Part A

Nonlinear Optics

An Introduction

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Text: **Photonics** Ralf Menzel, 2007

Chapter 4: NL Interactions of Light and Matter without Absorption

For additional reading:

1 Nonlinear Optics, R. W. Boyd, 3rd ed, 2007

2. Principles Of Nonlinear Optics, Y. R. Shen 2003

3. Introduction to Nonlinear Optics, G. New, 2011

4. Handbook of Optics, 3rd ed 2010, Chap. 10

5. Fundamentals of Photonics, B.E. Salih and M.C. Tiech 2nd ed, 2007

6. Handbook of Lasers and Optics, F. Träger (Ed.), 2007

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Introduction to Nonlinear Optics

Important Note

Understanding linear interactions is essential.

See Chapter 3

Photonics

Photonics” is the science and technology of light, with an emphasis on applications.

It is considered as one of the key technologies of the 21st century.

Photonics Applications

Typical **applications** areas are:

- Communications: free-space optical communications, optical data storage and optical computing …

- Medicine dentistry, and life sciences, e.g., diagnostics and therapy in ophthalmology and cancer research; biology, biotechnology …

r Optic

Photonics Applications

-Sensing: e.g., fiber-optic sensors, high-speed cameras, industrial process control …

-Industry and manufacturing: e.g. laser material processing, semiconductor chip manufacturing, printing …

-Lighting: e.g., energy-efficient LED illumination.

-Defense and space technology: e.g. satellite surveillance systems, night vision, missile guidance, anti-missile systems …

Nonlinear Optics (NLO)

NLO is the area of optics that studies the interaction of light with matter in the regime where the response of the material system to the applied electromagnetic field is <u>nonlinear</u> in the amplitude of this field.

At low light intensities, the properties of materials remain independent of intensity. Light waves can pass through materials or be reflected from boundaries and interfaces without interacting with each other.

Laser sources can provide sufficiently high light intensities to modify the optical properties of materials. Light waves can then interact with each other, exchanging momentum and energy. This interaction can result in the generation of optical fields at new frequencies.

The first nonlinear optical effect induced by laser was a second harmonic generation found in 1961 under irradiation of crystalline quartz by pulsed Ruby laser.

Peter Franken, 1961

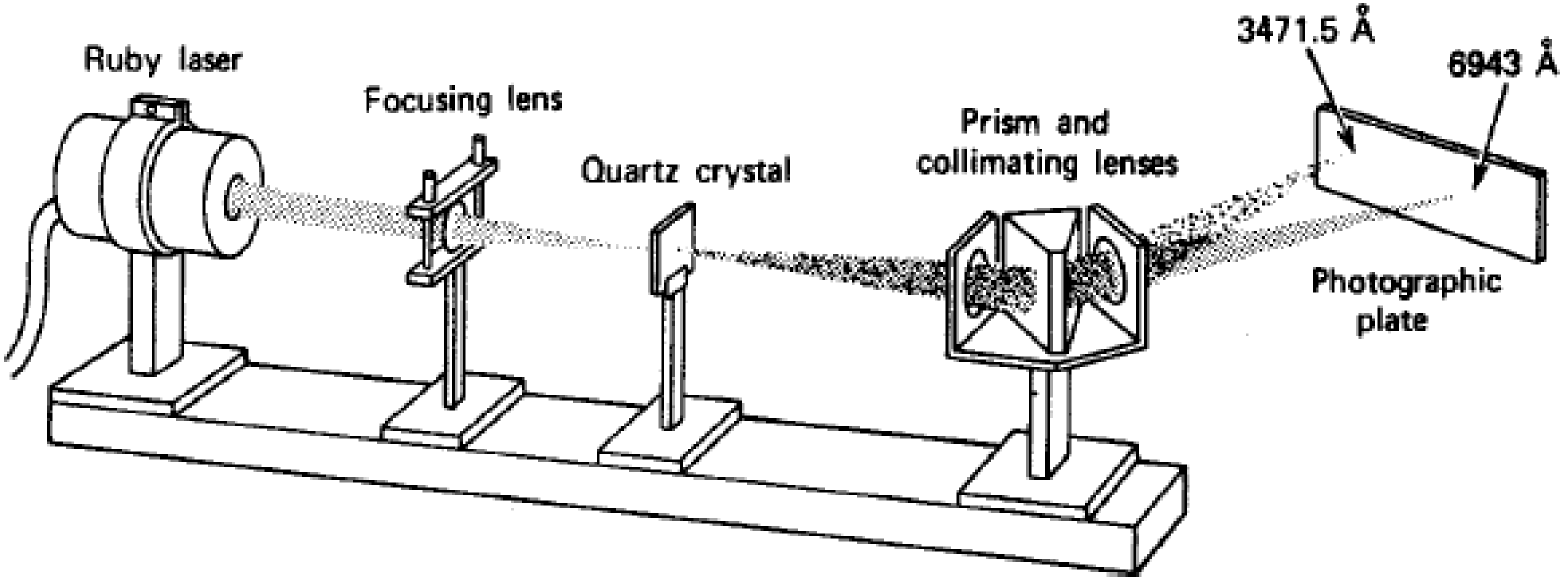


Fig: Frequency doubling of a Ruby laser: λ = 694.3 nm → λ = 347.1 nm as shown by Franken et al.[13]

Linear optics

- Optical phenomena commonly observed in nature such as reflection, refraction, and birefringence result from *linear* interactions with matter.
- In this conventional (linear) regime, the polarization (**P**) induced in the medium is linearly proportional to the electric field **E** of an applied optical wave:

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

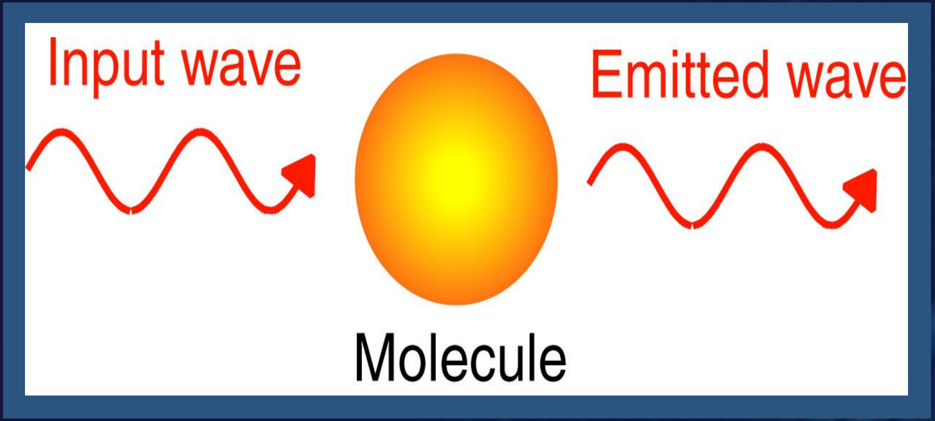
where ε_o is the *vacuum permittivity,* $\chi^{(1)}$ is the *linear susceptibility* (defined as the ability of the material to polarize the electrical field passing through it), and **E** is the electric field strength.

- In a linear medium the refractive index n_0 is a constant, independent of beam intensity for a given λ.

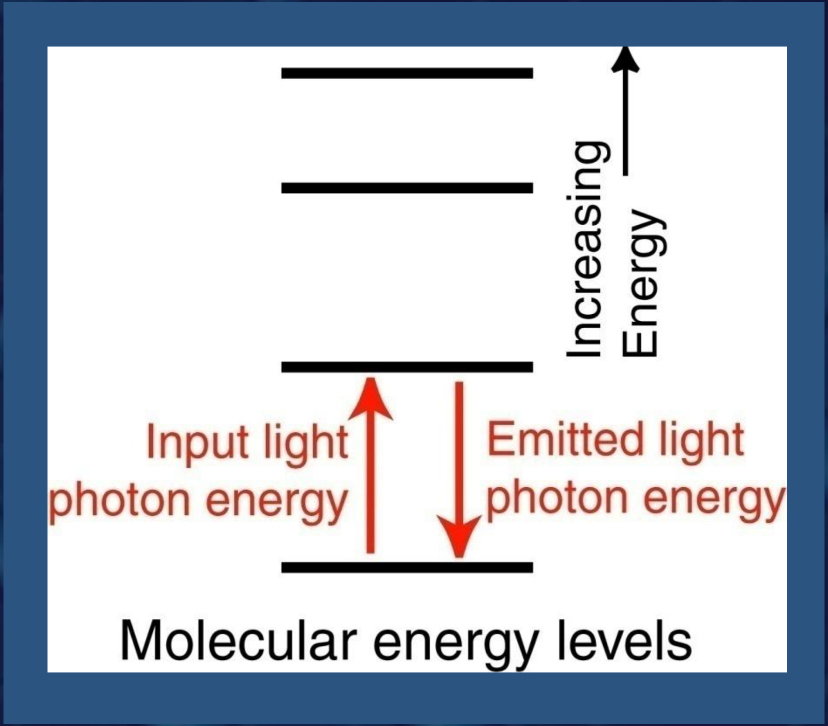
- Also, different ν of light encounter slightly different indices of refraction

- Some materials have two values of n depending on the polarization of the light. These are called n_o and n_e. This property is called **birefringence**

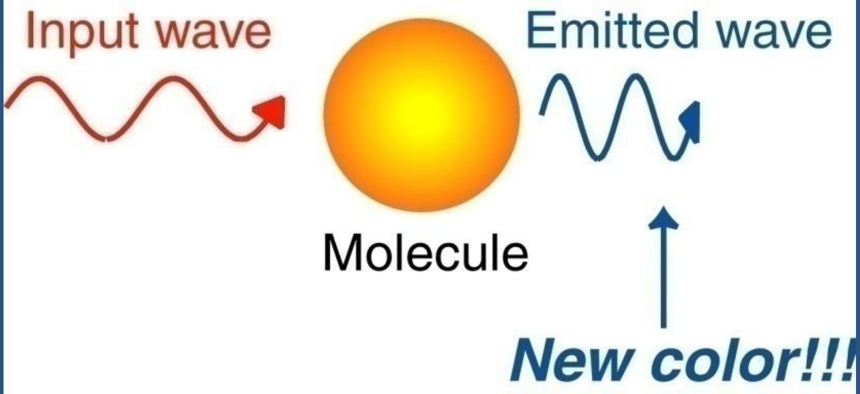
In Linear optics

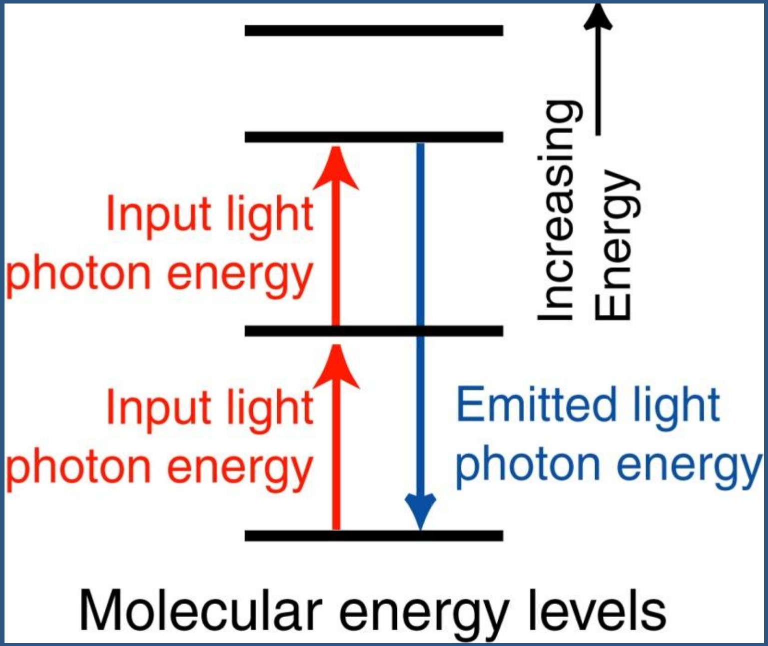


A light wave acts on a molecule, which vibrates and then emits its own light wave that interferes with the original light wave.



In Non-Linear Optics

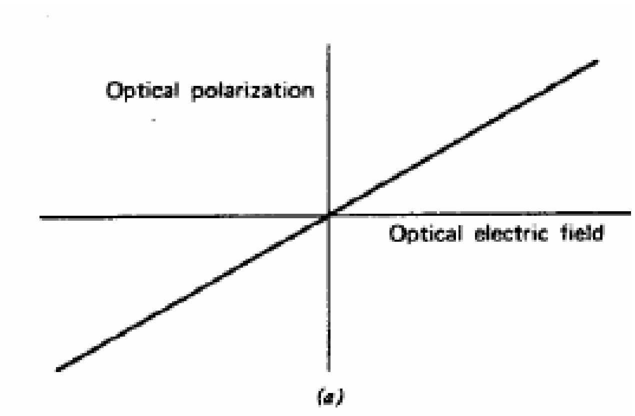




If irradiance is high enough vibrations at all frequencies corresponding to all energy differences between populated states are produced.

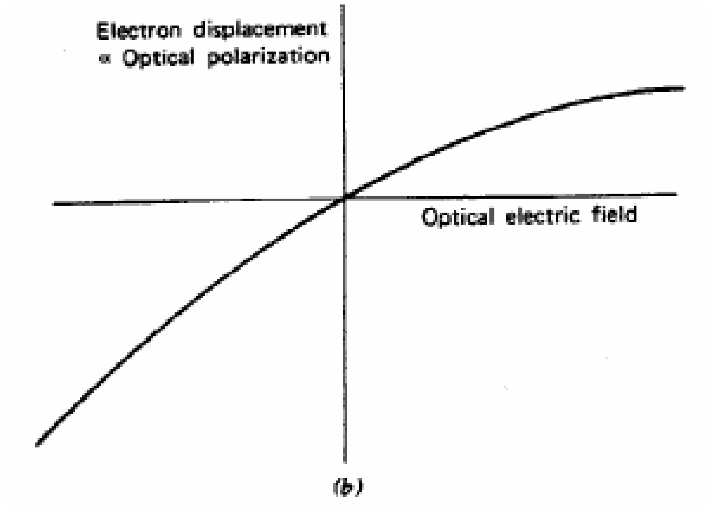
LINEAR RESPONSE

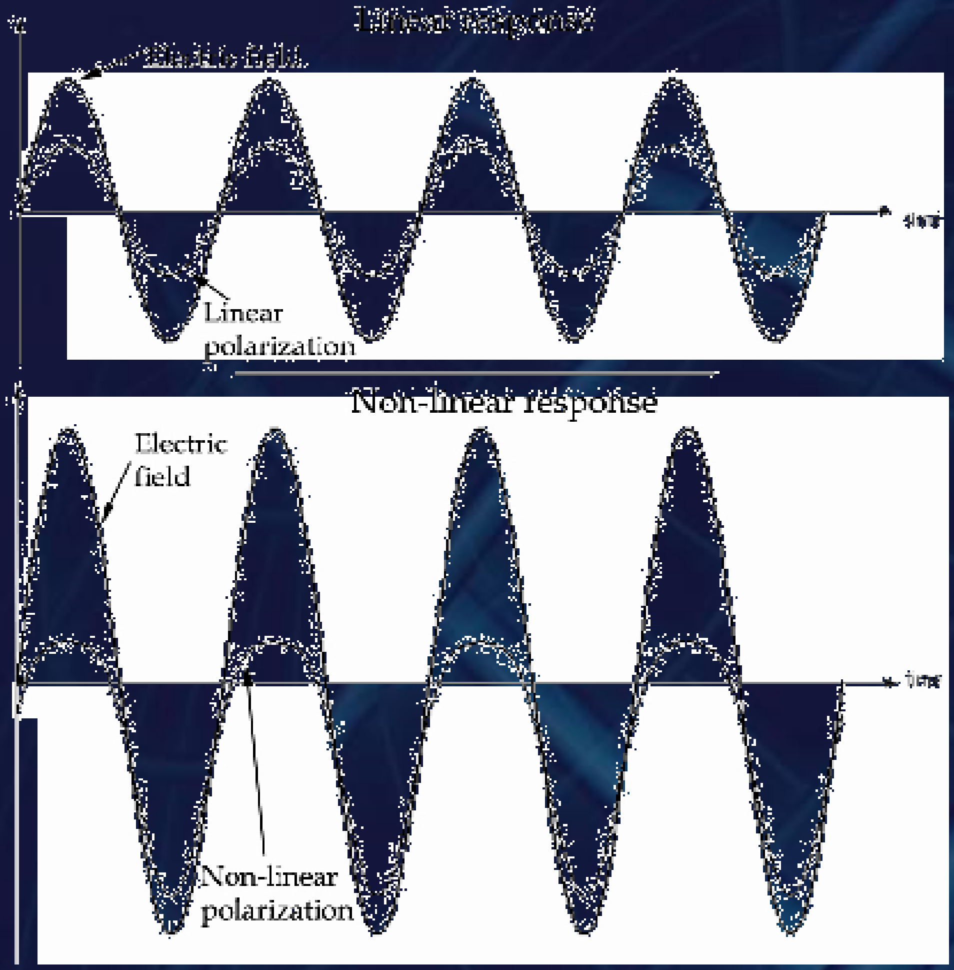
$$P = \chi^{(1)} E$$



NONLINEAR RESPONSE

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




Linear response
Electric field
time
Linear polarization
Non-linear response
Electric field
time
Non-linear polarization

In *non-resonant,* interactions (without absorption), the refractive index: $n = f(I) = f\{E(r, \lambda, t, \phi)\}$

In *resonant interactions*, the absorption coefficient:
$\alpha = f(I) = f\{E(r, \lambda, t, \phi)\}$

The transmission of a sample T is function of incident intensity depending on light and material parameters (n and α) in NL case.

Linear range:

$$I\,\text{inc}\,(r, \lambda, t, \phi) \longrightarrow \boxed{\begin{array}{l} n \neq f(I\,\text{inc}) \\ \alpha \neq f(I\,\text{inc}) \end{array}} \longrightarrow I\,\text{out}\,(r, \lambda, t, \phi)$$

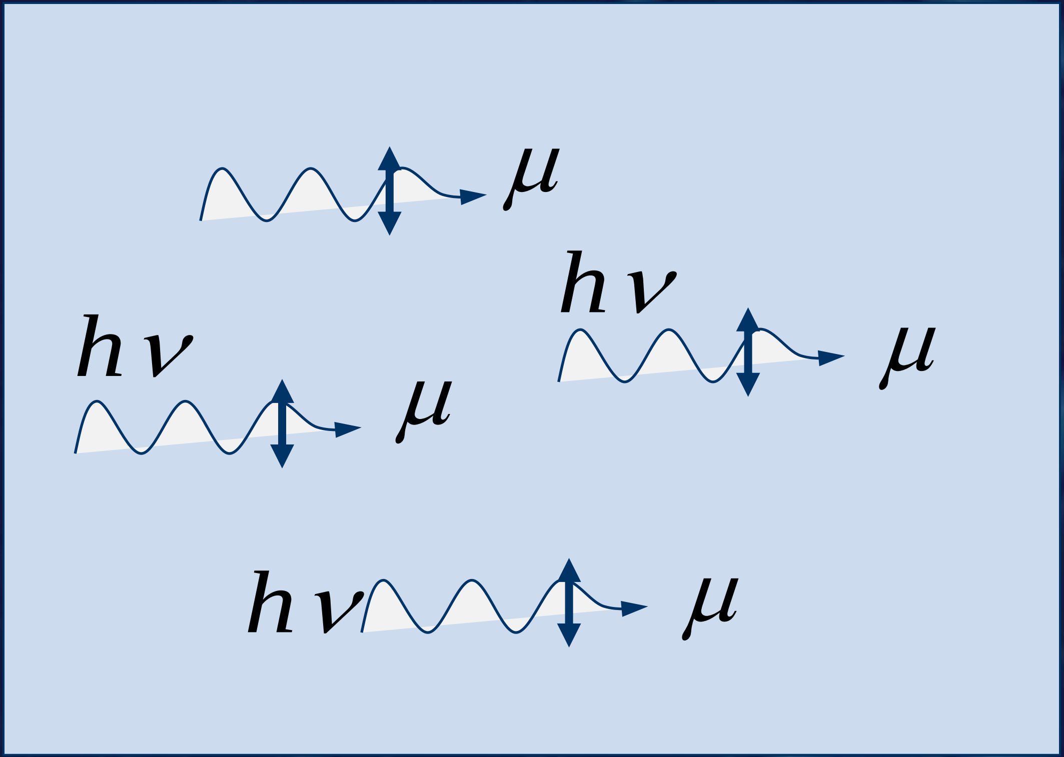
$T \neq f(I\,\text{inc}) = \text{constant}$

Nonlinear range:

$$I\,\text{inc}\,(r, \lambda, t, \phi) \longrightarrow \boxed{\begin{array}{l} n = f(I\,\text{inc}) \\ \alpha = f(I\,\text{inc}) \end{array}} \longrightarrow I\,\text{out}\,(r, \lambda, t, \phi)$$

$T = f(I\,\text{inc}) = \text{variable}$

The response of an optical medium to the incident electromagnetic field is the induced dipole moments inside the medium



The propagation of a wave through material produces changes in the spatial and temporal distributions of electrical charges as the electrons and atoms react to the e. m. fields of the wave.

The main effect of the forces exerted by the fields on the charged particles is a displacement of the valance electrons from their normal orbit. This perturbation creates electric dipoles whose macroscopic manifestation is the polarization, which depends on the strength of the electric field E and the dielectric susceptibility of the material. For small field strengths:

P_l

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

where ε_o is the vacuum permittivity. $\chi^{(1)}$ is the linear susceptibility, E is the electric field strength.

In the nonlinear case, the re-radiating comes from dipoles whose amplitudes do not reproduce the sinusoidal electric field that generates them. As a result, the distorted re-radiated wave contains different frequencies from that of the original.

Nonlinear effects occur with increasing the electric field strength, then

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

$$P_{tot} = P_{Lin} + P_{NL}$$

Dipole moment per unit volume or polarization

$$P_i = P_i^0 + \chi_{ij} E_j$$

The general form of polarization

$$P_i = P_i^0 + \chi_{ij}^{(1)} E_j + \chi_{ijk}^{(2)} E_j E_k + \chi_{ijkl}^{(3)} E_j E_k E_l + \cdots$$

Nonlinear Polarization

- Permanent Polarization
- First order polarization:
- Second order Polarization
- Third Order Polarization

$$P_i^0$$

$$P_i^1 = \chi_{ij}^{(1)} E_j$$

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

$$P_i^3 = \chi_{ijkl}^{(3)} E_j E_k E_l$$

When an electric field is applied to a dielectric material, the material becomes polarised (i.e. the electrons shift with respect to the nuclei). The resulting polarisation depends on the strength of the electric field and the dielectric susceptibility, χ

$$P = D - \varepsilon_0 E = \chi \, \varepsilon_0 E$$

where

$$D = \varepsilon_0 \, \varepsilon_r \, E, \quad \text{i.e.} \quad \chi = \varepsilon_r - 1$$

In isotropic materials the above relationship may be scalar (i.e. P parallel to E). However, in general, crystals are anisotropic and the relationship between P and E is best described by a tensor.

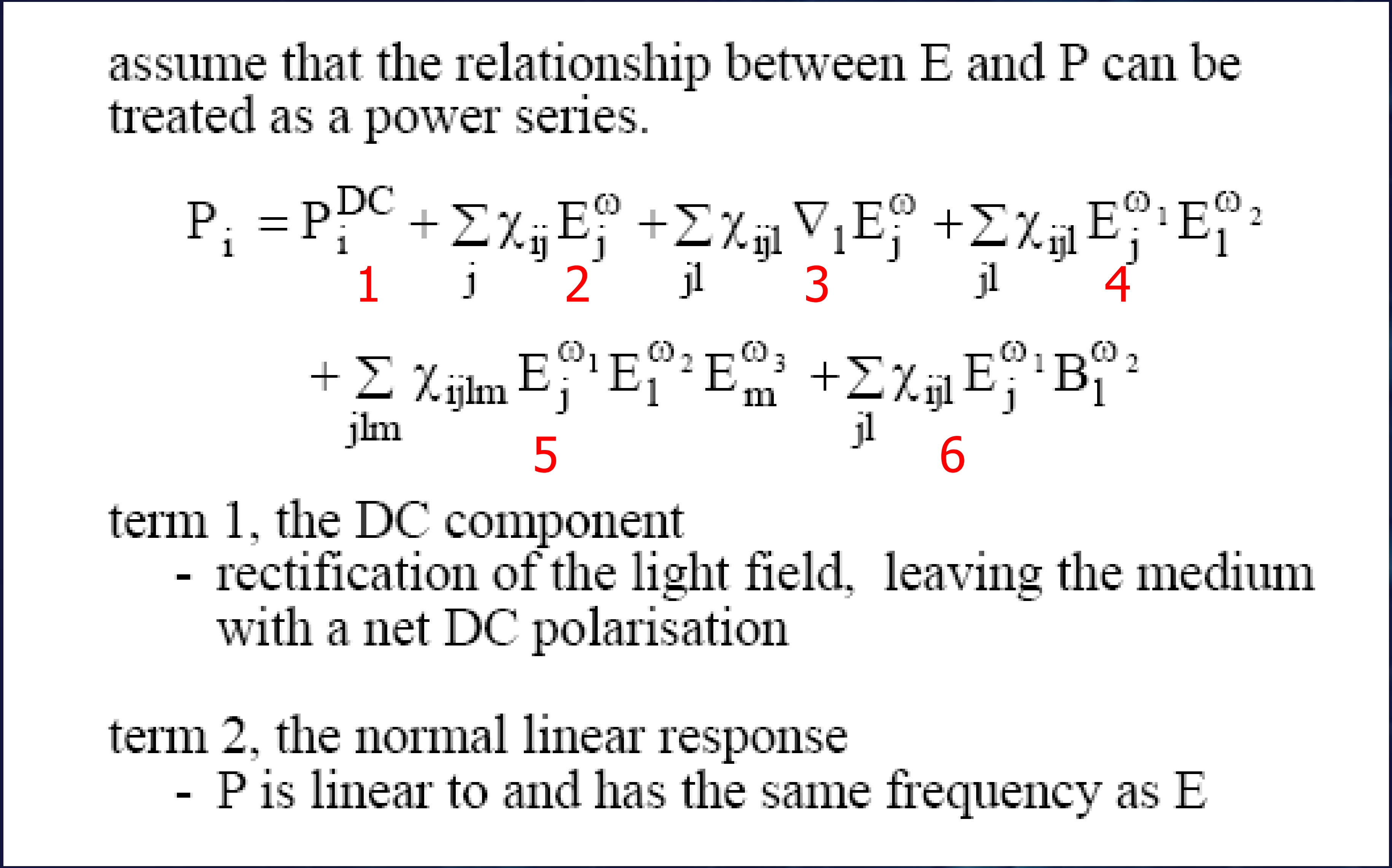
$$\left(P_x, P_y, P_z\right) = \varepsilon_0 \begin{pmatrix} \chi_{1,1} & \chi_{1,2} & \chi_{1,3} \\ \chi_{2,1} & \chi_{2,2} & \chi_{2,3} \\ \chi_{3,1} & \chi_{3,2} & \chi_{3,3} \end{pmatrix} \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix}$$

note that the polarisation in the x-direction depends on the applied fields in the x , y and z-directions.

The nonlinear susceptibilities X$^{(n)}$ *(where n is the rank of the susceptibilities)* are in general complex tensors and thus the polarization will be complex.

The electric field (E) is related to the local power density (I) by

$$I = \frac{1}{2} \in_0 cE^2$$



assume that the relationship between E and P can be treated as a power series.

$$P_i = \underset{1}{P_i^{DC}} + \sum_j \underset{2}{\chi_{ij} E_j^{\omega}} + \sum_{jl} \underset{3}{\chi_{ijl} \nabla_l E_j^{\omega}} + \sum_{jl} \underset{4}{\chi_{ijl} E_j^{\omega_1} E_l^{\omega_2}}$$

$$+ \sum_{jlm} \underset{5}{\chi_{ijlm} E_j^{\omega_1} E_l^{\omega_2} E_m^{\omega_3}} + \sum_{jl} \underset{6}{\chi_{ijl} E_j^{\omega_1} B_l^{\omega_2}}$$

term 1, the DC component

- rectification of the light field, leaving the medium with a net DC polarisation

term 2, the normal linear response

- P is linear to and has the same frequency as E

$$P_i = \underset{1}{P_i^{DC}} + \underset{2}{\sum_j \chi_{ij} E_j^{\omega}} + \underset{3}{\sum_{jl} \chi_{ijl} \nabla_l E_j^{\omega}} + \underset{4}{\sum_{jl} \chi_{ijl} E_j^{\omega_1} E_l^{\omega_2}}$$

$$+ \underset{5}{\sum_{jlm} \chi_{ijlm} E_j^{\omega_1} E_l^{\omega_2} E_m^{\omega_3}} + \underset{6}{\sum_{jl} \chi_{ijl} E_j^{\omega_1} B_l^{\omega_2}}$$

term 3, the Curl E term
- responsible for optical activity

term 4, second order processes (rest of this course)
- proportional to the product of the two E-fields e.g.
- $\omega = \omega_1 + \omega_2$, sum frequency mixing
- $\omega_1 = \omega_2$, $\omega = \omega_1 + \omega_2$, second harmonic generation
- $\omega_2 = 0$, $\omega = \omega_1$, the Pockels effect
- $\omega = \omega_1 - \omega_2$, difference frequency mixing

term 5, third order response
- third harmonic generation
- Raman effect,
- optical Kerr effect
- four-wave difference frequency mixing

term 6, magneto optical effects
- $\omega_2 = 0$, the Faraday effect

Important Notations

$\mathbf{E}$	Electric Field [V/m]
$\mathbf{D}$	Displacement (or *Electric Flux Density*) [C/m^2]
$\mathbf{P}$	Electric Polarisation Density [C/m^2]
$\mathbf{H}$	Magnetic Field [A/m]
$\mathbf{B}$	Magnetic Flux Density [W/m^2]

All are vectors and functions of position vector $\mathbf{r}$ *and time* t

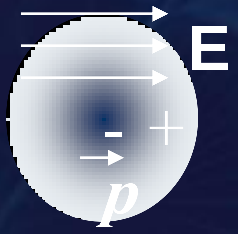
j = Current density

ρ = electrical charge density

μ_0 = magnetic permeability of free space

ε_o = electric permittivity of free space

D, *E*, P



The polarisation is induced by an E-field:

- $P = f(E)$
- $D = \varepsilon_0 E + P$

D is the electric flux density - caused by:
An E-field in the absence of the medium: $\varepsilon_0 E$,
Plus the field created by the response of the medium: P

- $P = 0$ in free space
- $P \neq 0$ in a dielectric

Nonlinear Optical Materials

Nonlinear optical (NLO) materials play a major role in nonlinear optics and in particular they have a great impact on information technology and industrial applications.

Some materials change light passing through them, depending upon orientation, temperature, light wavelength etc. NLO materials typically have a distinct crystal structure, which is an isotropic with respect to the electromagnetic radiation.

Many good NL crystals are now available for optical parametric oscillators and amplifications. New nonlinear crystals are being developed constantly.

Properties of Media

- Homogeneous: Ex. Glass
  - P doesn't vary with location
- Inhomogeneous: Ex. Graded-index fibre
  - P = P($\mathbf{r}$)
- Linear: Ex. Glass, low laser intensity
  - P = $\varepsilon_0 \chi$ E (where χ is the *electric susceptibility*)
- Non-linear: Ex. Glass, high laser intensity
  - P $\propto$ E ^{n} (where $n > 1$)
- Isotropic: Ex. Glass
  - P doesn't depend on direction of E
- Anisotropic: Ex. Calcite crystal
  - P = P$_i$ (where i = 1,2,3 for x,y,z)
- Dispersive (and Absorptive) media: Ex. Na Vapour
  - P has a memory: resonance

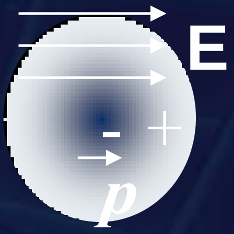
Linear Susceptibility

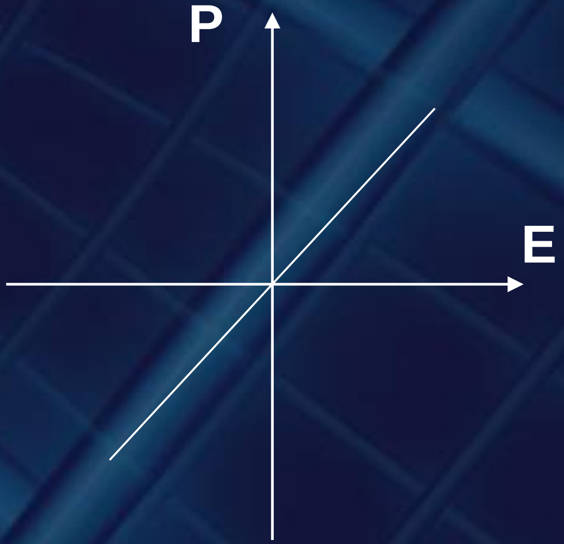
- Linear:
  - **P** = $\varepsilon_0\,\chi\,$**E**

    χ = the electric susceptibility (dimensionless)
    (dependent on the frequency)
    = 0 for vacuum
    = 0.00059 for air
    = 1.28 for crown glass

    $\varepsilon = \varepsilon_0\,(1+\chi)$ = permittivity of the material
    resulting in **D** = ε **E** (includes **P**)

    $n = (1+\chi)^{1/2}$ = refractive index = c_0/c
    (not valid near a resonance)





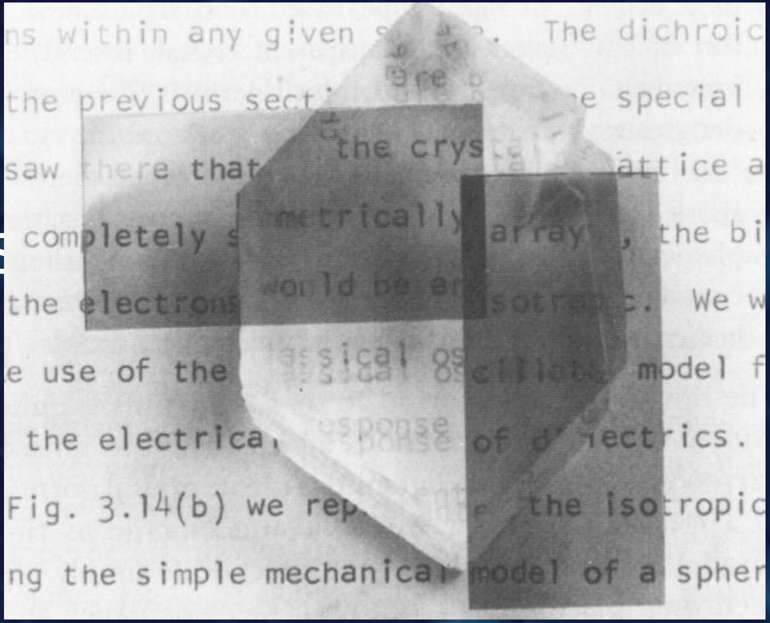
  - **E** is the total field
    - includes the *applied* field and the *field produced by* **P** !
    - **E** induces **P** which changes **E** which changes **P** , and so on...

Non-linear Susceptibility

- Non-linear effects:
  - Intense electric fields of one frequency can generate a **P** at another
  - $\mathbf{P} = \varepsilon_0 (\chi \mathbf{E} + \chi^{(2)} \mathbf{E}^2 + \chi^{(3)} \mathbf{E}^3 + \ldots)$
  - $\chi^{(2)}$ = 2nd order susceptibility
    - Only media lacking a centre of symmetry (no gases, liquids)
  - $\chi^{(3)}$ = 3rd order susceptibility
    - All dielectric media
  - Generally ($\chi \mathbf{E} >> \chi^{(2)} \mathbf{E}^2 >> \chi^{(3)} \mathbf{E}^3$)

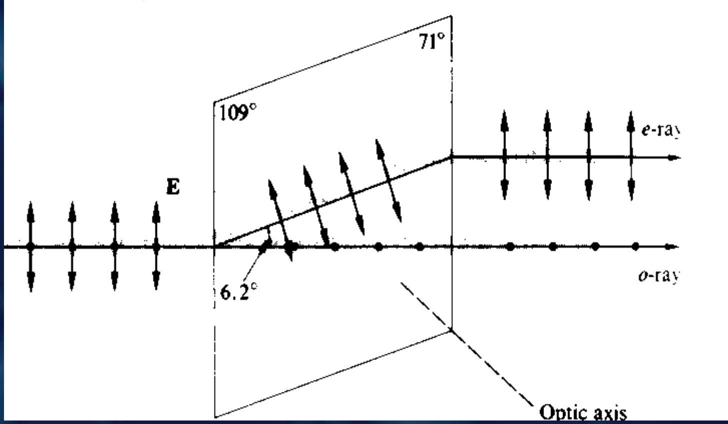
Anisotropic Media

- **P** depends on direction of input field(s)
  i.e., Polarisation of input wave(s
  Independent n_x, n_y, n_z



Calcite Crystal

- $\mathbf{P}_i = \varepsilon_0 \sum_j \chi_{ij} \mathbf{E}_j$
  - where i,j = 1,2,3 for x,y,z
  - χ_{ij} is a 3 x 3 tensor with 6 independent variables ($c_{ji} = c_{ij}$
  - Special case: uniaxial crystals
    - $n_x = n_y = n_o$ and $n_z = n_e$
    - Known as "**birefringence**"



Reaching nonlinearity

If the response of the media is caused by electrons in non-resonant case for $\chi^{(n)}$ the following ratio is valid:

$$\boxed{\frac{\chi^{(n+1)}}{\chi^{(n)}} = \frac{1}{E_A}}$$

where E_A is an interatomic field. For hydrogen $E_A \approx 10^9\, V/cm$.

Nonlinear effects can be observed at relatively high light intensities, e.g. **pulse lasers**.

Nonlinear optics experiments became real after innovation of **Q-switched laser** with pulse duration of 10^{-8} s and intensities of 10^{10}-10^{11} W/cm^2.

fs lasers generate pulses with duration of 5-30 x10^{-15} s at the intensity up to 10^{17}-10^{20} W/cm^2. In this case the electric field in the light wave exceeds the value of E_A.

Nonlinear Effects

Second order effects

- Second harmonic generation (SHG) and
- Optical parametric oscillation (OPO)
- Optical parametric amplification (OPA).
- Pockel's effect.
- Electro-optical beam deflection.
- Optical rectification.

Third order effects

- Third harmonic generation (THG).
- Raman effect.
- Kerr effect - induced birefringence.
- Four- wave difference frequency mixing (FWM).
- Self- diffraction
- Self-phase modulation
- Stimulated Brillouin scattering

Higher 0rder effects

- High harmonic generation

2nd Order Effects

Two different electric fields **E_1 E_2** are superimposed and generate the NL polarization **$P^{(2)}$** $= \varepsilon_0 \chi^{(2)}$ **E_1 E_2**

The incident optical field

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

Nonlinear polarization contains the following terms

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

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

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

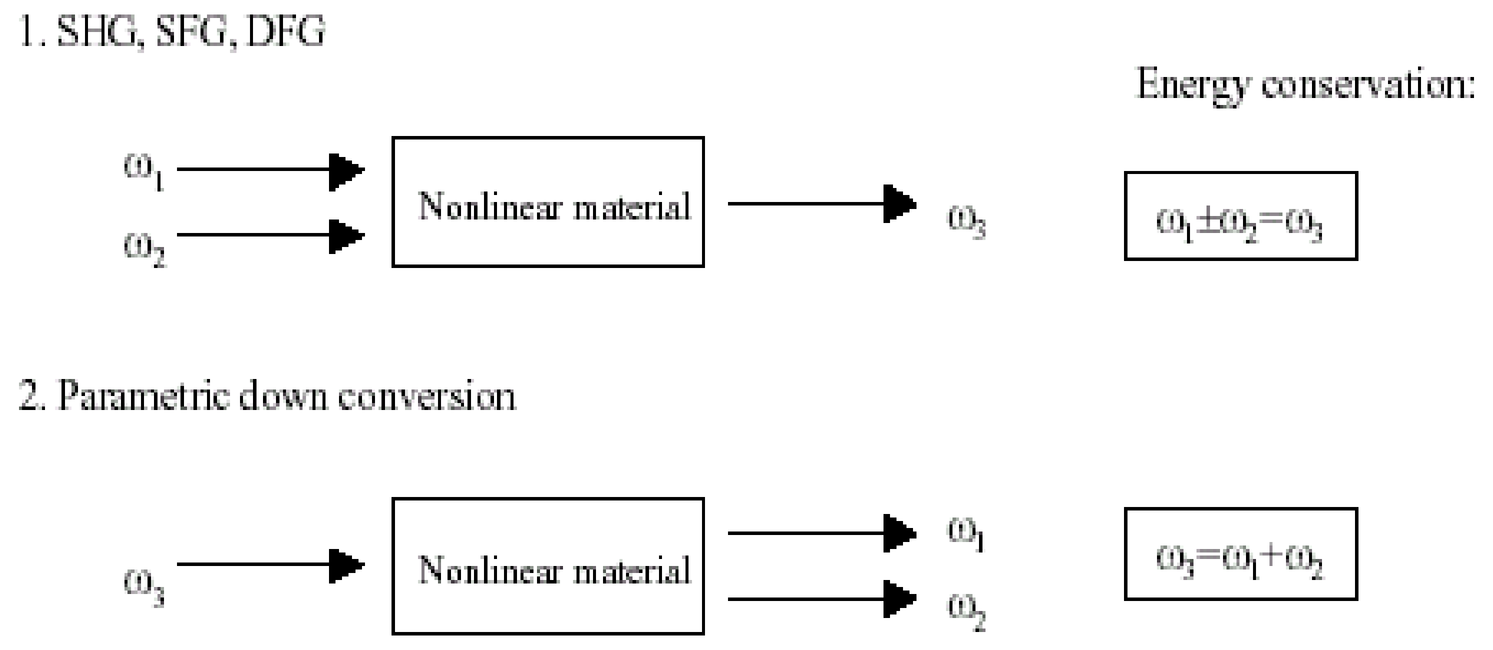
$$P(\omega_1 - \omega_2) = 2\chi^{(2)} E_1 E_2^* \qquad \text{(DFG)}$$

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

$\in_0$

Frequency Conversion

An important consequence of nonlinear properties is that several electro-magnetic waves of different frequencies can interact with each other in the material under the condition of energy conservation. In this way, it is possible to obtain frequency conversion.



Second Harmonic Generation (SHG)

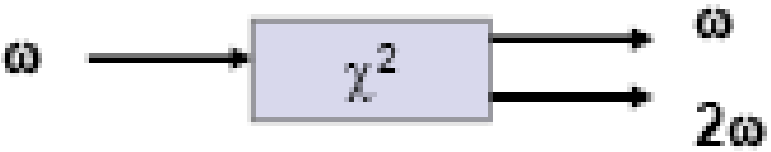
Two equal monochromatic light waves with same polarization, frequency and direction.

The 2nd order NL polarization is determined by the product of the two electric fields. The result shows two terms; zero or 2ω.

$$
\begin{aligned}
P^{(2)} &= \varepsilon 0\, \chi^{(2)}\, E_1\, E_2 \\
&= \varepsilon 0\, \chi^{(2)}\, E^2 \\
&= P^{(2)}(0) + P^{(2)}(2\omega_{inc})
\end{aligned}
$$

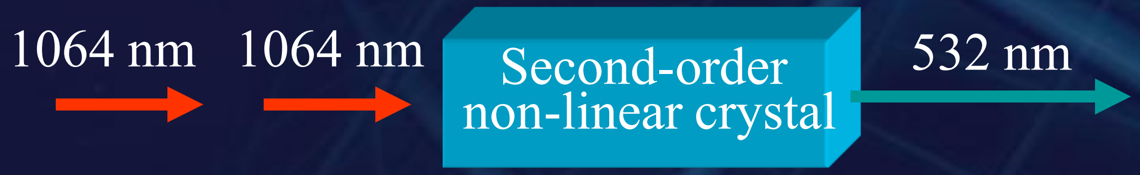
Second Harmonic Generation (SHG)

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



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

Second Harmonic Generation (SHG)

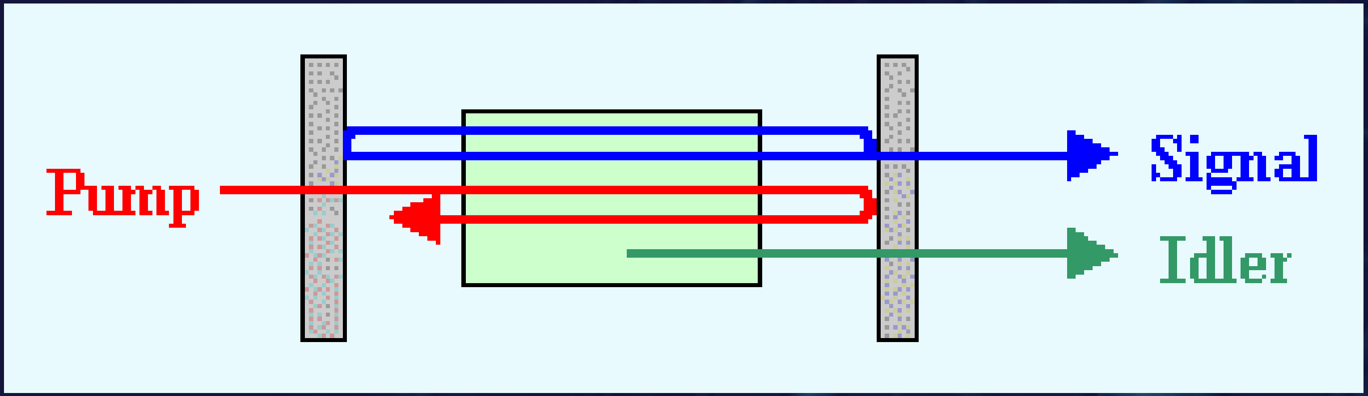


I_{532} = constant • (I_{1064})^2

- Note: 9,398 cm^{-1} + 9,398 cm^{-1} = 18,796 cm^{-1}

Optical Parametric Oscillator (OPO)

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




Q-switched Solid-state laser resonator

Pumping wavelength

NLO crystal

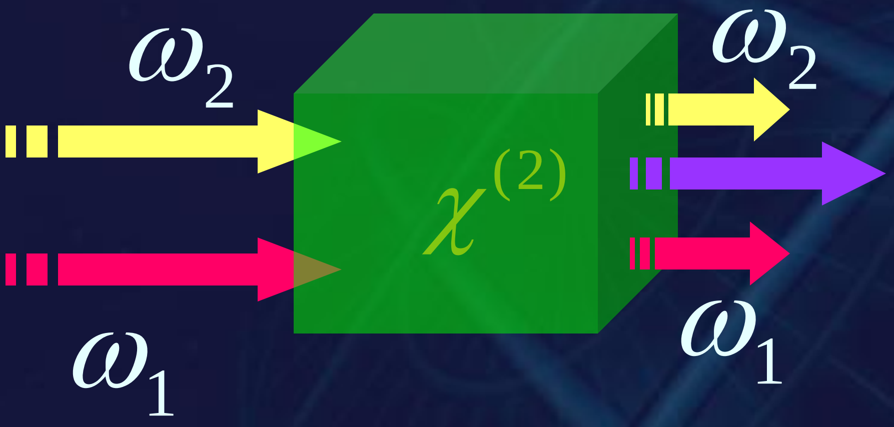
Signal wavelength

M 1

M 2

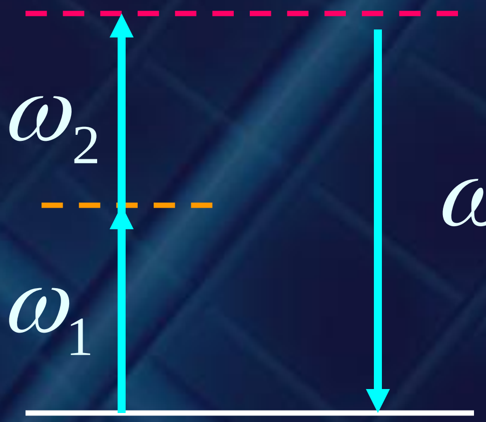
An OPO with return pump beam

Sum Frequency Generation SFG

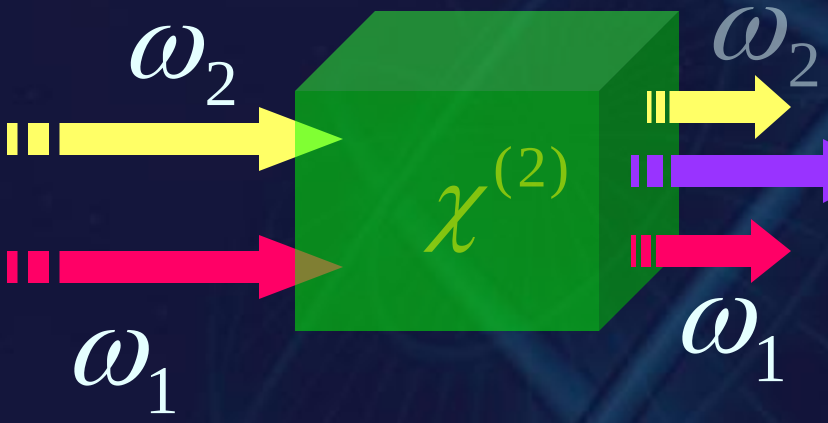


$$\omega_3 = \omega_1 + \omega_2$$

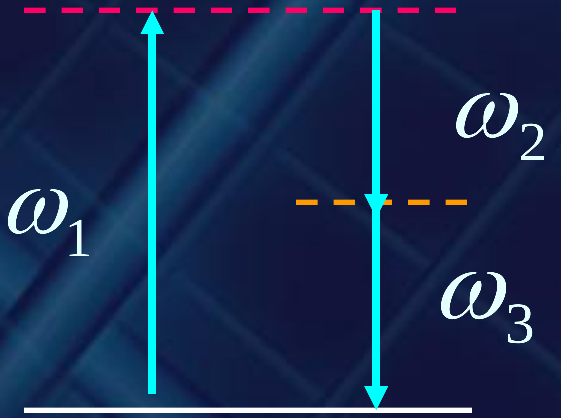
Application:
Tunable radiation in the UV Spectral region.



Difference Frequency Generation DFG



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

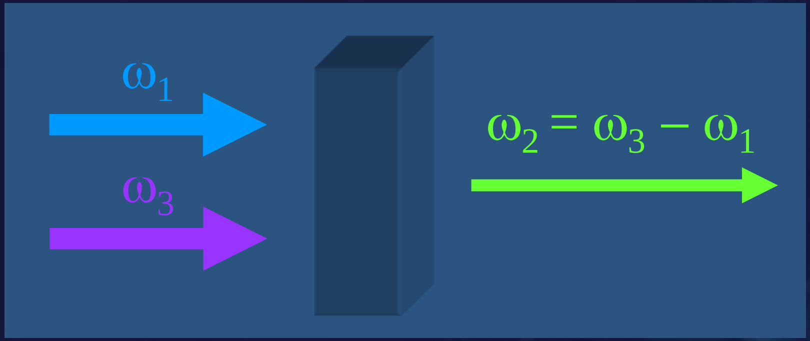


Application:

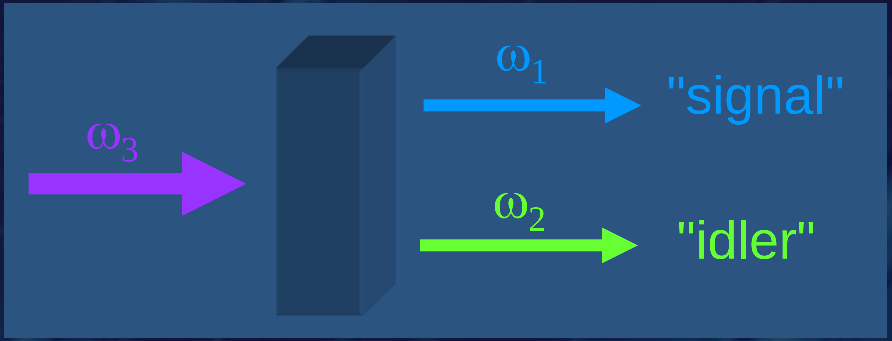
The low frequency photon, ω_2 amplifies in the presence of high frequency beam ω_1 . This is known as parametric amplification.

Difference-Frequency Generation: **Optical Parametric Generation, Amplification, Oscillation**

Difference-frequency generation takes many useful forms.

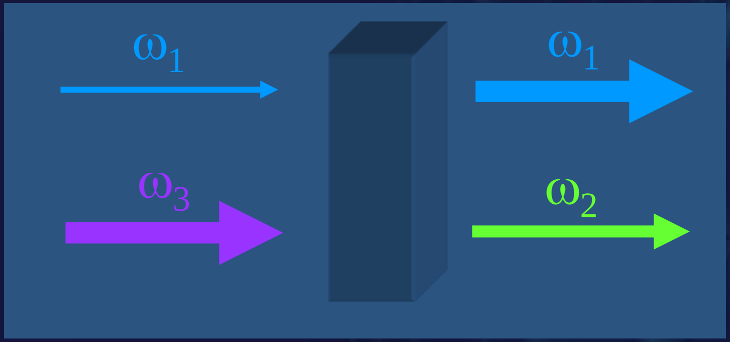


Parametric Down-Conversion
(Difference-frequency generation)

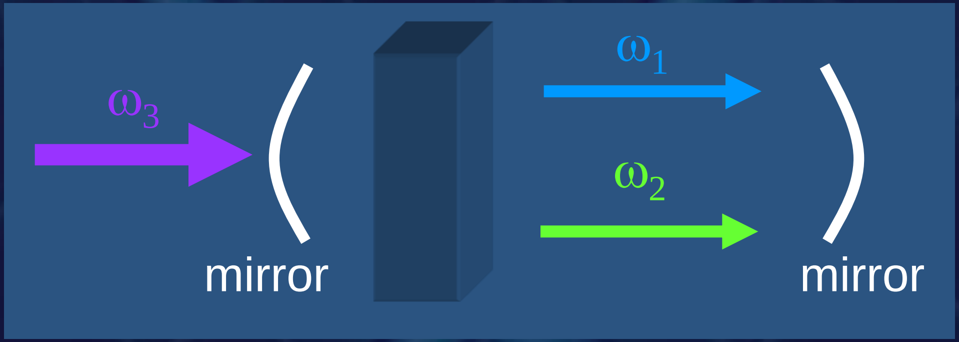


Optical Parametric
Generation (OPG)

By convention:
$\omega_{signal} > \omega_{idler}$

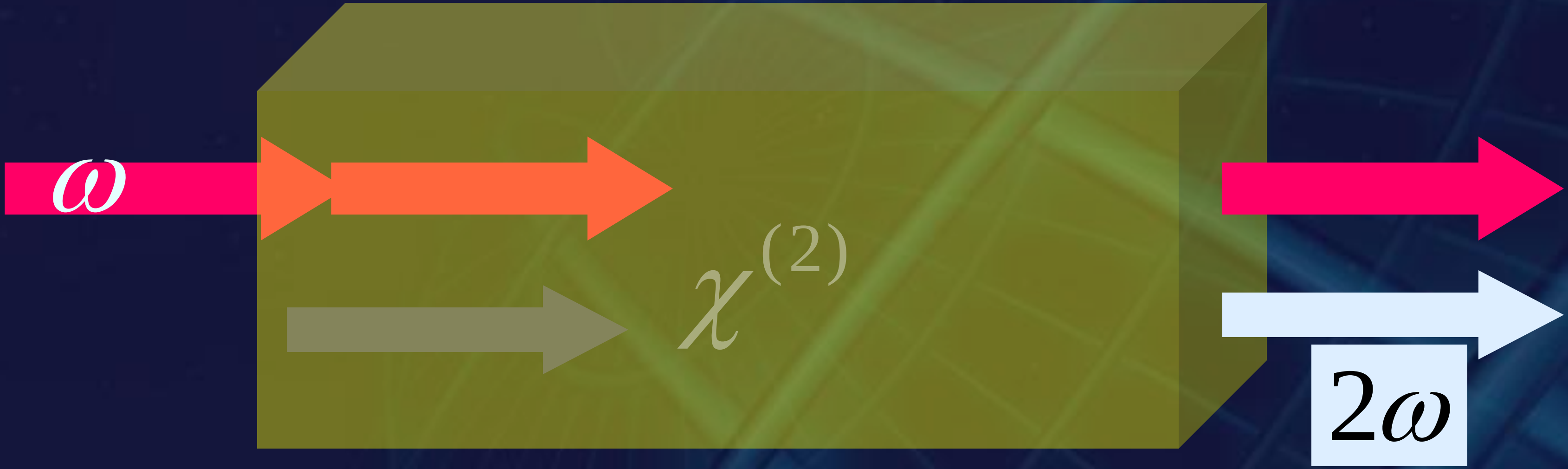


Optical Parametric
Amplification (OPA)



Optical Parametric
Oscillation (OPO)

Phase Matching

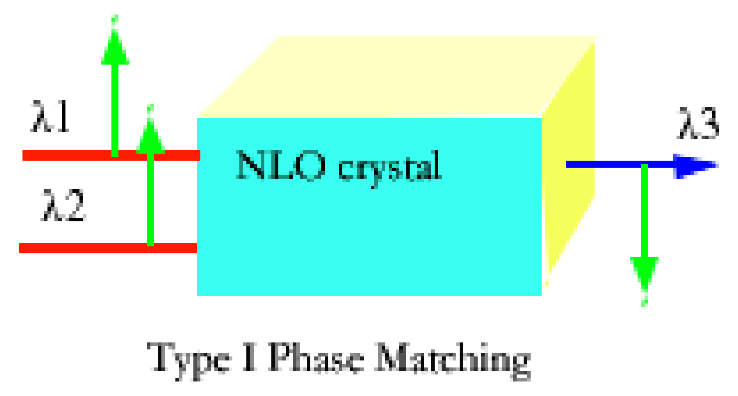


- Since the optical (NLO) media are dispersive, The fundamental and the harmonic signals have different propagation speeds inside the media.

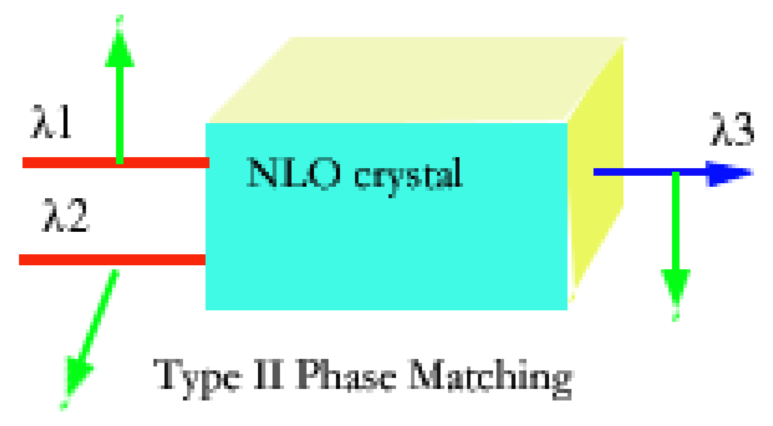
- The harmonic signals generated at different points interfere destructively with each other.

ϵ_0

The generation of new frequencies via nonlinear polarization in matter is more efficient the better the incident light waves and the newly generated waves are in suitable phase over the interaction length. This can be achieved in crystals by choosing a suitable orientation of the crystal with respect to the light beam and is called ***phase matching***



Type I Phase Matching



Type II Phase Matching

It can be achieved with anisotropic materials like crystals and is based on the birefringence in these materials.

Phase matching

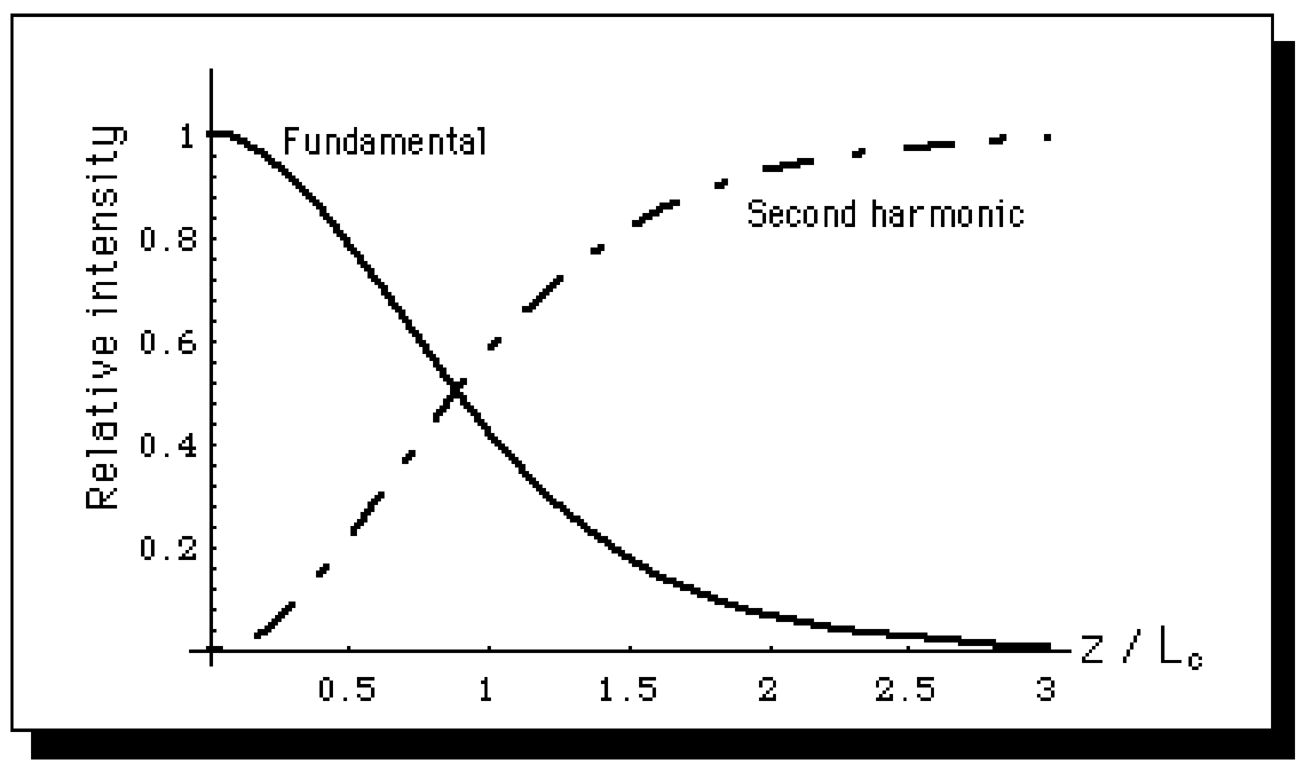


Fig. : Intensities of incident and new generated light waves in SHG as a function of interaction length in material with phase matching.

Pockel's Effect

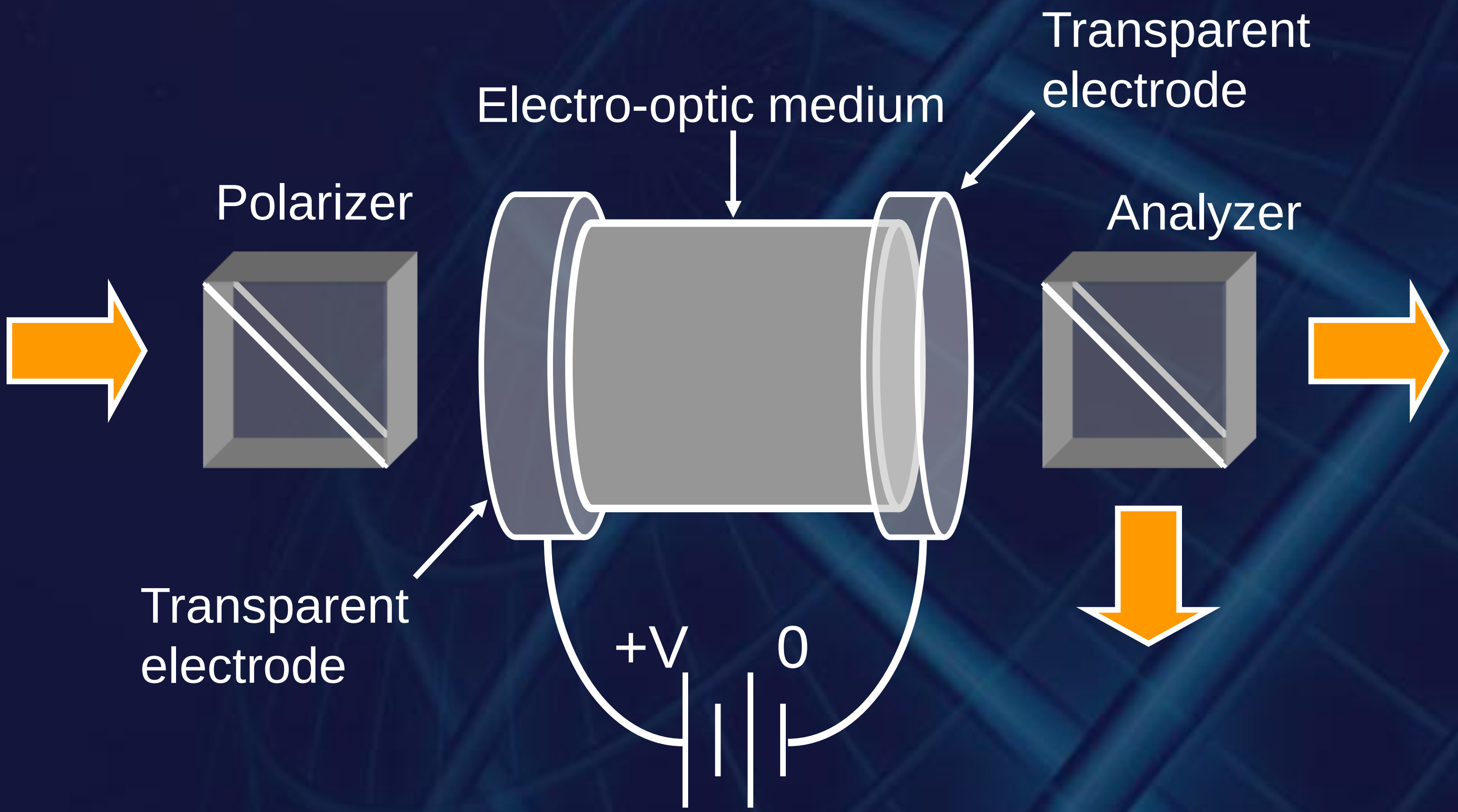
In a NL material, the electric filed of the light wave is superimposed on the externally applied filed and resulting NL polarization. Hence it's a *field –field* interaction.

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

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

The Pockels' Effect

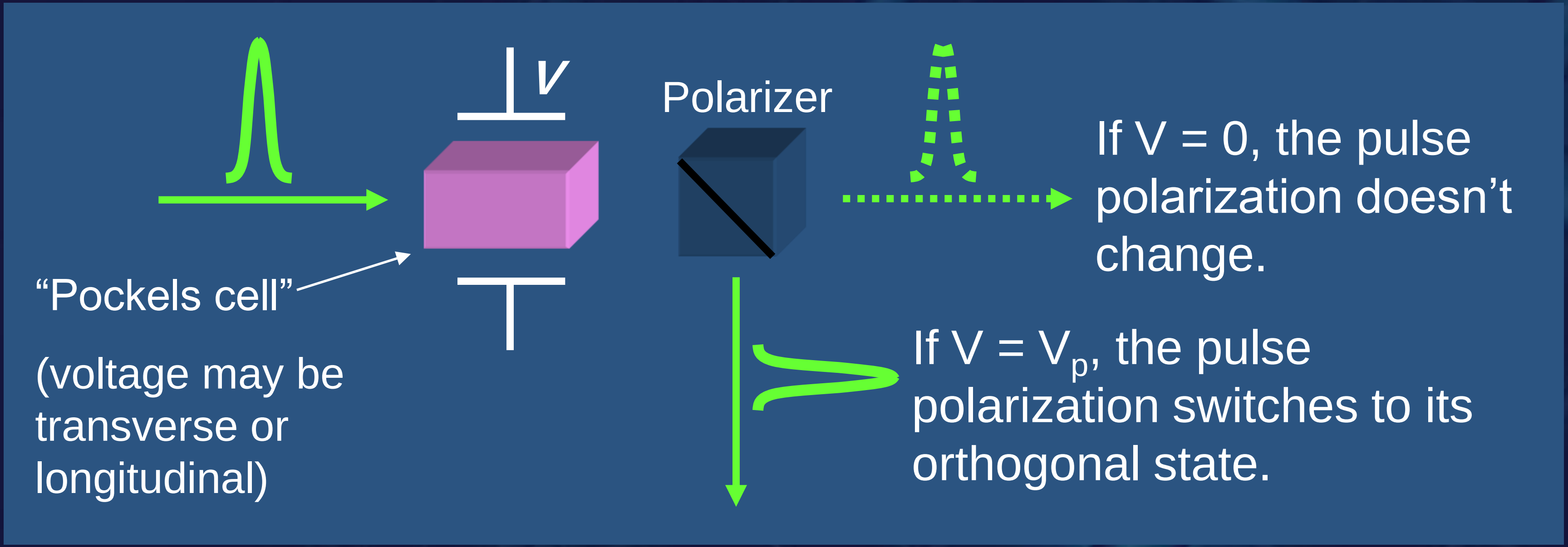
An electric field can induce birefringence.



The Pockels' effect allows control over the polarization rotation.

Applying a voltage to a crystal changes its refractive indices and introduces birefringence. In a sense, this is sum-frequency generation with a beam of zero frequency (but not zero field!).

A few kV can turn a crystal into a half- or quarter-wave plate.



If V = 0, the pulse polarization doesn't change.

"Pockels cell"

(voltage may be transverse or longitudinal)

If $V = V_p$, the pulse polarization switches to its orthogonal state.

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

Third Order Effects

- When the general form of the incident electric field is in the following form,

$$\tilde{E}(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + E_3 e^{-i\omega_3 t}$$

The third order polarization will have 22 components which their frequency dependent are

$$\omega_i, 3\omega_i, (\omega_i + \omega_j + \omega_k), (\omega_i + \omega_j - \omega_k)$$
$$(2\omega_i + \omega_j), (2\omega_i - \omega_j), i, j, k = 1,2,3$$

Third harmonic generation

If the medium possesses cubic nonlinearity, under the action of two monochromatic waves ω_1 and ω_2 the polarization $P^{(3)}$ would contain the components with frequencies:

$$3\omega_1, 3\omega_2, 2\omega_1 \pm \omega_2, 2\omega_2 \pm \omega_1 .$$

The Intensity Dependent Refractive Index

- 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

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

One can define an effective susceptibility

$$\chi_{\mathrm{eff}}=\chi^{(1)}+4\pi\,|E(\omega)|^2\,\chi^{(3)}$$

The refractive index can be defined as usual

$$n^2=1+4\pi\chi_{\mathrm{eff}}$$

By definition

$$n = n_0 + n_2 I$$

where

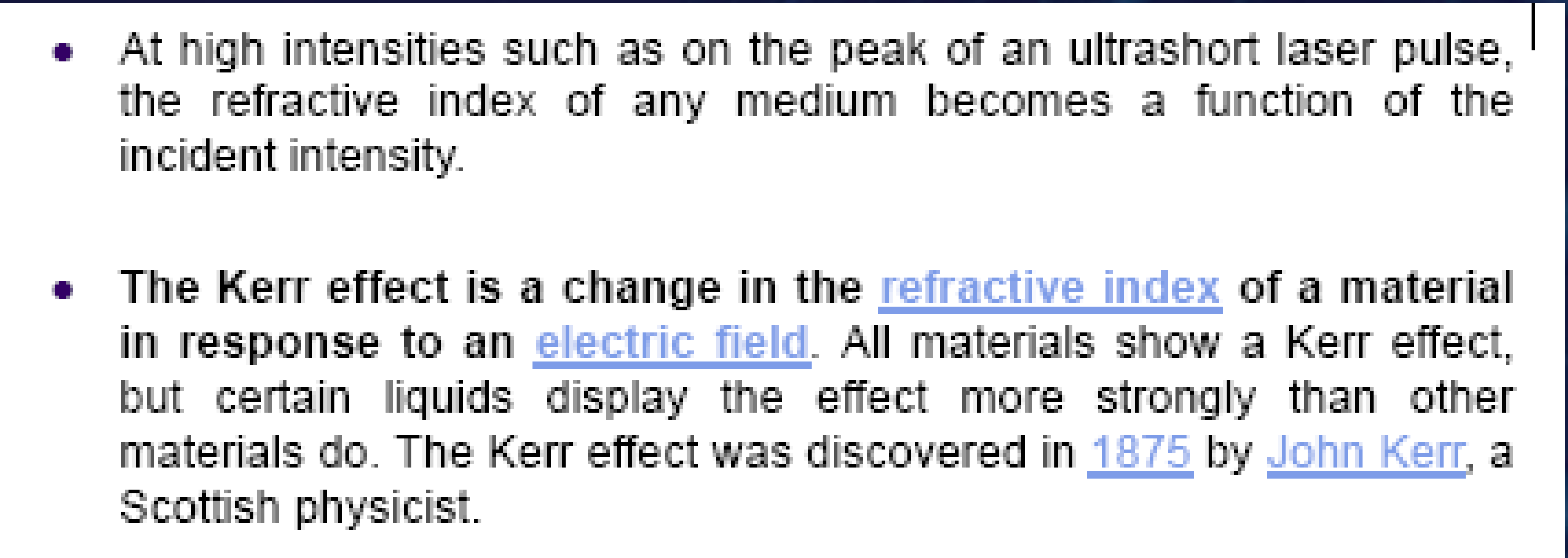
$$I = \frac{n_0 c}{2\pi} |E(\omega)|^2$$

$$n_2 = \frac{12\pi^2}{n_0^2 c} \chi^{(3)}$$

Typical values of nonlinear refractive index

Mechanism	n_2(cm^2/W)	$\chi^{(3)}_{1111}$ (esu)	Response time (sec)
Electronic Polarization	10^{-16}	10^{-14}	10^{-15}
Molecular Orientation	10^{-14}	10^{-12}	10^{-12}
Electrostriction	10^{-14}	10^{-12}	10^{-9}
Saturated Atomic Absorption	10^{-10}	10^{-8}	10^{-8}
Thermal effects	10^{-6}	10^{-4}	10^{-3}
Photorefractive Effect	large	large	Intensity dependent

Kerr effect



- At high intensities such as on the peak of an ultrashort laser pulse, the refractive index of any medium becomes a function of the incident intensity.

- The Kerr effect is a change in the refractive index of a material in response to an electric field. All materials show a Kerr effect, but certain liquids display the effect more strongly than other materials do. The Kerr effect was discovered in 1875 by John Kerr, a Scottish physicist.

- A strong applied electric field can induce birefringence in materials which are isotropic without the filed. The refractive index becomes a function of light intensity.

- $$n_{non-linear} = \sqrt{(1+\chi^{(1)}+\chi^{(3)})} \cong n_0 + n_2 I$$

The Kerr effect: the polarization rotation is proportional to the Kerr constant and E^2

$$\Delta n = \lambda_0 K E^2$$

where:
Δn is the induced birefringence, E is the electric field strength, K is the **Kerr constant** of the material.

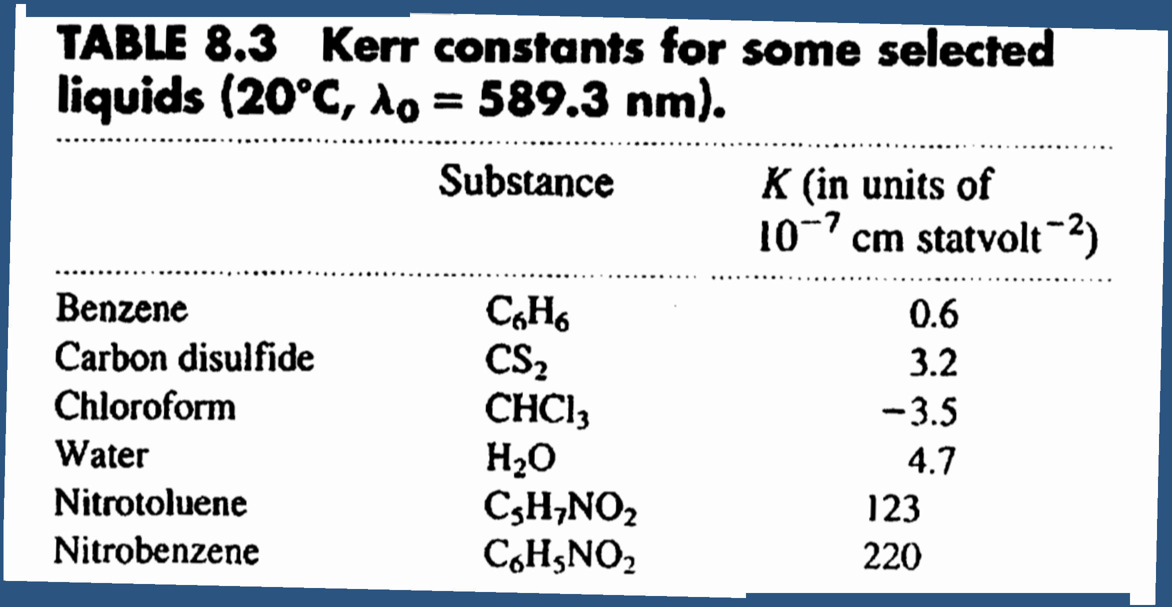


TABLE 8.3 Kerr constants for some selected liquids (20°C, λ_0 = 589.3 nm).

	Substance	K (in units of 10^{-7} cm statvolt^{-2})
Benzene	C_6H_6	0.6
Carbon disulfide	CS_2	3.2
Chloroform	$CHCl_3$	−3.5
Water	H_2O	4.7
Nitrotoluene	$C_5H_7NO_2$	123
Nitrobenzene	$C_6H_5NO_2$	220

Use the Kerr effect in isotropic media, where the Pockels' effect is zero

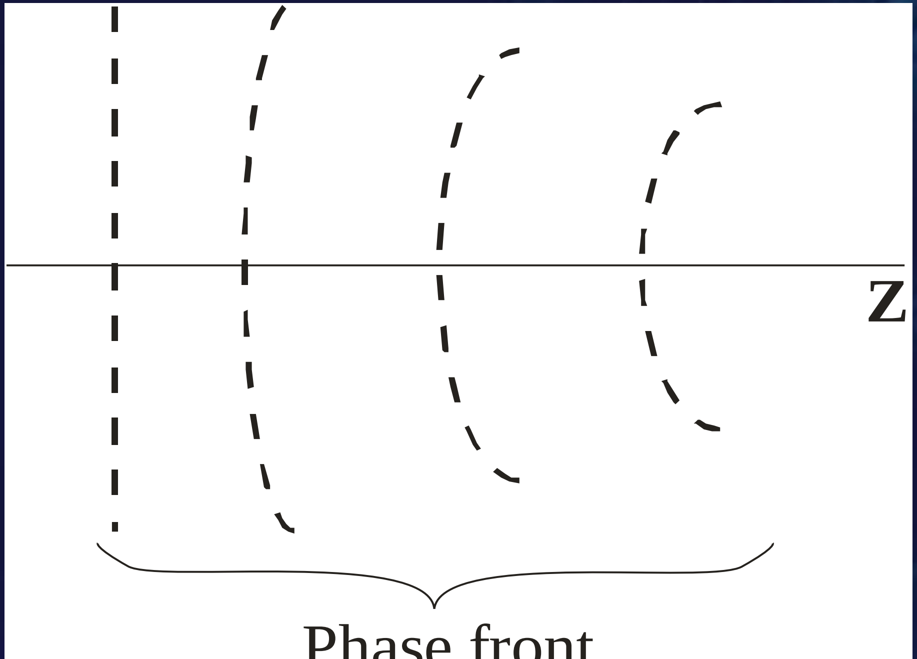
The **AC Kerr Effect** creates birefringence using intense fields of a light wave. Usually very high irradiances from ultrashort laser pulses are required to create quarter-wave rotations.

Self focusing

If the intensity of a laser beam is high enough instead of diffraction an opposite effect of self focusing takes place. Phase velocity depends on the intensity through nonlinear refractive index:

$$V_{ph} = c / n_0 + n_2 I$$

If $n_2 > 0$ the phase velocity at the axis of the beam is lower and nonlinear medium is working as a lens.



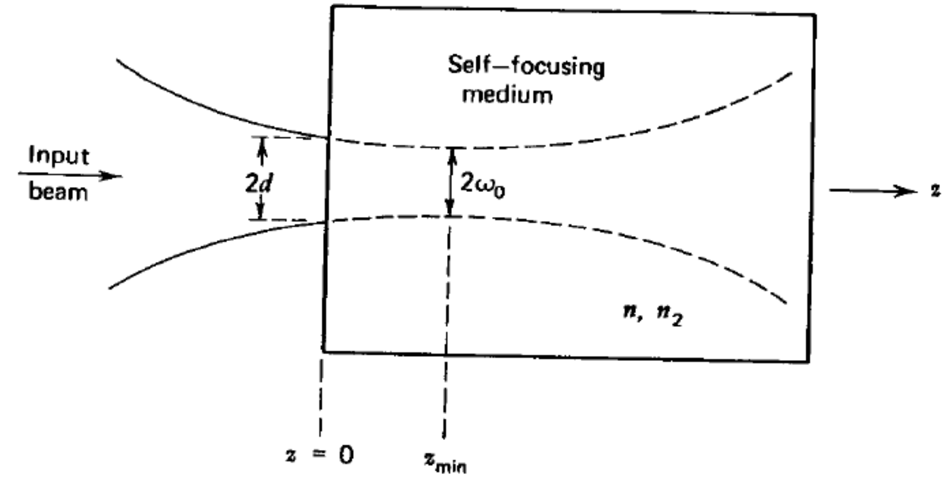
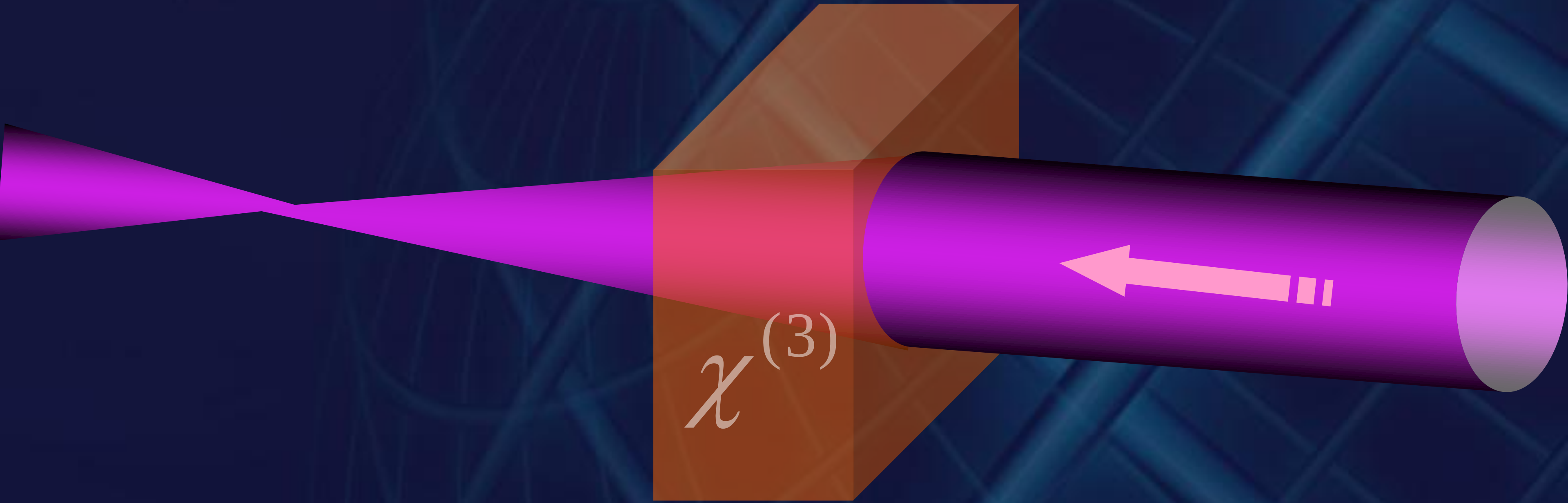


FIGURE 18.13 A Gaussian beam entering a slab of material that has a dielectric constant $\varepsilon = n^2 + nn_2 |E|^2$. The dashed curve gives the beam radius without self-focusing ($n_2 = 0$).

Self Focusing and Self Defocusing

- The laser beam has Gaussian intensity profile. It can induce a Gaussian refractive index profile inside the NLO sample.



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Optical Phase Conjugation

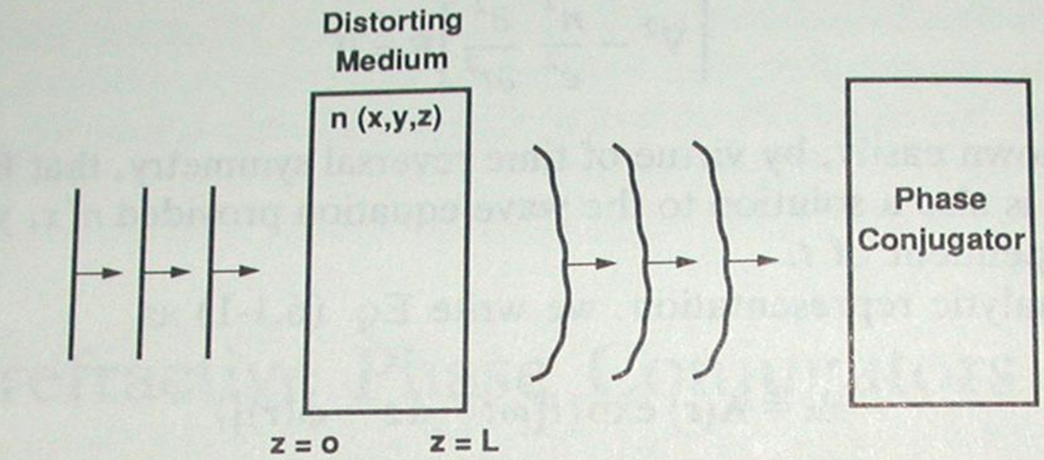
- Generation of a time-reversed replica of the wave, like a mirror. It can be used for improving the laser beam quality.

- $E = A \cos(\omega t - kz - \phi)$

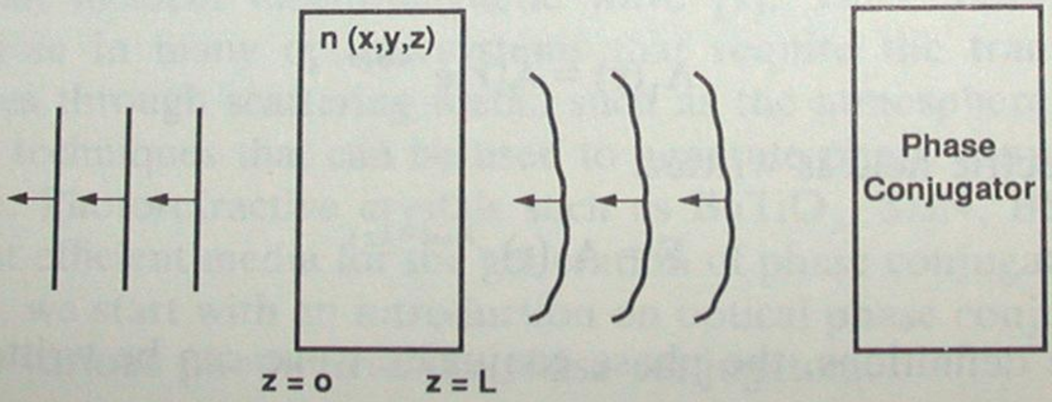
  $E_c = A \cos(\omega t + kz + \phi)$

- Lensless imaging, distortion correction, associated with a frequency shift
- With nonlinear techniques this can be done real-time

Phase Conjugation



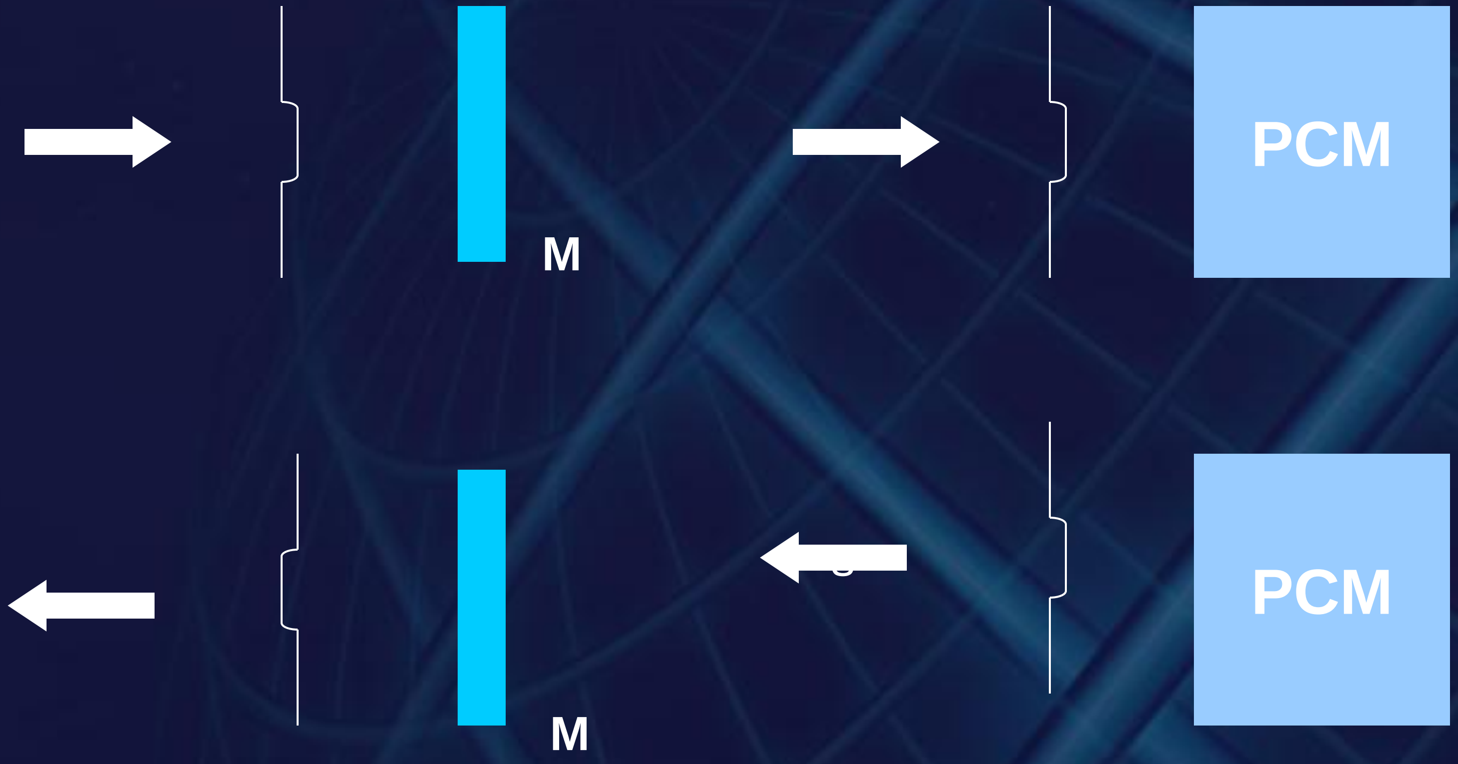
(a) Incident Wave



(b) Phase Conjugate Wave

Optical Phase Conjugation

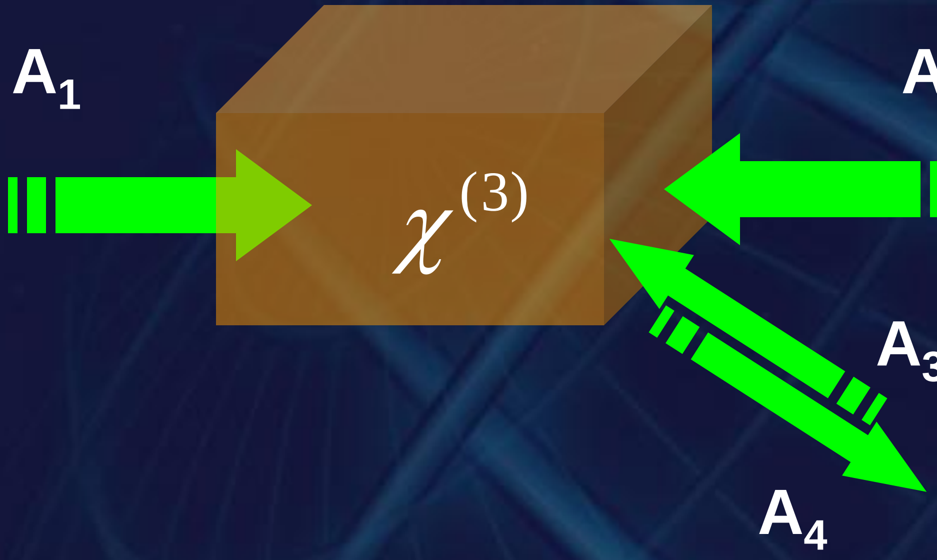
- Phase conjugation mirror



Degenerate Four Wave Mixing (DFWM)

- A 3rd order nonlinear optical process
- The induced grating by 2 input waves scatters the 3rd and generates the 4th
- Inputs are 2 antiparallel, high-power pumps and a weaker probe wave, the output could be amplified besides being conjugated
- In conventional holography gratings are recorded in a photographic emulsion

DFWM



- All of the three incoming beams A1, A2 and A3 should be originated from a coherent source.
- The fourth beam A4, will have the same Phase, Polarization, and Path as A3.
- It is possible that the intensity of A4 be more than that of A3

Raman and Brellouin Processes

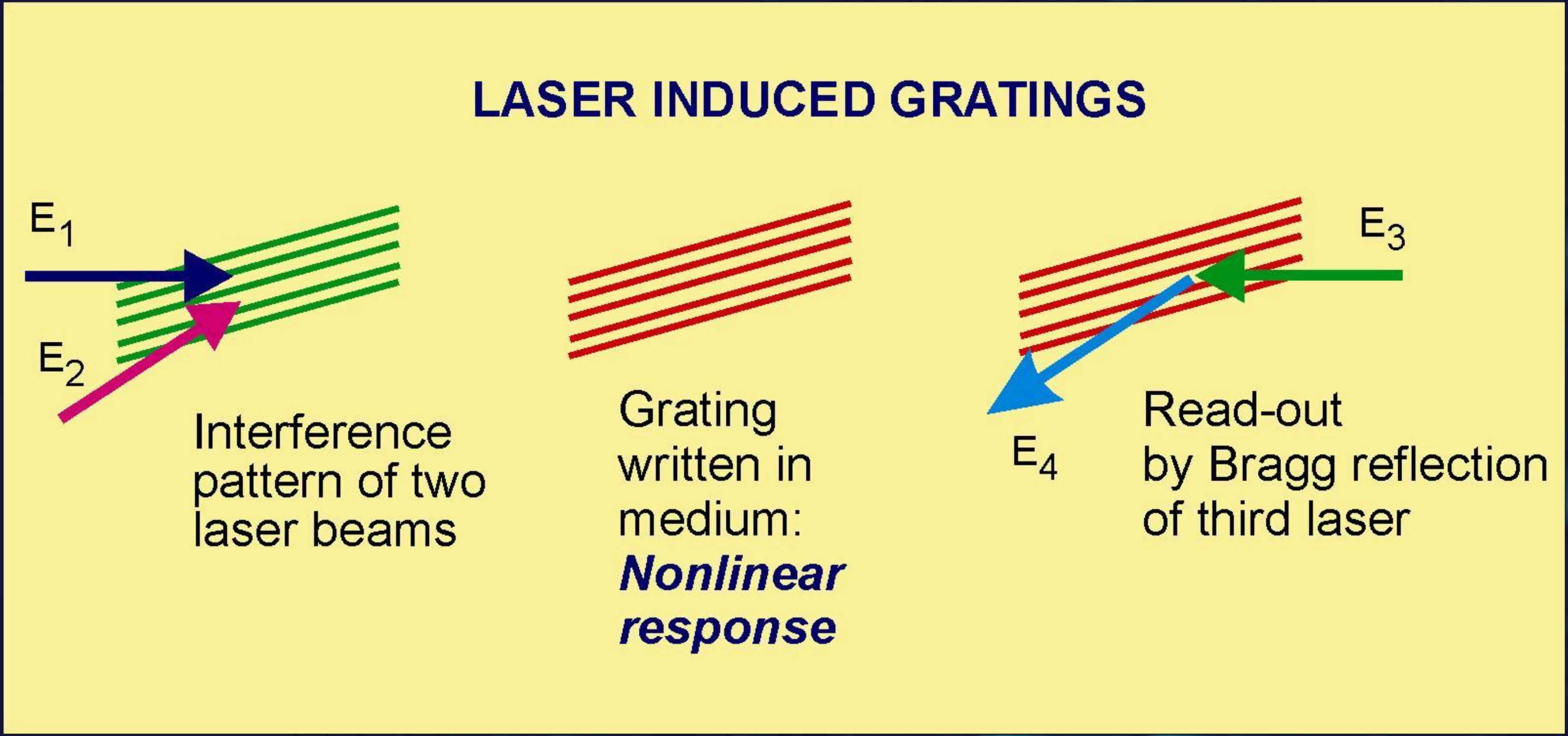
- Mixing of mechanical oscillations with a light beam in a nonlinear media
- Freq spectrum of light is modified while phonons are emitted or absorbed

Brillouin Scattering

- “n” is a function of density. Acoustic wave scatters light with a Doppler shift)
- In turn electronic polarization creates pressure variations
- Light can pump a sound wave
- Then it is scattered back with slightly different frequency.

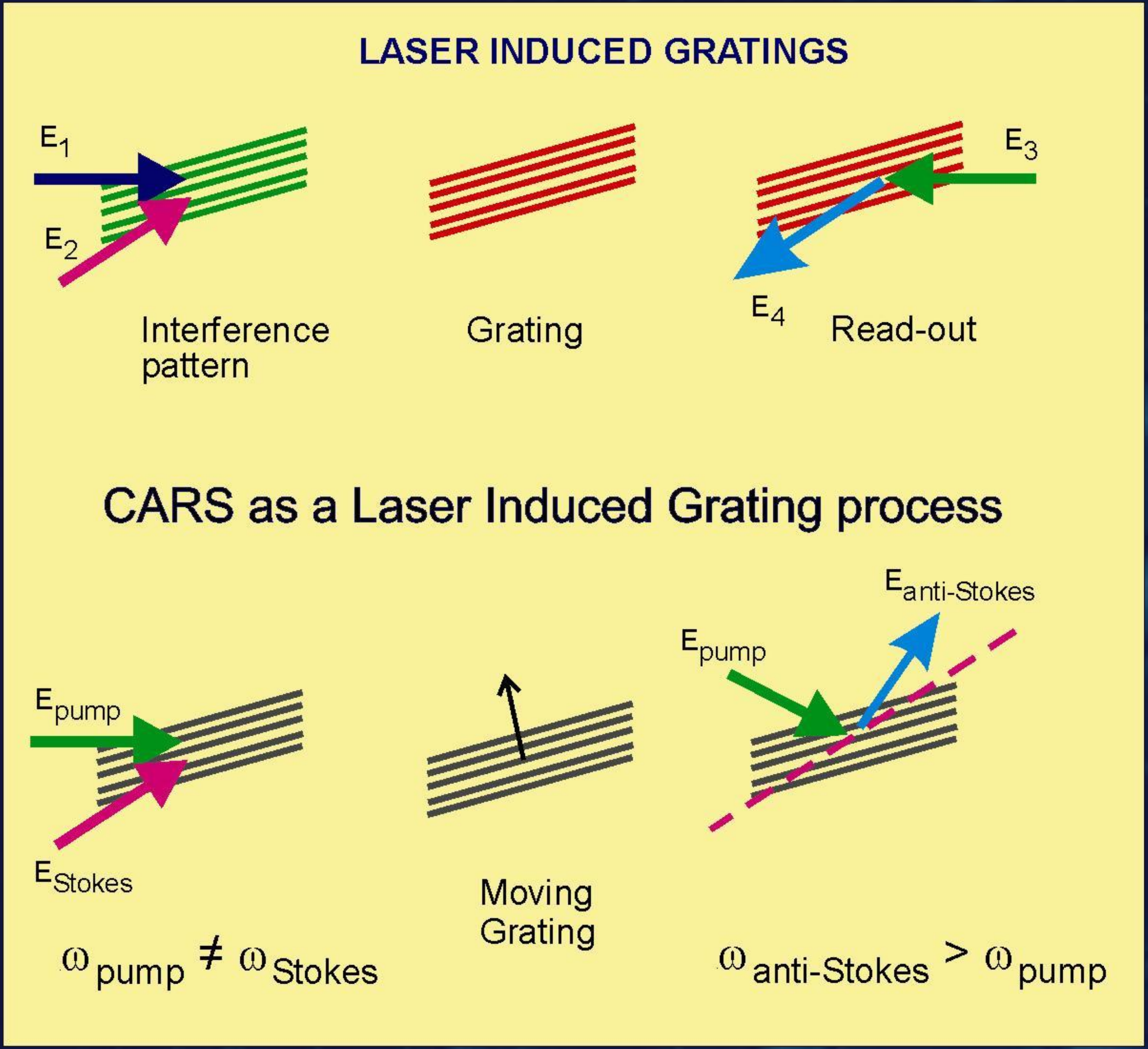
Raman Scattering

- Intra-atomic distance within the molecule is changed when the electron cloud is displaced
- Involves the vibration state of molecules

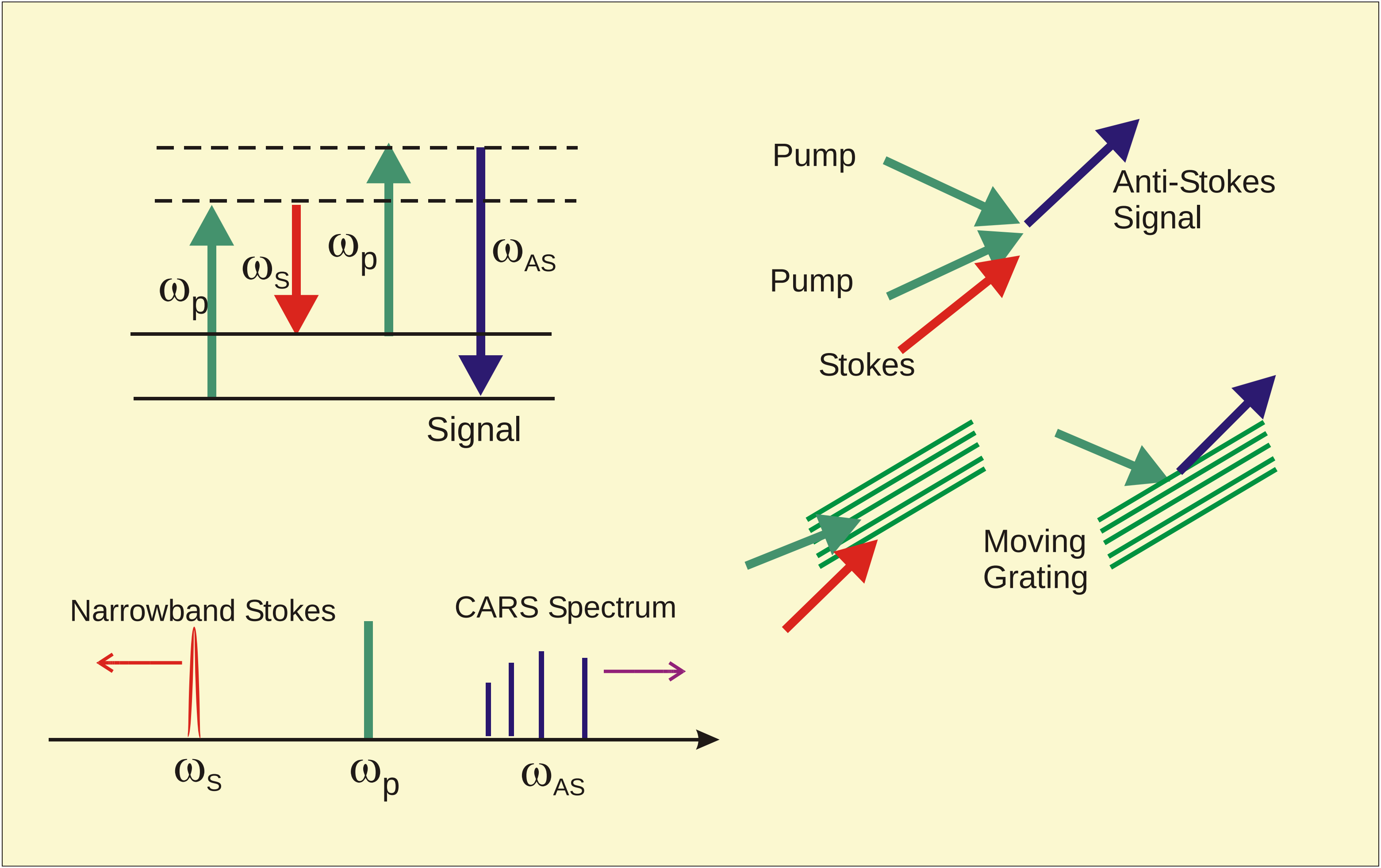


Four Wave Mixing techniques

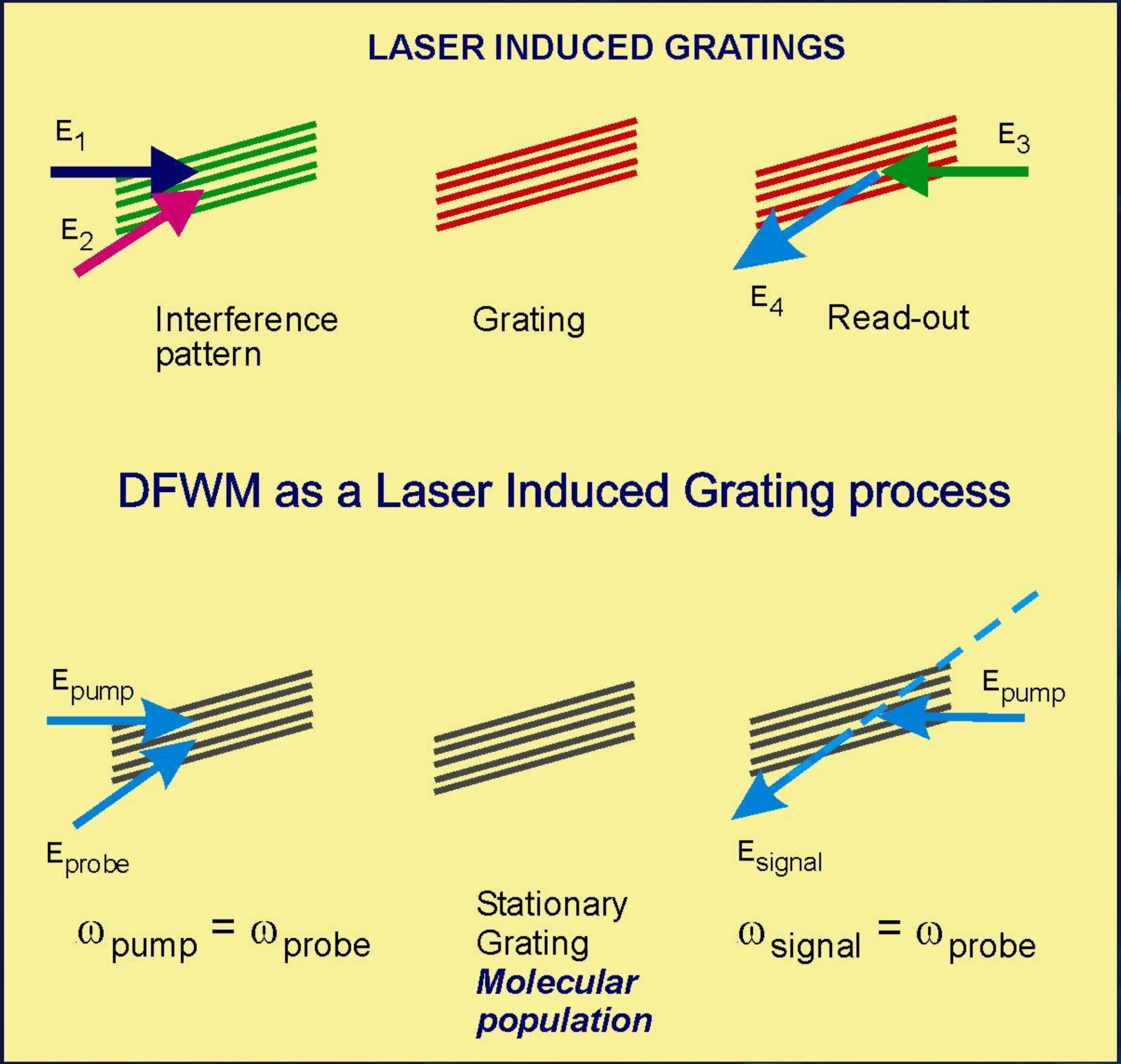
CARS Coherent Anti-Stokes Raman Scattering



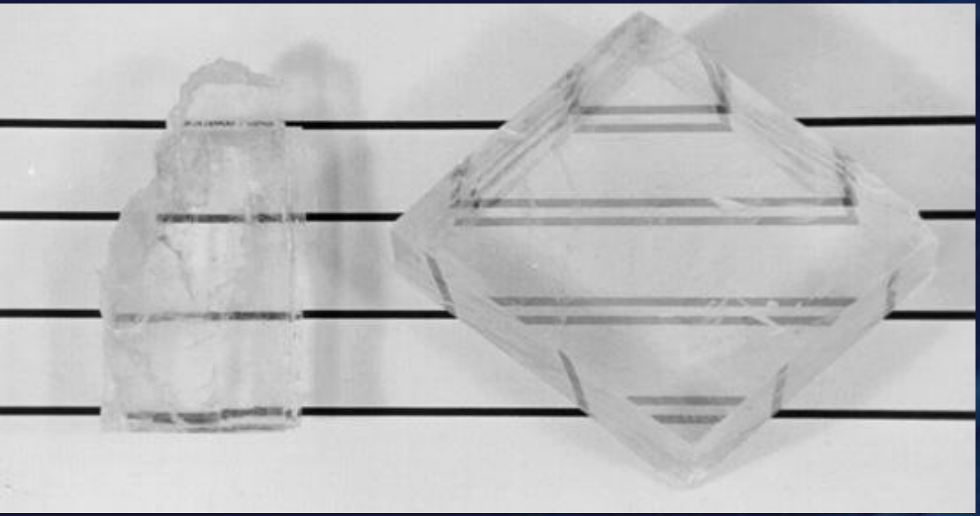
Coherent Anti-Stokes Raman Scattering, CARS: Narrowband



DFWM: Degenerate Four Wave Mixing

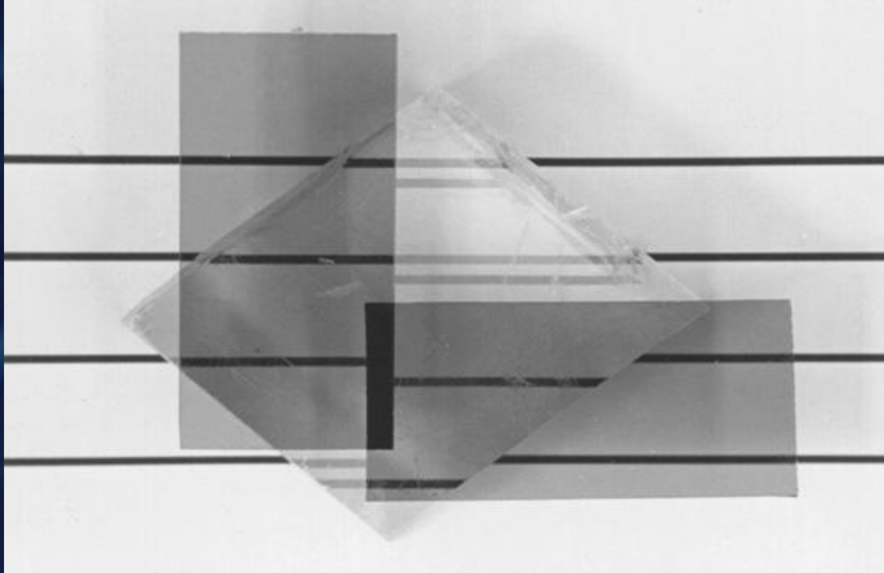


Birefringence



Halite (cubic sodium chloride crystal, optically isotropic)

Calcite (optically anisotropic)



Calcite crystal with two polarizers at right angle to one another

Birefringence was first observed in the 17th century when sailors visiting Iceland brought back to Europe calcite cristals that showed double images of objects that were viewed through them.

This effect was explained by Christiaan Huygens (1629 - 1695, Dutch physicist), as double refraction of what he called an ordinary and an extraordinary wave.

With the help of a polarizer we can easily see what these ordinary and extraordinary beams are.

Obviously these beams have orthogonal polarization, with one polarization (ordinary beam) passing undeflected throught the crystal and the other (extraordinary beam) being twice refracted.

Birefringence

$n^2 = 1 + \chi = \varepsilon$ [1] and $D = \varepsilon \cdot E$ [2]

as n depends on the direction, ε is a tensor

optically isotrop crystal (cubic symmetry) → $n_x = n_y = n_z$ → constant phase delay

uniaxial crystal (e.g. quartz, calcite, MgF_2) → $n_x = n_y \neq n_z$ → Birefringence

extraordinary / optic axis

linear anisotropic media:

$$D_i = \sum_j \varepsilon_{ij} E_j \quad [3]$$

$$\varepsilon_{ij} = \varepsilon_{ji}$$

principal axes coordinate system:

off-diagonal elements vanish,
D is parallel to E

$$D_x = \varepsilon_{11} E_x \qquad D_y = \varepsilon_{22} E_y \qquad D_z = \varepsilon_{33} E_z$$

inverting [3] yields:

$$E = \varepsilon^{-1} D$$

defining

$$\eta = \frac{1}{\varepsilon}$$

in the pricipal coordinate system η is diagonal with principal values:

$$\frac{1}{\varepsilon_i} = \frac{1}{n_i^2} \quad [4]$$

Birefringence

the index ellipsoid

a useful geometric representation is:

the index ellipsoid:

$$\sum_{ij}\eta_{ij}x_i x_j = 1 \qquad [5]$$

is in the principal coordinate system:

$$\frac{x_1^2}{n_1^2}+\frac{x_2^2}{n_2^2}+\frac{x_3^2}{n_3^2}=1 \qquad [6]$$

uniaxial crystals ($n_1=n_2\neq n_3$):

$$\frac{1}{n^2(\theta)}=\frac{\cos^2(\theta)}{n_0^2}+\frac{\sin^2(\theta)}{n_e^2} \qquad [7]$$

$n_a = n_0$ $\qquad$ $n_b = n(\theta)$

$\rightarrow$ $n(0^\circ)=n_0$ $\qquad$ $n(90^\circ)=n_e$

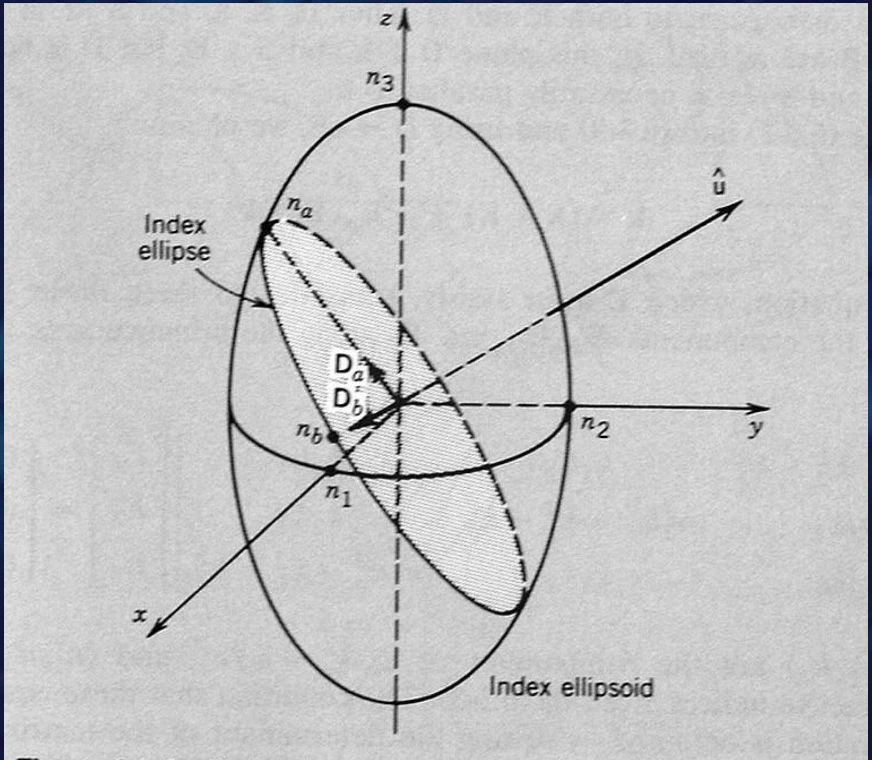


Figure 6.3-6 Normal modes determined from the index ellipsoid.

Birefringence

double refraction

refraction of a wave has to fulfill the **phase-matching condition** (modified Snell's Law):

$$n_{air}\cdot\sin(\theta_1)=n(\theta)\cdot\sin(\theta)$$

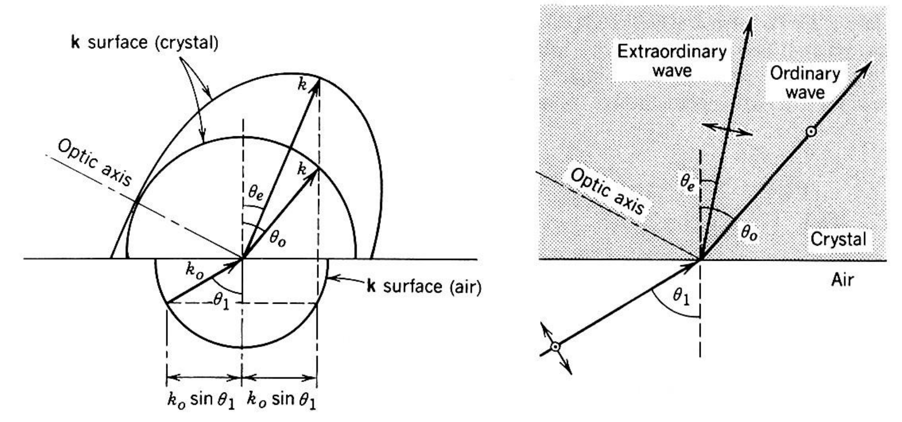


Figure 6.3-13 Determination of the angles of refraction by matching projections of the **k** vectors in air and in a uniaxial crystal.

two solutions do this:

- ordinary wave:

$$n_1\cdot\sin(\theta_1)=n_0\cdot\sin(\theta_0)$$

- extraordinary wave:

$$n_1\cdot\sin(\theta_1)=n(\theta_e)\cdot\sin(\theta_e)$$

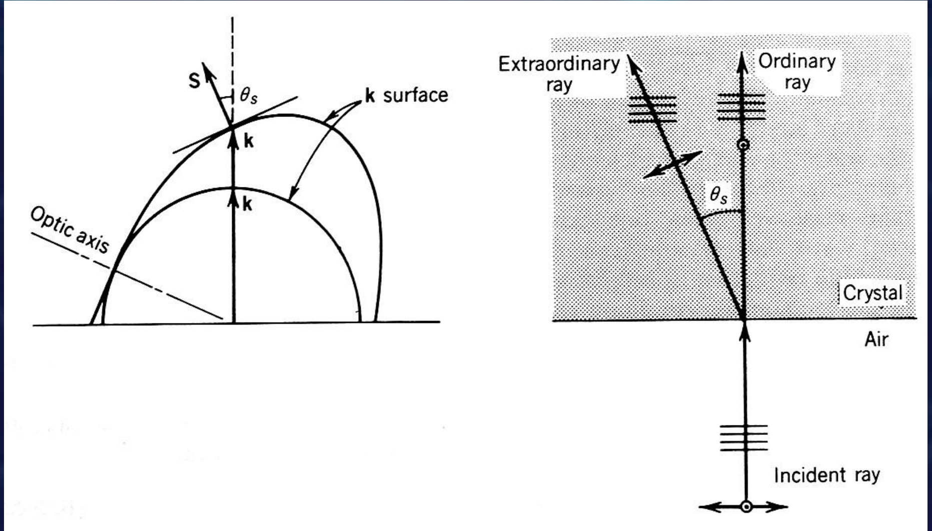


Figure 6.3-14 Double refraction at normal incidence.

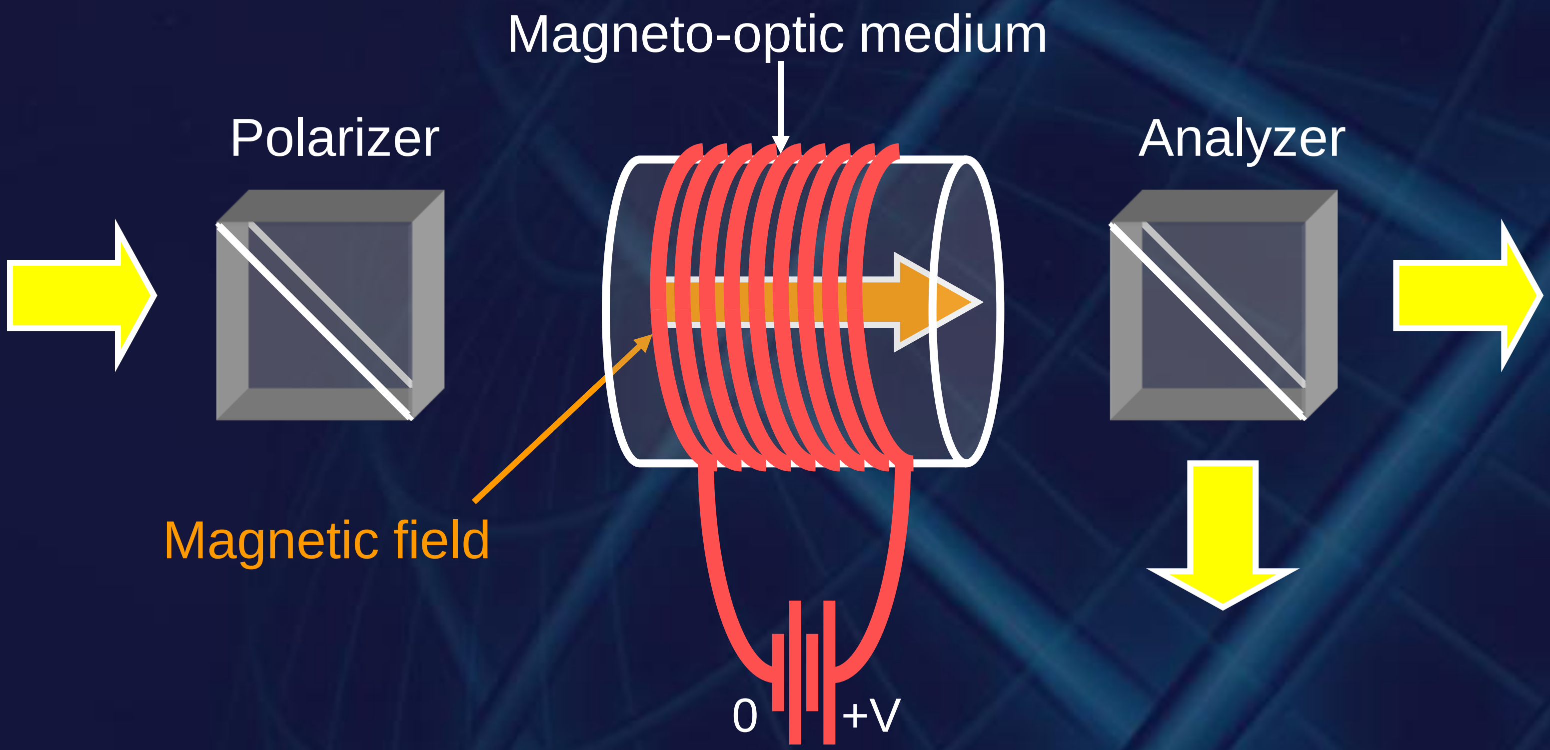
Optical Activity (also called Chirality)

Unlike birefringence, optical activity rotates polarization, but maintains a linear polarization throughout. The polarization rotation angle is proportional to the distance. Optical activity was discovered in 1811 by Arago.

Some substances rotate the polarization clockwise (dextrorotatory) and some produce a counterclockwise rotation (levorotatory).

The Faraday Effect

A magnetic field can induce optical activity.



Magneto-optic medium

Polarizer

Analyzer

Magnetic field

0

+V

The Faraday effect allows control over the polarization rotation.

The Faraday effect: the polarization rotation is proportional to the Verdet constant.

$$\beta = V B d$$

where:

β is the polarization rotation angle,

B is the magnetic field strength,

d is the distance,

V is the **Verdet constant** of the material.

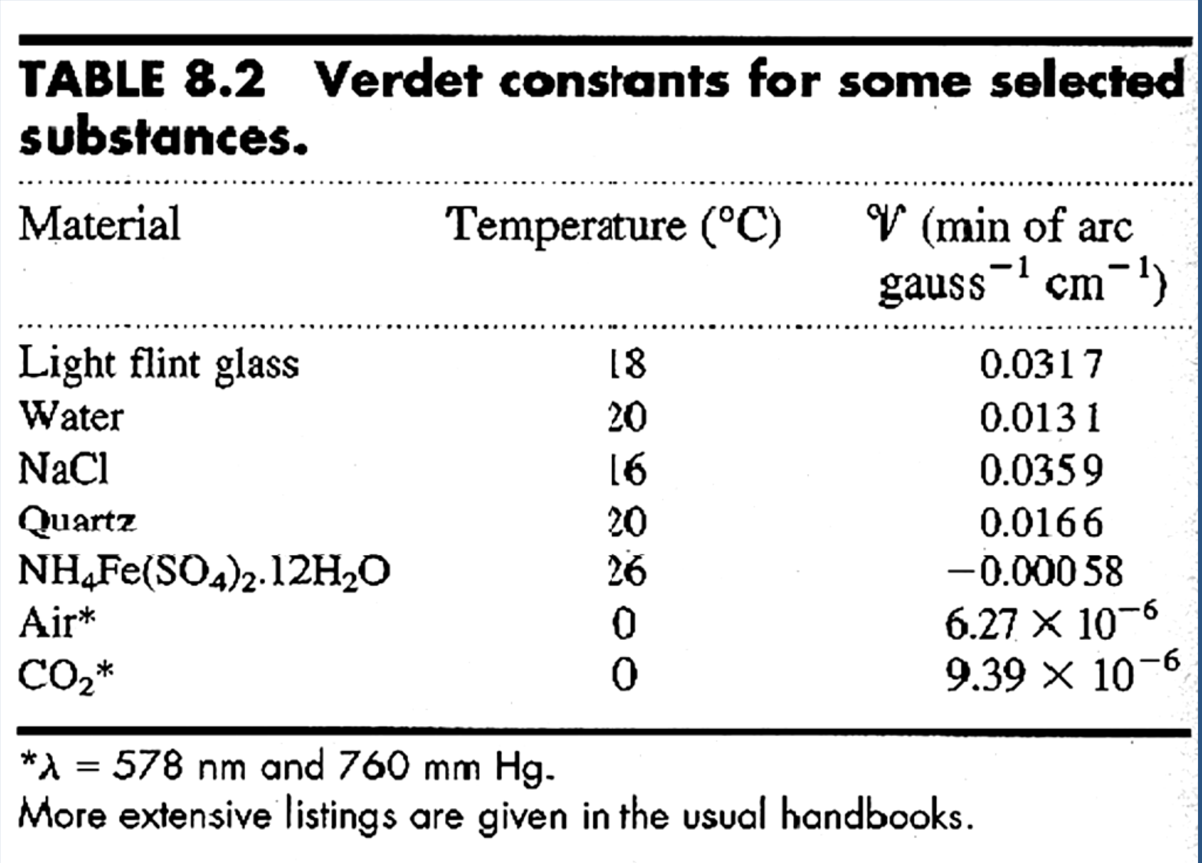


TABLE 8.2 Verdet constants for some selected substances.

Material	Temperature (°C)	$\mathcal{V}$ (min of arc gauss^{-1} cm^{-1})
Light flint glass	18	0.0317
Water	20	0.0131
NaCl	16	0.0359
Quartz	20	0.0166
$NH_4Fe(SO_4)_2.12H_2O$	26	-0.00058
Air*	0	6.27×10^{-6}
CO_2*	0	9.39×10^{-6}

*λ = 578 nm and 760 mm Hg.

More extensive listings are given in the usual handbooks.