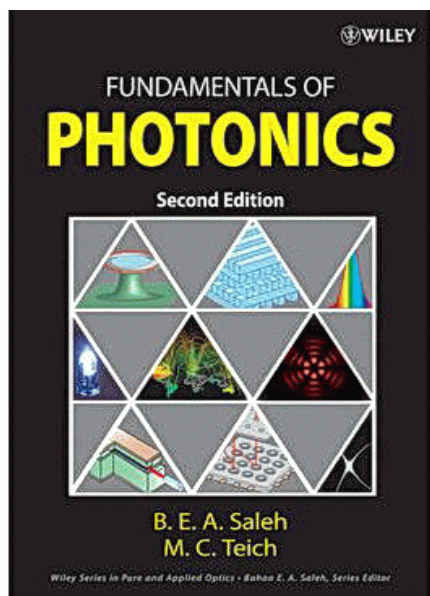


# Quantum Electronics

## Lecture 4



Introduction to  
nonlinear optics

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# Contents

- ◆ **Nonlinear polarization - physical origin**
- ◆ **Wave mixing - Complex notation**
- ◆ **Conservation laws for elastic NL interactions**
- ◆ **Second harmonic generation**
- ◆ **Birefringence and Quasi-Phase Matching**
- ◆ **Four Wave Mixing**
- ◆ **Stimulated Raman Scattering**



# Motto

*“Physics would be dull and uninteresting and life most unfulfilling if all physical phenomena around us were linear. Fortunately we are living in a non-linear world. While linearization beautifies physics, non-linearity provides excitement”*

Y.R. Shen in “Non-Linear Optics”, Wiley



# Where from the nonlinear effects come?

In contrast to electrons photons can **only** interact through polarization of the medium

Incident photons make an electron cloud or a molecule oscillate

Those oscillations in turn result in photon emission

**Linear regime:** (small deviations from equilibrium) the emitted photons have the same frequency as the incident ones – there is no effective interaction between photons

**Nonlinear regime:** (more photons, large oscillation amplitude) different frequency (higher harmonics) are generated – photons effectively "interact"

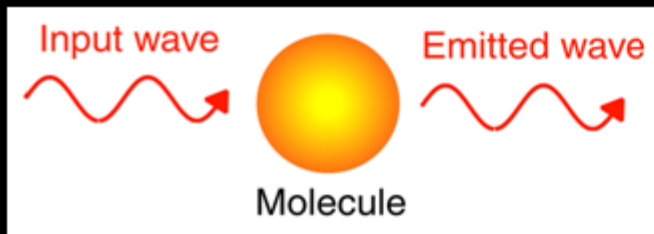
All media are nonlinear

However, for low light intensities the nonlinear response of the medium is typically negligible in analogy with a harmonic oscillator

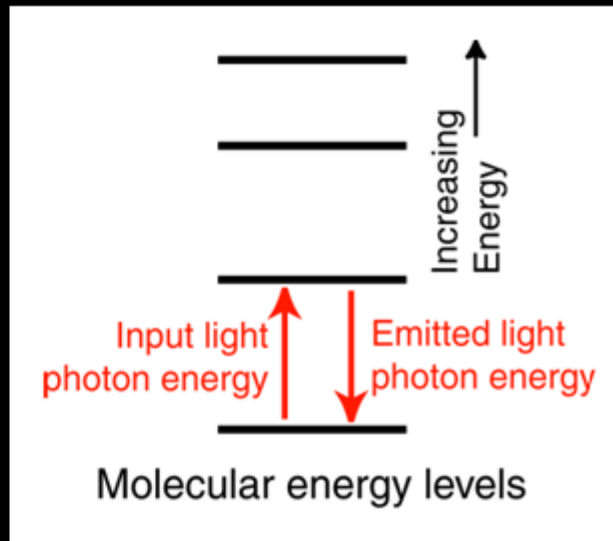


# Linear response of the medium

Recall that, in normal linear optics, a light wave acts on a molecule, which vibrates and then emits its own light wave that interferes with the original light wave.

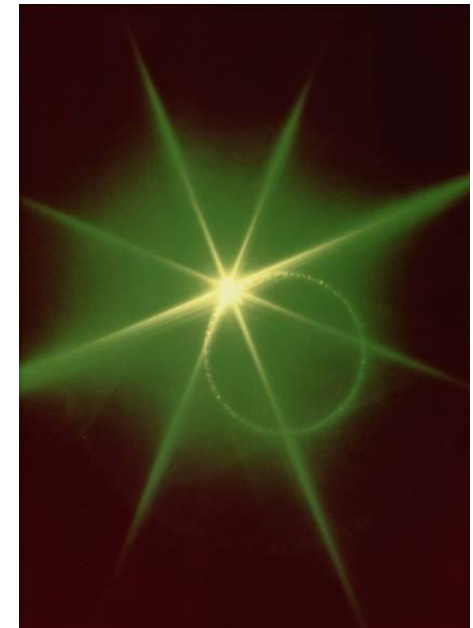
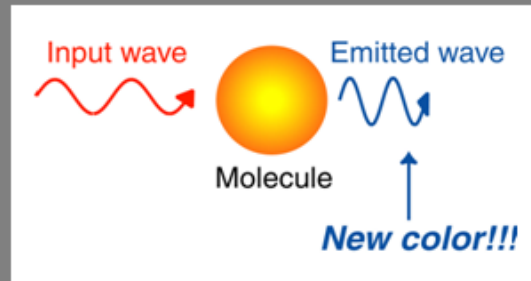
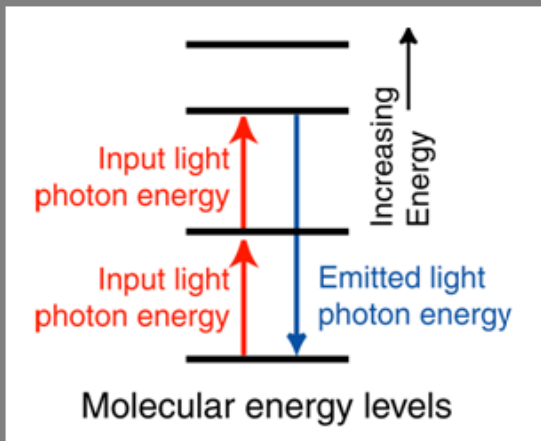


We can also imagine this process in terms of the molecular energy levels, using arrows for the photon energies:



# Origin of nonlinear effects

Now, suppose the irradiance is high enough that many molecules are excited to the higher-energy state. This state can then act as the lower level for additional excitation. This yields vibrations at all frequencies corresponding to all energy differences between populated states.

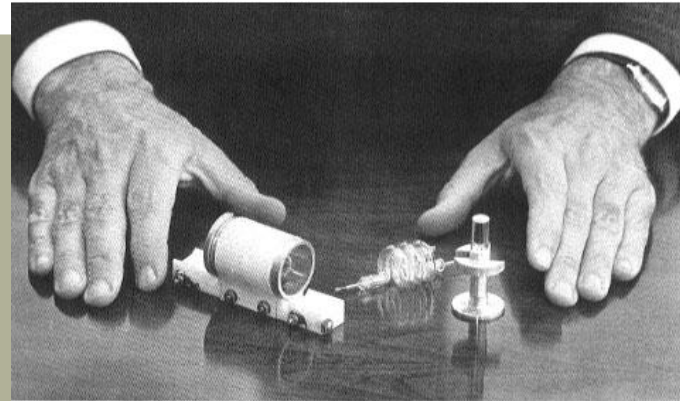
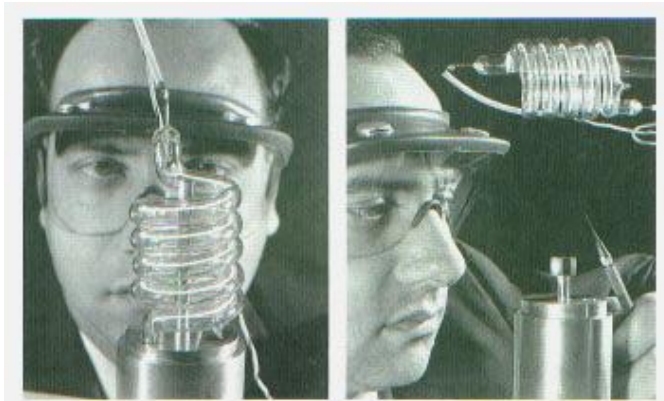


*Green light obtained by illuminating the crystal with infrared light*

Gatech

# Invention of the first lasers

(1960) **Theodore Maiman** Invention of the first Ruby Laser



(1960) **Ali Javan**  
The first He-Ne Laser

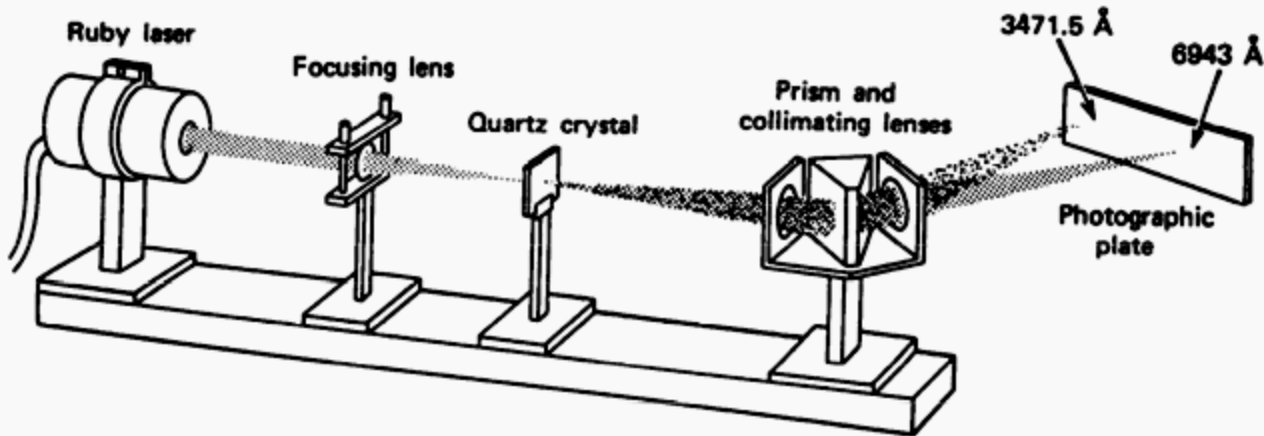


# Birth of nonlinear optics

The advent of the laser as an intense, coherent light source gave **birth to nonlinear optics**

**Optical Second-Harmonic Generation – the first nonlinear effect observed with coherent input generating coherent output**

P.A. Franken, et al, Physical Review Letters 7, p. 118 (1961)



Peter Franken

**Figure 12.1.** Arrangement used in the first experimental demonstration of second-harmonic generation [1]. A ruby-laser beam at  $\lambda = 0.694 \mu\text{m}$  is focused on a quartz crystal, causing the generation of a (weak) beam at  $\frac{1}{2}\lambda = 0.347 \mu\text{m}$ . The two beams are then separated by a prism and detected on a photographic plate.



# First theoretical description of nonlinear phenomena

**N. Bloembergen, Harvard, Cambridge (a book “Nonlinear Optics”, 1964)**

**R.V. Khokhlov and S.A. Akhmanov, Moscow University (a book “Problems of Nonlinear Optics”, 1964)**



Nicolas Bloembergen



Rem Khokhlov



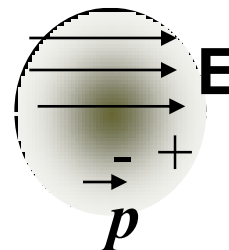
Sergey Akhmanov

# Induced polarization – medium response

The polarization is induced by E-field:

$$\mathbf{P} = f(\mathbf{E})$$

$$\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P}$$



$\mathbf{D}$  is the electric flux density - caused by:

- E-field in the absence of the medium:  $\varepsilon_0 \mathbf{E}$ ,
- Plus the field created by the response of the medium:  $\mathbf{P}$ 
  - $\mathbf{P} = 0$  in free space
  - $\mathbf{P} \neq 0$  in a dielectric

***Polarization is a driving term for the wave equation:***

$$\frac{\partial^2 E}{\partial x^2} - \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P}{\partial t^2}$$



# Nonlinear polarization

For high intensity fields oscillatory motion of bound electrons becomes anharmonic in analogy to a simple pendulum motion: sufficiently small oscillations are harmonic - the larger ones include **higher harmonics**



**Higher order (nonlinear) terms** in the induced polarization **P**:

$$P = \varepsilon_0 \left[ \chi^{(1)} E + \chi^{(2)} EE + \chi^{(3)} EEE + \dots \right]$$

$\uparrow$   
 $P_L$

$\uparrow$   
 $P_{NL}$

$\chi^{(n)}$  -  $n$ th order susceptibility **tensor**

Generally  $(\chi^{(1)} \mathbf{E} \gg \chi^{(2)} \mathbf{E} \mathbf{E} \gg \chi^{(3)} \mathbf{E} \mathbf{E} \mathbf{E})$

In centrosymmetric media:  $P(-E) = -P(E) \Rightarrow \chi^{(2n)} = 0$

$\chi^{(2)}$  - only in media lacking a centre of symmetry (i.e. crystals)  
 $\chi^{(3)}$  - in all dielectric media

*Note alternative definition in some books (e.g. Saleh) :  $\varepsilon_0 \chi^{(2)} \rightarrow 2d$   $\varepsilon_0 \chi^{(3)} \rightarrow 4\chi^{(3)}$*



# Wave mixing by 2nd order nonlinearity

Let us look at **polarization** frequencies generated by a sum of two waves:

$$E = E_1 \cos(\omega_1 t - k_1 x) + E_2 \cos(\omega_2 t - k_2 x) = \frac{1}{2} \left[ E_1 e^{i\omega_1 t} e^{-ik_1 x} + E_2 e^{i\omega_2 t} e^{-ik_2 x} + E_1^* e^{-i\omega_1 t} e^{ik_1 x} + E_2^* e^{-i\omega_2 t} e^{ik_2 x} \right]$$

$$P \propto \chi^{(2)} EE$$

$$P \propto \chi^{(2)} \left[ E_1^2 e^{i2\omega_1 t} e^{-i2k_1 x} + E_2^2 e^{i2\omega_2 t} e^{-i2k_2 x} \right. \\ \left. + 2E_1 E_2 e^{i(\omega_1 + \omega_2)t} e^{-i(k_1 + k_2)x} \right. \\ \left. + 2E_1 E_2^* e^{i(\omega_1 - \omega_2)t} e^{-i(k_1 - k_2)x} \right. \\ \left. + |E_1|^2 + |E_2|^2 \right]$$

**2nd-harmonic generation**

**Sum-frequency generation**

**Difference-frequency generation**

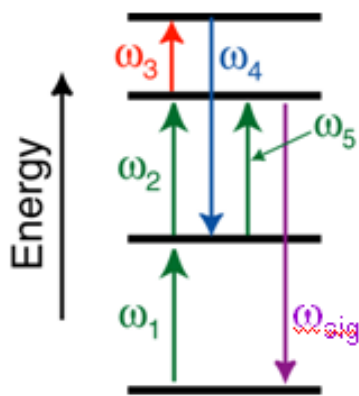
**dc rectification**

Can light at those frequencies be efficiently generated?

Answer: only if the conservation laws are fulfilled – next slide



# Conservation laws for elastic nonlinear processes



N-wave-mixing (or N-photon) mixing

N - number of photons involved (including the emitted one)

$\chi^{(2)}$  can mix 3 waves,  $\chi^{(3)}$  - 4 waves, etc

Absorbed photons

Emitted photons



- contribute to **complex conjugate** of their field

$$P \propto E_1 E_2 E_3 E_4^* E_5$$

Energy must be conserved:  $\omega_1 + \omega_2 + \omega_3 - \omega_4 + \omega_5 = \omega_{sig}$

**Automatic !**

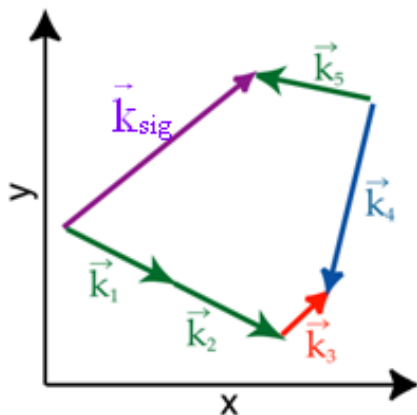
( $\hbar$ 's are cancelled)

Momentum must

also be conserved:

$$\vec{k}_1 + \vec{k}_2 + \vec{k}_3 - \vec{k}_4 + \vec{k}_5 = \vec{k}_{sig}$$

Typically,  $\vec{k}_{sig}$  DOES NOT correspond to a light wave at frequency  $\omega_{sig}$  !



Satisfying these two relations simultaneously is called **"phase-matching"**

# Phase-matching problem for SHG

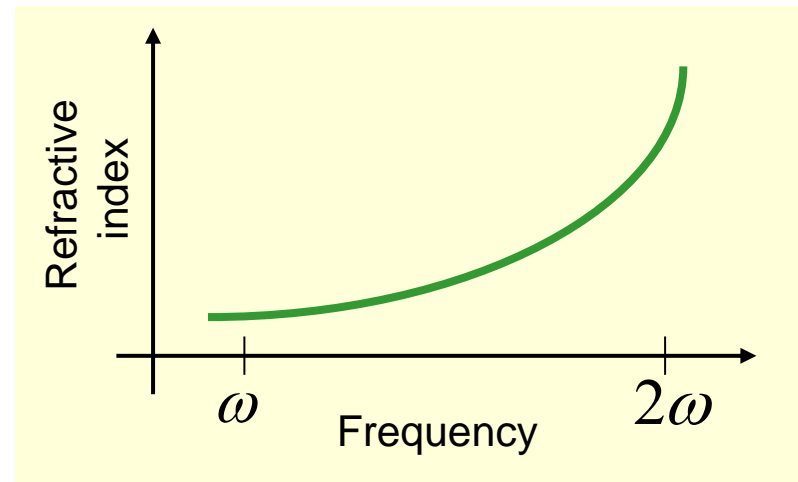
$k$ -vector of the polarization:  $k_{sig} = 2k_{\omega} = 2 \frac{\omega}{c} \underline{n(\omega)}$

$k$ -vector of the second harmonic:  $k_{2\omega} = \frac{2\omega}{c} \underline{n(2\omega)}$

For phase-matching they should be equal:  $k_{sig} = k_{2\omega}$   
which requires:  $n(2\omega) = n(\omega)$

Unfortunately, dispersion prevents this from ever happening!

Dispersion couples energy and momentum conservation



”Tricks” to enforce phase matching will be discussed later

# 2nd order NL tensor – contracted notation

*Due to intrinsic permutation symmetries one can reduce the NL polarization tensor to its contracted form. For the 2nd order polarization it is:*

$$\begin{array}{c} \left| \begin{array}{c} P_x \\ P_y \\ P_z \end{array} \right| = \left| \begin{array}{cccccc} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{array} \right| \left| \begin{array}{c} E_x^2 \\ E_y^2 \\ E_z^2 \\ 2E_z E_y \\ 2E_z E_x \\ 2E_x E_y \end{array} \right| \end{array}$$

$$P_i = \chi_{ijk}^{(2)} E_j E_k \equiv 2d_{ijk} E_j E_k \equiv \sum_{jK} E_j E_K = 2d_{iK} (EE)_K$$

# Units of $d$ (F/m or m/V)

- There are different conventions in the use of  $d$
- in some texts you will find that the polarisation is written as

$$P = \epsilon_0 d E^2$$

- while in other texts you will find polarisation written as

$$P = d E^2$$

(we use  $d$  in italics here simply to distinguish it from the  $d$  above)

- recall that the polarisation  $P$  is the dipole moment (C.m) per unit volume ( $m^3$ ). Units of  $P$  are  $C.m^2$
- units of  $E$  are  $V.m^{-1}$
- units of  $\epsilon_0$  are  $F.m^{-1}$  or  $C.V^{-1}.m^{-1}$
- units of  $d$  are  $d = P / (\epsilon_0 E^2) = C.m^2 / (C.V^{-1}.m^{-1}.V^2.m^{-2}) = m.V^{-1}$
- units of  $d$  are  $d = P / E^2 = C.m^2 / (V.m^{-1}) = F.m^{-1}$





# Three-Wave Mixing - mathematical description

Maxwell equations:

$$\left. \begin{aligned} \nabla_{\mathbf{x}} \bar{\mathbf{E}} &= -\frac{\partial \bar{\mathbf{B}}}{\partial t} \\ \nabla_{\mathbf{x}} \bar{\mathbf{B}} &= \mu_0 \frac{\partial}{\partial t} (\epsilon_0 \bar{\mathbf{E}} + \bar{\mathbf{P}}) \\ \bar{\mathbf{P}} &= \epsilon_0 \chi^{(2)} \bar{\mathbf{E}} + \bar{\mathbf{P}}_{NL} \end{aligned} \right\} \Rightarrow \boxed{\nabla^2 \mathbf{E}} = \mu_0 \frac{\partial^2}{\partial t^2} (\epsilon_0 \mathbf{E} + \mathbf{P}) = \boxed{\mu_0 \epsilon \frac{\partial^2 \mathbf{E}}{\partial t^2} + \mu_0 \frac{\partial^2 \mathbf{P}_{NL}}{\partial t^2}}$$

Wave equations:

Consider interaction of three harmonic fields via  $\chi^{(2)} \equiv 2d_{ijk}$  with  $\omega_3 = \omega_1 + \omega_2$  Crystal axes

Express each of the fields :  $E_i^{\omega_1}(t) = \frac{1}{2} (E_{0i}^{\omega_1} e^{i(\omega_1 t - k_1 z)} + \text{c.c.}) = \frac{1}{2} (a_{1i} E_1 e^{i(\omega_1 t - k_1 z)} + \text{c.c.})$  ( $i = x', y', z'$ )

Unit polarization vectors

Mix the total field:  $E_i(t) = E_i^{\omega_1}(t) + E_i^{\omega_2}(t) + E_i^{\omega_3}(t)$

to obtain nonlinear polarization:  $P_{iNL} = P_i = 2d_{ijk} E_j E_k$

$$P_i(t) = 2d_{ijk} \frac{1}{2} (E_{0j}^{\omega_1} e^{i\omega_1 t} + E_{0j}^{\omega_2} e^{i\omega_2 t} + \text{c.c.}) \times \frac{1}{2} (E_{0k}^{\omega_1} e^{i\omega_1 t} + E_{0k}^{\omega_2} e^{i\omega_2 t} + \text{c.c.})$$

Group terms at different frequencies:

$$[P_{NL}^{\omega_3 - \omega_2}(z, t)]_i = d_{ijk} a_{3j} a_{2k} E_3 E_2^* e^{i[(\omega_3 - \omega_2)t - (k_3 - k_2)z]} + \text{c.c.}$$

$$[P_{NL}^{\omega_3 - \omega_1}(z, t)]_i = d_{ijk} a_{3j} a_{1k} E_3 E_1^* e^{i[(\omega_3 - \omega_1)t - (k_3 - k_1)z]} + \text{c.c.}$$

$$[P_{NL}^{\omega_1 + \omega_2}(z, t)]_i = d_{ijk} a_{1j} a_{2k} E_1 E_2 e^{i[(\omega_1 + \omega_2)t - (k_1 + k_2)z]} + \text{c.c.}$$

# Coupled equations for Three Wave Mixing (1)

Substituting nonlinear polarizations to the wave equation, and applying Slowly Varying

Amplitude Approximation:  $\frac{d^2}{dz^2} E_s \ll k_s \frac{d}{dz} E_s \quad (s = 1, 2, 3)$

we obtain, after a few steps of algebra,

$$\begin{aligned}\frac{d}{dz} E_1 &= -i\omega_1 \sqrt{\frac{\mu_0}{\epsilon_1}} d E_3 E_2^* e^{-i(k_3 - k_2 - k_1)z} \\ \frac{d}{dz} E_2^* &= +i\omega_2 \sqrt{\frac{\mu_0}{\epsilon_2}} d E_1 E_3^* e^{+i(k_3 - k_2 - k_1)z} \\ \frac{d}{dz} E_3 &= -i\omega_3 \sqrt{\frac{\mu_0}{\epsilon_3}} d E_1 E_2 e^{+i(k_3 - k_2 - k_1)z}\end{aligned}$$

where  $d$  is the effective second-order nonlinear coefficient

$$d = \sum_{ijk} d_{ijk} a_{1i} a_{2j} a_{3k}$$

Energy is conserved:

$$\frac{d}{dz} \left( \sqrt{\epsilon_1} |E_1|^2 + \sqrt{\epsilon_2} |E_2|^2 + \sqrt{\epsilon_3} |E_3|^2 \right) = 0$$



# Coupled equations for Three Wave Mixing (2)

Introducing variables:  $A_m = \sqrt{\frac{n_m}{\omega_m}} E_m$ ,  $m = 1, 2, 3$   $n$ -index of refractio  $|A_m|^2 \propto$  photon flux  
 one obtains:

$$\begin{aligned} \frac{d}{dz} A_1 &= -i\kappa A_3 A_2^* e^{-i\Delta k z} \\ \frac{d}{dz} A_2^* &= +i\kappa A_1 A_3^* e^{+i\Delta k z} \\ \frac{d}{dz} A_3 &= -i\kappa A_1 A_2 e^{+i\Delta k z} \end{aligned}$$

$\Delta k = k_3 - (k_1 + k_2)$  Momentum mismatch

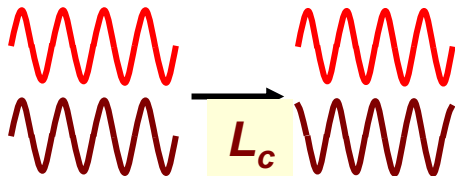
Coupling coefficient:

$$\kappa = d \sqrt{\frac{\mu_0 \omega_1 \omega_2 \omega_3}{\epsilon_0 n_1 n_2 n_3}} = \left( \sum_{ijk} d_{ijk} a_{1i} a_{2j} a_{3k} \right) \sqrt{\frac{\mu_0 \omega_1 \omega_2 \omega_3}{\epsilon_0 n_1 n_2 n_3}}$$

Interaction most efficient for:  $\Delta k=0$  (phase matching)

Collinear propagation - large field overlaps and large interaction lengths

Constructive interaction only possible over coherence length  $L_c$ :



$$\frac{1}{L_c} \propto \Delta k = \left| \vec{k}_p - (\vec{k}_s + \vec{k}_i) \right|$$

# Conservation laws

$$\Rightarrow \boxed{\frac{\partial}{\partial z} I_1 + \frac{\partial}{\partial z} I_2 + \frac{\partial}{\partial z} I_3 = 0} \quad \text{energy conservation} \quad I_i = \frac{1}{2} \sqrt{\frac{\mu_0}{\epsilon_0}} n_i |E_i|^2 = \frac{1}{2} \sqrt{\frac{\mu_0}{\epsilon_0}} \omega_i |A_i|^2$$

$$\text{and : } \boxed{\frac{\partial}{\partial z} \left( \frac{I_1}{\omega_1} \right) = \frac{\partial}{\partial z} \left( \frac{I_2}{\omega_2} \right) = - \frac{\partial}{\partial z} \left( \frac{I_3}{\omega_3} \right)} \quad \text{Manley-Rowe relations}$$

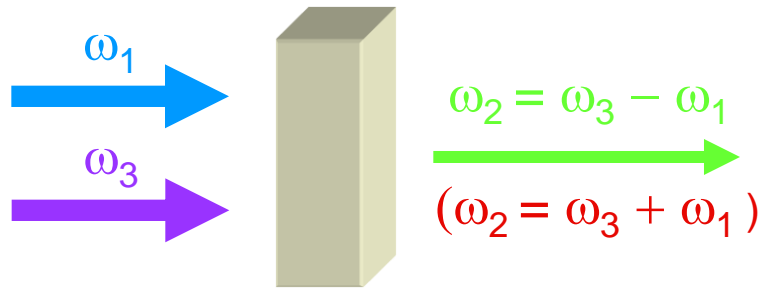
*note* : the ratio of intensity  $I$  and freq.  $\omega$  is proportional to the number of photons :  $I = \frac{N \hbar \omega}{\Delta t \Delta A} \propto N \omega$

$\Rightarrow$  interpretation of the Manley-Rowe relation : the generation of one photon at freq.  $\omega_3$  requires the annihilation of one photon at  $\omega_1$  and one photon at  $\omega_2$

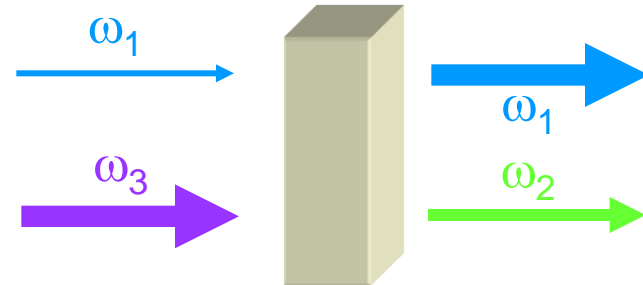
$$\Phi = \frac{I}{h\omega} \quad \text{Photon flux density}$$



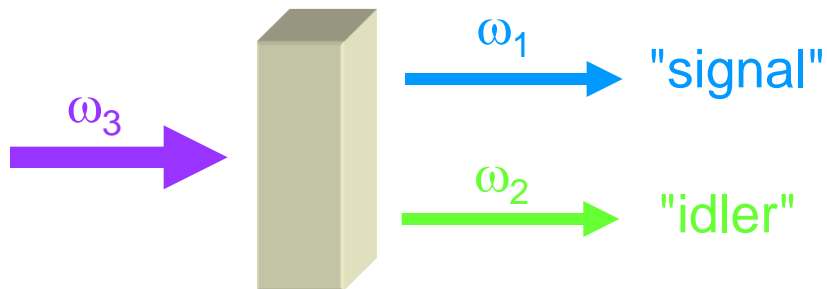
# Examples of three waves mixing effects



Parametric Down (Up)-Conversion  
 [Difference (Sum)-frequency generation]

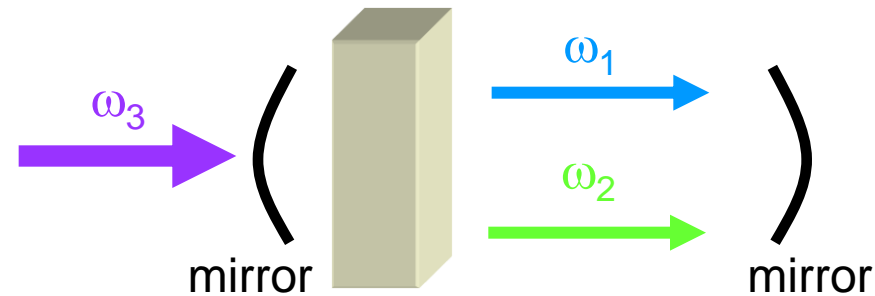


Optical Parametric Amplification (OPA)



Optical Parametric Generation (OPG)

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



Optical Parametric Oscillation (OPO)

**Strong pump laser at  $\omega_3$  amplifies a weak, phase matched, signal at  $\omega_1$**

# Second Harmonic Generation

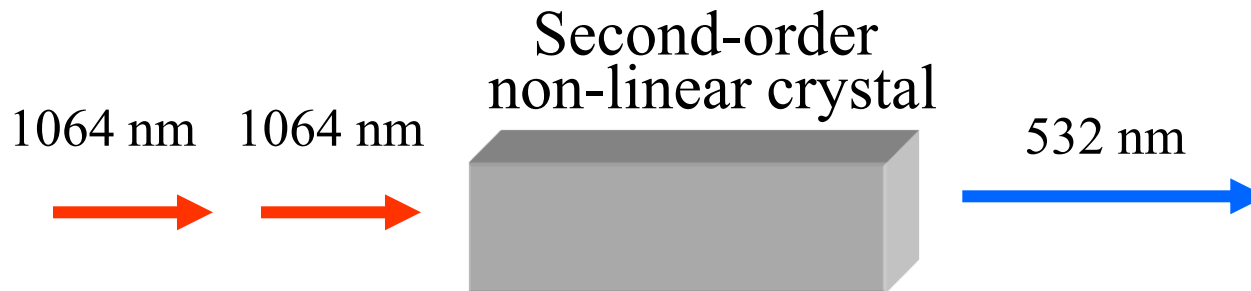
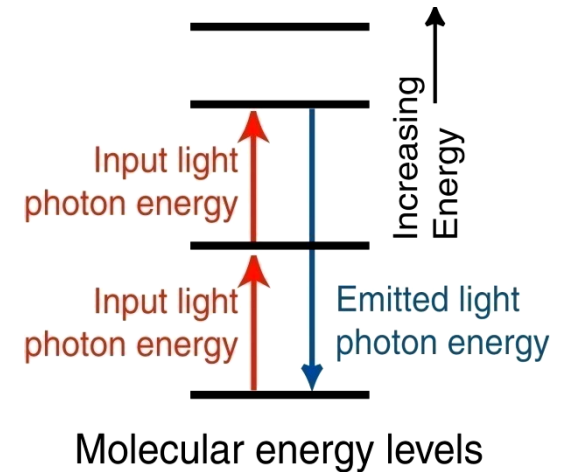
Input intense beam at frequency  $\omega$  to 2<sup>nd</sup> order nonlinear crystal

Since  $E(t) \propto E_0 \exp(i\omega t) + E_0^* \exp(-i\omega t)$ ,

$$E(t)^2 \propto E_0^2 \exp(2i\omega t) + 2|E_0|^2 + E_0^{*2} \exp(-2i\omega t)$$



**$2\omega = 2\text{nd harmonic!}$**



# Second Harmonic Generation - SHG

## Undepleted pump solution

$$\omega_1 = \omega_2 \equiv \omega \quad \omega_3 = 2\omega \quad A_1 = A_2 \equiv A \approx \text{const}$$

$$\frac{dA_3}{dz} \approx -i \frac{1}{2} \kappa A^2 e^{i\Delta k z}$$

$$A_3(L) = -i \frac{1}{2} \kappa A^2 \frac{e^{i\Delta k L} - 1}{i\Delta k} = -i \frac{1}{2} \kappa A^2 L e^{i(\Delta k/2)L} \left[ \frac{\sin[(\Delta k/2)L]}{(\Delta k/2)L} \right]$$

## Conversion efficiency for SHG

$$\eta_{SHG} = \frac{I^{(2\omega)}}{I^{(\omega)}} = \frac{2\omega^2 d^2 L^2}{n^3} \left( \frac{\mu_0}{\epsilon_0} \right)^{3/2} \left[ \frac{\sin[(\Delta k/2)L]}{(\Delta k/2)L} \right]^2 I^{(\omega)}$$

Note interaction length and intensity dependence



# Coherence length and Beat length

$$A_3(L) = -iA^2 \frac{\kappa}{\Delta k} e^{i(\Delta k/2)L} \sin[(\Delta k/2)L] = \begin{cases} A^2 \frac{\kappa}{\Delta k} & \text{for } \underline{(\Delta k/2)L = \pi/2} \\ 0 & \text{for } \underline{(\Delta k/2)L = \pi} \end{cases}$$



$$L_C = \frac{\pi}{\Delta k}$$

**Coherence length**

$$L_B = \frac{2\pi}{\Delta k}$$

**Beat length**

$\rightarrow \infty$  for  $\Delta k \rightarrow 0$



# Depleted Pump SHG

In the treatment of SHG so far was assumed that the input intensity at  $\omega$  was not affected by the interaction, i. e. that the pump remained undepleted. This limits the validity of the result to situations where the fraction of the power converted from  $\omega$  to  $2\omega$  is small.

Generally the conversion efficiency for **phase matched** SHG is:

$$\eta \equiv \frac{I^{(2\omega)}}{I^{(\omega)}} = \tanh^2 \left[ \frac{A_1(z=0)}{\sqrt{2}} \kappa z \right],$$

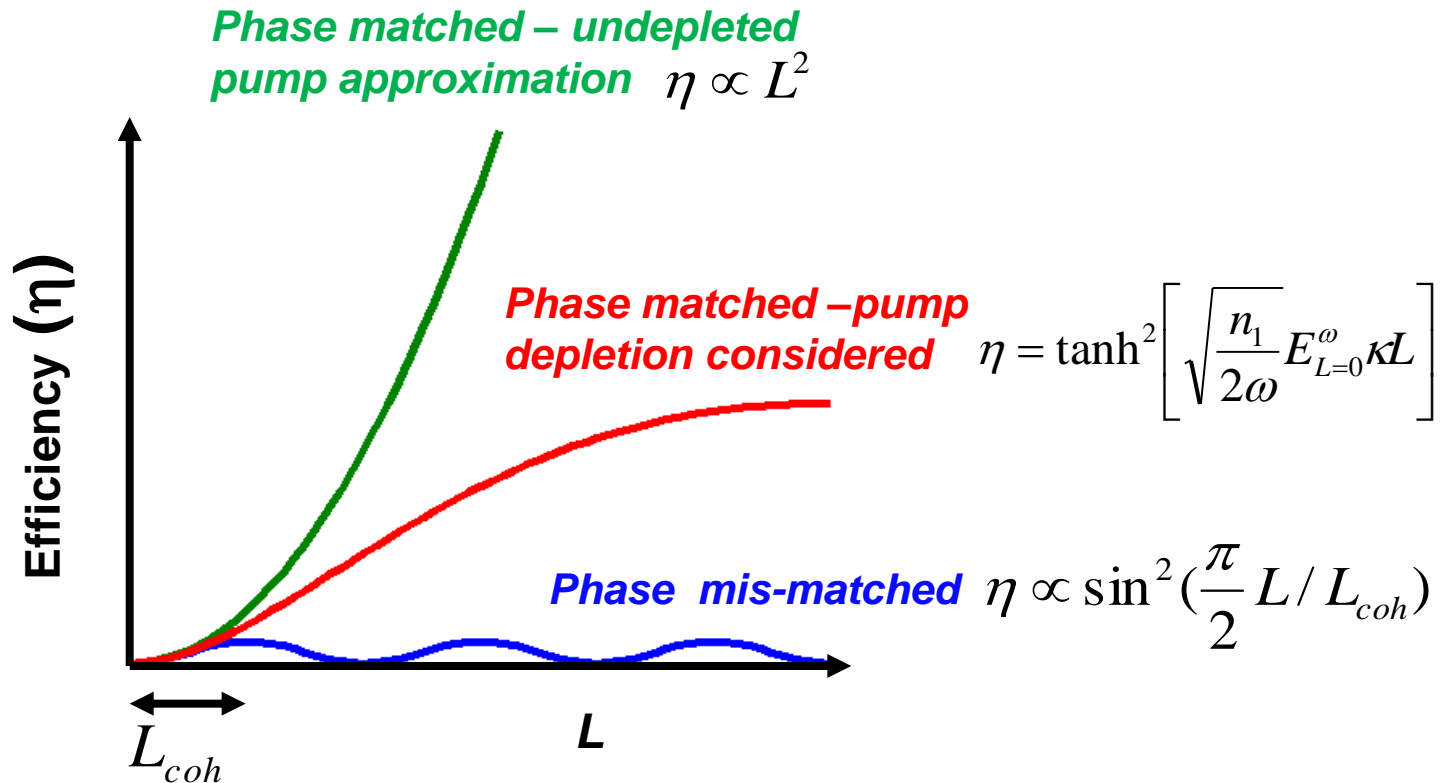
$$\text{where } A_1 = \sqrt{\frac{n_1}{\omega}} E_1^\omega,$$

$$\text{and } \kappa \equiv d \sqrt{\frac{\mu_0}{\epsilon_0} \frac{\omega_1 \omega_2 \omega_3}{n_1 n_2 n_3}}.$$

$$\text{with: } \begin{aligned} \omega_1 &= \omega_2 = \omega \\ \omega_3 &= 2\omega \end{aligned}$$

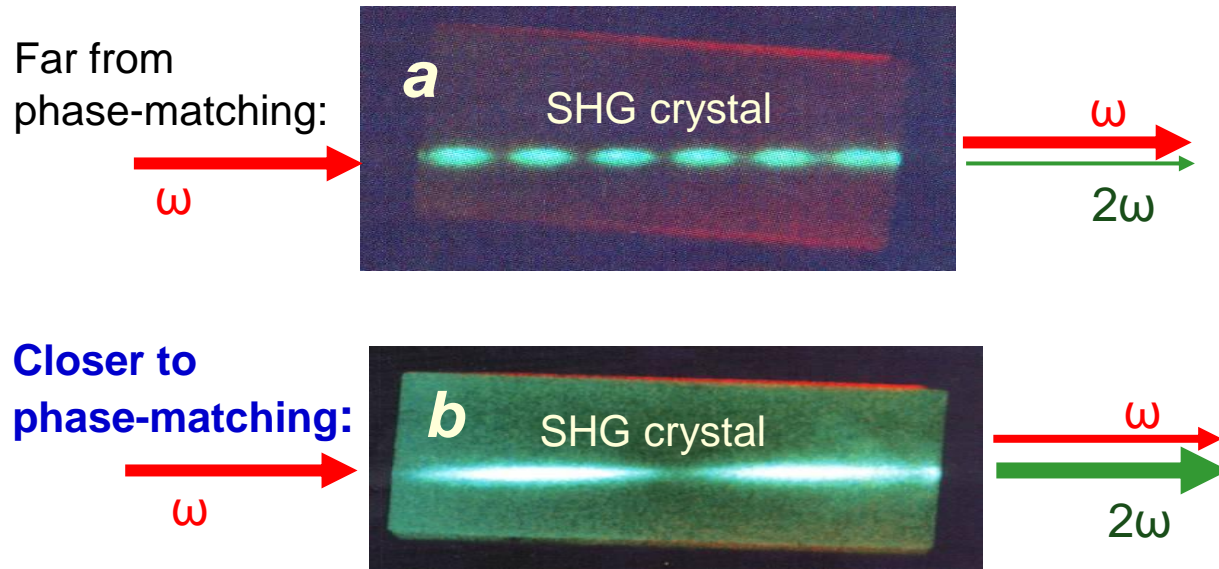


# SH conversion efficiency vs phase mismatch



# SH evolution in nonlinear crystals

$$L_B = \frac{2\pi}{\Delta k}$$



Oscillation period (beat length) increases and the SH beam becomes brighter  
 - conversion efficiency increases

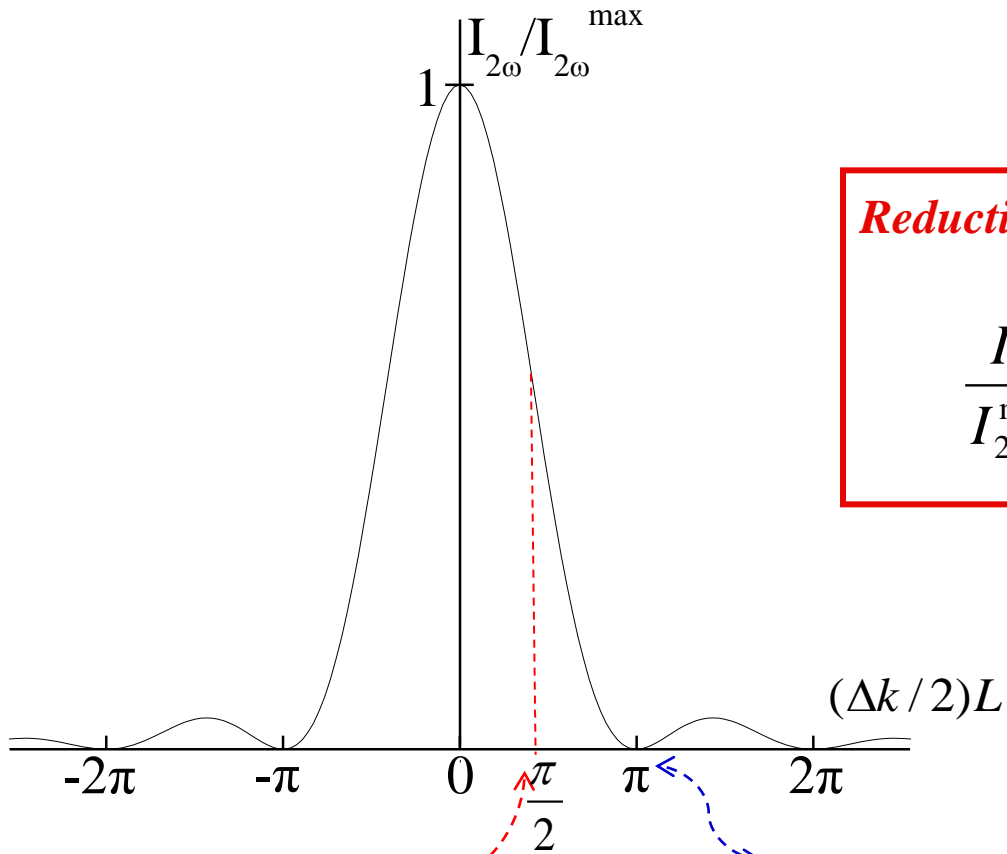
$$n_a(\omega) = n_b(\omega), \quad n_a(2\omega) \neq n_b(2\omega)$$

$$\Delta_a n \equiv n_a(2\omega) - n_a(\omega), \quad \Delta_b n \equiv n_b(2\omega) - n_b(\omega)$$

$$\Delta_b k \equiv k_0 \Delta_b n < \Delta_a k \equiv \Delta_a n$$

# Phase mismatch penalty for SHG

## for fixed crystal length



**Reduction** of SH output by the factor:

$$\frac{I_{2\omega}}{I_{2\omega}^{\max}} = \left[ \frac{\sin[(\Delta k / 2) L]}{(\Delta k / 2) L} \right]^2$$

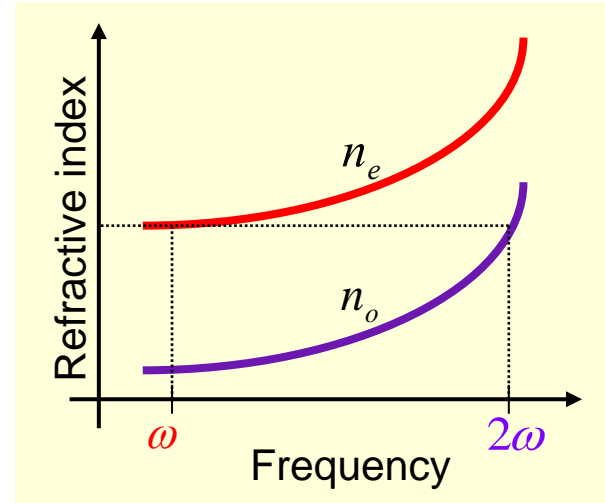
$\Delta k = 2\pi / L_B$  Mismatch at which the crystal length = *beat length*

$\Delta k = \pi / L_C$  Mismatch at which the crystal length = *coherence length*

# SHG – birefringence phase-matching

One can utilize this for satisfying phase-matching condition

In positive uniaxial crystals  $n_e > n_o$  chose the extraordinary polarization for  $\omega$  and the ordinary for  $2\omega$ .

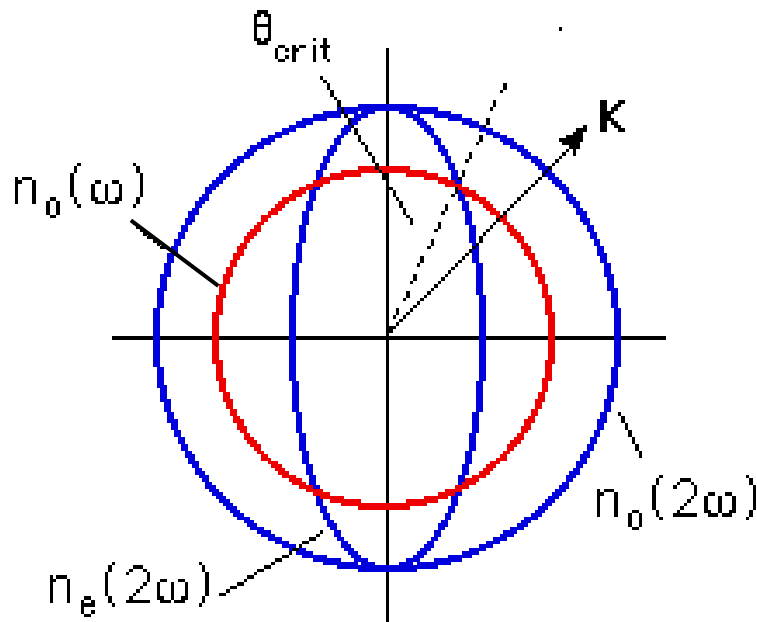


$n_e$  depends on propagation angle, so we can tune for a given  $\omega$ .

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



# Birefringence Phase-Matching



If  $n_e < n_o$ , (negative uniaxial crystal) there exists an angle  $\theta_{crit}$  at which  $n_e^{2\omega}(\theta_{crit}) = n_o^\omega$ . So if the fundamental beam (at  $\omega$ ) is launched along  $\theta_{crit}$  as an ordinary ray, the second-harmonic beam will be generated along the same direction as an extraordinary ray

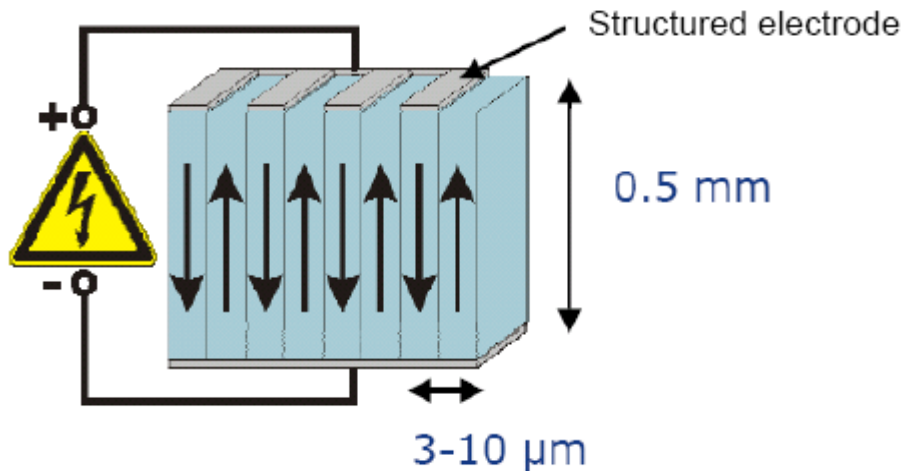
$$\frac{1}{n_e^2(\theta)} = \frac{\cos^2 \theta}{n_o^2} + \frac{\sin^2 \theta}{n_e^2}$$

# Quasi-Phase Matching (QPM)

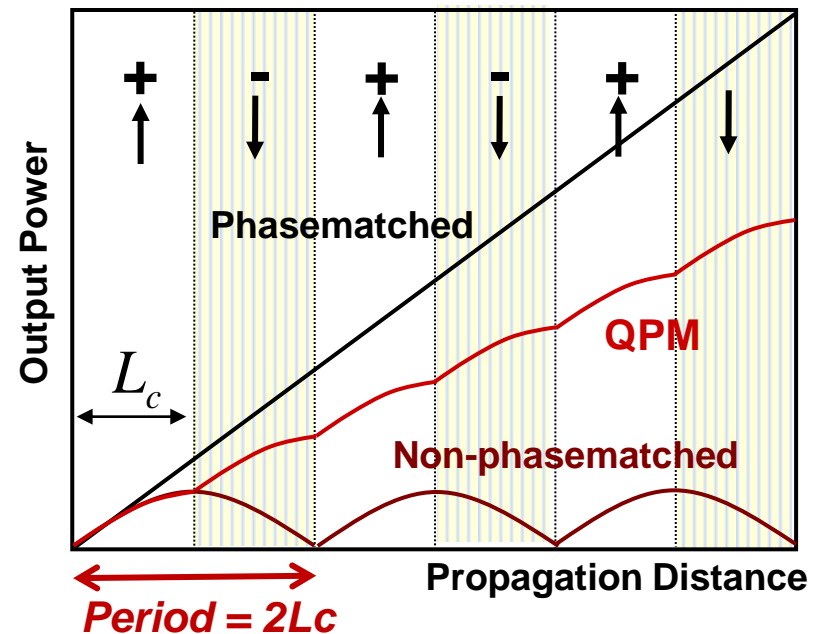
Reverse sign of nonlinear coefficient every coherence length: rephase waves

$$\vec{k}_p = \vec{k}_s + \vec{k}_i + \vec{K}_g$$

$$\vec{K}_g = 2\pi / 2L_c = \Delta k$$



In ferroelectrics (e.g., LiNbO<sub>3</sub> or KTP), reversed permanent electric dipole moment is obtained by applying high voltage of electric field – so called "periodic poling"



# Third order nonlinear effects

$$E_n = A_n e^{i\omega_n t} e^{-i\beta_n z}$$

$$P_{\omega_n}^{NL} = \chi^{(3)} A_k A_L A_m^* e^{i(\omega_k + \omega_L - \omega_m)t} e^{-i(\beta_k + \beta_L - \beta_m)z} \Rightarrow A_n e^{i\omega_n t} e^{-i\beta_n z}$$

**ENERGY CONSERVATION:**  $\omega_k + \omega_L - \omega_m = \omega_n$

**MOMENTUM CONSERVATION:**  $\beta_k + \beta_L - \beta_m \cong \beta_n$  — PHASE MATCHING

Self-phase  
matched {

$$\beta_k + \beta_k - \beta_k = \beta_k$$

SPM — self-phase modulation

$$\beta_k + \beta_L - \beta_L = \beta_k$$

XPM — cross-phase modulation

SRS — stimulated Raman scattering

SBS — stimulated Brillouin scattering

$$\beta_k + \beta_L - \beta_m \cong \beta_n$$

FWM — four-wave mixing  
(with small frequency shift)

Requires  
phasematching

**Phase-matched effects build up**





# Self-Phase Modulation - SPM

The incident optical field:  $\tilde{E}(t) = E(\omega)e^{-i\omega t} + \text{c.c.}$

Third order nonlinear polarization:

$$P^{(3)}(\omega) = 3\chi^{(3)}(\omega = \omega + \omega - \omega) |E(\omega)|^2 E(\omega)$$

The total polarization can be written as:

$$P^{\text{TOT}}(\omega) = \chi^{(1)} E(\omega) + 3\chi^{(3)}(\omega = \omega + \omega - \omega) |E(\omega)|^2 E(\omega)$$

One can define an effective susceptibility:  $\chi_{\text{eff}} = \chi^{(1)} + 4\pi |E(\omega)|^2 \chi^{(3)}$

The refractive index can be defined as usual:  $n^2 = 1 + 4\pi\chi_{\text{eff}}$

Define:  $n = n_0 + n_2 I$  with:  $n_2 = \frac{12\pi^2}{n_0^2} \chi^3$   $I = \frac{n_0 c}{2\pi} |E(\omega)|^2$

**Intensity dependent refractive index**

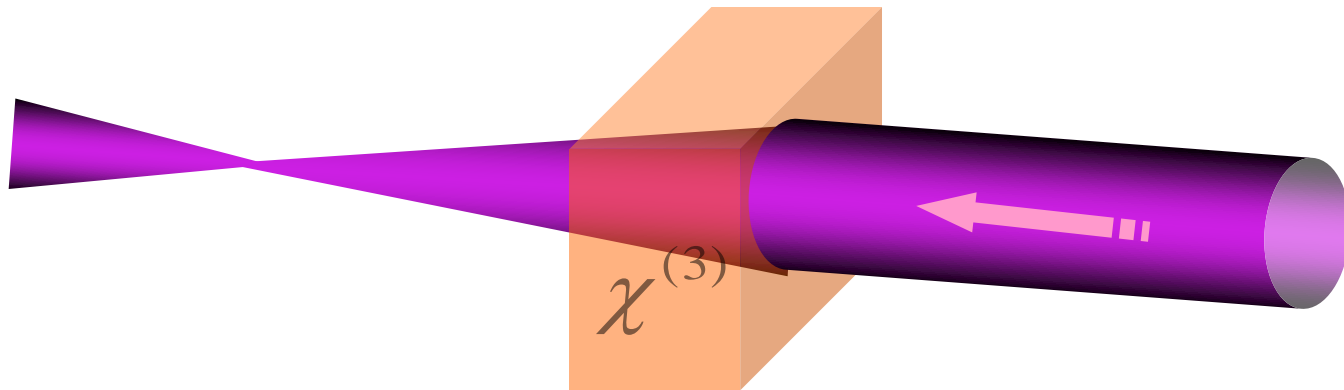
**Optical Kerr effect**



# Self focusing due to SPM

Optical Kerr effect:  $n = n_0 + n_2 I$

The laser beam has Gaussian intensity profile. It can induce a Gaussian refractive index profile inside the NLO sample – a “**Kerr lens**” !

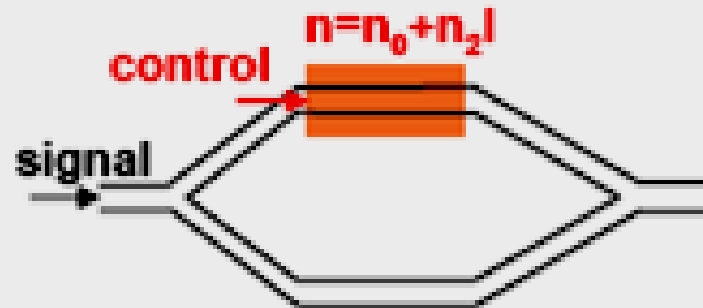


**Self-focusing** of Gaussian beam in **Kerr medium**

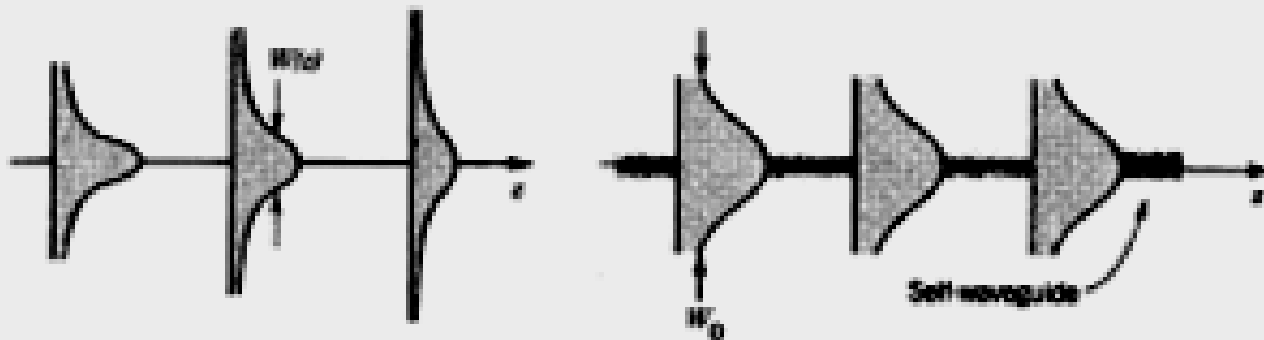
**Utilized e.g. for passive mode-locking**

# Kerr effect - examples of other applications

- All-optical switching:



- Spatial solitons: Self-focusing balances diffraction



# Spectral pulse broadening due to SPM

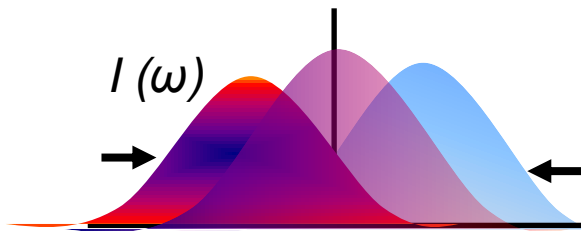
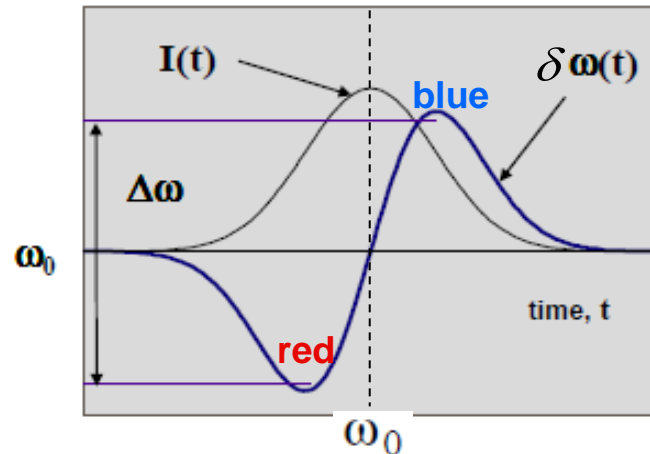
$$E_{sig}(z, t) = E_{sig}(0, t) \exp[i n k z] = E_{sig}(0, t) \exp\{i[n_0 + n_2 I(t)]k z\}$$

$$\phi(z, t) = n_2 k z I(t)$$

The phase is modulated according to the pulse **time envelope**  $I(t)$ , and increases with the **propagation distance**  $z$

Instantaneous frequency shift:

$$\delta\omega = \omega(t) - \omega_0 = \frac{d\phi}{dt} = -n_2 k z \frac{dI}{dt}$$



**Spectral broadening**  
**Generation of new frequencies**

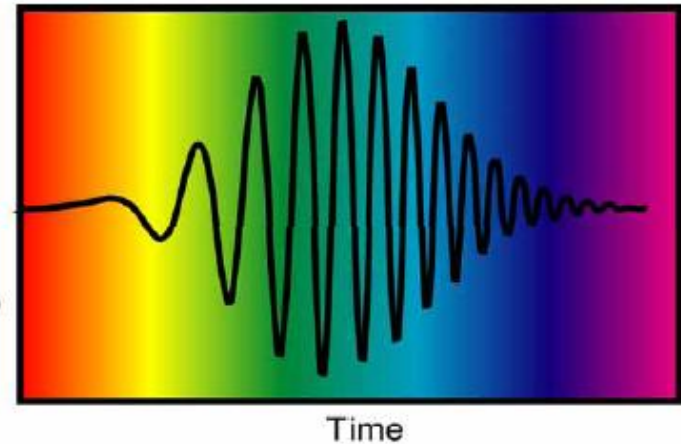
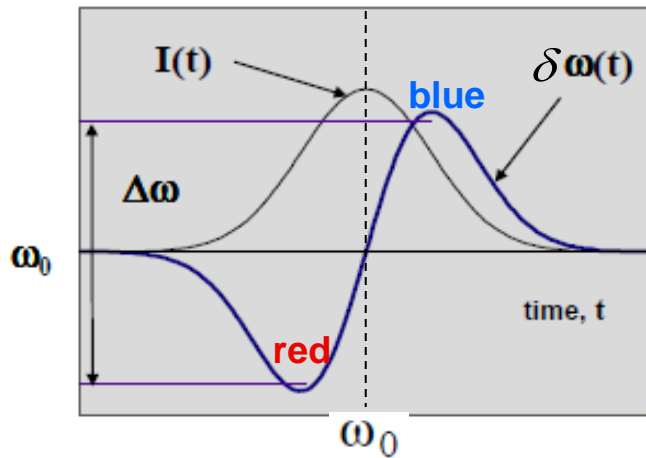
$$\Delta\omega = 2n_2 k z \left[ \frac{dI}{dt} \right]_{\max}$$

**In the absence of GVD only spectrum broadens - temporal shape is preserved !**

# SPM – frequency chirp

Pulse frequency varies in time - the leading edge is shifted to lower frequencies (**red shift**), the trailing edge to the higher ones (**blue shift**)

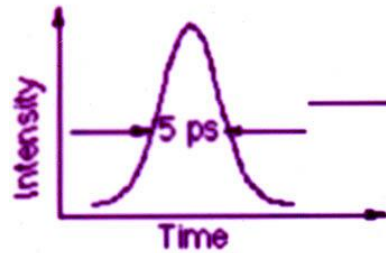
In analogy to bird sounds the pulse is called **”chirped”**



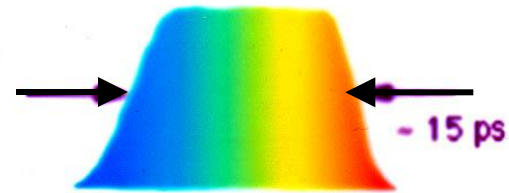
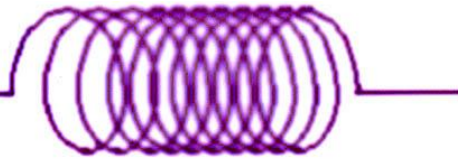
Frequency chirp:

$$C = \frac{d\omega}{dt} = \frac{d^2\phi}{dt^2} = -n_2 k_z \frac{d^2 I}{dt^2}$$

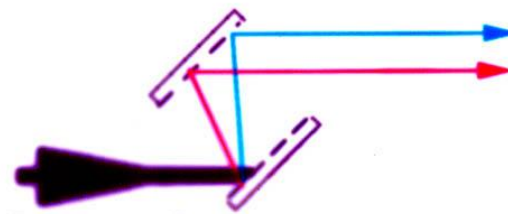
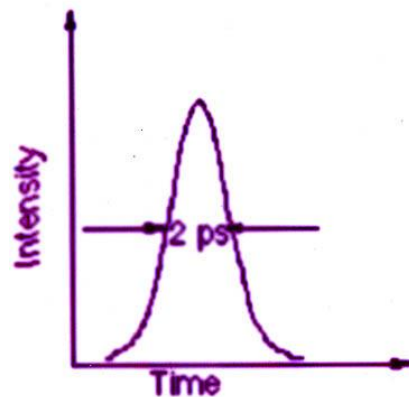
# SPM and GVD induced frequency chirp



Transform limited laser pulse

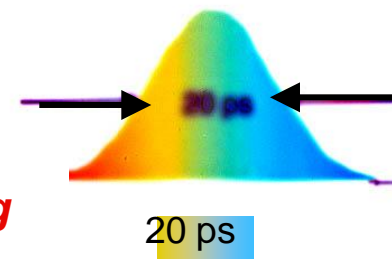


Frequency chirp generated by SPM in optical fiber  
**With normal GVD - enhanced pulse broadening**



Temporal broadening but **no spectral broadening**

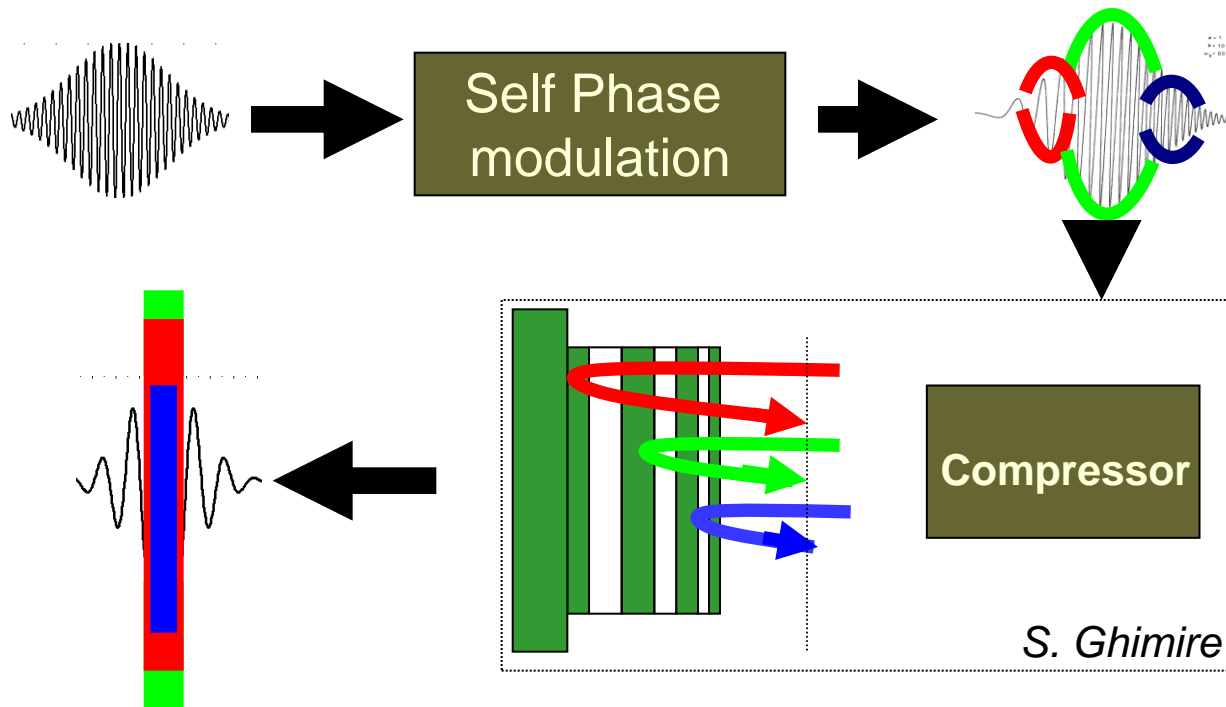
Frequency chirp generated by a grating or prism pair



# Pulse compression by SPM+GVD

With normal GVD – enhanced pulse broadening

With anomalous GVD – **pulse compression!**

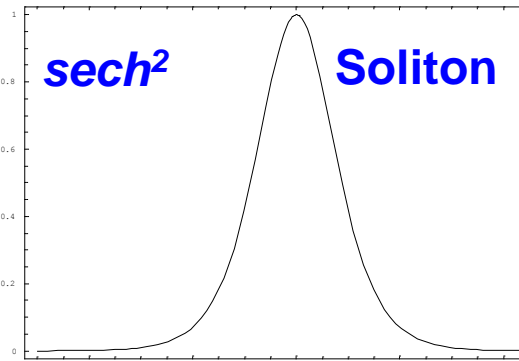


Note: at a given spectral width the shortest possible pulse is unchirped (transform limited)

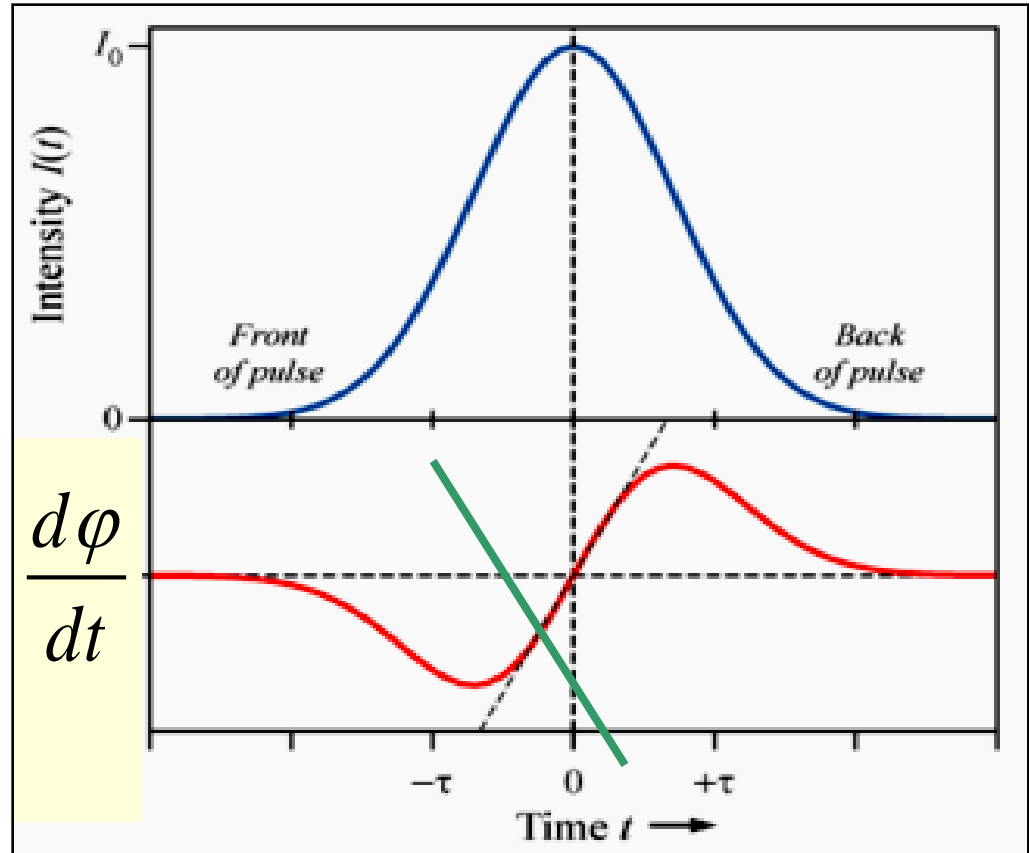
Chirped pulses have a "compression potential"

# Optical soliton

Perfect balance between  
**GVD** and **SPM**



Transform-limited (no chirp)  
In lossless media preserves  
its shape !



**Negative (anomalous) dispersion needed to form solitons**



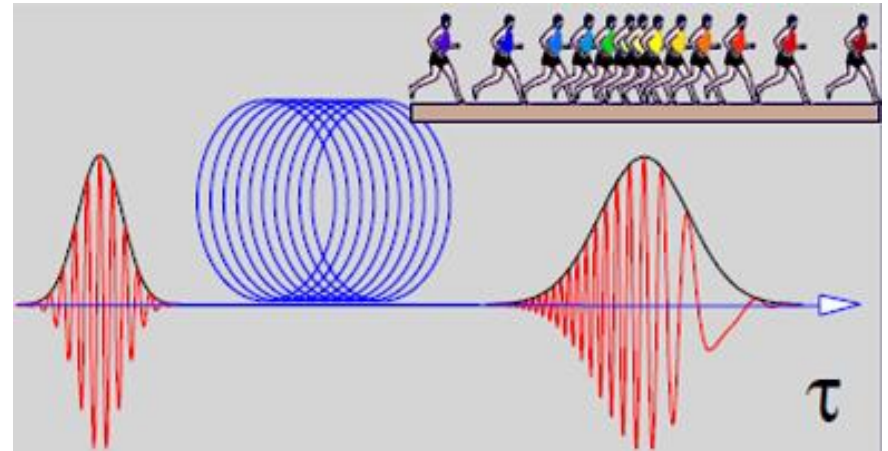
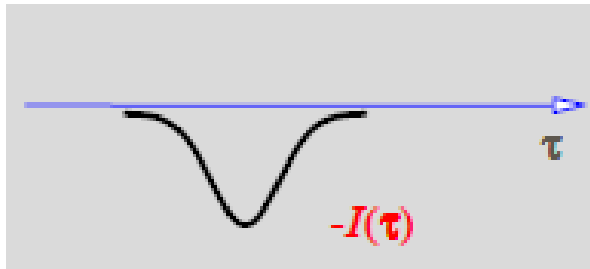
# Soliton – NL Schrödinger Equation

GVD  $\longrightarrow$

$$i \frac{\partial \tilde{E}}{\partial z'} = -\frac{\beta_2}{2} \frac{\partial^2 \tilde{E}}{\partial \tau^2} + \Gamma |\tilde{E}|^2 \tilde{E}$$

SPM

In anomalous dispersion regime  
pulse forms a bounding potential



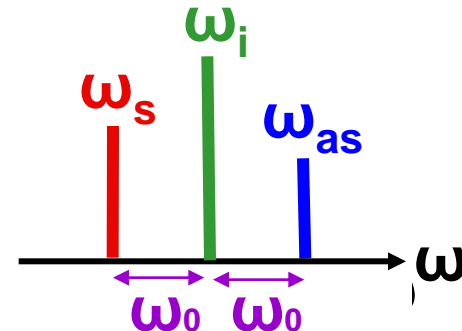
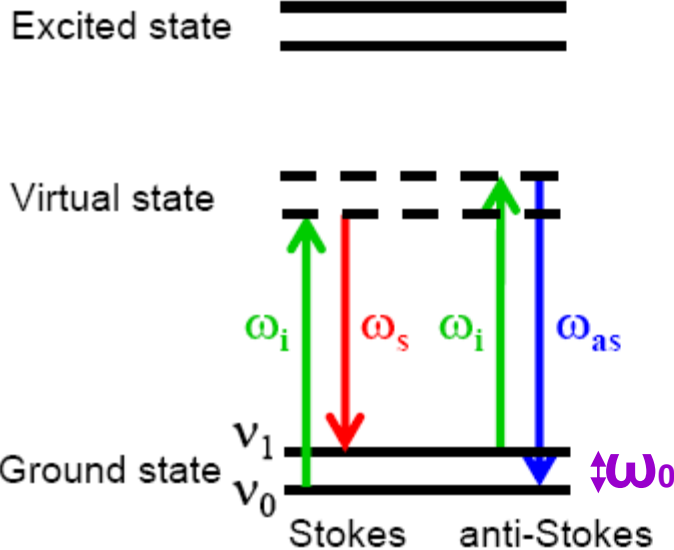
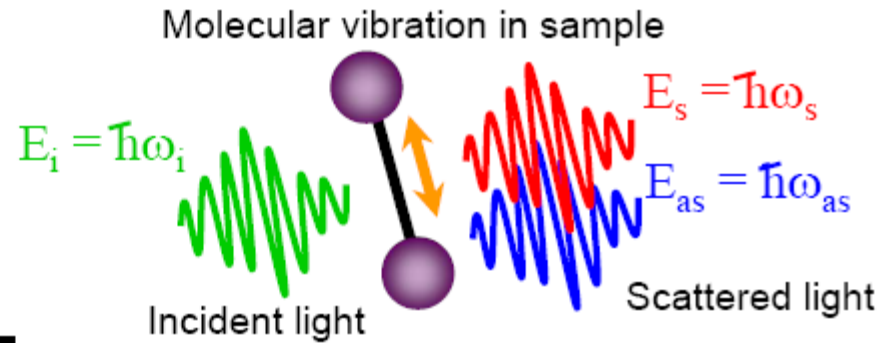
**Pulse is trapped in time (shape does not change)**

# Raman scattering

Interaction of photons and molecular vibrations (optical phonons)



C.V. Raman  
1930 Nobel Prize



+ Boltzmann distribution broadening  
 $\omega_{as} - \omega_i = \omega_i - \omega_s = \omega_0$



# Stimulated Raman scattering

New phenomenon observed in 1962 by **Gisela Eckhardt et al:**  
for intense pump the Stokes wave rapidly grows

Spontaneous Raman Scattering provides a weak signal that is amplified by the pump

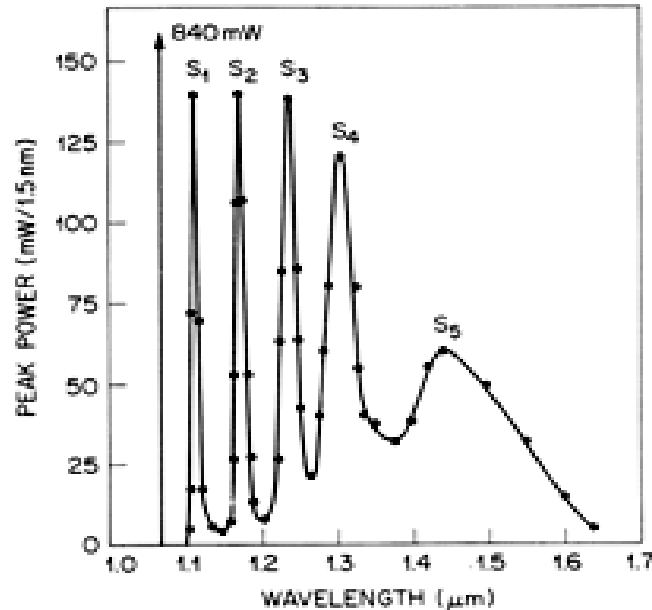


Stimulated Raman Scattering  
**SRS**

When the Stokes power becomes large enough it can act as a pump to the next order Stokes



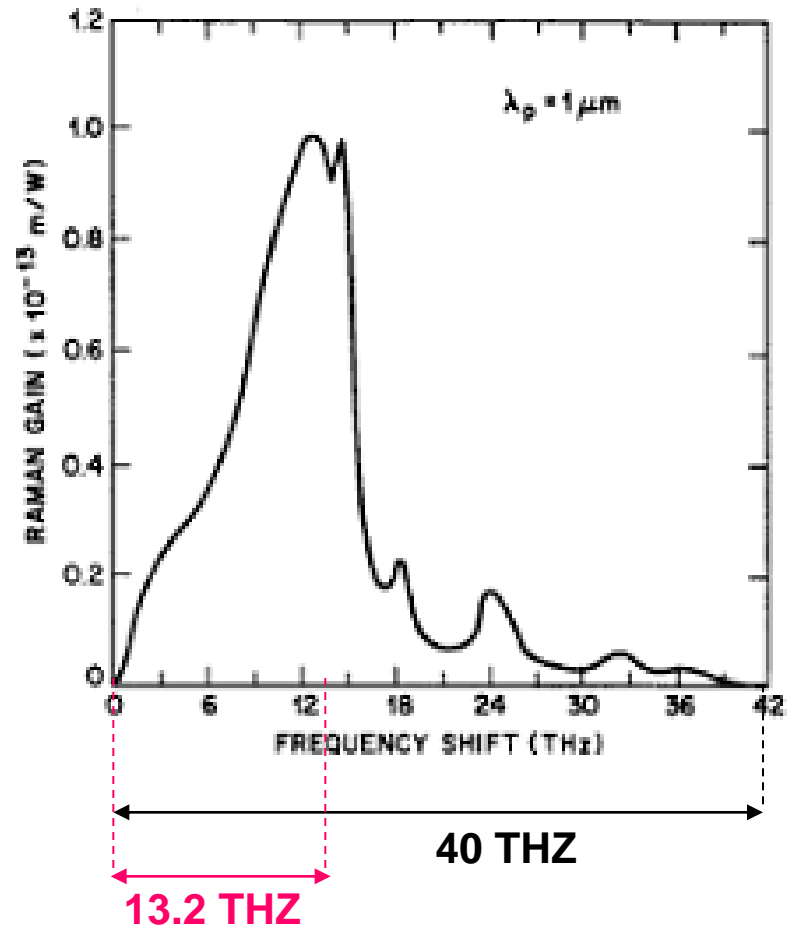
**SRS** is utilized for single pass or cascaded **amplifiers** and **lasers**



# Raman gain in optical fibers

Raman amplification in an optical fiber was first observed and measured in 1973 by **Stolen and Ippen**

- ✘ Raman gain peak is shifted from the pump by resonance frequency of molecular vibrations
- ✘ Gain spectrum in optical fibers is broad and smooth due to amorphous nature of glass
- ✘ For a given fiber the normalized gain profile does not depend on pump wavelength (in contrast to EDFA)



# Single-pass Raman amplifier

