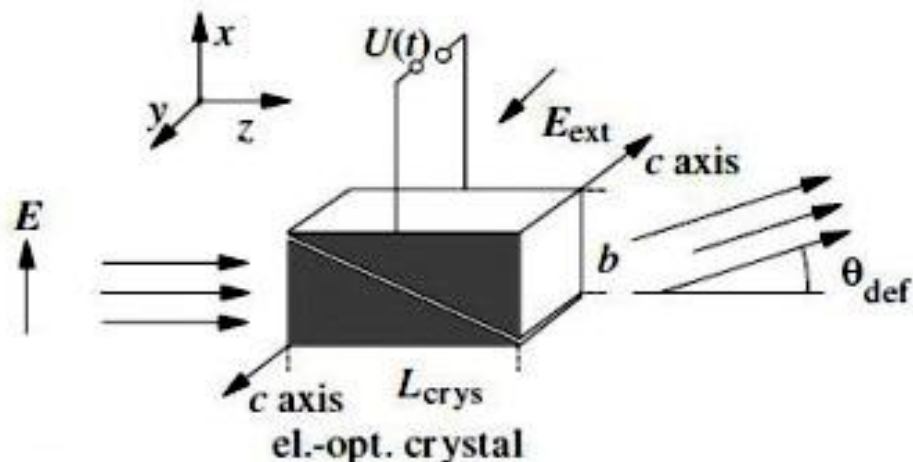


Fig. 4.20. Beam deflection via the electro-optical effect at the border of two antiparallel oriented crystals

The optical axes of the two crystals, e.g. KDP, are antiparallel and perpendicular to the incident light beam. If a voltage is applied along the optical axes of the two crystals the change of the refractive indices will add at the boundary surface. As a result the incident beam will experience different optical paths as a function of its height  $x$  in the vertical direction. The refractive index of crystal 1 will be approximately  $n_1 = n_0 - d_{36}E_{\text{ext}}/2n_0$  and of crystal 2 will be  $n_2 = n_0 + d_{36}E_{\text{ext}}/2n_0$ . Thus the total refractive index difference will be:

$$\Delta n = \frac{1}{n_0} d_{36} E_{\text{ext}}. \quad (4.79)$$

The path length difference for the upper and lower beams in Fig. 4.20 as a consequence of the refractive index difference lead to a deflection of the light beam with angle:

$$\text{deflection angle } \theta_{\text{def}} = \frac{L_{\text{crys}}}{b_{\text{crys}}} \Delta n = -\frac{L_{\text{crys}}}{b_{\text{crys}}} \frac{d_{36}}{n_0} E_{\text{ext}} \quad (4.80)$$

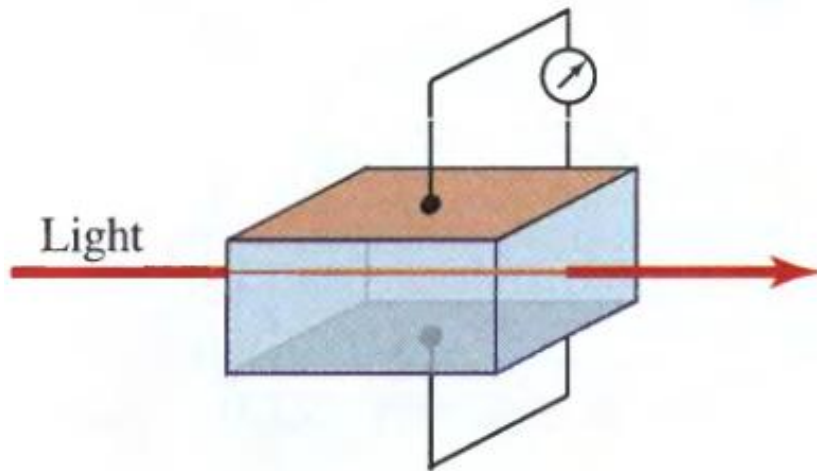
If this deflection is used for scanning diffraction-limited laser beams the resolution  $A$  is given by the ratio of the deflection angle and the divergence angle of the beam  $\theta_{\text{beam}}$ :

$$A = \frac{\theta_{\text{def}}}{\theta_{\text{beam}}} = -\frac{\pi L_{\text{crys}} d_{36}}{2 \lambda_{\text{beam}} n_0} E_{\text{ext}}. \quad (4.81)$$

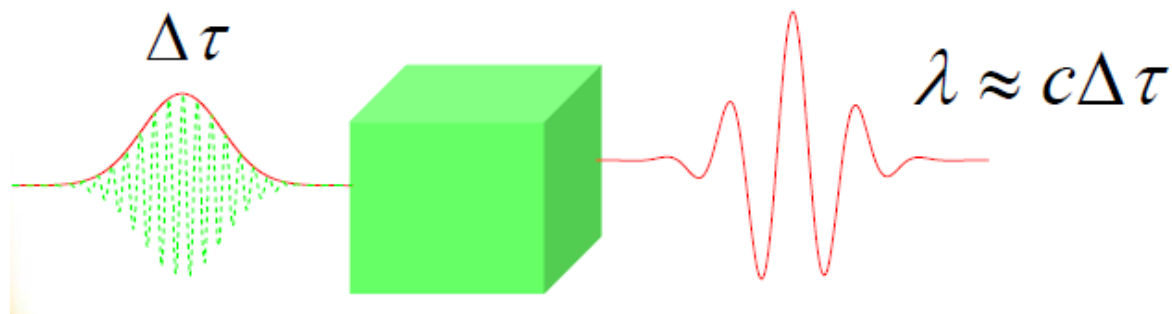
In practical applications angles below  $1^\circ$  can be obtained with crystals of several 10 mm length and thus resolutions of many thousands are possible. The advantage of this type of scanning compared to rotated mirrors or other mechanical scanners is the very high scanning speed of the electro-optical beam deflection.

# Optical Rectification

The component  $P_{NL}(0)$  corresponds to a steady (non-time varying) polarization density that creates a dc potential difference across the plates of a capacitor within which the nonlinear material is placed.



An optical pulse of several MW peak power, may generate a voltage of mV scale.

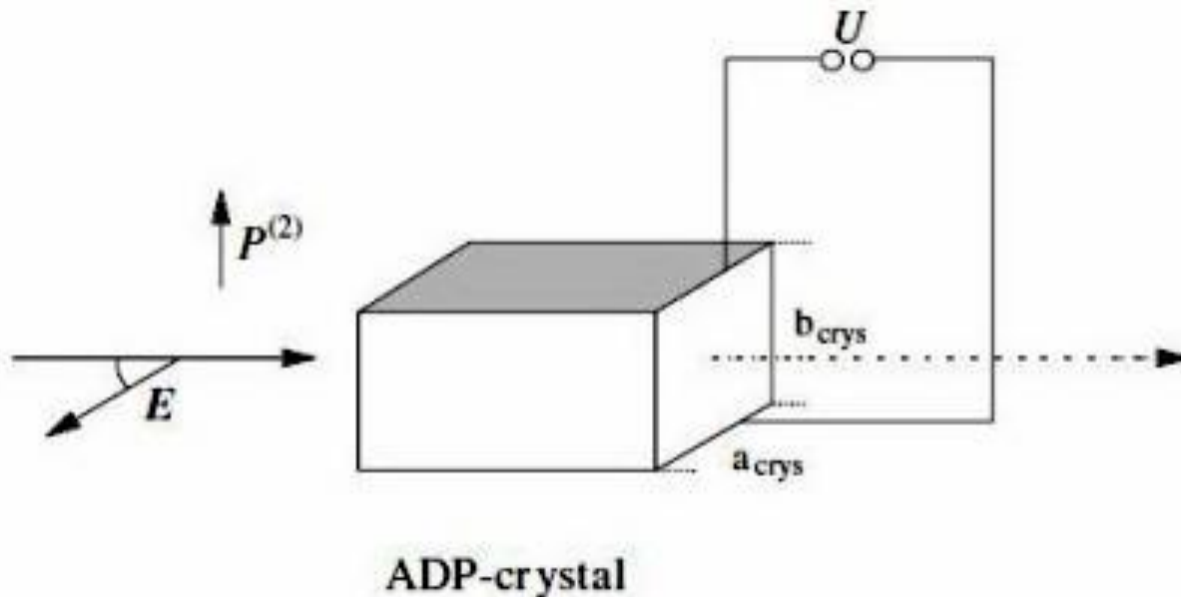


## Optical Rectification

the application of high electric light fields can produce a second-order nonlinear effect with frequency 0, which is a nonoscillating electric field. The physical background for this effect is the displacement of the charges in the temporal average as a consequence of the anharmonic potential.

The calculation of the second-order nonlinear polarization components  $P_i^{(2)}$  for this rectification with  $\nu = 0$  follows by analogy to (4.19) from:

$$\begin{pmatrix} P_x^{(2)}(\nu = 0) \\ P_y^{(2)}(\nu = 0) \\ P_z^{(2)}(\nu = 0) \end{pmatrix} = \varepsilon_0 \begin{pmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{pmatrix} \cdot \begin{pmatrix} E_x E_x^* \\ E_y E_y^* \\ E_z E_z^* \\ 2E_y E_z^* \\ 2E_x E_z^* \\ 2E_x E_y^* \end{pmatrix} \quad (4.82)$$



**Fig. 4.21.** Experimental setup for rectification of light via second-order nonlinear polarization in a suitable nonlinear crystal. The cw voltage can be detected in this example perpendicular to the electric field vector of the light

The cw polarization  $P_x^{(2)}(0)$  in the  $x$  direction which is vertical in Fig. 4.21 can be calculated from (4.82) as:

$$P_x^{(2)}(\nu = 0) = 2\varepsilon_0 d_{14}^{(0)} E_y E_z^* = \varepsilon_0 d_{14} E_{\text{inc}}^2 \quad (4.83)$$

which becomes maximal for  $E_y = E_z = \frac{1}{\sqrt{2}} E_{\text{inc}}$  as assumed in this equation.

This nonlinear polarization generates a charge separation  $Q_{\text{crys}}$  in the crystal of:

$$Q_{\text{crys}} = a_{\text{crys}} L_{\text{crys}} P_x^{(2)}(0) = C_{\text{crys}} U. \quad (4.84)$$

This charge  $Q_{\text{crys}}$  leads to an externally observable voltage  $U$  depending on the capacity of the crystal  $C_{\text{crys}}$ :

$$C_{\text{crys}} = \varepsilon_0 \varepsilon_{\text{crys}} \frac{L_{\text{crys}} a_{\text{crys}}}{b_{\text{crys}}}. \quad (4.85)$$

If finally the light beam power  $P_{\text{beam}}$  as a function of the electric field amplitude  $E_{\text{inc}}$  is introduced:

$$P_{\text{beam}} = \frac{1}{2} \varepsilon_0 c_0 n_{\text{crys}} a_{\text{crys}} b_{\text{crys}} E_{\text{inc}}^2 \quad (4.86)$$

the voltage at the crystal can be expressed as a function of this power by:

$$U = \frac{2d_{14}^{(0)}}{\varepsilon_0 \varepsilon_{\text{crys}} c_0 n_{\text{crys}} a_{\text{crys}}} P_{\text{beam}}. \quad (4.87)$$

In practical cases this voltage is very small. In [4.243] with a 1 MW pulsed ruby laser light source at 694 nm in a  $1 \times 1 \times 2 \text{ cm}^3$  ADP crystal a voltage of 24 mV was observed during the pulse duration of 25 ns.