Chapter 1
Introduction

This chapter discusses the importance of nonlinear optics in physics, optics and photonics technology, the physical meaning of nonlinear optics, the research contents, the research history and the development trend of nonlinear optics, and the situation and prospect of application of nonlinear optics.

1.1 Importance of Nonlinear Optics

1.1.1 Status of Nonlinear Optics in Modern Physics

As is well-known,the modern physics was found on two headstones of the quantum physics and the relativity physics in the early 20th century. The quantum physics studies the movement theory of microscopic particles including molecules, atoms, nucleons and elemental particles; the relativity physics studies the movement theory of high speed object near light velocity and the gravitational interaction among objects with big quality. Because the quantum theory is found on the corpuscular property of light, and the relativity theory is found on the principle of constancy of light velocity, so the optics is the basic of quantum theory and relativity theory.

Some people think that the nonlinear physics founded in the latter half of the 20th century also is a headstone of the modern physics. The nonlinear physics is study of the nonlinear relationship between action and reaction (response) of objects under a strong interaction. These phenomena contain in different fields of physics, to form the nonlinear mechanics, nonlinear acoustics, nonlinear thermodynamics, nonlinear electromagnetism, and nonlinear optics, respectively. Nonlinear optics is one of branches of the nonlinear physics.

C. Li, *Nonlinear Optics*, DOI 10.1007/978-981-10-1488-8_1

1.1.2 Status of Nonlinear Optics in Modern Optics

Since the invention of laser in 1960, the modern optics was born. We call the optics based on the common light source with the spontaneous radiation as traditional optics; and the optics based on the laser source with the stimulated radiation as modern optics (we can call it as Photonics). In the latter half of the 20th century, the modern optics developed very quickly, surround the laser research and application to generate several subdisciplines of modern optics. Table 1.1 lists a number of relatively mature subdisciplines of modern optics and their research object and main application [1].

Among the subdisciplines of modern optics, the laser physics and the nonlinear optics are most important. The laser physics mainly studies the principle of laser devices and properties of laser; and nonlinear optics studies various nonlinear effects in the interaction between laser and matter. There are two aspects: one is that the pump light induces the change of matter's macroscopic parameters (susceptibility, dielectric coefficient, refractive index, absorption coefficient, etc.) or the change of matter's microscopic structure, to realize "controlling matter by light";

Table 1.1 Research object and main application of subdisciplines of modern optics (Photonics)

Subdisciplines	Research object	Main application
Laser physics	Laser device theory and properties of laser	Design of different types of laser devices with different parameters of output laser beams. Laser induces strong field and extreme physical environment; laser control particles and cell. Using laser for illumination, measuring, display, sensing, communication and laser processing
Nonlinear optics	Interaction between laser and matter	Light pulse compress, frequency conversion, elimination of distortion in transmission medium, nonlinear communications, all-optical switch, optical storage, slow light, high resolution spectrum analyze, and digital optical information processing
Fourier optics	Fourier optics theory and optical information processing	Holography, holographic storage, optical picture processing, simulation optical computing, and information safety
Guided wave optics	Light transmission and control in fiber and planar waveguide	Light transmission in fiber and planer waveguides; dispersion, loss, polarization, and power control, active and passive waveguide devices, coupling between fiber and device
Quantum optics	Non-classical optical phenomena	Using squeezed state to suppress the noise in optical communication; using quantum entangled state for quantum secrecy communication, quantum storage, quantum computing and quantum information processing

the other one is that the change of matter induces the change of parameters of the signal light propagating in the matter (frequency, power, wavevector, phase, pulsewidth, frequency spectrum, group velocity, propagation direction, etc.), thereby the "Controlling light with light" can be realized. In short, the laser physics studies the laser generation method; and the nonlinear optical studies the laser controlling method. Both are the basis of laser application. Therefore, the optics specialized graduate students must study nonlinear optics, besides study laser physics.

1.1.3 Nonlinear Optics Is a Basis of Photonic Technology

Figure 1.1 gives a comparison of development roadmaps of photonics and electronics; we find both have spectacular similarity [1].

Optics was founded after Newton published a book "Optics" to summary the achievement of geometrical optics in 1704, and Fresnel used the Huygens-Fresnel principle to explain the wave optics phenomena in 1818. Electromagnetism was founded after Coulomb established the Coulomb law in 1785 and Maxwell summarized the electromagnetic field theory in l873.

Electromagnetism and Optics are subdisciplines of physics, both born in 18–19th century. Electrotechnology was generated due to the application of Electromagnetism; and Engineering Optics was generated due to the application of Optics.

In 1906 human invented electronic valve to solve the problem of "Controlling electrons", then the Elections start established. However, in that time the electronic valve is the vacuum electronic valve, so the electronics was called "Vacuum electronic valve electronics". Utile in 1948 the semiconductor transistor was invented; the "Solid state electronics" was born.

Since 1960 the micrometer semiconductor integrated circuit was invented, the "Microelectronics" and "Microelectronic technology" were generated. The next

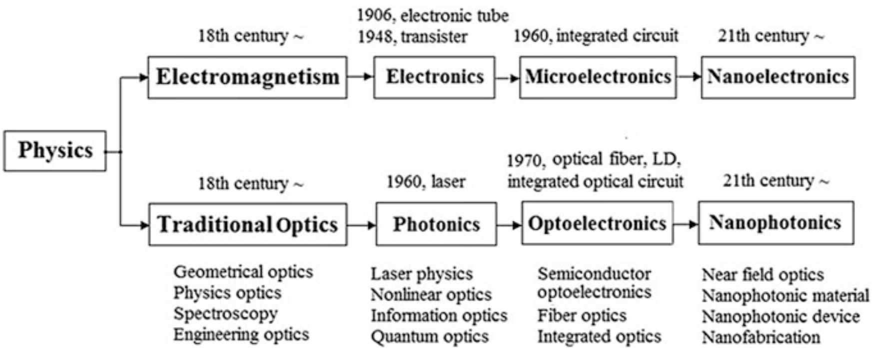


Fig. 1.1 Comparison between two development roadmaps of photonics and electronics

step of electronics should be “Nanoelectronics” and “Nanoelectronic technology”. The nanoelectronic devices, such as nanotube and graphite devices will be used. Due to the inherent limitation in the bandwidth, capability, transmission speed, etc. the electronic technology meets a bottleneck for further developing; it cannot satisfy the requirement of current modern science and technology. Therefore developing photonic science and technology is necessary tendency.

The traditional optics start from 18th century is based on the nature light source. Since 1960 the invention of laser, the modern optics based the stimulated radiation light source was born. The appearance of the different lasers, such as gases, solid, dye, semiconductor and chemical lasers and their widespread application formed a new “Laser technology”.

Laser is a generator of photons at same state. So that the born of laser means the born of photonics. Photonics is a science to study photon’s generation, transmission, control, detect, display, storage, and interaction with matter, it is the core content of modern optics.

After laser born, over 10 years development, it appeared low-loss silica fiber, room temperature heterojunction semiconductor laser diode, and various semiconductor photoelectrical integrated devices and waveguide devices, for instance, lasers, optical amplifiers, modulators, detectors, optical switch and grating sensors, and different fibers, these devices have micrometer size. Therefore, the photonics in this stage can call Microphotonics (corresponding to Microelectronics). Because the main feature of Microphotonics are “controlling light with electricity” for the devices and “optical and electrical hybrid” for the systems, so the photonics in this stage can also be called as “Optoelectronics”, the corresponding technology is optoelectronic technology. The optoelectronic technology contains following four parts: the information optoelectronic technology; the energy optoelectronic technology; the material optoelectronic technology and the biomedical optoelectronic technology. All of these technologies are related to the interaction between laser and matter, so nonlinear optics is a basis of optoelectronic technology.

The next stage of photonics is “Nanophotonics”. In that stage will adopt the nanophotonic technology and the combinative ultrafast-photonic technology, the target of Nanophotonics is to realize the “Controlling light with light”, namely the all-optical technology. Because photons are not charged, it cannot directly realize “Controlling light with light” by interaction between photons similar to electrons in the transistor. The only way is use nonlinear optics method indirectly to realize “Controlling light with light”. That is through a pump light to change the parameters of medium or the micro-stricture of medium to control the frequency, amplitude, phase, polarization, or group speed of the signal light, which is propagating in the medium. Up to now, using nonlinear optics method people can control the frequency (or wavelength) of the signal light, but cannot control the amplitude (or power) of the signal light, that means the all-optical switches cannot be made. The all-optical switching is a basic technology for optical digital information processing. The all-optical switch is a basic device of all-optical communication and all-optical computer in the future.

In the latter half of 20th century, laser source problem has well solved, but the laser controlling problem is not solved, this is an important problem to be solved. For this purpose, nonlinear optics will give play to key action. In short, the nonlinear optics is an important basis of photonics and its applications.

1.2 Physical Meaning of Nonlinear Optics

What is the nonlinear optics? We can explain his physical meaning from the viewpoint of that the light induces the polarization of medium.

1.2.1 Phenomenon Related with High-Order Polarization

When a common light with an electrical field strength (or amplitude) $\boldsymbol{E}$ irradiates the medium, under the action of light electrical field, the electric charge of molecule and atom in the medium occurs overall or relative displacement, induced a secondary light electrical field, which is described by a physical quantity-electric polarization $\boldsymbol{P}$, the induced polarization depends linearly on the electrical field amplitude, the relationship between $\boldsymbol{P}$ and $\boldsymbol{E}$ is

$$\boldsymbol{P} = \varepsilon_0 \boldsymbol{\chi}^{(1)} \cdot \boldsymbol{E}, \tag{1.2.1}$$

where the constant of proportionality $\boldsymbol{\chi}^{(1)}$ is the linear susceptibility, which is a complex number tensor for the anisotropic medium; ε_0 is the dielectric coefficient in the vacuum.

If the incident light is a laser, its intensity is much higher than common light in several orders of magnitude. We can make the expansion of electric polarization into a power series of light electrical field amplitude, the relationship between $\boldsymbol{P}$ and $\boldsymbol{E}$ for the anisotropic medium is

$$\boldsymbol{P} = \varepsilon_0 \boldsymbol{\chi}^{(1)} \cdot \boldsymbol{E} + \varepsilon_0 \boldsymbol{\chi}^{(2)} : \boldsymbol{EE} + \varepsilon_0 \boldsymbol{\chi}^{(3)} \vdots \boldsymbol{EEE} + \cdots, \tag{1.2.2}$$

where $\boldsymbol{\chi}^{(1)}$ is linear susceptibility, $\boldsymbol{\chi}^{(2)}$ and $\boldsymbol{\chi}^{(3)}$ are the second- and third-order nonlinear susceptibilities, respectively. $\boldsymbol{\chi}^{(1)}$, $\boldsymbol{\chi}^{(2)}$ and $\boldsymbol{\chi}^{(3)}$ are second-, third- and fourth-rank tensors, respectively. The symbols "·", ":" and "⋮" are denoted second-, third- and fourth-rank tensor multiplication operations, respectively. In the right side of Eq. (1.2.2), the first item is linear polarization, the second item, third item, and so on are high-order nonlinear polarizations, so Eq. (1.2.2) can be denoted by electric polarizations:

$$\begin{aligned} \boldsymbol{P} &= \boldsymbol{P}^{(1)} + \boldsymbol{P}^{(2)} + \boldsymbol{P}^{(3)} + \cdots \\ &= \boldsymbol{P}_L + \boldsymbol{P}_{NL}, \end{aligned} \tag{1.2.3}$$

where first item is linear polarization, which is denoted by

$$\boldsymbol{P}_L = \boldsymbol{P}^{(1)}, \tag{1.2.4}$$

the following items are high-order nonlinear polarization, which is denoted by

$$\boldsymbol{P}_{NL} = \boldsymbol{P}^{(2)} + \boldsymbol{P}^{(3)} + \cdots. \tag{1.2.5}$$

In short, nonlinear optics studies the phenomenon related with the high-order nonlinear polarization of the medium.

1.2.2 Nonlinear Response of Medium to the Optical Field

Now we suppose the medium is an isotopic homogeneous medium, in Eq. (1.2.2), the $\boldsymbol{\chi}$ can be written to the scalar quantity, $\boldsymbol{P}$ and $\boldsymbol{E}$ are the vector quantities, then Eq. (1.2.2) can be expressed as

$$\begin{aligned} \boldsymbol{P} &= \varepsilon_0\chi^{(1)}\boldsymbol{E} + \varepsilon_0\chi^{(2)}|\boldsymbol{E}|\boldsymbol{E} + \varepsilon_0\chi^{(3)}|\boldsymbol{E}|^2\boldsymbol{E} + \cdots \\ &= \boldsymbol{P}^{(1)} + \boldsymbol{P}^{(2)} + \boldsymbol{P}^{(3)} + \cdots, \end{aligned} \tag{1.2.6}$$

The linear polarization, second-order nonlinear polarization and third-order nonlinear polarization can be expressed as

$$\boldsymbol{P}^{(1)} = \varepsilon_0\chi^{(2)}\boldsymbol{E}, \tag{1.2.7}$$

$$\boldsymbol{P}^{(2)}=\varepsilon_0\chi^{(2)}|\boldsymbol{E}|\boldsymbol{E}, \tag{1.2.8}$$

$$\boldsymbol{P}^{(3)} = \varepsilon_0\chi^{(3)}|\boldsymbol{E}|^2\boldsymbol{E}. \tag{1.2.9}$$

We can see that in the case of linear polarization, the polarization of medium is proportional to the field strength; however in the case of second-order nonlinear polarization, the polarization of medium is proportional to the square of field strength; in the case of third-order nonlinear polarization, the polarization of medium is proportional to the triple of field strength. In conclusion, in the nonlinear polarization case, the polarization of medium is not proportional to the field strength.

Therefore, Nobel laureate Bloembergen [2], an authoritative scholar in nonlinear optics, gave a strict scientific definition of nonlinear optics: “If the response of matter to the impressed electromagnetic field is not a linear function of the impressed electromagnetic field strength, this optical phenomenon belongs to the field of nonlinear optics".

It is worth noting that the definition said the electromagnetic field, namely the light field, which is composed by electrical field and magnetic field. Actually, the light magnetic field also has effect to the magnetic polarization of the medium. Especially in the material composed by asymmetry chiral molecules the effect is stronger. However, for the common materials, the effect of light magnetic field to the medium is very weak, it can be ignored. Therefore, in the general nonlinear optics books, people only consider the contribution of light electrical field to the nonlinear optical effects of the medium.

1.2.3 Parameters of Medium Are Function of Optical Field

If we only consider the linear optical effect and third-order nonlinear optical effect, the total polarization of the medium is

$$\boldsymbol{P} = \varepsilon_0(\chi^{(1)} + \chi^{(3)}|\boldsymbol{E}|^2)\boldsymbol{E} = \varepsilon_0(\chi^{(1)} + \Delta\chi^{(1)})\,\boldsymbol{E}, \tag{1.2.10}$$

where

$$\Delta\chi^{(1)} = \chi^{(3)}|\boldsymbol{E}|^2 \propto \chi^{(3)}\boldsymbol{I} \tag{1.2.11}$$

is the variation quantity of first-order susceptibility induced by the electrical field of impressed light, which is proportional to the square of light field amplitude, i.e., the light intensity $\boldsymbol{I}$.

Because the third-order nonlinear susceptibility is a complex number, it can be written to real and imaginary two parts:

$$\chi^{(3)} = \chi^{(3)\prime} + i\chi^{(3)\prime\prime}. \tag{1.2.12}$$

In Chap. 2 we will prove that, the reflective index and the absorption coefficient of the medium also can be divided into linear and nonlinear two parts:

$$n = n_0 + \Delta n, \tag{1.2.13}$$

$$\alpha = \alpha_0 + \Delta\alpha, \tag{1.2.14}$$

where n_0 is the linear reflective index; Δn is the nonlinear reflective index; α_0 is the linear absorption coefficient; $\Delta\alpha$ is the nonlinear absorption coefficient. And the nonlinear reflective index is related with the real part of susceptibility; the nonlinear absorption coefficient is related with the imaginary party of susceptibility; and both are proportional to the light intensity $I \propto |E|^2$:

$$\Delta n \propto (\chi^{(3)\prime})I, \quad (1.2.15)$$

$$\Delta\alpha \propto (\chi^{(3)\prime\prime})I. \quad (1.2.16)$$

In conclusion, for the third-order nonlinear effect, the variation of susceptibility, the reflective index and the absorption coefficient are all linearly related with light intensity; in other word, they are the function of optical field amplitude. Therefore, we can generally say that the nonlinear optical phenomenon is the phenomenon in which the variation of optical parameters of the medium is the function of the optical field amplitude.

1.3 Research Content of Nonlinear Optics

1.3.1 Typical Nonlinear Optical Effects

What is difference between nonlinear optics and linear optics? The basic difference has two: firstly, the light sources of both are different: the linear optics is used the incoherent and low power nature light; the nonlinear optics is used the coherent and high power laser. Secondly, in the case of linear optics, the optical parameters of the medium are independent with the light field; but in the case of nonlinear optics, the parameters of medium, such as refractive index, absorption coefficient, susceptibility, etc. are the function of the light electrical field amplitude.

Now we list the following 10 nonlinear optical effects to further explain the difference between the nonlinear optics and the linear optics.

1. **Optical Kerr Effect**

When a laser beam passes through a medium, the refractive index of the medium will be changed. The variation quantity is proportional to the light intensity, that is

$$n = n_0 + n_2 I, \quad (1.3.1)$$

where n_0 is the linear refractive index of medium. I is light intensity in medium. n_2 is the nonlinear refraction coefficient. n_2 can be positive or negative, it depends on the medium. So the refractive index of medium can be increased or decreased with increase of light intensity. However, in the linear optics case, when the light beam passes though the medium, the refractive index of medium cannot be changed. It will keep the constant n_0.

2. **Self-focusing and Self-defocusing**

When a Gaussian-type laser beam passes through a medium, due to the radial distribution of light intensity is ununiformed, the Kerr effect induced the refractive index in radial direction is gradually changed, to form a like-convex lens

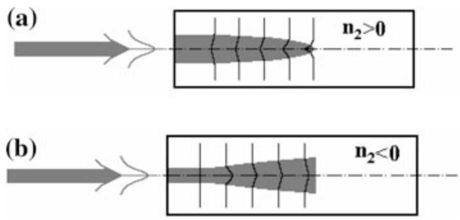


Fig. 1.2 Nonlinear refraction phenomena: **a** self-focusing; **b** self-defocusing

(for $n_2 > 0$) effect or a like-concave lens effect (for $n_2 < 0$), so that the transverse dimension of light beam in the medium becomes small and small, i.e., self-focusing, or becomes large and large, i.e., self-defocusing. These two nonlinear refraction effects are show in Fig. 1.2.

However, in the linear optics case, the refractive index not changes with light intensity, so the self-focusing and self-defocusing cannot occur.

3. **Nonlinear Absorption**

In the condition of resonance interaction between laser and medium, the absorption coefficient of medium α can be changed with increase of light intensity until saturation. The absorption-coefficient change is different for different material: if α decreases with increase of light intensity, i.e., the saturation absorption as shown in Fig. 1.3a; if α increase with increase of light intensity, i.e., the reverse saturation absorption, as shown in Fig. 1.3b.

In the linear optics case, the light passes though the medium, the absorption coefficient cannot be change with the light intensity; it is a constant of α_0.

4. **Multiwave Mixing**

In nonlinear optics, when a number of laser beams transmit in the medium, it is possible occurrence of the energy (or frequency) interconversion among the laser beams, in the same time the light with new frequency will be generated. This is the multiwave mixing effect. Usually we can see the three-wave mixing or the four-wave mixing. For example, two light beams at same frequency input a nonlinear crystal in the same time to produce a new light beam at the double frequency. This is optical frequency-doubled effect as shown in Fig. 1.4a. In the another

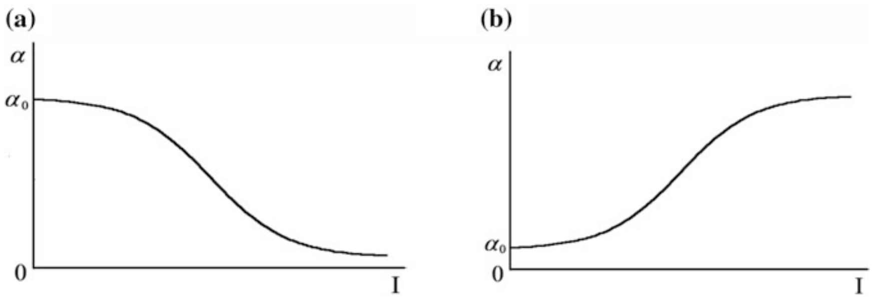


Fig. 1.3 Nonlinear absorption phenomena: **a** saturable absorption; **b** revers saturable absorption

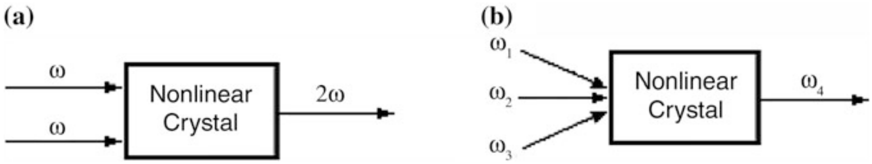


Fig. 1.4 Multiwave mixing phenomena: **a** optical frequency doubling; **b** four-wave mixing

example, when three lights at different frequency input a nonlinear crystal, it is possible to produce a new light at the frequency that is the sum of the frequencies of three input beams, the four-wave mixing is shown in Fig. 1.4b.

However in the linear optics case, when a number of light beams cross transport in the medium, the light beams cannot reciprocally interchange their energies; even cannot produce a new light.

5. **Phase Conjugation**

When a number of laser beams transmit in the medium, they could mutual transfer their phase signals, and it is possible to lead the phases of two beams become a conjugation relation. Figure 1.5 shows that when we input a signal light $(\boldsymbol{E}_{\mathrm{p}})$ at frequency ω into a nonlinear medium, and at the same time reversely input two strong-power pump lights $(\boldsymbol{E}_1 \text{ and } \boldsymbol{E}_2)$ into the medium, in the reverse direction of signal light can produce a new light at same frequency ω, with an amplitude that is the conjugated amplitude of the signal light. The new light is called the conjugated light $(\boldsymbol{E}_{\mathrm{c}})$. The conjugated two lights $\boldsymbol{E}_{\mathrm{p}}$ and $\boldsymbol{E}_{\mathrm{c}}$ are expressed in the right side of Fig. 1.5.

However in the linear optics case, a number of light beams transit in the medium, they cannot mutually transfer their phase signals and the phase conjugation effect cannot appear.

6. **Stimulated Raman Scattering**

A laser beam inputs the medium and interacts with the molecules of medium to generate a series of stimulated-radiation scattered lights at different frequencies, this is stimulated Raman scattering. The spectrogram of stimulated Raman scattering is shown in Fig. 1.6. The longer dotted line at the center of spectrum is the incident laser. The spectral lines in both sides are multi-order scattering lights. The lights at lower frequency in the left side are called Stokes scattering lights; the lights at higher frequency in the right side are called anti-Stokes scattering lights.

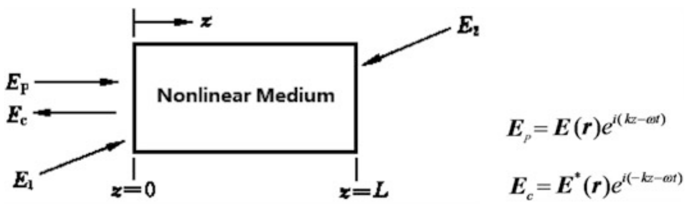


Fig. 1.5 Schematic diagram of phase conjugation effect

Fig. 1.6 Stimulated Raman scattering spectrogram

The common light interacting with molecules also can generate the scattering lights at different frequencies, but these scattering lights are just common lights. And only appear the first-order stokes scattering light and the first-order anti-stokes scattering light, without multi-order scattering phenomenon.

7. **Optical Bistability**

A laser beam with the intensity I_{i0} inputs into an F–P optical cavity containing a nonlinear medium, it can produce two possible transmitted light intensities I_{t1} and I_{t2}, such device is the optical bistable device. The characteristic curve of optical bistability is shown in Fig. 1.7.

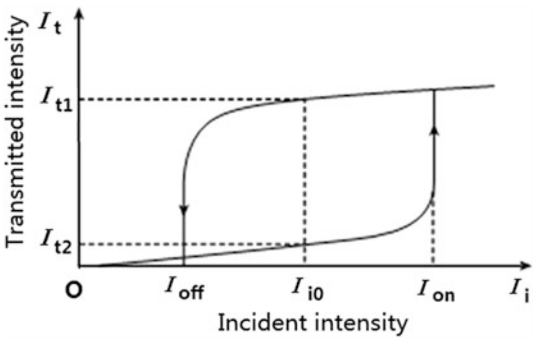
However in the case of linear optics, when a common light passes through an F–P cavity, the transmitted light intensity is a linear function of the incident light intensity only.

8. **Nonlinear Optical Limiting**

A laser beam passes through an optical device containing a nonlinear material; its transmittance may decreases with increase of optical intensity. When the light intensity is strong enough, the transmittance can reduce to the zero, as shown in Fig. 1.8. This is the characteristic curve of a nonlinear optical limiter. The slop of the curve is the transmittance $T = \frac{dP_T}{dP_I}$, which changes with the incident power P_I. When $P_I = 0, T = T_0$, T_0 is linear transmittance. When $P_I = P_{I0}, T = 0$, the transmitted power P_T is limited below P_{T0}.

However in the case of linear optics, when a light beam passes through the material, the transmittance is a constant; it cannot be changed with the light intensity.

Fig. 1.7 Characteristic *curve* of the optical bistability



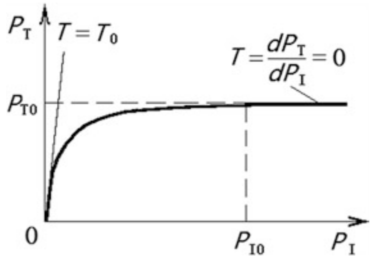


Fig. 1.8 Characteristic *curve* of nonlinear optical limiting

9. **Optical Soliton**

When a common light pulse takes a long-distance transmission in the optical fiber, its pulsewidth will be broaden with increase of the transmission distance due to the dispersion of fiber, as shown in Fig. 1.9a. However, if it is a laser pulse, and transmission fiber is selected by an anomalous dispersion fiber, the pulsewidth can keep a constant, that is because the nonlinear self-phase modulation counteracting the group dispersion of fiber to form a time optical soliton, as shown in Fig. 1.9b.

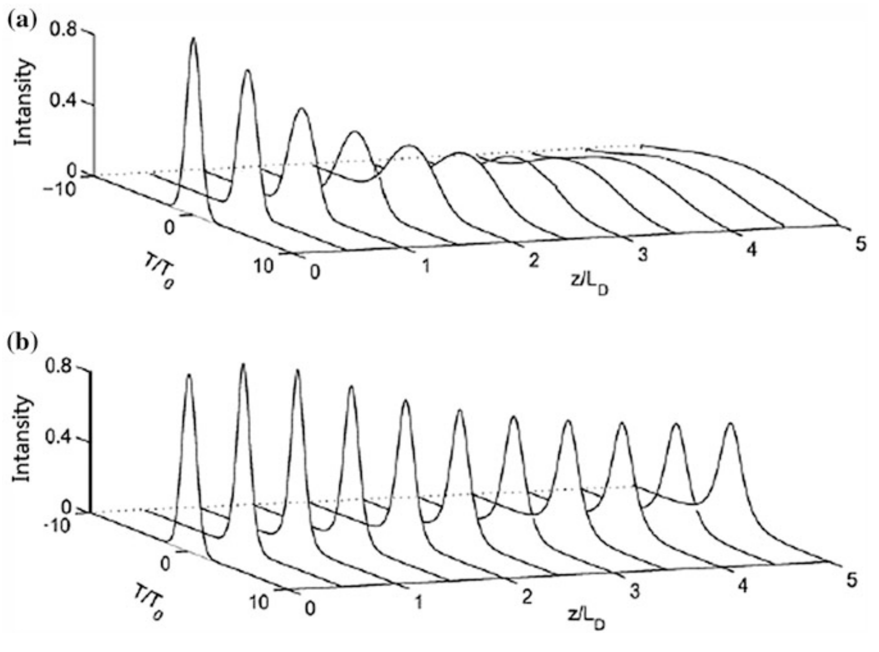


Fig. 1.9 The formation of time optical soliton in the fiber: **a** the dispersion of common light pulse makes the pulsewidth broaden; **b** the self-phase modulation of laser pulse balances the dispersion, to keep the pulsewidth as a constant

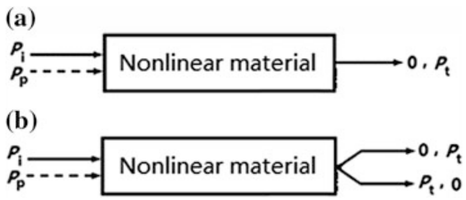


Fig. 1.10 Schematic diagram of all-optical switches: **a** intensity-type all-optical switch; **b** space-type all-optical switch

10. **All-optical Switch**

Because the photon is not charged, it cannot use a light directly to control the intensity or direction of the other light. However, in the nonlinear optics case, we can use a strong laser (pump light) to change the optical parameter of the medium, and then realize the controlling of the signal light passing through the medium, changing its intensity or propagation direction as shown in Fig. 1.10a, b, respectively. In the figure, P_P is the power of the pump light, P_i and P_t are the input and output powers of the signal light, respectively.

1.3.2 Two Kinds of Nonlinear Optical Effects

According to the difference of energy conversion process we can divide the nonlinear optics into two kinds: the passive nonlinear optics and the active nonlinear optics.

1. **Passive Nonlinear Optics**

The feature of the passive nonlinear optics is that in the nonlinear optical process, as a result the energy exchange between light and medium is nonoccurrence; however the energy exchange among the light fields at different frequencies is occurrence, in addition the light field at a new frequency may produce. All the lights including the original lights and the produced new light in the mixing process obey the energy conservation law and the momentum conservation law. The passive nonlinear optical process includes, for example, the frequency doubling, three-wave mixing, parameter processing, four-wave mixing, phase conjugation, etc. in which, the frequency doubling and the four-wave mixing have shown in Fig. 1.4.

2. **Active Nonlinear Optics**

The feature of the active nonlinear optics is that the energy exchange between light and medium is occurrence; and the change of optical parameter of medium is associated with the light electrical field amplitude, for example, the nonlinear absorption (saturable absorption, reverse saturable absorption, two-photon absorption, etc.); the nonlinear refraction (optical Kerr effect, self-focusing and self-defocusing, saturable refraction and reverse saturable refraction, two-photon refraction, etc.); the nonlinear scattering (stimulated Raman scattering, stimulated

Brillouin scattering, etc.), optical bistability and optical limiting, etc. The most of these active nonlinear optical processes have explained in above section.

1.3.3 Nonlinear Optical Materials

The nonlinear optical materials with different nonlinear mechanisms have many kinds. Table 1.2 lists some typical nonlinear optical materials and their nonlinear optical mechanisms.

What kind material is good nonlinear material? According to requirements of the device made by nonlinear materials, which should be:

1. The material has large nonlinear refraction coefficient n_2;
2. The material has low linear absorption coefficient α_0, with high optical transparency;
3. The material has short response time to impressed light field;
4. The material has simple and low-cost fabrication processing.

However, above requirements are contradicted with each other. the materials with strong nonlinearity often has large absorption loss, for instance, the compound semiconductors have the strong nonlinearity, but they work in the large absorption spectrum area; and the materials with strong nonlinearity usually have slow response time, for example, the liquid crystals have strong nonlinearity, but they have slow response time, because its response is rely on the molecular rotation; the materials with high transparency and fast response time often have low nonlinearity, such as silicon and silica, because they have symmetrical molecular structure. Therefore, the selection of material should comprehensively consider the material properties and the requirements of device.

Table 1.2 Nonlinear optical materials and nonlinear mechanisms (or structures)

Nonlinear optical material	Nonlinear mechanism (or structure)
Semiconductor	Electron or exciton mechanism
Organic and polymer	Electron or molecular polarization
Electro-optical crystal	External electro-optical effect
Photorefractive material	Internal electro-optical effect
Liquid crystal	Molecular orientation polarization
Cluster material(C_{60}, etc.)	Molecular polarization
Chiral molecule material	Molecular electric moment and magnetic moment
Quantum confinement (quantum well, quantum wire, quantum dot)	Semiconductor with periodic alternative large and low energy gap structure
Photonic crystal (1D, 2D and 3D)	Dielectrics with periodic alternative high and low refractive index structure
Surface plasmon polaritons	Meter-dielectric interface nano structure

1.4 Development History of Nonlinear Optics

1.4.1 Brief History of Nonlinear Optics

The development history of nonlinear optics can be divided into the following several stages:

1. **Initial Foundation Stage of Nonlinear Optics**

Before laser invention, there are several theory predictions of the nonlinear optical phenomenon, for example, 1931 Goeppert and Mayer theoretically predicted the two-photon absorption; 1956 Buckingham proposed the theory of Kerr effect. However, in that time it is difficult to experimentally verify these theory predictions.

Since laser invention in 1960, scientists carried through a series of experimental studied and found a lot of nonlinear optical effects. In 1965 Nobel praise winner Bloembergen [3] has written a book "Nonlinear Optical Phenomena" to summarize the research achievements in that period time, and then founded the new discipline "Nonlinear optics". So from 1961 to 1965 is the initial foundation stage of nonlinear optics. The important events in this stage are:

1961, the discovery of frequency doubling in ruby laser by Franken et al. [4];

1962–1964, the discovery of stimulated Raman scattering and stimulated Brillouin scattering [5–7];

1962–1965, the discovery of sum frequency, difference frequency, parametric oscillation, four-wave mixing [8–10];

1963–1966, the discovery of saturable absorption, two photon absorption [11, 12];

1962–1964, the discovery of self-focusing and self-phase modulation [13, 14];

1965, Bloembergen [3] published the book "Nonlinear Optical Phenomena".

2. **Mature Stage of Nonlinear Optics**

After 1965, the scientists discovered many new nonlinear optical effects in experiments and made the theoretical explains. In 1984 the famous professor Shen [15] published a book "The Principles of Nonlinear Optics" to summary the research achievements in that period time. From 1965 to 1984 for near 20 years is mature stage of nonlinear optics. The significative events in this stage are:

1962–1975, the discovery of transient-state coherent optics effects (photon-echo, optical nutation, self-inductance transparency, etc.) [16];

1963–1983, the discovery of degenerate four-wave mixing and optical phase conjugation [17–19];

1964–1974, the study of optical Kerr effect and the verification by the experiment [20, 21];

1975–1985, the discovery of optical bistability and optical chaos [22, 23];

1972–1987, the study of nonlinear optical properties in fiber and optical soliton [24–26];

1984, Professor Shen [15] published the book "The Principles of Nonlinear Optics".

3. **Application Development Stage of Nonlinear Optics**

After 1984 scientists continually found many new nonlinear optical phenomena, the theory of nonlinear optics became more mature. The application of nonlinear optics has a large development, especially in the applications of the laser science and technology, the information science and technology and the material science and technology. Since 1984–2015 for 30 years is the application development stage of nonlinear optics. The major events in this stage are:

1982–1998, the study of nonlinear optical properties of semiconductor quantum confinement materials [27, 28];

1985–1989, the discovery of high efficient nonlinear optical crystal materials BBO and LBO [29, 30];

1985–1991, the study of nonlinear optical properties of organic and polymer [31, 32];

1987–1995, the discovery of reverse saturable absorption and optical limiting effects [33–35];

1979–1993, the study of photorefractive effect and its nonlinear optical theory [36, 37];

1985–1997, the application in quantum optics (squeezed state and quantum entanglement, etc.) [38, 39];

1985–1999, the application in nonlinear interferometer-type all-optical switches [40–42];

1984–2001, the application in optical fiber communication, optical soliton communication and nonlinear communication, etc. [43, 44];

1979–2002, the application in photonic crystal [45, 46];

1995–2005, the application in chiral molecular materials [47, 48];

1995–2006, the application in light speed controlling (slow light and fast light) [49–52];

2006–2015, the application in surface plasmon polaritons technology [53, 54].

1.4.2 Development Tendency of Nonlinear Optics

In this half of century, the nonlinear optics continually and rapidly developed in depth and breadth, its development tendency displayed on the several respects: the study objects, the study in time and space scales, and the nonlinear optical materials.

1. **Study Objects**

The development tendency of the nonlinear optics study is from strong light source to the weak light source; from the resonance area to the non-resonance area; from the fundamental state-excited state transition to the excited state-excited state transition; from the two-energy mode to the multi-energy mode; from the local area to the non-local area, etc.

2. **Space Scale**

For different space scale of material system, the law of interaction between light and material is different. It needs use different theory to study. The macroscopic scale system should use the wave optics theory; the microscopic scale system should use the quantum optics theory; now the studied heat point is turned to the nano-scale material system, in the nano-scale material system we should use the near-field optics theory. However, the nonlinear optics theory in the nano-scale system is not mature yet, it needs further development.

3. **Time Scale**

The study of nonlinear optics is developed from the steady-state process to the quasi-steady-state process, and to the transient-state (dynamic state) process; the different process should use different theory. To study the steady-state process based on the continues light or the quasi-steady-state process based on the microsecond-pulsed light, we can use the steady-state nonlinear wave equations, which are not contained the time derivative; but to study the transient-state process based on the nanosecond, picosecond or femtosecond short-pulsed light, we should use nonlinear wave equations contained first-order or second-order derivatives.

4. **Nonlinear Optical Material**

From the inorganic material to the organic material; from the amorphous material to the crystalline material; from the symmetric material to the asymmetric material (chiral material); from the homogenous material to the composite material; form the high dimensional material to the low dimensional material, such as 3D balk, 2D surface and thin film, ID line, and OD particle; from macroscopic material to nano-structure material, such as semiconductor quantum well, quantum line, and quantum dot; 3D, 2D, and 1D photonic crystal; metal-dialectic intersurface; nanotube, nano sphere and nano cluster, etc.

1.5 Applications of Nonlinear Optics

What are the applications of nonlinear optics? From the current point of view, the applications of nonlinear optics mainly have following three aspects: in the laser technology; in the information technology and in the materials technology [55].

1.5.1 Application in Laser Technology

1. **New Laser Device**

Based on the principles of nonlinear optics, scientists have made various new type laser devices, such as the ultrashort pulsed lasers, the wavelength or power tunable lasers, the stimulated Raman laser, the nonlinear fiber laser, the soliton laser, the terahertz laser, the nanoscale laser, etc. In addition, the nonlinear optics also can use for the laser mode selection, the laser power stabilization and the laser parameter measurement, etc.

2. **Laser Pulse Compression**

The Q-switching and mode-locking technologies are based on the nonlinear optics. These technologies are used for compression of the laser pulsewidth to generate the picosecond, femtosecond and attosecond ultrashort pulsed lasers.

3. **Laser Frequency Conversion**

Using nonlinear optical technologies, such as the frequency doubling; sum frequency; difference frequency; parametric amplification and oscillation; third harmonic generation; four-wave mixing; stimulated scattering, etc., the various laser-frequency conversions are realized.

4. **Laser Transportation**

The important application in adaptive optical technology is based on the optical phase conjugation of nonlinear optics, for example, the compensation of the light beam distortion in the laser atmospheric transmission; also can be used to solve the problem of light beam distortion in laser nuclear fusion, etc.

5. **Laser Protection**

It has significance in militarily to protect the laser blind weapons, which are using the laser pulses to damage the human eye and the photodetectors. The nonlinear optical limiters based on the principle of nonlinear optics are advantageous in compare with the linear optical limiters.

1.5.2 Application in Information Technology

1. **Optical Communication**

In currant optical communication including the fiber optical communication and the space optical communication, there are many technologies based on nonlinear optics, for instance, the semiconductor laser, Raman optical amplifier, optical modulator, self-focusing lens, optical switch, wavelength convertor, optical delayer, optical add drop multiplexer, optical switching, optical cross connect, etc. Novel optical communication technology, for example, the coherent optical communication, optical soliton communication, optical chaos communication, optical quantum communication, and future all-optical communication are all associated with nonlinear optics.

2. **Optical Computing**

The digital optical information processing is the development direction of future information processing technology. The key technologies used in future optical computer, such as the all-optical computer, optical logic gate, optical number arithmetic device, 3D two-photon optical storage, optical amplifier, all-optical switch, optical clock-signal generator, etc., they all establish on the basis of nonlinear optics.

3. **Optical Sensing**

The distributed fiber sensor based on Raman and Brillouin fibers is a new type fiber sensing technology. In order to realize all-optical sensing network (internet of things), the all-optical switch made by nonlinear fiber grating is a key device. In the laser remote sensing technology, the nonlinear optics also plays an important role.

1.5.3 Application in Material Technology

1. **High Resolution Spectrum Analysis**

Taking advantage of ultrafast laser pulse excited stimulated Raman scattering, four-wave mixing, second harmonics, two-photon absorption, etc. nonlinear optical effects, people have developed the nonlinear spectrum analysis technology with high space resolution and high time resolution, extensively used to analyze and measure the state and structure of the atoms and molecules, the hyperfine structure of energy level, and change of physical and chemical cluster structure, activity of biological cell, etc. The combination of the nonlinear spectrum technology and the near-field optical microscopy forms imaginable near-field spectrum technology, to be used for analyzing state change of nano-structure, the luminescence process of matter and chemical reaction process, etc.

2. **Micromachining of Material**

The laser direct writing high precision processing by using multiphoton absorption (such as two-photon absorption) and multiphoton ionization has been used in the preparation of nanometer materials and the process of nano structure. In order to improve the luminous efficiency of solid light-emitting devices (LED) and the absorption efficiency of solar cells, to make microstructure graphics on the surface of the device with a laser micro machining method is an effective measure.

3. **Investigation of New Material**

The nonlinear optics method can be used for investigation of the following materials: the surface, intersurface, cluster etc. low dimensional materials; the quantum well, quantum line and quantum dot, etc. quantum confinement materials; 1D, 2D and 3D photonic crystal materials; the surface plasmon polariton material; the nanosphere, nanocavity, nanotube etc. nanoscale materials; the negative refractive index materials; the chiral materials; the biologic cell materials, etc. and measuring the nonlinear optical parameters of these new materials.

At present the applications of nonlinear optics in laser technology include the laser device technology, the laser frequency conversion technology, the laser transmission technology, etc. are comparative maturity, however, the laser protection technology (nonlinear limiter) is not well maturity; in the application of material technology, the extensive application is high resolution nonlinear spectrum analysis technology. The laser microprocessor technology has had a lager progress. In respect of nonlinear parameter measuring, Z-scan technology has had larger development, but the measurement accuracy and the stability should be enhanced. In the respect of information technology application, nonlinear optics has big potential. But the optical digital processing technology, such as all-optical switch and its application in all-optical communication, all-optical computer and all-optical sensing network, is still in foundational research stage. We hope after the research of nanometer devices obtains a great progress in the near future, it will have a successful application.

In shout, nonlinear optics is the basis of nowadays and future photonic technology, it will powerfully promote the information photonic technology, the energy photonic technology, the material photonic technology and the biologic technology development in an all-around way.

Review Questions of Chapter 1

1. What is the important significance to study nonlinear optics?
2. Take examples to explain what is the difference between linear optics and nonlinear optics?
3. What are relationships between the polarization of medium and the electrical field amplitude of incident light in two-order and three-order nonlinear optical effects?
4. What is difference between the passive nonlinear optics and the active nonlinear optics?
5. How many kinds of the nonlinear optical materials there are? What is their nonlinear mechanism?
6. Please introduce the development stages of nonlinear optics. What is the development trend of nonlinear optics?
7. To compare the development processes of photonics and electronics, what are their similarities and differentia?
8. Why nonlinear optics is an important basis of photonics? What are the mature applications and the development potential?

References

1. S. Liu, C. Li, *Photonics: Technology and Application* (Guangdong Science and Technology Publishing house, Guangzhou, China, 2006)
2. N. Bloembergen, *Nonlinear Spectroscopy, Proceedings* (North-Holland, Amsterdam, 1977)
3. N. Bloembergen, *Nonlinear Optics* (Benjamin, New York, 1965)
4. P.A. Franken, C.W. Hill et al., Generation of optical harmonics. Phys. Rev. Lett. **7**(4), 118–119 (1961)
5. G. Eckhardt, R.W. Hellwarth et al., Stimulated Raman scattering from organic liquids. Phys. Rev. Lett. **9**(11), 455–457 (1962)
6. E. Garmire, E. Pandarese, C.H. Townes, Coherently driven molecular vibrations and light modulation. Phys. Rev. Lett. **11**(4), 160–163 (1963)
7. R.Y. Chiao, C.H. Townes, B.P. Stoicheff, Stimulated Brillouin scattering and coherent generation of intense hypersonic waves. Phys. Rev. Lett. **12**(21), 592–595 (1964)
8. N. Bloembergen, J. Ducuing, Experimental verification of optical laws of non-linear reflection. Phys. Lett. **6**(1), 5–6 (1963)
9. S.A. Akhmanov, A.I. Kovrigin, M.M. Strukov, Frequency dependence of threshold of optical breakdown in air. JETP Lett. **1**(1), 25–29 (1965)
10. J.A. Giordmaine, R.C. Miller, Tunable coherent parametric oscillation in $LiNbO_3$ at optical frequencies. Phys. Rev. Lett. **14**(24), 973–976 (1965)
11. A. Szoke, A. Javan, Isotope shift and saturation behavior of the 1.15-μ transition of Ne. Phys. Rev. Lett. **10**(12), 521–524 (1963)
12. W.F. Kosonocky, S.E. Harrison, Saturation of absorption and fluorescence in solutions of phthalocyanines. J. Appl. Phys. **37**(13), 4789–4797 (1966)
13. G.A. Askaryan, Interaction between laser radiation and oscillating surfaces. Sov. Phys. JETP **15**(6), 1161–1162 (1962)
14. M.J. Hercher, Laser-induced damage in transparent media. J. Opt. Soc. Am. **54**(4), 563–569 (1964)
15. Y.R. Shen, *The Principles of Nonlinear Optics* (Wiley, New York, 1984)
16. B.I. Stepanov, E.V. Ivakin et al., Recording two-dimensional and three-dimensional dynamic holograms in transparent substances. Sov. Phys. Doklady **16**, 46 (1971)
17. A. Yariv, D.M. Pepper, Amplified reflection, phase conjugation, and oscillation in degenerate four-wave mixing. Opt. Lett. **1**(1), 16–18 (1977)
18. L. Allen, J.H. Eberly, *Optical Resonance and Two-level Atoms* (Wiley, New York, 1975)
19. R.A. Fisher (ed.), *Optical Phase Conjugation* (Academic Press, New York, 1963)
20. P.D. Maker, R.W. Terhune, C.M. Savage, Intensity-dependent changes in the refractive index of liquids. Phys. Rev. Lett. **12**(18), 507–509 (1964)
21. G.K.L. Wong, Y.R. Shen, Transient self-focusing in a nematic liquid crystal in the isotropic phase. Phys. Rev. A **10**(4), 1277–1284 (1974)
22. S.L. McCall, H.M. Gibbs, T.N.C. Venkatesan, Optical transistor and bistability (A). J. Opt. Soc. Am. **65**, 1184 (1975)
23. H.M. Gibbs, *Optical Bistability: Controlling Light with Light* (Academic Press, 1985)
24. R.H. Stolen, E.P. Ippen, A.R. Tynes, Raman oscillation in glass optical waveguide. Appl. Phys. Lett. **20**(2), 62–64 (1972)
25. L.F. Mollenauer, R.H. Stolen, The soliton laser. Opt. Lett. **9**(1), 13–15 (1984)
26. V.M. Mitev, L.M. Kovechev, Soliton shaping by stimulated Raman process in silica optical fibers. Opt. Commun. **63**(6), 421–424 (1987)
27. D.A.B. Miller, D.S. Chemla et al., Large room temperature optical nonlinearity in $GaAs/Ga_{1-x}Al_x$ As multiple quantum well structures. Appl. Phys. Lett. **41**(8), 679–681 (1982)
28. H. Haug (ed.), *Optical Nonlinearities and Instability in Semiconductors* (Academic Press, 1998)
29. C.T. Chen, B.C. Wu, J. Aidong, A new-type ultraviolet SHG crystal β-BaB_2O_4. Sci. Sin **B28**, 235–243 (1985)

30. C.T. Chen, Y.C. Wu, J. Aidong, New nonlinear-optical crystal: LiB_3O_5. J. Opt. Soc. Am. B **6** (4), 616–621 (1986)
31. D.J. Williams, D.S. Chemla, J. Zuss (eds.), *Nonlinear Optical Properties of Organic Molecules and Crystals*, vol. 1 (Academic Press, New York, 1987)
32. P.N. Prasad, K.J. Williams, *Introduction to Nonlinear Optical Effects and Polymers* (Wiley, New York, 1991)
33. R. Adair, L.L. Chase, S.A. Payne, Nonlinear refractive-index measurements of glasses using three-wave frequency mixing. J. Opt. Soc. Am. B **4**(6), 875–881 (1987)
34. L.W. Tutt, S.W. McCahon, Reverse saturable absorption in metal cluster compounds. Opt. Lett. **15**(12), 700–702 (1990)
35. Chunfei Li, Jinhai Si, Miao Yang et al., Excited-state nonlinear absorption in multi-energy-level molecular systems. Phys. Rev. A **51**(1), 569–575 (1995)
36. N.V. Kukhtarev, V.B. Markov, et al., Holographic storage in electrooptic crystals. II. Beam coupling—light amplification. Ferroelectries **22**(1), 961–964 (1978)
37. P. Yeh, *Introduction to Photorefractive Nonlinear Optics* (Wiley, New York, 1993)
38. R.E. Slusher, L.W. Hollberg et al., Observation of squeezed states generated by four-wave mixing in an optical cavity. Phys. Rev. Lett. **55**(22), 2409–2412 (1985)
39. D. Bouwmeester, J.-W. Pan et al., Experimental quantum teleportation. Nature **90**(11), 575–579 (1997)
40. H.M. Gibbs, *Optical Bistability: Controlling Light with Light* (Academic Press, New York, 1985)
41. M.G. Kane, W. Glesk, J.P. Sokoloff, P.R. Prucnal, Asymmetric optical loop mirror: analysis of all-optical switch. Applied Opt. **33**(29), 6833–6841 (1994)
42. J.E. Heebner, R.W. Boyd, Enhanced all-optical switching by use of nonlinear fiber ring resonator. Opt. Lett. **24**, 847–849 (1999)
43. W. Zhao, E. Bourkoff, Propagation properties of dark solitons. Opt. Lett. **14**(13), 703–705 (1989)
44. G.P. Agrawal, *Application of Nonlinear Fiber Optics* (Academic Press, New York, 2001)
45. H.G. Winful et al., Theory of bistability in nonlinear distributed feedback structures. Appl. Phys. Lett. **35**(5), 379–381 (1979)
46. R.E. Slusher, B.J. Eggleton (eds.) *Nonlinear Photonic Crystals* (Springer, New York, 2002)
47. J.J. Maki, A. Persoons, Surface second-harmonic generation from chiral materials. Phys. Rev. B **51**(3), 1425–1434 (1995)
48. X. Wang, J. Li, C. Li, Three-coupled-oscillator model for nonlinear optical response of chiral molecules with tripod-like structure. Chem. Phys. **320**(1), 37–44 (2005)
49. V. Hau, S.E. Harris, Z. Dutton, C.H. Behroozi, Light speed reduction to 17 metres per second in an ultracold atomic gas. Nature **397**(18), 594–598 (1999)
50. K. Olga, Y. Rostovtsev, M.O. Scully, Stopping light via hot atoms. Phys. Rev. Lett. **86**(4), 628–631 (2001)
51. M.S. Bigelow, N.N. Lepeshkin, R.W. Boyd, Superluminal and slow light propagation in a room temperature solid. Science **301**, 200–202 (2003)
52. C.J. Chang-Hasnain, S.L. Chuang, Slow and fast light in semiconductor quantum-well and quantum-dot devices. J. Light-wave Technol. **24**(12), 4642–4654 (2006)
53. G.A. Wurtz, R. Pollard, A.V. Zayats, Optical bistability in nonlinear surface-plasmon polaritonic crystals. Phys. Rev. Lett. **97**(5), 057402-1–057402-4 (2006)
54. M. Kauranen, A.V. Zayats, Nonlinear plasmonics. Nat. Photon. **6**, 737 (2012)
55. E. Garmire, Nonlinear optics in daily life. Opt. Express **21**(25), 30532–30544 (2013)