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### **Introduction to nonlinear optics**

- •The refractive index of a medium depends on the dielectric polarization which is determined by the arrangement of atoms and distribution of electrons in it.
- •The linear and nonlinear terms refer to the conditions when the dielectric polarization (hence the refractive index) depends linearly and nonlinearly on the electric field or the light intensity.

$$\vec{P} = \varepsilon_0 \chi^{(1)} \vec{E} + \varepsilon_0 \chi^{(2)} \vec{E} \vec{E} + \varepsilon_0 \chi^{(3)} \vec{E} \vec{E} \vec{E} + \dots$$

### Linear optical effect

If the intensity of incident light beam is small (electric field  $E < 10^7 \text{ V/cm}$ ) compared to the intra-atomic fields ( $10^7 - 10^{10} \text{ V/cm}$ ) of the material, the material response i.e. dielectric polarization P is proportional to electric field of the light and the refractive index is a constant for the material.

$$\vec{P}_{linear} = \varepsilon_0 \chi^{(l)} \vec{E}$$
$$\vec{D} = \varepsilon_0 \vec{E} + \vec{P}_{linear}$$
$$\varepsilon \vec{E} = \varepsilon_0 (l + \chi^{(l)}) \vec{E}$$
$$n_0 = \sqrt{l + \chi^{(l)}}$$
Ma

Material constant (independent of E)

### Nonlinear optical effect

After the invention of lasers (E ~  $10^7 - 10^{14}$  V/cm), the interaction of strong coherent radiation can greatly modify the distribution of electrons. This leads to the change in refractive index with intensity of light. It is called nonlinear material because the material respond nonlinearly to the amplitude of the electric field. Materials like BaTiO<sub>3</sub>, LiNbO<sub>3</sub>, BSO, BGO, GaAs, InP etc. respond to intense light beam nonlinearly. Nonlinearity gives rise to two major effects, e.g., Electro-optic effect and Photorefractive effect.

$$\vec{P}_{nonlinear} = \varepsilon_0 \chi^{(2)} \vec{E} \vec{E}$$
$$\vec{D} = \varepsilon_0 \vec{E} + \vec{P}_{nonlinear}$$
$$\varepsilon \vec{E} = \varepsilon_0 (1 + \chi^{(1)}) \vec{E} + \varepsilon_0 \chi^{(2)} E \vec{E}$$
$$\varepsilon / \varepsilon_0 = (1 + \chi^{(1)}) + \chi^{(2)} E$$
$$n^2 = n_0^2 + \chi^{(2)} E$$

$$n = \sqrt{n_0^2 + \chi^{(2)}E}$$

Nonlinear dependence on E

### **Electro-optic effect:**

Optical properties of certain materials change when subjected to an electric field. This is caused by forces that distort the positions, orientations, or shapes of the molecules constituting the material. The electro-optic effect is the change in the refractive index resulting from the application of a dc (or low frequency of order kHz) applied electric field.

(a) <u>Linear electro-optic effect or Pockels effect</u> (Friedrich Carl Alwin Pockels in 1893):

It produces <u>birefringence</u> in an optical medium induced by a constant or varying <u>electric field</u>. This effect can occur only in non-centrosymmetric materials. It allows the polarisation properties of light to be controlled electrically and, in combination with a polarising beam splitter, and also is used to create an optical switch.

#### $\Delta n \propto E$

**(b)** Quadratic electro-optic effect or Kerr effect (Scottish physicist John Kerr in 1875 ):

The Kerr effect is that the induced index change is directly proportional to the square of the electric field instead of varying linearly with it. All materials show a Kerr effect, but certain liquids display it more strongly than others.

### $\Delta n \propto E^2$ or higher order

## **Photorefractive effect and Photorefractive materials**

The photorefractive effect can be described by the following five physical processes:

- (1) Photoionization of impurities and the generation of charge carriers.
- (2) Transport of these charge carriers due to diffusion or drift (electric forces
- (3) Trapping of the charge carriers and the formation of space charge density.
- (4) Due to this charge redistribution, a space charge electric field is generated.
- (5) Formation of the refractive index grating via the linear electro optic effect.

> A photorefractive material exhibits both the photoconductive and electro-optic behaviour and has the ability to detect and store spatial distribution of optical intensity in the form of spatial pattern of altered refractive index.

First experimental demonstration of non-linear phenomena

The first nonlinear optics experiment as shown in Fig., is often taken to be the discovery of second-harmonic generation by Franken et al.(1961), shortly after the demonstration of the first working laser by Maiman in 1960. It should be noted that some nonlinear effects were discovered prior to the advent of the laser.

In this experiment, a ruby laser was focused into a slab of crystalline quartz to discover if the nonlinear response of the medium to the intense optical frequency radiation at 694.3 nm was strong enough to create a detectable second harmonic component at a wavelength of 347.15 nm.



Fig. Schematic diagram of first second harmonic generation experiment

### **Second-Harmonic Generation**

In the process of second-harmonic generation (SHG), a laser beam whose electric field strength is represented as

 $\tilde{E}(t) = Ee^{-i\omega t} + \text{c.c.}$ 

is incident upon a crystal for which the second-order susceptibility  $\chi(2)$  is nonzero. The nonlinear polarization that is created in such a crystal is given by

$$\tilde{P}^{(2)}(t) = 2\epsilon_0 \chi^{(2)} E E^* + (\epsilon_0 \chi^{(2)} E^2 e^{-i2\omega t} + \text{c.c.}).$$

From above Eq. we can see that the second-order polarization consists of a contribution at zero frequency (the first term) and a contribution at frequency  $2\omega$ (the second term), lead to the generation of radiation at the second-harmonic frequency. It should be noted that the first contribution in above Eq. does not lead to the generation of electromagnetic radiation (because its second time derivative vanishes); it leads to a process known as optical rectification, in which a static electric field is created across the nonlinear crystal.

### Fig. (a) Geometry of SHG

(b) Energy-level diagram describing second-harmonic generation



- SHG is used to convert the output of a fixed-frequency laser to a different spectral region. For example, the Nd:YAG laser operates in the near infrared at a wavelength of 1.06 μm.
- SHG is also used to convert the wavelength of the radiation to 0.53µm, in the middle of the visible spectrum.

#### Sum-Frequency Generation

The optical field incident upon a second-order nonlinear optical medium consists of two distinct frequency components, The process of sumfrequency generation (SFG) is illustrated in given Fig. and the complex amplitude of the nonlinear polarization describing this process is given by

$$P(\omega_1 + \omega_2) = 2\epsilon_0 \chi^{(2)} E_1 E_2.$$

In many ways the process of SFG is analogous to that of SHG, except that in SFG the two input waves are at different frequencies.

SFG is used to produce tunable radiation in the UV spectral region by selecting one of the input waves to be the output of a fixedfrequency visible laser and the other to be the output of a frequencytunable visible laser.



Fig.- (a) Geometry of SFG(b) Energy-level description.

### **Difference-Frequency Generation**

The process of difference-frequency generation (DFG) as shown in Fig., and is described by a nonlinear polarization of the form

(a)

$$P(\omega_1 - \omega_2) = 2\epsilon_0 \chi^{(2)} E_1 E_2^*$$

where the frequency of the generated wave is the difference of those of the applied fields. In this process, the conservation of energy requires that for every photon that is created at the difference frequency  $\omega 3=\omega 1-\omega 2$ , a photon at the higher input frequency ( $\omega 1$ ) must be destroyed and a photon at the lower input frequency ( $\omega 2$ ) must be created. Thus, the lower frequency input field is amplified by the process of DFG. For this reason, the process of DFG is also known as optical parametric amplification.

DFG can be used to produce tunable infrared radiation by mixing the output of a frequency-tunable visible laser with that of a fixed-frequency visible laser.



Fig.- (a) Geometry of the DFG (b) Energy-level description

# **Third Harmonic Generation**

We consider the third-order contribution to the nonlinear polarization

$$\tilde{P}^{(3)}(t) = \epsilon_0 \chi^{(3)} \tilde{E}(t)^3.$$
$$\tilde{P}^{(3)}(t) = \frac{1}{4} \epsilon_0 \chi^{(3)} \mathcal{E}^3 \cos 3\omega t + \frac{3}{4} \epsilon_0 \chi^{(3)} \mathcal{E}^3 \cos \omega t.$$

Since the applied electric field  $\tilde{E}(t)$  is monochromatic and is given by

$$\tilde{E}(t) = \mathcal{E}\cos\omega t.$$

The second term in above Eq. describes a nonlinear contribution to the polarization at the frequency of the incident field; this term hence leads to a nonlinear contribution to the refractive index experienced by a wave at frequency  $\omega$ .

The first term, 
$$\frac{1}{4}\epsilon_0\chi^{(3)}\mathcal{E}^3\cos 3\omega t$$

describes a response at frequency  $3\omega$  that is created by an applied field at frequency  $\omega$ . This term leads to the process of third-harmonic generation (THG), which is illustrated in Fig. According to the photon description of this process, shown in part (b) of the Fig., three photons of frequency  $\omega$  are destroyed and one photon of frequency  $3\omega$  is created in the microscopic description of this process.

