Module 4 : Third order nonlinear optical processes

Lecture 20 : Nonlinear Refraction and absorption

Objectives

In this lecture

1. We will analyze the effects of third order nonlinear polarization on the optical properties of the medium.
2. Relation between coefficients of intensity dependent index of refraction and intensity dependent absorption coefficients and third order nonlinear susceptibility will be derived.

Nonlinear Refraction and absorption

All third order nonlinear optical phenomena stem from the nonlinear polarization $P_{NL}^{(3)}(\vec{r},t)$ induced by frequency mixing of three interacting fields. The nonlinear polarization $P_{NL}^{(3)}$ is related to the incident light fields by the general equation which is essentially the nonlinear convolution of the material response function and the interacting fields (Lecture 8)

$$P_{NL}^{(3)}(\vec{r},t)=\varepsilon_0\int\int_{-\infty}^{\infty}\int R^{(3)}\left((z-t_1),(t-t_2),(t-t_3)\right)E(\vec{r},t_1)E(\vec{r},t_2)E(\vec{r},t_3)\,\mathrm{d}t_1\mathrm{d}t_2\mathrm{d}t_3 \qquad (20.1)$$

The material response function is related to the third order nonlinear susceptibility by its Fourier transform in frequency domain. If we assume material response to be instantaneous like Dirac delta function, the nonlinear polarization induced in the medium can be written as

$$\vec{P}_{NL}^{(3)}=\varepsilon_0\,\hat{\chi}^{(3)}:\vec{E}\vec{E}\vec{E} \qquad (20.2)$$

where $\hat{\chi}^{(3)}$ is the tensor of rank 4. In this lecture we will deal with the consequences of $P_{NL}^{(3)}$ which are responsible for self-(or) cross-action effects. In other words, we will focus on those manifestations of $P_{NL}^{(3)}$ which modify optical properties of the medium refractive index and absorption coefficient. Consequently, these affect the propagation characteristics of the field responsible for it and others as well. For simplicity, we consider linearly polarized transverse fields propagating along *z* axis all at the same frequency to ω.

These can be either *x* or *y* polarized. Thus if we consider the induced polarization along *x* axis

$$\left(P_{NL}^{(3)}\right)_x=\varepsilon_0\,\chi_{xe_1e_2e_3}^{(3)}E_{e_1}E_{e_2}E_{e_3} \qquad (20.3)$$

where indices e_i, represent the directions of field polarization. Contributions to $\left(P_{NL}^{(3)}\right)_x$ require that either all fields be *x* polarized or one *x* and two *y* polarized i.e. contributing nonlinearity coefficients are

$\chi_{xe_1e_2e_3}^{(3)}\equiv\chi_{xxxx}^{(3)}$ or $\chi_{xxyy}^{(3)}$ or $\chi_{xyyx}^{(3)}$ or $\chi_{xyxy}^{(3)}$

corresponding to $E_xE_xE_x$; $E_xE_yE_y$; $E_yE_yE_x$ and $E_yE_xE_y$ products in fields.

Similarly for $\left(P_{NL}^{(3)}\right)_y$ following tensor coefficients contribute

$\chi_{xe_1e_2e_3}^{(3)}\equiv\chi_{yyyy}^{(3)}$ or $\chi_{yyxx}^{(3)}$ or $\chi_{yxxy}^{(3)}$ or $\chi_{yxyx}^{(3)}$

Far from resonance, indices are freely permutable (Lecture 9) i.e.
Following pairs of indices
xxyy, *xyyx, xyxy* and *yyxx* etc. are all equivalent.

To further simplify our discussion we assume that the medium is isotropic and the nonlinearity is of electronic origin

Then

$$\chi^{(3)}_{xxxx} = \chi^{(3)}_{yyyy} = \chi^{3} \tag{20.4}$$

and for *i jkl* involving *xy* pairs we have

$$\chi^{(3)}_{xy-pairs} = \frac{1}{3}\chi^{(3)} \tag{20.5}$$

Hence, the medium with electronic nonlinearity is described by only one independent tensor component which can be regarded as a scalar.
Thus, we can write

$$P^{(3)}_{NL} = \varepsilon_0 \chi^{(3)} EEE \tag{20.6}$$

Let us consider the electromagnetic field of the form

$$\tilde{E}(t) = \hat{x}\left[E(\omega)\,\mathrm{e}^{-\mathrm{i}\omega t} + c.c.\right] \tag{20.7}$$

and the induced nonlinear polarization oscillating at frequency ω

$$P^{(3)}_{NL}(t) = \hat{x}\left[P^{(3)}_{NL}(\omega)\,\mathrm{e}^{-\mathrm{i}\omega t} + c.c.\right] \tag{20.8}$$

Total nonlinear polarization

$$P^{(3)}_{NL}(t) = \varepsilon_0 \chi^{(3)} \hat{x}\left[E(\omega)\,\mathrm{e}^{-\mathrm{i}\omega t} + c.c.\right]^3 \tag{20.9}$$

Polarization part oscillating at frequency ω can be identified as

$$P^{(3)}_{NL}(\omega) = 3\varepsilon_0 \chi^{(3)} |E|^2 E \tag{20.10}$$

Restricting our discussion to centrosymmetric systems $(\chi^{(2)} = 0)$, induced total polarization in the medium

$$P_{total} = P^{(1)}_{L} + P^{(3)}_{NL}$$

$$= \varepsilon_0 \left(\chi^{(1)} + 3\chi^{(3)}|E|^2\right) E$$

$$= \varepsilon_0 \chi_{eff} E \tag{20.11}$$

Since the refractive index of the medium is related to its susceptibility, we can write

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

$$= 1 + \chi^{(1)} + +3\chi^{(3)}|E|^2 \tag{20.12}$$

This means that the refractive index of the medium becomes intensity dependent which can be written as

$$n = n_0 + n_2\left\langle \left|\tilde{E}(t)\right|^2 \right\rangle \tag{20.13}$$

where n_0 is the linear refractive index and n_2 is the coefficient of nonlinear index of refraction.
Angular bracket implies the time average over one optical cycle

$$\left\langle \left|\tilde{E}(t)\right|^2 \right\rangle = 2\left|E(\omega)\right|^2 \tag{20.14}$$

Thus

$$\left(n_0 + 2n_2\left|E\right|^2\right)^2 = 1 + \chi^{(1)} + 3\,\chi^{(3)}\left|E\right|^2$$

$$= n_0^2 + 3\,\chi^{(3)}\left|E\right|^2 \tag{20.15}$$

In writing the above equation we have dropped the frequency argument of the electric field of light. Retaining only lower order terms in nonlinearity,

$$n_0 + 4n_0 n_2\left|E\right|^2 = n_0^2 + 3\,\chi^{(3)}\left|E\right|^2 \tag{20.16}$$

The intensity dependent change in refractive index is

$$\Delta n = 4n_0 n_2\left|E\right|^2 = 3\,\chi^{(3)}\left|E\right|^2 \tag{20.17}$$

$$\therefore\ n_2 = \frac{3}{4n_0}\,\chi^{(3)} \tag{20.18}$$

From an analogy of equation 20.17 with D.C. Kerr effect, phenomenon of intensity dependent refractive index is also called optical Kerr effect and corresponding nonlinearity as Kerr -nonlinearity. Alternatively, the intensity dependent refractive index is also expressed as

$$n\left(I\right) = n_0 + n_2^I I \tag{20.19}$$

where n_2^I, is the coefficient of nonlinear index of refraction and *I* is the intensity

$$I = 2n_0 c\epsilon_0\left|E\right|^2 \tag{20.20}$$

From the equivalence of two descriptions, one can write

$$2n_2\left|E\right|^2 = n_2^I I \tag{20.21}$$

Using equation (20.20) we can write

$$n_2^I = \frac{n_2}{n_0 c\,\varepsilon_0} = \frac{3}{4n_0 c\,\varepsilon_0}\,\chi^{(3)} \tag{20.22}$$

Susceptibilities are in general complex quantities. It is easily seen that the real part of χ is related to the refraction properties while the imaginary part relates to the absorption coefficient
Of the medium

$$\alpha = \frac{\omega}{c}\,\mathrm{Im}\left[\,\chi\right] \tag{20.23}$$

While deriving equation (20.18), we have taken $\chi^{(3)}$ to be real. Allowing the complex nature of $\chi^{(1)}$ and $\chi^{(3)}$ in equation (20.17), it is easy to realize that the imaginary part of $\chi^{(3)}$ will contribute to the intensity dependent absorption.
Analogous to equation (20.19)

$$\alpha\left(I\right) = \alpha_0 + \beta I \tag{20.24}$$

where α_0 is the linear absorption coefficient and β is the intensity dependent absorption coefficient. One can immediately write in analogy with equation (20.22)

$$\beta = \frac{3\omega}{4 n_0 c^2 \varepsilon_0} \mathrm{Im}\left[\chi^{(3)}\right] \tag{20.25}$$

and

$$n_2^I = \frac{3}{4 n_0 c \varepsilon_0} \mathrm{Re}[\chi^{(3)}] \tag{20.26}$$

If a strong pump and a weak probe beam propagate, together the optical properties modified by the strong pump beam affect the propagation of the weak probe beam. This is called cross-action phenomenon. The effect is twice as large as that for the self- action for the single beam.

Thus third order nonlinear optical interaction leads to the intensity dependent refractive index and absorption in the medium. These two lead to some very interesting nonlinear optical processes and some important applications that we will discuss in the forthcoming lectures.

Recap

In this lecture

- We have analyzed how third order nonlinear polarization for the interaction of three degenerate fields with matter manifests in the intensity dependent optical properties of the medium.
- Relations between coefficients of intensity dependent index of refraction and intensity dependent absorption and third order nonlinear susceptibility have been derived for isotropic medium assuming nonlinearity to be of electronic origin.