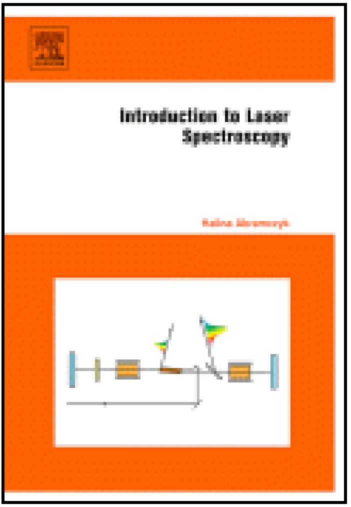

Introduction to Laser Spectroscopy

SPH 618
Optical and Laser Physics
University of Nairobi, Kenya
Lecture 4
Non-linear effects

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Linear and nonlinear optics

If several weak electric fields act on medium, in linear approximation the polarization is the sum of polarizations from the individual fields

$$\boldsymbol{P}(\boldsymbol{r},t) = \boldsymbol{P}_1(\boldsymbol{r},t) + \boldsymbol{P}_2(\boldsymbol{r},t) + \boldsymbol{P}_3(\boldsymbol{r},t) + \ldots$$

- It indicates that the weak electric fields obey the superposition principle according to which electromagnetic waves spread in medium independently, without interaction with one another. All optical phenomena undergoing this principle are called linear and belong to the field of linear optics. If electric fields are strong, this assumption is no longer valid. The intense electric fields violate the superposition principle because they interact with each other. All optical phenomena for which the superposition principle of electric fields is violated are called *nonlinear optic phenomena*

Weak electric field ***E*** induces in a material an electric induction ***D***,

$$\boldsymbol{D} = \boldsymbol{E} + 4\boldsymbol{\pi}\boldsymbol{P}$$

which depends on medium *polarization* ***P***. For small intensity of light the polarization induced in the material depends linear on the electric field intensity ***E***

$$\boldsymbol{P} = \chi^{(1)}\boldsymbol{E}$$

where $\chi^{(1)}$

is called the *electric susceptibility* of the first order. This term represents the linear response used in the conventional linear optics. The above expression arises from the fact that the medium polarization *P* is a sum of the orientational polarization and induced polarization.

$$\left(P^{orient} = N\frac{\mu^2}{3kT}E\right)$$

$$\left(P^{ind} = N\alpha E\right)$$

When the electric field ***E*** of the incident light increases, the polarization of medium is not longer linearly dependent on ***E*** and can be expanded in a power series

$$\boldsymbol{P} = \chi^{(1)}\boldsymbol{E} + \chi^{(2)}\boldsymbol{E}^2 + \chi^{(3)}\boldsymbol{E}^3 + \ldots$$

- where each term depends on the susceptibility of the n-th order. The susceptibility is a complex quantity and contains all of information about the optical properties of the dielectric material: its real part – about the index of refraction, the imaginary part – about the absorption coefficient.

Nonlinear optics

- The susceptibility is a tensor because the electric field applied in the *x* direction may result in an electron moving in the *y* or *z* directions as well. It results from the fact that electrons in the material oscillating in the direction of the light wave experience not only a restoring force, but also forces from the neighboring molecules. Thus, eq. (5.3) should be written in the following form

$$P_i = \chi_{ij}^{(1)} E_j + \chi_{ijk}^{(2)} E_j E_k + \chi_{ijkl}^{(3)} E_j E_k E_l + \ldots$$

where *i*, *j*, *k* denotes *x*, *y*, *z* cartesian components. For the repeated indices of the susceptibility χ and the field intensity *E* summation must be performed over the components of the electric field.

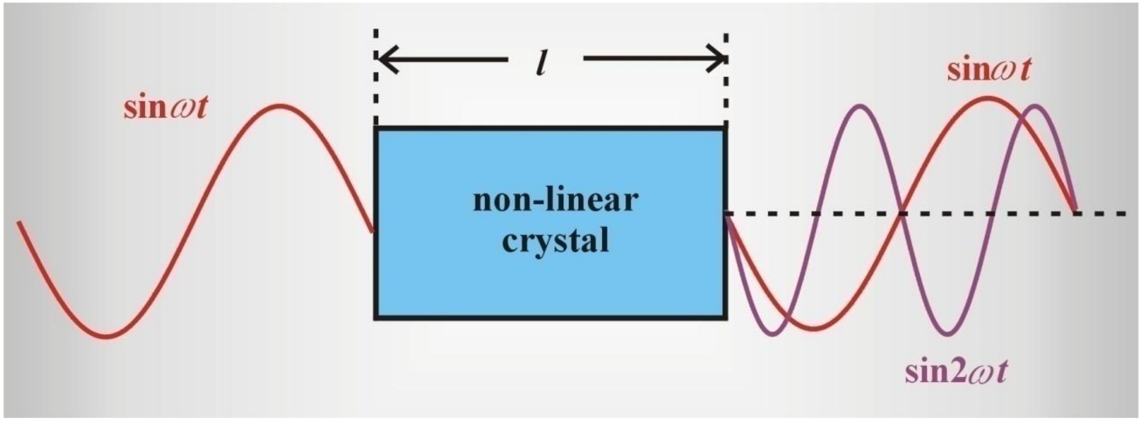
All optical phenomena for which the superposition principle of electric fields is violated are called *nonlinear optic phenomena*. One of the examples of nonlinear phenomena is the dependence of the index of refraction *n* on the light intensity.
For small intensities the index of refraction has a constant value. However, if liquid medium is illuminated with light of large Intensity the index of refraction depends directly on the electric field of the light E^2 and liquid becomes birefringent like uniaxial crystal. This effect is known as the *Kerr effect*. Another example is an absorption coefficient that in linear regime is expressed in the form of the *Lambert-Beer formula*:

$$T = I/I_0 = \exp\left(-\alpha c l\right)$$

$$A = \ln \frac{1}{T} = \alpha c l$$

where *T* is the transmission expressed as the ratio of the beam intensity *I* which is transmitted through the medium to the intensity of the incident beam I_0, α is the absorption coefficient, *c* is concentration of the absorbing medium, *l* is length of the optical path. In the regime of the linear optics the absorption coefficient does not depend on light intensity. For larger intensities this approximation is not valid any longer.

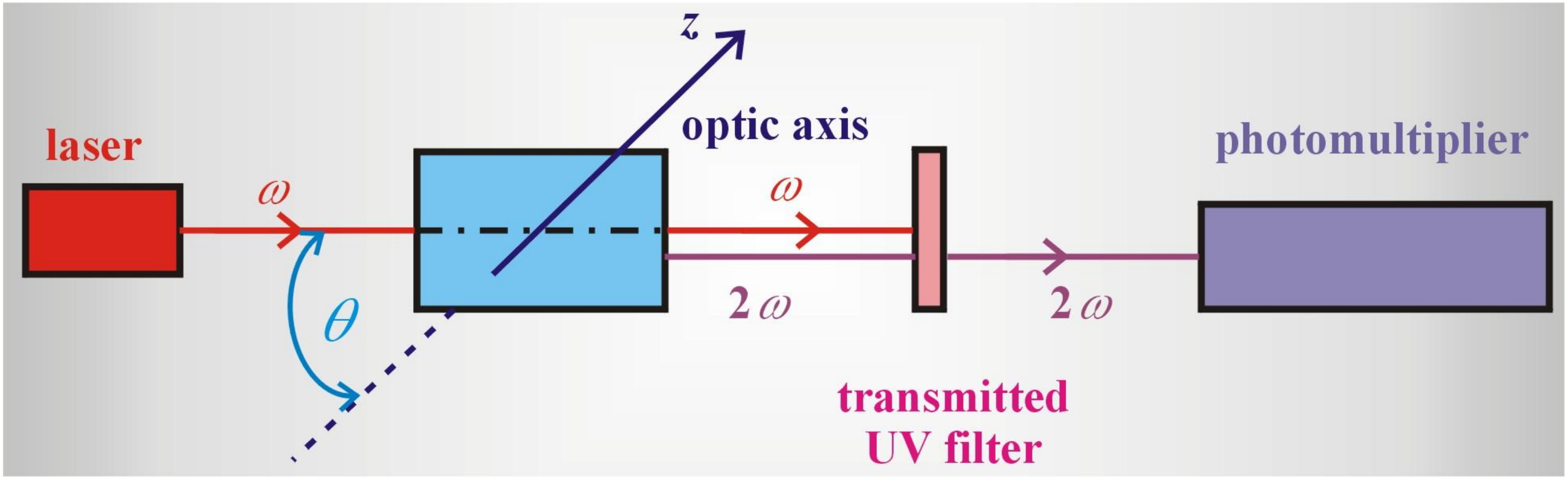
Second Harmonic Generation (SHG)



The nonlinear phenomena of the second order are described by the second term of the equation

$$P_i = \chi_{ijk}^{(2)} E_j E_k$$

The second harmonic generation was demonstrated for the first time in 1961 when the light from the ruby laser illuminating quartz produced coherent ultraviolet light. The light of the ruby laser at wavelength of 694.3 nm was directed onto a nonlinear crystal. By changing the angle between the direction of the laser beam and the direction of the crystal optic axis, it was noticed that at certain angles the output beam from the crystal has two components: 694.3 nm and 694.3 nm / 2 = 347.15 nm. Therefore, besides the fundamental component 694.3 nm, which does not change its frequency after passage through the crystal, an additional component appears with the frequency twice the fundamental frequency) called the second harmonic.



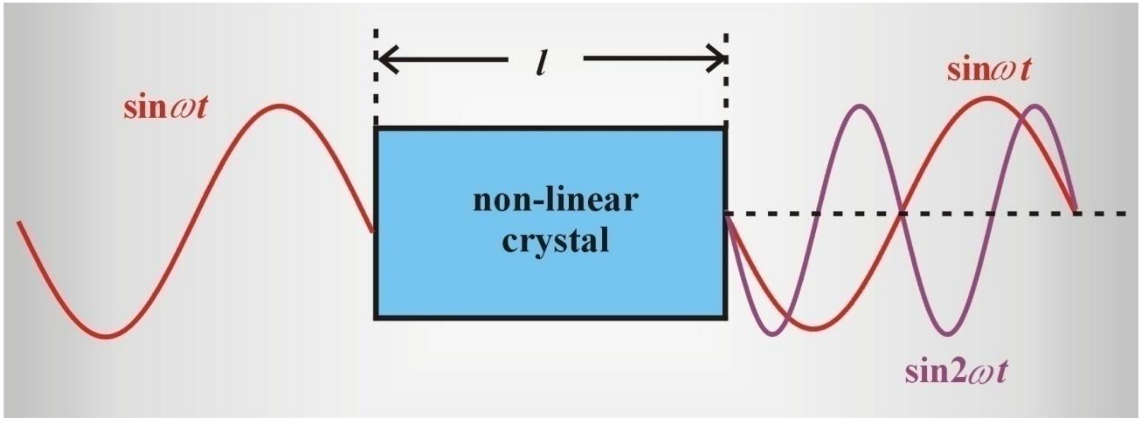
$$P_i^{(2)}(\boldsymbol{r},t)=\chi_{ijk}^{(2)}E_j(\boldsymbol{r},t)E_k(\boldsymbol{r},t)=\chi_{ijk}^{(2)}E_0^2\cos^2(\boldsymbol{k}_1\boldsymbol{r}-\omega_1 t)$$

using $\cos^2 x=\frac{1+\cos 2x}{2}$

$$P_i^{(2)}(\boldsymbol{r},t)=\chi_{ijk}^{(2)}(0)\frac{E_0^2}{2}+\frac{1}{2}\chi_{ijk}^{(2)}(2\omega_1,2\boldsymbol{k}_1)E_0^2\cos 2(\boldsymbol{k}_1\boldsymbol{r}-\omega_1 t)$$

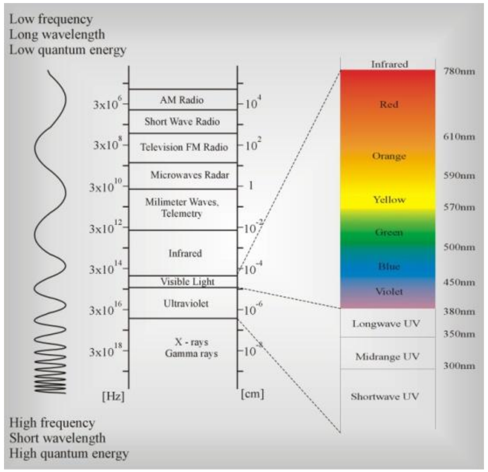
Thus, the polarization induced by interaction of two waves of frequency w_1 each consists of two terms: a constant time independent polarization $\frac{1}{2}\chi_{ijk}^{(2)}(0)E_0^2$ and the polarization modulated at the frequency $2\omega_1$.

Second Harmonic Generation (SHG)

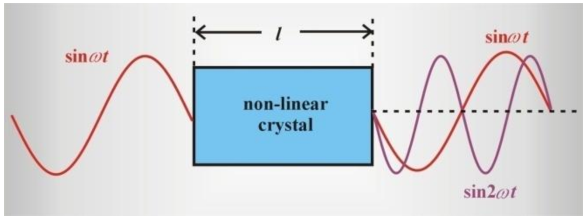


The nonlinear phenomena of the second order are described by the second term of the equation

$$P_i = \chi^{(2)}_{ijk} E_j E_k$$



The phase matching condition $(\Delta \boldsymbol{k} = 0)$



- The phase matching condition is fulfilled only when
- . $\boldsymbol{k}_2(2\omega_1) = \boldsymbol{k}_1(\omega_1) + \boldsymbol{k}_1(\omega_1)$
- The phase matching condition can be written more generally as
- $\boldsymbol{k}_3(\omega_3) = \boldsymbol{k}_1(\omega_1) + \boldsymbol{k}_2(\omega_2)$ (5.14)
- or $\Delta \boldsymbol{k} = \boldsymbol{k}_3(\omega_3) - \boldsymbol{k}_1(\omega_1) - \boldsymbol{k}_2(\omega_2) = 0$
- . (5.15)
- Since the wave vector ***k*** is related to the photon momentum ***p*** ($\boldsymbol{p} = \hbar \boldsymbol{k}$), this condition simply indicates that the energy exchange between the waves is possible only when the momentum conservation law for three photons participating in the process of frequency mixing is not violated. When all the beams are collinear, the condition (5.14) may be replaced by
- , $n_3\omega_3 = n_1\omega_1 + n_2\omega_2$ (5.16)
- where we employed the relation $\upsilon_{ph} = \frac{\omega}{k} = \frac{c}{n(\omega)}$ For the second harmonic with $\omega_1 = \omega_2$ and , eq. (5.16) takes the form
- . $n(2\omega_1) = n(\omega_1)$ (5.17)
- This indicates that the second harmonic may be generated efficiently only when the index of refraction at $2\omega_1$ is equal to the index of refraction at ω_1. In most cases it is impossible due to dispersion of the material, which simply means that two waves differing in the wavelength, have different indices of refraction as well. However, we can use some tricks related to the properties of birefringent crystals.
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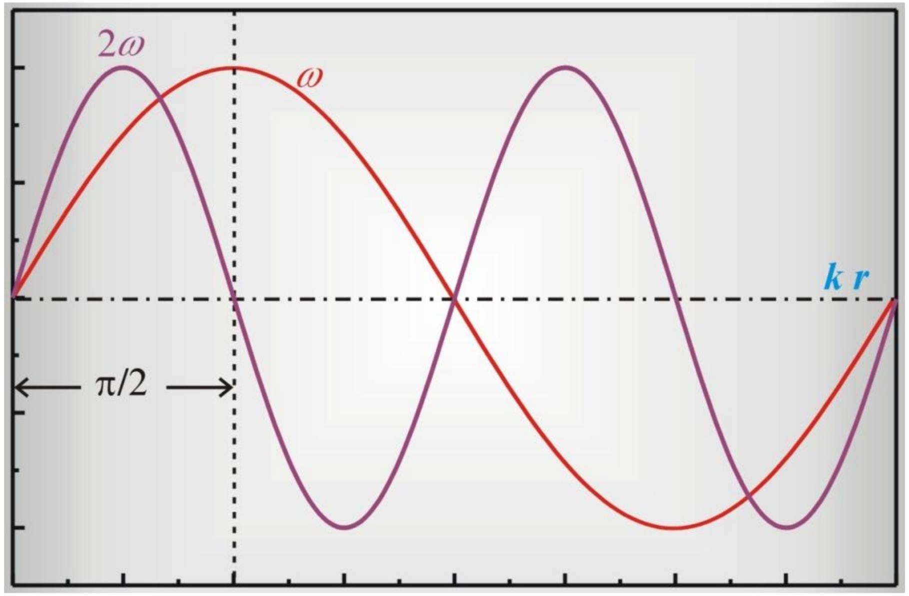
The phase matching condition



$$\left(\Delta \boldsymbol{k} = 0\right)$$

$$I(2\omega) = K(\chi_{eff}^{(2)})^2 l^2 \left(\frac{\sin(\Delta kl/2)}{\Delta kl/2}\right)^2 I_0^2(\omega)$$

where: *l* is a length of crystal, $I_0(\omega)$ is the intensity of incident radiation at frequency ω, $\chi_{eff}^{(2)}$

is the effective susceptibility of the second order, K is a constant for a given wavelength and a given material

How to achieve the high SHG efficiency?

$$I(2\omega) = K(\chi_{eff}^{(2)})^2 l^2 \left(\frac{\sin(\Delta k l / 2)}{\Delta k l / 2} \right)^2 I_0^2(\omega)$$

This can be achieved by employing:

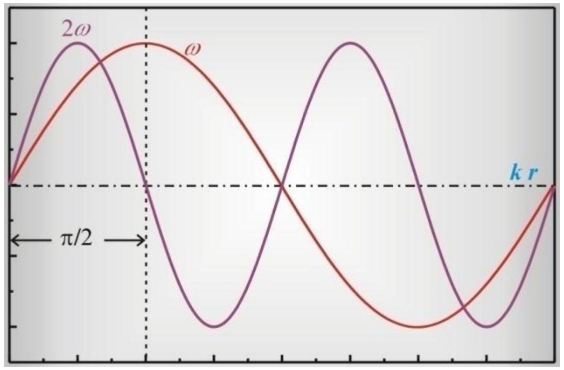
- laser pulses of high incident intensity I_0; the magnitude of I_0 are limited by a crystal damage threshold,
- nonlinear materials of high $\chi^{(2)}$ susceptibility,
- phase matching condition $\Delta \boldsymbol{k} = 0$,
- long optical path l.

However, material thickness l is limited by a certain critical value l_{coh}, called the *coherence length*.
One can see from fig. 5.3 that the fundamental wave at frequency ω and the SHG wave traveling along the crystal are more and more out of phase leading to destructive interference and reducing the second harmonic intensity.
The distance from the entrance surface of the crystal to the point where the second harmonic intensity has its first maximum has been termed the coherence length l_{coh}. If the crystal length happens to be an odd multiple of the coherence length, no second-harmonic light will be emitted.

The coherence length can be roughly estimated from its definition

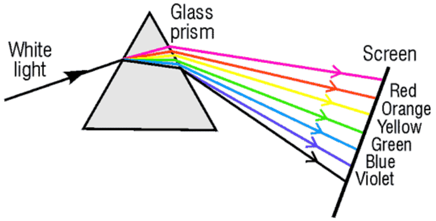
$$\Delta k \cdot l_{coh} = \frac{\pi}{2}$$

$$l_{coh} = \frac{\pi}{2\Delta k} = \frac{\lambda}{4(n(2\omega) - n(\omega))}$$

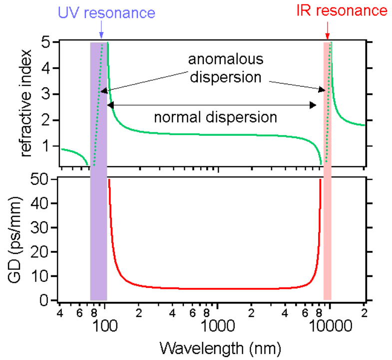


Scheme illustrating the crystal coherence length

How to achieve the condition of phase matching for SHG in dispersive media?

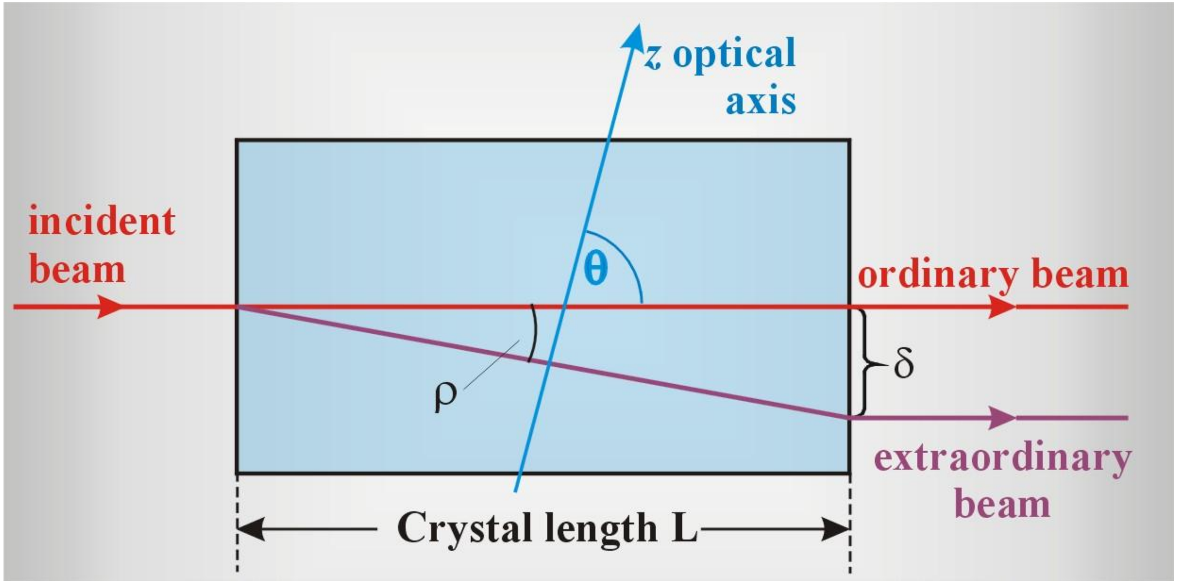


$$n(2\omega_1) = n(\omega_1)$$



birefringence

birefringent crystals



$$\frac{\sin\alpha}{\sin\beta} = \frac{n_1}{n_2}$$

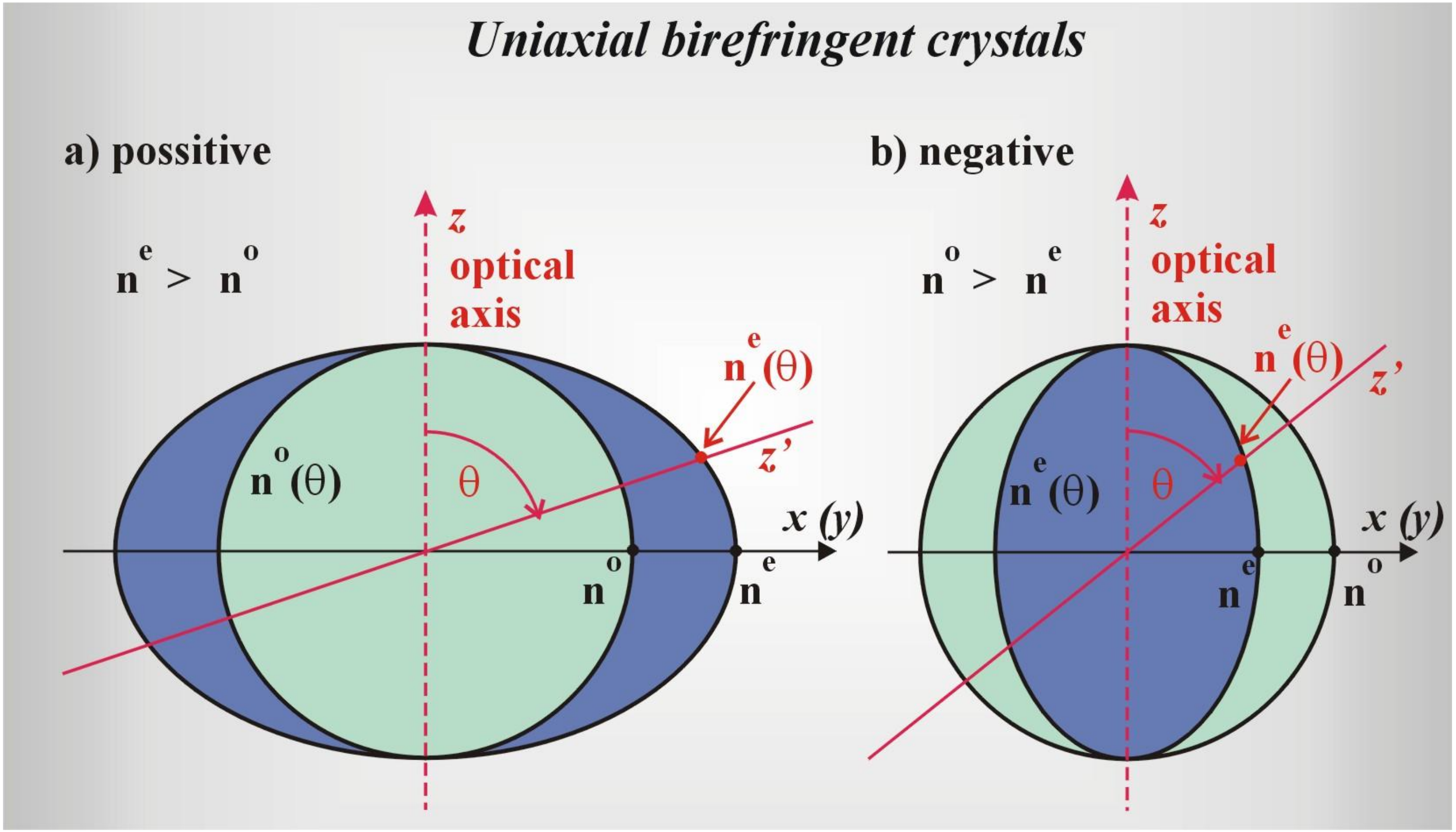
the ordinary and extraordinary beams do not propagate along the same line, the beam is divided onto two orthogonally polarized beams separated by δ called the *walk-off distance*

The *birefringence* can be observed when light passes through many crystals, including calcite, ice, quartz, mica, sugar, that are anisotropic and have the optic axes. In such crystals the light beam undergoing refraction divides into two rays in contrast to the isotropic media (liquids, gases, glasses) that show only one ray. The two rays are called the *ordinary* (o) and *extraordinary* (e) rays and have the different phase velocities and the different indices of refraction.

This phenomenon is called *double refraction* or *birefringence* and was discovered by Bartholinus and Huygens.

They found that both rays are polarized linearly in planes perpendicular to each other.

birefringent crystals



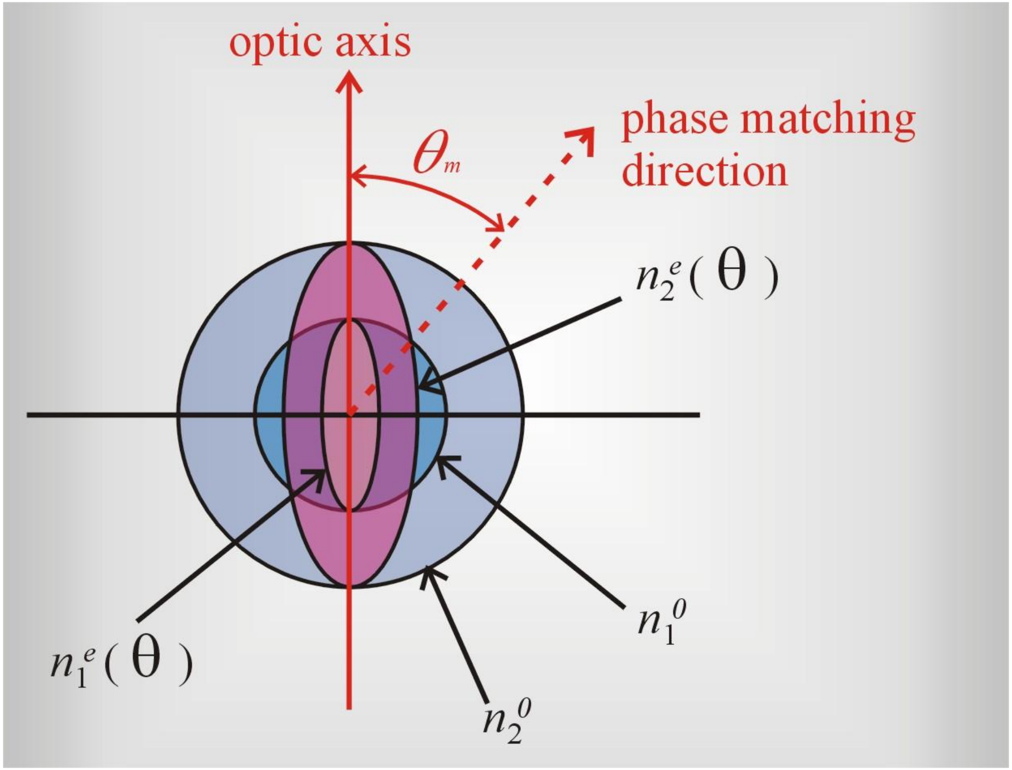
Phase Matching Methods- using birefrigence

$$n(2\omega_1) = n(\omega_1)$$

$$n^e(2\omega_1) = n^o(\omega_1)$$

One can notice that there are certain directions of propagation in the crystal for which the fundamental and second harmonic beams have the same refraction index in spite of their different wavelengths. These directions go along a line forming the angle ϑ_m with the optic axis for which the surfaces of the indices of refraction n_1^o and $n_2^e(\theta)$ intersect

$$n_1^o = n_2^e(\theta_m)$$



the *index matching technique*

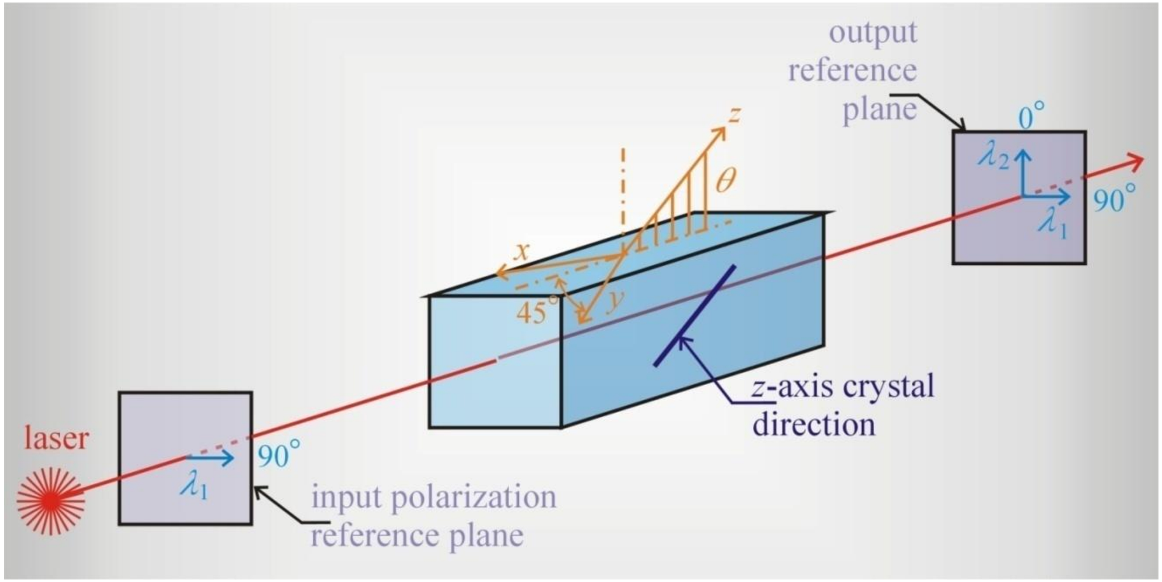
I type of phase matching

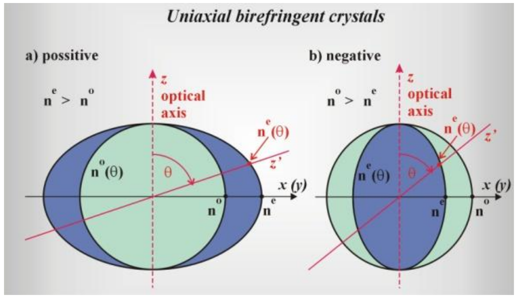
In uniaxial negative crystals the I type phase matching condition is

$$\boldsymbol{k}_1^o + \boldsymbol{k}_2^o = \boldsymbol{k}_3^e(\theta)$$

In uniaxial positive crystals the I type phase matching condition is

$$\boldsymbol{k}_1^e(\theta) + \boldsymbol{k}_2^e(\theta) = \boldsymbol{k}_3^o$$





o+o=e

e+e=o

I type of phase matching

- If the mixing waves at frequencies ω_1 and ω_2 have the same polarization and the sum frequency wave $\omega_3 = \omega_1+\omega_2$ (SFG) is polarized perpendicularly to their incident polarization, I type of phase matching takes place.

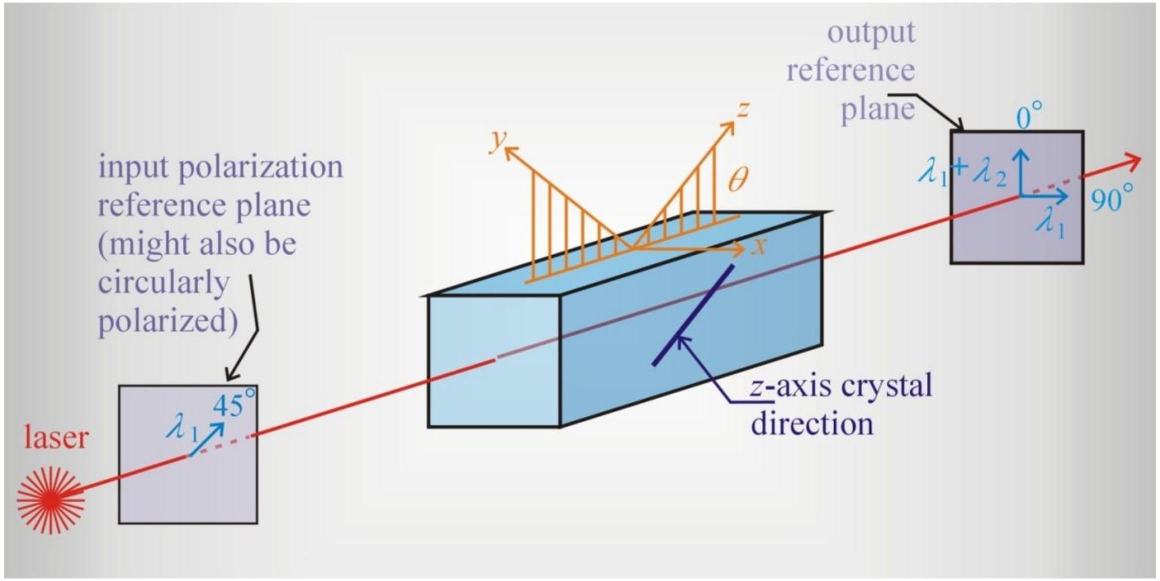
II type of phase matching

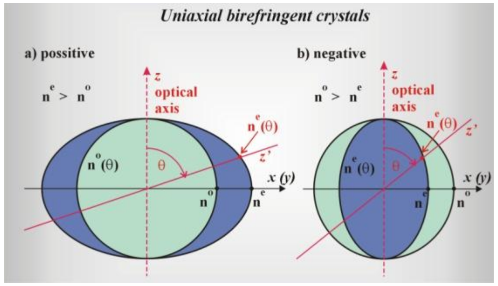
In uniaxial negative crystals the II type phase matching condition occurs when

$$\boldsymbol{k}_1^o + \boldsymbol{k}_2^e(\theta) = \boldsymbol{k}_3^e(\theta)$$

In uniaxial positive crystals the II type phase matching occurs when

$$\boldsymbol{k}_1^o + \boldsymbol{k}_2^e(\theta) = \boldsymbol{k}_3^o$$



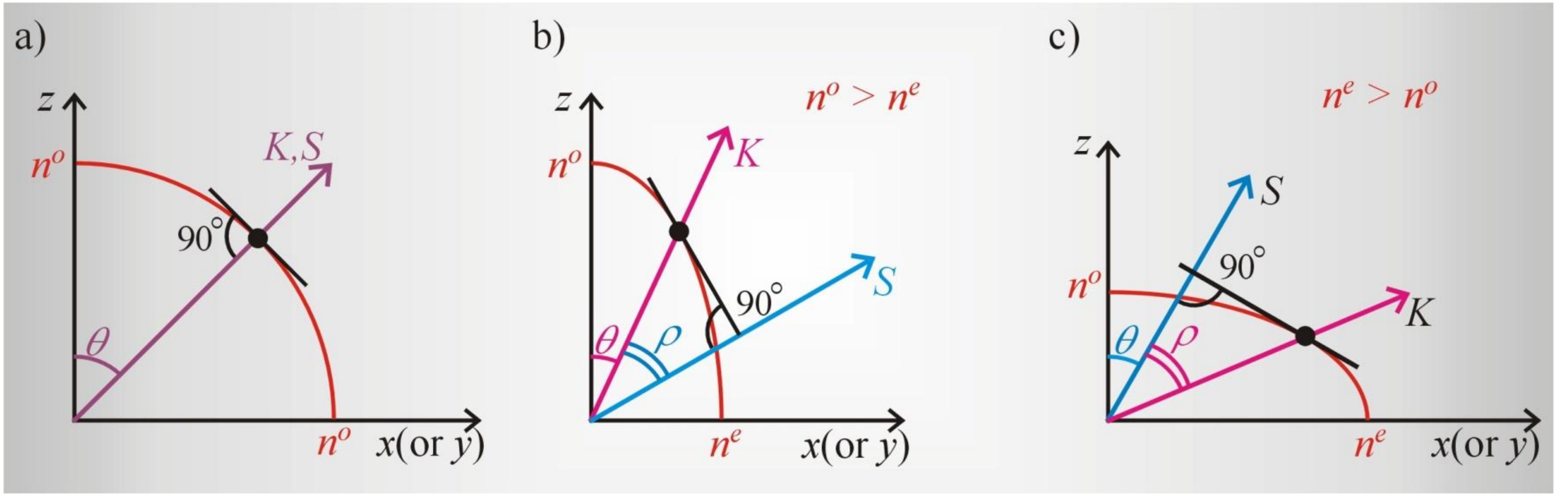


e+o=e

o+e=o

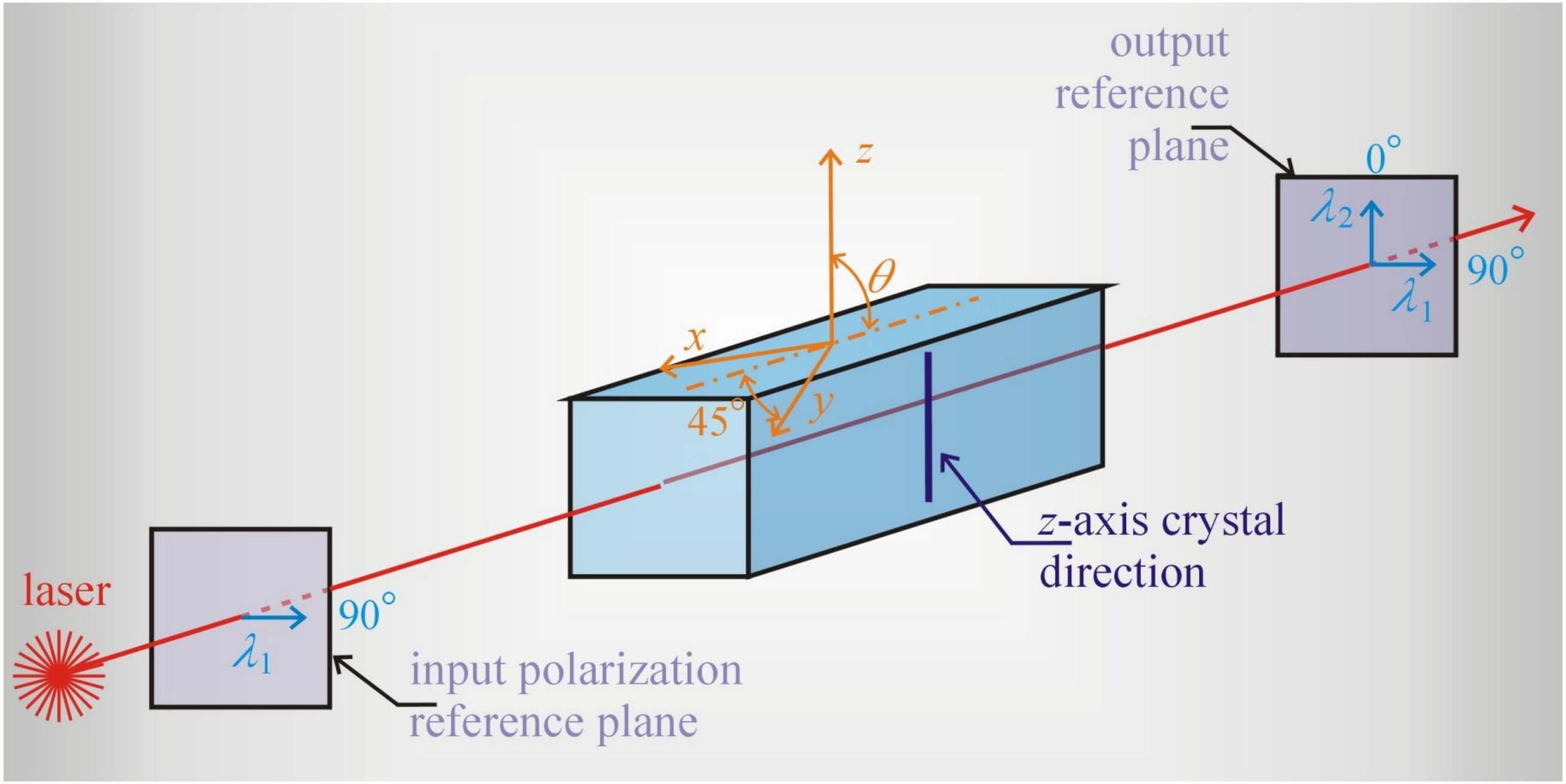
If the mixing waves at frequencies ω_1 and ω_2 are of orthogonal polarization, the II type phase matching occurs

However, an additional difficulty with SHG process is that the energy of the two beams, the fundamental and the second harmonic that are orthogonally polarized will propagate in different directions, characterized by the walk-off distance δ. This comes from the fact that in a birefringent crystal the direction of propagation of the wave phase (wave vector ***k***) generally does not coincide with that of the wave energy (Poynting vector ***s***). The direction of Poynting vector ***s*** is defined as the normal to the tangent drawn at the point of intersection of wave vector ***k*** with $n(\theta)$ cross section .
One can see that for the ordinary beam both vectors coincide in contrast to the extraordinary beam.



Directions of the wave vector ***k*** and the energy wave (Poynting vector) ***s*** for (a) ordinary beam; extraordinary beam in negative (b) and positive (c) uniaxial crystal

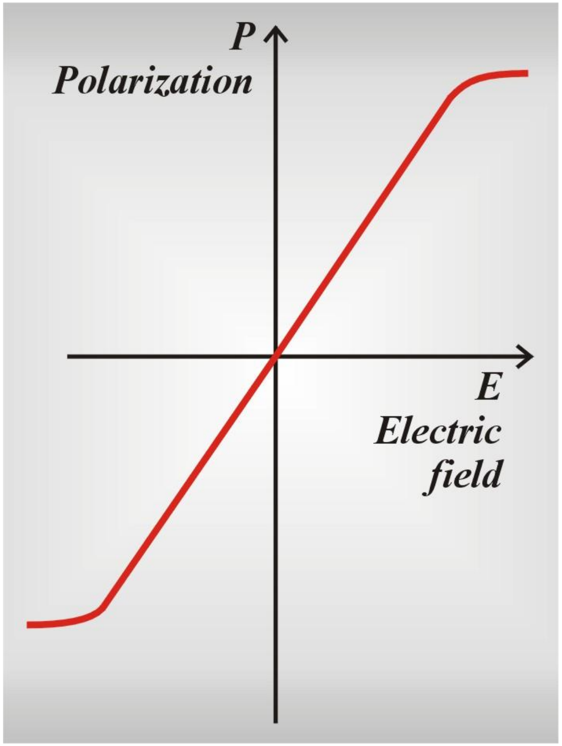
This disadvantage of the angle-tuned phase matching technique can be eliminated by employing crystals with the phase matching angle $\theta_m = 90^\circ$. In this case the direction of propagation and the optic axis is set to 90° and the angle between the Poynting vectors for the two beams is zero.

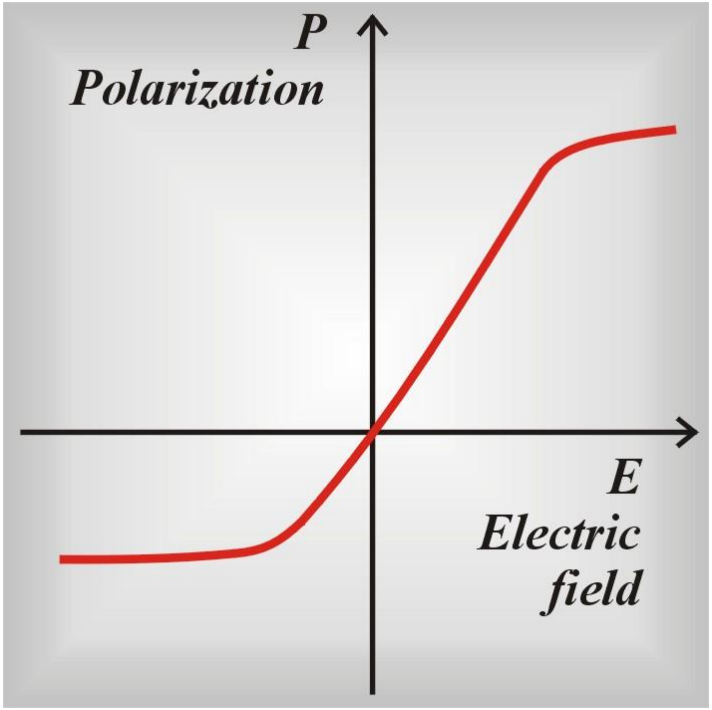


In systems having the macroscopic center of inversion (liquids, gases, crystals of some classes) it is impossible to generate the second harmonic. Indeed, if the system has the center of inversion, the polarization *P* = *f*(*E*) is an odd function of the electric field This indicates that the second term in equation

$$\boldsymbol{P} = \chi^{(1)}\boldsymbol{E} + \chi^{(2)}\boldsymbol{E}^2 + \chi^{(3)}\boldsymbol{E}^3 + \dots$$

has to vanish during one period of the electric field oscillation. The medium can produce the second harmonic only when *P* = *f*(*E*) dependence looks like that in figure on right that takes place only when there is no macroscopic center of inversion.
Therefore, the second harmonic generation is limited to:
some classes of crystals (without the macroscopic inversion symmetry),
surfaces of a medium, gas or liquid systems in which the isotropy has been broken by an electric field or or a density gradient.





Properties of important nonlinear materials and phase matching parameters [5.1]

Material	Phase-matching type	Effective nonlinear coefficient* [10^{-12} m/V]	index of refraction $n_0(\omega)$	Damage threshold [$\mathrm{GW/cm^2}$]	Absorption [$\mathrm{cm^{-1}}$]	Phase matching angle	Walk-off angle
KD*P	II	0.37	1.49	0.5	0.005	53.7°	1.45°
KTP	II	3.18	1.74	0.5	0.010	24.3°	0.26°
LBO	I	1.16	1.56	2.5	0.005	-	-
BBO	I	1.94	1.62	1.5	0.005	22.8°	3.19°
$LiNbO_3$ (5%MgO)	I	4.7	2.23	0.10	0.002	90° (1)	0°

* For 1064 nm to 532 nm second-harmonic generation, (1) at T = 107°C

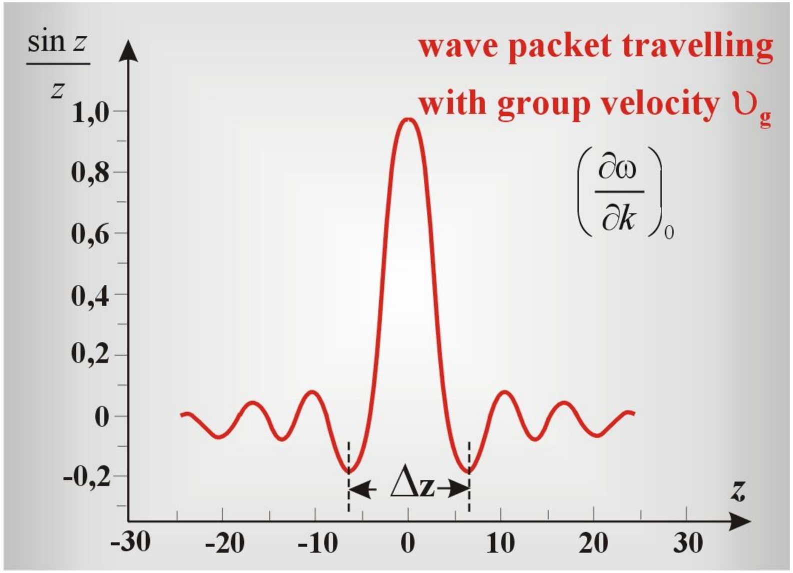
SHG for Pico- and Femtosecond Pulses

The mechanism of the second harmonic generation by femtosecond lasers is the same as discussed previously. However, in the case of such short pulses one should take into account additional effect, which is negligible for longer pulses or at continuous work. An ultrashort pulse corresponds to a large spectral width $\Delta\omega$. The phase matching condition should be obeyed not only for the center of the spectral distribution ω_0 for the fundamental but also for the whole spectral range of the pulse . If the phase matching is valid only around the center ω_0, the generated second harmonic will be spectrally narrowed and the pulse duration will become longer.

Group velocity

Phase velocity

$$\upsilon_{ph} = \frac{\omega}{k} = \frac{c}{n(\omega)}$$



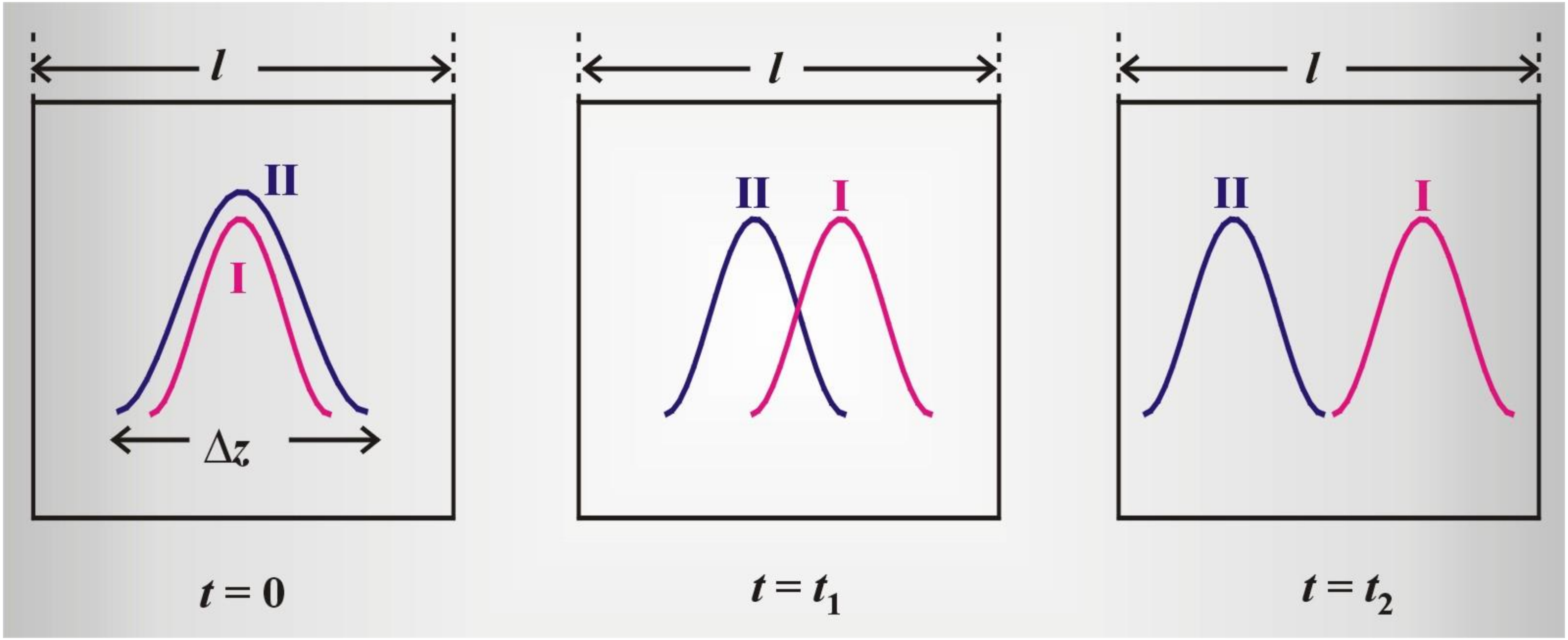
Another parameter important for ultrashort pulses related to dispersion properties is the *group delay*.

Assume the second harmonic is generated under the phase matching condition in a crystal of length *l* by pulses of duration t_p. It indicates that the phase velocities of the fundamental beam and the second harmonic are equal for a given component

$$\left(\upsilon_{ph}(\omega)=\upsilon_{ph}(2\omega)\right) \qquad \upsilon_{ph}=\frac{\omega}{k}=\frac{c}{n(\omega)}$$

It does not mean, however, that the group velocities are also equal. The group velocities of the fundamental beam and the second harmonic are usually different due to the dispersion properties of the crystal.

The consequence of the different group velocities is the different time t_g of a passage through a crystal of length *l* for the fundamental beam and the second harmonic.



This indicates that the second harmonic wave packet is delayed with respect to the fundamental wave packet by Δt

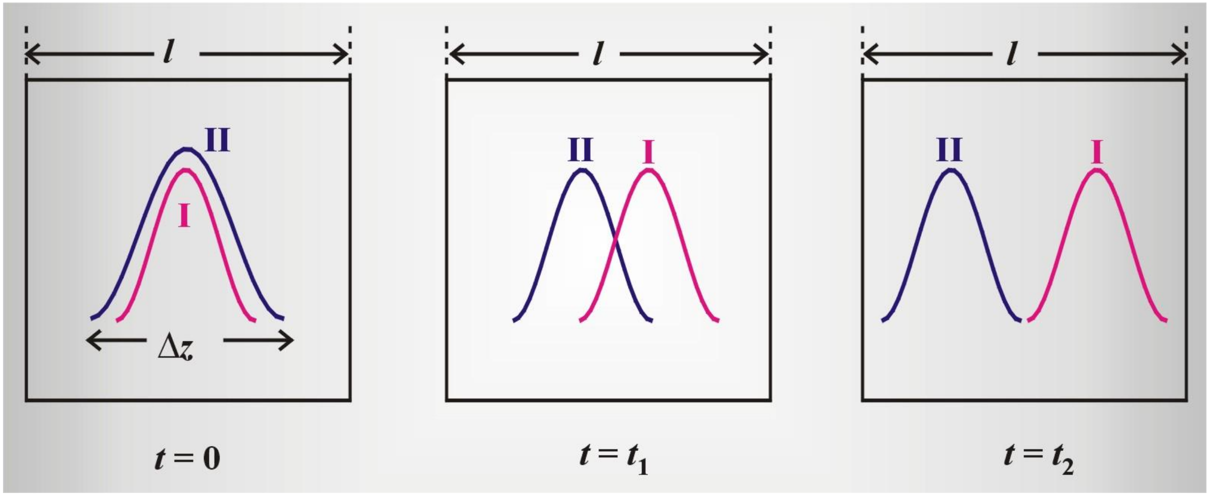
SHG for Pico- and Femtosecond Pulses

- The greater the *group delay* time, $\Delta t = t_g^2 - t_g^1$, the shorter the length of interaction between the fundamental and the second harmonic beams.

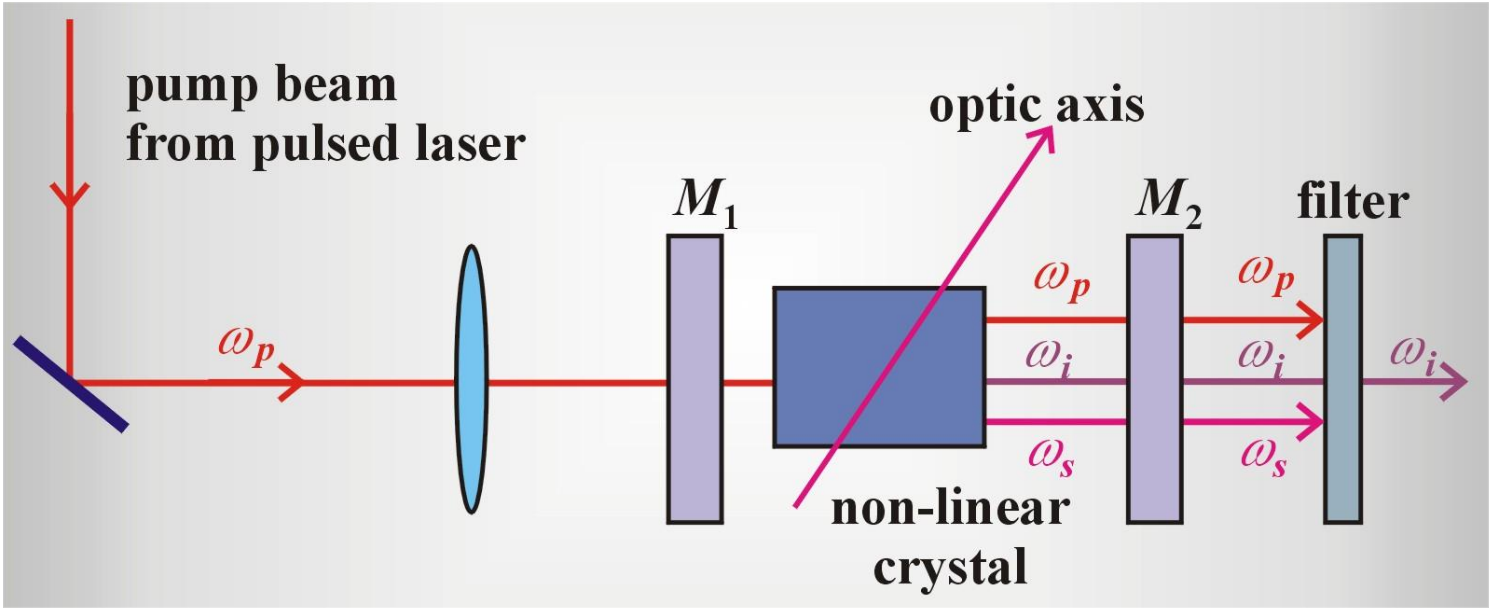
The interaction length is limited by the condition

$$\Delta t \le t_p$$

This condition defines the effective crystal length *l*. If the crystal is longer than *l* leading to the violation of the condition (5.41), the rest of the crystal is ineffective because the beams do not overlap any longer and the SHG process does not occur. For $\Delta t_g > t_p$ the second harmonic intensity does not increase with crystal length but a temporal pulse stretching occurs.



Optical Parametric Oscillator (OPO)



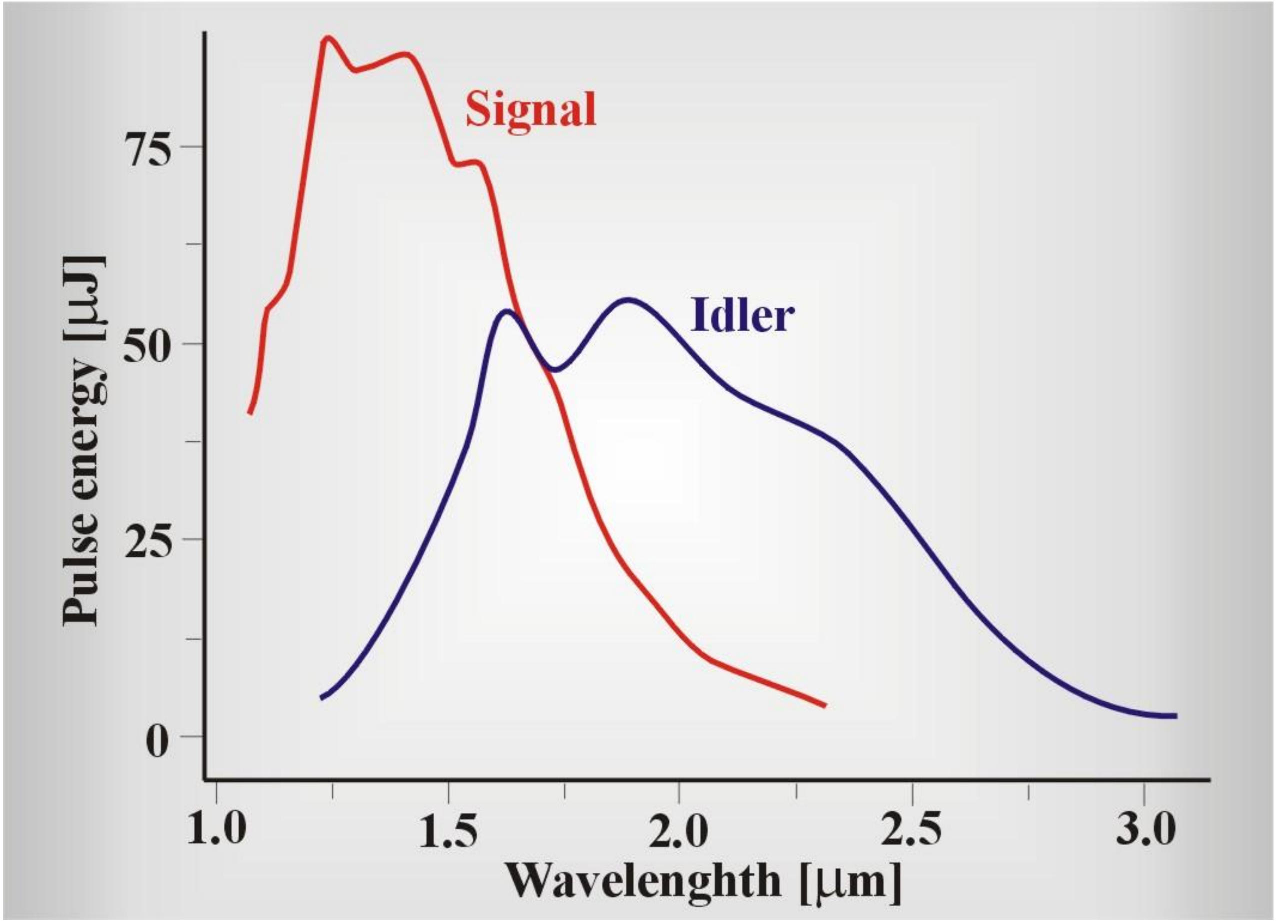
The parametric conversion can be described as inelastic scattering of a pumping photon $\hbar\omega_p$ on a crystal lattice. As a result of interaction, two photons: $\hbar\omega_s$ and $\hbar\omega_i$ are generated. The component with the frequency ω_i is called the *idler component*, and the component ω_s – *the signal component*. The energy conservation law requires

$$\omega_p = \omega_i + \omega_s$$

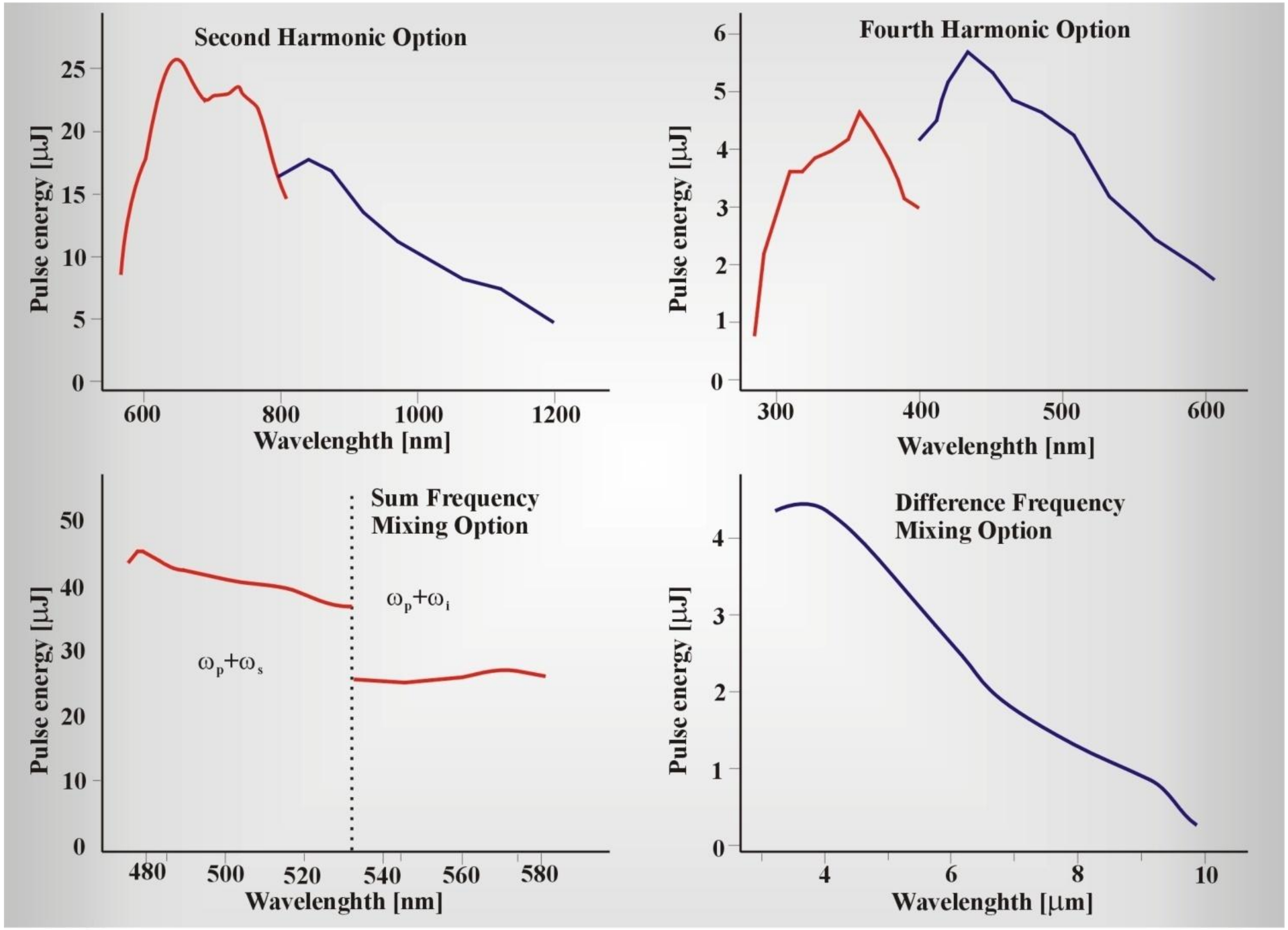
$$\boldsymbol{k}_p = \boldsymbol{k}_i + \boldsymbol{k}_s$$

$$\Delta k = \boldsymbol{k}_p - \boldsymbol{k}_i - \boldsymbol{k}_s = 0$$

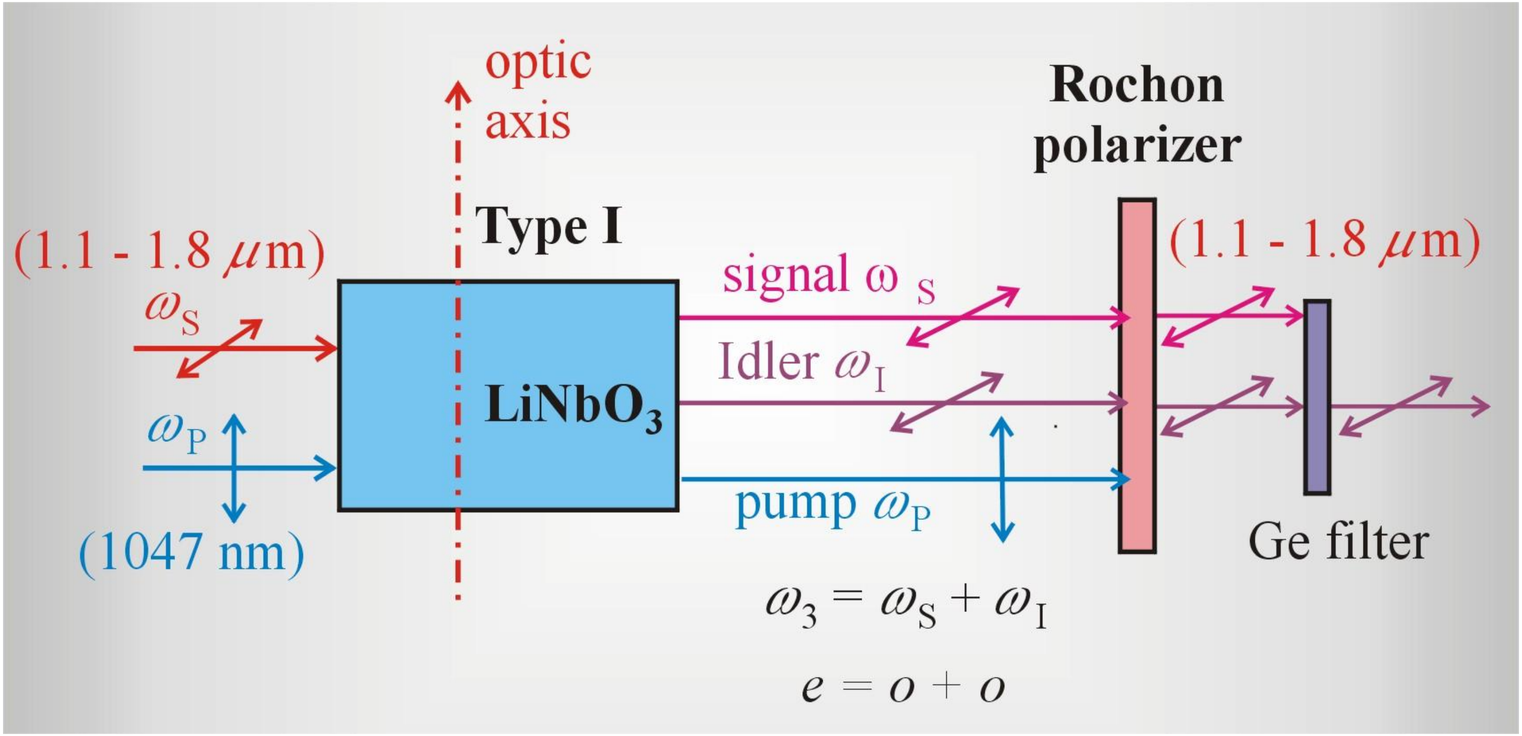
Typical femtosecond OPA systems use BBO crystals and offer tunability from 1.1 μm to 1.6 μm (signal) and 1.6 μm to 3.0 μm (idler). The output pulses are near transform-limited with less than 130 fs pulse width and typical energies about 100 μJ



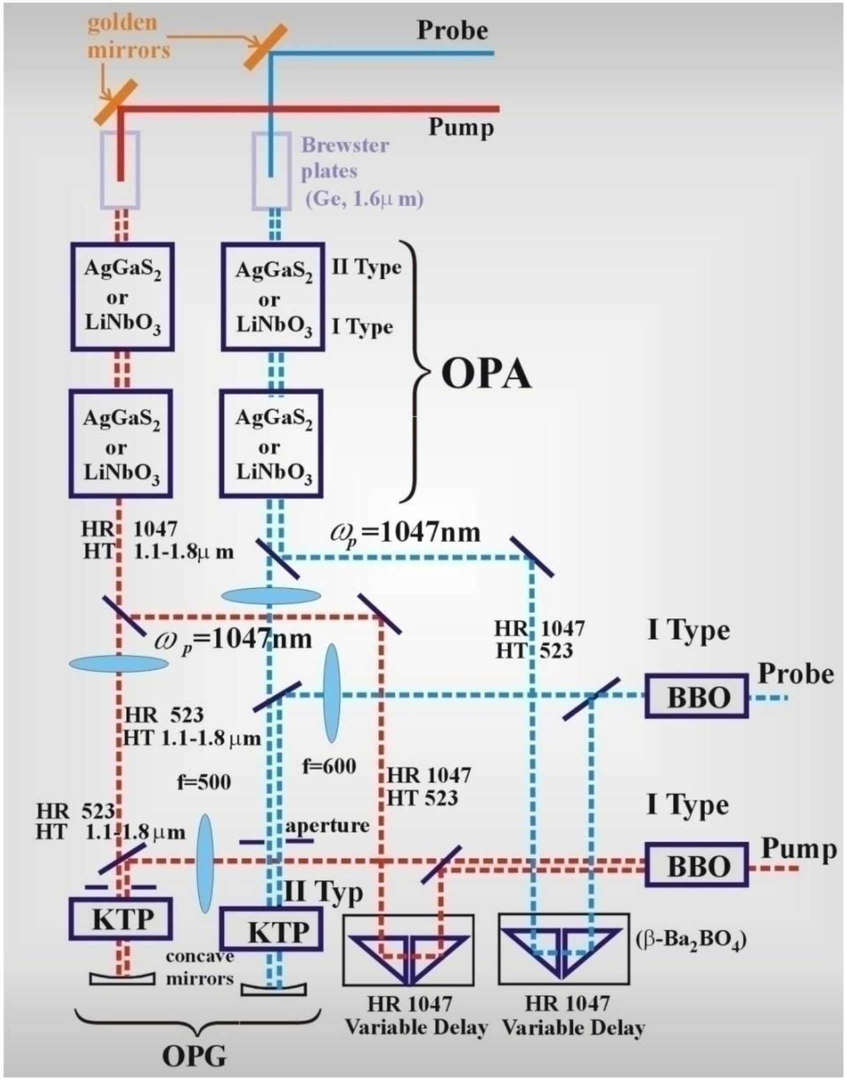
The wavelength coverage can be further extended from <300nm to >10 mm via harmonic generation, sum-frequency mixing or difference-frequency mixing. One can get the range between 480 and 580 nm for the sum-frequency mixing, 3-10 μm for the difference-frequency mixing, 600-1200 nm for the II harmonics of the signal and idler components and 300-600 nm for the IV harmonic of the signal and idler components.



optical parametric amplifier (OPA)

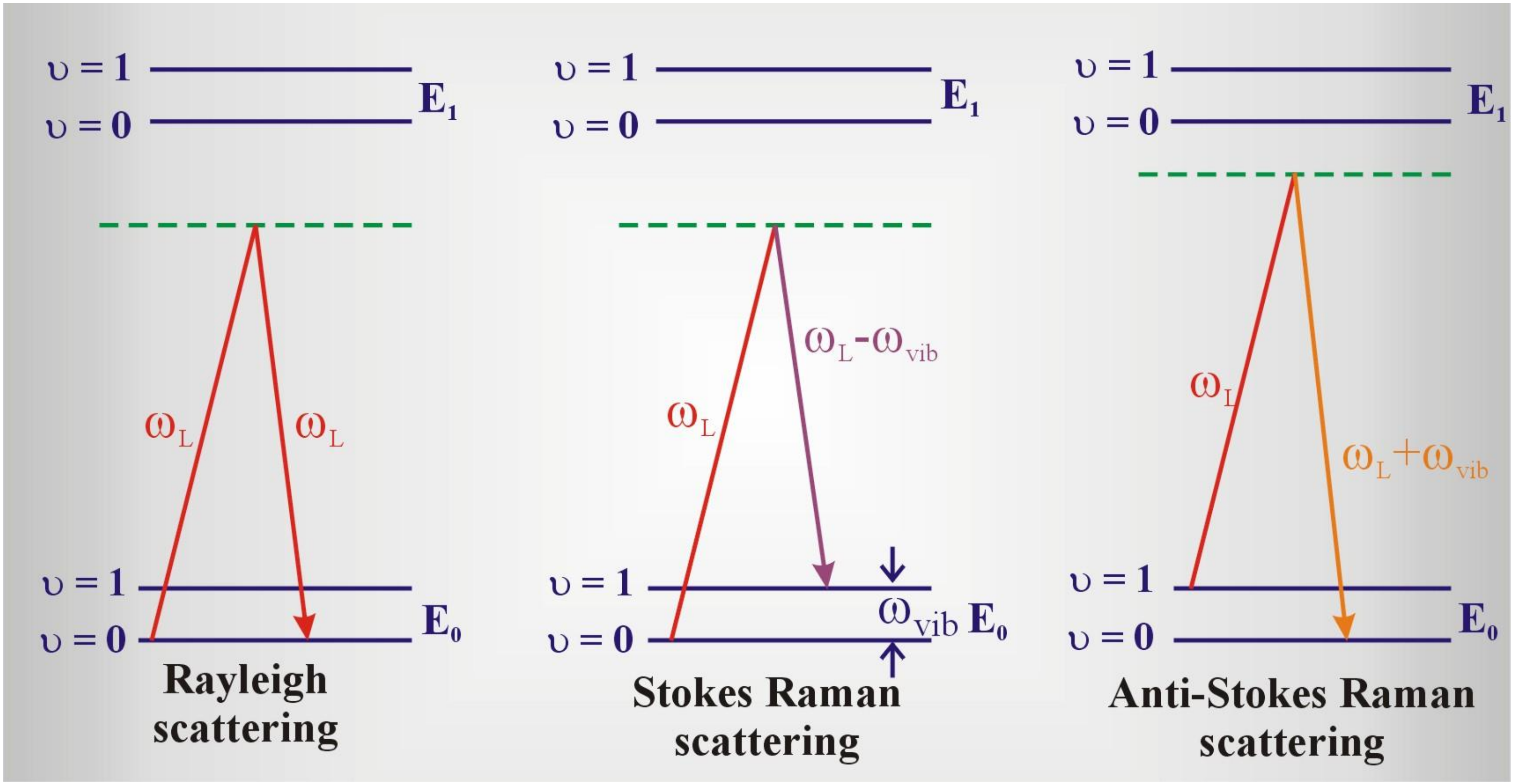


OPO, OPA, OPG


golden mirrors
Probe
Pump
Brewster plates (Ge, 1.6μ m)
$AgGaS_2$ or $LiNbO_3$
$AgGaS_2$ or $LiNbO_3$
$AgGaS_2$ or $LiNbO_3$
$AgGaS_2$ or $LiNbO_3$
II Type
I Type
OPA
HR 1047
HT 1.1-1.8μ m
ω_p=1047nm
ω_p=1047nm
HR 1047
HT 523
I Type
BBO
Probe
HR 523
HT 1.1-1.8 μm
f=600
f=500
HR 1047
HT 523
I Type
HR 523
HT 1.1-1.8 μm
aperture
BBO
Pump
II Typ
KTP
KTP
concave mirrors
(β-Ba_2BO_4)
HR 1047 Variable Delay
HR 1047 Variable Delay
OPG

The Third Order Nonlinear Processes

$$P_i = \chi^{(1)}_{ij} E_j + \chi^{(2)}_{ijk} E_j E_k + \chi^{(3)}_{ijkl} E_j E_k E_l$$



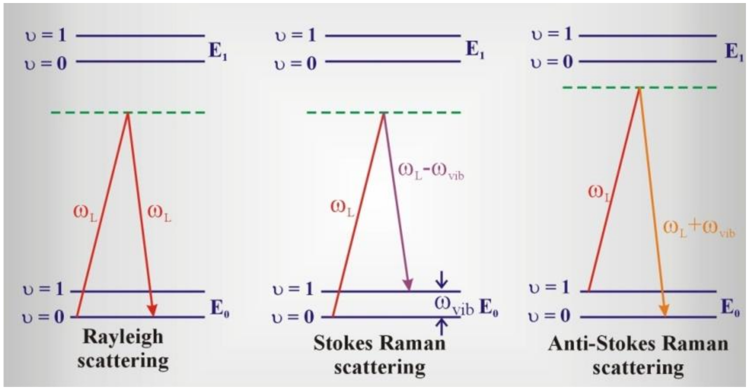
Rayleigh scattering

Stokes Raman scattering

Anti-Stokes Raman scattering

Spontaneous Raman scattering

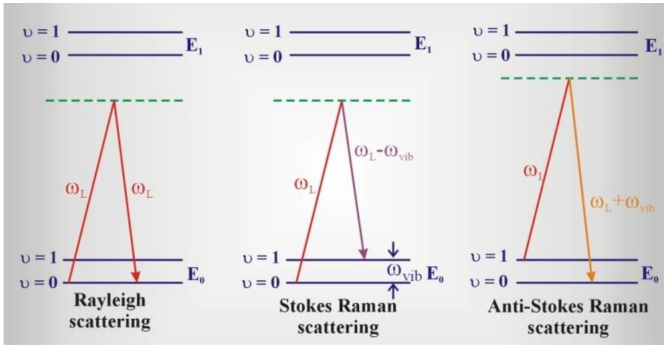
$$\boldsymbol{P} = \chi^{(1)}\boldsymbol{E}$$



- The levels denoted as E_0 and E_1 represent electronic energy levels while the levels numbered with a quantum number υ represent the vibrational levels. If the sample is illuminated with photons of energy , smaller than the resonance energy , all of the photons that interact with the sample are not absorbed, but cause the potential energy of the interacting molecules to be raised to virtual state, above the ground state. Almost immediately most molecules return to the ground state through the emission of photons of the same energy as the incident photons. This elastic scattering is called the *Rayleigh scattering*. A small fraction of the molecules drops back to the first excited vibrational state ($\upsilon = 1$) instead to the ground state. Since the energy of the incident and the scattered photons are different, the scattering is inelastic and the process is known as *Stokes Raman scattering* with the scattered radiation observed at lower energy . Molecules that are already in the excited vibrational state ($\upsilon = 1$) will undergo analogous effect when illuminated with a laser light. When the excited molecules drops back to the ground vibrational state ($\upsilon = 0$), the scattered radiation will be observed at higher energy . This scattering is known as *anti-Stokes Raman scattering*, The frequency ω_{vib} denotes the frequency of a given vibrational mode of the molecule.

Spontaneous Raman scattering

$$\boldsymbol{P} = \chi^{(1)}\boldsymbol{E}$$



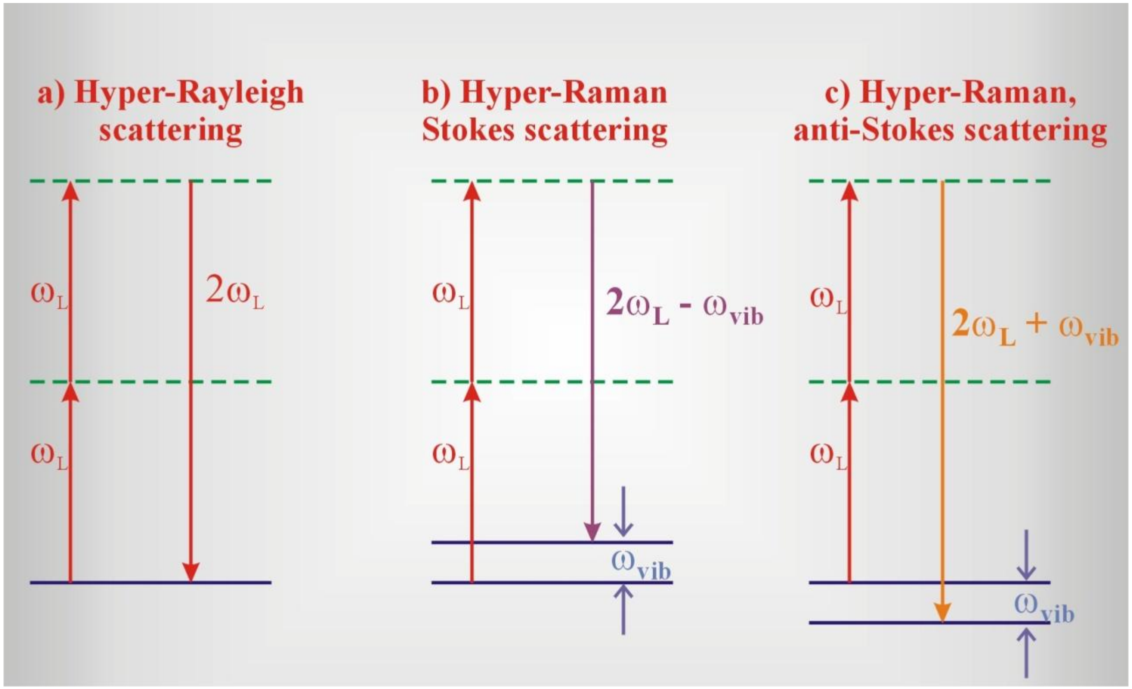
- To describe Raman scattering a fully quantum-mechanical theory is required but some intuitive description can be also obtained from a classical picture. The electric field $E_0\cos(\boldsymbol{k}\cdot\boldsymbol{r}-\omega_L t)$ drives the electron displacements that induce the polarization ***P*** in a medium modulated in time that in turn generates a wave at the same frequency ω_L (Rayleigh scattering). When the dipole oscillations are modulated additionally by the molecule vibrations at frequency ω_{vib} the waves at ($\omega_L - \omega_{vib}$) (S*tokes Raman scattering*) or ($\omega_L + \omega_{vib}$) (anti-S*tokes Raman scattering*) are generated.

- When more intense light is employed nonlinear Raman processes occur in the material
- *hyper Raman or*
- *stimulated Raman scattering*

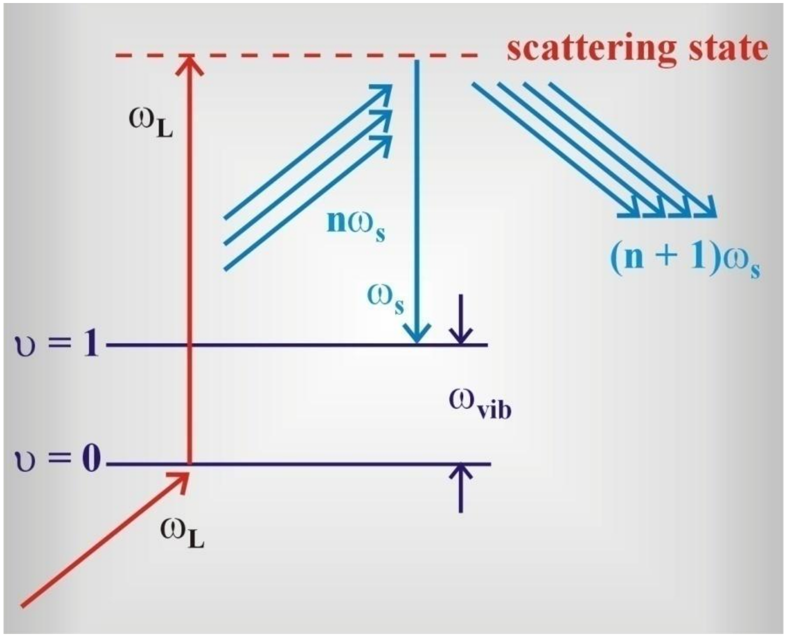
Hyper Raman or *Hyper Rayleigh scattering*

When the intensity of the incident radiation increases, the probability that two or three photons participate in the Raman and Rayleigh scattering also increases. Such phenomena are called the ***hyper Raman* or *hyper Rayleigh scattering***. Fig. illustrates hyper Rayleigh and hyper Raman scattering related to the second term of equation with involvement of two photons

$$P_i = \chi_{ij}^{(1)} E_j + \chi_{ijk}^{(2)} E_j E_k + \chi_{ijkl}^{(3)} E_j E_k E_l$$

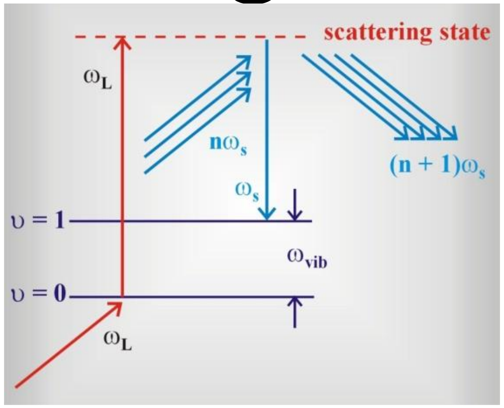


Stimulated Raman Scattering



The difference comes from the intensity of the laser incident beam. When the incident light exceeds a certain threshold in the material, it can enhance or stimulate the rate of Raman Stokes (or anti-Stokes) emission. This stimulated Raman scattering shows a similarity to the stimulated emission in the optically pumped laser. The intense incident beam at frequency ω_L generates the intense Stokes radiation at the frequency $\omega_s = \omega_L - \omega_{vib}$. The Stokes radiation, in turn, stimulates additional Stokes scattering from the virtual scattering state. In contrast to the linear Raman scattering, the stimulated Raman scattering can achieve 50% or higher conversion of the pump wavelength to the Stokes line. There is a full analogy to the phenomena of the stimulated emission. The only difference is that previously we considered the stationary real quantum energy state in contrast to the virtual scattering state. For the intensities above the material threshold the incident and the scattered radiation at ω_L and ω_s act simultaneously on a system of molecules leading to creation of phase coherent vibrations at the frequency of $\omega_{vib} = \omega_L - \omega_s$

Stimulated Raman Scattering



The stimulated Raman scattering is a special case of *four-wave interaction*

$$\omega_4 = \omega_1 + \omega_2 \pm \omega_3 \qquad \Delta\boldsymbol{k} = \boldsymbol{k}_1 + \boldsymbol{k}_2 \pm \boldsymbol{k}_3 - \boldsymbol{k}_4 = 0$$

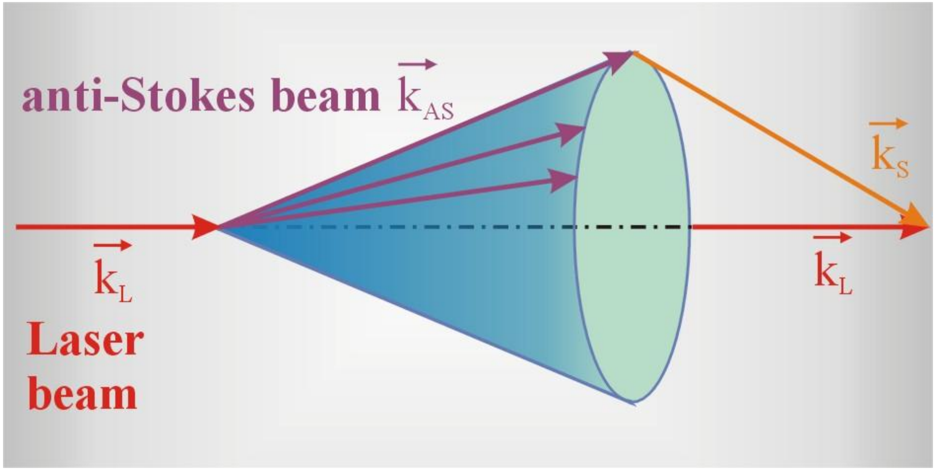
$$I_S^{SRS} = const \cdot \left(\left(\chi^{(3)}\right)\right)^2 I_L^2 I_S l^2 \left(\frac{sin\Delta kl/2}{\Delta kl/2}\right)^2$$

For the stimulated Stokes scattering the phase matching condition is given by

$$\Delta\boldsymbol{k} = \boldsymbol{k}_1 + \boldsymbol{k}_2 \pm \boldsymbol{k}_3 - \boldsymbol{k}_4 = 0$$

is established automatically

Direction of stimulated anti-Stokes scattering



the direction of wave propagation of the stimulated anti-Stokes scattering is limited to the cone surface with the axis parallel to the direction of the incident laser beam

$$\boldsymbol{k}_{AS} = 2\boldsymbol{k}_L - \boldsymbol{k}_S$$

The stimulated anti-Stokes scattering in turn, can be described as fully non-degenerated four-wave interaction

$$\omega_{AS} = \omega_L + \omega_{vib} = \omega_L + (\omega_L - \omega_s)$$

$$\Delta\boldsymbol{k} = \boldsymbol{k}_L + \boldsymbol{k}_L - \boldsymbol{k}_S - \boldsymbol{k}_{AS} = 2\boldsymbol{k}_L - \boldsymbol{k}_S - \boldsymbol{k}_{AS}$$

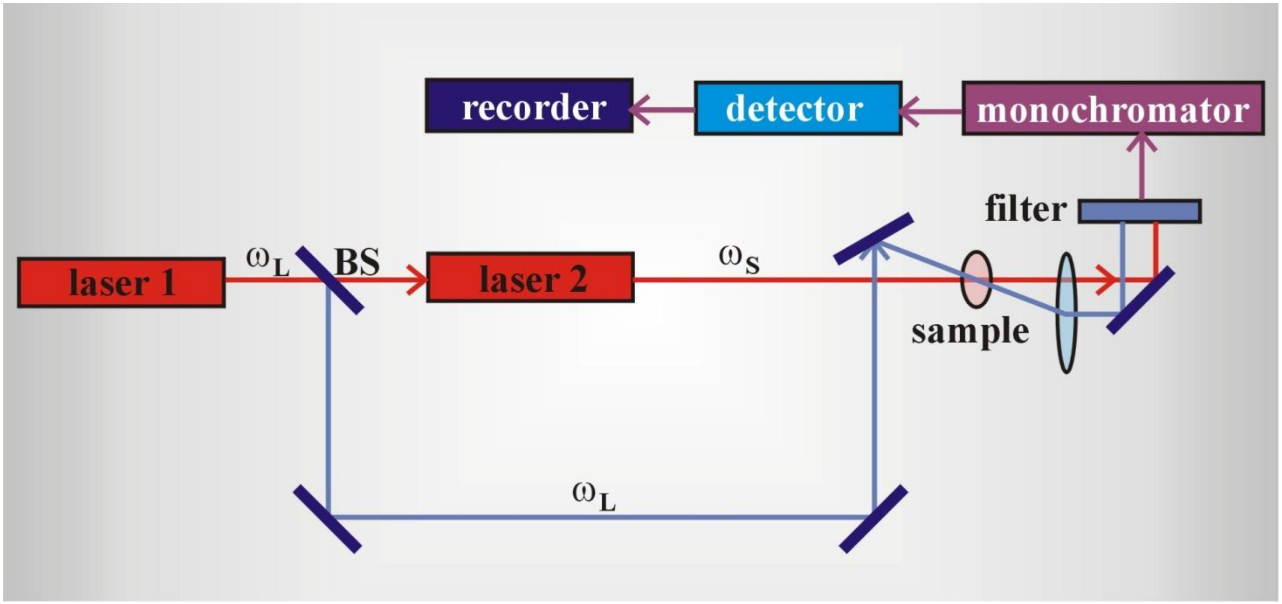
This indicates that the phase matching condition is achieved only for certain directions for which the relation occurs

$$\boldsymbol{k}_{AS} = 2\boldsymbol{k}_L - \boldsymbol{k}_S$$

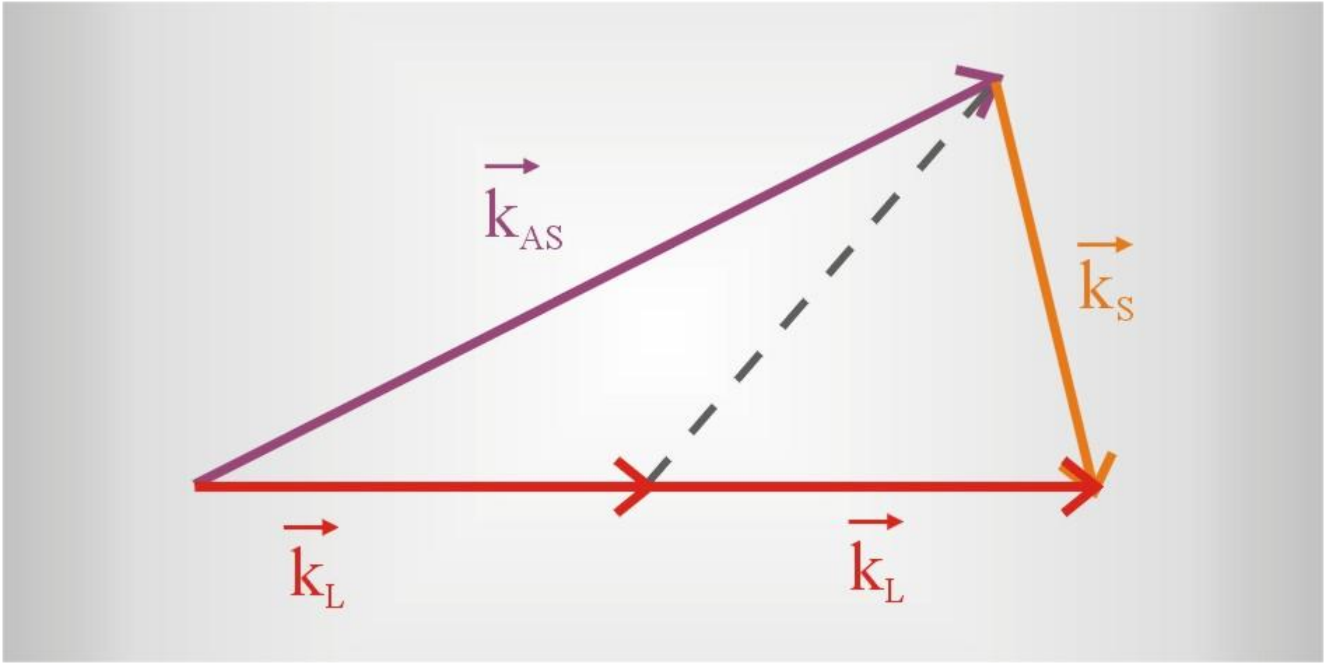
- For the stimulated Stokes scattering the phase matching condition $(\Delta\mathbf{k} = 0)$ occurs for all directions and the scattered light can be emitted in every direction. The stimulated anti-Stokes scattering is observed in the directions of $\mathbf{k}_{\mathrm{AS}}$ determined by the phase matching condition $2\mathbf{k}_L - \mathbf{k}_{\mathrm{S}} = \mathbf{k}_{\mathrm{AS}}$.

CARS- Coherent anti-Stokes Raman scattering

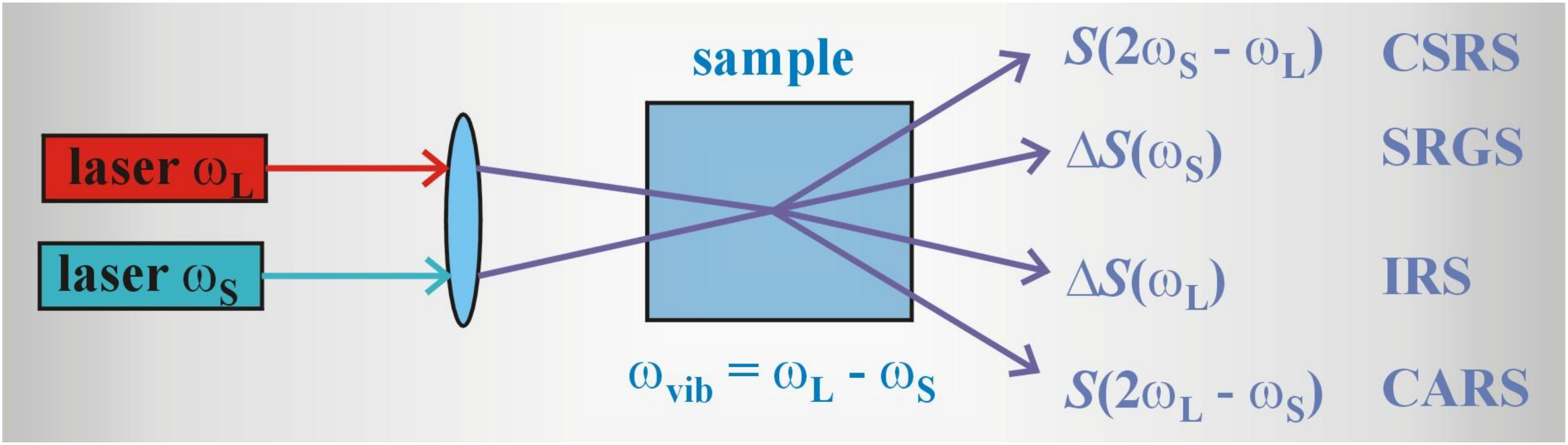
CARS (and CSRS) are techniques based on SRS (stimulated Raman Scattering) that use two laser beams to excite a sample: at the frequency ω_L and ω_S (instead of one at the frequency ω_L in SRS). The phenomena are similar to those described in the previous section, but the CARS (and CSRS) signals are much stronger. Moreover, by tuning the frequency ω_S (or ω_L) to the resonance with a molecular vibration $\omega_L - \omega_S = \omega_{vib}$ it is possible to excite most of the vibrations in contrast to SRS for which only the strongest vibrational lines could be observed.



The stimulated anti-Stokes scattering that is generated with only one beam at the frequency ω_L is observed on the cone surface with the cone axis parallel to the wave vector in contrast to CARS where the stimulated anti-Stokes scattering that is generated with two beams ω_L and ω_S is observed only in one direction determined by the plane of the vectors and

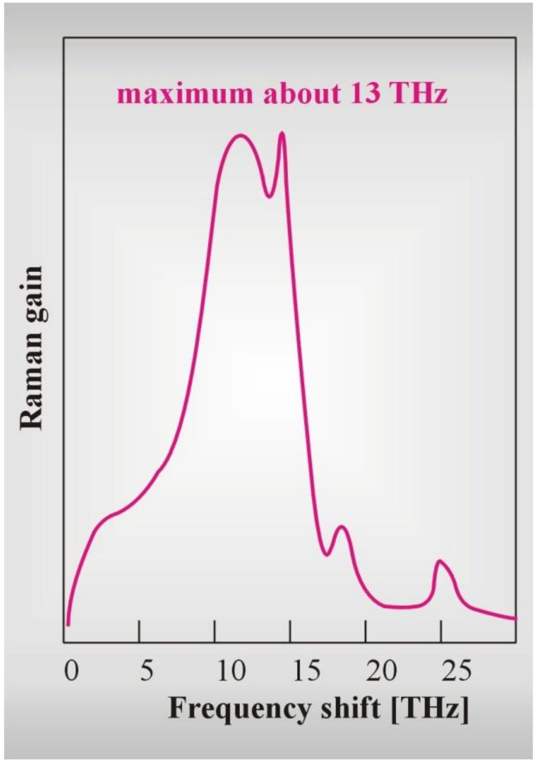


Besides the coherent stimulated anti-Stokes (CARS) and Stokes (CSRS) Raman scattering there are many other techniques of the coherent stimulated Raman effect, such as SRGS (*Stimulated Raman Gain Spectroscopy*) and IRS (*Inverse Raman Scattering*). The nonlinear four-wave interaction of the incident beams at frequencies ω_L or ω_S with the electric field originating from the coherent vibrations of molecules at the frequency ω_{vib} can cause the gain not only at the frequency $2\omega_S - \omega_L$ (CSRS) and $2\omega_L - \omega_S$ (CARS) but also the Stokes gain or the intensity loss of the incident beam . The former method related to the Stokes gain is known as SRGS (Stimulated Raman Gain Stokes) method and the latter is known as IRS (Inverse Raman Scattering). The schematic diagram of the most commonly used Raman techniques is presented in fig.



RAMAN AMPLIFIER in OPTICAL FIBER TECHNOLOGY

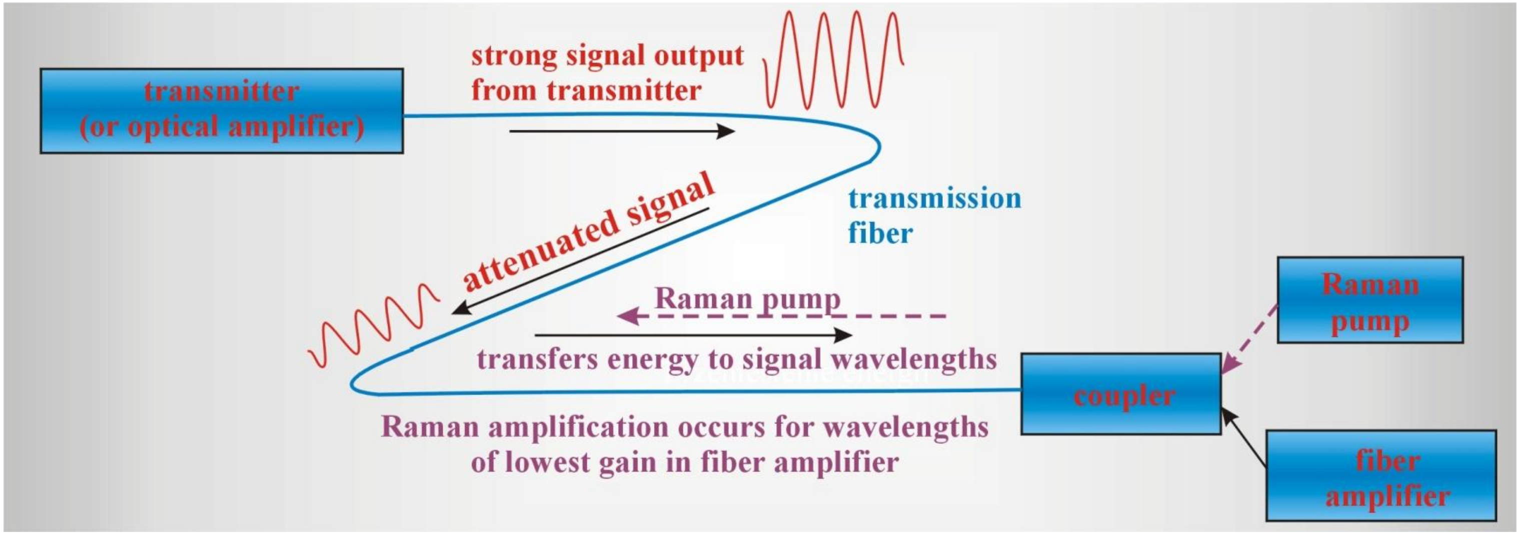
The stimulated Raman scattering is emerging as a crucial technology for high-speed, long-distance fiberoptic transmission. The stimulated Raman scattering amplifies the input signal in an optical fiber in the same way as the stimulated emission from erbium atoms amplifies signals in EDFA (erbium-doped fiber amplifiers). The crucial difference comes from the fact that the wavelength of the Raman gain is shifted to higher wavelengths (Stokes component) with respect to the pump pulse $\omega_S = \omega_p - \omega_{vib}$ (; where ω_{vib} is the vibrational frequency of molecules in the fiber - glass itself or a dopant).



Cross section for stimulated Raman scattering in silica shows the shift peaking at a frequency 13 THz from the stimulating wavelength, by about 100 nm in the 1550 nm window

RAMAN AMPLIFIER in OPTICAL FIBER TECHNOLOGY

In the usual configuration the Raman amplification occurs in the final length of fiber before the receiver or EDFA The signal and pump beams travel in the opposite directions, with the pump coupled into the fiber at the receiver end. A coupler directs the pump light into the transmitting fiber, while diverting signals arriving through the fiber to the receiver or EDFA.



Distributed Raman amplification amplifies signals passing through fiber in the transmission cable by transferring energy from a strong pump beam to the less powerful signal wavelengths

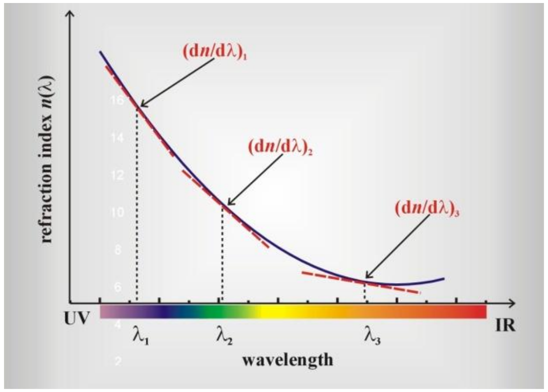
Nonlinear dispersion phenomena affecting picosecond and femtosecond pulse duration

The dispersion properties of an active medium become more and more important for short pulses.
For femtosecond lasers, particularly for pulses shorter than 100 fs, the dispersion properties of the active medium and the optical elements in the resonator begin to affect significantly the pulse duration.
Moreover, the output beam emitted from the laser usually travels through additional optical elements (mirrors, prisms, crystals, beamsplitters, filters, etc) before it reaches a detector.
The dispersion has a tremendous influence on the pulse duration.

There are two main mechanisms resulting in modification of the pulse:
•group velocity dispersion (GVD) and
•self phase modulation (SPM).

There are many methods to compensate the GVD effect

- prism pairs for the femtosecond pulses
- Gires-Tournois interferometer for picosecond pulses
- Pair of diffraction gratings
- Chirped mirrors



Perfect modelocking

The magnitude of the product $\Delta t \cdot \Delta E$

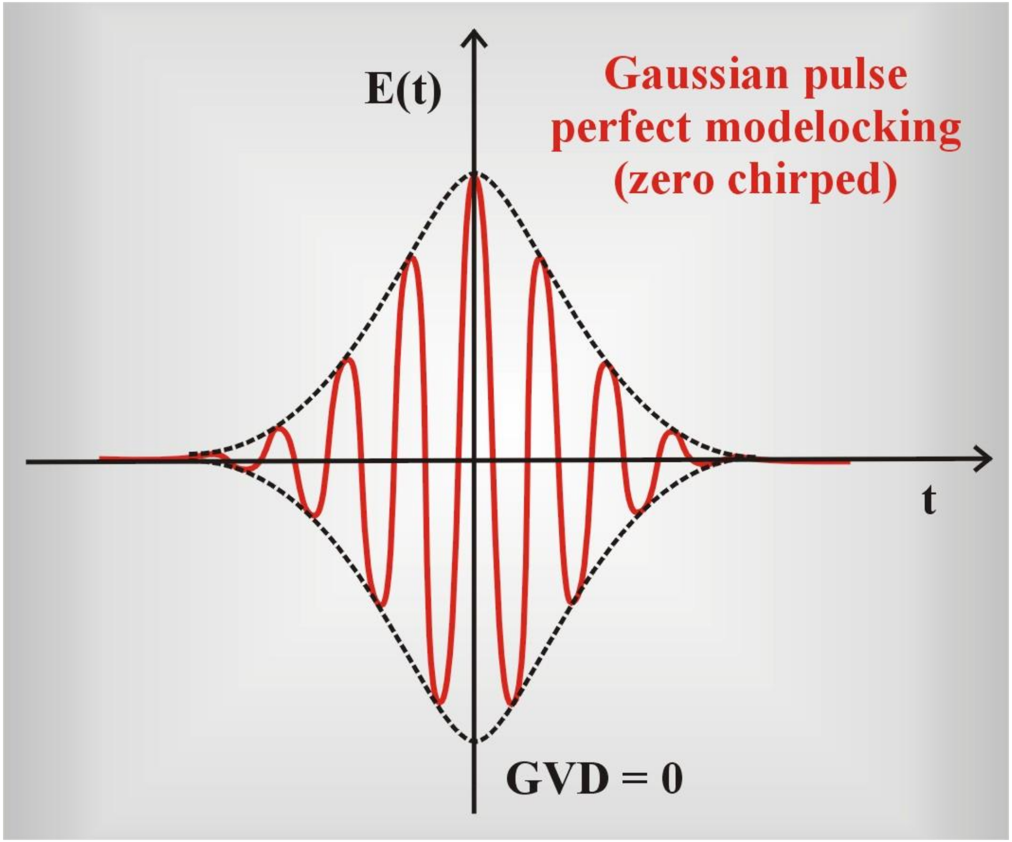
depends on a temporary pulse shape. Assume that the temporary pulse shape is described by a Gaussian function

$$E(t) = \frac{E_0}{\tau} \exp\left(-\frac{t^2}{2\tau^2} \right)$$

The frequency spectrum $E(\omega)$

can be obtained from the Fourier transform

$$E(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} E(t) e^{-i\omega t} dt = \frac{E_0}{2\pi} \exp\left[-\frac{\tau^2}{2} (\omega - \omega_0)^2 \right]$$

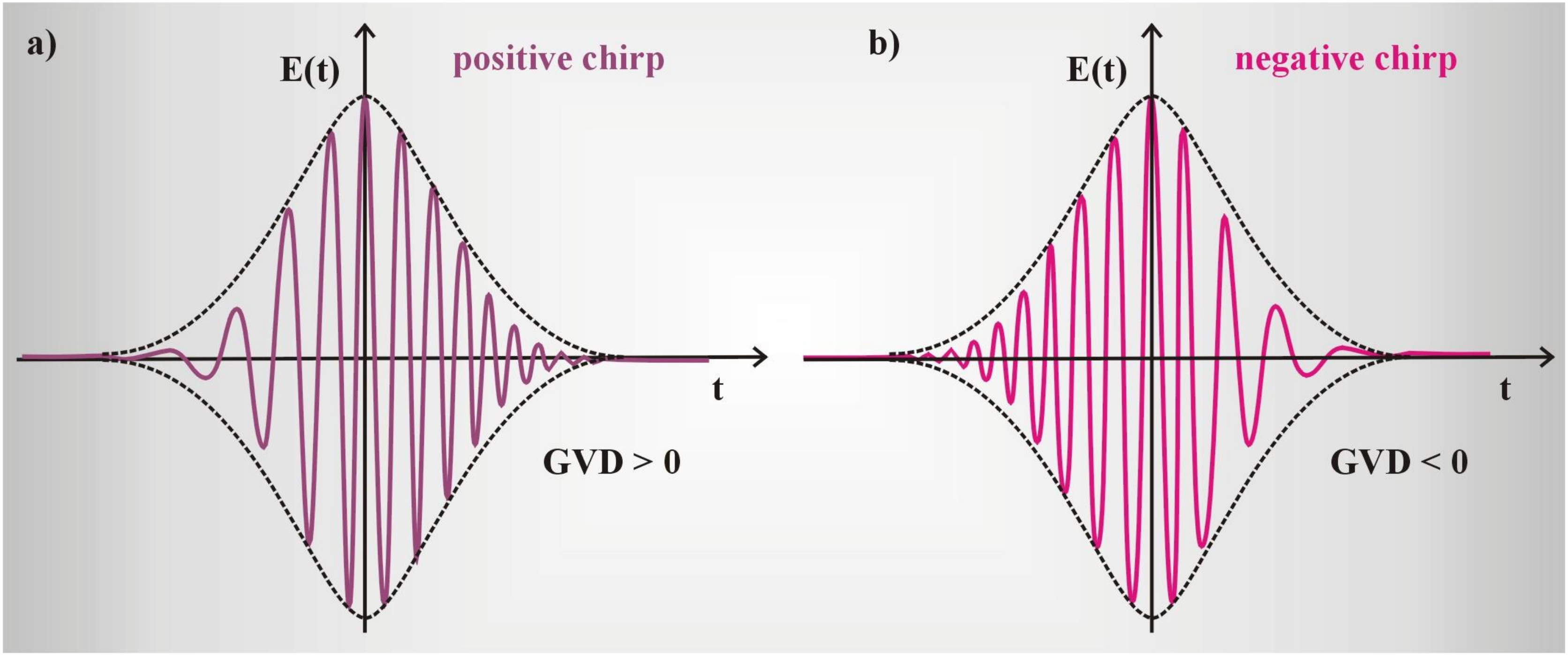


Thus, for the Gaussian profile the product $\Delta t \cdot \Delta E$ is equal to $\Delta t_{FWHH} \cdot \Delta \nu_{FWHH} = 0.441$

The relation derived in eq. corresponds to an ideal situation of a perfectly modelocked laser with a pulse called the *Fourier-transform limited pulse*. Such a pulse is the shortest pulse Δt_{FWHH} that can be generated for a given gain spectrum $\Delta \nu_{\mathrm{FWHH}}$

In practice such pulses are seldom produced.

Real world (dispersion)



To produce pulses as short as possible, dispersion in the cavity must be compensated for by adding optical elements – typically pairs of prisms or gratings, specially coated mirrors or a length of optical fiber

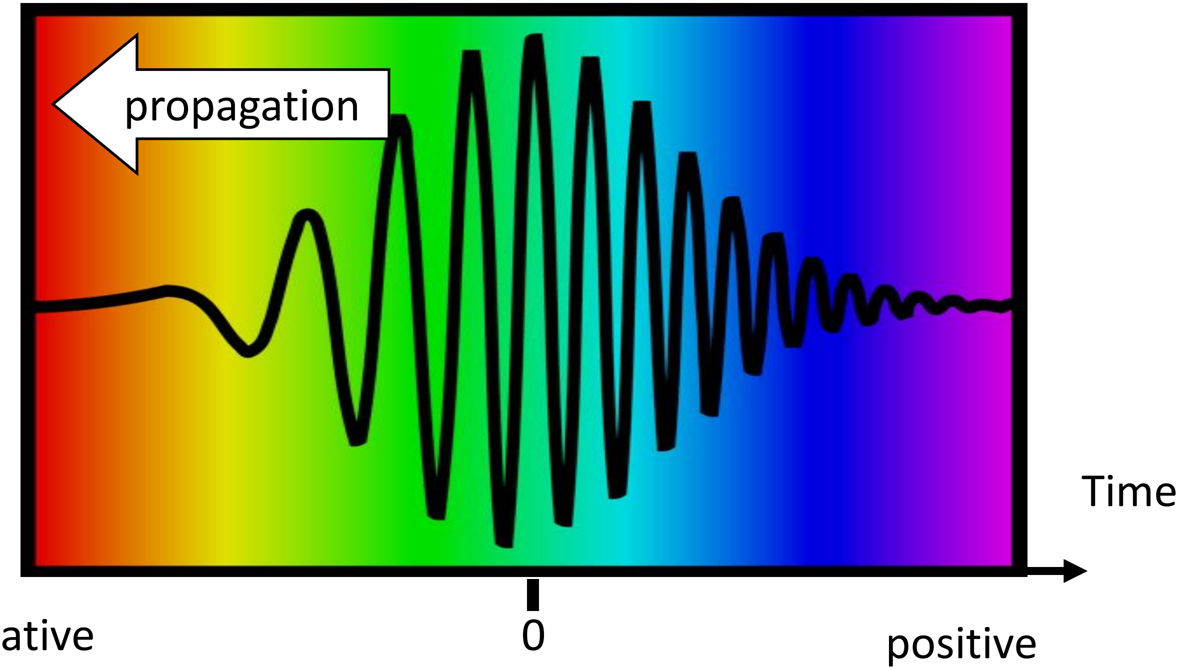
The Chirp (Instantaneous frequency)

G. Steinmeyer, European Virtual University, http://mitr.p.lodz.pl/evu

The temporal phase, $\phi(t)$, contains frequency-vs.-time information.

The pulse *instantaneous angular frequency*, $\omega_{inst}(t)$, is defined as:

$$\boxed{\omega_{inst}(t) \equiv \omega_0 - \frac{d\phi}{dt}}$$



This pulse increases its frequency linearly in time (from red to blue).

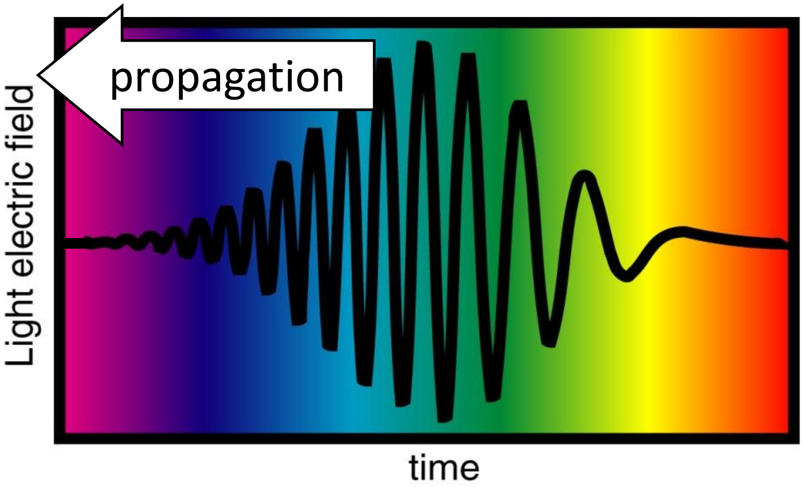
In analogy to bird sounds, this pulse is called a "chirped" pulse.
This pulse is positively chirped, i.e., red leading blue, as from material dispersion !

The Negatively Chirped Pulse

- We have been considering a pulse whose frequency *increases*
- linearly with time: a *positively* chirped pulse.

- One can also have a *negatively*
- chirped (Gaussian) pulse, whose
- instantaneous frequency
- *decreases* with time.

- We simply allow β to be *negative*
- in the expression for the pulse:



$$E(t) = \operatorname{Re} E_0 \exp\left[-(t/\tau_G)^2\right] \exp\left[i\left(\omega_0 t + \beta t^2\right)\right]$$

- And the instantaneous frequency will decrease with time:

$$\omega_{inst}(t) = \omega_0 + 2\beta t = \omega_0 - 2|\beta| t$$

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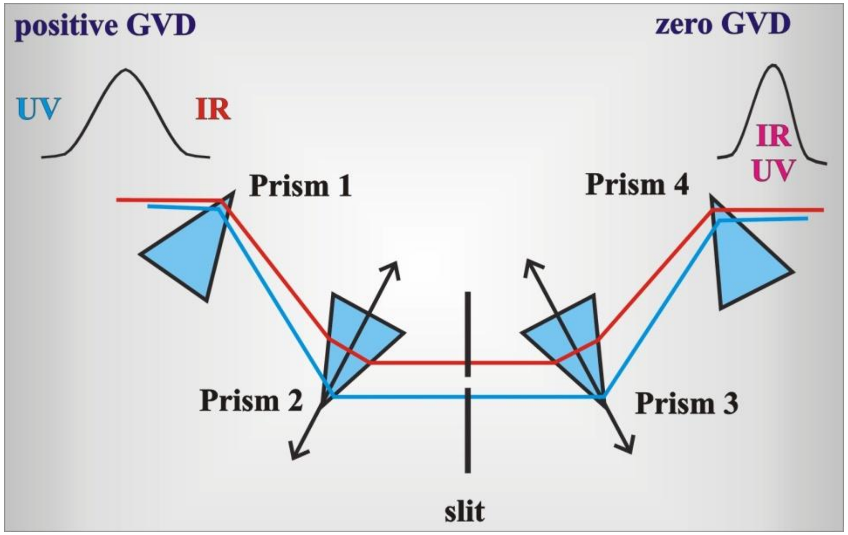
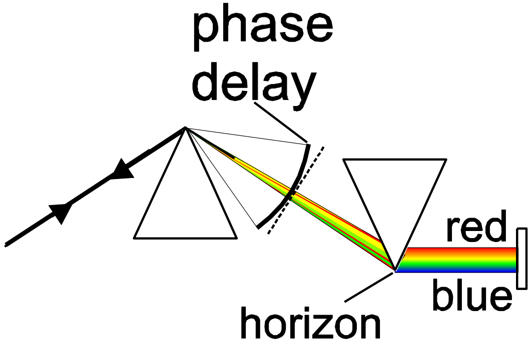
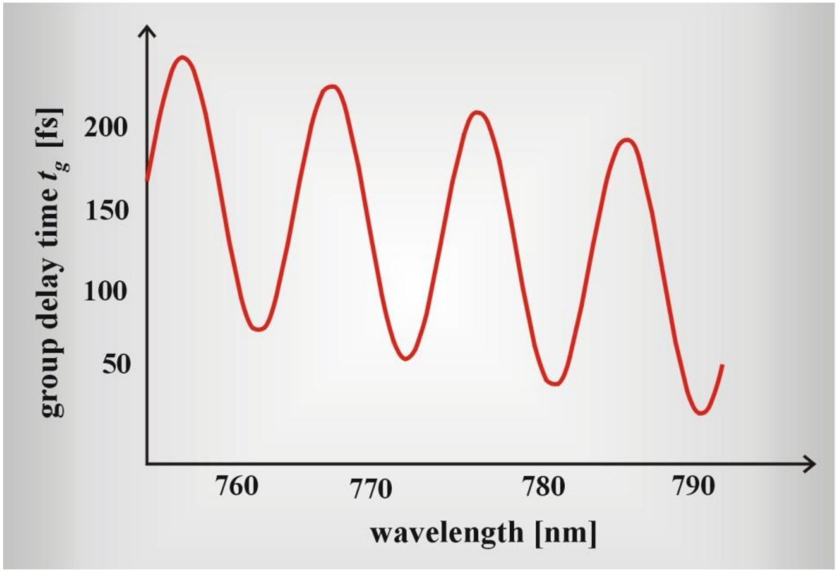
Prism pairs for the femtosecond pulses





A positively chirped pulse from the cavity of the resonator travels to the prism P_1, where the different spectral components of the pulse are separated. Then, the broadened pulse enters the prisms P_2 and P_3 at the Brewster angle (to minimize losses). Since the glass of the prisms exhibits a positive GVD, the red wavelengths exhibit larger group velocities than the blue wavelengths of the pulse. However, the red wavelengths travel a longer path length in the prisms P_2 and P_3 than the blue wavelengths. By translating prisms P_2 and P_3 perpendicularly to their bases, it is possible to select such a length of the optical path for which the group velocity dispersion will be compensated. As a consequence, all spectral components of the pulse reach the prism P_4 at the same time, the pulse is free of chirping. As a consequence the output pulse is shorter than the input pulse and has an ideal shape, free of GVD effects. The prism P_4 liquidates the spatial distribution of the spectral components.

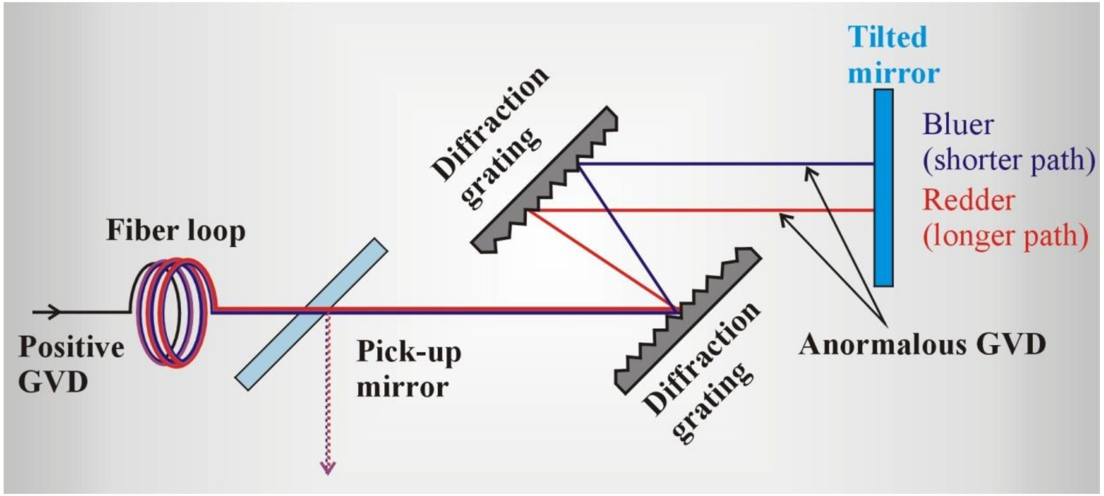
Gires-Tournois interferometer for picosecond pulses



Group delay time t_g as a function of wavelength for a Gires-Tournois interferometer

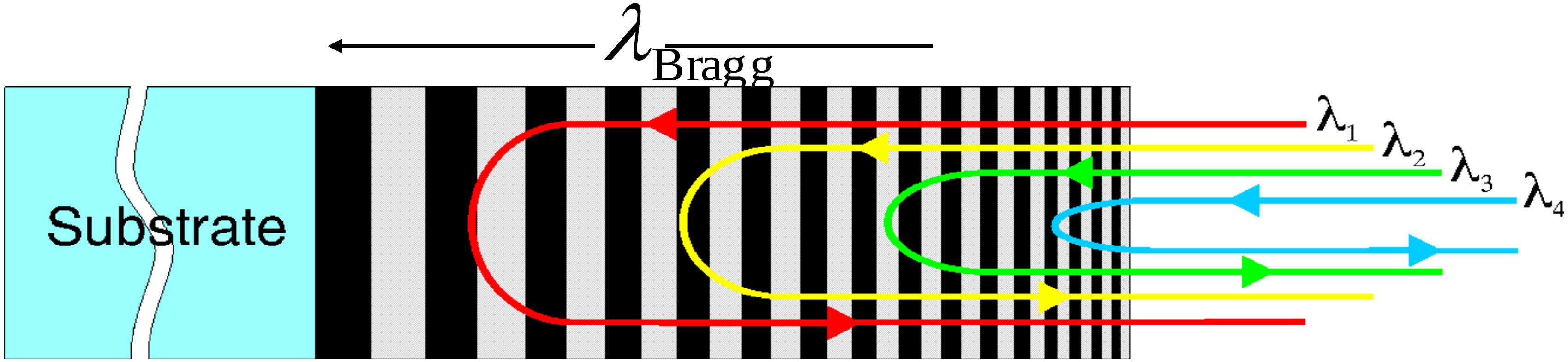
In picosecond lasers GVD compensation is usually performed with a Gires-Tournois interferometer. The Gires-Tournois interferometer consists of a pair of parallel surfaces spaced by *d*. One surface is a partial reflector (reflectivity *r* <<100%), while the other one has 100% reflectivity. Typical spacing is on the order of a few dozen micrometers, and the reflectivity *r* is on the order of several percent

pair of diffraction gratings



During the propagation in the fiber the pulse is affected both by SPM and GVD. The resulting pulse has its spectral width significantly increased with a significant positive chirp, which is almost linear. Then, the positively chirped pulse passes through an external pair of gratings, which are designed to produce negative GVD. The negative GVD designed pair of grating offset the positive chirp from the positive GVD and SPM by making the optical path length of the redder components substantially greater than the path for the bluer components. Therefore, the trailing bluer edge of the pulse catches up to the leading redder edge, and all components begin to travel at the same velocity. As a result, the output pulse exhibits zero GVD, and is compressed to less than its initial value.

Chirped mirrors



A chirped mirror is a dielectric mirror with chirped spaces—spaces of varying depth designed to reflect varying wavelengths of lights—between the dielectric layers (stack). Chirped mirrors are used in applications like lasers to reflect a wider range of light wavelengths than ordinary dielectric mirrors, or to compensate for the dispersion of wavelengths that can be created by some optical elements

R. Szipöcs et al., *Opt. Lett.* 19, 201 (1994)

G. Steinmeyer, European Virtual University, http://mitr.p.lodz.pl