

Chapter 2

Third-Order Nonlinear Effects

2.1 Introduction

The book focuses on utilizing the third-order nonlinear effects (including nonlinear absorption and refraction) to break the diffraction limit and form super-resolution nanoscale spot. In this chapter, let us give a brief introduction to the characteristics of the third-order effects.

When a light beam with a frequency of ω is incident on the isotropic nonlinear medium, the nonlinear effect occurs, and the second-order nonlinear susceptibility $\chi^{(2)}$ can be neglectable. The whole polarization is expressed as [1]

$$P[E(\omega)] = P^{(1)} + P^{(3)} = \epsilon_0 \left[\chi^{(1)} + 3\chi^{(3)}|E(\omega)|^2 \right] E(\omega) \quad (2.1)$$

where $P^{(1)}$ and $P^{(3)}$ the linear and third-order nonlinear polarization. $\chi^{(1)}$ and $\chi^{(3)}$ are the linear and third-order nonlinear susceptibility, respectively. The polarizations are marked into the real and imaginary parts as follows

$$\begin{cases} P^{(1)} = P_R^{(1)} + iP_I^{(1)} \\ P^{(3)} = P_R^{(3)} + iP_I^{(3)} \end{cases} \quad (2.2)$$

The $\chi^{(1)}$ and $\chi^{(3)}$ can be also marked into real and imaginary parts as follows

$$\begin{cases} \chi^{(1)} = \chi_R^{(1)} + i\chi_I^{(1)} \\ \chi^{(3)} = \chi_R^{(3)} + i\chi_I^{(3)} \end{cases} \quad (2.3)$$

where $\chi_R^{(1)}$ and $\chi_R^{(3)}$ are real part and directly related with the refraction. $\chi_I^{(1)}$ and $\chi_I^{(3)}$ are imaginary parts and related with absorption.

2.2 Nonlinear Refraction

For isotropic (homogeneous) materials, the nonlinear refraction can be considered to be from a four-wave interaction and the third-order nonlinearity is mainly field dependent. Accordingly, the nonlinear polarization is written as $P^{(3)}[E(\omega)] = 3\epsilon_0\chi_R^{(3)}|E(\omega)|^2E(\omega)$. Assuming that the refractive index is measured using a single laser beam method, such as single beam z-scan technique, the four-order and higher terms can be neglectable. Then, the total polarization of the material system is

$$P_R[E(\omega)] = P_R^{(1)} + P_R^{(3)} = \epsilon_0 \left[\chi_R^{(1)} + 3\chi_R^{(3)}|E(\omega)|^2 \right] E(\omega) \equiv \epsilon_0 \chi_R^{\text{eff}} E(\omega) \quad (2.4)$$

where $\chi_R^{(1)}$ is the real part of linear susceptibility. Here, one defines the effective susceptibility as $\chi^{\text{eff}} = \chi_R^{\text{eff}} + i\chi_I^{\text{eff}}$, where the χ_R^{eff} and χ_I^{eff} are the real and imaginary parts of effective susceptibility, respectively. According to formula (2.4), the χ_R^{eff} is written as

$$\chi_R^{\text{eff}} = \chi_R^{(1)} + \chi_{eR}^{(3)}|E(\omega)|^2 \quad (2.5)$$

with

$$\chi_{eR}^{(3)}|E(\omega)|^2 = 3\chi_R^{(3)}|E(\omega)|^2 \quad (2.6)$$

If all of the nonlinear susceptibility is thought to be effective susceptibility, one can obtain

$$\chi_{eR}^{(3)}|E(\omega)|^2 \sim \chi_R^{(3)}|E(\omega)|^2 \quad (2.7)$$

The electric displacement vector is

$$D = \epsilon_0 E + P = \epsilon_0 \left[1 + \chi_R^{(1)} + \chi_R^{(3)}|E(\omega)|^2 \right] E(\omega) \quad (2.8)$$

Based on $D = \epsilon E$, the total permittivity is

$$\epsilon = \epsilon_0 \left[1 + \chi_R^{(1)} + \chi_R^{(3)}|E(\omega)|^2 \right] \quad (2.9)$$

The relative permittivity is

$$\epsilon_r = \frac{\epsilon}{\epsilon_0} = 1 + \chi_R^{(1)} + \chi_R^{(3)}|E(\omega)|^2 \quad (2.10)$$

The refractive index is defined as

$$n = \sqrt{\epsilon_r \mu_r} \quad (2.11)$$

where μ_r is relative permeability and generally equals to 1 for non-magnetic materials. Thus

$$n = \sqrt{\epsilon_r} \quad (2.12)$$

Substituting formula (2.10) into formula (2.12), one has

$$n = \sqrt{1 + \chi_R^{(1)} + \chi_R^{(3)} |E(\omega)|^2} \quad (2.13)$$

The linear refractive index is defined as

$$n_0^2 = 1 + \chi_R^{(1)} \quad (2.14)$$

Thus, formula (2.13) is rewritten as

$$n = n_0 \left[1 + \frac{\chi_R^{(3)} |E(\omega)|^2}{n_0^2} \right]^{\frac{1}{2}} \quad (2.15)$$

Generally, the $\chi_R^{(3)} |E(\omega)|^2 / n_0^2$ term is much smaller than 1. Using the Taylor series expansion, the formula (2.15) can be expressed as

$$n \approx n_0 + \frac{\chi_R^{(3)}}{2n_0} |E(\omega)|^2 \quad (2.16)$$

Generally, the refractive index can also be written as

$$n = n_0 + \Delta n = n_0 + \gamma I \quad (2.17)$$

where Δn is the third-order nonlinear susceptibility-induced refractive index change and γ is nonlinear refraction coefficient. I is light intensity and can be defined as

$$I = \frac{v\epsilon_r\epsilon_0}{2} \langle \tilde{E}(\omega, t)^2 \rangle \quad (2.18)$$

where $v = c/n_0$ is light speed in the medium. $\tilde{E}(\omega, t)$ is time-dependent optical field, and $\epsilon_r \sim n_0^2$, thus

$$I \approx \frac{1}{2} \epsilon_0 c n_0 \langle \tilde{E}(\omega, t)^2 \rangle \quad (2.19)$$

If the optical field is of the form [2]

$$\tilde{E}(\omega, t) = E(\omega) \exp(-i\omega t) + c.c., \quad (2.20)$$

one has

$$\langle \tilde{E}(\omega, t)^2 \rangle = 2E(\omega)E^*(\omega) = 2|E(\omega)|^2 \quad (2.21)$$

According to formulas (2.19) and (2.21), one has

Table 2.1 Typical values of γ and response time for linearly polarized light [2]

Mechanism	$\gamma (m^2/W)$	Response time (s)
Electronic polarization	$\sim 10^{-20}$	$\sim 10^{-15}$
Molecular orientation	$\sim 10^{-18}$	$\sim 10^{-12}$
Electrostriction	$\sim 10^{-18}$	$\sim 10^{-9}$
Saturated atomic absorption	$\sim 10^{-14}$	$\sim 10^{-8}$
Thermal effects	$\sim 10^{-10}$	$\sim 10^{-3}$
Photorefractive effects ^a	Large	Intensity dependent

^a The photorefractive effect can cause large nonlinear effect, which cannot usually be described with nonlinear susceptibility $\chi^{(3)}$ (or γ). The nonlinear polarization process of the photorefractive effect is not the same as the other physical mechanisms listed

$$|E(\omega)|^2 = \frac{1}{\epsilon_0 c n_0} I \quad (2.22)$$

Substituting formula (2.22) into formula (2.16), one obtains

$$n \approx n_0 + \frac{\chi_R^{(3)}}{2\epsilon_0 c n_0^2} I \quad (2.23)$$

Based on formulas (2.17) and (2.23), the nonlinear refraction coefficient is written as

$$\gamma = \frac{\chi_R^{(3)}}{2\epsilon_0 c n_0^2} \quad (2.24)$$

According to formula (2.24), one can obtain γ value. The different physical mechanisms can cause different γ values. The typical γ value and response time are listed in Table 2.1. In formula (2.24), $\gamma < 0$ corresponds to self-defocusing effect and $\gamma > 0$ can lead to self-focusing effect. The self-focusing effect can generate the nanoscale spot, which is very useful for nanolithography and high-resolving light imaging, etc.

2.3 Nonlinear Absorption

Similar to the real part of polarization, the imaginary part of polarization is

$$P_I[E(\omega)] = P_I^{(1)} + P_I^{(3)} = i\epsilon_0 \left[\chi_I^{(1)} + \chi_I^{(3)} |E(\omega)|^2 \right] E(\omega) \quad (2.25)$$

For isotropic medium, on the basis of the *slowly varying-envelope approximation*, one has

$$\frac{dE}{dz} = \frac{i\omega}{2\epsilon_0 c n_0} P_I \quad (2.26)$$

Substituting formulas (2.22) and (2.25) into formula (2.26), one has

$$\frac{dE}{dz} = - \left(\frac{\omega}{2cn_0} \chi_I^{(1)} + \frac{\omega}{2\epsilon_0 c^2 n_0^2} \chi_I^{(3)} I \right) E \quad (2.27)$$

One defines the following

$$\frac{\alpha}{2} = \frac{\alpha_0}{2} + \frac{\Delta\alpha}{2} = \frac{\omega}{2cn_0} \chi_I^{(1)} + \frac{\omega}{2\epsilon_0 c^2 n_0^2} \chi_I^{(3)} I \quad (2.28)$$

From the formula (2.28), the formula (2.27) is rewritten as

$$\frac{dE}{dz} = -\frac{\alpha}{2} E \quad (2.29)$$

So

$$E(L) = E_0 \exp\left(-\frac{\alpha}{2} L\right) \quad (2.30)$$

where L is medium thickness. The intensity decaying accordingly is

$$I(L) = I_0 e^{-\alpha L} \quad (2.31)$$

Thus,

$$\begin{cases} \alpha_0 = \frac{\omega}{cn_0} \chi_I^{(1)} \\ \Delta\alpha = \frac{\omega}{\epsilon_0 c^2 n_0^2} \chi_I^{(3)} I \end{cases} \quad (2.32)$$

Thus, α_0 is defined as linear absorption coefficient, and $\Delta\alpha$ is nonlinearity-induced intensity-dependent absorption coefficient change and is

$$\Delta\alpha = \beta I \quad (2.33)$$

where β is nonlinear absorption coefficient and is

$$\beta = \frac{\omega}{\epsilon_0 c^2 n_0^2} \chi_I^{(3)} \quad (2.34)$$

For some materials, such as semiconductors, $\beta < 0$ is saturation absorption, and $\beta > 0$ means reverse saturation absorption or multi-photon absorption.

As described in formula (2.31), the light goes through the nonlinear materials, and the intensity changes with the traveling distance z inside the materials. The intensity decaying process can be rewritten as

$$\frac{\partial I}{\partial z} = -\alpha_0 I - \beta I^2 \quad (2.35)$$

The exiting light intensity from the materials is [3]

$$I_e = \frac{I_0 \exp(-\alpha_0 L)}{1 + \beta I_0 L_{\text{eff}}} \quad (2.36)$$

with

$$L_{\text{eff}} = \frac{1 - \exp(-\alpha_0 L)}{\alpha_0}$$

For $\alpha_0 \rightarrow 0$, the linear absorption of materials is neglectable, and the third-order nonlinear absorption is dominant. In this case, $\exp(-\alpha_0 L) \sim (1 - \alpha_0 L)$, thus $L_{\text{eff}} \sim L$. Based on formula (2.36), the exiting light intensity becomes

$$I_e \sim \frac{I_0}{1 + \beta I_0 L} \quad (2.38)$$

An alternative way to obtain the exiting light intensity is through the formula (2.35) with $\alpha_0 = 0$, which yields

$$\frac{\partial I}{\partial z} = -\beta I^2 \quad (2.39)$$

Formula (2.38) can be solved by the separation of variables, which yields

$$I(z) = \frac{I_0}{1 + \beta I_0 z} \quad (2.40)$$

Formula (2.40) is consistent with formula (2.38) and can be used to calculate the light spot intensity profile after passing through the nonlinear absorption materials with weak linear absorption characteristics ($\alpha_0 \rightarrow 0$).

The nonlinear refraction can induce self-focusing super-resolution effect (for $\gamma > 0$), and the nonlinear absorption can lead to the generation of aperture-type super-resolution effect (for $\beta < 0$), or subwavelength energy absorption region (for $\beta > 0$), which can be applied to the nanolithography and nanoscale optical data storage, etc.

References

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