Nonlinear Interaction of Light and Matter without Absorption

1-1 General classification

1-2 Nonresonant interactions

1-3 Nonlinear polarization of the medium

1-4 Second-order effects

1-4-1 Generation of second harmonic

1-4-2 Phase matching

1-4-3 Frequency mixing of two monochromatic fields

1-4-4 Parametric amplifier and oscillators

1-4-5 Pockel's effect

1-4-6 Electro-optical beam deflection

1-4-7 Optical rectification

1-5 Third-order effects

1-5-1 Generation of the third harmonic

1-5-2 Kerr effect

1-5-3 Self-focusing

1-5-4 Spatial solitons

1-5-5 self-diffraction

1-5-6 Self-focusing in weakly absorbing samples

1-5-7 Self-phase modulation

1-5-8 Generation of temporal solitons: soliton pulses

1-5-9 Stimulated Brillouin Scattering (SBS)

1-5-10 Stimulated Thermal Brillouin Scattering (STBS)

1-5-11 Stimulated Rayleigh (SRLS) and thermal Rayleigh (STRS) scattering

1-5-12 Stimulated Rayleigh Wing Scattering (SRWS)

1-5-13 Stimulated Raman Techniques

1-5-14 Optical Phase Conjugation via Stimulated Scattering

1-6 Higher-order Nonlinear Effects

1-7 Materials for Nonresonant Nonlinear Interaction

1-7-1 Inorganic Crystals

1-7-2 Organic Materials

1-7-3 Liquids

1-7-4 Liquid Crystals

1-7-5 Gases

Ref : Photonics Auther: Ralf Menzel

Nonlinear interaction of light and matter without absorption

- Nonlinear interactions are the basis of photonics
- The direct influence of one light beam or another is not practical with today's sources because of the extremely small photon-photon interaction cross-section

Nonresonant interaction $n = f(I) = f[E(r, \lambda, t, \varphi)]$

- The superposition of light in matter will produce new physical effects in the nonlinear regime.

Linear range: $I_{inc}(r,\lambda,t,\varphi) \rightarrow n \neq f(I_{inc}) \rightarrow I_{out}(r,\lambda,t,\varphi)$

$T \neq f(I_{inc})$

Nonlinear range: $I_{inc}(r,\lambda,t,\varphi) \rightarrow n = f(I_{inc}) \rightarrow I_{out}(r,\lambda,t,\varphi)$

$T = f(I_{inc})$

- All these nonlinear effects first start from the linear interaction.

General Classification

- There are three useful approaches to the description of the nonlinear interaction of light with matter:

Nonlinear polarization: $P_{nl}[E(r,\lambda,t,\varphi)]$

Density matrix formalism: $\rho_{ij}[E(r,\lambda,t,\varphi)]$

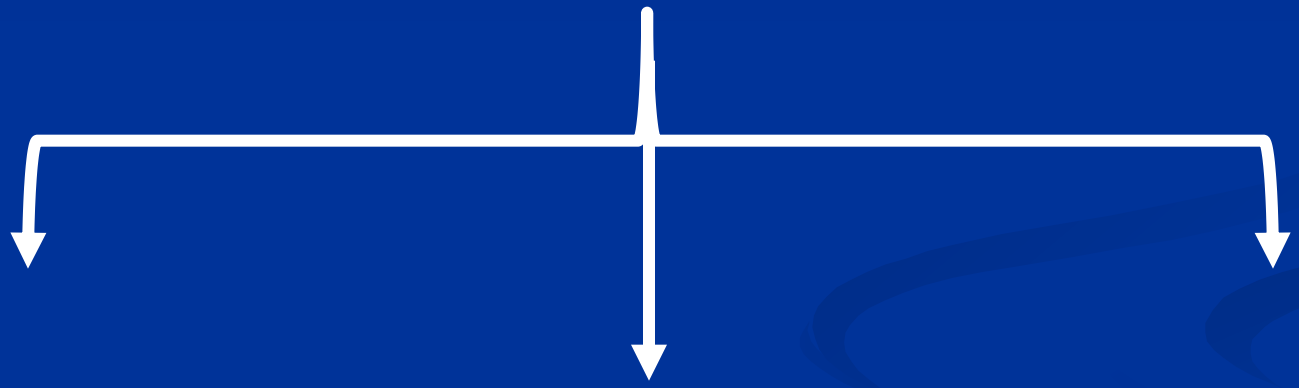
Rate equations : $N_i[I(r,\lambda,t,\varphi)]$

Types of nonlinear optical interactions of light with matter

Matter	Light	Useful description	Example
Nonresonant (transparent)	Incoherent	Maxwell's equations	Self-focusing of light
Nonresonant (transparent)	Coherent	Maxwell's equations	Frequency conversion in crystals
Resonant (absorbing)	Incoherent	Rate equations	Passive Q-switching
Resonant (absorbing)	Coherent	Density matrix formalism	Self induced transparency

Nonresonant interactions

Nonresonant interactions are useful for



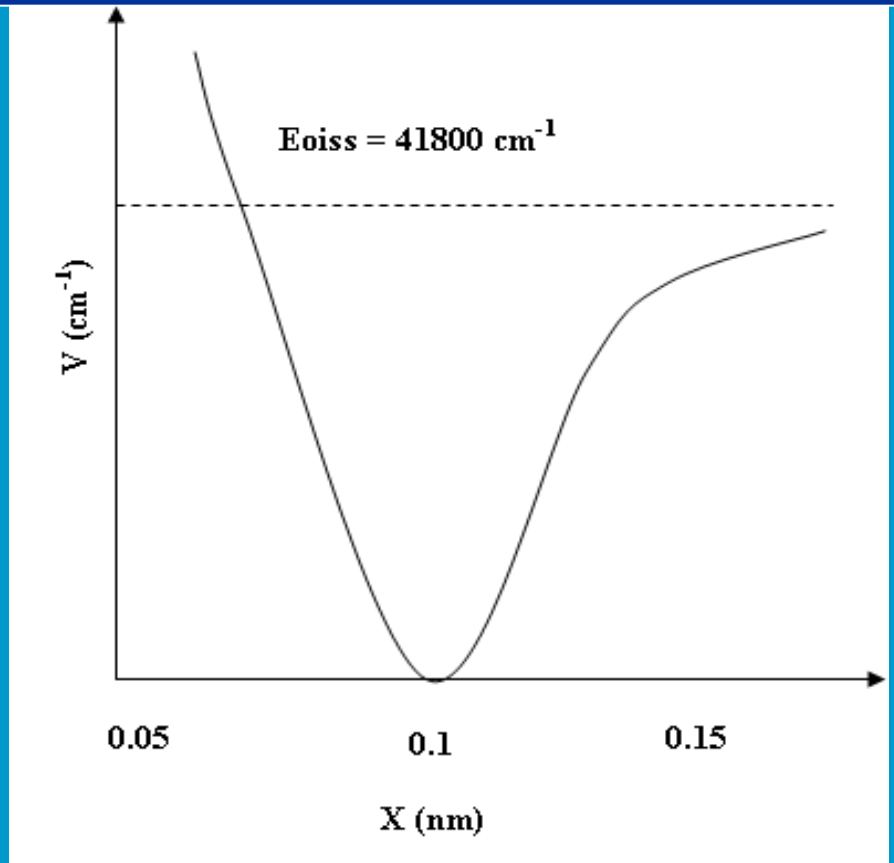
Optical phase conjugating

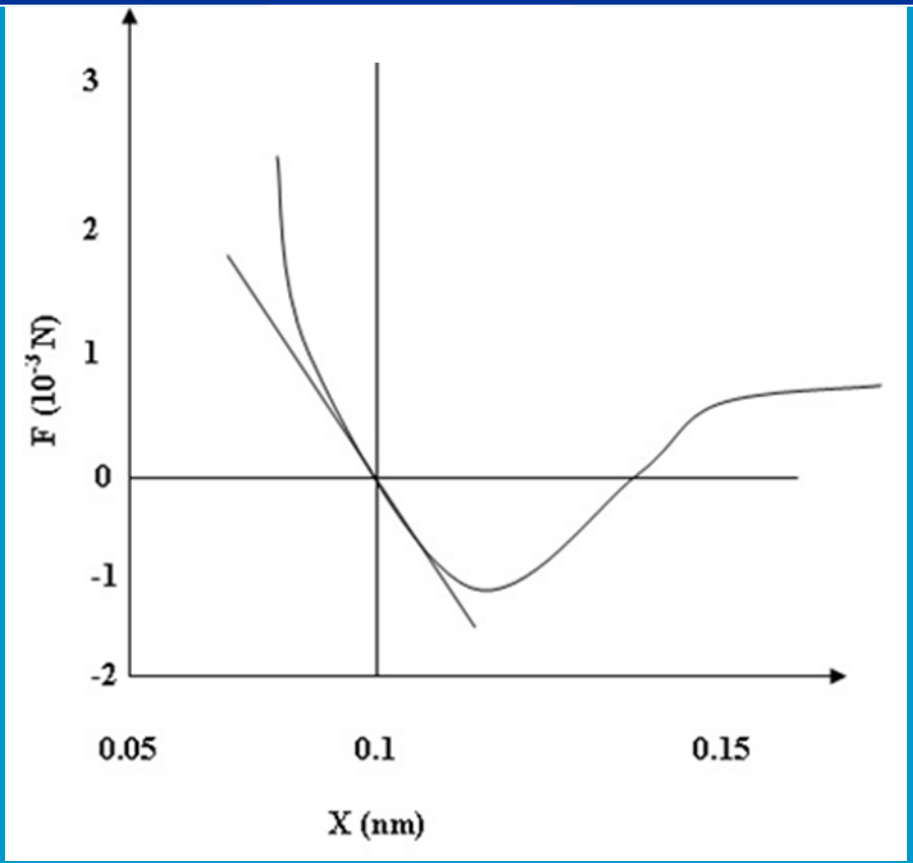
Wave mixing

Wavelength conversion of laser light

They can be applied for high average powers with high frequencies because of the negligible absorption almost no energy will be stored in the material

- The nonresonant nonlinear interaction may be understood as the reaction of electric dipoles built by electrons and the positivity charged atomic cores in the matter under the influence of high electric light fields.





- E for conventional light sources $1 Vcm^{-1}$ ⟶ Elongations $< 10^{-16}m$

Atomic or molecular diameters of 10^{-10}-$10^{-7}m$

- E for laser radiation $10^4 Vcm^{-1} \approx MWcm^{-2}$
- These are second - order effects, third-order effect, higher - order effects.

Nonlinear Polarization of the Medium

- Based on Maxwell's equations, the reaction of the matter under the influence of the electric field of the light E(r,t) can be described by the polarization P(r,t).

Linear polarization: $P_1(r,t) = \varepsilon_o \chi\ E_1(r,t)$

$$P(r,t) = \varepsilon_o \chi^{(1)} E\ (r,t) + \varepsilon_o \chi^{(2)} E^2(r,t) + \varepsilon_o \chi^{(3)} E^3(r,t) + ...$$

$$= P_{lin}(r,t) + P_{nl}(r,t)$$

Nonlinear polarization:

$$P_{nl}(r,t) = \varepsilon_o \chi^{(2)} E^{(2)}(r,t) + \varepsilon_o \chi^{(3)} E^{(3)}(r,t) + ...$$

- Using this nonlinear polarization the equation for the electric field of the light wave as:

$$\Delta E - \mu_o \varepsilon_o \frac{\partial^2 E}{\partial t^2} - grad\ div E = \mu_o \frac{\partial^2 P_{nl}}{\partial t^2}$$

For plane wave

(div E=0)

$$\Delta E - \mu_o \varepsilon_o \frac{\partial^2 E}{\partial t^2} = \mu_o \frac{\partial^2 P_{nl}}{\partial t^2}$$

Now

$$E_{gen}(z,t) = E_{gen,o}(z)\cos(2\pi \upsilon_{gen} t - k_{gen} z)$$

Using the assumption of slowly varying amplitudes (SVA) with

SVA approximation $\dfrac{\partial E_{gen,o}}{\partial z} << k_{gen} E_{gen,o}$

- The differential equation for the amplitude of the new light wave produced by the nonlinear polarization can be written as:-

$$\frac{\partial E_{gen,o}}{\partial z} = i\frac{\mu_o}{2k_{gen}}\frac{\partial^2 P_{nl}(E)}{\partial t^2}e^{-i(2\pi\nu_{gen}t - k_{gen}z)} \quad **$$

- The nonlinear susceptibilities χ^m and derived coefficients are in general functions of material and the frequencies of the applied light wave:

$$\chi^m = f(\upsilon_p, k_p, matter)$$

$$P_{nl,real} = \frac{1}{2}(P_{nl} + P^*_{nl})$$

- If nonresonant interaction are considered and the susceptibility is replaced by $n = \sqrt{1+x}$, the nonlinear wave equation can be simplified:

$$\nabla^2 E - \frac{1}{c^2}\frac{\partial^2}{\partial\, t^2}[(n_o + \Delta n_{nl})^2 E] = 0$$

- If the field E is assumed to be a monochromatic cylindrically symmetric beam propagating along the Z axis then:

$$E = E_o(r, z, \zeta)\ e^{i(2\pi\nu t - kz)}$$

- With time variable $\zeta = t - \frac{z n_o}{c_o}$ considering the propagation of the beam with the velocity c_o/n_o in the matter .
- With the amplitude varying slowly compared to, the nonlinear wave equation can be simplified:

$$\left(\frac{\partial^2}{\partial\, r^2} + i2k\frac{\partial}{\partial\, z}\right)E_o(r,z,\zeta) + 2k^2\left(\frac{\Delta n_{nl}}{n_o}\right)E_o(r,z,\zeta) = 0$$

- In this simple form the nonlinear wave equation may be analyzed for changes in the amplitude and the phase of the beam in the nonlinear medium.
- For complicated functions: $\Delta n_{nl} = f[E_o(r,z,\zeta)]$

Second-order Effects

- The second _order nonlinear susceptibility $\chi^{(2)}$ of used crystals reaches values of up to about

$$I = 0.1\text{-}1\ GWcm^{-2}$$

- These values ≈ the change threshold of optical surfaces, thus new materials are developed.
- In second-order nonlinear optical effects two different light waves .

$$P^{(2)} = \varepsilon_o \chi^{(2)}\ E_1 E_2$$

- In Cartesian coordinates x , y , z described by the indices m , p , q the spatial components Px , Py , Pz of the vector $P^{(2)}$ can be calculated from :

$$P_m^{(2)} = \varepsilon_o \sum_{p,q} \chi^{(2)}{}_{mqp} E_p E_q$$

Generation of the Second Harmonic

- In the case of two equal monochromatic light waves with same polarization, υ_{inc} and direction K_{inc} which can be two shares of the same wave.

$$P^{(2)} = \varepsilon_o \chi^{(2)} E_1 E_2 = \varepsilon_o \chi^{(2)} E^2$$
$$= \varepsilon_o \chi^{(2)} \{E_o(k,\varphi)\cos(2\pi\upsilon_{inc}t)\}^2$$
$$= P^{(2)}(0) + P^{(2)}(2\upsilon_{inc})$$

- The energy efficiency of SHG is < 1

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

$$\begin{pmatrix} P_x^{(2)}(2\upsilon_{inc}) \\ P_y^{(2)}(2\upsilon_{inc}) \\ P_z^{(2)}(2\upsilon_{inc}) \end{pmatrix} = \varepsilon_o \begin{pmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{pmatrix} \begin{pmatrix} E_x^{\,2} \\ E_y^{\,2} \\ E_z^{\,2} \\ 2E_yE_z \\ 2E_xE_z \\ 2E_xE_y \end{pmatrix}$$

- These matter parameters drs are functions of the applied frequencies because of the symmetry of commonly used crystals this number is reduced further.
- For efficient generation of SHG phase matching conditions have to be fulfilled.

Phase Matching

- The generation of frequencies via nonlinear polarization in matter is more efficient the better the incident light waves and the newly generated waves are in suitable phase over the interaction length.
- Phase matching is a suitable orientation of the crystal w.r.t the light beam.

Phase Matching of SHG

- The increasing amplitude of the SHG wave can be calculated from:

$$\frac{\partial E_{gen,o}}{\partial z} = i\frac{\mu_o}{2k_{gen}}\frac{\partial^2 P_{nl}(E)}{\partial t^2}e^{-i(2\pi\nu_{gen}t - k_{gen}z)} *$$

- The nonlinear polarization for this wave is:

$$P_{nl}^{(2)}(2\upsilon_{inc}) = \frac{1}{2}\varepsilon_o \chi^{(2)} E^2{}_{inc,o}(z) + e^{i2(2\pi\nu_{inc}t - k_{inc}z)}$$

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

$$\frac{\partial\ E_{SHG,}(z)}{\partial\ z} = -i\frac{k_{SHG}\chi^{(2)}}{4n^2{}_{SHG}}\ E_{inc}^2(z)\ e^{-i\Delta kz}$$

- Which cannot easily be solved.

- Assuming an undepleted incident wave, it describes an oscillation of the amplitude of the electric field of the generated SH light with z as a function of $\Delta k = |k_{SHG} - k_{inc}|$
- This oscillation of the intensity of the SHG light can be calculated as:

$$I_{SHG}(z) = I_{inc}^2 \frac{8\pi^2 d^2}{\varepsilon_0 c_o \lambda_{inc}^2 n_{inc}^2 n_{SHG}} \left[\frac{\sin(\frac{\Delta k z}{2})}{(\frac{\Delta k}{2})} \right]^2$$

- Where d as the relevant matrix element of the d matrix of mater .
- 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 Zip.

$$Zip = \frac{\pi}{\Delta k} m = \frac{\lambda_{inc}}{4(n_{SHG} - n_{inc})} m$$

- And out of phase after a path length Zoop of :

$$Zoop = \frac{2\pi}{\Delta k} m = \frac{\lambda_{inc}}{2(n_{SHG} - n_{inc})} m$$

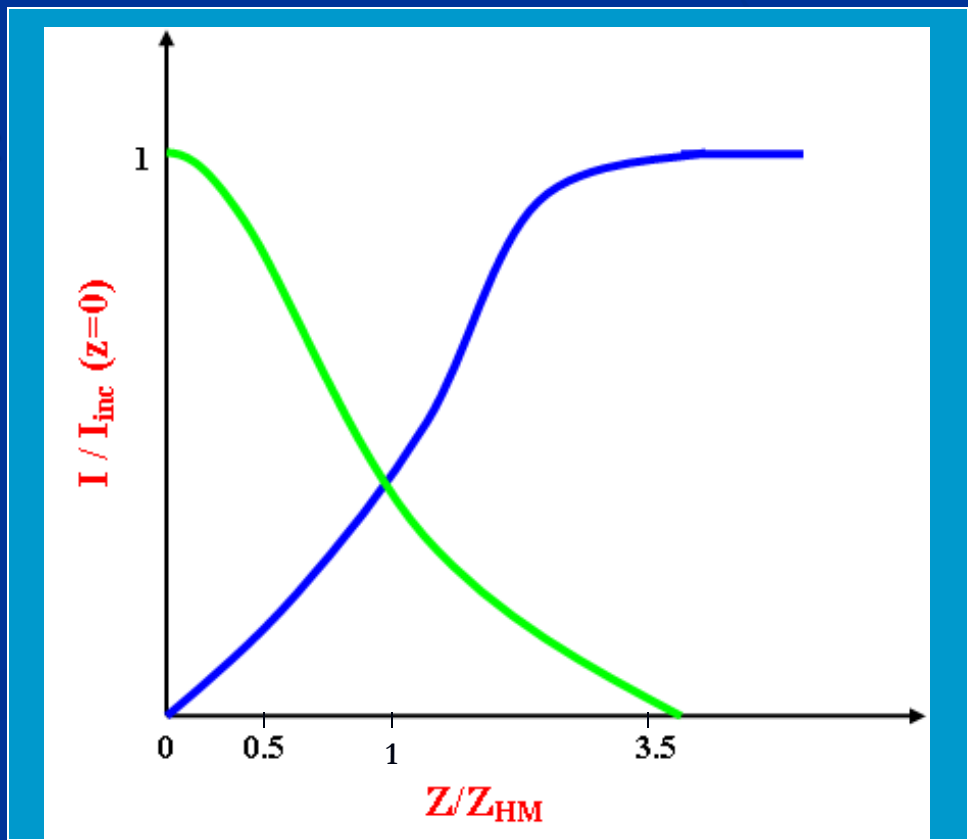
- For energy conservation the amplitude of the incident wave will oscillate too. This problem can be avoided by choosing:

$$Phase\ matching = \left| k_{SHG} - k_{inc} \right| = 0$$

- Applying a suitable crystal orientation (see examples)

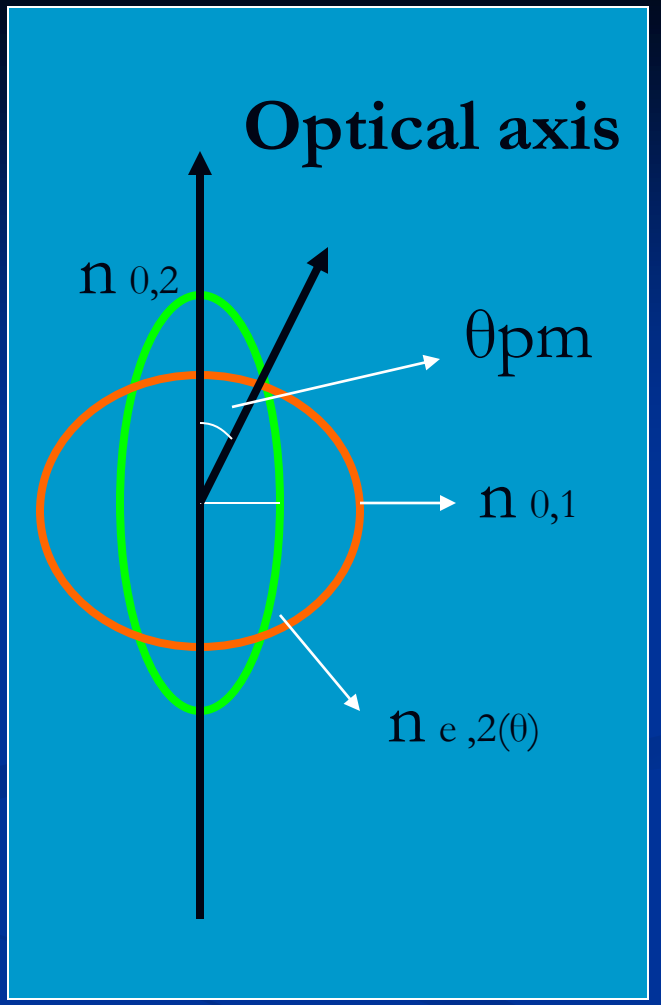
- The incident intensity Iinc will decrease with z in non-absorbing materials by:

$$I_{inc}(z) = I_{inc}(z=0) - I_{SHG}(z)$$



- Phase matching can be achieved with anisotropic material like crystals and is based on the birefringence in these materials .
- The orientation and the temperature of the crystal has to be chosen for equal refractive indices for the incident and generated waves .
- Often P of the incident and generated waves are perpendicular as a result of $\mathbf{P}_{nl}$ in the crystal and thus different refractive index ellipsoids are available for phase matching. For example, in the case of SHG
- It is possible to match n for the incident fundamental wave as an ordinary beam with the SH wave as an extraordinary beam even in a uniaxial crystal.

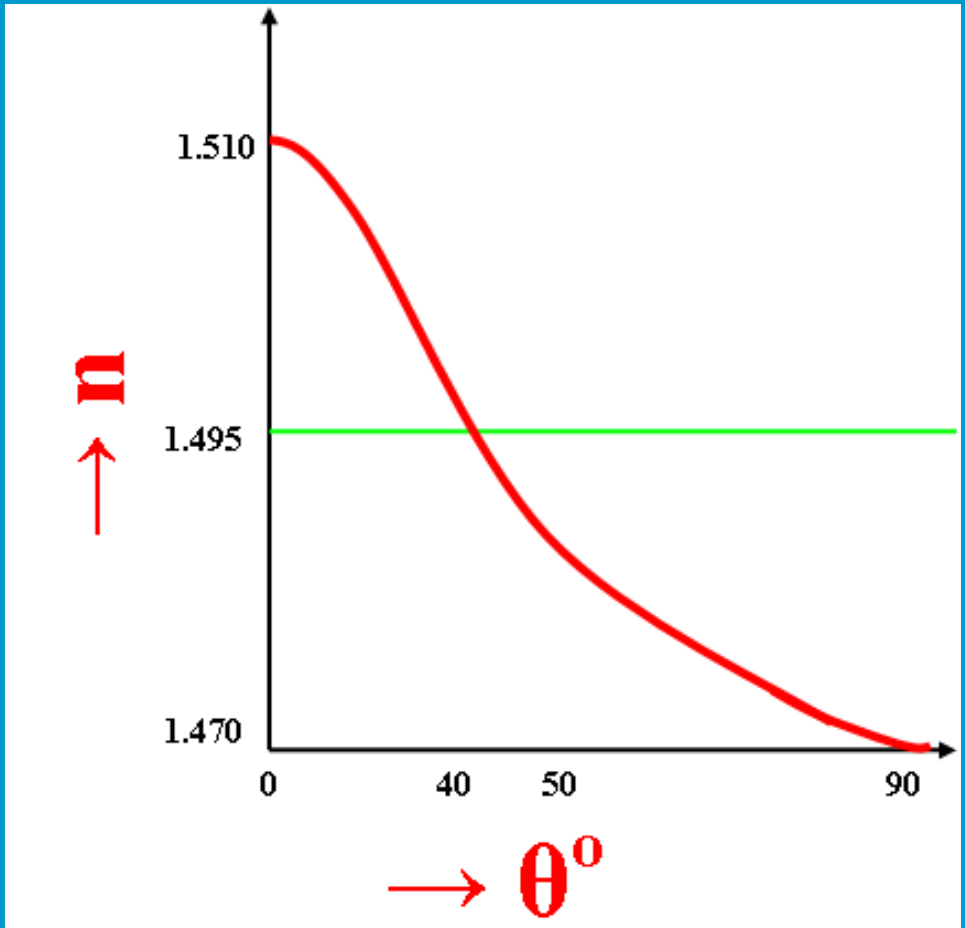
- If an intersection of the two refractive index surfaces for the two waves exists, then phase matching can be achieved.
- In this example, the phase matching angle θpm follows the definitions:



$$\text{phase matching angle } Sin^2\,\theta_{pm} = \frac{\frac{1}{n_{0,1}^2} - \frac{1}{n_{0,2}^2}}{\frac{1}{n_{e,2}^2} - \frac{1}{n_{0,2}^2}}$$

- And reaches of 40 – 60° in crystals KDP or KTP

- The two refractive indices for KDP



ne (λ=0.532 μm,θ)

no (λ=1.064 μm)

Dispersion of crystals: Sellmeier coefficients

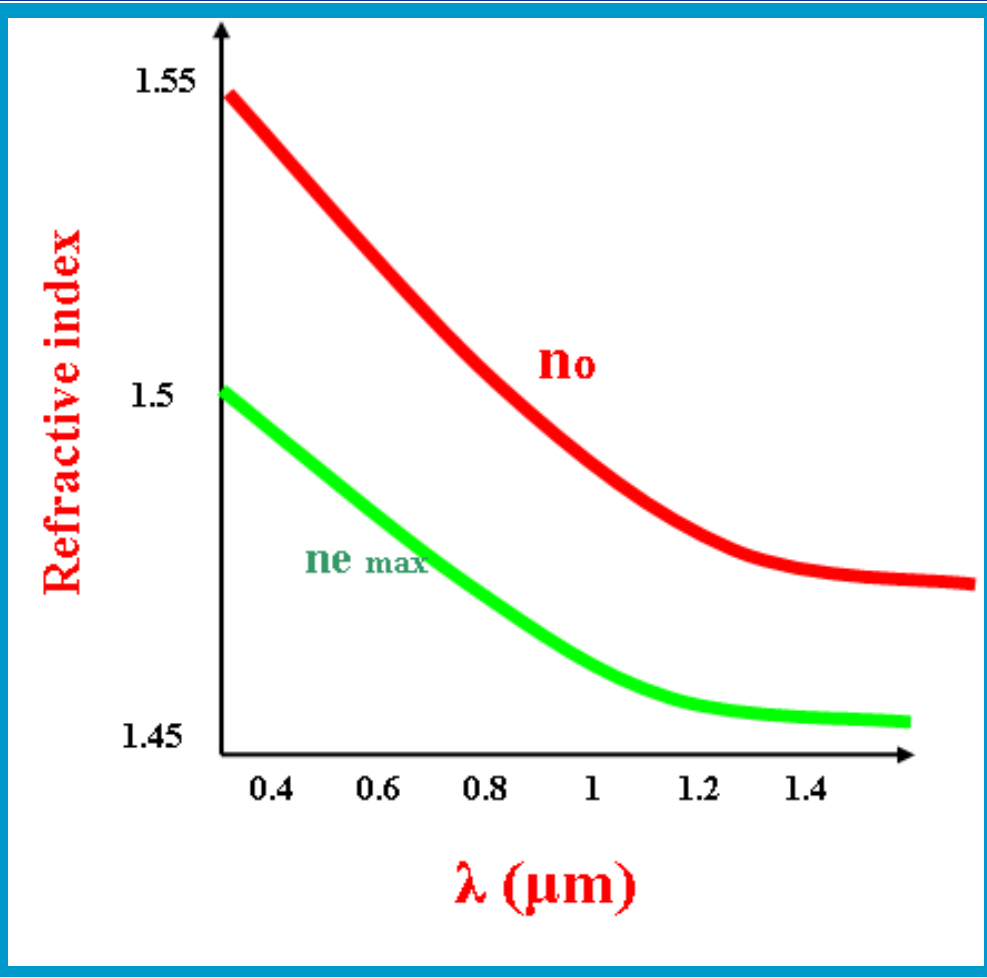
- The dispersion of relevant crystals is available from the phenomenological Sellmeier formulas

$$\text{dispersion } n^2(\lambda_{sell}) = A_{sell} + \frac{B_{sell}}{\lambda_{sell}^2 - C_{sell}} + \frac{D_{sell}\lambda_{sell}^2}{\lambda_{sell}^2 - E_{sell}}$$

- With the unit of measure for the wavelength λ in this equation: $[\lambda_{sell}] = \mu m$ (see example for these coefficients)

- The refractive index ne is the value of the index ellipsoid perpendicular to the optical axis and thus the extreme value. (see an example for Nd lasers and their harmonics).

- The refractive indices for the two beams for KDP as a function of the wavelength .



- To get appropriate phase matching angle n_e can be varied between $n_{e,max}$ and n_o to et the same value but for different wavelengths.
- But not all materials allow phase matching for the required combination of wavelengths. New crystals are being developed.

Walk-Off angle

- Even in case of perfect phase matching , the directions of the beam propagation of new and old frequencies and propagation direction of their energy (Poynting vector) can be different because E and D are not parallel in case of extraordinary beam .

- In this type of critical phase matching the interaction length is limited by (walk - off) between the beam and the energy propagation directions:

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

- With a value of 1.4 for KDP and $\lambda = 1064$ nm .
- In (noncritical phase matching) with $\theta_{pm} = 90^{o}$ no walk-off occurs.
- For high efficiency of frequency conversion
- Intensity ↑ → stronger focusing reduces the interaction length phase matching ↓ due to higher divergence of incident light.

- The acceptance angle $\Delta\theta$pm is:

$$\Delta\theta_{pm} = \frac{0.442\lambda_{inc} n_{0,1}}{n_o^2 (n_{0,2} - n_{e,2}) L_{crystal} \sin 2\theta_{pm}}$$

- As the full angle around θpm for half intensity of SHG, It results in values of (a few 0.1^o for KTP and 25^o for KDP) .
- In noncritical phase matching this equation reduces to:

$$\Delta\theta_{pm} = n_{0,1} \sqrt{\frac{\lambda_{inc}}{(n_{0,2} - n_{e,2}) L_{crystal}}}$$

- From both equations the critical λ range for the incident beam can be the dispersion

of the crystal. $\frac{\partial_{ni}}{\partial\lambda_{inc}}$

- 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 .
- The phase matching can also be tuned by temperature variation for certain crystals .

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 temp. sensitivity which changes the refractive index ellipsoids.

$$\textbf{Optimal crystal length Lcrystal} = 2.9\frac{\pi\, w_o^2}{\lambda_{inc}}$$

For a Gaussian b

- The optimal length can be shorter for short pulses with durations, fs , ps

Type I and Type II phase matching

- There are many different types of phase matching principles.
- The incident wave was used as the ordinary beam for the quadratic nonlinear effect and the SH as the extraordinary beam in materials.

$$Type\ I\ \ phase\ matching\ k_{SHG}(e) = k_{inc}(o) + k_{inc}(o)$$

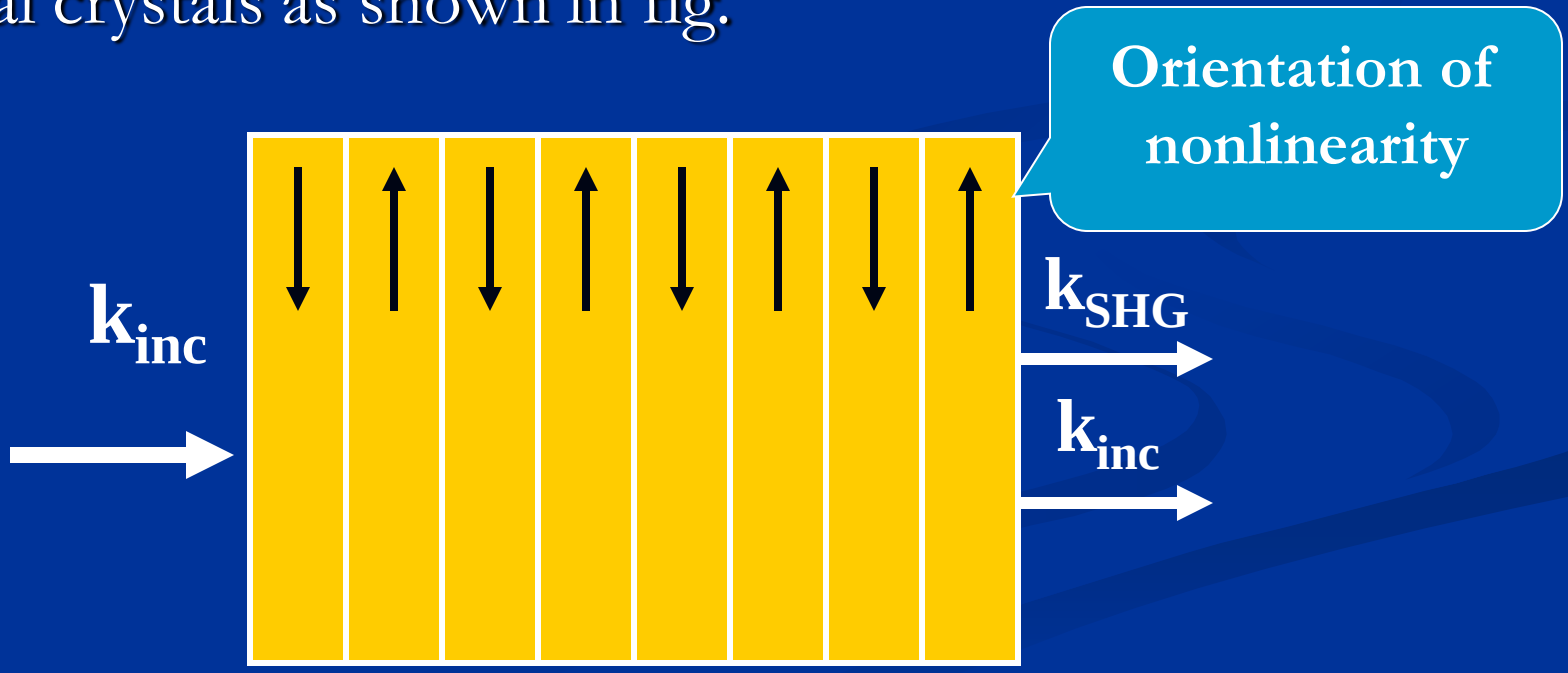
- If the incident wave is used as a mixture of an ordinary and an extra ordinary beam producing an extraordinary SHG.

$$Type\ \mathrm{II}\ \ phase\ matching\ k_{SHG}(e) = k_{inc}(o) + k_{inc}(e)$$

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

Quasi-phase matching (qpm)

- The phase mismatch between the original light wave and the nearly generated waves as a result of the different refractive indices for different λ's can also be compensated by grating structures of the orientation of the nonlinearity of the material.
- The grating is typically produced by periodic poling of ferro electrical crystals as shown in fig.



- The grating period Λqpm depends on the mismatch of the refractive indices of the incident and SH waves:

$$qpm\ period\ \Lambda_{qpm} = m\frac{2\pi}{|k_{inc} - k_{SHG}|} = m\frac{2\pi}{|n_{inc} - n_{SHG}|}$$

- With m as the order of the periodic poling . At (m=1) → the phase mismatch between two waves after half the period is π.
- The optimum case is obtained if the sign of the nonlinearity is reversed every $\pi/|k_{inc}-k_{SHG}|$ second.
- Typical poling periods are in the range around are μm.
- The advantage of this qpm compared to conventional phase matching is the higher efficiency from crystals such as LiNbOs, KTP. They can be applied in optimal direction showing max. nonlinearity, which can not be achieved with conventional phase matching.
- A disadvantage of this method is the limited size of crystals of about 0.5 mm in the direction of poling field

Frequency mixing of two monochromatic fields

- Two light waves with different frequencies υ_1, and υ_2 can be mixed and new frequencies generated.

Sum frequency $\upsilon_{sum} = \upsilon_1 + \upsilon_2$

Difference frequency $\upsilon_{diff} = |\, \upsilon_1 - \upsilon_2 \,|$

- E.g., two collinear waves with parallel polarization combining to give the total field E:

$$E = E_1 + E_2 = E_{0,1} \cos(2\pi \upsilon_1 t - k_1 z) + E_{0,2} \cos(2\pi \upsilon_2 t - k_2 z).$$

- The nonlinear polarization results form:

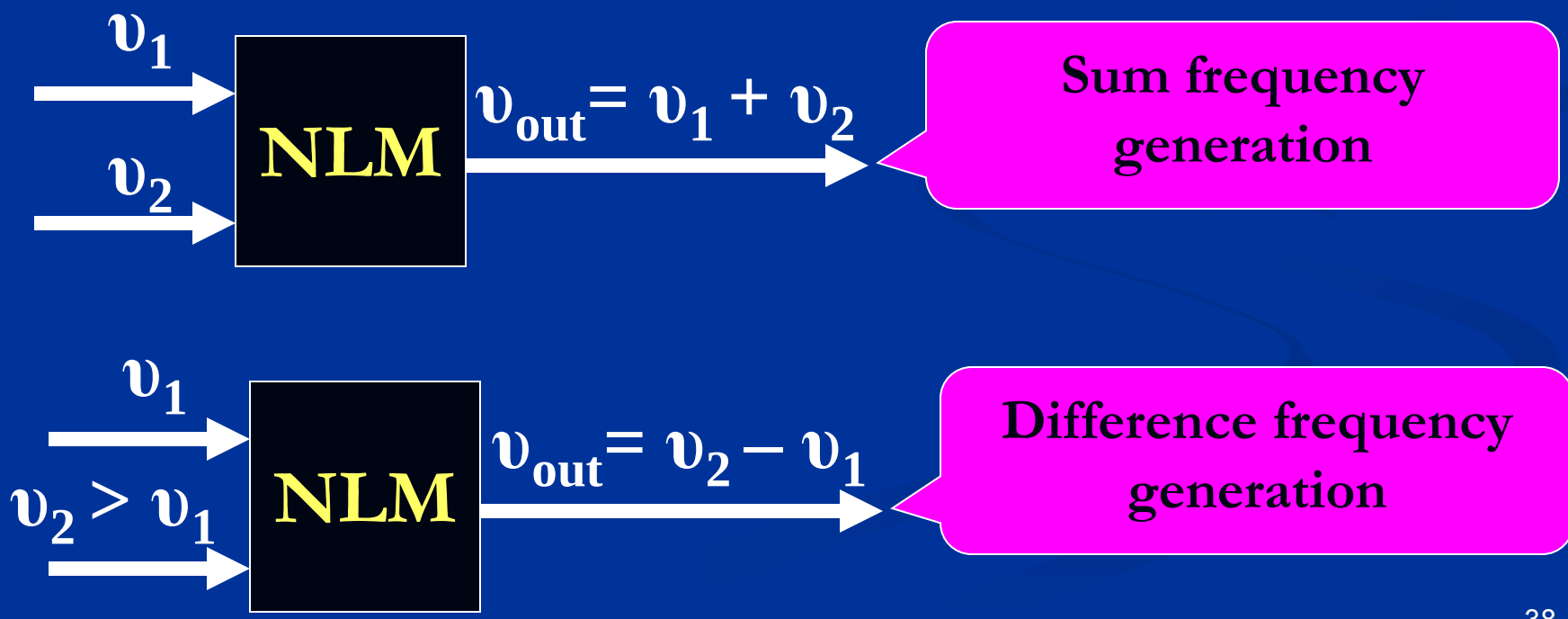
$$\begin{pmatrix} P_x^{(2)} \\ P_y^{(2)} \\ P_z^{(2)} \end{pmatrix} = 2\varepsilon_o [d] \begin{pmatrix} E_x(\upsilon_1)\ E_x(\upsilon_2) \\ E_y(\upsilon_1)\ E_y(\upsilon_2) \\ E_z(\upsilon_1)\ E_z(\upsilon_2) \\ E_y(\upsilon_1)\ E_x(\upsilon_2) + E_y(\upsilon_2)\ E_z(\upsilon_1) \\ E_x(\upsilon_1)\ E_z(\upsilon_2) + E_x(\upsilon_2)\ E_z(\upsilon_1) \\ E_x(\upsilon_1)\ E_y(\upsilon_2) + E_x(\upsilon_2)\ E_y(\upsilon_1) \end{pmatrix}$$

With the matrix (d) as used before .

These new frequencies can be achieved with parametric oscillators or amplified in parametric amplifiers.

Parametric amplifiers and oscillators

- If two beams with frequencies υ_1 , and υ_2 are superimposed in a suitable nonlinear material (NLM) with a total intensity reaching the nonlinear regime the additional sum frequency υ_{sum} or υ_{diff} (or both) occur with I_{sum} and I_{diff}.



- 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.
- If one of the two incident beam, e.g. a signal beam with frequency υ_{signal} is originally weak, it can be amplified at the expense of the other strong incident pump light beam intensity with υ_{pump}.
- A new frequency υ_1 , will occur in a so-called (idler beam) for photon energy conservation.

$$\text{If } \upsilon_{signal} < \upsilon_{pump} \quad \rightarrow \quad \upsilon_{idler} = \upsilon_{pump} - \upsilon_{signal}$$

- This arrangement is called an Optical parametric amplified OPA . This process can be applied e.g. (LiNbO3).
- Laser pump light → visible range.
- Signal and idler frequencies → IR region up to 7 μm .

- If only one beam with one frequency υ_{pump} is used as incident light, these photons can be divided into two photons with the same energy as the pump photon.

$$\upsilon_{signal} > \upsilon_{idler} \quad \rightarrow \quad \upsilon_{signal} + \upsilon_{idler} = \upsilon_{pump}$$

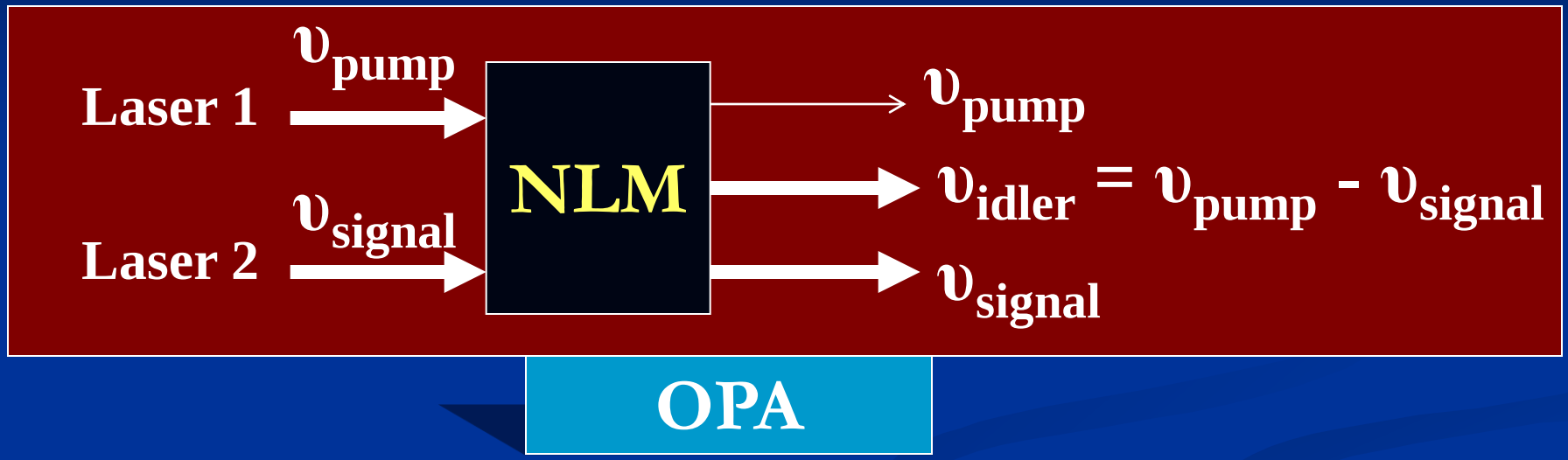
- In addition the phase matching condition giving momentum conservation has to be fulfilled.
- For a negative uniaxial crystal:

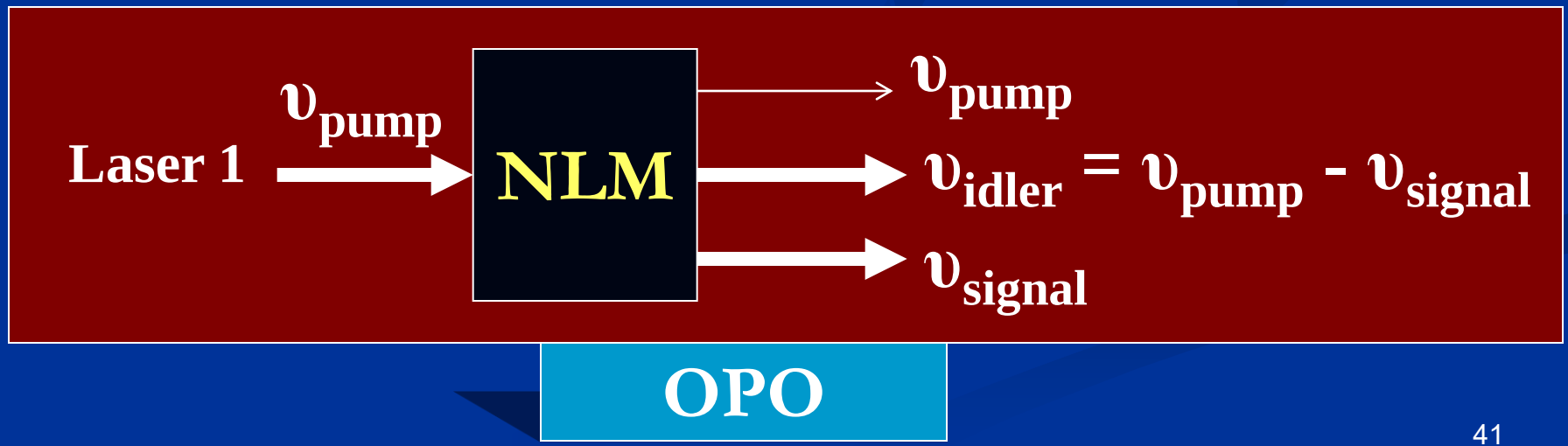
$$Type\ I \quad k^{e}_{pump} = k^{o}_{signal} + k^{o}_{idler}$$

$$Type\ \mathrm{II} \quad k^{e}_{pump} = k^{o}_{signal} + k^{e}_{idler}$$

$$k^{e}_{pump} = k^{e}_{signal} + k^{o}_{idler}$$

- This arrangement is called an Optical Parametric Oscillator OPO





- Several crystals are useful for OPA and OPO (see examples).
- The phase matching can be reached by angle or temp. tunning of the crystal.
- Because the OPO process starts from noise, the resulting new beams are usually not as spectrally narrow and not of as good beam quality as can be reached with the OPA.
- But in both cases the power P or pulse energy E is higher the higher frequency of the light.
- If no absorption occurs energy conservation is fulfilled, the Menley _ Rowe condition with the light power P for CW and pulse energy E for the light pulses are valid:

$$\frac{(P\,or\,E)_{pump}}{\nu_{pump}} = \frac{(P\,or\,E)_{signal}}{\nu_{signal}} = \frac{(P\,or\,E)_{idler}}{\nu_{idler}}$$

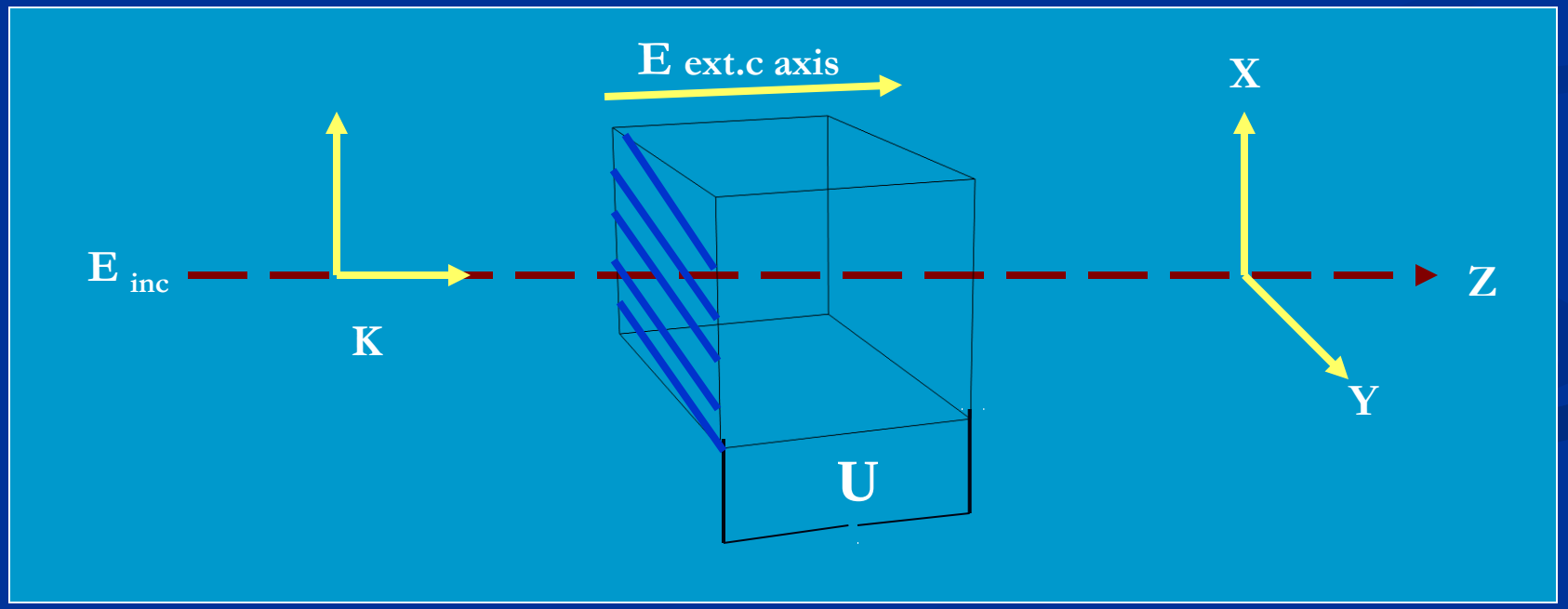
and

$$(P\,or\,E)_{pump,inc} = (P\,or\,E)_{signal} + (P\,or\,E)_{idler} + (P\,or\,E)_{pump\ -residual}$$

- The efficiency of these frequency conversion techniques depends on the bam quality of the pump beam, its spectrum, the degree of polarization and the pulse duration, the focusing and the nonlinear material.

Pockel's Effect

- Some electro-optical second-order effects are important in photonics.
- In NLM the electric field of the light wave is superimposed on the externally applied electric filed and will be influenced by the resulting nonlinear polarization.
- The Pockel's effect rotates the polarization of the incident light as a function of the externally applied field. This can be acquired longitudinally or transversally w.r.t the wave vector of the light beam. As shown in Fig. below.



- The electric light field of the incident monochromatic planar wave has components Ex and Ey:

$$E_{inc,x} = E_{inc,x,o} e^{i(2\pi\nu_{inc} - k_{inc} z)}$$

and

$$E_{inc,y} = E_{inc,y,o} e^{i(2\pi\nu_{inc} - k_{inc} z)}$$

- If a uniaxial crystal is used and its optical axis has the same direction as the external electric field E_{ext}, the resulting nonlinear polarization can be calculated. For this geometry only the two terms with Ey.Ez and Ex.Ez will be nonzero. Thus the quadrate nonlinear polarization with υ_{inc} has components:

$$P_x(w) = 2\varepsilon_o d_{14}^{(o)} E_{inc,y,o} E_{ext} \cos(2\pi\upsilon_{inc} t - k_{inc} z)$$

and

$$P_y(w) = 2\varepsilon_o d_{25}^{(o)} E_{inc,x,o} E_{ext} \cos(2\pi\upsilon_{inc} t - k_{inc} z)$$

  - With the condition **$d_{14} = d_{25}$, e.g. KDP**.

- If this nonlinear polarization is applied to (**) the amplitude of the electric field of the light wave $E_{light,x}(z)$ and $E_{light,y}(z)$ can be calculated:

$$\frac{\delta E_{light,x}(z)}{\delta z} = \frac{-ik_{inc} d_{14}^{(o)} E_{inc,y,o} E_{ext}}{n^2}$$

and

$$\frac{\delta E_{light,y}(z)}{\delta z} = \frac{-ik_{inc} d_{14}^{(o)} E_{inc,x,o} E_{ext}}{n^2}$$

- With the ordinary refractive index of the crystal.
- These equations can be solved in the case of $E_{inc,y,o}=0$

$$E_{light,x}(z) = E_{inc,x,o} \cos\varphi_{pockels}$$

and

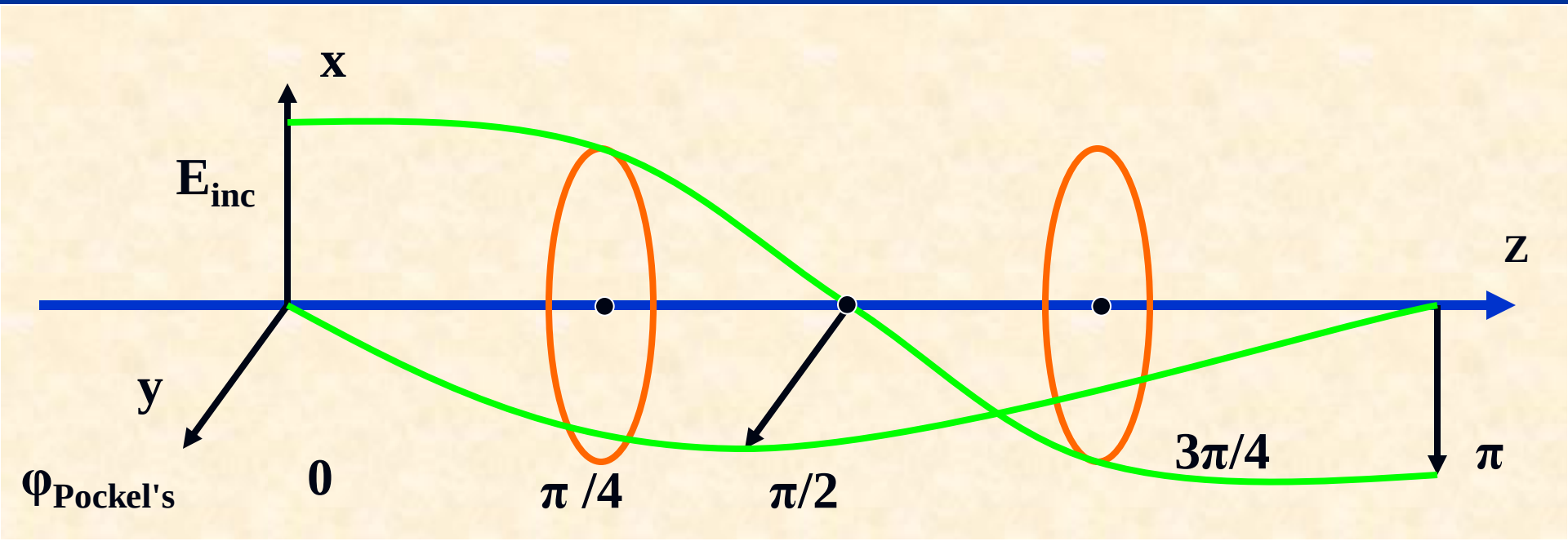
$$E_{light,y}(z) = -iE_{inc,x,o} \sin\varphi_{pockels}$$

- $-i$ indicates a phase shift by 90°

- The angle $\varphi_{\text{Pockel's}}$ results from:

$$\varphi_{pockels} = \frac{k_{inc} d_{14}^{(o)}}{n^2} E_{ext} z$$

- Thus in the Pockel's effect the incident linearly polarized light is convert to circular polarization, to linear at 90°, to circular polarization, to linear at 180° and so on. See fig

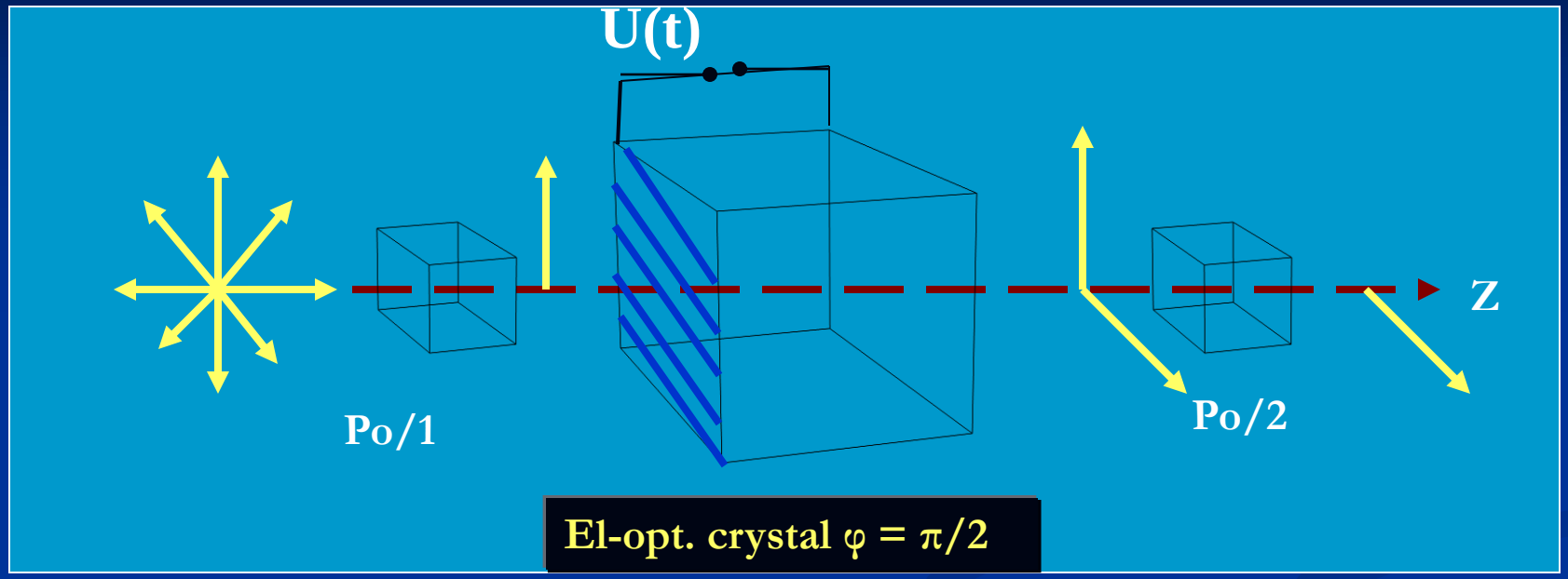


- Thus in the Pockel's effect, a certain Eext, the crystal work as a quarter wave plate and for twice this field as a half-wave plate, producing linear but 90° rotated light
- The necessary voltages Ui are: $U_{\lambda/4} = \frac{n\lambda_{inc}}{8d_{14}^{(o)}}$ **and** $U_{\lambda/2} = \frac{n\lambda_{inc}}{4d_{14}^{(o)}}$
- Crystals with less symmetry such as e.g. a two-axial material can also be used.
- The electro-optical effect is based on the deformation of the refractive ellipsoid in the matter by the electric field.
- The change of the refractive index of the matter is in the first approximation as linear function of the external electric field. Thus, this type of second-order nonlinear electro-optical effect is sometimes called linear. This linear change of the refractive index Δnm can be described by:

$$\Delta n_m = \frac{\overline{n^3}}{2} \sum_p r_{mp} E_p$$

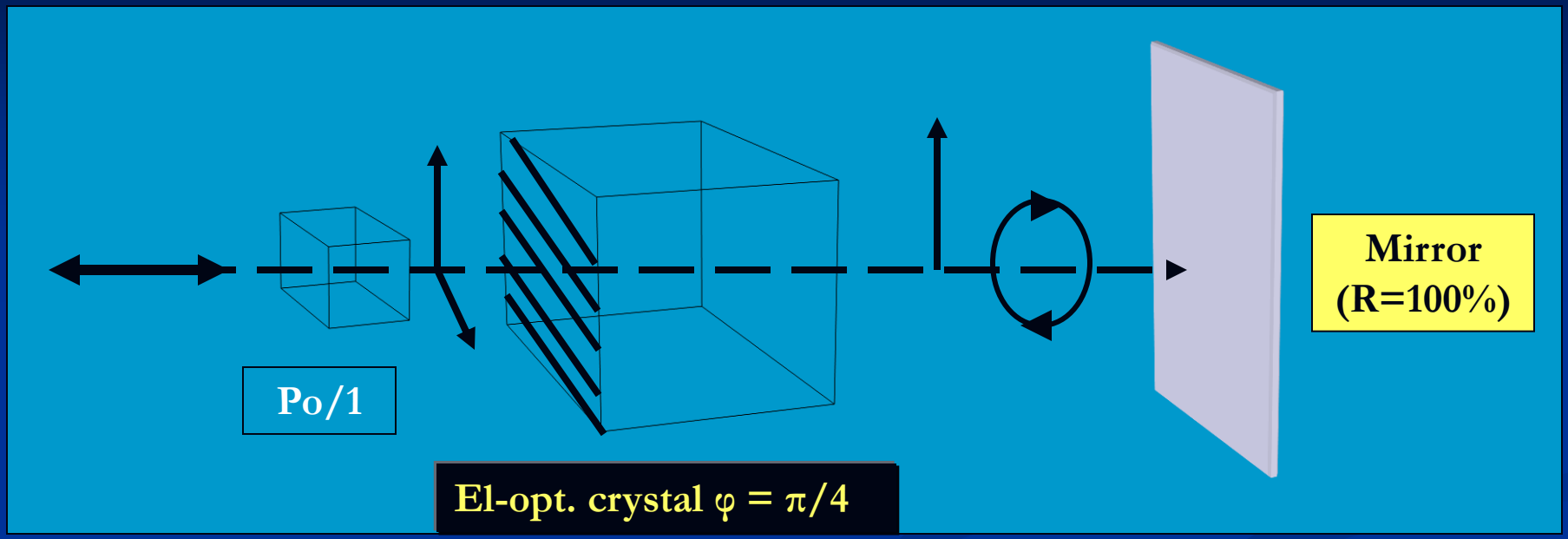
Where r_{mp} is is the electro-optical coefficients and resulting from: $\mathbf{r_{mp} = -d_{pm}^{(0)} / n^{-4}}$ which are functions of applied .

- The Pockel's effect can be applied for light modulation and optical switching if the Pockel's crystal is combined with conventional polarizer as shown in Fig. below.



- The incident light is unpolarized → vertically linearly polarized behind Pol.1. If no voltage applied → stay vertical, cannot pass through Pol.2. If the half-wave voltage Uλ/2 is applied → the light is changed to horizontal → the beam passes Pol.2.Aay voltage between 0 and Uλ/2 will let part of light intensity transmit.
- If the incident light is well enough linearly polarized, Pol.1 is of course not necessary.

- This scheme can be simplified for Q-switched of laser resonator at lower voltages as given in Fig. below.

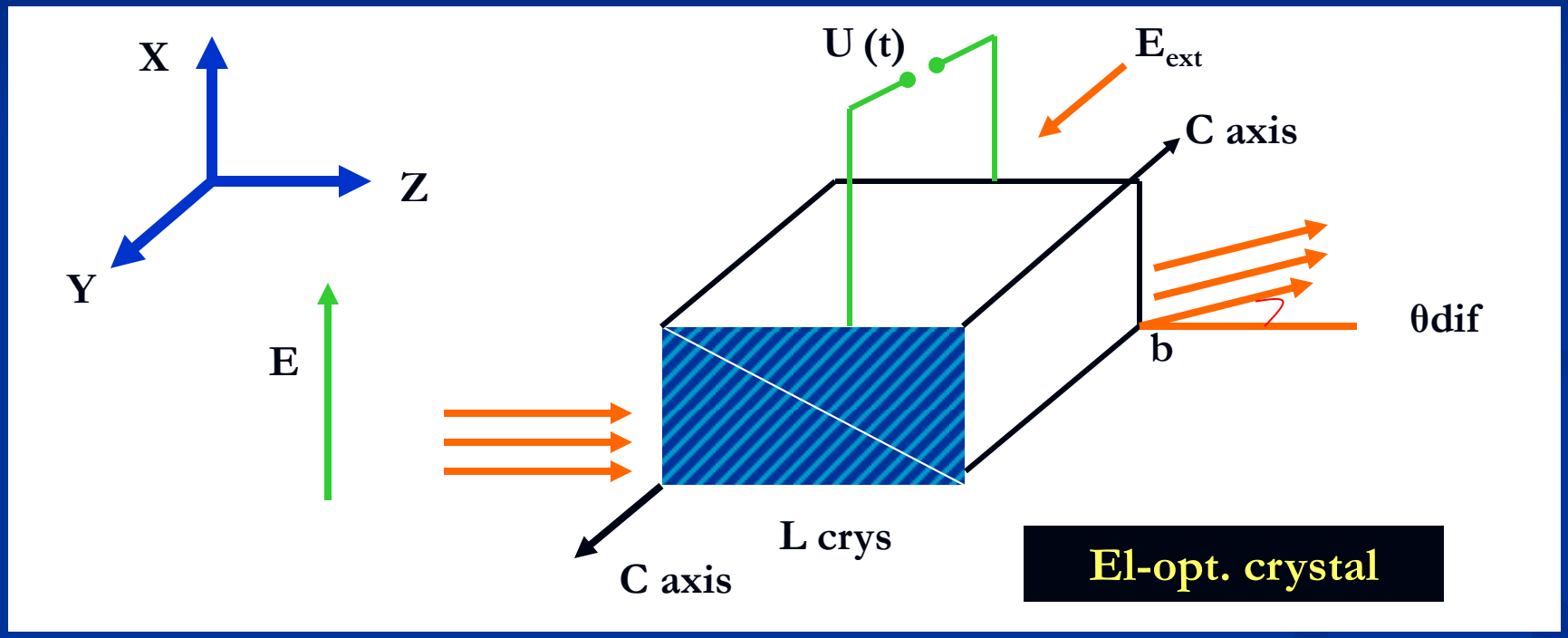


- In this case a 100% reflecting mirror is placed at the position Pol.2. if no voltage is applied, the system will reflect linearly polarized light, but if the Uλ/4 is used the light beam will be circularly polarized behind the nonlinear crystal. After reflection a second pass through the crystal the light will be horizontally linearly polarized. Then it cannot pass the Pol.1. Thus, the set-up works as a mirror with electrically tunable R between 0 and 100%.

- This arrangement can be used in laser resonator for modulation of the output. In particular, it is applied for the generation of pulses with ns duration in solid-state laser via Q-switching.
- The reaction time of the useful crystal is faster than 10^{-10} s and therefore the switching time is limited by the electric transient time for the necessary voltages. Transient times of less than 1ns are possible.
- Because of isolating properties of the crystals the necessary electrical energy is quite 10 W. It is mostly determined by impendence of the electric circuit for reaching of the crystal capacity.
- This is typically a few pF, e.g. 5 pF for KD*P which has a $U\lambda/2$ of 3.4 kV for 1.06 μm.

Electro-optical beam deflection

- Another possibility of switching or modulation light based on the electro-optical effect uses a reflection index step at the border of two differently oriented nonlinear crystals. See Fig. below.



- The optical axis of two crystals is antiparallel, perpendicular to the incident beam. If a voltage is applied along the optical axis of two crystals, the change of n will add at boundary surface.

- As a result the incident beam will experience different optical paths as a function of its height x in vertical direction
- The reflective index of crystal 1 is $\mathbf{n_1 = no - d36\ E_{ext} / 2no}$
- The reflective index of crystal 2 is $\mathbf{n_2 = no + d36\ E_{ext} / 2no}$
- Thus the total reflective index difference will be:

$$\Delta n = \frac{1}{n_o} d_{36} E_{ext}$$

The path length difference for upper and lower beam lead to a deflection of the light beam with angle:

$$deflection\ angle\ \theta_{def} = \frac{L_{crys}}{b_{crys}} . \Delta n = \frac{L_{crys}}{b_{crys}} . \frac{d_{36}}{n_o} E_{ext}$$

- The resolution **A** is given by

$$A = \frac{\theta_{def}}{\theta_{beam}} = \frac{\pi\ L_{crys} . d_{36}}{2 \lambda_{beam} n_o} E_{ext}$$

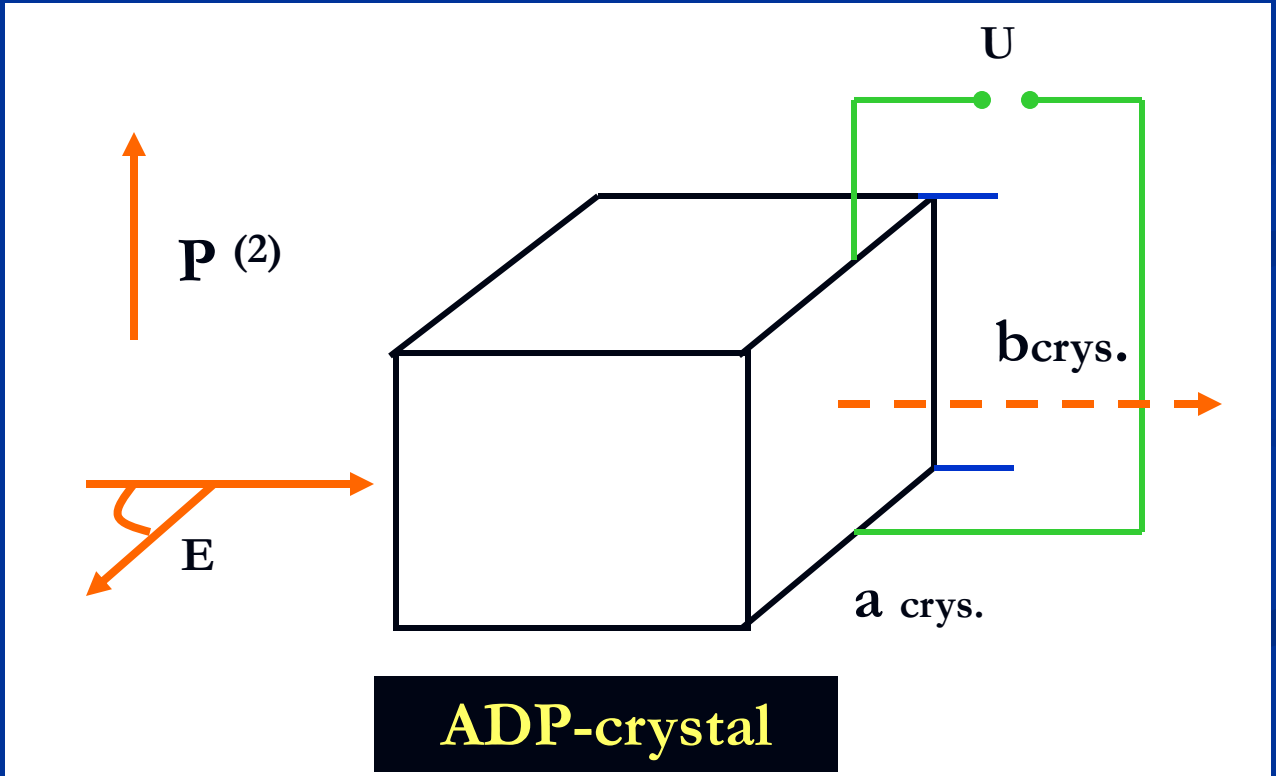
- In practical, angles below 1o can be obtained for crystals of several 10 mm length.

Optical Rectification

- The application of high E produce a second-order nonlinear effect with υ=0, which is a nonoscillating electric field. The physical background for this effect is the displacement of the changes in the temporal average as a sequence of the anharmonic potential.
- The calculation of the second-order nonlinear polarization component $Pi^{(2)}$ for this rectification with υ=0 to form,

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

- With the same relations between the components of the d matrix, but the d values are the same as for the electro-optical effects.
- They are different from the values because of the dispersion and the different frequencies of both processes.
- The experimental setup is sketched in this Fig.



- The CW polarization $P_x^{(2)}(0)$ in the x direction can be calculated as:

$$P_x^{(2)}(\upsilon = 0) = 2\varepsilon_o d_{14}^{(0)} E_y E_z^{*} = \varepsilon_o d_{14} E_{inc}^2$$

- Which becomes maximal for **Ey = Ez = (1/√2)Einc** as assumed in this equation.
- The nonlinear polarization generates a charge separation **Qcrys** in the crystal of:

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

- This change **Qcrys** leads to an externally observable voltage U depending on the capacity of the crystal **Ccrys** :

$$C_{crys} = \varepsilon_o \varepsilon_{crys} \frac{L_{crys} a_{crys}}{b_{crys}}$$

- If finally the light beam power P_{beam} as a function of the electric field amplitude E_{inc} is introduced.

$$P_{beam} = \frac{1}{2}\varepsilon_o c_o n_{crys} a_{crys} b_{crys} E_{inc}^2$$

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

$$U = \frac{2d_{14}^{(0)}}{\varepsilon_o \varepsilon_{crys} c_o n_{crys} a_{crys}} P_{beam}$$

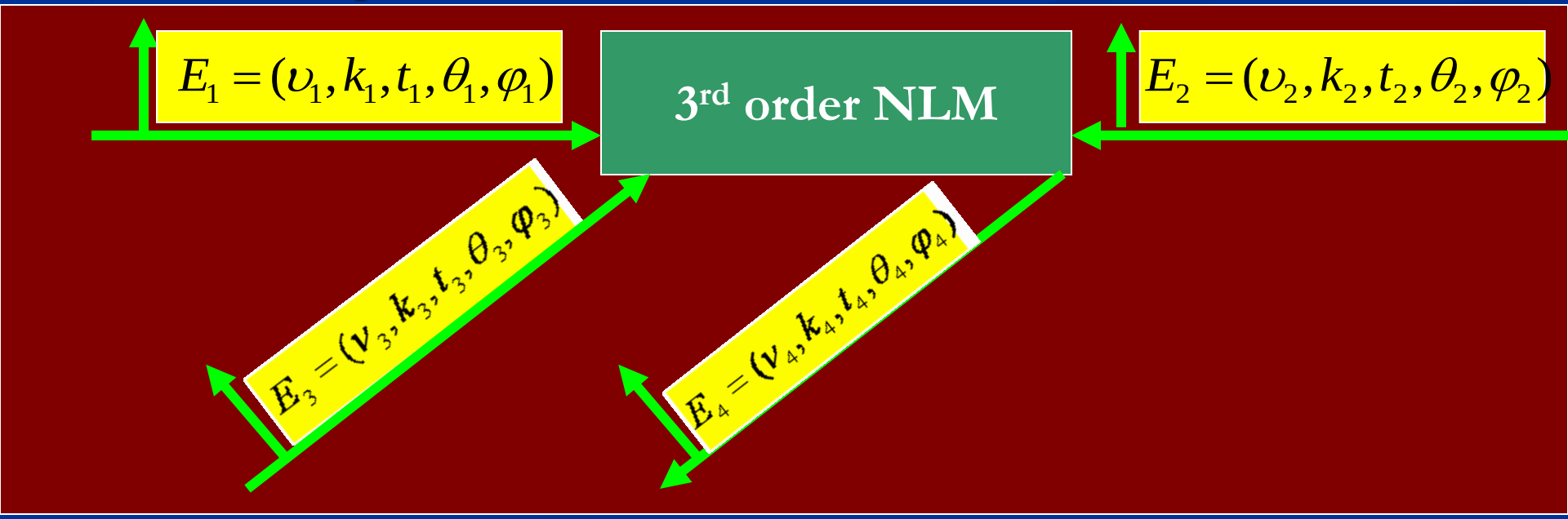
- In practical cases this voltage is very small.
- In a 1 MW pulsed ruby laser light source at 694 nm in a 1x1x2 cm^3 ADP crystal a voltage of 24 mV was observed during the pulse duration of 25 ns.
- In another experiment, in $LiTaO_3$ a laser pulse with duration of 1ps and energy of 1μJ produced a peak current of 0.3A.

Third-Order Effect

- Third-order nonlinear polarization $P^{(3)}$ is a function of the third power of the incident Einc:

$$P^{(3)} = \varepsilon_o \chi^{(3)} E_{inc,1} E_{inc,2} E_{inc,3}$$

- $\chi^{(3)}$Is a four dimensional tensor which can in general be complex.
- The three Einc,i can be components of the same light beam but can also be 3 different light beams which, overlap in the NLM. If a fourth beam is used to detect the changes in the third-order NLM, four wave mixing (FWM) takes place.



- By choosing different υ_m, k_m, t_m, φ_i, φ_m more than hundred prominent schemes of FWM can be applied.
- If one of the simplest cases all light waves have the same frequency and this process is called degenerate four wave mixing (DFWM).
- The three components of $P^{(3)}$ in x,y,z direction are:

$$P_m^{(3)} = \frac{\varepsilon_o}{4}\left[\sum\left(\chi_{mpqr}^{(3,1)}E_pE_qE_r + \chi_{mpqr}^{(3,2)}E_p^{*}E_qE_r + \chi_{mpqr}^{(3,3)}E_pE_q^{*}E_r + \chi_{mpqr}^{(3,4)}E_pE_qE_r^{*}\right)\right]$$

- For nonabsorbing materials the susceptibility tensor $\chi(3)$ has 81 real components and the complex products of E components disappear .

- In second-order nonlinearity for symmetry reasons of the allowed permutations of p,q,r of the E vectors, the number of the distinguisble tensor components of $\chi^{(3)}$ is reduced to 30.

$$\begin{pmatrix} P_x^{(3)} \\ P_y^{(3)} \\ P_z^{(3)} \end{pmatrix} = \varepsilon_o \begin{pmatrix} e_{11} & e_{12} & e_{13} & e_{14} & e_{15} & e_{16} & e_{17} & e_{18} & e_{19} & e_{110} \\ e_{21} & e_{22} & e_{23} & e_{24} & e_{25} & e_{26} & e_{27} & e_{28} & e_{29} & e_{210} \\ e_{31} & e_{32} & e_{33} & e_{34} & e_{35} & e_{36} & e_{37} & e_{38} & e_{39} & e_{310} \end{pmatrix} \begin{pmatrix} E_x^3 \\ E_y^3 \\ E_z^3 \\ 3E_xE_y^2 \\ 3E_xE_z^2 \\ 3E_yE_z^2 \\ 3E_yE_x^2 \\ 3E_zE_y^2 \\ 3E_zE_x^2 \\ 6E_xE_yE_z \end{pmatrix}$$

With the internal relations between the e matrix elements.

- $e_{12} = e_{24}$ $e_{13} = e_{25}$ $e_{14} = e_{26}$ $e_{15} = e_{38}$ $e_{16} = e_{21}$
- $e_{17} = e_{25} = e_{310}$ $e_{19} = e_{210} = e_{34}$ $e_{110} = e_{28} = e_{36}$
- $e_{18} = e_{31}$ $e_{23} = e_{37}$ $e_{27} = e_{30}$ $e_{29} = e_{32}$

- Considering the further symmetry rules $\chi_{mpqr} = \chi_{pmqr}$
- Finally 15 components are relevant for describing 3rd nonlinear process which are function of applied light frequency.
- If the nonabsorbing crystals are of cubic symmetry, only two different values are distinguishable.

$$P_m^{(3)} = \varepsilon_o E_m \left\{ e_{11} E_m^2 + 3e_{14} (E_p^2 + E_q^2 \right\} \text{ with } m \neq p \neq q = x, y, z$$

- If the material is isotropic the additional relation $e_{11}=3e_{14}$ is valid and $P_m^{(3)}$ reduced to:

$$P_m^{(2)} = \varepsilon_o e_{11}(EE) \;\; with \; m = x, y, z$$

- Materials of cubic symmetry (isotropic) will not shell any second-order nonlinear effect.

Generation of The Third Harmonic

- The third order nonlinearity of the previous equation can be applied for the third harmonic generation (THG).

$$THG \; I_{out}(3\upsilon_{inc}) \underleftarrow{\chi^{(3)}_{THG}} I_{inc}(\upsilon_{inc})$$

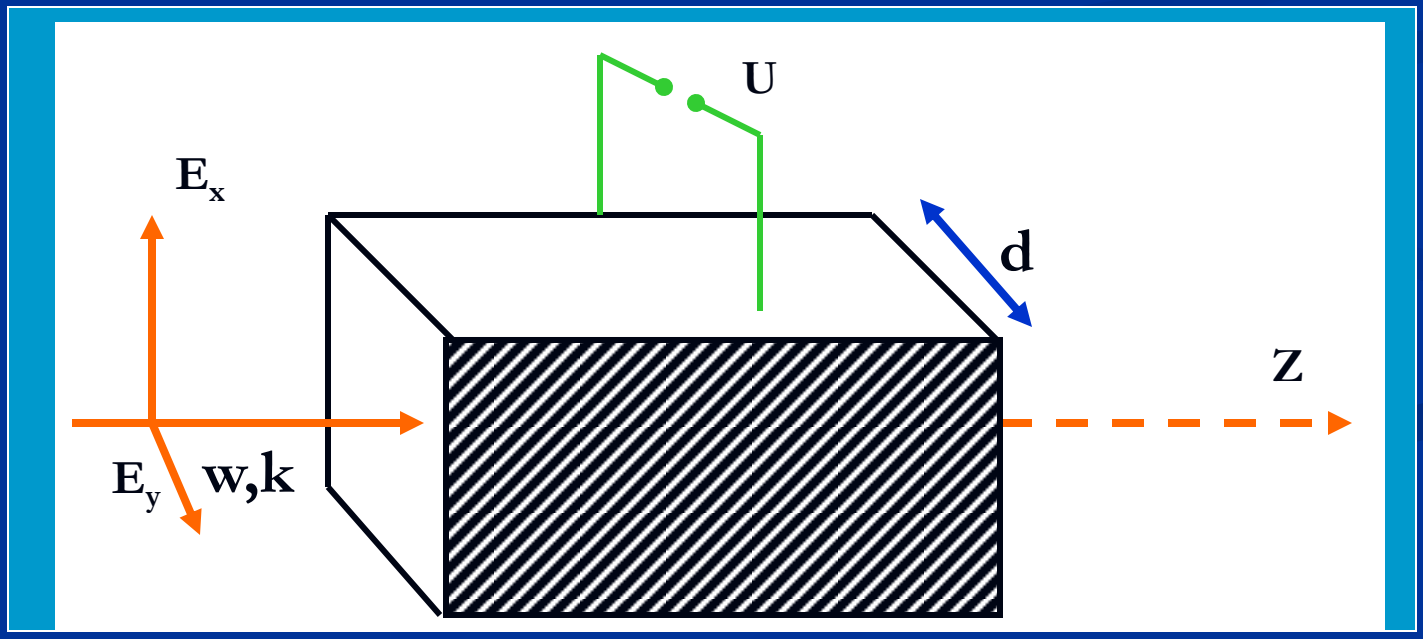
- If this process is based on $\chi^{(3)}$, the efficiency is quite low **($<10^{-14}$)** .
- In photonics the most commonly used generation of 3rd harmonic light is based on two-step procedure of (SHG) in a first step and frequency mixing of this SH with the residual incident wave as the second step.

$$THG \; \text{II}_{out} I_{THG}(3\upsilon_{inc}) \underleftarrow{\chi^{(2)}_{mix}} I_{SHG}(2\upsilon_{inc}) \underleftarrow{\chi^{(2)}_{SHG}} I_{inc}(\upsilon_{inc})$$

- The total efficiency for this type of third harmonic generation can be as high as about 80%.

Kerr Effect

- A strong applied electric field can induced optical birefringence in materials which are optically isotropic without the field.
- Thus, the refractive index tensor of the material becomes a function of light intensity. This electric field $E_{ext}=U/d$ can be applied as an oscillating field of a strong incident light wave or as an external field as in the Pockel's effect but with transversal orientation to the light propagation direction as shown in fig.



- The total polarization $\mathbf{P^{(tot)}}$ is the sum over the linearly induced polarization and the 3rd order nonlinear polarization which both have a component in the x-direction:

$$P_m^{(tot)}(\upsilon_{inc}) = P_{lin,x}^{(1)} + P_{\ln,x}^{(3)}$$

$$= \varepsilon_o \chi^{(1)} E_{inc,x} + \varepsilon_o \chi^{(2)} E_{inc,x} \left(\frac{3}{4} E_{inc,x}^2 + E_{ext}^2\right)$$

$$= \varepsilon_o E_{inc,x} \left[\chi^{(1)} + \chi^{(3)} \left(\frac{3}{4} E_{inc,x}^2 + E_{ext}^2\right)\right]$$

$$P_y^{(tot)}(\upsilon_{inc}) = P_x^{(tot)}(\upsilon_{inc}) = 0$$

$$with \quad P_{nl,x}^{(2)}(\upsilon_{inc}) = \varepsilon_o e_{11} E_{inc,x} \left(\frac{3}{4} E_{inc,x}^2 + E_{ext}^2\right)$$

- The refractive index $n=\sqrt{1+x}$ in the x-direction is changed by this nonlinear polarization :

$$n_{nl,x} = \sqrt{n_o + e_{11}(\frac{3}{4}E_{inc,x}^2 + E_{ext}^2)}$$

- And is constant in the y and x directions.
- The small change Δn_x in the x direction results in:

$$\Delta n_x = \frac{e_{11}}{2n_o}(\frac{3}{4}E_{inc,x}^2 + E_{ext}^2)$$

- Assuming low light intensities the change in the refractive index as a function of the external cw electric field E^2_{ext} results in:

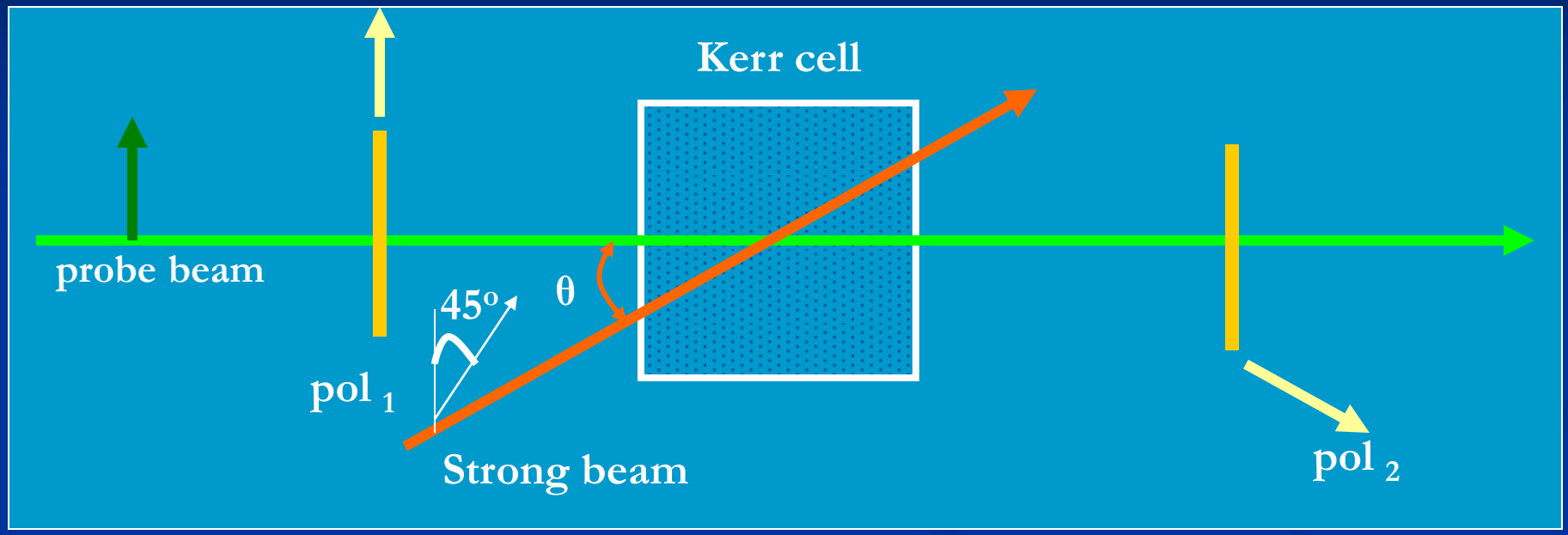
$$\Delta n_x = \frac{e_{11}}{2n_o}E_{ext}^2 = k_{\ker r}\lambda_{inc}E_{ext}^2$$

With Kerr constant

Kerr constant

$$k_{\ker r} = \frac{e_{11}}{2n_o\lambda_{inc}}$$

- With the external E the birefringence of the Kerr material can be changed and used in a setup of a Kerr cell between two crossed polarizers, as shown in fig.



- This is an electrically controlled light gate, a Kerr shutter.
- The necessary E can be determined from the Kerr constant .
- Using this type of induced optical birefringence of the Kerr effect very fast optical switches can be made.

- **In the setup,**
- The incident probe beam polarized in the direction of Pol.1 can not pass the setup. A short and strong light pulse is applied to switch on the transparency. The direction of its polarization has to be 45°. The angel should be as small as possible. If the intensity of the switch beam is suitably chosen, the probe beam will be circularly polarized behind the NL cell.
- Thus, 50% of the incident probe beam can pass the second Pol.
- For a cell length or interaction length Lcell, the necessary intensity follows from:

$$I_{switch} = \frac{n_o c_o \varepsilon_o}{3 k_{\mathrm{ker}\, r} L_{cell}}$$

Self-Focusing

- If the intensity of a transmitted light beam is sufficiently high, almost gases, liquids or solids will show a nonlinear interaction.
- The refractive index change will modify the light propagation not only w.r.t the polarization but in its geometrical properties too. As a consequence for high-intensity beams with long interaction length in the matter, self-focusing can occur, and for short interaction lengths self-diffraction or self-defocusing may be obtained.
- Assuming a Gaussian transversal beam profile of the incident light the intensity across the beam radius r perpendicular to the beam propagation direction is given as a function of the total power P_{tot} by

$$I_{inc,r}(r,t) = \frac{2}{\pi w_{beam}^2} e^{-2r^2 / w_{beam}^2 P_{tot}(t)}$$

- The refractive index n(r,I) in the material will be modified across the beam diameter:

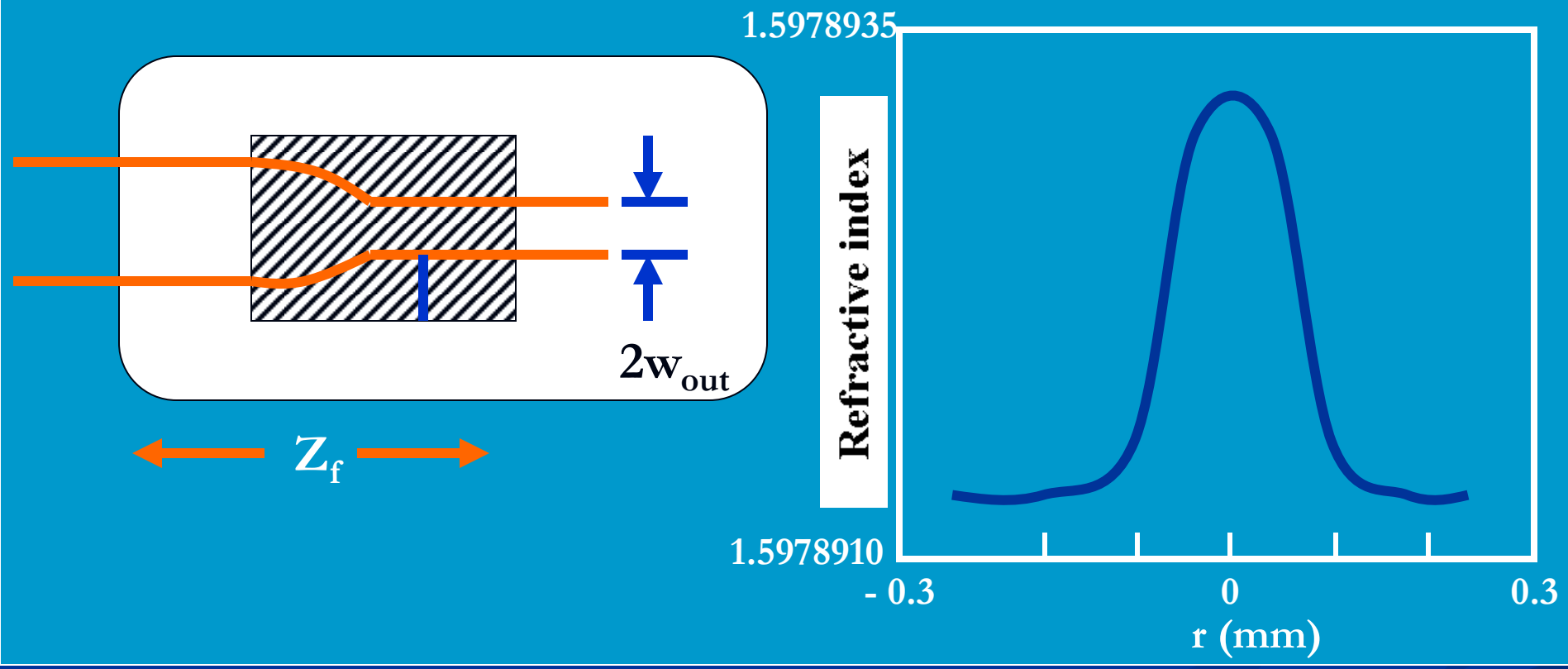
$$n_{(r,I)} = n_o + \Delta n_{nl,r} = n_o + \frac{1}{2} e_{11} c_o \varepsilon_o E_{inc,r}^2$$
$$= n_o + \gamma(\upsilon_{inc}) E_{inc,r}^2$$
$$= n_o + \gamma_1(\upsilon_{inc}) I_{inc,r}$$

- With coefficients γ and γ_1 valid for linearly polarized light.
- In the cases of unpolarized or circularly polarized light the effective coefficients have to be reduced to 2/3 of given values:

$$\gamma(\upsilon_{inc}) = \frac{3}{8} \frac{e_{11}}{n_o}, \quad \gamma(\upsilon_{inc})_{circ} = \frac{1}{4} \frac{e_{11}}{n_o}$$

- This Gaussian refractive index profile is shown in fig. resulting from a Gaussian shaped incident beam.

- The refractive index profile acts as a lens analogous to the quadratic refractive index profile and will focus the beam as shown in Fig.



- This focusing increases I and thus n will be changed even more.
- This position feed back for more and more focusing is limited by the increased divergence of the Gaussian beam with smaller diameter.

- After a certain length Z_f an equilibrium between focusing and defocusing is reached and the beam will propagates as in a waveguide with constant diameter d_{sf}.
- As a hint fir the order of magnitude of self-focusing effect the critical power was estimated under different assumption e.g. of aberration free-focusing, to give:

Critical power of self-focusing $P_{cr} = \frac{\varepsilon_o c_o \lambda_{inc}^2}{4\pi\gamma}$

- As this critical power an incident plane wave front will stay planar or in other works the incident beam will be waveguided with unchanged diameter.
- Thus, this power gives self-trapping of the beam.

- At higher powers the incident beam will be focused with a focal length Z_f which was estimated in the same way as P_{cr}:

Self focusing focus length

$$Z_f = \frac{\pi}{\lambda_{inc}\left(\sqrt{\frac{p}{p_{er}} - 1} + 1\right)}$$

- But the Gaussian index profile leads to aberration-during focusing.
- Thus, a more precise solution was numerically produced.

$$Z_f = \frac{0.734\pi w_{inc}^2}{\lambda_{inc}\left(\sqrt{\left(\left(\frac{p}{p_{er}}\right)^{1/2} - 0.852\right)^2 - 0.0219}\right)}$$

- In practical cases the focus length Z_f is in the range of several 10 cm. the spot size is theoretically zero but practically minimum diameter of a bout 5μm were obtained.
- The high intensities are even more easily achievable with short pulses.
- In this cases the self-focusing will be a function of times and the effect of "temporarily moving foci" has to be discussed.
- It seems that the mechanism are different for ns, ps, and fs.
- Self-focusing is used in white light generation to achieve high intensities and large interaction lengths at the same time which is not possible by simple focusing.
- Besides reorientation, induced dipole moments and other nonlinear effects on molecular and atomic scale causing self-focusing, additional thermal effects can also produce refraction index changes. This thermal self-focusing or defocusing is based on $\mathbf{dn/dT \neq 0}$.
- The thermal effect is produced by absorption of the light and thus, this is at least a partially resonant process.

Spatial Solitons

- The nonlinear refractive index change in a Kerr material can establish a waveguiding effect which compensates the self-diffraction of a propagating beam. As a result the light beam can propagates through the matter with constant beam profile and diameter. But this effect needs a certain beam profile different from the Gaussian shape. Such a beam is called a "spatial soliton".
- A solution for the intensity of a spatial soliton $\mathbf{I_{sol}}$ related to the nonlinear wave equation is:

$$I_{sol}(r) = \frac{1}{2} c_o \varepsilon_o n_o E_{o,sol}^2(r_{sol}) \operatorname{sech}^2\left(\frac{r}{r_{sol}}\right)$$

- With the characteristic beam radius r_{sol} where the intensity is 42% of the I_{max} in the middle of the beam at r =0.

- The intensity $I_{o,sol}$ is a function of the beam diameter $\mathbf{r_{sol}}$:

Spatial soliton intensity

$$I_{o,sol} = \frac{1}{k^2 \gamma_1} \frac{1}{r_{sol}^2}$$

- With Kerr coefficient γ .
- The cross-section of a spatial soliton with radius $\mathbf{r_{sol}}$ **=3 μm** which is about the fundamental mode diameter in an optical fiber (see example).
- The phase velocity $\mathbf{C_{sol}}$ of the spatial soliton is given by:

Spatial soliton velocity

$$C_{sol} = \frac{c}{1 + \frac{\lambda^2}{8\pi^2 r_{sol}^2}}$$

- Which is smaller than C (about 0.15% in the example) and approaches the velocity of the linearly diffracted light for $\mathbf{r_{sol} >> \lambda}$.

Self Diffraction

- If the refractive index change is produced in a thin silica of matter by a tightly focused laser beam a lens-like index profile will occur. At this induced refractive index profile the laser beam will be diffracted. (see fig. in the text)
- The number of diffraction rings N_{rings} can be used to determine the maximum change of the refractive index in the matter $\Delta n_{nl,max}$ at the center of the Gaussian beam for the normal incidence as:

Maximum index change

$$\Delta n_{nl,\max} = \frac{\lambda_{beam}}{L_{mat.}} N_{rings}$$

- N counts the number of 2π phase shift for the transmitted light from the center to the wings.

Self-focusing in weakly absorbing samples

- The refractive index change can be produced by very weakly absorbing sample via thermally induced refractive index changes.
- The absorption coefficient produces under steady-state conditions a thermal lens with a focal length $\mathbf{f_{def}}$

$$f_{def} = k_{mat}\left[\frac{dn_o}{dT}\, p_{inc}\left(1-e^{-\alpha L_{mat}}\right)\right]^{-1}$$

- With K_{mat} thermal conductivity, $\mathbf{P_{inc}}$ incident power.
- For outweighing diffraction and defocusing of a parallel input beam the incident power has a critical value $\mathbf{P_{cr,def}}$:

$$P_{cr,def} = \lambda_{inc} k_{mat}\left[\frac{dn_o}{dT}\left(1-e^{-\alpha L_{mat}}\right)\right]^{-1}$$

- This type of self-focusing can be applied for the determination of α for thin films or $\alpha < 10^{-6}$ cm^{-1}.

Self Phase-Modulation

- If a light pulse of high I transmits through a material n becomes a function of the temporally changing incident E. The refractive index will be a function of time following the pulse shape of I as a function of **t, $I_{inc}(t)$.** This temporally changed n will change λ in the matter and "self-phase modulation takes place.
- As a consequence the frequency of the transmitted light will be tuned during the pulse, it has a "chirp".
- The pulse duration **Δt_{FWHM}** can be long compared to the interaction tome of the matter (few ps). This may or may not lead to steady-state condition. In non-stationary self-phase modulation the reaction of the matter will be delayed.
- A Gaussian pulse shape with the duration is assumed :

$$\Delta t_{FWHM} = 2\Delta\tau\sqrt{(\ln 2)}$$

$$I(t) = I_o \; e^{-(t-t_{\max}/\Delta\tau)^2}$$

- During this pulse n_{max} will change under steady-state conditions instantaneously with I (see fig. in the text).
- Almost monochromatic light is presupposed can be achieved with $1/\Delta\tau << \nu_{inc}$
- If the beam is propagating in the z-direction through a material with $n_{nl}(I)$ and length L_{mat}, the electric field behind the material is given by:

$$E(t,z) = E_o e^{-\frac{1}{2}(t-t_o/\Delta\tau)^2 + i\varphi}$$

With phase factor:

$$\varphi(t, L_{mat}) = 2\pi\upsilon_{inc}t - kL_{mat} = 2\pi\upsilon_{inc}\left(t - \frac{n_{nl}(I)L_{mat}}{c_o}\right)$$

- This phase shift leads to a temporal compression and expansion of the light wave (see fig. in the text)

- The n will be changed by the light pulse, presupposing reaction times of the nonlinear material are short compared to pulse duration

$$n_{nl}(I) = n_o + \Delta n_{nl} = n_o + \gamma I(\upsilon_{inc}) I_{inc,\tau}$$
$$= n_o + \gamma I(\upsilon_{inc}) I_o e^{-(t - t_{\max}/\Delta\tau)^2}$$

- The light frequency behind the sample can be calculated :

$$\upsilon_m = \frac{d\varphi}{dt} = \upsilon_{inc}\left(1 - \frac{L_{mat}}{c_o}\frac{dn_{nl}}{dt}\right) = \upsilon_{inc} - \Delta\upsilon_{spm}$$

- Leading to a time-dependant frequency shift $\Delta\nu_{spm}$ caused by the steady-state self-phase modulation :

Chirp frequency

$$\Delta\upsilon_{spm} = \upsilon_{inc}\gamma I_o \frac{L_{mat}}{c_o}\frac{2(t - t_{\max})}{\Delta\tau^2} e - (t - t_{\max}/\Delta\tau)$$

- Which is (-) at the leading edge of the pulse and (+) after its peak.

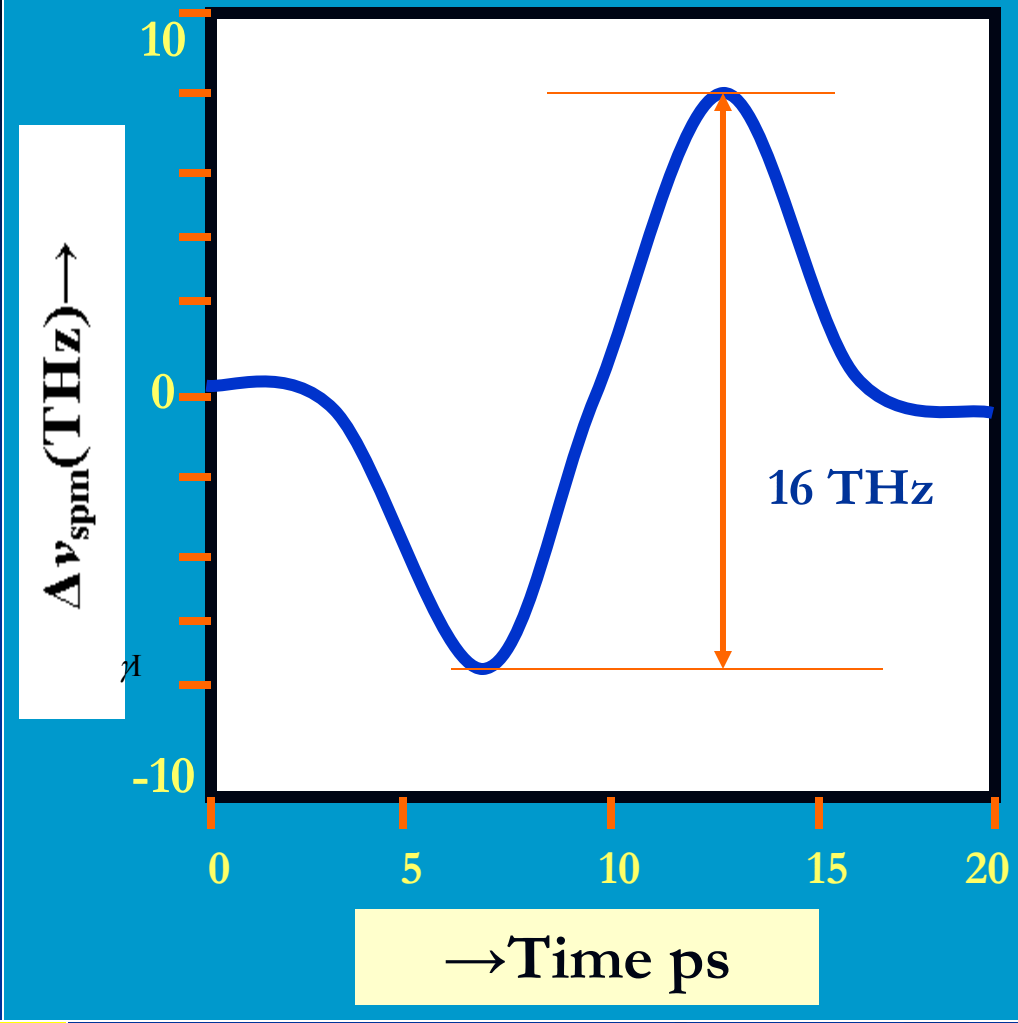
The maximum shift reached at a time:

$$t_{spm,\max/\min} = t_{\max} \pm \frac{\Delta t_{FWHM}}{2\sqrt{2\ln^2}}$$

And reaches a total value between the frequency max. and min. of :

$$\Delta\upsilon_{spm,tot} = \Delta\upsilon_{spm,\max} - \Delta\upsilon_{spm,\min}$$

$$= \upsilon_{inc}\gamma I_{inc}\frac{L_{mat}}{c_o}\frac{4\sqrt{2\ln^2}}{\sqrt{e}\Delta t_{FWHM}}$$



Thus, during the main part of the pulse intensity the frequency is shifted from "red" to "blue".

Generation of Temporal Solitons :

Soliton Pulses:

- Any linear dispersion will lead to an increase of the pulse length because pulses cannot be perfectly monochromatic. Thus over long distances, as in fiber communications, this will limit the possible modulation frequency.
- The third-order nonlinearity of the materials can be used to compensate this effect and transmit specially designed pulses, "longitudinal or temporal solitons", with out any change in the temporal pulse shape over long distances.
- Nonlinear induced dispersion by self-phase modulation can delay the shorter λ part of the pulse only. Thus for the soliton effect an anomalous linear dispersion with $\mathbf{dn/d\lambda > 0}$ is necessary.

Assuming:

- SVA of the pulse
- Weak dispersion
- Small NL effect ($n = n_o + \gamma_I I$)

- The differential equation for the temporal envelope of the pulse propagating in the z-direction can be derived as:

$$\frac{\partial E}{\partial z}+\frac{1}{c_g}\frac{\partial E}{\partial t}-i\frac{\pi}{c_o}\left\{2\left|\frac{dn}{d\upsilon}\right|+\left|\frac{d^2n}{d\upsilon^2}\right|\right\}\frac{\partial^2 E}{\partial t^2}-i\pi\varepsilon_o n_o \upsilon_o \gamma\left|E\right|^2 E=0$$

- This equation can be transformed to an expression similar to the nonlinear Schrödinger equation then solved by the following function for the envelope of the electric field $\mathbf{E_{sol}}$:

$$E_{sol}(z,t)=E_{sol,o}\,\mathrm{sech}\left(\frac{t-(z/c_g)}{\Delta t_{sol}}\right)e^{iz/4z_o}$$

- With the temporal pulse width of $\mathbf{\Delta t_{sol}}$ which is measured at 65% of the maximum filed amplitude.

- The intensity of the soliton pulse $\mathbf{I_{sol}}$ is given by:

$$I_{sol}(t) = I_{sol,o}\,\mathrm{sech}\left(\frac{t-(z/c_g)}{\Delta t_{sol}}\right)e^{iz/2z_o}$$

- Which shows a half-width of $\mathbf{\Delta t_{FWHM,sol} = 1.76\,\Delta t_{sol}}$
- For the fundamental soliton it has to fulfill the following conditions

Fundamental soliton

$$I_{sol,o} = \frac{1}{\Delta t_{sol}^2}\frac{2}{\upsilon_o \gamma}\left|\frac{dn}{d\upsilon}\right|$$

- With the important consequence of quadratically increasing soliton pulse energy for shorter pulse duration. (see fig. in the text).
- This fundamental soliton propagates without any changes in shape and energy through the matter, which is assumed to be without absorption.

- Pulse energies between these discrete values may complicated.
- The characteristic soliton period Z_{per} is:

soliton period

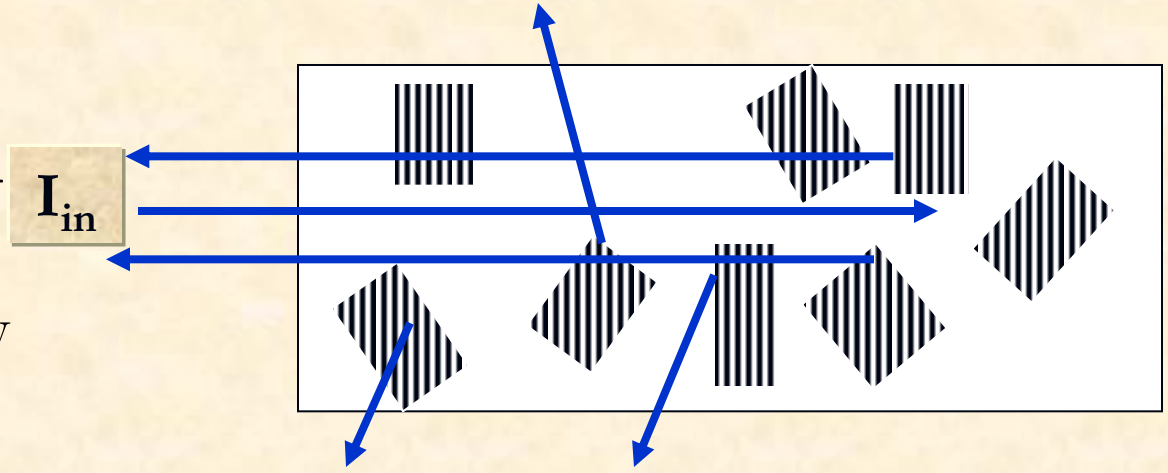
$$Z_{per} = \pi\, z_o = \frac{\pi c_o^2}{4}\left|\frac{dn}{d\upsilon}\right|^{-1} \Delta t_{sol}^2$$

- Which describes periodically the return of the original pulse shape for pulses different from the fundamental soliton by conditioning more energy. In between, the pulse may be split into different pulses and may recover its original shape.
- Solitons in optical fibers can be used for transmitting optical signals over long distances with very high bit rates based on very short pulses.
- They can also be applied for the generation of short pulses with a duration of a few fs in soliton laser.

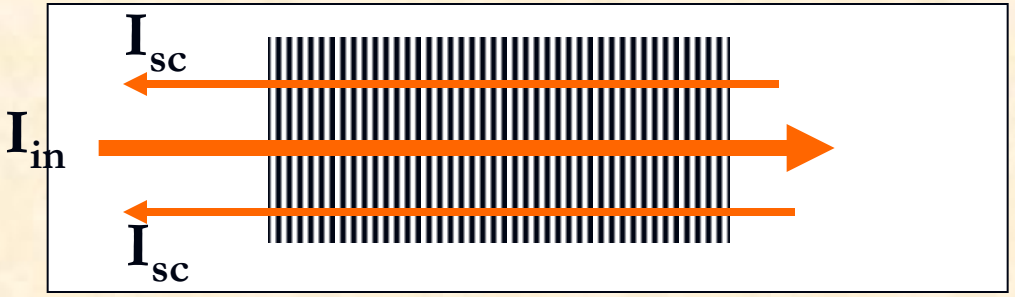
Stimulated Brillouin Scattering "SBS"

- It occurs as amplified spontaneous Brillouin scattering at high intensities of the incident light wave, which increases the sound wave amplitude. It occurs also as spontaneous scattering in the nonresonant spectral range. But SBS is an inelastic optical scattering process with very small energy loss.
- SBS is applied in phase conjugating mirrors (PCM).
- The third-order nonlinear process can be imagined in four steps which take place simultaneously:

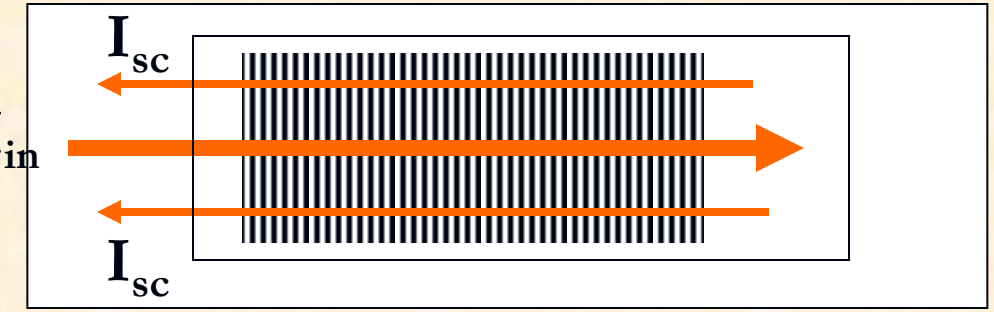
1) A small share of the spontaneous scattered light will exactly be back-reflected towards the incident beam (frequency shifted).



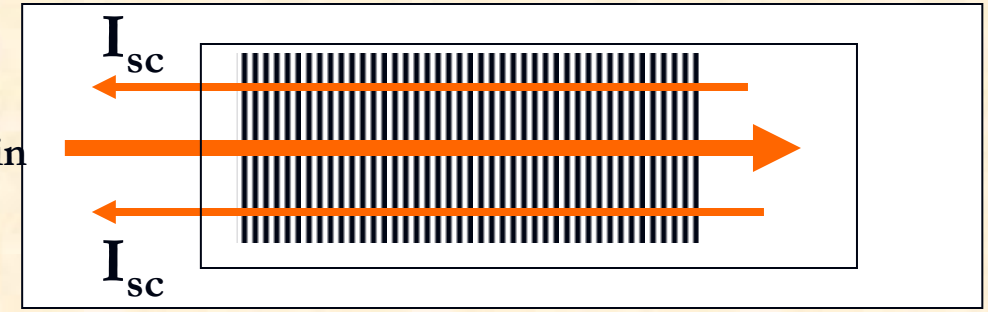
2) Incident and reflected light waves interfere and generate a moving intensity grating.



3) An intensity grating amplifies the suitable sound wave for back-scattering via electrostriction.



4) The amplified sound wave increases the share of the exactly back-scattered light which leads to more interference and process continues



- Because of the perfect back scattering in SBS the scattering angle is 180^o and λ of the sound wave Λ_{sound} results from the λ of the incident light wave λ_{inc} :

Sound wavelength

$$\Lambda_{sound} = \frac{1}{2}\lambda_{inc}$$

- The sound wave frequency Ω_{sound} follows from sound velocity υ_{sound}:

Sound frequency

$$\Omega_{sound} = 2\frac{\upsilon_{sound}}{\lambda_{inc}} = 2\frac{\upsilon_{sound} n \upsilon_{inc}}{c_o}$$

- And is in the range of several 100 MHz for gases and up to several 10 GHz for liquids and solids. The energy loss (10^{-4} – 10^{-6}) can be neglected.
- The potential linewidth of the SBS $\Delta\Omega_{sound}$ measured at 1/e of the peak value is a function of the life time of sound wave τ_{sound}:

SBS linewidth

$$\Delta\Omega_{sound} = \frac{1}{2\pi\tau_{sound}}$$

- Which is twice the lifetime of the phonons:

Phonon lifetime

$$\tau_{SBS} = \frac{1}{2}\tau_{sound} = \frac{k_{sound}^2 \eta}{\rho_o}\alpha\frac{1}{\lambda_{inc}^2}$$

- η : is the material viscosity
- The reciprocal quadratic λ dependency is fulfilled in near UV to IR.
- The theoretical description is based on the wave equation of nonlinear optics:

$$\Delta E - \frac{1}{c_o^2}\frac{\partial^2 E}{\partial t^2} - grad\ div\ E = \frac{1}{\varepsilon_o c_o^2}\frac{\partial^2 p_{nl}}{\partial t^2}$$

- P_{nl} : represents the modulation of the material density ρ by electrostriction from E_{tot} of the interference pattern of the incident and the scattered light by:

$$P_{nl} = \varepsilon_o E_{tot}\left(\frac{\partial \chi^{(2)}}{\partial \rho_{mat}}\right)_T \bar{\rho}_{mat} = \varepsilon_o E_{tot}\frac{\gamma^e}{\rho_{mat,o}}\bar{\rho}_{mat}$$

- $\bar{\rho}_{mat}$: The average density modulation, $\rho_{mat,o}$ the average density and γ^2 the coefficient of electrostriction.
- It is assumed that the variation of X with T can be neglected compared to the density modulation

$$T\left(\partial\chi/\partial T\right)_{\rho} << \rho_{mat}\left(\partial\chi/\partial\rho_{mat}\right)$$

- $\bar{\rho}_{mat}$: can be calculated for small changes from "Navier-Stokes" equation including the equation of continuity:

$$-\frac{\partial^2 \bar{\rho}_{mat}}{\partial t^2}+\left\{\frac{C_v}{Cp}\upsilon_{sound}^2+\frac{\eta}{p_{mat,o}}\frac{\partial}{\partial t}\right\}\Delta\bar{\rho}_{mat}=\frac{\varepsilon_o\gamma^e}{2}\Delta\left|E_{total}\right|^2$$

The material viscosity

$$\eta=(4/3)_{7/8}+n_b$$

- The total E results from the interference of the incident and reflected light beams which are assumed monochromatic with υ_{inc} and $\upsilon_{scat.}$ and the wave vector values ki propagating in the z-direction:

$$E_{total} = E_{inc}(r,t)e^{i(2\pi\upsilon_{inc}t - k_{inc}z)} + E_{scat}(r,t)e^{i(2\pi\upsilon_{scalt}t + k_{sat}z)}$$

- The polarization of the light is not changed in the SBS process.
- The resulting sound wave will show **(½) λ_{inc}** and a wave vector value of:

$$k_{SBS} = k_{inc} + k_{scatt} \approx 2k_{inc}$$

- The sound wave density will be replaced by a normalized sound wave amplitude S.

Sound wave amplitude

$$S(r,t) = \frac{2\pi\upsilon_{inc}\gamma^{e}}{2c_{o}n\rho_{mat,o}}\rho_{mat}(r,t)$$

Stationary Brillouin gain

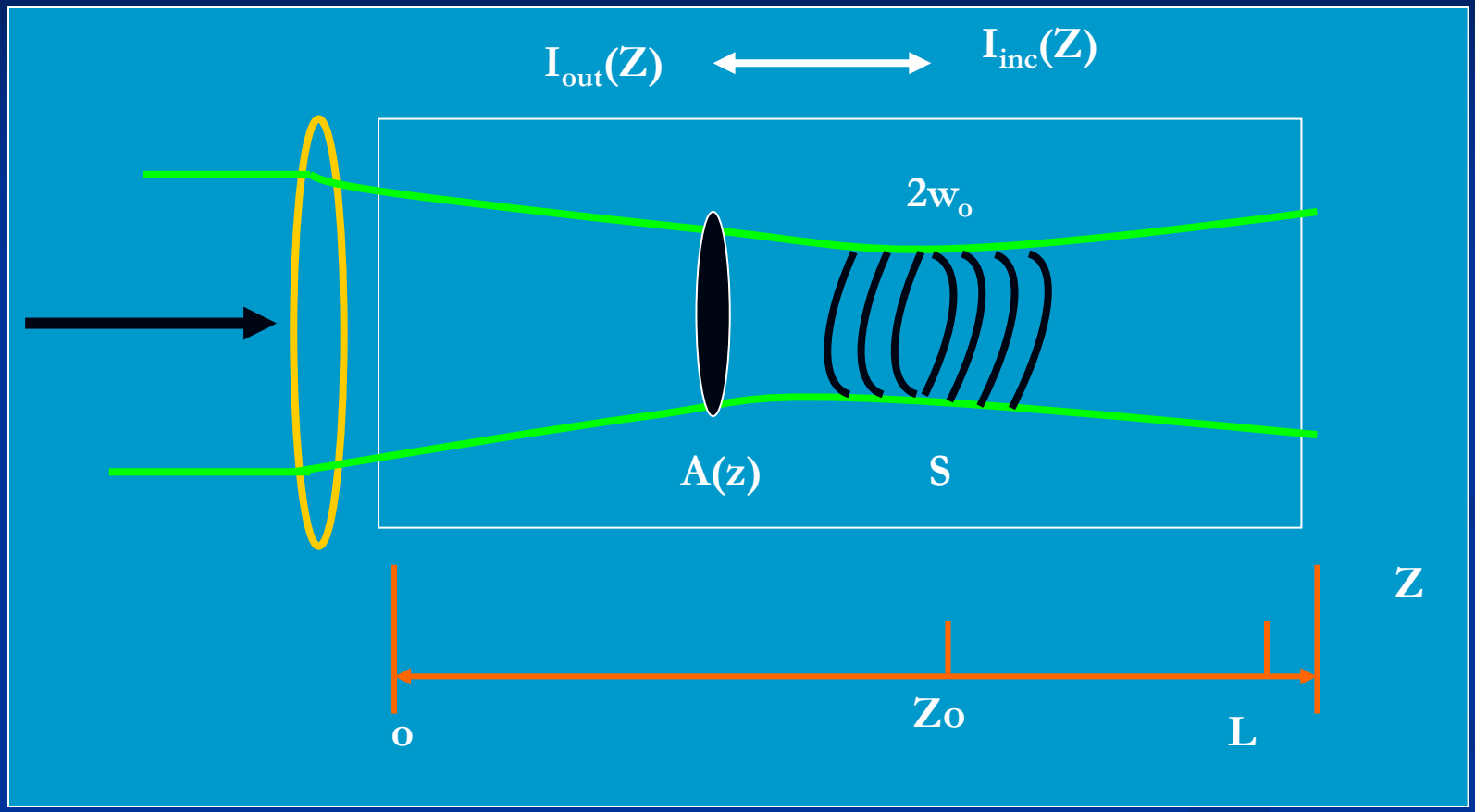
$$g_{SBS} = \frac{(2\pi\Omega_{sound}\gamma^{e})\tau_{sound}}{c_o^2 n \rho_{mat,o} \upsilon_{sound}}$$

- Which has the same **Lorentzian** spectral profile as the spontaneous scattering Brillouin line and the spontaneous sound amplitude S_o relevant for SBS process to start the self pumped scattered from noise.
- For thermodynamic calculation:

$$S_o = \left(e^{(1-h\Omega_{sound}/kT)} + 1\right) g_{SBS} h\Omega_{sound} \frac{1}{\tau_{sound}} \frac{L_{\text{interaction}}}{4A_{\text{interaction}}}$$

- Useful parameter for common SBS material are given in table (see text).
- The sound wave life time scales to a good approximation with λ^2_{inc} over the range of UV to near IR. For gases g_{SBS} is proportional to $p^2_{mat,0}$ and the life time is proportional to p_{mat} .

- For practical solution including focusing the fig. was used.



- The following equation are obtained:

$$\frac{\partial I_{inc}(z,t)}{\partial z} - \frac{n}{c_o}\frac{\partial I_{inc}(z,t)}{\partial t} = -S(z,t)\sqrt{I_{inc}I_{scat}} - \frac{I_{inc}}{A(z)}\frac{\partial A(z)}{\partial z}$$

$$\frac{\partial I_{scat}(z,t)}{\partial z} - \frac{n}{c_o}\frac{\partial I_{scat}(z,t)}{\partial t} = -S(z,t)\sqrt{I_{inc}I_{scat}} - \frac{I_{scat}}{A(z)}\frac{\partial A(z)}{\partial z}$$

$$\frac{\partial S(z,t)}{\partial t} = \frac{1}{2\tau_{sound}}\left\{ g_{SBS}\sqrt{I_{inc}I_{scat}} - \left[S(z,t) - S_o\right]\right\}$$

- With the assumption of a coherence length longer than the interaction length. For spatial and temporal distributions, (see examples in text).
- It was shown that the reflectivity is almost not a function of the focusing as long as the coherence length is longer than interaction time. (see example and fig. in the text).

- In the case of stationary SBS the differential equation can be approximated for nondepleted incident light.

$$I_{sat}\left(L_{interaction}\right) = I_{sat.,spont}(z_o)e^{g_{SBS}I_{inc}L_{interaction}}$$

- Assuming the incident beam and the reflected beam interface coherently.
- The total stationary **SBS-gain G_{SBS}** results from:

Stationary SBS-gain

$$G_{SBS} = g_{SBS}I_{inc}L_{interaction}$$

- The stationary "SBS –threshold" power P_{th} can be estimated from this formula by considering the spontaneous reflectivity useful for starting the SBS.

Stationary SBS-threshold

$$P_{th} \approx 20\frac{A_{interaction}}{g_{SBS}L_{interaction}}$$

- Typical $A_{interaction}$ in SBS with focused beam are of 10^{-5} and the interaction lengths are a few mm.
- For gases and solids 100 kW to MW and for liquids as low as 10 kW.
- In nonstationary SBS with pulses shorter than the life time of the sound wave the threshold increases with the ratio of the photons lifetime divided by the pulse width. Thus, SBS with ps or fs laser pulses shows very small reflectivities.
- The $A_{interaction}$ /$L_{interaction}$ ratio can be decreased many order of magnitudes by using waveguide structures as SBS reflectors. (see example in the text).
- For application of SBS mirrors in OPC the "SBS threshold" is an important parameter.
- SBS reflection in general does not change the polarization and works best for linearly polarized light.
- For several other properties of SBS materials (see the text).
- The coherence length of the pump laser light limits the maximum interaction length and then the reflectivity.

Stimulated Thermal Brillouin Scattering "STBS"

- If a small absorption in the SBS matter is present at the wavelength of the incident light the scattering can be enhanced by a temperature grating in addition to the usual sound wave or density grating.
- The wave equation can be written as:

$$\Delta E - \frac{1}{c_o}\frac{\partial^2 E}{\partial t^2} - \frac{a}{c_o}\frac{\partial E}{\partial t} - grad\ divE = \frac{1}{\varepsilon_o c_o^2}\frac{\partial^2 p_{nl}}{\partial t^2}$$

- a Is the absorption coefficient.

- The Navie-Stokes equation including the equation of continuity then:

$$-\frac{\partial^2 \bar{\rho}_{mat}}{\partial t^2}+\left\{\frac{C_v}{C_p}\upsilon_{sound}^2+\frac{\eta}{\rho_{mat,o}}\frac{\partial}{\partial t}\right\}\Delta\bar{\rho}_{mat}+\frac{C_v}{C_p}\upsilon_{sound}^2 B_T \rho_{mat,o}\Delta\tau$$
$$=\frac{\varepsilon_o\gamma^e}{2}\Delta\left|E_{tot}\right|^2-\frac{\varepsilon_o}{2}\left(\frac{\partial\chi^{(3)}}{\partial T}\right)_\rho\nabla(E^2\nabla T)$$

- B_T Is the coefficient of thermal expansion.
- After mathematical treatments, the equation for the electric fields of the incident and scattered light beams in the plane wave:

$$\frac{\partial E_{scat}(r,t)}{\partial z}+\frac{n}{c_o}\frac{\partial E_{scat}(r,t)}{\partial z}+\frac{a}{2}E_{scat}(r,t)$$
$$=i\frac{2\pi^2\varepsilon_o\upsilon_{scat}}{c_o n}\left[\frac{\gamma^e}{\rho_{mat,o}}\rho_{mat}\left(\frac{\partial\chi^{(3)}}{\partial T}\right)_{\rho_{mat}}T\right]E_{scatt}$$

Notice : same equation for $E_{inc}(r,t)$

- It may be noticed that the sum of the incident and scattered light is no longer constant along the z axis because of absorption.
- A simple approximation can be reached using the phenomenological equation:

$$\frac{\partial I_{scat}(z,t)}{\partial z} - \frac{n}{c_o}\frac{\partial I_{scat}(z,t)}{\partial t} - aI_{scat}(z,t)$$
$$= -S(z,t)\sqrt{I_{inc}I_{scat}} - \frac{I_{scat}}{A(z)}\frac{\partial A(z)}{\partial z}$$

Notice : same equation for I_{inc}.

- The SBS gain factor g_{SBS} contains two parts from electrostriction g^e_{SBS} and from absorption g^a_{SBS} :

$$g_{SBS} = g^e_{SBS} + g^a_{SBS}$$

With the additional components:

$$g^a{}_{SBS} = a\frac{\pi \upsilon_{inc}\tau_{SBS}\gamma^e B_T}{c_o n \rho_{mat,o} c_p}$$

This part shows asymmetric wavelength dependence

Stimulated Rayleigh "SRLS" and Thermal Rayleigh Scattering "STRS" :

- It takes place in the direct spectral neighborhood of the incident light frequency, thus it is almost elastic.
- The stimulation results from the change in matter density via electrostriction or thermal effects.
- The stationary gain for stimulated Rayleigh scattering g_{SRLS} is:

$$g_{SRLS} = (g^{e}_{SRLS,o} - g^{a}_{SRLS}) \frac{4\pi \upsilon_{inc} \tau_{RL}}{1 + (4\pi \upsilon_{inc} \tau_{RL})^2}$$

- τ_{RL} the life time of the Rayleigh scattering as the reciprocal spectral width:

$$\tau_{RL} = \frac{1}{2\pi \Delta \upsilon_{RL}} = \frac{\rho_{mat,o} C_p \lambda^2_{inc}}{16\pi^2 \Lambda_T (\sin^2(\theta_{scat/2}))}$$

- Which is in the range of 10 ns and with the two peak gains:

$$g^{e}_{SRLS,o} = \frac{4\pi^2 \upsilon^2_{inc} \tau_{RL} \gamma^e \gamma^{RL}}{c^3 n \upsilon_{sound} \rho_{mat,o}} \quad and \quad g^{a}_{SRLS,o} = \frac{4\pi^2 \upsilon^2_{inc} \tau_{RL} \gamma^e \gamma^a}{c^3 n \upsilon_{sound} \rho_{mat,o}}$$

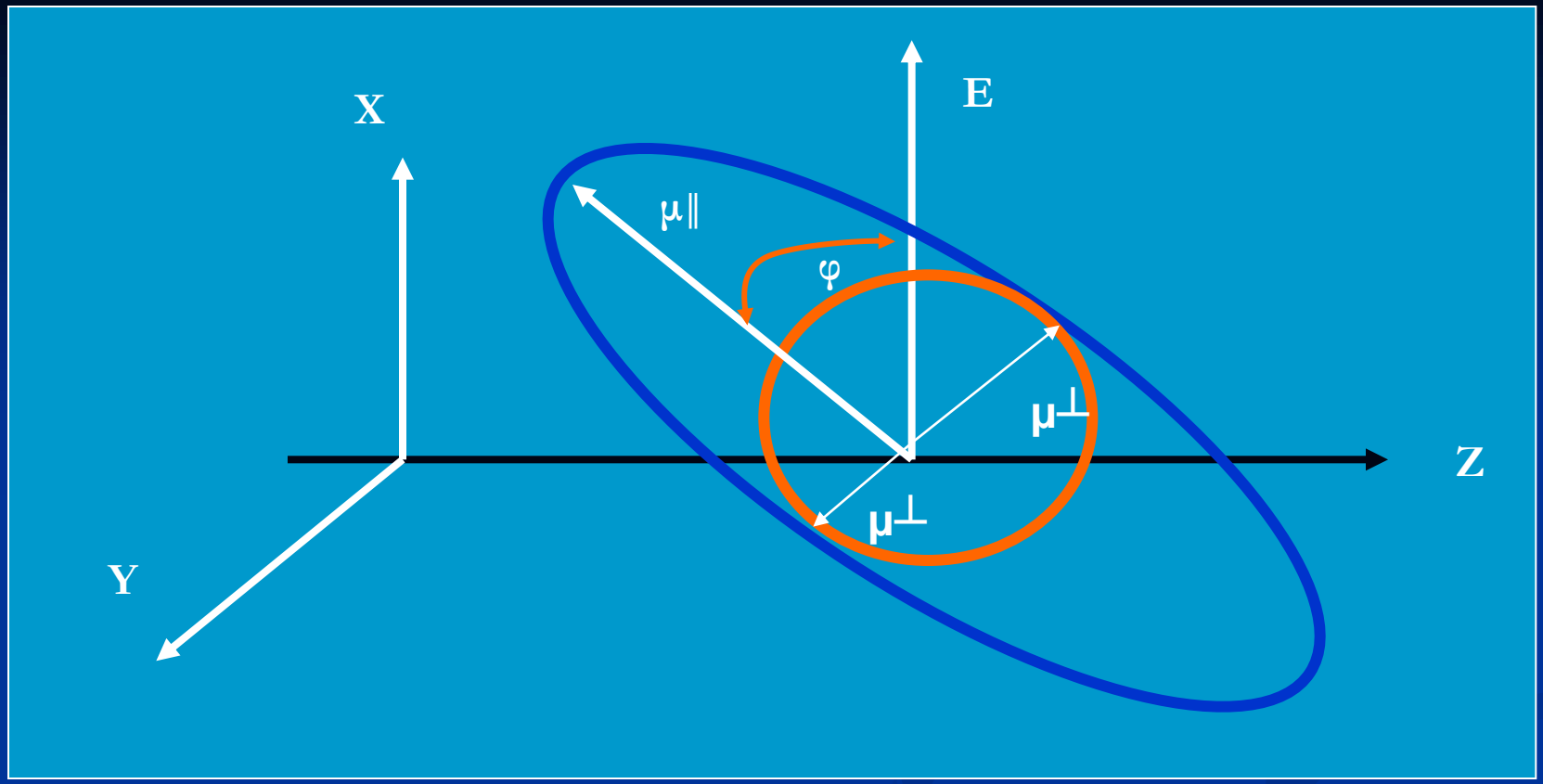
With the coefficients:

$$\gamma^{RL} = \frac{\left(c_p / c_o - 1\right) c_o \gamma^e}{8\pi\, n \upsilon_{sound} \upsilon_{inc} \tau^{RL}} \quad and \quad \gamma^a = a \frac{c_o^2 \upsilon_{sound} \beta_T}{2\pi\, c_p \upsilon_{inc}}$$

- Even for small absorption $a \approx 10^{-3}\ cm^{-1}$ the absorptive term can dominate the gain factor.
- Backward scattering will show shorter τ and smaller g than forward scatter.
- Because of the long lifetimes the Rayleigh scattering will be nonstationary.
- Because of small gain coefficients, stimulated Rayleigh scattering is almost not observable in nonabsorbing material.

Stimulated Rayleigh Wing Scattering (SRWS):

- It produced by orientation fluctuations of particles with nonisotropic induced dipole moment (e.g. anisotropic molecules) can be easily stimulated with high light fields and SRWS can be observed.
- The anisotropic E vector of the light beam changes the angular orientation of these particles and thus a local variation of n will be induced.
- The driving force for the SRWS results from the interaction of the E vector $\mathbf{E}_{\mathbf{inc}}$ with the vector of the polarizability μ_{mol} of the molecular resulting in a torque M as shown in Fig.



- The torque can be calculated:

$$M = -\left[\mu_{mol,ll} - \mu_{mol,\perp}\right] \left|E_{inc}\right|^2 \cos\varphi \sin\varphi$$

- For a simple theoretical description of SRWS, light linearly polarized in x direction and propagating in z direction is assumed. The angular distribution function $\mathbf{f_{mol}(\varphi)}$ for the molecular orientation has to fulfill:

$$\frac{\partial f_{mol}}{\partial t} - \frac{1}{2\tau_{orientation}\sin\varphi}\frac{\partial}{\partial\varphi}\left[\sin\varphi\left(\frac{\partial f_{mol}}{\partial\varphi} - \frac{M}{kT}f_{mol}\right)\right] = 0$$

- $\boldsymbol{\tau_{Orientation}}$ is the relaxation time of the orientation of molecules.
- For known f the $\mathbf{P_{nl}}$ of matter as a function of E can be written:

$$P_{nl} = 2\pi\varepsilon_o N_{mol} E_{inc}\int_{-1}^{1} f_{mol}(\varphi, E_{inc})\cos^2\varphi\, d(\cos\varphi)$$

- $\mathbf{N_{mol}}$ is the density of molecules.:

- The total E consists of the incident wave, the Stokes wave with the index S and the anti-stokes wave with the index aS:

$$E_{tot} = E_{inc,o} e^{i(2\pi \upsilon_{inc} t - k_{inc} r)} + E_{\partial,o} e^{i(2\pi \upsilon_s t - k_s r)} + E_{as,o} e^{i(2\pi \upsilon_{as} t - k_s r)}$$

- And under the assumption of a nondepleted strong incident field two coupled equations can be derived:

$$2ik_s^2 \frac{dE_s}{dz} - (k_s^x)^2 E_s = -\frac{\Delta n_{SRWS}^2 \pi^2 \upsilon_s^2}{c_o^2} \frac{|E_{inc}|^2 E_s + E_{inc}^2 E_{as}^*}{1 + i2\pi \Delta \upsilon_{RW} \tau_{RW}}$$

- And similar equation for Eas.
- With the change of permittivity Δn_{SRWS} :

$$\Delta n_{SRWS}^2 = \frac{\varepsilon_o N_{mol} (\mu_{11} - \mu_1)}{90kT}$$

- And the relations:

$$\partial \upsilon_{Rw} = \upsilon_{inc} - \upsilon_s = \upsilon_{as} - \upsilon_{inc}$$

$$k_s^x = -k_{as}^x << k_{inc}$$

$$\frac{C_o^2 (k_i^2)^2}{4\pi^2 \upsilon_i^2} - n_o^2 = \frac{1}{2} \Delta n_{SRWS}^2 \left| E_{inc} \right|^2 \quad with \;\; i = S, aS$$

- If large scattering angles between the Stokes and the anti-Stokes waves are investigated, the coupling between them can be neglected.
- The gain factors for the case of parallel polarization of incident and scattered waves are:

$$g_{SRWS,S} = \frac{4\pi^2 \upsilon_s \Delta n_{SRWS}^2}{c_o^2 n_o^2} \frac{2\pi \Delta \upsilon_{Rw} \tau_{Rw}}{1 + (2\pi \Delta \upsilon_{Rw} \tau_{Rw})^2} \approx -g_{SWRS,aS}$$

- The gain factors are comparable to SBS gain factors and high enough for experiments with pulsed lasers.
- The orientation relaxation times are in the ps range.
- In the near-forward direction Stokes and anti-Stokes photons will be generated by the scattering of two incident photons.
- The scattering is a maximum for an angle $\theta_{SRWS,max}$ of :

$$\theta_{SRWS,\max} = \pm \frac{1}{\sqrt{2}} \frac{\Delta n_{SRWS} |E_{inc}|}{n_o}$$

- Which is in the range of a few mrad.

Stimulated Raman Techniques

- Stimulated Raman Scattering "SRS" is usually applied in the nonabsorbing material, the energy change of the scattered photons compared to the incident photons can be as large as 10%.
- SRS process can be understood as scattering coupled with a matter transition between two vibrational energy states with energy difference $E_{vib}=h\upsilon_{vib}$ via a virtual energy states of more than times this energy.

Stimulated Raman Scattering "SRS"

- It takes place with the excitation of the vibration or of an optical phonon (Stokes SRS results in smaller photon energy) or by its depletion (anti-Stokes SRS)

Energy condition :

$$\upsilon_{scatt,SRS} = \upsilon_{inc} \neq m\upsilon_{mb} \quad with\ m = 1,2,3,....$$

Whereas subsequent scattering or nonlinear coupling of molecular vibration allows multiple frequency shift with m > 1.

The intensity of scattered light in SRS can be amplified by many orders of magnitude from spontaneous Raman scattering up to 10 % of the intensity of the incident light.

Under certain circumstances strength of the anti-Stokes SRS can be comparable to the Stokes SRS.

SRS is useful in cases of difficult spontaneous Raman scattering measurements for improving the S/N ration and shortening the measuring time by many order of magnitude.

The amplification in SRS strongly depends on the scattering cross-section $d\sigma_{SRS}/d\Omega_{scatt}$, the incident intensity I_{inc} and the interaction length $L_{interaction}$.

- Therefore the amplification can be described by the Raman gain coefficient g_{SRS}. Below incident intensities causing saturation, which starts roughly at 10^{-1} of the saturation limit:

Below saturation

$$I_{Scatt,SRS} = I_{Scatt,Spont} e^{g_{SRS} I_{inc} L_{interaction}}$$

- $\mathbf{I_{scatt,spont}}$: is about $10^{-6} – 10^{-10}$ of the saturation.
- The stimulated new Raman photon has the same direction, frequency and polarization as the stimulated one.
- Typically SRS gain coefficients are in the order of $\mathbf{g_{SRS} \approx 10\ cm\ GW^{-1}}$
- Thus, incident intensities of several 100 $MWcm^{-2}$ are necessary for strong effects with interaction lengths in the cm range.
- (see the table in the text)
- Because of the nonlinear interaction the linewidth of the SRS is smaller then the linewidth of spontaneous Raman scattering.

- Besides, energy conservation for stimulated anti-Stokes Raman scattering.

Energy condition

$$2\upsilon_{inc} = \upsilon_{SRS,stokes} + \upsilon_{SRS,anti-stokes}$$

- The conservation momentum has to be considered.
- Thus, the wave vectors have to fulfill the condition:

$$k_{inc} + k_{inc} = k_{SRS,stokes} + k_{SRS,anti-stokes}$$

- With the wave vectors:

$$k_m = \frac{2\pi\, n(\upsilon_m)\upsilon_m}{c_o} e_m$$

- Pointing in the propagation direction e_m of the mth beam.

Anti-stokes angle

$$\theta_{SRS,anti-stokes} = arc\ \cos\left[\frac{4k_{inc}^2 - k_{SRS,anti-stokes}^2 - k_{SRS,stokes}^2}{4k_{inc}k_{SRS,anti-stokes}}\right]$$

- Which is different from zero as a consequence of different n`s λ`s for different (see numerical examples in the text)
- The electric field amplitude E in combination with $\mathbf{P_{nl}}$ but including some absorption with the coefficient a and assuming plane wave, the wave equation is:

$$\Delta E - \mu_o \varepsilon_o \frac{\partial^2 E}{\partial t^2} - a\sqrt{\mu_o \varepsilon_o}\frac{\partial E}{\partial t} = \mu_o \frac{\partial^2 P_{nl}}{\partial t^2}$$

- For the electric field E of the light beam with planar wave front propagating in the z-direction:

$$E(z,t) = \sum_m E_{j,o} e^{i(2\pi \upsilon_m t - k_m z)}$$

- And for nonlinear polarization $\mathbf{p_{nl}}$:

$$P_{nl}(z,t) = \sum_m P_{\mathrm{nln},o} e^{i(2\pi \upsilon_m t)}$$

- It follows that for the spectral components j using SVA approximation and neglecting the second time derivatives of E:

$$\frac{\partial E_m}{\partial z} - \mu_o \varepsilon_o \frac{\partial^2 E_m}{\partial t^2} - \frac{a}{2} E_m = i \frac{\mu_o \pi \upsilon_m}{c_o} \frac{\partial^2 p_{nlm}}{\partial t^2} e^{ik_m z}$$

- The interaction of the light with matter is obtained from the forced expectation value of the displacement q_{vib} in the vibration and relative population differences $\mathbf{\Delta N=(N_{ground}-N_{exc}) / N_{ground}}$ in the vibrational two level system. The displacement $\mathbf{q_{vib}}$ from a damped wave equation:

$$\frac{\partial^2 q_{vip}}{\partial t^2} + \frac{1}{\tau_{vib}} \frac{\partial q_{vip}}{\partial t} + 4\pi^2 \upsilon_{vib}^2 q_{vib} = \frac{1}{2m_{vib}} \frac{\partial \mu_{vip}}{\partial q_{vib}} \Delta N E^2$$

- With the lifetime of the vibration $\tau_{vib}.\partial\mu_{vib}/\partial q_{vib}$ gives the polarizability of this vibration. **ΔN** Can be calculated from a rate equation.
- The nonlinear polarization follows from the solution of this equation by:

$$P_{nl} = N_{tot} \frac{\partial \mu_{vip}}{\partial q_{vib}} q_{vib} E$$

- N_{tot}: Is the total density of vibrating particles per unit volume.
- Assuming forward (fw) and backward (bw) scattering along z-axis.
- The electric field resulting from Stokes scattering of SRS is:

$$E = E_{inc} e^{i(2\pi \upsilon_{inc} t - k_{inc} r)} + E_{stokes,\,fw} e^{i(2\pi \upsilon_{stokes} t - k_{stokes,\,fw} z)} + E_{stokes,bw} e^{i(2\pi \nu_{stokes} t - k_{stokes,bw} z)}$$

- With all E vectors pointing in the direction of the incident field and:

$$q_{vib} = \frac{1}{2}\left\{q_{fw}e^{i(2\pi\upsilon_{vib}t - k_{vib,fw}z)} + q_{bw}e^{i(2\pi\upsilon_{vib}t - k_{vib,bw}z)} + c.c\right\}$$

- Using the differential equations of the electric fields:

$$\frac{n_{inc}}{c_o}\frac{\partial E_{inc}}{\partial t} + \frac{\partial E_{inc}}{\partial z} = \frac{i2\pi^2\upsilon_{inc}}{n_{inc}c_o}\Delta N\frac{\partial \mu_{vip}}{\partial q_{vib}}(q_{fw}E_{fw} + q_{bw}E_{bw}) - \frac{a}{2}E_{inc}$$

$$\frac{n_{stokes,s}}{c_o}\frac{\partial E_{fw}}{\partial t} + \frac{\partial E_{fw}}{\partial z} = \frac{i2\pi^2\upsilon_{stokes}}{n_{stokes}c_o}\Delta N\frac{\partial \mu_{vip}}{\partial q_{vib}}q^*_{fw}E_{inc} - \frac{a}{2}E_{fw}$$

$$\frac{n_{stokes,s}}{c_o}\frac{\partial E_{bw}}{\partial t} + \frac{\partial E_{bw}}{\partial z} = \frac{i2\pi^2\upsilon_{stokes}}{n_{stokes}c_o}\Delta N\frac{\partial \mu_{vip}}{\partial q_{vib}}q^*_{bw}E_{inc} - \frac{a}{2}E_{bw}$$

- The vibrational displacements for the Stokes Raman frequency for the two propagation directions are:

$$\frac{\partial q_{fw}}{\partial t} + \frac{1}{2\tau_{vib}} q_{fw} = \frac{i}{8\pi \upsilon_{vib} m} \frac{\partial \mu_{vip}}{\partial q} E_{inc} E^*_{fw}$$

$$\frac{\partial q_{bw}}{\partial t} + \frac{1}{2\tau_{vib}} q_{bw} = \frac{i}{8\pi \upsilon_{vib} m} \frac{\partial \mu_{vip}}{\partial q} E_{inc} E^*_{bw}$$

- If heavily damped vibrations are found, the temporal derivatives of the displacements can be neglected:

$$\frac{\partial q_{fw}}{\partial t} << \frac{q_{fw}}{2\tau_{vib}} \quad and \quad \frac{\partial q_{bw}}{\partial t} << \frac{q_{bw}}{2\tau_{vib}}$$

- Introducing the intensities Im of the different light beams:[2]

$$SRS\ equations\ \frac{n_{stokes}}{c_o}\frac{\partial I_{fw}}{\partial t}+\frac{\partial I_{fw}}{\partial z}=g_{stokes}I_{fw}I_{inc}-aI_{fw}$$

$$\frac{n_{stokes}}{c_o}\frac{\partial I_{bw}}{\partial t}+\frac{\partial I_{bw}}{\partial z}=g_{stokes}I_{bw}I_{inc}-aI_{bw}$$

The gain coefficients g_{SRS} follows with the relation :

$$g_{stokes}=\frac{g_{inc}\nu_{stokes}}{\nu_{inc}}$$

from :

$$g_{SRS}=g_{stokes}=\frac{2c_oN}{h\pi\upsilon_{vib}^3n^2\Delta\upsilon_{SRS}}\left(\frac{d\delta_{SRS}}{d\Omega}\right)_{ll}$$

$$=\frac{8\pi^2\upsilon_{stokes}\Delta N2\tau_{vib}}{n^2c_o^2m\upsilon_{SRS}}\left(\frac{\partial\mu_{SRS}}{\partial q_{vib}}\right)^2\frac{1}{1+B}$$

- The gain depends on the parameters the cross-section and vibrational lifetime or linewidth of the vibration. The cross-section σ_{SRS} can be calculated from:

$$\frac{d\delta_{SRS}}{d\Omega} = \frac{2\pi^2 \upsilon_{stokes} h}{c_o^4 m \upsilon_{SRS}} \left(\frac{\partial \mu_{vib}}{\partial q_{vib}} \right)^2$$

- And the linewidth $\Delta\nu_{vib}$ is related to the lifetime of the vibration τ_{vib} by:

$$\Delta\upsilon_{vib} = \frac{1}{2\pi\tau_{vib}}$$

  - Much stronger SRS can be observed if the frequency of the incident laser is tuned to an absorption transition of the material.
  - Strongly SRS is reached in the materials with large gains (i.e. large dipole moment) and long lifetime.

- Saturation can be reached for high intensities, thus the SRS intensity can be 10^{11} times larger than spontaneous Raman scattering.
- In the case of stationary Stokes scattering with almost undepleted incident light ($I_{fw} << I_{inc}$) and insignificant backward scattering ($I_{bw} << I_{fw}$) can be solved to give:

Stationary small SRS

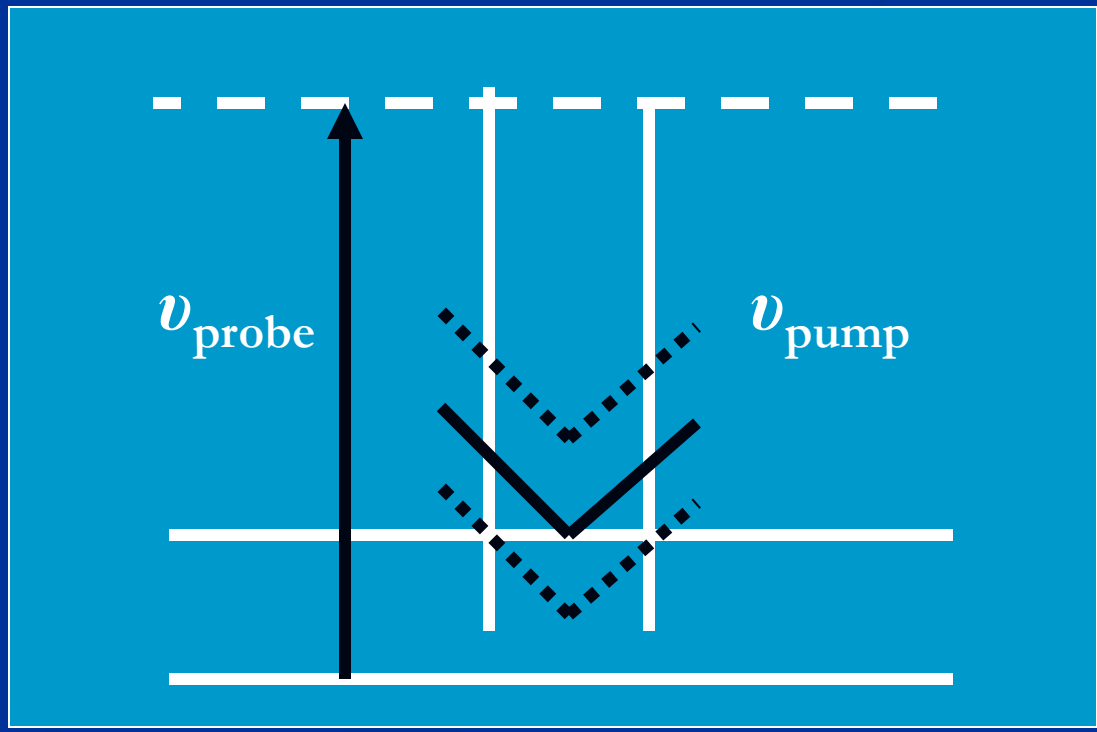
$$I_{fw}(z) = I_{fw}(z=0)e^{(g_{SRS}I_{inc}z - az)}$$

- A most important application of SRS is frequency conversion. If the SRS cell is placed inside a suitable resonator for Stokes or anti-Stokes signal gain may be sufficient for laser action at the wavelengths $\nu_{stokes} = \nu_{inc} - \nu_{vip}$ or $\nu_{anti\text{-}stokes} = \nu_{inc} + \nu_{vip}$
- On the other hand SRS is used for spectroscopic investigations.
- Another important application field may be based on surface-enhanced Raman spectroscopy (SERS).
- From measurements of anti-stokes lines the temperature distribution of samples can be determined with this optical method, e.g. inside combustion energies.

Inverse Raman Spectroscopy (IRS)

- It is obtained if the depletion of a weak probe light signal in the linear intensity range at the frequency of the SRS pump transition is measured while a strong laser is tuned across the Stokes frequency of the matter. (see fig. in the text).

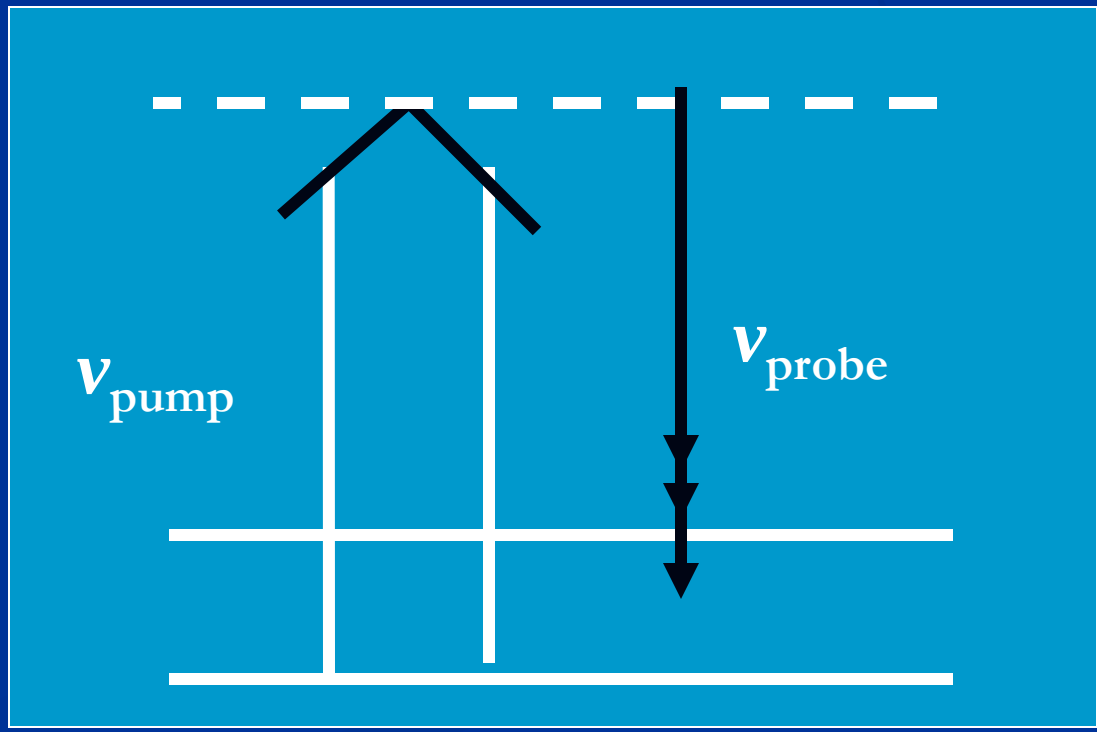
$$T\upsilon_{probe} = f(\upsilon_{pump})$$



Stimulated Raman Gain Spectroscopy (SRGS)

- Another spectroscopic possibility is the measurements of the amplification via the gain coefficient g_{probe} of a weak tunable probe signal with the light frequency around the Raman Stokes signal with the light frequency around the Raman Stokes signal while strong laser pumping with a suitable frequency for this Stokes signal (see Fig.)

$$g_{probe} = f(\upsilon_{pump})$$



Coherent Anti-Stokes Raman Scattering (CARS):

- The combination of simultaneous stimulated Stokes and anti-Stokes Raman scattering leads to the interaction of four photons in the matter
- In coherent anti-Stokes Raman scattering (CARS) two strong laser beams with frequencies υ_{inc} are applied (see fig.)

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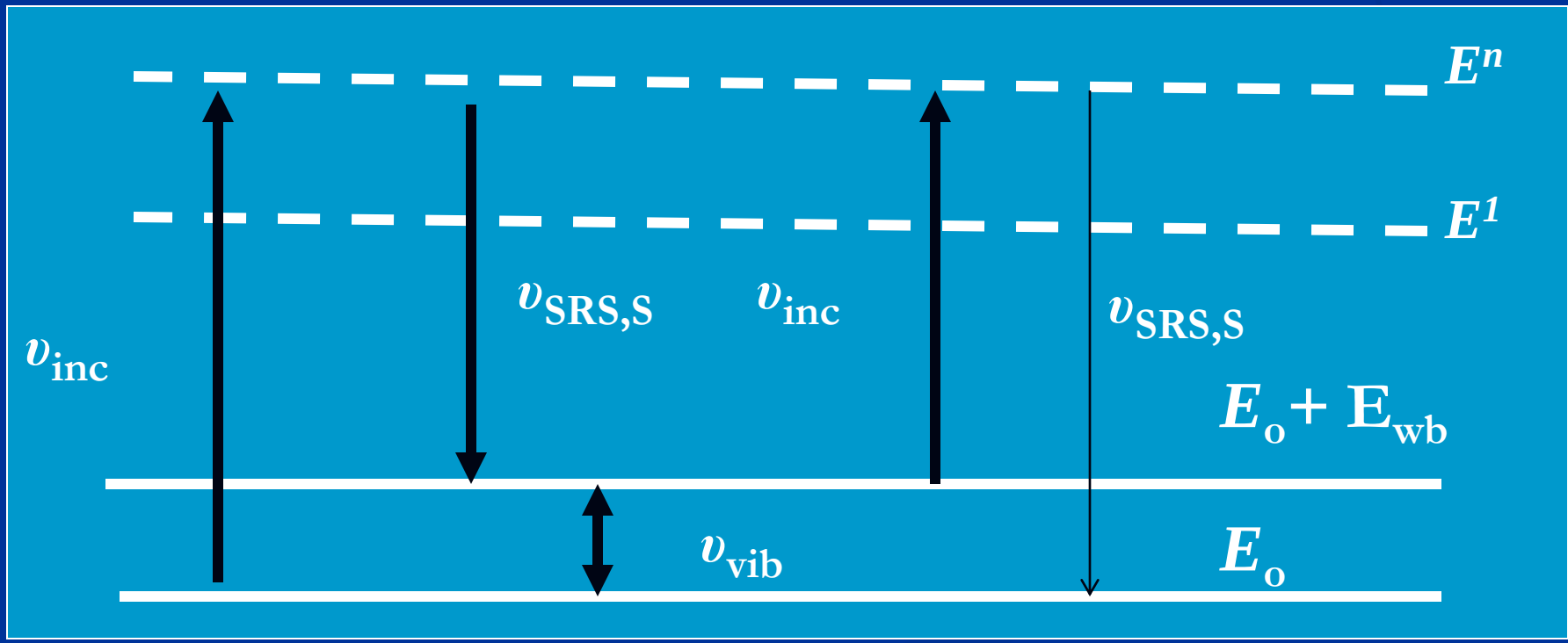
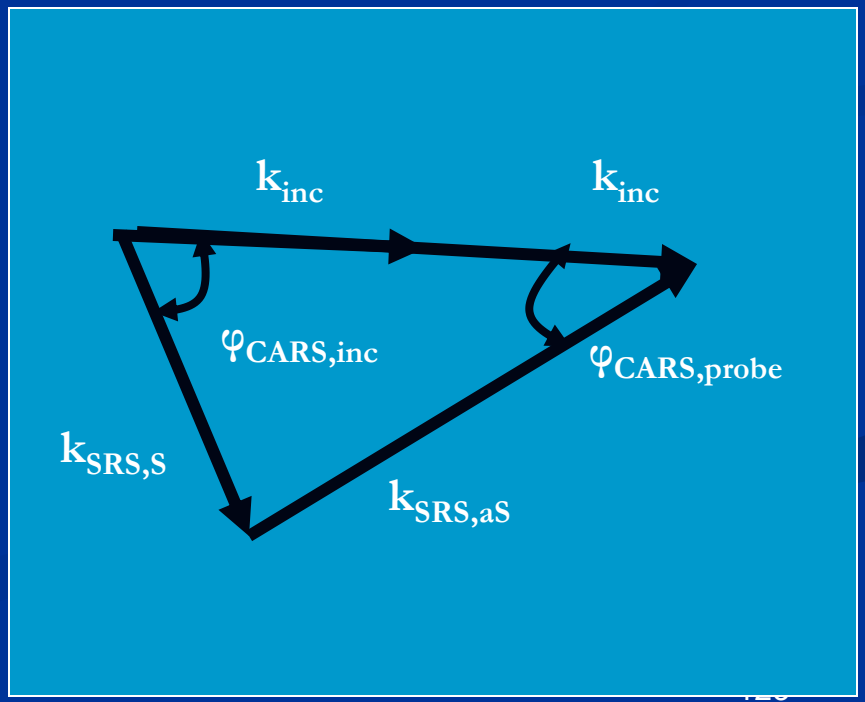


- The coherent scattering of these photons in (CARS) can be applied with very short pulses and allows highly sensitive measurements if phase matching is a achieved. In addition a high spatial resolution in the μm range is possible. CARS is a four-wave mixing (FWM) process.
- Phase matching is achieved if the momentum of the four attended photons are conserved and thus the wave vectors of the incident laser light $\mathbf{k}_{inc}$ and of the Raman Stokes light $\mathbf{K}_{stokes}$ and anti-Stokes light k have to fulfill the angle condition.

Therefore the two incident laser beams have to be enclosed in the angle $\varphi_{CARS,inc}$ and the anti-Stokes Raman light beam can be observed at the angle $\varphi_{CARS,inc}$ to the laser with $\mathbf{K}_{inc}$ in the forward direction.



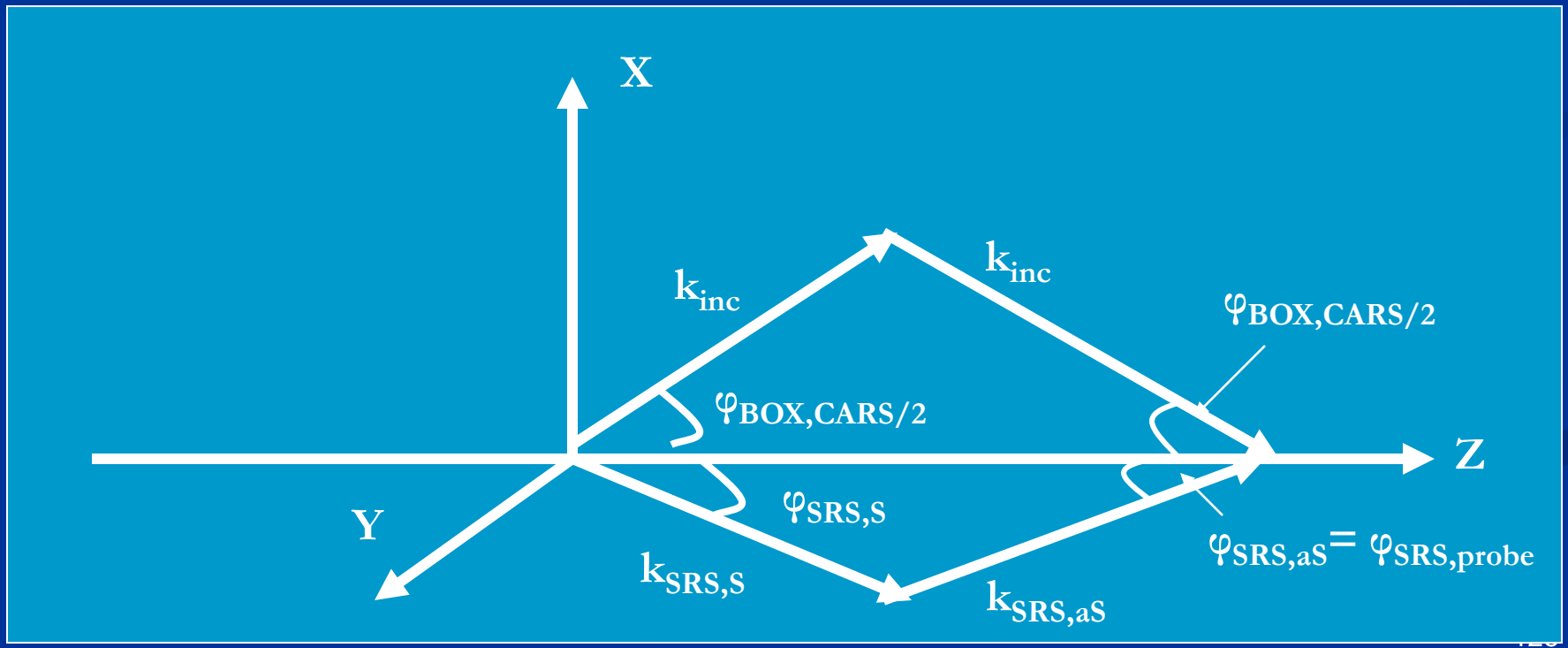
- The scattering intensity $\mathbf{I_{CAR,as}}$ is proportional to :

$$I_{CARS,aS} \ \alpha \ I_{inc}^{2} I_{SRS,S} N_{mat}^{2}$$

- With the pumped laser intensities $\mathbf{I_{inc}}$ of the incident and $\mathbf{I_{SRS,S}}$ of the tuned light and $\mathbf{N_{mat}}$ is particle density.
- The scattering efficiency can be increased by many orders of magnitude if the pump laser energy matcher the electronic transitions of the material (resonant (CARS).
- In this case the virtual Raman levels of the energy schemes above will be real energy states of the matter.

BOX CARS:

- If the CARS scattering angles are too small for safe splitting of the different signals the BOX CARS technique can be used.
- The incident laser beam is split into two beam which are applied at the angle $\varphi_{\text{Box CARS}}$ FIG. below.



- The two pump laser beams with frequency ν_{inc} are applied at the angle $\varphi_{\text{Box CARS}}$ symmetrically in the xz plane each making the angle $\varphi_{\text{Box CARS}\backslash 2}$ each with the z axis the third laser beam with frequency $\nu_{SRS,S} = \nu_{inc} - \nu_{vib}$ can be applied in the yz plane at the angle with the z axis.
- The resulting angle $\varphi_{SRS,aS}$ for detecting the newly generated anti - stoker Raman light with frequency $\nu_{SRS,S}$ can be calculated from:

$$\varphi_{SRS,aS} = arc\ \cos\left\{\frac{1}{1+\left(\upsilon_{vib}\lambda_{inc}/2\right)}\left[2\cos\left(\frac{\varphi_{Box,CARS}}{2}\right)-\left(1+\frac{\upsilon_{vib}\lambda_{inc}}{c}\right)\cos\varphi_{SRS}\right]\right\}$$

- The three beams with their different directions have to overlap.

Optical phase conjugation via stimulated scattering

- The conjugation of the phase of an optical wave is equivalent to inversion of the wave front of the light beam.
- It allows the realization of phase conjugating mirrors (PCMs) which are used in laser for improving the beam quality.
- If the incident light beam is described by:

Incident light $$E_{inc}(r,t) = R_e\left(E_o(r)\ e^{i2\pi\nu t}\right)$$

With the complex amplitude $$E_o(r) = A_o(r)e^{-i(kr+\varphi)}$$

The phase conjugate is given by: $$E_{inc}(r,t) = R_e\ \left(E_o^*(r)e^{i2\pi\nu t}\right)$$

- With the complex conjugated amplitude:

conjugated amplitude

$$E_o^*(r) = A_o(r)\ e^{+i(kr+\varphi)}$$

- Where the sign of the spatial phase is changed.
- The conjugate can also be written as:

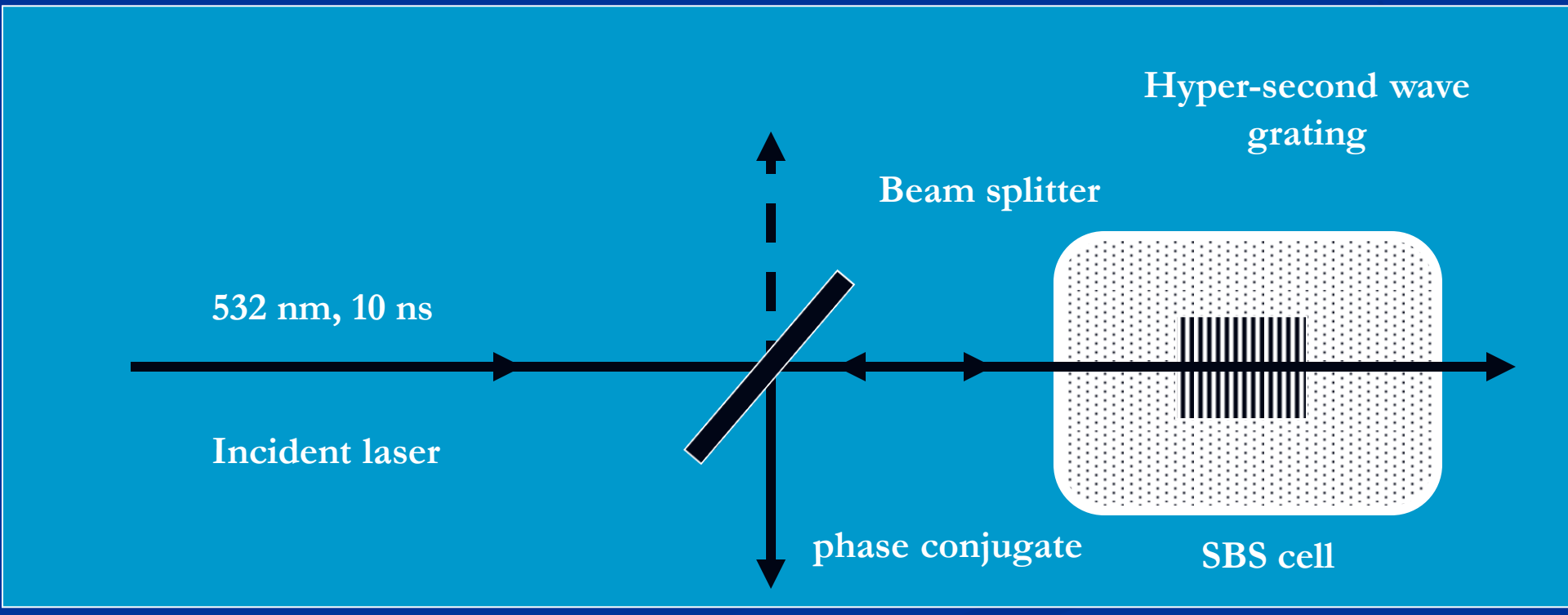
$$E_{inc}(r,t) = R_e\left(E_o(r)e^{-i2\pi\upsilon t}\right)$$

- Where the unchanged spatial amplitude:

$$E_o(r) = A_o(r)\ e^{-i(kr+\varphi)}$$

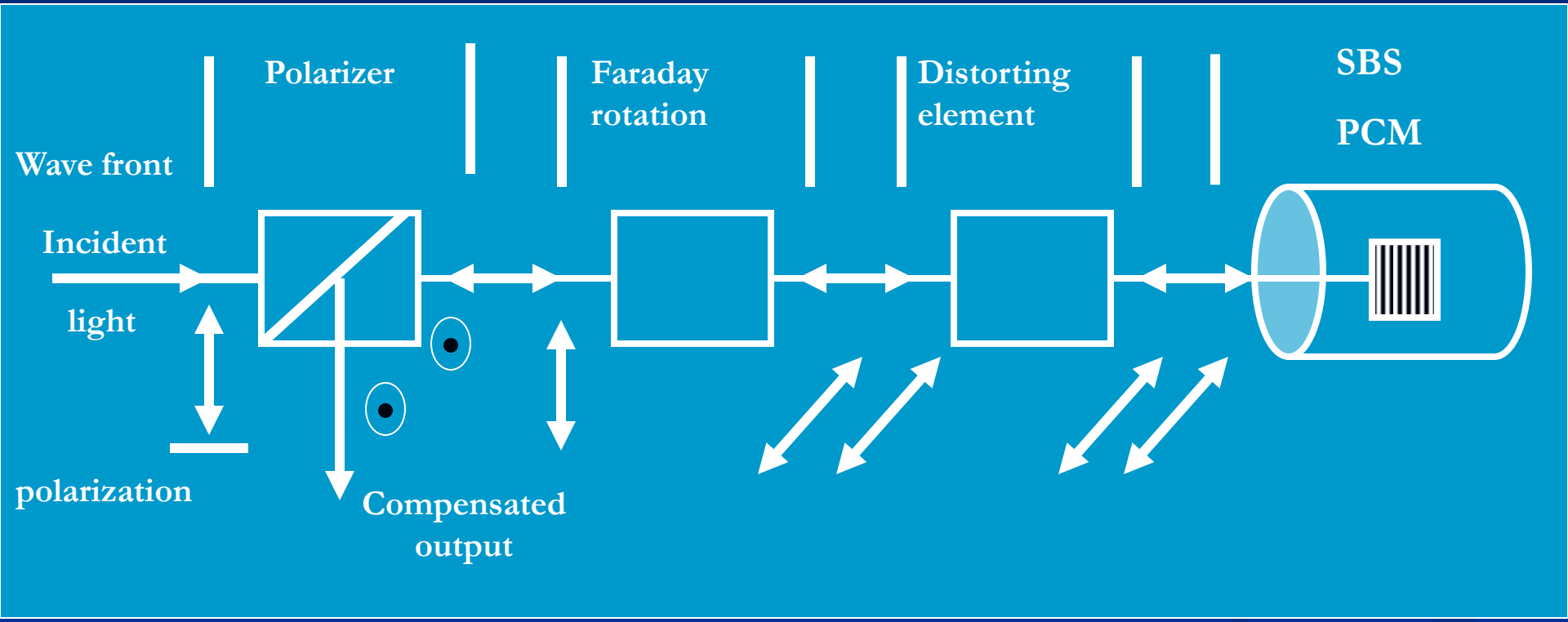
  - But there is a change of sign of the temporal phase.
  - This corresponds formally to a change in time direction indicating that the phase front is moving perfectly backwards.

- This phase conjugation can be achieved with nonlinear back-reflection of the beam via stimulated scattering (.e.g. SBS) or via four-wave mixing (FWM) . Such a volume reflector is called a phase conjugating mirror (PCM). (see e.g. in the text).
- Stimulated Brillouin scattering such as a self-pumped process allows very easy realization of this process.



- A phase conjugating mirror shows unusual properties compared to a conventional mirror (see a fig. of the reflectivity properties of PCM in the text).
- Most obviously the light is perfectly back-reflected by the PCM independent of the direction of the incident light beam.
- More precisely the complete wave front is inverted in the PCM with the important consequence of possible compensation of phase directions in optical element by applying the PCM (see fig. in the text).
- A third important property of the PCM is the treatment of the polarization of the light. (see Fig. in the text).
- In PCMs the linear polarization of light is unchanged and for many PCM processes linear polarized light is most efficient.
- In perfect PMs the circular or elliptic polarization is conserved.
- Thus this type is called (vector phase conjugation). It can be in four-wave mixing schemes only.
- The phase conjugating mirror based on the SBS can be applied for almost perfect compensation of the phase distortions as they are caused by highly pumped solid state laser rods using a double pass arrangement.

- Compensation of the phase distortions takes place if the disturber does not change during the round trip of the light via the PCM which is usually fulfilled if thermal processes are involved.



- The more complicated scheme of **Basov** can be used. (see the text).

Why allows reflection via SBS optical phase conjugation?

- The sound wave rating is stimulated by the intensity interference pattern of the incident and the reflected light. Thus these intensity maximum are higher the better the overlap of the wave fronts of the incident and the reflected beams. Therefore the reflection will reach its highest values if the wave front of the reflected light is identical to the wave front of the incident light although the propagation direction is inverted. This is (optical phase conjugation).
- This effect can be described mathematically under the assumption of a mode mixture for both the incident and reflected light:

$$E_{inc}(r) = \sum_j C_{inc,j}(z) A_{o,inc,j}(r)$$

$$E_{scatt}(r) = \sum_j C_{scatt,j}(z) A_{o,scatt,j}(r)$$

- With the electric fields E_{inc} and E_{scatt} for the incident and back scattered light. The cofficiente Cinc/scatt,j(z) determine the share of the jth mode with the shape Ao , inc/scat,j.
- After mathematical treatments, we can find the build up of the reflected light by scattering is a function of the overlap of the wave front of the incident and scatted light.
- In cases with similar phase fronts:
- Similar phase front

$$A_{o,scatt,J}(r) \approx A_{o\ inc,m(r)}$$

The reflectivity will be high.
The quality of phase conjugation is given by the (fidelity):

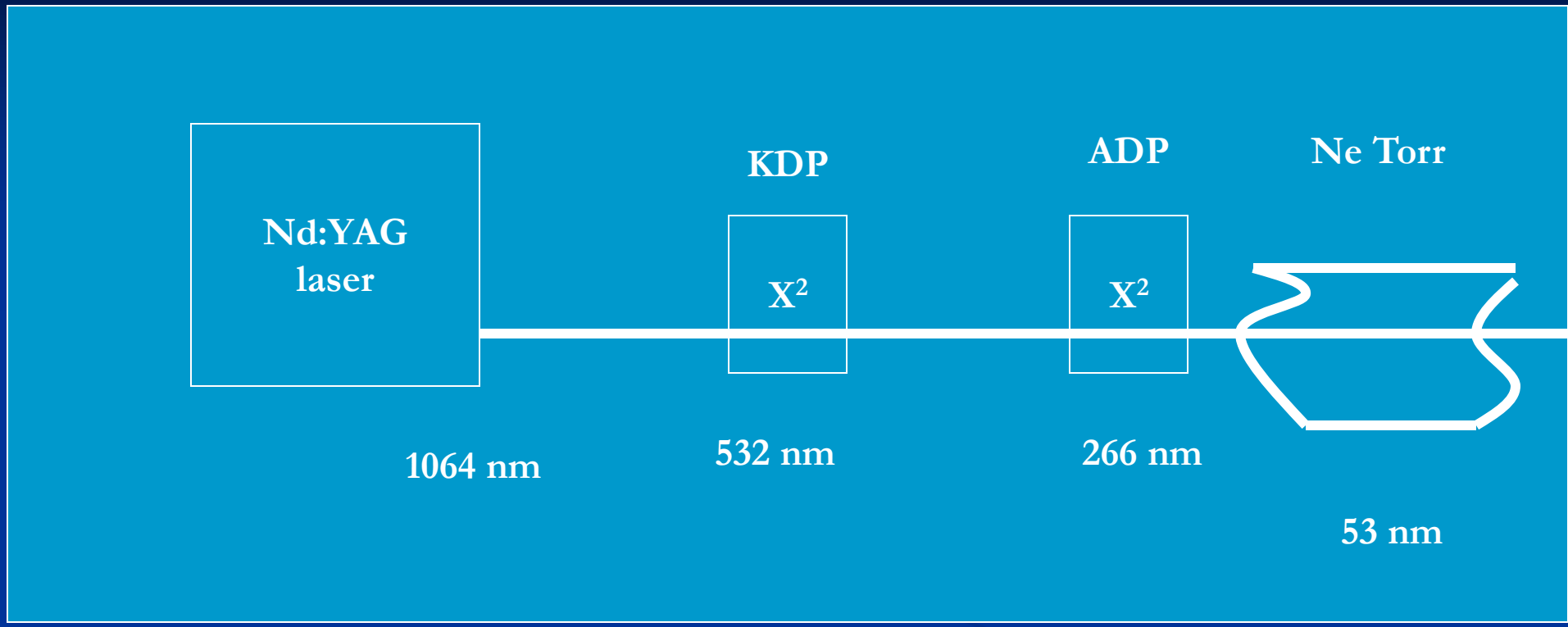
$$Fidelity\ \ F = \frac{\left|\int E_{o,scatt}(r)E_{o,inc}(r)dxdy\right|^2}{\int\left|E_{o,scatt}(r)\right|^2 dxdy\int\left|E_{o,inc}(r)\right|^2 dxdy}$$

- This correlation function F is 1 for perfect phase conjugation and 0 for random scattering. Values above 95% are possible.
- The fidelity of phase conjugation of Gaussian beam is measured frequently with the (energy in the bucket) method. (see the text).

Higher-order Nonlinear effects:

- Nonlinear effects of higher order than 2 or 3 are present in many high-power laser experiments but are not dominant and are thus difficult to detect.
- Explicity reported are the generation of higher-order frequency hormonics mainly in noble gases.
- The usually applied atom vapors for generation of higher frequencies do not automatically give phase matching. Thus by tuned mixing of different atoms with different refractive indices at the wavelength of the fundamental and the high harmonics, phase matching can be achieved in isotropic materials.

- The generation of fifth harmonic in Ne-vapor (see fig.)



- It is described, which in combination with the generation of twice the second harmonic, finally results in the generation of the 20th harmonics.
- (other examples should be observed in the text).

Materials for Nonresonant Nonlinear Interaction:

- It is still a need for better suitable materials with higher nonlinear coefficients, higher damage threshold, lower casts and higher reliability. This is especially true for wavelengths in the IR above 1.2μm and for short wavelength below 0.3 μm. .

Inorganic crystals

- Crystals are used for all kinds of frequency transformation technologies such as harmonic generation, frequency mixing and electro-optical effects. They may be classified into two groups:
- • Grown from solution.
- • Grown form melt. (see the text)

Organic materials

Organic materials can show very high nonlinear coefficients, high damage threshold and good transparency at short wavelengths.

Liquids

Organic liquids or solutions are applied in nonresonant photonic applications for (white light) generation, optical phase conjugation and Raman shifting of the incident light.

Liquid crystal

The geometrical orientation and order of molecular systems in liquid crystals can be applied in photonics for changing the polarization of a transmitting light beam.

Gases

Noble gases and gases of organic molecules are used in a way similar to liquids for optical phase conjugation and Raman shifting.