CHAPTER 1
Introduction to Nonlinear Optics

Laser technology has been developed for several decades. The progress of laser research has shown that our knowledge and understanding about the generation and control of coherent and intense optical radiation have reached a much higher level than before. Compared to various ordinary light sources, laser devices can provide intense coherent light beams with high directionality, high monochromaticity, high brightness, and high photon degeneracy. Based on the nonlinear interaction of laser radiation with matter, a great number of new effects and novel phenomena have been discovered. Studies on these new effects and the related novel techniques are the major issues of nonlinear optics.

1.1 DEFINITION OF NONLINEAR OPTICS

Nonlinear optics is a study that deals mainly with various new optical effects and novel phenomena arising from the interactions of intense coherent optical radiation with mater. There is a historical reason why this new branch of optical physics is termed 'nonlinear optics'.

Before 1960's, in the area of conventional optics many basic mathematical equations or formulae manifested a linear feature. To show this linear feature of conventional optics, we can consider the following three examples.

First, in order to interpret the refraction, reflection, dispersion, scattering, as well as birefringence of light propagation in a medium, we should consider an important physical quantity, the electric polarization induced in the medium. In the regime of conventional optics, the electric polarization vector **P** is simply assumed to be linearly proportional to the electric field strength **E** of an applied optical wave , i.e.,

$$\mathbf{P} = \varepsilon_0 \chi \mathbf{E}, \qquad (1.1\text{-}1)$$

where ε_0 is the free-space permittivity, χ is the susceptibility of a given medium. Based on this linear assumption, Maxwell's equations lead to a set of linear differential equations in which only the terms proportional to the first power of the field **E** are involved. As a result, there is no coupling between different light beams or between different monochromatic components when they pass through a medium. In other words, if there are several monochromatic optical waves of different frequencies passing through a medium simultaneously, no coherent radiation at any new frequency will be generated.

Second, in conventional optics, the attenuation of an optical beam propagating in an absorptive medium can be described as

$$\frac{dI}{dz} = -\alpha I, \tag{1.1-2}$$

where I is the beam intensity, z is the variable along the propagation direction, and α is a constant for a given medium. The physical meaning of Eq. (1.1-2) is that the decrease of the beam intensity in a unit propagation length is linearly proportional to the local intensity itself. From Eq. (1.1-2) we obtain a well known exponential attenuation expression

$$I(z) = I(0) \cdot e^{-\alpha z}. \tag{1.1-3}$$

This expression implies that for a given propagation length of $z=l$, the transmitted intensity $I(l)$ is linearly proportional to the initial intensity of $I=I(0)$.

The third example is related to the Fabry-Perot (F-P) interferometer that plays a vital role in modern optics. The transmission T of this device is determined by [1]

$$T = \frac{1}{1 + F\sin^2(\delta/2)}. \tag{1.1-4}$$

Here F is a constant determined by the reflectivity of the two mirrors of the interferometer, and δ is a phase-shift factor determined by

$$\delta = \frac{4\pi}{\lambda} n_0 d \cos\theta, \tag{1.1-5}$$

where λ is the wavelength of the incident beam, d is the spacing between the two mirrors, θ is the angle between the beam and the normal of the mirrors, and finally n_0 is the refractive index of the medium inside the F-P cavity. In the regime of conventional optics, n_0 is a constant independent of the incident beam intensity for a given wavelength. Therefore, the transmission T of the whole device is also a constant for given values of λ, θ, and d. In this case the transmitted intensity I_t is linearly proportional to the incident intensity I_0, i.e.,

$$I_t = T \cdot I_0 \propto I_0. \tag{1.1-6}$$

So far we have given three examples that manifest a simple linear feature as shown by Eqs. (1.1-1), (1.1-2), and (1.1-6), respectively. These simple linear assumptions or conclusions given by the conventional optics were widely accepted, and verified by most experimental observations and measurements based on the use of ordinary light sources. However, these situations have been changed radically since the beginning of 1960's.

Shortly after the demonstration of the first laser device (a pulsed ruby laser) in 1960,[2] it was found that these simple linear assumptions or conclusions described above were no longer adequate for circumstances in which an intense laser beam was incident on certain types of optical media. For the sake of clarity, we shall stay with our three examples and show why some higher-order approximations should be employed when an intense laser field interacts with an optical medium.

The first breakthrough was achieved in 1961 when a pulsed laser beam was sent into a piezoelectric crystal sample. In this case researchers, for the first time in the history of optics, observed the second-harmonic generation at an optical frequency.[3] Shortly after this discovery, several other coherent optical frequency-mixing effects (such as optical sum-frequency generation, optical difference-frequency generation, and optical third-harmonic generation) were observed. The researchers realized that all these new effects could be reasonably explained if replaced the linear term on the right-hand side of Eq. (1.1-1) by a power series

$$\mathbf{P} = \varepsilon_0[\chi^{(1)}\mathbf{E} + \chi^{(2)}\mathbf{EE} + \chi^{(3)}\mathbf{EEE} + \cdots]. \qquad (1.1\text{-}7)$$

Here, $\chi^{(1)}$, $\chi^{(2)}$, and $\chi^{(3)}$ are the first-order (linear), second-order (nonlinear), and third-order (nonlinear) susceptibility and so on. They are material coefficients and in general are tensors. Substituting Eq. (1.1-7) into Maxwell's equations leads to a set of nonlinear differential equations that involve high-order-power terms of optical electric field strength; these terms are responsible for various observed coherent optical frequency-mixing effects.[4]

In the same time period, researchers also found that the depletion behavior of an intense laser beam propagating in an absorptive optical medium did not follow the description indicated by Eq. (1.1-2) or Eq. (1.1-3). For instance, in a one-photon absorptive medium, if the intensity of the incident beam is high enough, the attenuation coefficient α is no longer a constant and may become a variable that depends on the incident intensity. Therefore, the exponential attenuation formula like Eq. (1.1-3) can not be applied and the linear relationship between $I(z=l)$ and $I(0)$ does not hold. In this case, either a saturable absorption or a reverse-saturable absorption effect may take place. Moreover, if there is a two-photon absorption process involved in the medium, the attenuation of an intense incident beam should be described as

$$\frac{dI}{dz} = -\alpha I - \beta I^2, \qquad (1.1\text{-}8)$$

where β is the two-photon absorption coefficient, which could be viewed as a constant only if the saturation or reverse-saturation effect can be neglected. In more general cases, if we further extend our consideration to include multi-photon (three-photon or more) absorption processes, then Eq. (1.1-8) should be generalized to the following form:

$$\frac{dI}{dz} = -\alpha I - \beta I^2 - \gamma I^3 - \cdots. \qquad (1.1\text{-}9)$$

Here γ is the three-photon absorption coefficient and so on.

Now let us return to the transmission behavior of a F-P device under the action of an intense laser beam. In this case, Eq. (1.1-6) is no longer applicable. The prediction that the refractive index of a medium at given wavelength is a constant, arises from the linear assumption of the electric polarization expressed by Eq. (1.1-1). However, based on the more general assumption expressed by Eq. (1.1-7), the refractive index for centrosymmetric or isotropic media can be written as (see Chapter 5)

$$n = n_0 + n_2' I, \tag{1.1-10}$$

where the first term n_0 is the linear refractive index independent of the beam intensity, the second term corresponds to an additional nonlinear refractive index contribution proportional to the beam intensity, and n_2' is a proportionality coefficient. When the beam intensity is quite low, the second term in Eq. (1.1-10) can be neglected. However, when the intensity of the incident laser beam is high enough, the second term may no longer be negligible. In fact, the intensity-dependent refractive-index change is the basic mechanism for many major nonlinear optical effects. For a F-P device interacting with an intense laser beam, the phase-shift factor, according to Eq. (1.1-10), now is determined by

$$\delta = \frac{4\pi}{\lambda} d \cos\theta \cdot (n_0 + n_2' I_i), \tag{1.1-11}$$

where I_i is the intracavity intensity of the incident laser beam. In this case, the transmission T of the F-P device is no longer a constant even for the given values of λ, d, and θ; in other words, there will be a complicated nonlinear relationship between the incident intensity and the transmitted intensity. The nonlinear response of a F-P device containing a nonlinear medium is one of the major issues of optical bistability studies.

Based on these comparisons described above, we can conclude that the main concern in conventional optics is the propagation and interaction with matter of the light from ordinary light sources, wherein the intensities of the light beams are so low that even a simple linear approximation is enough to give a good theoretical explanation for the related optical effects and phenomena. In this sense, the conventional optics may also be called 'linear optics' or 'optics of weak light'. On the other hand, 'nonlinear optics' mainly deals with the interaction of intense laser radiation with matter. In the latter case, the intensities of laser beams can be so high that a great number of new effects and novel phenomena can be observed, and some high-order nonlinear approximations have to be employed to explain these new effects and phenomena. In this sense, nonlinear optics may also be called 'optics of intense light'. In general, the contents of nonlinear optics are much more extensive than that of linear optics and, accordingly, the theories of the former are more complicated than that of the latter.