



# Quantum theory of light-matter interaction: Fundamentals

## Lecture 4 Three- and four-wave mixing

Attila Czirják

University of Szeged, Dept. of Theoretical Physics, 2014



„Ágazati felkészítés a hazai ELI projekttel  
összefüggő képzési és K+F feladatokra ”

TÁMOP-4.1.1.C-12/1/KONV-2012-0005 projekt

SZÉCHENYI 2020



MAGYARORSZÁG  
KORMÁNYA

Európai Unió  
Európai Strukturális  
és Beruházási Alapok



BEFEKTETÉS A JÖVŐBE

# Outline/Contents

- 1 Overview
- 2 Light propagation in a nonlinear medium: General method
- 3 Three-wave mixing
  - Couplings due to a second-order nonlinear medium
  - Frequency addition
  - Phase matching
  - Coupled dynamics of three-wave mixing
  - Parametric amplification
  - Frequency doubling with pump depletion
- 4 Four-wave mixing in a nutshell
- 5 Summary

# Overview

- Three- and four-wave mixing denote a set of methods and techniques that utilize nonlinear optical media to create and control light beams by other light (usually strong laser) beams.
- Three-wave mixing: involves 3 light beams, uses second order non-linearity.
- Four-wave mixing: involves 4 light beams, uses third order non-linearity.
- These methods are of fundamental importance in modern laser science.
- Common terms: frequency doubling, sum frequency generation, optical rectification, parametric amplification, phase matching, OPO, OPA, etc.

## Reminder

Polarization and nonlinear susceptibility in a nonlinear medium:

$$P_i = \epsilon_0 \sum_j \chi_{ij}^{(1)} E_j + \epsilon_0 \sum_{j,k} \chi_{ijk}^{(2)} E_j E_k + \epsilon_0 \sum_{j,k,l} \chi_{ijkl}^{(3)} E_j E_k E_l + \dots,$$

Wave equation with the linear and nonlinear polarization as source:

$$\Delta \mathbf{E}(\mathbf{r}, t) - \frac{1}{c^2} \partial_t^2 \mathbf{E}(\mathbf{r}, t) = \frac{1}{c^2 \epsilon_0} \partial_t^2 (\mathbf{P}_L(\mathbf{r}, t) + \mathbf{P}_{NL}(\mathbf{r}, t))$$

Analogous wave equation for the complex fields (or "analytic signals", or "positive frequency parts")  $\mathbf{E}^{(+)}(\mathbf{r}, t)$  and  $\mathbf{P}^{(+)}(\mathbf{r}, t)$ .

Definition of the complex electric field:

$$\mathbf{E}^{(+)}(\mathbf{r}, t) = \frac{1}{\sqrt{2\pi}} \int_0^\infty d\omega e^{-i\omega t} \frac{1}{\sqrt{2\pi}} \int_{-\infty}^\infty dt' e^{i\omega t'} \mathbf{E}(\mathbf{r}, t'),$$

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}^{(+)}(\mathbf{r}, t) + \mathbf{E}^{(-)}(\mathbf{r}, t), \quad \mathbf{E}^{(-)}(\mathbf{r}, t) = \left[ \mathbf{E}^{(+)}(\mathbf{r}, t) \right]^*$$

## General method

To solve the wave equation for the complex electric field:

$$\Delta \mathbf{E}^{(+)}(\mathbf{r}, t) - \frac{1}{c^2} \partial_t^2 \mathbf{E}^{(+)}(\mathbf{r}, t) = \frac{1}{c^2 \epsilon_0} \partial_t^2 \left( \mathbf{P}_L^{(+)}(\mathbf{r}, t) + \mathbf{P}_{NL}^{(+)}(\mathbf{r}, t) \right)$$

expand the complex electric field over monochromatic plane waves which already account for the linear polarization of the medium:

$$\mathbf{E}^{(+)}(\mathbf{r}, t) = \sum_{\ell} \epsilon_{\ell} \mathcal{E}_{\ell}(\mathbf{r}) \exp [i(\mathbf{k}_{\ell} \mathbf{r} - \omega_{\ell} t)]$$

with wave vector  $\mathbf{k}_{\ell}^2 = n_{\ell}^2 \omega_{\ell}^2 / c^2$ , where  $n_{\ell}^2 = 1 + \chi^{(1)}(\omega_{\ell})$  is the linear refractive index, and  $\epsilon_{\ell} \cdot \mathbf{k}_{\ell} = 0$ .

Note that the amplitudes  $\mathcal{E}_{\ell}(\mathbf{r})$  still have spatial dependence. To simplify, we shall only consider the case in which all the waves propagate in the same direction, along the  $z$ -axis.

## General method

Decompose the nonlinear polarization into the same monochromatic components:

$$\mathbf{P}_{\text{NL}}^{(+)}(z, t) = \sum_{\ell} \epsilon_{\ell} \mathcal{P}_{\text{NL}}^{\omega_{\ell}}(z) \exp[-i\omega_{\ell}t]$$

Substitute these sums into the wave equation and apply the slowly varying amplitude approximation. Equating terms of the same frequency we obtain a set of equations like

$$\frac{\partial \mathcal{E}_{\ell}(z)}{\partial z} \exp[ik_{\ell}z] = \frac{i\omega_{\ell}}{2\epsilon_0 n_{\ell} c} \mathcal{P}_{\text{NL}}^{\omega_{\ell}}(z)$$

Then express  $\mathcal{P}_{\text{NL}}^{\omega_{\ell}}(z)$  in terms of the amplitudes  $\mathcal{E}_m(z)$  using the nonlinear susceptibilities, to obtain a system of coupled first-order differential equations that can be solved to yield the amplitudes  $\mathcal{E}_{\ell}(z)$  of the different waves.

## General method

Self-consistency of the equations, Born approximations:

The incoming light wave generates a polarization density in the nonlinear medium, which generates a new light wave, which again, generates another polarization density in the nonlinear medium, etc. The previous equations have to be solved such that self-consistency is achieved. This can be done usually by successive approximation.

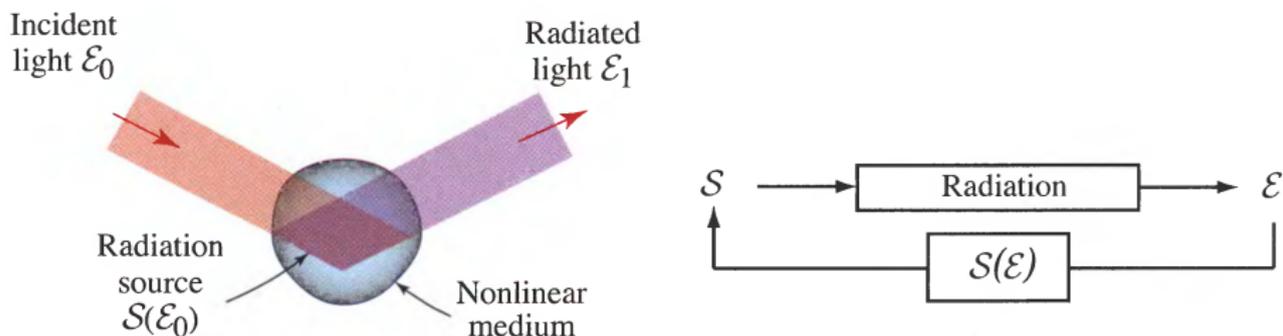


Figure: From [Saleh and Teich]

## What is three-wave mixing?

Consider two intense waves, called pump waves, propagating in the  $z$  direction in a second-order nonlinear medium. The frequency dependent second-order susceptibility gives rise to the following terms in the nonlinear polarization, associated with the physical processes of frequency doubling, addition or difference, and optical rectification (Figure from [Saleh and Teich]):

$\chi^{(2)}(-2\omega_1; \omega_1, \omega_1)$	$(\mathcal{E}_1^2 e^{-2i\omega_1 t} + \text{c.c.})$	Frequency doubling
$\chi^{(2)}(-2\omega_2; \omega_2, \omega_2)$	$(\mathcal{E}_2^2 e^{-2i\omega_2 t} + \text{c.c.})$	Frequency doubling
$2\chi^{(2)}(0; \omega_1, -\omega_1)$	$ \mathcal{E}_1 ^2$	Optical rectification
$2\chi^{(2)}(0; \omega_2, -\omega_2)$	$ \mathcal{E}_2 ^2$	Optical rectification
$2\chi^{(2)}(-\omega_1 - \omega_2; \omega_1, \omega_2)$	$(\mathcal{E}_1 \mathcal{E}_2 e^{-i(\omega_1 + \omega_2)t} + \text{c.c.})$	Frequency addition
$2\chi^{(2)}(-\omega_1 + \omega_2; \omega_1, -\omega_2)$	$(\mathcal{E}_1 \mathcal{E}_2^* e^{-i(\omega_1 - \omega_2)t} + \text{c.c.})$	Frequency difference

We shall consider frequency addition in detail.

# Frequency addition

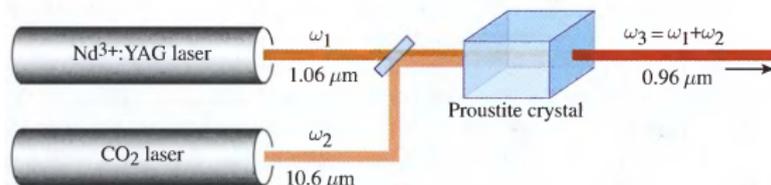


Figure: From [Saleh and Teich]

Two pump waves,  $\mathcal{E}_1(z) \exp [i(k_1z - \omega_1t)]$  and  $\mathcal{E}_2(z) \exp [i(k_2z - \omega_2t)]$ , propagating in a second-order nonlinear medium, generate the following polarization amplitude at the sum frequency  $\omega_3 = \omega_1 + \omega_2$

$$\mathcal{P}_{\text{NL}}^{\omega_3}(z) = 2 \epsilon_0 \chi^{(2)}(-\omega_3; \omega_1, \omega_2) \mathcal{E}_1(z) \mathcal{E}_2(z) \exp [i(k_1 + k_2)z]$$

This nonlinear polarization creates a field  $\mathcal{E}_3(z)$  at frequency  $\omega_3$ , which evolves according to

$$\frac{\partial \mathcal{E}_3(z)}{\partial z} \exp [ik_3z] = \frac{i\omega_3}{2\epsilon_0 n_3 c} \mathcal{P}_{\text{NL}}^{\omega_3}(z)$$

## Frequency addition

Substituting the polarization, we get the following first-order ODE that governs the spatial variation of the sum-frequency wave amplitude:

$$\frac{\partial \mathcal{E}_3(z)}{\partial z} = i \frac{\omega_3 \chi^{(2)}}{n_3 c} \mathcal{E}_1(z) \mathcal{E}_2(z) \exp [i \Delta k z]$$

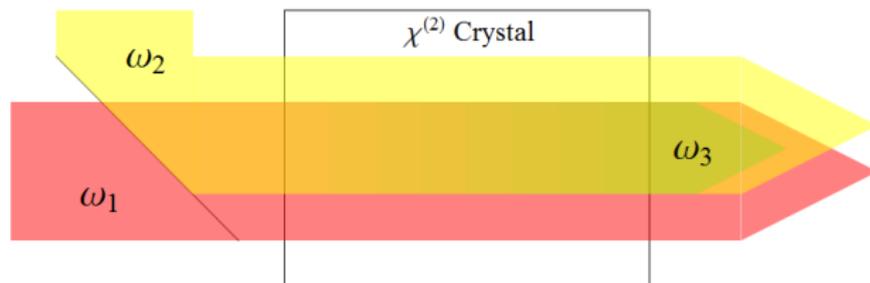
where  $\Delta k = k_1 + k_2 - k_3$  is the wavenumber mismatch. The usual initial condition is  $\mathcal{E}_3(0) = 0$ .

To a first approximation, we assume that  $\mathcal{E}_1$  and  $\mathcal{E}_2$  are constant over the length  $L$  of the nonlinear material. If this is also thin, i.e.  $0 < z < L \ll 1/\Delta k$ , then the above ODE can be simplified to

$$\frac{\partial \mathcal{E}_3(z)}{\partial z} = i \frac{\omega_3 \chi^{(2)}}{n_3 c} \mathcal{E}_1 \mathcal{E}_2$$

which means that  $\mathcal{E}_3(z)$  increases linearly with  $z$ .

# Frequency addition



Therefore the intensity of the generated beam  $I_3$  scales with the square of the length of the crystal:

$$I_3 = CL^2 I_1 I_2$$

with  $C = (\omega_3 \chi^{(2)})^2 / (2\epsilon_0 c^3 n_1 n_2 n_3)$ .

Orders of magnitude: KTP crystal:  $\chi^{(2)} = 5 \cdot 10^{-12} \text{ m/V}$ ,  $L = 1 \text{ cm}$ , pump waves of 1 W power around  $\lambda = 1 \mu\text{m}$  wavelength, focused on an optimal spot of  $10^{-2} \text{ mm}^2$ , the sum-frequency beam has power 0.2 mW.

## Phase matching

If we can achieve  $\Delta k = 0$ , called perfect phase matching, then the previous results hold for any  $z$  as long as the pump beams can be considered constant. However, because of dispersion,  $k_3 = k_1 + k_2$  is usually not fulfilled in a medium.

If  $\Delta k \neq 0$ , then the spatial dependence of the sum-frequency beam:

$$\mathcal{E}_3(z) = \frac{\omega_3 \chi^{(2)}}{n_3 c} \mathcal{E}_1 \mathcal{E}_2 \frac{\exp [i \Delta k z] - 1}{\Delta k}$$

i.e. its intensity leaving the medium of length  $L$  is proportional to

$$|\mathcal{E}_3(L)|^2 = \left( \frac{2 \omega_3 \chi^{(2)}}{n_3 c} \right)^2 |\mathcal{E}_1|^2 |\mathcal{E}_2|^2 \left( \frac{\sin [\Delta k L / 2]}{\Delta k} \right)^2$$

which is maximal if the length of the medium is set as  $L_{\text{opt}} = \pi / |\Delta k|$ . The resulting optimal intensity is still less by a factor of  $4/\pi^2$  than in the case of perfect phase matching.

# Phase matching

How can perfect phase matching be achieved?

In a dispersive material  $k(\omega) = n(\omega)\omega/c$ , thus for collinear propagation  $k_3 = k_1 + k_2$  can be achieved using a birefringent material, where the refractive index depends on the polarization.

For example, in type-I case the waves 1 and 2 have the same polarization, and the direction of propagation through a uniaxial crystal can be chosen such, that perfect phase matching is fulfilled.

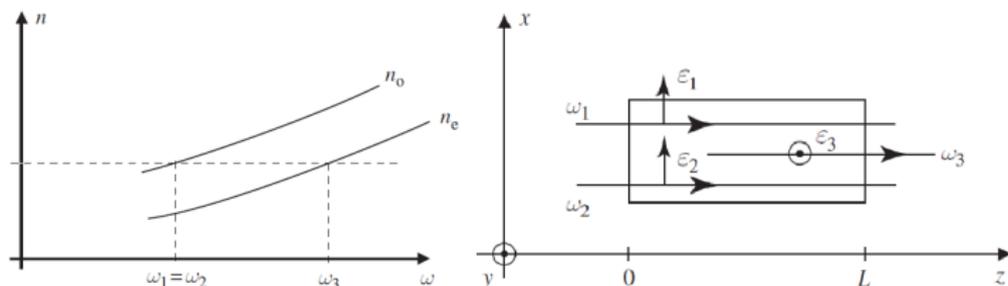


Figure: From [Gilbert, Aspect, and Fabre]

# Phase matching

If we drop the collinearity requirement, perfect phase matching requires  $\mathbf{k}_3 = \mathbf{k}_1 + \mathbf{k}_2$  which is more easily achieved:



Figure: From [Gilbert, Aspect, and Fabre]

Disadvantage: poor beam overlap limits the interaction length.

Quasi-phase matching:

For long interaction lengths (guided propagation), phase matching for the whole length is a problem. Quasi solution: several pieces of the medium of length  $\pi/|\Delta k|$  set end-to-end, with alternating signs of  $\chi^{(2)}$ , this compensates phase mismatch. PPLN: periodically poled lithium niobate

# Coupled dynamics of three-wave mixing

Consider the second-order nonlinear material, three collinear coupled waves, and perfect phase matching:

$$k_3 = k_1 + k_2, \quad \omega_3 = \omega_1 + \omega_2$$

Let us now relax the assumption of constant pumps, i.e. we go beyond the Born approximation: all the amplitudes are subject to spatial variation during propagation.

Clearly, if beams 1 and 2 generated already an up-converted beam 3, this can combine with beam 1 or 2 to generate two other beams by down-conversion. What about the many other possibilities, like  $\omega_4 = \omega_3 + \omega_2$ ,  $\omega_5 = 2\omega_3$ , etc.?

They can not be phase matched simultaneously with the phase matching for 1, 2 and 3, thus they are suppressed by these, they can be neglected in the modelling.

## Coupled dynamics of three-wave mixing

The closed system of coupled ODEs that govern the 3 efficient processes of collinear phase matched three-wave mixing:

$$\begin{aligned}\frac{\partial \mathcal{E}_1(z)}{\partial z} &= i \frac{\omega_1 \chi^{(2)}}{n_1 c} \mathcal{E}_3(z) \mathcal{E}_2^*(z), \\ \frac{\partial \mathcal{E}_2(z)}{\partial z} &= i \frac{\omega_2 \chi^{(2)}}{n_2 c} \mathcal{E}_3(z) \mathcal{E}_1^*(z), \\ \frac{\partial \mathcal{E}_3(z)}{\partial z} &= i \frac{\omega_3 \chi^{(2)}}{n_3 c} \mathcal{E}_1(z) \mathcal{E}_2(z),\end{aligned}\quad (\text{coupled})$$

they have a complicated general solution in terms of elliptic functions. Instead, look for conserved quantities: energy is a good guess, the Poynting vector's magnitude is intensity,

$$\Pi = |\mathbf{\Pi}| = |\mu_0^{-1} \mathbf{E} \times \mathbf{B}| = 2n\epsilon_0 c \mathcal{E}^* \mathcal{E}$$

## Coupled dynamics of three-wave mixing

Calculate the spatial derivative of  $\Pi$  for all the 3 waves and add them:

$$\frac{\partial}{\partial z} (\Pi_1(z) + \Pi_2(z) + \Pi_3(z)) = i(\omega_1 + \omega_2 - \omega_3)2\epsilon_0\chi^{(2)}(\mathcal{E}_1^*\mathcal{E}_2^*\mathcal{E}_3 - \text{c.c.}) = 0$$

which expresses energy conservation. No energy is transferred to the nonlinear medium, which serves only to facilitate the coupling process.

The second invariant of the coupled ODEs comes also from the spatial derivatives of the intensity, since

$$\frac{1}{\omega_1} \frac{\partial \Pi_1}{\partial z} = \frac{1}{\omega_2} \frac{\partial \Pi_2}{\partial z} = -\frac{1}{\omega_3} \frac{\partial \Pi_3}{\partial z},$$

therefore

$$\frac{\partial}{\partial z} \left( \frac{\Pi_1}{\omega_1} - \frac{\Pi_2}{\omega_2} \right) = 0.$$

This is the Manley-Rowe relation, which has a simple interpretation in the quantum theoretical description, in terms of photons.

## Optical Parametric Amplification

A very important special case of the previous description of coupled dynamics of three-wave mixing is when a strong pump beam of frequency  $\omega_3$  and a weak signal of frequency  $\omega_1$  (to be amplified) enters the nonlinear medium with perfect phase matching for  $\omega_2 = \omega_3 - \omega_1$ . We make the constant amplitude approximation for the pump beam, which leads to the following ODEs for beams 1 and 2:

$$\frac{\partial^2 \mathcal{E}_1(z)}{\partial z^2} = \gamma^2 \mathcal{E}_1(z), \quad \frac{\partial^2 \mathcal{E}_2(z)}{\partial z^2} = \gamma^2 \mathcal{E}_2(z), \quad \gamma^2 = \left( \frac{\chi^{(2)} |\mathcal{E}_3|}{c} \right)^2 \frac{\omega_1 \omega_2}{n_1 n_2}$$

still coupled by the following initial conditions:  $\mathcal{E}_1(z=0) = \mathcal{E}_1(0)$  and  $\mathcal{E}_2(z=0) = 0$ . The solution is

$$\mathcal{E}_1(z) = \mathcal{E}_1(0) \cosh(\gamma z), \quad \mathcal{E}_2(z) = i \sqrt{\frac{\omega_2 n_1}{\omega_1 n_2}} \frac{\mathcal{E}_3}{|\mathcal{E}_3|} \mathcal{E}_1^*(0) \sinh(\gamma z)$$

i.e. the signal is amplified ca. exponentially, and an amplified complementary wave appears at  $\omega_2$ , called the idler.

## Frequency doubling with pump depletion

Frequency doubling is a degenerate case of sum-frequency generation: only one incident beam of frequency  $\omega_1$ , and one generated wave of frequency  $\omega_3 = 2\omega_1$ . The nonlinear polarization is then proportional to the square of the incident field, which makes a change of a factor of 2 in the coupled equations for the two fields. Assuming phase matching  $k_3 = 2k_1$ ,

$$\frac{\partial \mathcal{E}_1(z)}{\partial z} = i \frac{\omega_1 \chi^{(2)}}{n_1 c} \mathcal{E}_3(z) \mathcal{E}_1^*(z), \quad \frac{\partial \mathcal{E}_3(z)}{\partial z} = i \frac{\omega_3 \chi^{(2)}}{2n_3 c} \mathcal{E}_1^2(z),$$

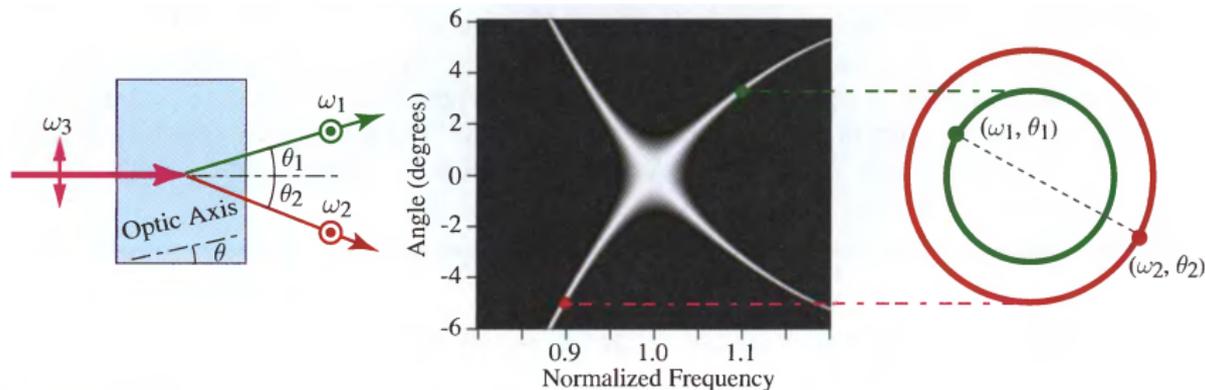
yield the solution for the initial conditions  $\mathcal{E}_1(z=0) = E_1$  (assumed real), and  $\mathcal{E}_3(z=0) = 0$

$$\mathcal{E}_1(z) = \frac{E_1}{\cosh(\gamma' z)}, \quad \mathcal{E}_3(z) = i E_1 \tanh(\gamma' z), \quad \gamma' = \chi^{(2)} E_1 \frac{\omega_1}{n_1 c}.$$

# Frequency doubling with pump depletion

For  $z \ll 1/\gamma'$  the power of the second harmonic wave grows as the square of the pump power and as the square of the interaction length, like for constant pump. At  $\gamma'z \approx 1$  the process saturates and tends to  $|\mathcal{E}_3| = E_1$ , while the pump field tends to zero, which means total conversion of the pump into the second harmonic if the medium is thick enough. Even in reality, one can achieve extremely high conversion efficiencies using pulsed lasers with very high peak powers: frequency doubling of the Megajoule laser with an efficiency of around 80%.

# Parametric fluorescence



**Figure 21.2-13** Tuning curves for non-collinear Type-I o-o-e spontaneous parametric downconversion in a BBO crystal at an angle  $\theta = 33.53^\circ$  for a 351.5-nm pump (from an  $\text{Ar}^+$ -ion laser). Each point in the bright area of the middle picture represents the frequency  $\omega_1$  and angle  $\theta_1$  of a possible down-converted wave, and has a matching point at a complementary frequency  $\omega_2 = \omega_3 - \omega_1$  with angle  $\theta_2$ . Frequencies are normalized to the degenerate frequency  $\omega_o = \omega_3/2$ . For example, the two dots shown represent a pair of down-converted waves at frequencies  $0.9\omega_o$  and  $1.1\omega_o$ . Because of circular symmetry, each point is actually a ring of points all of the same frequency, but each point on a ring matches only one diametrically opposite point on the corresponding ring, as illustrated in the right graph.

Figure: From [Saleh and Teich]

# Optical parametric devices

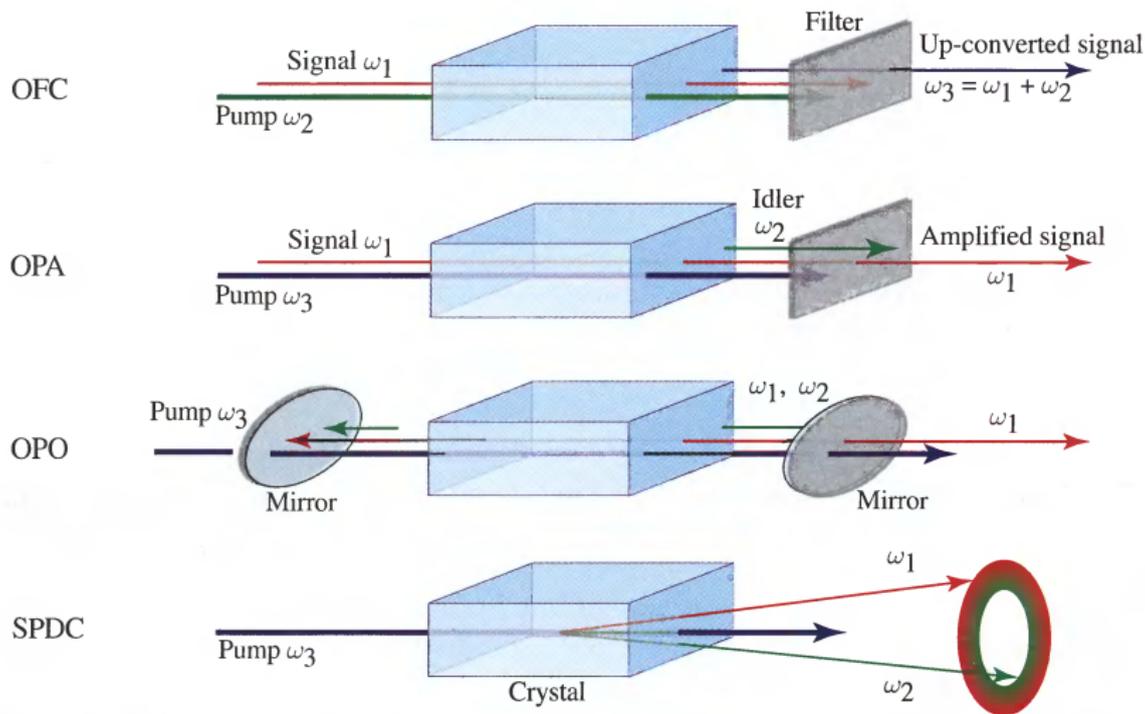


Figure: From [Saleh and Teich]

# ELI-ALPS laser system design

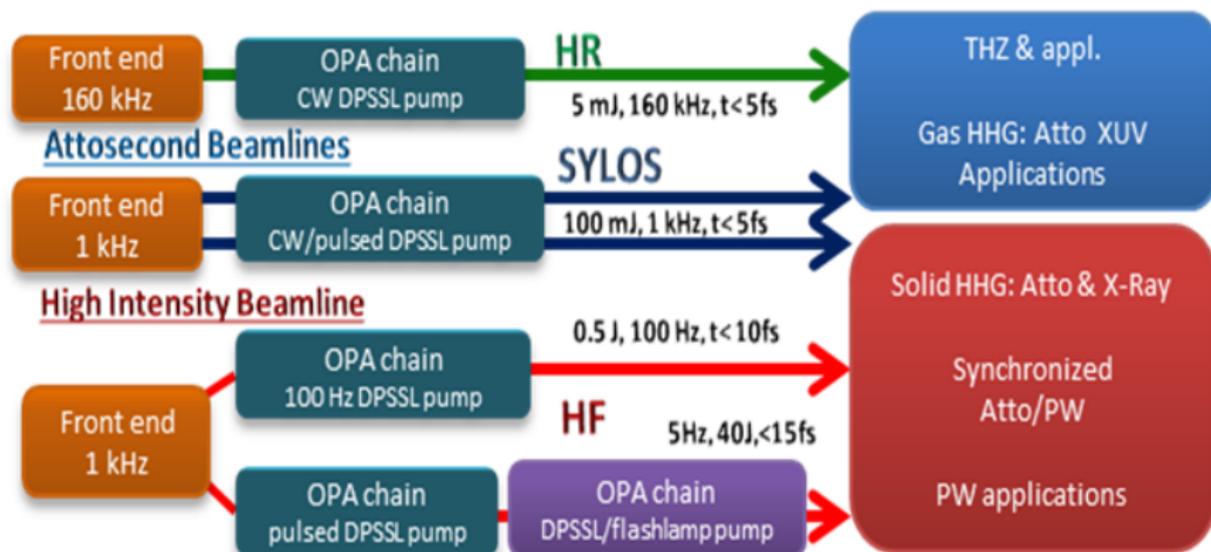


Figure: From [www.eli-alps.hu](http://www.eli-alps.hu)

# Third-order nonlinear response of two-level atoms

$$\mathbf{P}^{(+)}(\mathbf{r}, t) = \varepsilon_0 \chi \mathbf{E}^{(+)}(\mathbf{r}, t)$$

$$\chi = \chi' + i\chi''$$

$$\chi' = \frac{N}{V} \frac{d^2}{\varepsilon_0 \hbar} \frac{\omega_0 - \omega}{\frac{\Gamma_{\text{sp}}^2}{4} + \frac{\Omega_1^2}{2} + (\omega_0 - \omega)^2}$$

$$|\omega_0 - \omega| \gg \Gamma_{\text{sp}}$$

$$\chi' = \frac{N}{V} \frac{d^2}{\varepsilon_0 \hbar} \frac{\omega_0 - \omega}{\frac{\Omega_1^2}{2} + (\omega_0 - \omega)^2}$$

$$\chi' = \chi'_1 + \chi'_3 I + \dots$$

$$\chi'_1 = \frac{N}{V} \frac{d^2}{\varepsilon_0 \hbar (\omega_0 - \omega)}$$

$$\chi'_3 = -\frac{N}{V} \frac{d^4}{\varepsilon_0 \hbar^3 (\omega_0 - \omega)^3}$$

$$\mathbf{P}_L^{(+)} = \varepsilon_0 \chi'_1 \mathbf{E}^{(+)}$$

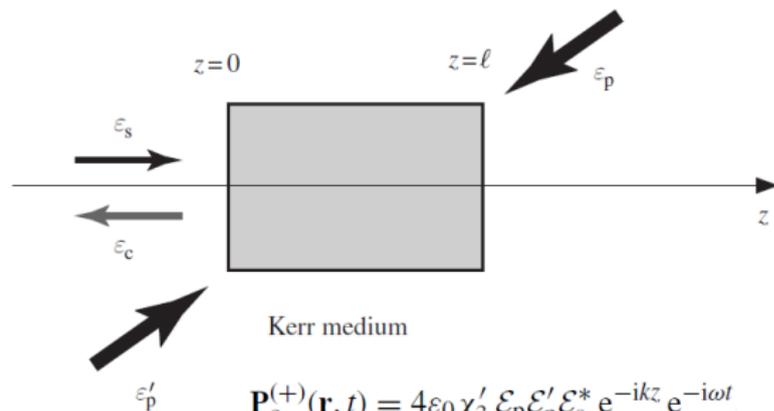
$$\mathbf{P}_{\text{NL}}^{(+)} = \varepsilon_0 \chi'_3 I \mathbf{E}^{(+)}$$

Figure: From [Gilbert, Aspect, and Fabre]

# Degenerate four-wave mixing

$$\mathbf{E}^{(+)}(\mathbf{r}, t) = \epsilon \left[ \mathcal{E}_p e^{i\mathbf{k}\cdot\mathbf{r}} + \mathcal{E}'_p e^{-i\mathbf{k}\cdot\mathbf{r}} + \mathcal{E}_s e^{ikz} \right] e^{-i\omega t}.$$

$$\mathbf{P}_{\text{NL}}^{(+)}(\mathbf{r}) = 2\epsilon_0 \chi'_3 |E^{(+)}(\mathbf{r}, t)|^2 E^{(+)}(\mathbf{r}, t).$$



$$\mathbf{P}_c^{(+)}(\mathbf{r}, t) = 4\epsilon_0 \chi'_3 \mathcal{E}_p \mathcal{E}'_p \mathcal{E}_s^* e^{-ikz} e^{-i\omega t}$$

$$\frac{d\mathcal{E}_c}{dz} = -i \frac{2\omega}{n_0 c} \chi'_3 \mathcal{E}_p \mathcal{E}'_p \mathcal{E}_s^*$$

Figure: From [Gilbert, Aspect, and Fabre]

# Phase conjugation

$$E_s^{(+)}(z, t) = \mathcal{E}_s(z) e^{-i\omega t} = A e^{i\varphi_s} e^{ikz} e^{-i\omega t}$$

$$E_c^{(+)}(z, t) = \mathcal{E}_c(z) e^{-i\omega t} = A' e^{-i\varphi_s} e^{-ikz} e^{-i\omega t}$$

$$E_c(z, t) = 2A' \cos(\omega t + kz + \varphi_z)$$

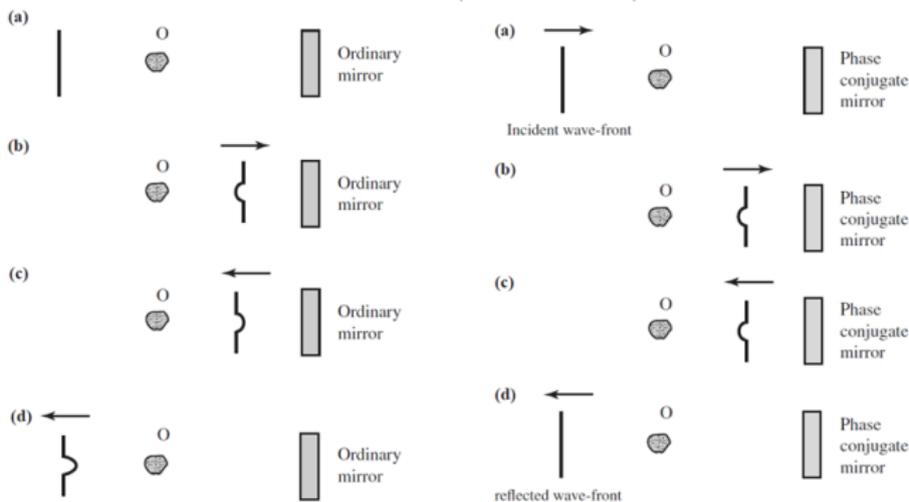


Figure: From [Gilbert, Aspect, and Fabre]

# Further reading

- References:

[Gilbert, Aspect, and Fabre]: G. Gilbert, A. Aspect, C. Fabre: Introduction to Quantum Optics, Cambridge University Press, 2010.

[Saleh and Teich]: B. E. A. Saleh and M. C. Teich: Fundamentals of Photonics, 2nd. ed., Wiley, 2007.

# Questions

- 1 Explain the self-consistency problem of light propagation in non-linear media.
- 2 How does the intensity of the sum-frequency beam scale with the length of the medium in the case of perfect phase matching and collinear propagation?
- 3 How does the intensity of the sum-frequency beam scale with the intensities of the pump beams?
- 4 Why is perfect phase matching in collinear propagation usually not possible in isotropic media?
- 5 Why is one of the electric field amplitudes conjugate in two of the ODEs governing the coupled field dynamics of three-wave mixing, but not in the third ODE?

# Questions

- 6 Explain why it is sufficient to have 3 ODEs to describe the coupled field dynamics of three-wave mixing? What about the processes like  $\omega_4 = \omega_3 + \omega_1$  ?
- 7 What are the invariants of the coupled dynamics of three-wave mixing?