

Introduction to nonlinear optics and its applications to laser photonics, integrated optics, and quantum photonics (I)

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Outline

- Background Introduction
- Brief principles of Nonlinear Optics
- Interesting nonlinear-optical devices and phenomena
- Applications

Background Introduction

Development history of nonlinear optics

1877: DC Kerr effect ($\propto \chi^{(3)} E(0)^2 \tilde{E}(\omega)$)

1883: Pockels (linear electro-optic; EO) effect ($\propto \chi^{(2)} E(0) \tilde{E}(\omega)$)

1960: The demonstration of the first laser device (Ruby laser)



1961: The first observation of the second-harmonic generation (SHG) by frequency-doubling the Ruby laser (694.2 nm) with a quartz crystal, two-photon absorption (TPA)



Phys. Rev. Lett., 7, 118 (1961)

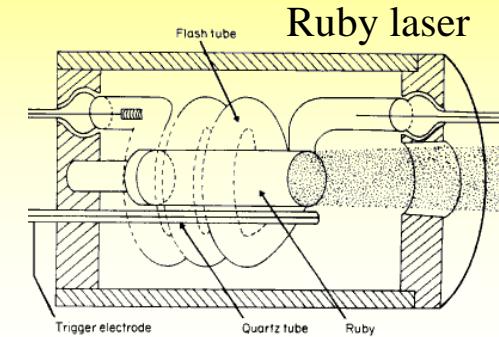
FIG. 1. A direct reproduction of the first plate in which there was an indication of second harmonic. The wavelength scale is in units of 100 Å. The arrow at 3472 Å indicates the small but dense image produced by the second harmonic. The image of the primary beam at 6943 Å is very large due to halation.

1962: Sum frequency generation (SFG), third-harmonic generation (THG), optical rectification (OR), concept of quasi-phase matching (QPM) technique



1962: Stimulated Raman scattering (SRS)

1963: Difference frequency generation (DFG)

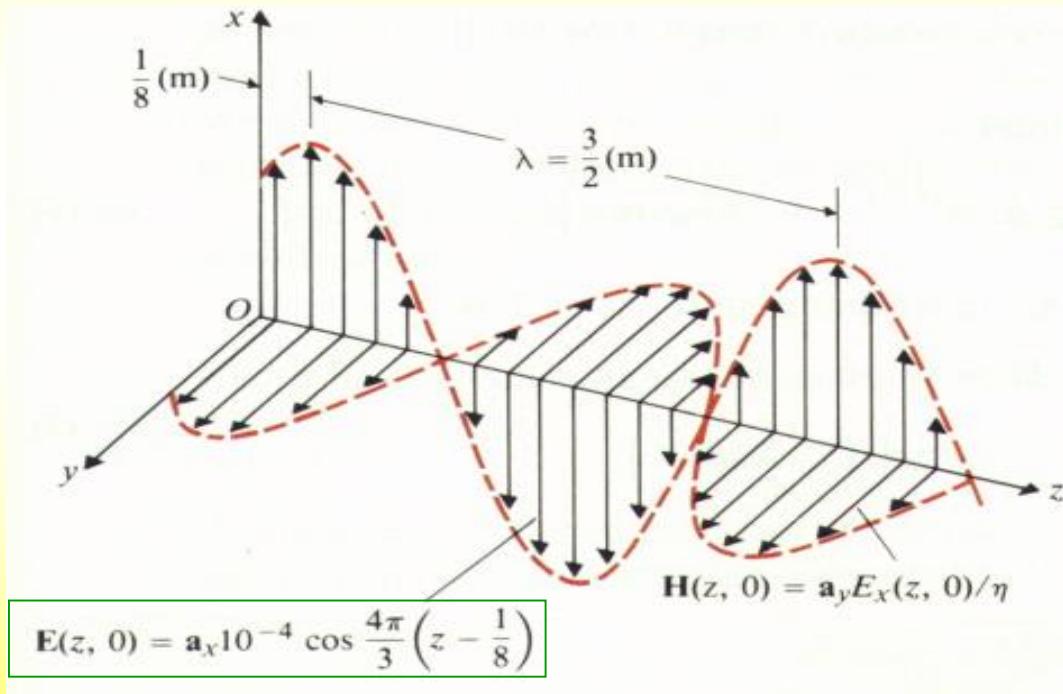


Background Introduction

- 1964: Stimulated Brillouin scattering, self-focusing, saturable absorption
- 1965: Optical parametric generation and oscillation (OPG and OPO)
- 1967: Self-phase modulation
- 1970: Doppler-free two-photon absorption spectroscopy
- 1971: Coherent Raman spectroscopy
- 1976: Inverse Raman spectroscopy, Raman gain spectroscopy, laser polarization spectroscopy
- 1975: Optical bistability
- 1980's: No noticeable new effects or novel phenomena were discovered.
Continuous advanced on applying existing nonlinear-optic techniques to ultra-high resolution spectroscopy, ultra-fast lasers, photorefractive effects, wavelength conversions, optical soliton, quantum optics, etc.
-  1995: Maturity of Quasi-phase matching (QPM) technique (periodically poled QPM crystals; PP-QPM crystals)
- 1995- : Applications in light speed controlling, surface plasmon polaritons, etc.

Background Introduction

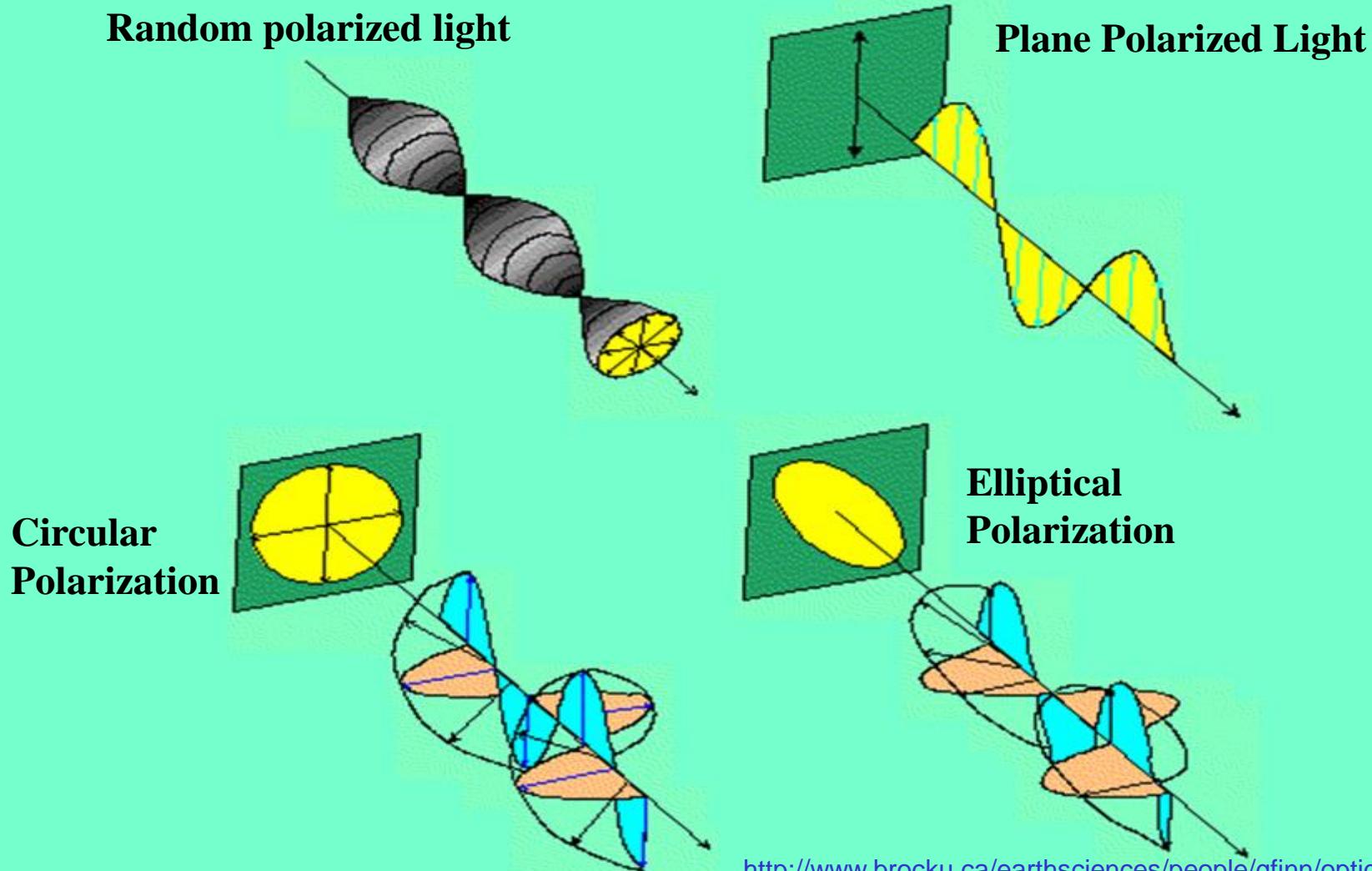
Light is an Electro-Magnetic (EM) wave



The polarization

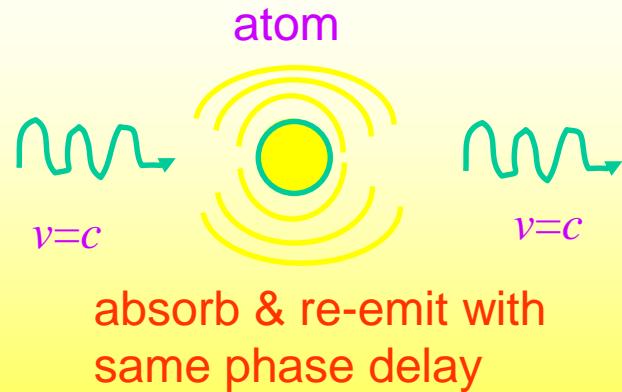
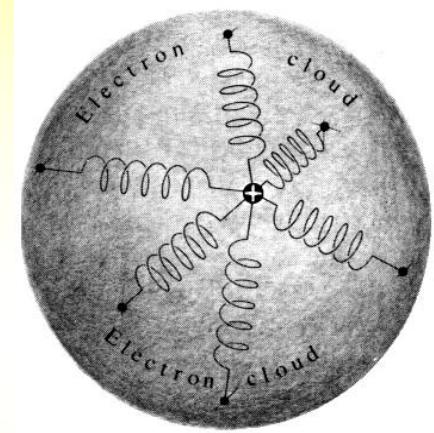
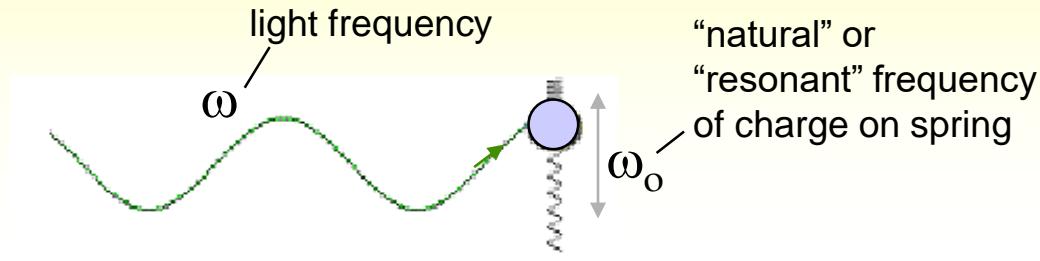
Background Introduction

- Polarization of EM waves



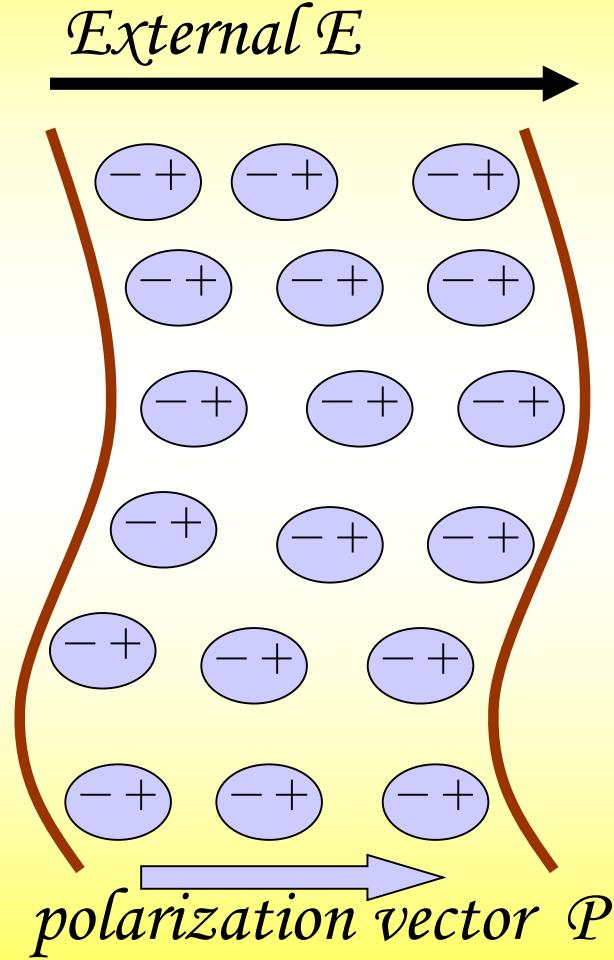
Background Introduction

- “Charge on spring” description of electrons ‘bound’ to atoms in materials



$$v = c/n$$

Background Introduction



$$\vec{D} = \epsilon_0 \vec{E} + \vec{P}$$

$$\vec{D} = \epsilon_0 \vec{E} + \epsilon_0 \chi \vec{E}$$

$$\vec{D} = \epsilon_0 (1 + \chi) \vec{E}$$

$$\vec{D} = \epsilon \vec{E}$$

$$\Rightarrow \epsilon = \epsilon_0 (1 + \chi)$$

$$\Rightarrow n = \left(\frac{\epsilon}{\epsilon_0} \right)^{1/2} = (1 + \chi)^{1/2}$$

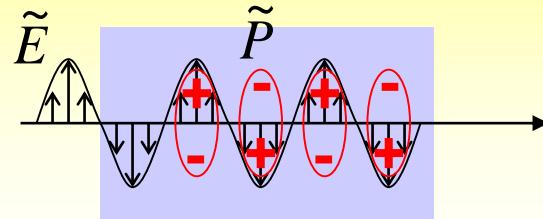
n : refractive index

ϵ : permittivity

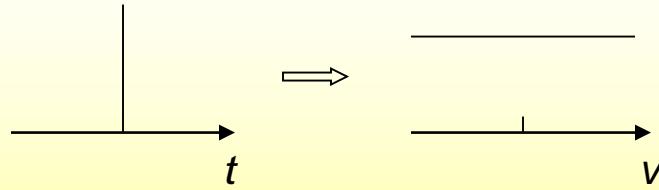
χ : susceptibility

Background Introduction

Interaction of EM waves with dielectric media



- Linear: $\tilde{P}(r,t) \propto \tilde{E}(r,t)$
- Nondispersive: $\tilde{P}(r,t)$ has an instantaneous response to an impulse of $\tilde{E}(r,t)$
- Homogeneous: $\tilde{P}(r,t)$ is independent of the position upon the excitation of $\tilde{E}(r,t)$
- Isotropic: $\tilde{P}(r,t)$ is in parallel with $\tilde{E}(r,t)$

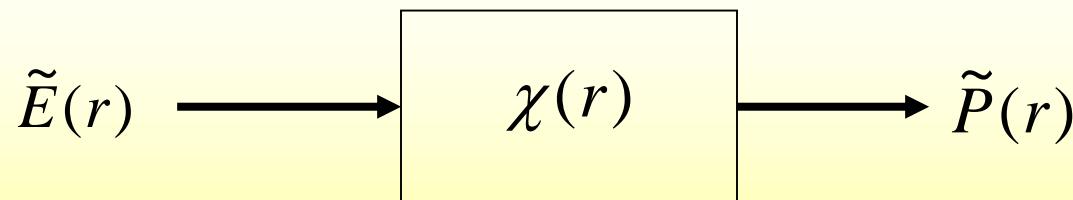


Background Introduction

Interaction of EM waves with dielectric media -- Inhomogeneous only media

The electric susceptibility, and thus the permitivity $\epsilon(r)$ and the refractive index $n(r)$ are position-dependent

$$\tilde{P}(r,t) = \epsilon_0 \chi(r) \tilde{E}(r,t)$$



Background Introduction

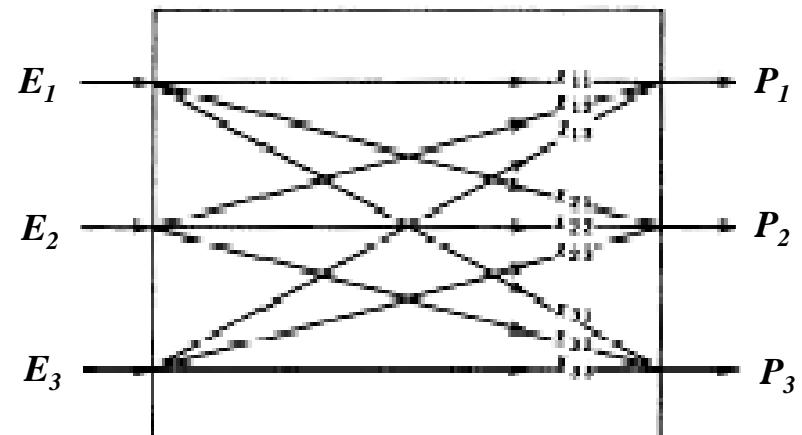
Interaction of EM waves with dielectric media -- Anisotropic only media

The polarization density vector now depends on electric field components that are not parallel to it.

$p_i = \sum_j \epsilon_0 \chi_{ij} E_j$, where χ_{ij} is the susceptibility tensor element.

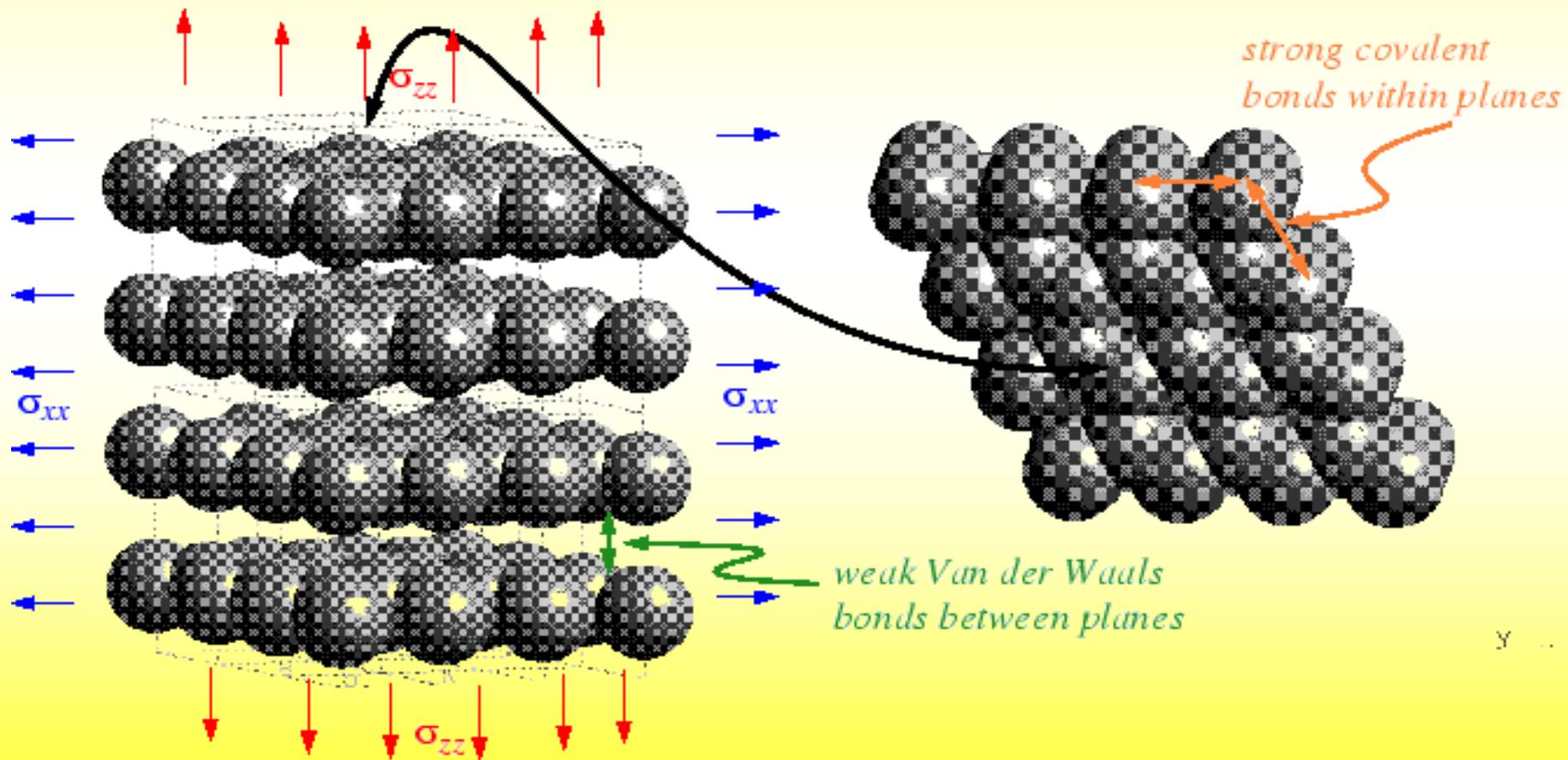
In tensor form,

$$\begin{bmatrix} p_x \\ p_y \\ p_z \end{bmatrix} = \epsilon_0 \begin{bmatrix} \chi_{xx} & \chi_{xy} & \chi_{xz} \\ \chi_{yx} & \chi_{yy} & \chi_{yz} \\ \chi_{zx} & \chi_{zy} & \chi_{zz} \end{bmatrix} \begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix}$$



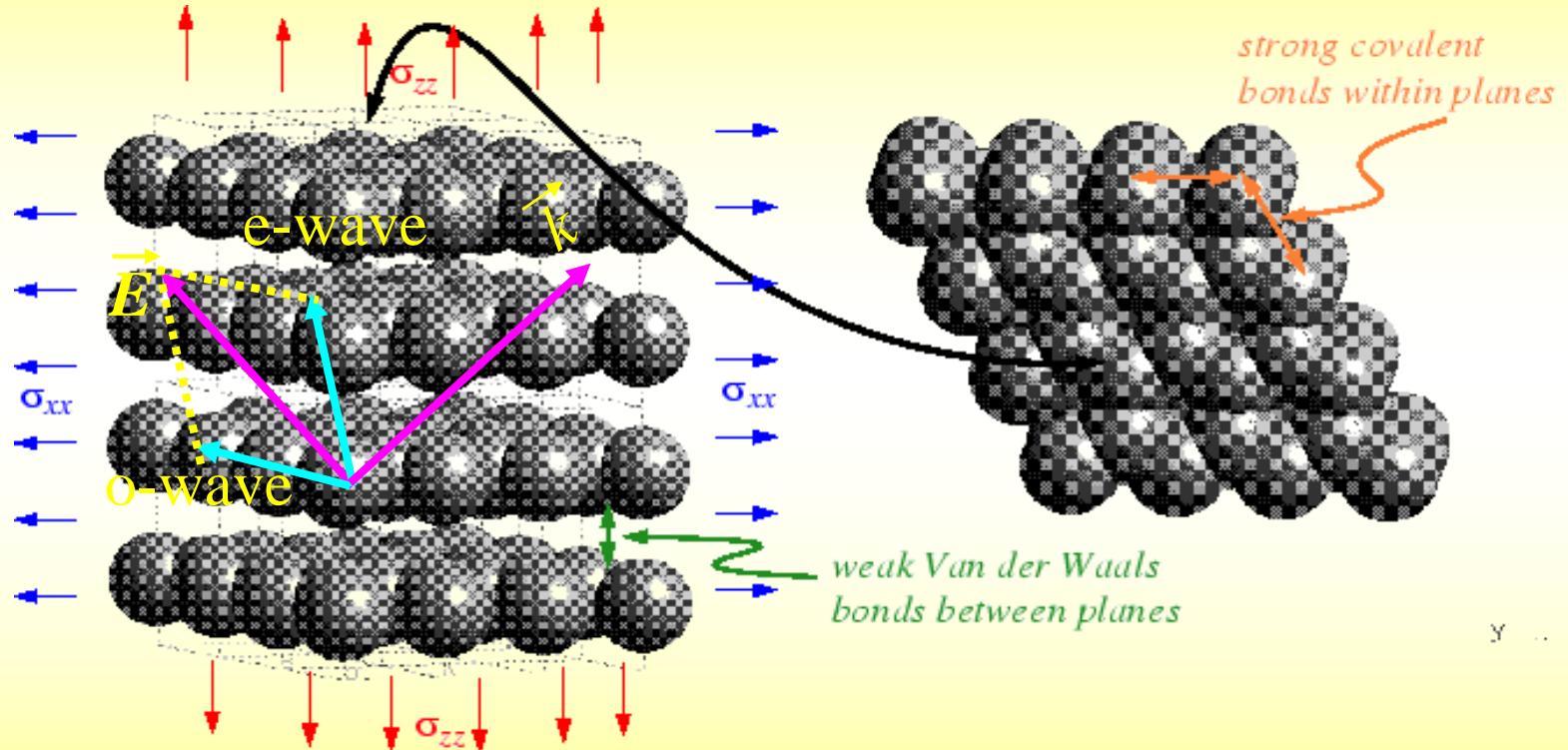
Background Introduction

Interaction of EM waves with dielectric media -- Anisotropic only media



Background Introduction

Anisotropic media

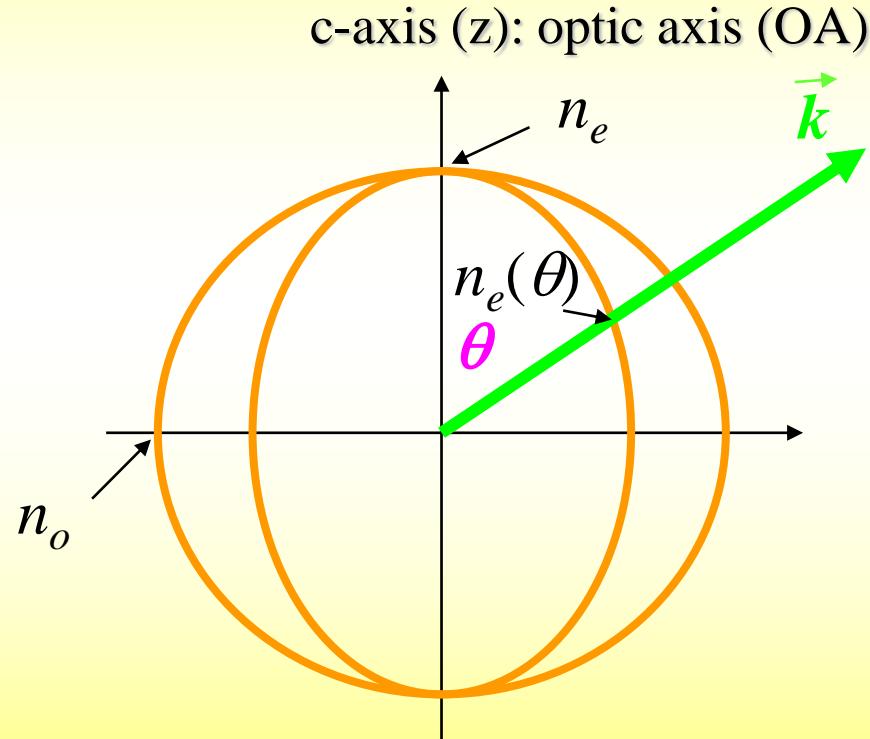


$$\vec{P}_i = \epsilon_0 \chi_i \vec{E}_i$$

$$n_i = (1 + \chi_i)^{1/2}$$

Background Introduction

Birefringence materials



Index ellipse in k space

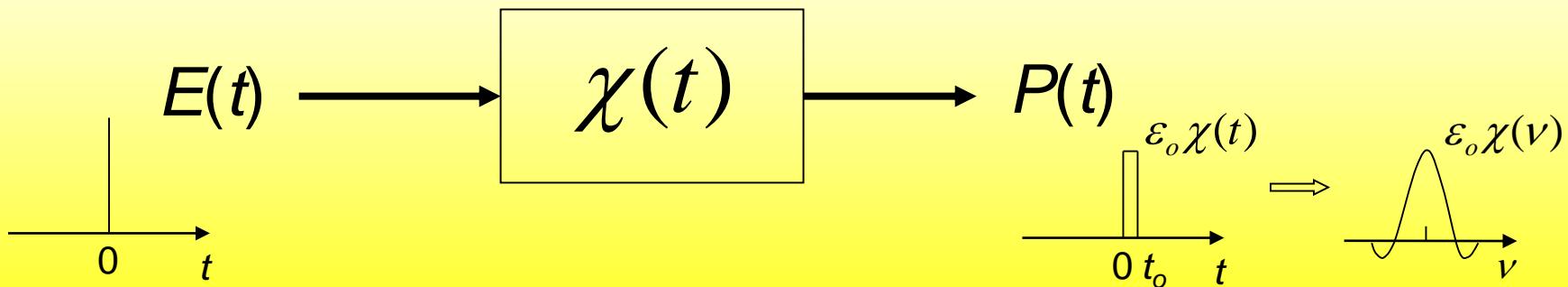
Background Introduction

Interaction of EM waves with dielectric media -- Dispersive only media

$\tilde{P}(r,t)$ has a finite response time upon the input of $\tilde{E}(r,t)$

From a linear system point of view, if $\varepsilon_o\chi(t)$ is the impulse response of the polarization density due to an impulse electric field excitation $E(t)=\delta(t)$, the polarization density due to an arbitrary excitation $E(t)$ is then the convolution

$$P(t) = \varepsilon_o \int_{-\infty}^{\infty} \chi(t-t') E(t') dt'$$



Background Introduction

Interaction of EM waves with dielectric media -- Nonlinear only media

The polarization density function is a nonlinear function of E :

$$p(t) = a_1 E + a_2 E^2 \dots = \epsilon_0 \chi E + p_{NL} = p_L + p_{NL}$$

Using the relation $\vec{D} = \epsilon_0 \vec{E} + \vec{p}$ and the vector identity

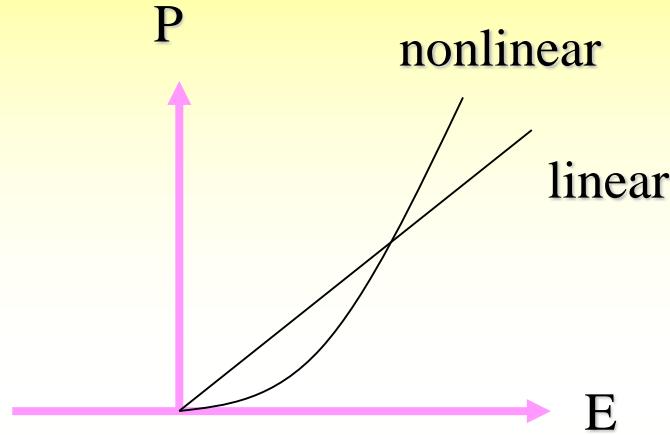
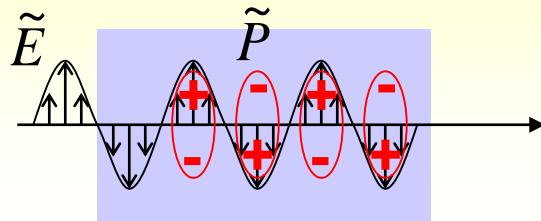
$\nabla \times \nabla \times \vec{E} = \nabla(\nabla \cdot \vec{E}) - \nabla^2 \vec{E}$, we can obtain the wave equation for a nonlinear media

$$\underbrace{\nabla^2 \vec{E} - \frac{1}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2}}_{\text{Free-space wave equation}} = \mu_0 \underbrace{\frac{\partial^2 \vec{p}_{NL}}{\partial t^2}}_{\text{Driving force (source) term}}, \text{ where } c = \frac{c_0}{\sqrt{1 + \chi}}$$

Free-space wave equation

Driving force (source) term

Brief principles of Nonlinear Optics



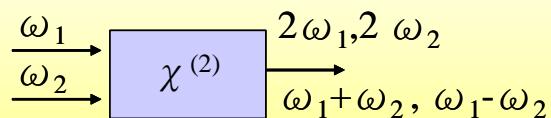
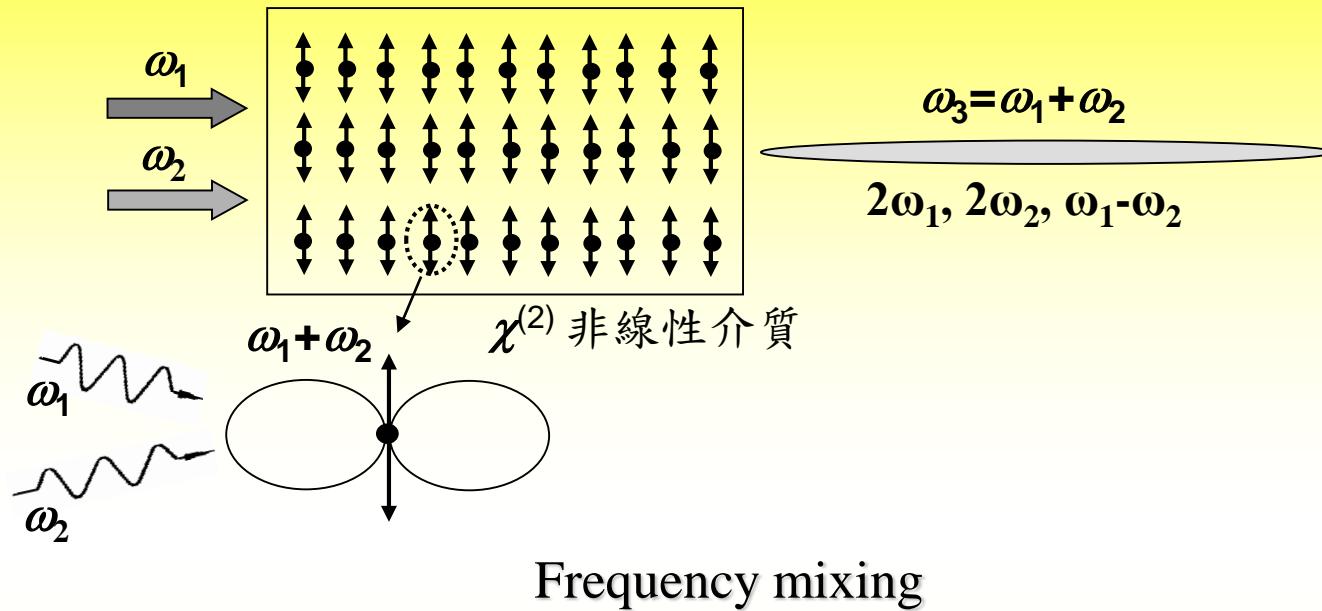
In linear optics

$$\tilde{P} = \epsilon_0 \chi^{(1)} \tilde{E}$$

In nonlinear optics

$$\tilde{P} = \epsilon_0 \{ \chi^{(1)} \tilde{E} + \chi^{(2)} \tilde{E}^2 + \chi^{(3)} \tilde{E}^3 + \dots \}$$

Brief principles of Nonlinear Optics



$$E = E_1 \cos \omega_1 t + E_2 \cos \omega_2 t$$

$$P_{NL} = \epsilon_0 \chi^{(2)} \{E_1 \cos \omega_1 t + E_2 \cos \omega_2 t\}^2$$

$$= \epsilon_0 \chi^{(2)} \left\{ \frac{1}{2} [1 + \cos 2\omega_1 t] E_1^2 \right\} \text{rectification}$$

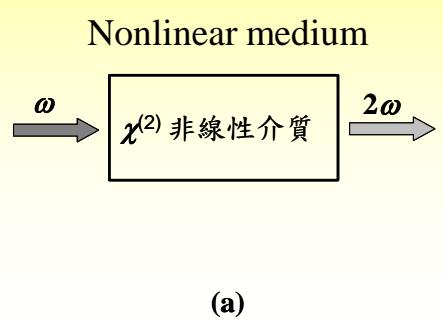
$$+ \frac{1}{2} [1 + \cos 2\omega_2 t] E_2^2 \text{ Second harmonic generation}$$

$$+ \frac{1}{2} \cos(\omega_1 t + \omega_2 t) E_1 E_2 + \frac{1}{2} \cos(\omega_1 t - \omega_2 t) E_1 E_2$$

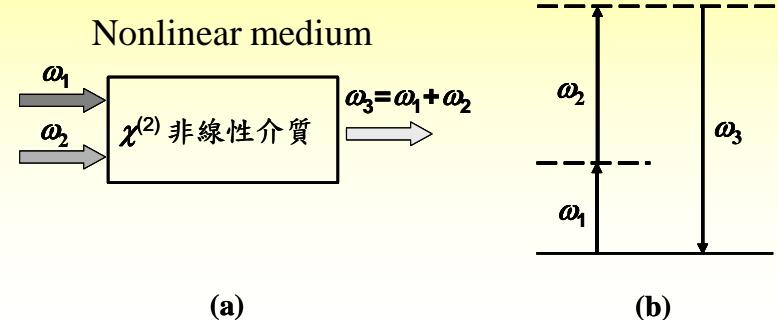
Sum frequency Difference frequency

Brief principles of Nonlinear Optics

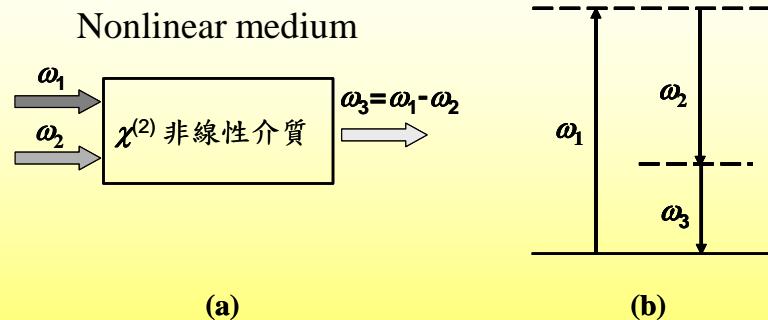
2nd order nonlinear wavelength conversion processes



Second harmonic generation (SHG)



Sum frequency generation (SFG)



Difference frequency generation (DFG)



Optical parametric oscillator (OPO)

Brief principles of Nonlinear Optics

★ Frequency conversion essential I: nonlinear material with reasonable large nonlinearity

$$\begin{aligned}\tilde{P} = \varepsilon_0 & \{ \chi^{(1)} \tilde{E} \\ & + \chi^{(2)} \tilde{E}^2 + \chi^{(3)} \tilde{E}^3 + \dots \}\end{aligned}$$

$$\varepsilon_0 \chi^{(1)} E \approx \varepsilon_0 \chi^{(2)} E^2 \approx \varepsilon_0 \chi^{(3)} E^3 \rightarrow E \sim 1/\chi^{(2)} \sim \sqrt{1/\chi^{(3)}} \sim 10^{11-12} V/m$$

$P > TW/cm^2 !!! \Rightarrow$ Only a tightly focused pulsed laser can achieve such a value!

Brief principles of Nonlinear Optics

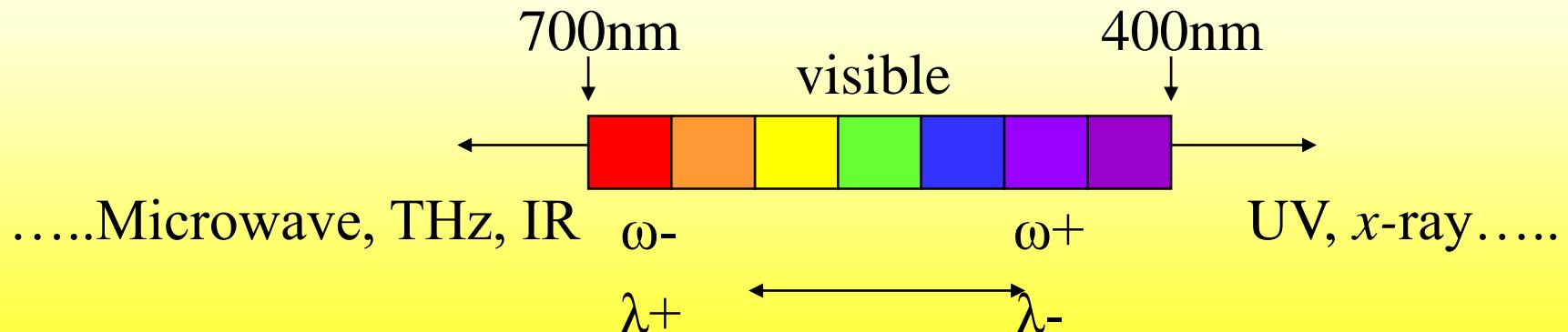
★ Frequency conversion essential II: energy conservation

$$\omega_3 = \omega_1 + \omega_2 \text{ or } \lambda_3^{-1} = \lambda_1^{-1} + \lambda_2^{-1}$$

Second harmonic generation (SHG): $\omega_3 = \omega_1 + \omega_1 = 2\omega_1$

Sum frequency generation (SFG): $\omega_3 = \omega_1 + \omega_2$

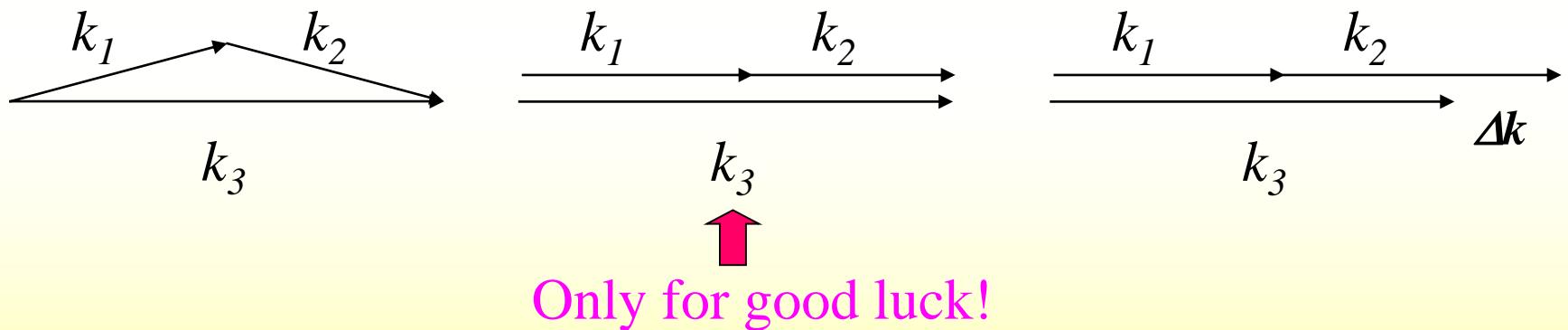
Difference frequency generation (DFG): $\omega_2 = \omega_1 - \omega_3$



Brief principles of Nonlinear Optics

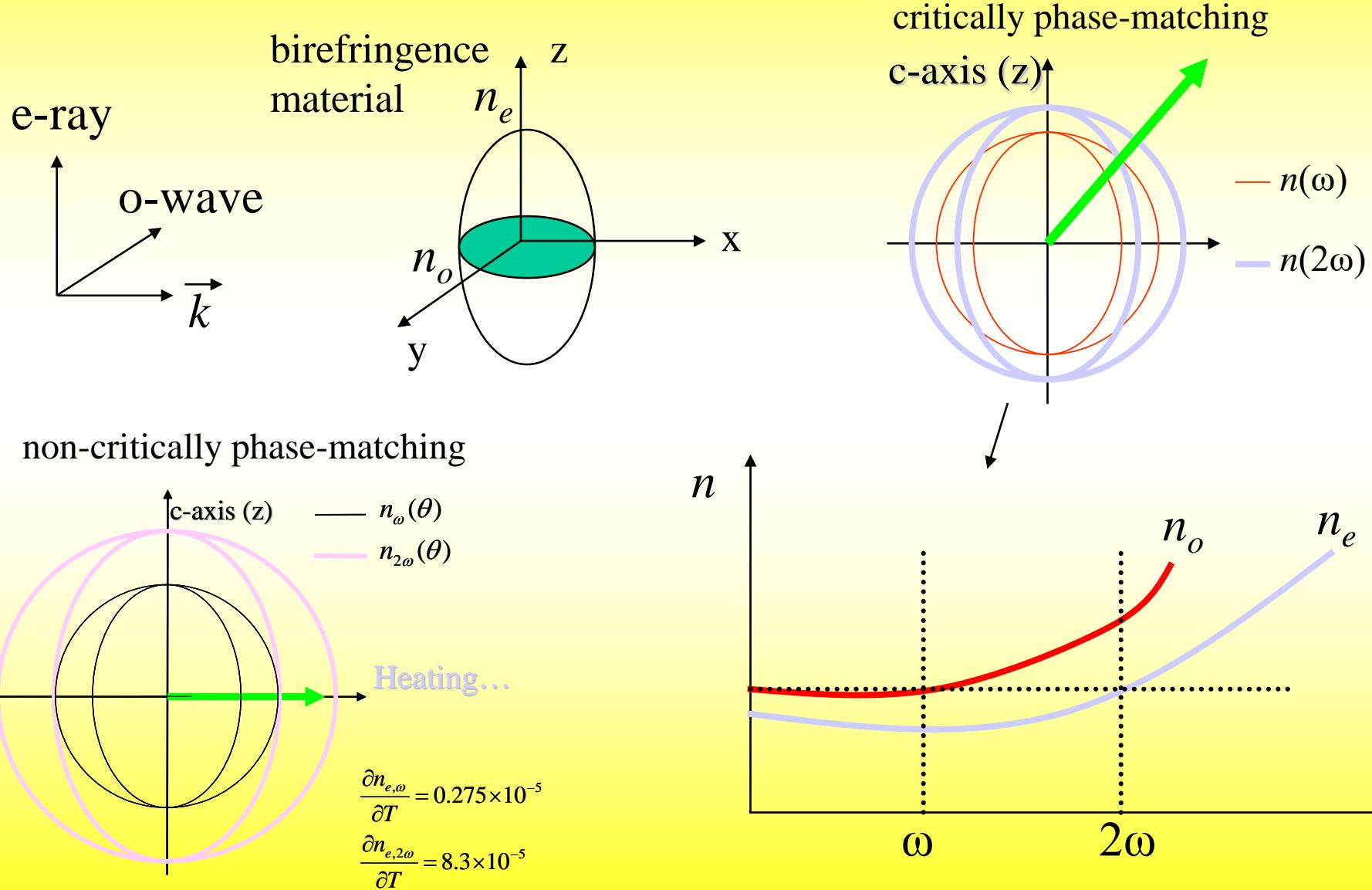
★ Frequency conversion essential III: momentum conservation or Phase matching

$$\Delta k = k_1 + k_2 - k_3 = 0 \quad \text{or} \quad n_1\omega_1 + n_2\omega_2 = n_3\omega_3 \quad (\text{if collinearly})$$



For SHG, $\omega_1=\omega_2, \omega_3=2\omega_1 \rightarrow k_{2\omega} - 2k_\omega = 0 \Rightarrow n(2\omega) = n(\omega) !?$

Brief principles of Nonlinear Optics



Brief principles of Nonlinear Optics

Disadvantages of conventional Birefringence phasematching

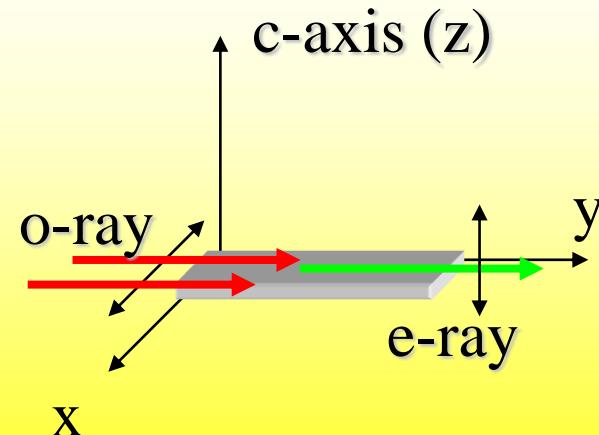
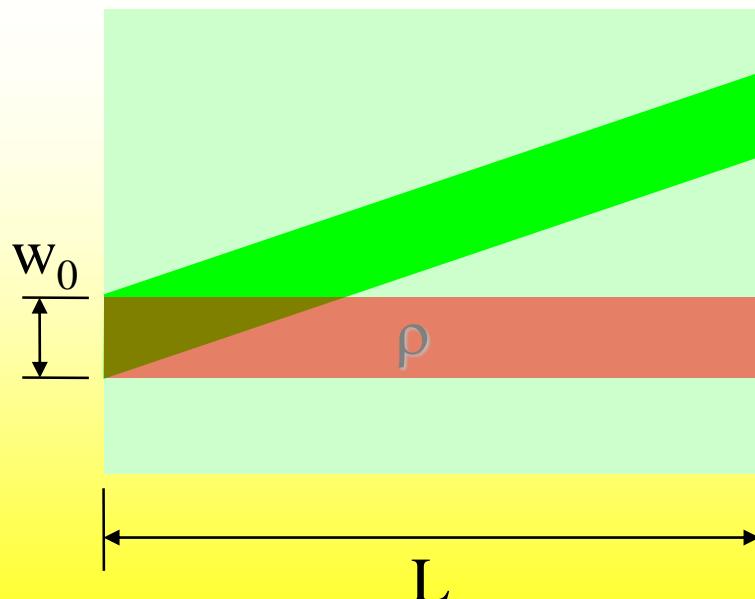
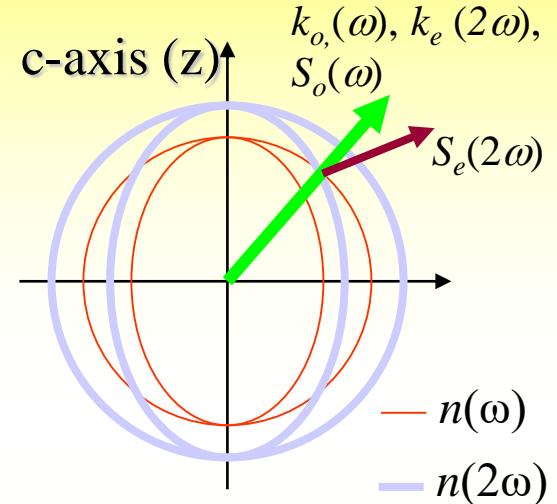
1. Poynting vector walk-off
2. Critical phasematching angle

e.g. $\lambda=1\mu\text{m}$, $L=1\text{cm}$, $\theta=45^\circ \Rightarrow \Delta\theta=0.6\text{mrad}=0.02^\circ$

3. Low accessible nonlinear coefficient

e.g. LiNbO₃ $d_{31}=d_{zxx}=4.7\text{pm/V}$ (ooe)

cp. $d_{33}=d_{zzz}=27\text{pm/V}$ (eee)



Brief principles of Nonlinear Optics

Quasi-phasematching (QPM)

$$k_1 \quad \quad k_2 \\ \hline \quad \quad \quad K = \frac{2\pi}{\Lambda} \\ k_3$$

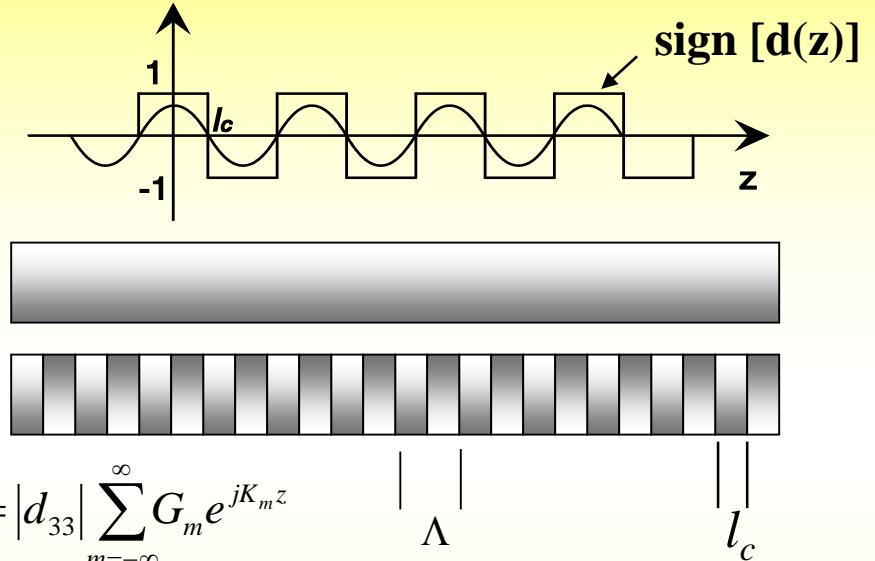
Phase compensation

$$\frac{dE_{2\omega}}{dz} \propto d(z) |E_\omega|^2 e^{-j\Delta kz}$$

$$= \frac{2}{\pi} |d_{33}| |E_\omega|^2 [e^{-j(\Delta k - \frac{2\pi}{\Lambda})z} - \frac{1}{3} e^{-j(\Delta k - \frac{2\pi}{\Lambda} \times 3)z} + \frac{1}{5} e^{-j(\Delta k - \frac{2\pi}{\Lambda} \times 5)z} + \dots]$$

first-order third-order fifth-order

$$\Delta k_{QPM}(\lambda, T, \theta) = k_{2\omega} - 2k_\omega - \frac{2m\pi}{\Lambda}$$



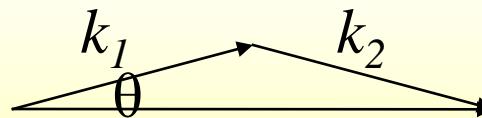
Brief principles of Nonlinear Optics

Quasi-phasematching (QPM)

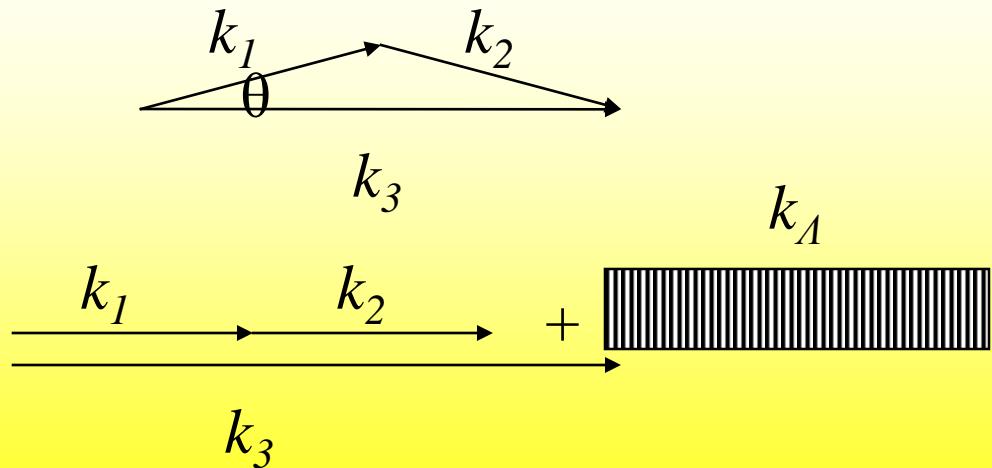
Advantages:

- (1) Noncritical phase-matching with engineerable QPM gratings → full usage of the wide spectral application range (PPLN: 0.4-4.5μm)
- (2) No walk-off problem
- (3) Access of the best nonlinear coefficient, d_{33}

Critical phase-matching:

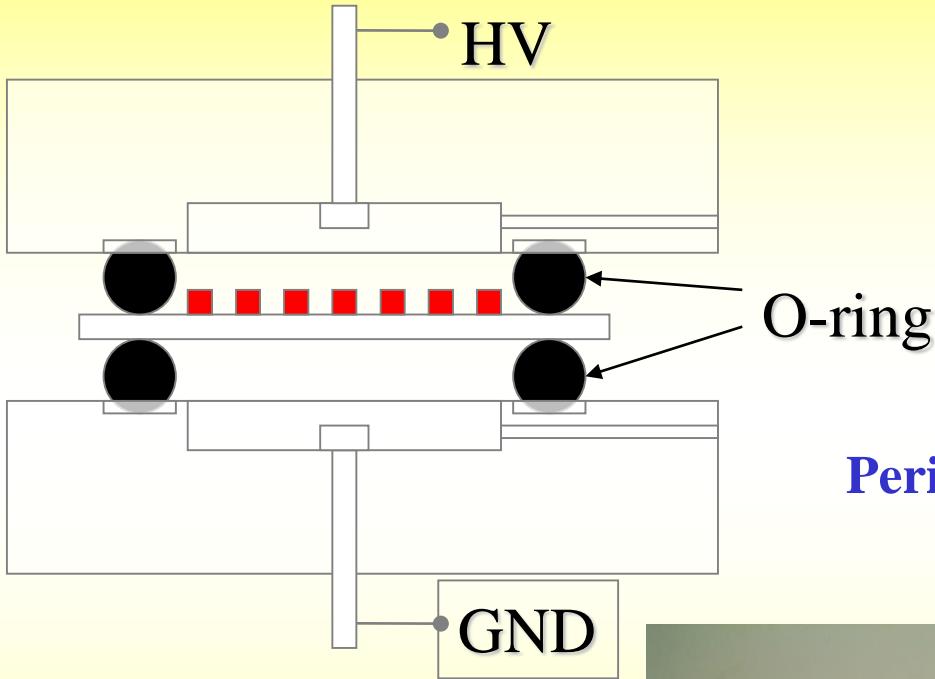


Quasi-phase-matching:

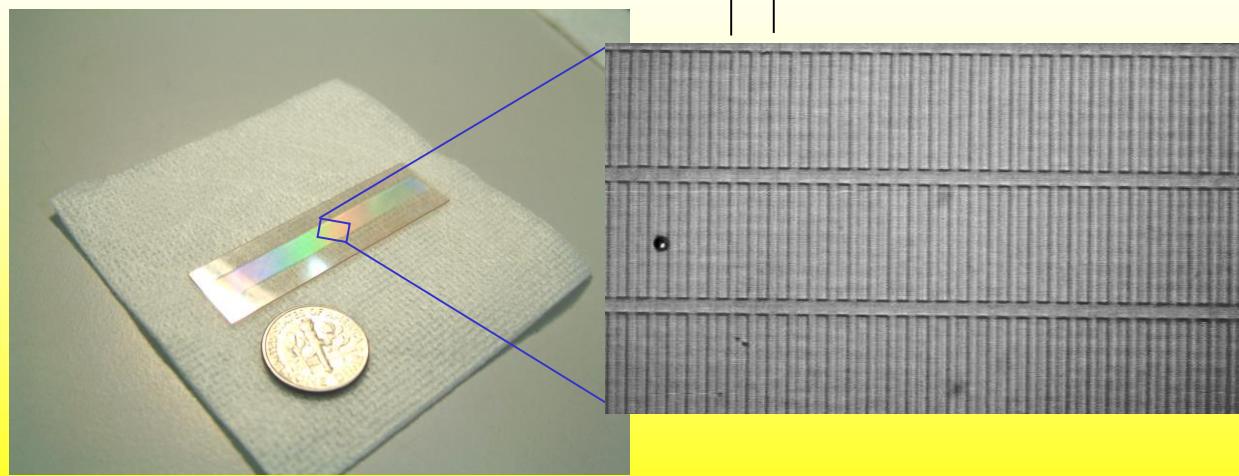


Brief principles of Nonlinear Optics

Quasi-phase-matching (QPM) technology



Periodically Poled Lithium Niobate (PPLN)

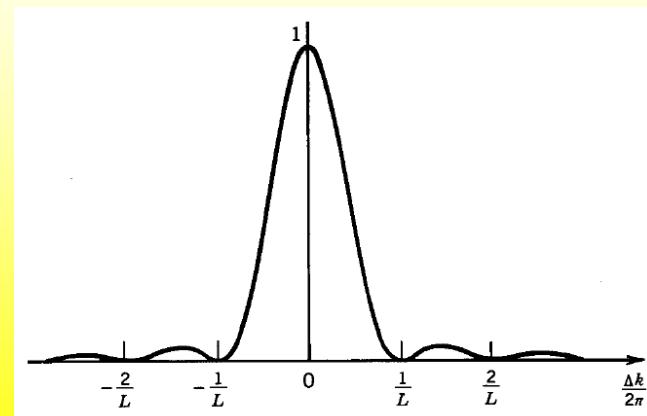


Brief principles of Nonlinear Optics

QPM Materials: LiNbO₃ and KTP

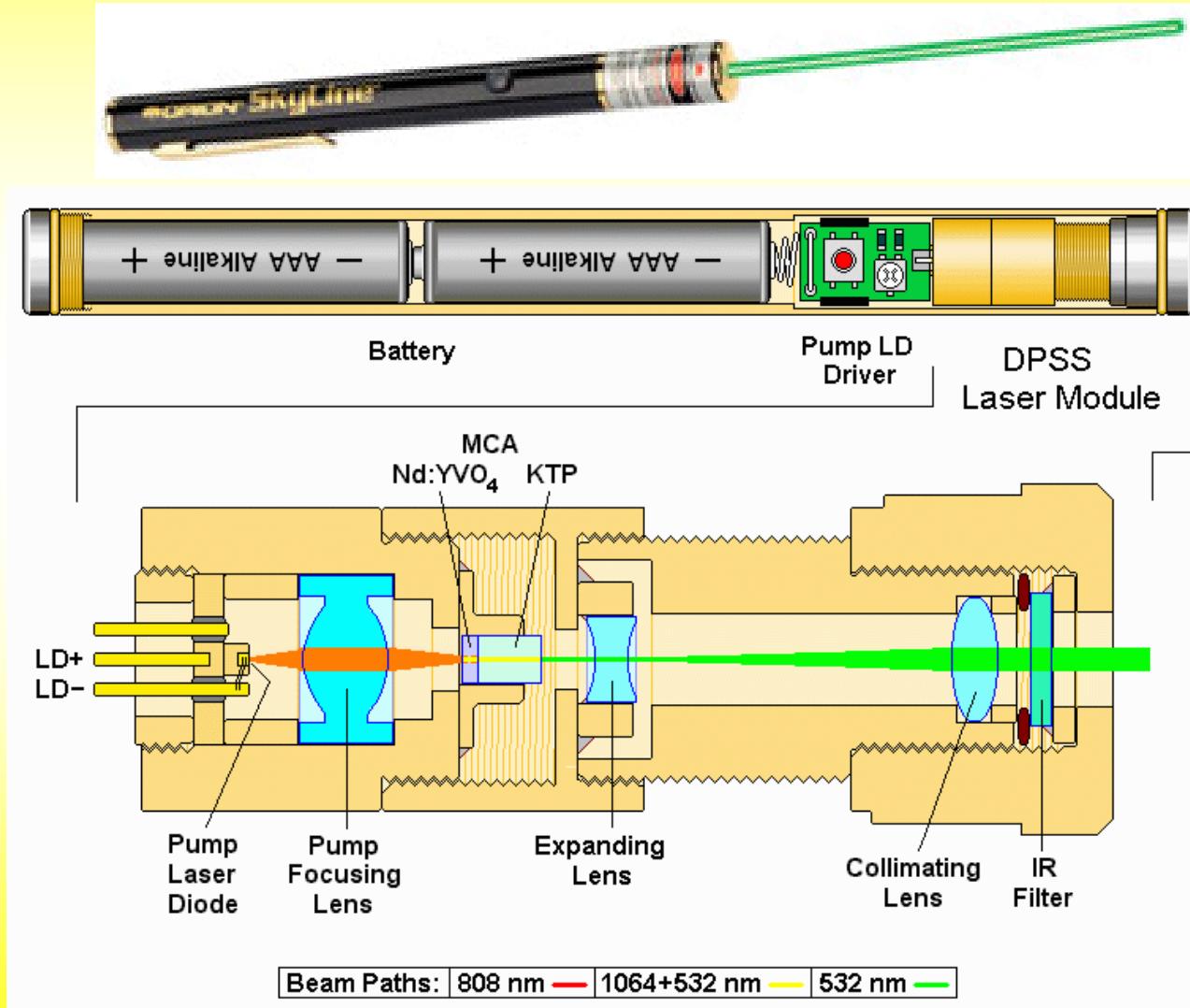
	LiNbO ₃	KTP
transparency	400~>4000 nm	350~4500 nm
d_{eff} for QPM	17.6 pm/V	9.5 pm/V
coercive field	21 kV/mm	2 kV/mm
common thickness	0.5 mm	1 mm
T_c	1200 °C	936 °C
damage threshold	700 MW/cm²	1~10 GW/cm²

$$\eta_{SHG} \propto d_{eff}^2 I_{pump} L^2 \sin c^2(\Delta kx / 2)$$



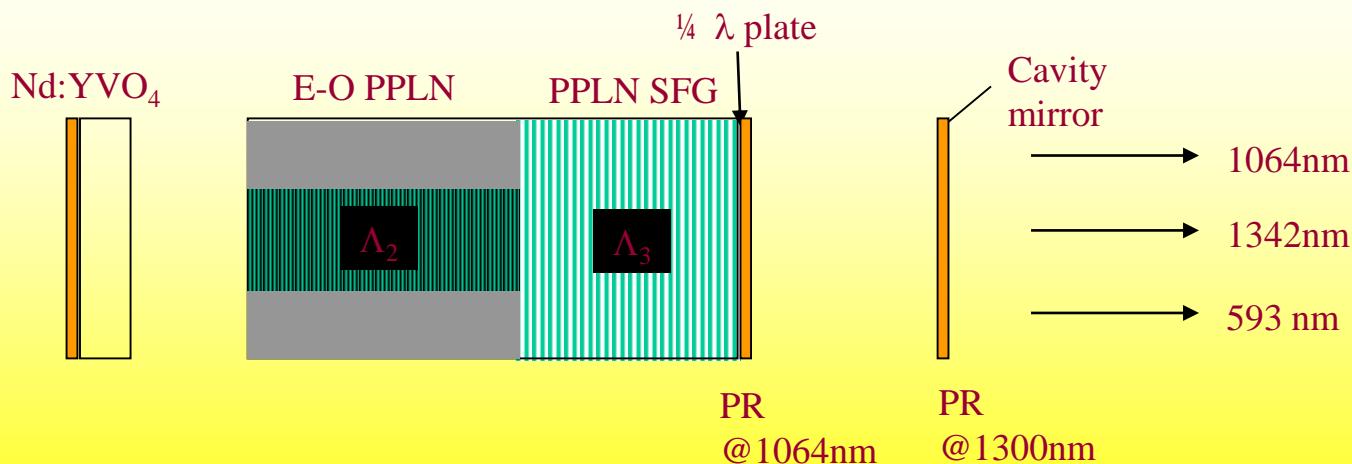
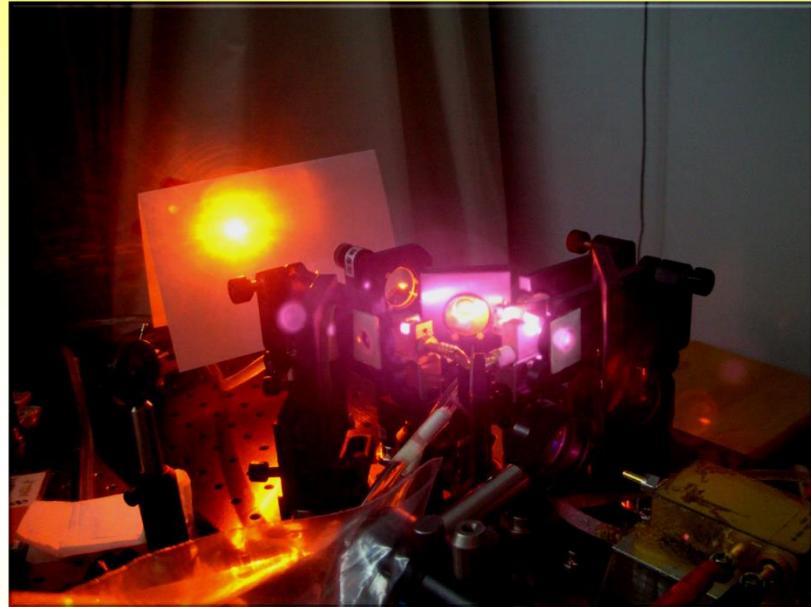
Interesting nonlinear-optical devices

Green laser pointer



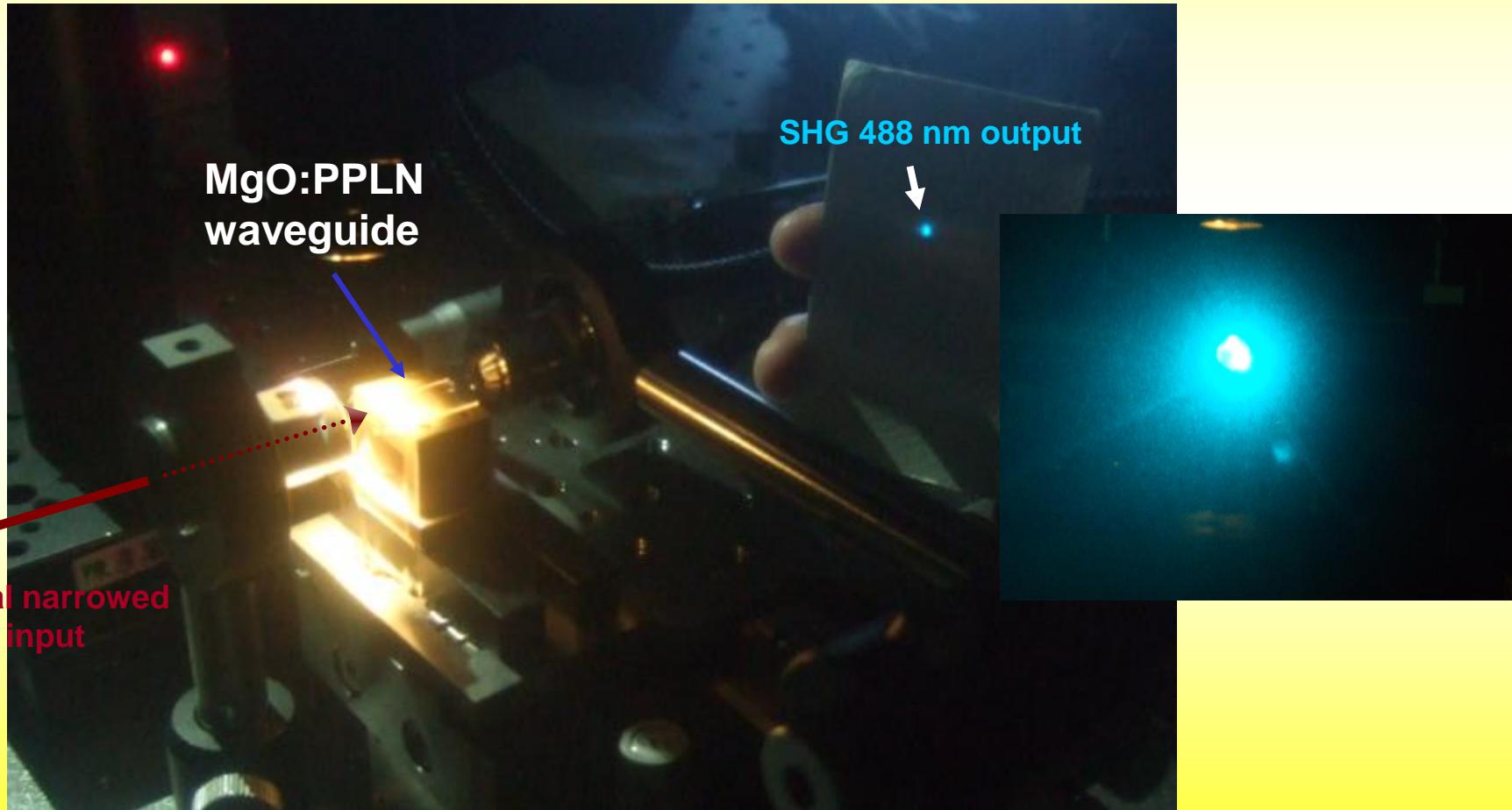
Interesting nonlinear-optical devices

Yellow-orange lasers

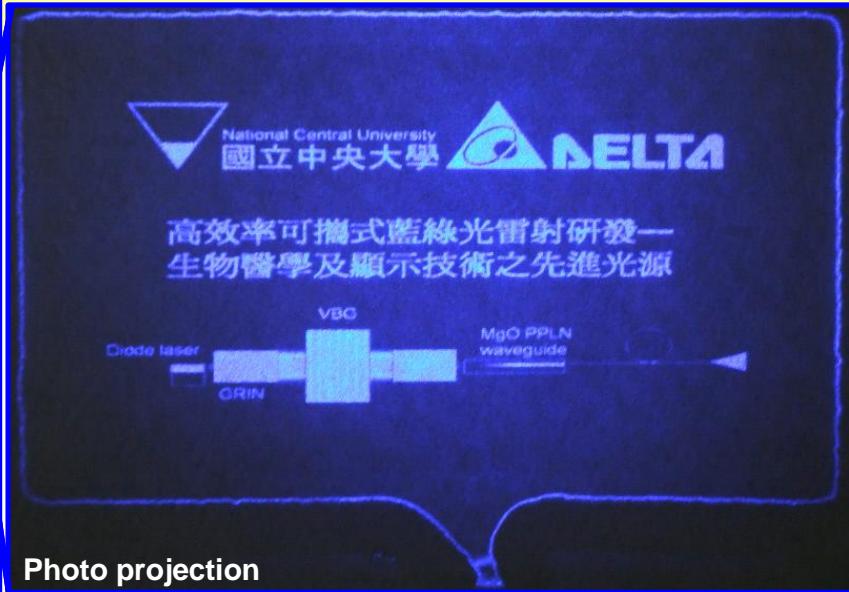
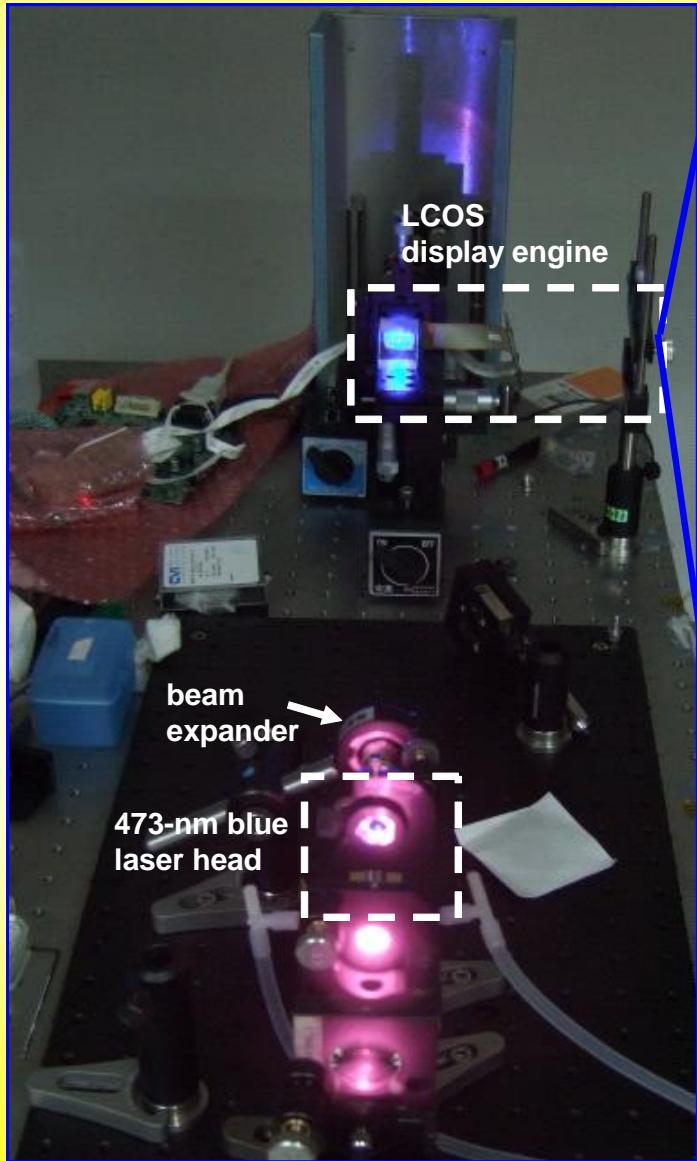


Interesting nonlinear-optical devices

Blue waveguide laser



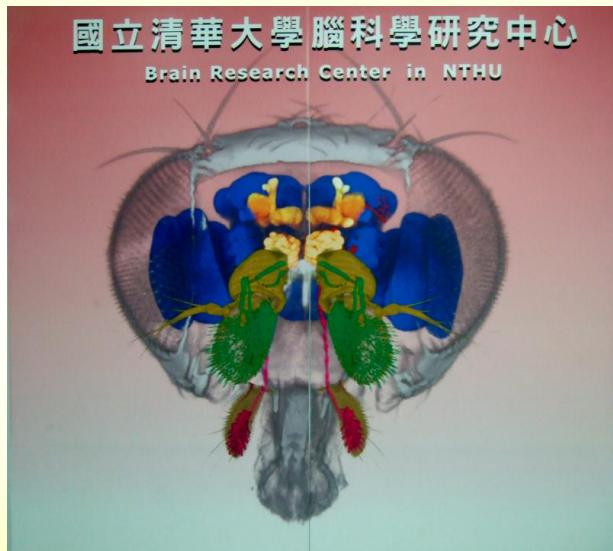
Interesting nonlinear-optical devices



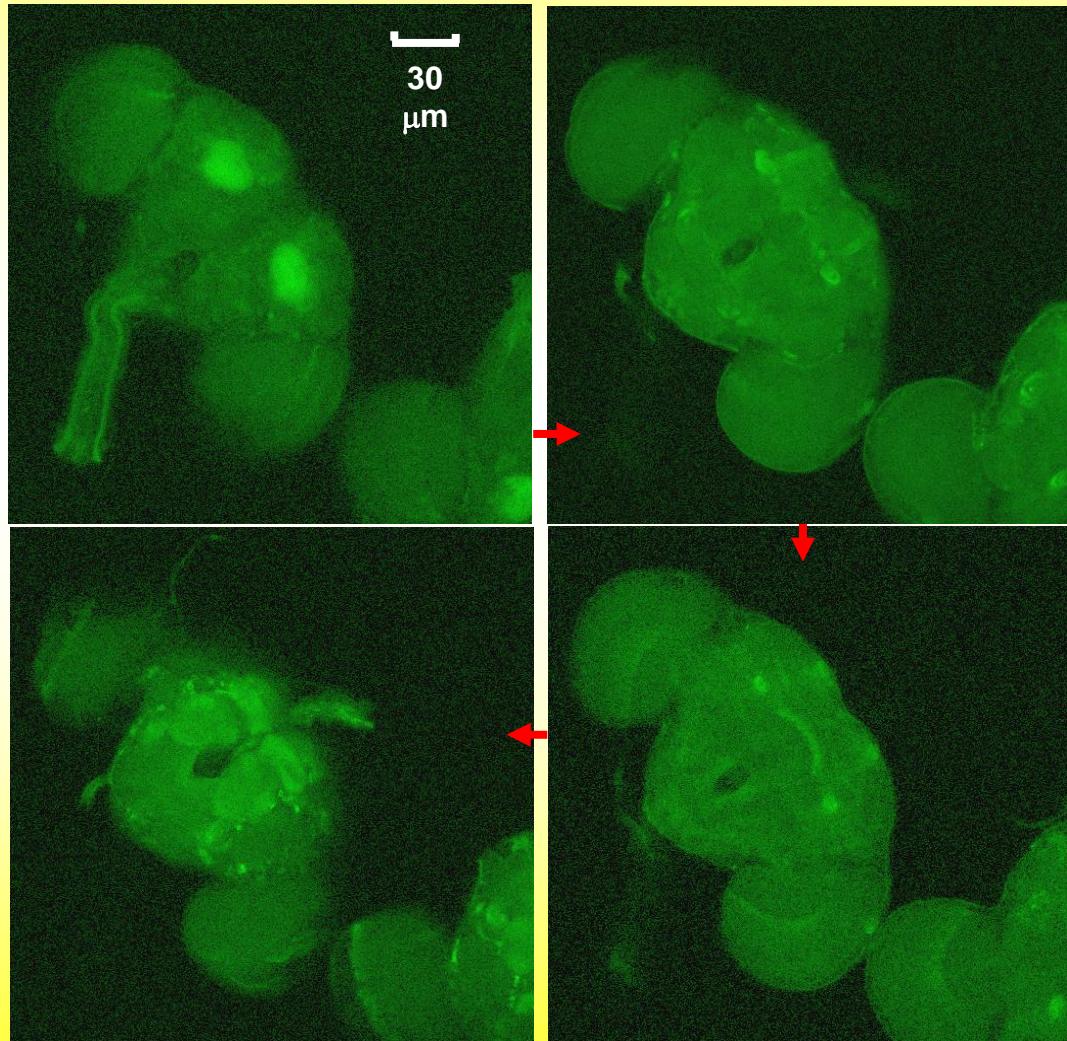
Laser display using
intracavity SHG technology

Interesting nonlinear-optical devices

Blue laser bio-image

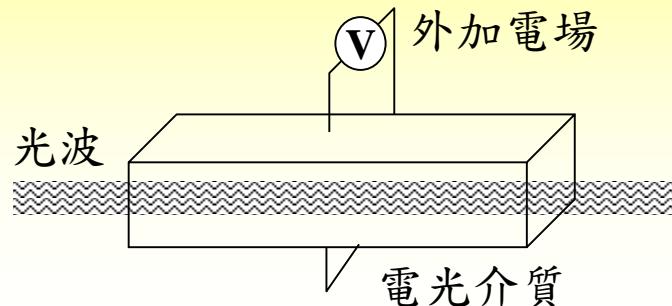


共焦顯微術 (confocal
microscopy) 果蠅 (*Drosophila*)
腦結構造影掃瞄

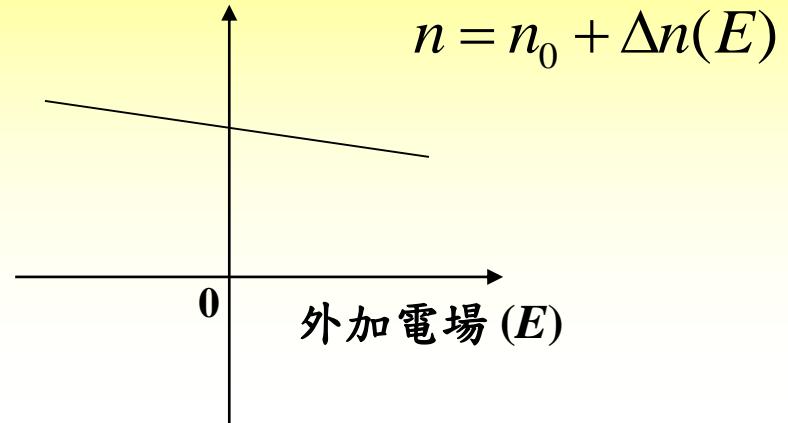


Interesting nonlinear-optical phenomena

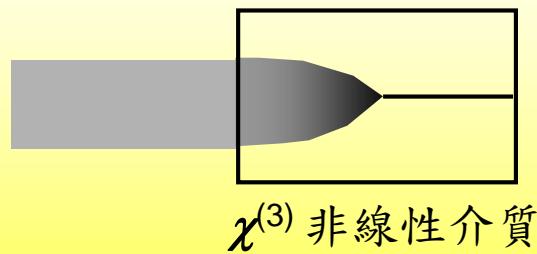
Electro-optic (EO) effect ($\chi^{(2)}$)



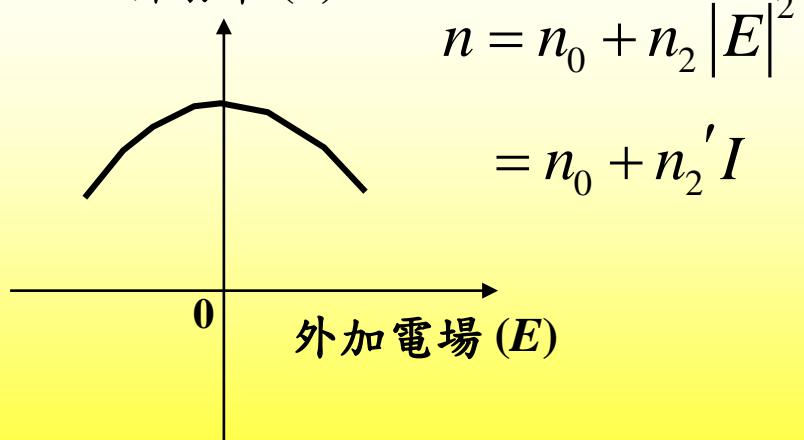
折射率 (n)



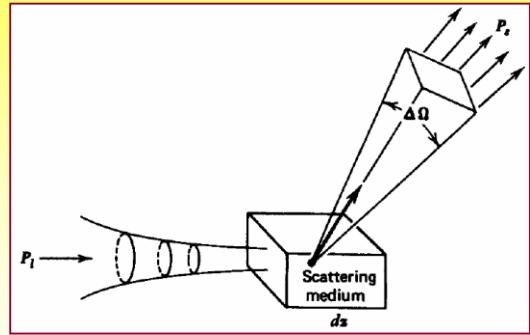
Kerr effect ($\chi^{(3)}$)



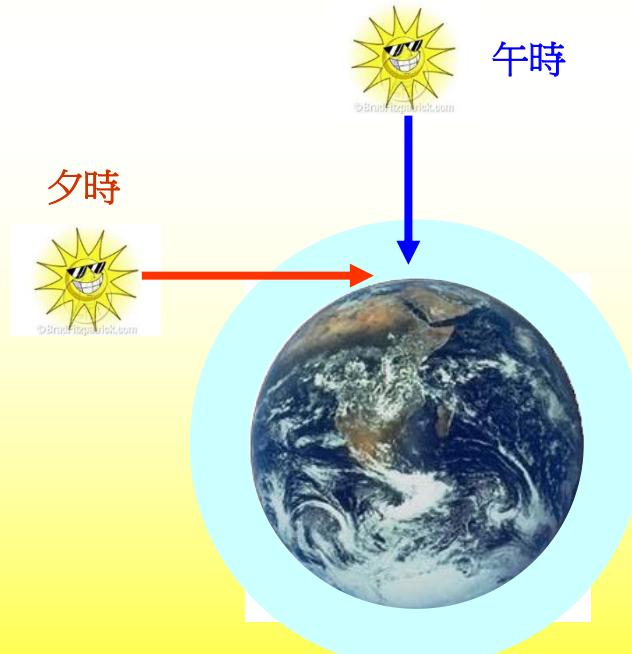
折射率 (n)



Interesting nonlinear-optical phenomena



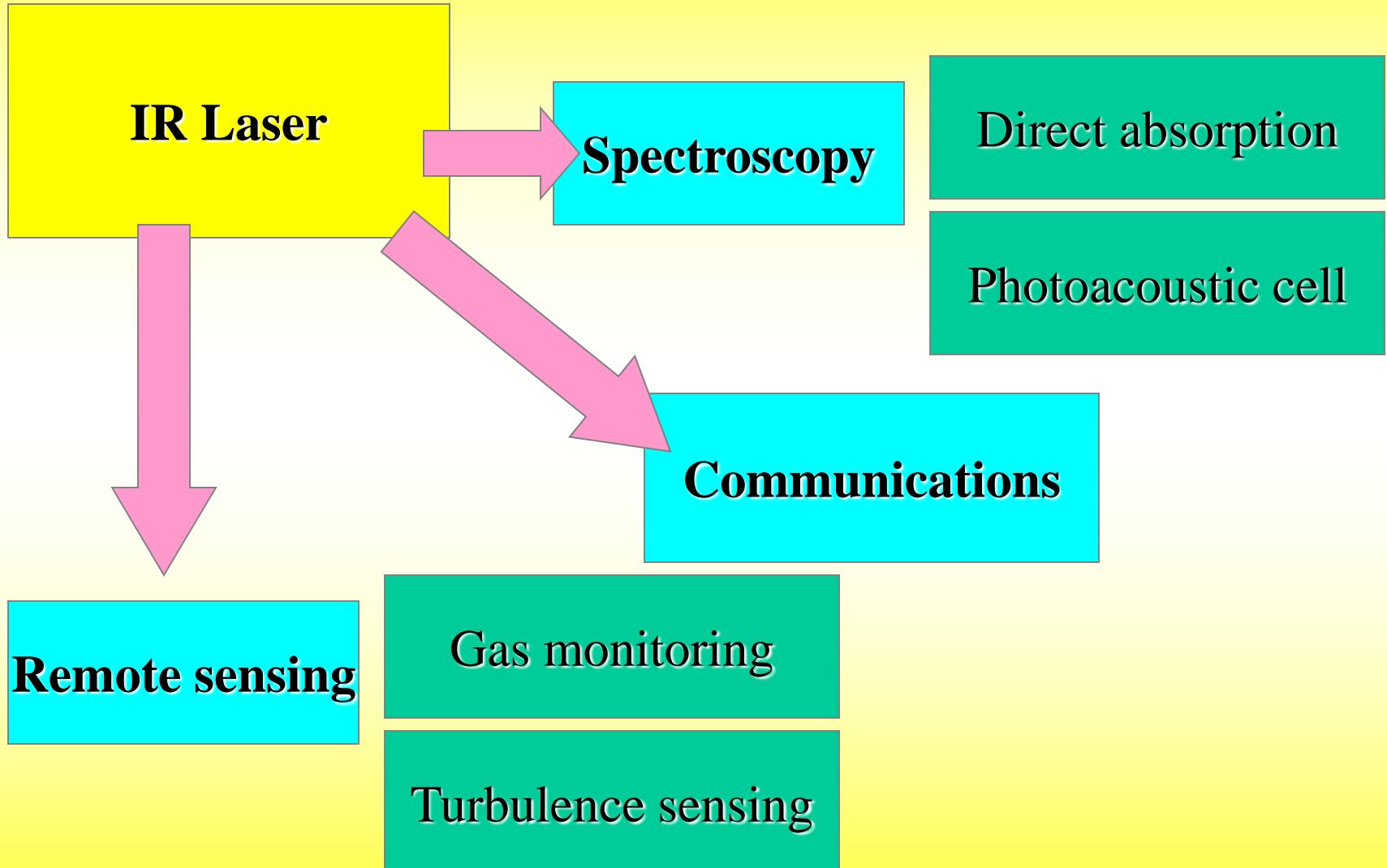
Rayleigh scattering



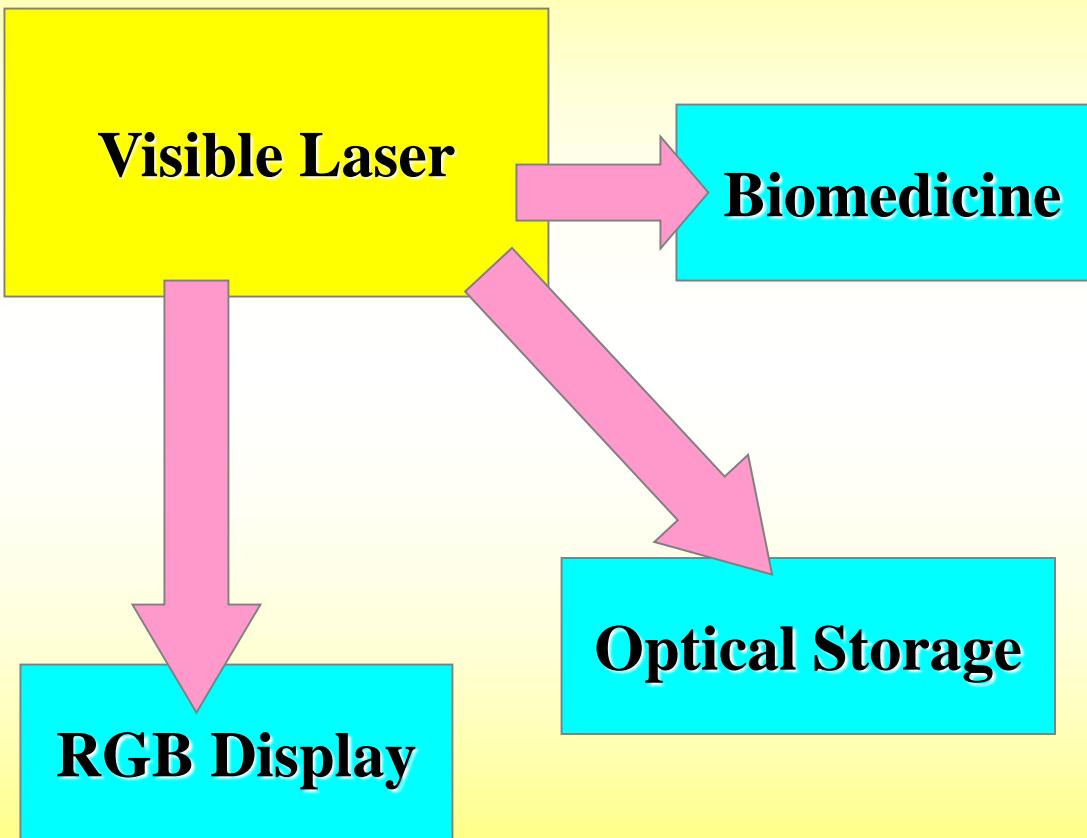
Why the sky is blue in the daytime?



Applications



Applications



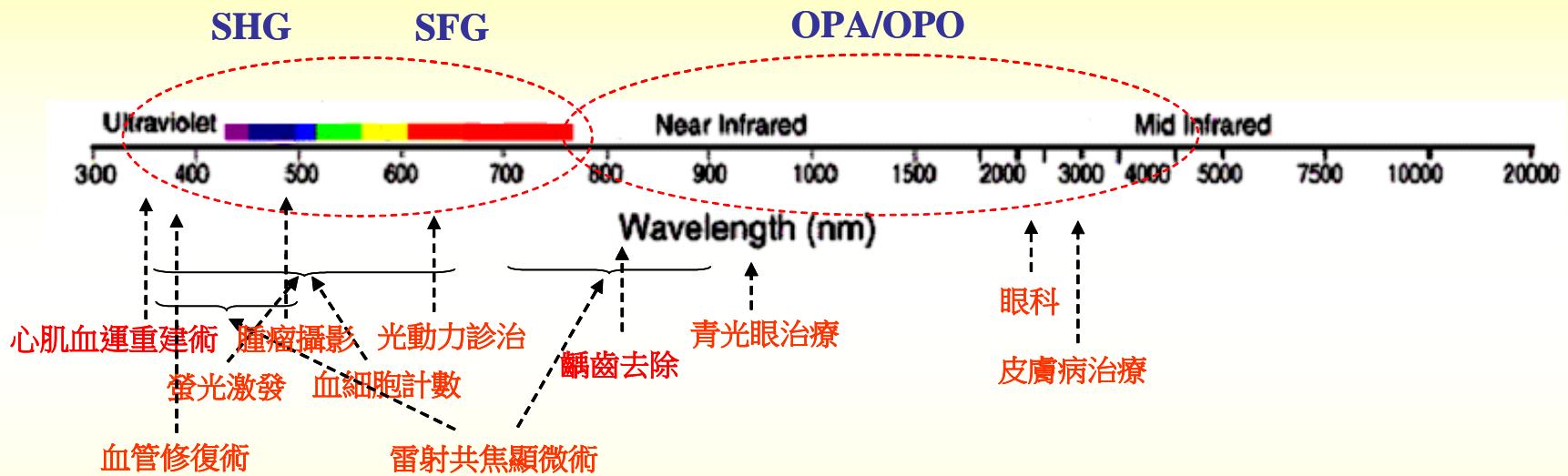
Fluorescence

Microscopy

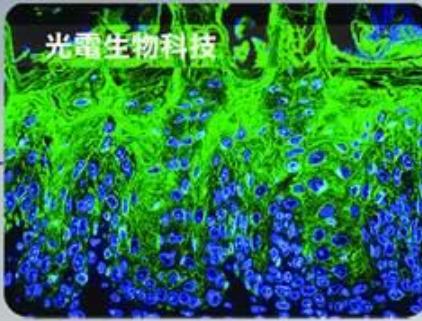
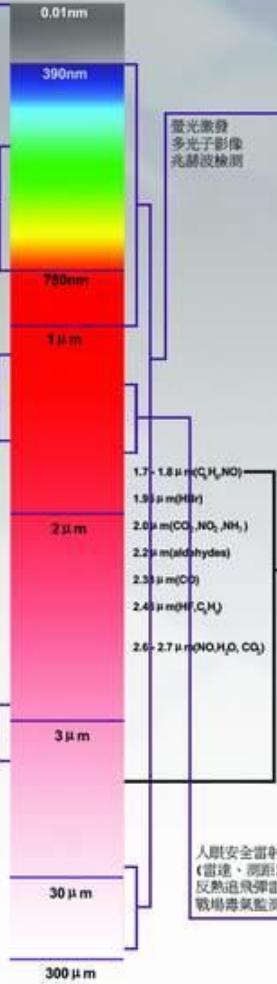
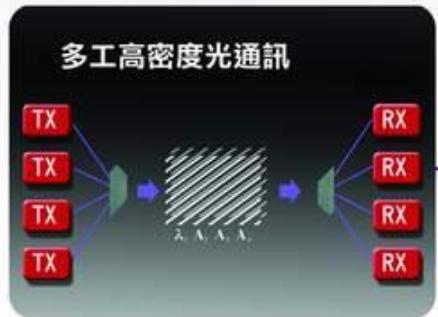
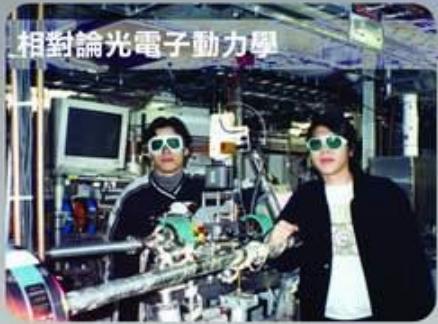
Biosensing

Applications

Biomedicine



Applications





Thank You for Your Kind Attention

中央大學非線性積體雷射光學實驗室

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