Chapter 4
Optical Four-Wave Coupling Process

In this chapter we will start to study the third-order nonlinear optical phenomena. Because susceptibility $\chi^{(3)}$ is much smaller than $\chi^{(2)}$, the third-order nonlinear effect is much weaker than second-order nonlinear effect. However all mediums are common existence of the third-order nonlinear effect. This chapter mainly discuss the passive third-order nonlinear optics phenomena, in which the energy exchange only happens among light waves, these phenomena include the third harmonic, four-wave mixing and phase conjugation, etc. Subsequent chapters will discuss active third-order nonlinear optics phenomena, in which existing the energy exchange between light and medium, these phenomena include the optical Kerr effect, stimulated light scattering, nonlinear absorption and refraction, optical bistability, optical soliton, and nonlinear all-optical switch, etc.

4.1 Introduction to Third-Order Nonlinear Optical Effects

At first we generally discuss which specific nonlinear optical effects does the third-order nonlinear optical process contain?

Suppose that the incident light field $\boldsymbol{E}(t)$ is consisted of three monochromatic light fields at different frequencies propagating along a same direction:

$$\boldsymbol{E}(t) = \boldsymbol{E}_1(\omega_1)e^{-i\omega_1 t} + \boldsymbol{E}_2(\omega_1)e^{-i\omega_2 t} + \boldsymbol{E}_3(\omega_1)e^{-i\omega_3 t} + c.c. \tag{4.1.1}$$

The corresponding time-domain third-order nonlinear polarization in the isotropic medium is expressed as

$$\boldsymbol{P}^{(3)}(t) = \varepsilon_0\chi^{(3)}\boldsymbol{E}^3(t). \tag{4.1.2}$$

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Substituting Eq. (4.1.1) into Eq. (4.1.2), and combining the items with the same frequency, we obtain

$$\boldsymbol{P}^{(3)}(t) = \sum_i \boldsymbol{P}^{(3)}(\omega_i) e^{-i\omega_i t}, \tag{4.1.3}$$

where i is the positive integer, $\boldsymbol{\omega}_i$ contains all kinds of frequency components, which is the frequency of polarization field composed of frequencies of three monochromatic light fields $\boldsymbol{\omega}_1, \boldsymbol{\omega}_2, \boldsymbol{\omega}_3$ in different way.

For different third-order nonlinear optical effect, there is different susceptibility $\chi^{(3)}(\omega_i)$ corresponding to the nonlinear polarization in the frequency domain:

$$\boldsymbol{P}^{(3)}(\omega_i) = D\varepsilon_0 \chi^{(3)}(\omega_i)\boldsymbol{E}(\omega_1)\boldsymbol{E}(\omega_2)\boldsymbol{E}(\omega_3), \tag{4.1.4}$$

where D is degeneration factor, for the third-order nonlinear effect, $n = 3$, if taking the degeneracy of m, then $D = n!/m! = 6/m!$. There are three values: when $m = 1$, $D = 6$; when $m = 2$, $D = 3$; when $m = 3$, $D = 1$. Below is a list of several typical third-order nonlinear optical effects, their polarization field frequency ω_i is equal to the combination of following different incident field frequency:

Third harmonic

$$(\boldsymbol{\omega}_1 + \boldsymbol{\omega}_1 + \boldsymbol{\omega}_1) \quad D = 1$$

Four wave mixing

$$(\boldsymbol{\omega}_1 + \boldsymbol{\omega}_2 + \boldsymbol{\omega}_3) \quad D = 6$$

Degenerate four wave mixing

$$(\boldsymbol{\omega}_1 - \boldsymbol{\omega}_1 + \boldsymbol{\omega}_1) \quad D = 3$$

Four wave mixing phase conjugation

$$(\boldsymbol{\omega}_1 + \boldsymbol{\omega}_2 - \boldsymbol{\omega}_3) \quad D = 6$$

Self-phase modulation optical Kerr effect

$$(\boldsymbol{\omega}_1 - \boldsymbol{\omega}_1 + \boldsymbol{\omega}_1) \quad D = 3$$

(Self-focusing, saturable absorption)
Cross phase modulation optical Kerr effect

$$(\boldsymbol{\omega}_2 - \boldsymbol{\omega}_2 + \boldsymbol{\omega}_1) \quad D = 6$$

(Two photon absorption)
Stocks Raman scattering

$$(\omega_1 - \omega_1 + \omega_2) \quad D = 6$$

Anti-Stocks Raman scattering

$$(\omega_1 + \omega_1 - \omega_2) \quad D = 3$$

In above third-order nonlinear optics phenomena, former 4 kinds belong to the passive nonlinear effects; later 4 kinds belong to the active nonlinear effects.

In general the passive third-order nonlinear optics process is a four-wave coupling process, there is the interaction of 4 light fields, which includes three extraneous light fields $E(\omega_1), E(\omega_2), E(\omega_3)$ and one polarization light field $E(\omega_i)$, in principle we need establish 4 nonlinear coupling equations to simultaneously solve these 4 light field amplitudes.

If some nonlinear optics process exists the small signal approximation, namely its polarization field is much weaker than the pump light field, in this case its nonlinear conversion efficiency is not high, we can regard that the pump light amplitude is unchangeable with the change of propagation distance in the nonlinear process, so we can omit the equation of that pump light field. In this way the number of the coupling equations can be reduced.

4.2 Optical Third Harmonic and Optical Four-Wave Mixing

4.2.1 Optical Third Harmonic

Optical third harmonic generation is a third-order nonlinear optics effect that the original light field at the frequency ω inputs the medium and generates a polarization light field at the frequency 3ω.

The principle of optical third harmonic effect is shown in Fig. 4.1. The three foundational frequency lights inputted into the third-order nonlinear medium are at the same frequency, namely, $\omega_1 = \omega_2 = \omega_3 = \omega$. According to the principle of conservation of energy, the frequency of new generated third harmonic light is $\omega_1 + \omega_2 + \omega_3 = 3\omega$.

Considering that, in the third harmonic effect, the foundational frequency light field $E(z, \omega)$ and the third harmonic light field $E(z, 3\omega)$ both are monochromic

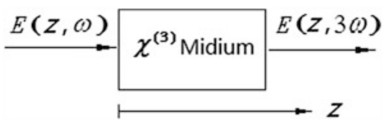

Fig. 4.1 Schematic diagram of optical third harmonic effect

plane waves propagating along z direction, the medium is non-absorption isotropic medium, we only consider the small signal approximation case, and the foundational frequency light field amplitude has no change in the z direction, i.e.

$$E(z,\omega) = E(0,\omega) = constant. \tag{4.2.1}$$

So we can omit 3 equations related 3 foundational frequency light field, only need consider the slowly-varying-amplitude approximation nonlinear wave equation corresponding to the third harmonic light field. If we adopt following notations:$E_{3\omega}(z) = E(z,3\omega)$,$E_{\omega}(z) = E(z,\omega)$, $P^{(3)}_{3\omega}(z) = P^{(3)}(z,3\omega)$, and $D = 1$, the wave equation can be written as

$$\frac{\partial E_{3\omega}(z)}{\partial z} = i\frac{3\omega}{2\varepsilon_0 c n_{3\omega}} P^{(3)}_{3\omega}(z)e^{-i\Delta kz}, \tag{4.2.2}$$

where the nonlinear polarization of third harmonic effect

$$P^{(3)}_{3\omega}(Z) = \varepsilon_0\chi^{(3)}(3\omega;\omega,\omega,\omega)E^3_{\omega}(Z). \tag{4.2.3}$$

Substituting Eq. (4.2.3) into Eq. (4.2.2), we obtain

$$\frac{\partial E_{3\omega}(Z)}{\partial z} = i\frac{3\omega}{2cn_{3\omega}}\chi^{(3)}(3\omega;\omega,\omega,\omega)E^3_{\omega}(Z)e^{-i\Delta kz}, \tag{4.2.4}$$

where

$$\Delta k = k_{3\omega} - 3k_{\omega} = \frac{3\omega}{c}(n_{3\omega} - n_{\omega}). \tag{4.2.5}$$

In Eq. (4.2.4), $\chi^{(3)}(3\omega;\omega,\omega,\omega)$ and $E_{\omega}(z) = E_{\omega}(0)$ are constant, we can make the equation variables separation, and then directly integrate to solve it. Setting the length of crystal is *L,* the result of integration is

$$E_{3\omega}(L) = -\frac{3\omega}{2cn_{3\omega}\Delta k}\chi^{(3)}E^3_{\omega}(0)(e^{-i\Delta kL} - 1). \tag{4.2.6}$$

To fine $|E_{3\omega}(L)|^2 = E_{3\omega}(L)\cdot E^*_{3\omega}(L)$, and use the intensity equations:

$$I_{3\omega}(L) = \frac{1}{2}\varepsilon_0 cn_{3\omega}|E_{3\omega}(L)|^2, \tag{4.2.7}$$

$$I_{\omega}(0) = \frac{1}{2}\varepsilon_0 cn_{\omega}|E_{\omega}(0)|^2, \tag{4.2.8}$$

we obtain the outputted intensity of the third harmonic light:

$$I_{3\omega}(L) = \frac{9\omega^2 L^2}{\varepsilon_0^2 c^4 n_\omega^3 n_{3\omega}} \left|\chi^{(3)}\right|^2 I_\omega^3(0) sinc^2\left(\frac{\Delta kL}{2}\right). \tag{4.2.9}$$

where the shape of function $\mathrm{sinc}^2(\Delta kL/2)$ is same as that of the frequency doubling under small signal approximation as shown in Fig. 3.5. The power conversion efficiency of the third harmonic light is

$$\eta = \frac{P_{3\omega}(L)}{P_\omega(0)} = \frac{9\omega^2 L^2}{\varepsilon_0^2 c^4 n_{3\omega} n_\omega^3} \left|\chi^{(3)}\right|^2 \left(\frac{P_\omega(0)}{S}\right)^2 sinc^2\left(\frac{\Delta kL}{2}\right). \tag{4.2.10}$$

If we define a coherent length $L_c = \pi/\Delta k$, it has $(\Delta kL_c/2) = \pi/2$. When $L \geq L_c$, the third harmonic efficiency drops down quickly.

Under phase matching condition, i.e. $\Delta k = 0$, $\mathrm{n}_{3\omega} = \mathrm{n}_\omega$, in this case there is a maximum of the third harmonic conversion efficiency:

$$\eta = \frac{9\omega^2 L^2}{\varepsilon_0^2 c^4 n_\omega^4 S^2} \left|\chi^{(3)}\right|^2 (P_\omega(0))^2 \tag{4.2.11}$$

where $P_\omega(0)$ is the foundational frequency light power, S is the cross sectional area of incident foundational frequency light beam.

After the discovery of the frequency doubling effect by using the ruby laser inputted in the quartz crystal soon, people found the third harmonic effect in some transparent nonlinear crystals. In general nonlinear crystal material, the second-order susceptibility of $\left|\chi^{(2)}\right|$ is about $10^{-3} \sim 10^{-8}$ esu, but the third order susceptibility $\left|\chi^{(3)}\right|$ is only $10^{-12} \sim 10^{-15}$ esu,so the third harmonic effect is much weaker than the second harmonic effect.

The solid crystal material is generally difficult to meet the requirement of phase matching condition, so it is hard to realize the third harmonic effect. In addition, for the common solid crystal, the ability to withstand the laser damage is very low, only the calcite crystal has stronger ability to withstand the laser damage due to it has birefringence characteristic. So the calcite crystal was used to get better third harmonic light, however its third harmonic conversion efficiency is low, the highest conversion efficiency is only 3×10^{-6}.

Researches show that the alkali metal steam has very strong nonlinear resonant enhancement effect in the visible light region, thus it has larger third-order susceptibility and stronger third harmonic conversion efficiency. In addition, the ultimate strength of the laser damage resistance of gas medium is higher than that of solid medium in several orders of magnitude, so the gas medium are often used to produce third harmonic. For example, using YAG laser at wavelength of 1.06 μm as the foundational frequency light, it is very easy to observe the third harmonic laser at wavelength of 355 nm in sodium steam. When the incident light power reaches to 300 MW, the third harmonic conversion efficiency can achieve 3.7 % [1–3].

4.2.2 Optical Four-Wave Mixing

Now we consider the frequency mixing of 4 light waves at different frequency in the third-order nonlinear medium, as shown in Fig. 4.2, where we set the incident light field amplitudes are $\boldsymbol{E}_2 = \boldsymbol{E}(\boldsymbol{r}, \omega_2), \boldsymbol{E}_1 = \boldsymbol{E}(\boldsymbol{r}, \omega_1)$ and $\boldsymbol{E}_3 = \boldsymbol{E}(\boldsymbol{r}, \omega_3)$, and the polarization light field amplitude is $\boldsymbol{E}_4 = \boldsymbol{E}(\boldsymbol{r}, \omega_4)$.

In the optical four-wave mixing process, if the phase matching condition is satisfied, the relations of energy and momentum conservation of photons are respectively:

$$\omega_4 = \omega_1 + \omega_2 + \omega_3, \tag{4.2.12}$$

$$\boldsymbol{k}_4 = \boldsymbol{k}_1 + \boldsymbol{k}_2 + \boldsymbol{k}_3 \tag{4.2.13}$$

The third-order nonlinear polarization generated by polarization field at frequency ω_4 in the medium is

$$\boldsymbol{P}^{(3)}(\boldsymbol{r}, \omega_4) = 6\varepsilon_0 \boldsymbol{\chi}^{(3)}(\omega_4;\omega_1,\omega_2,\omega_3)\boldsymbol{E}(\boldsymbol{r}, \omega_1)\boldsymbol{E}(\boldsymbol{r}, \omega_2)\boldsymbol{E}(\boldsymbol{r}, \omega_3). \tag{4.2.14}$$

Assuming 4 waves are the monochromic plane waves, and all waves propagate along z direction, the first-order coupling wave equation corresponding to the light field at frequency ω_4 is

$$\frac{\partial E(z, \omega_4)}{\partial z} = i\frac{\omega_4}{2\varepsilon_0 c n_4} P^{(3)}(z, \omega_4) e^{-i\Delta k z}. \tag{4.2.15}$$

Substituting the z-direction expression of Eq. (4.2.14) into Eq. (4.2.15), then the equation is written to

$$\frac{\partial E(z, \omega_4)}{\partial z} = i\frac{3\omega_4}{c n_4} \chi^{(3)}(\omega_4; \omega_1, \omega_2, \omega_3) E(z, \omega_1) E(z, \omega_2) E(z, \omega_3) e^{-i\Delta k z}, \tag{4.2.16}$$

where

$$\Delta k = k_4 - (k_1 + k_2 + k_3). \tag{4.2.17}$$

In the same way we can write other 3 coupling equations corresponding to the light waves at frequencies of ω_1, ω_2 and ω_3.

In the four wave mixing, for the 3 original light field frequencies:ω_1, ω_2 and ω_3, except above compound mode of $\omega_4 = \omega_1 + \omega_2 + \omega_3$, there are other compound modes, for example it can exist the following process: the difference frequency

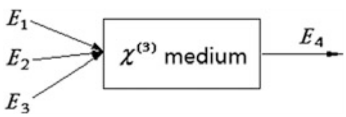

Fig. 4.2 schematic diagram of optical four-wave mixing

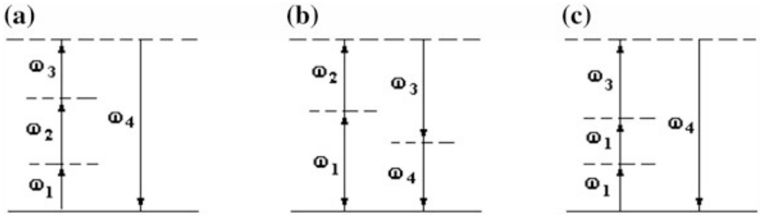

Fig. 4.3 The Energy-level diagrams for 3 kinds of four-wave mixing processes: **a** $\omega_4 = \omega_1 + \omega_2 + \omega_3$; **b** $\omega_4 = \omega_1 + \omega_2 - \omega_3$; **c** $\omega_4 = 2\omega_1 + \omega_3$

$\omega_4 = \omega_1 + \omega_2 - \omega_3$ and the sum frequency $\omega_4 = 2\omega_1 + \omega_3$ $(\omega_1 = \omega_2)$. We can explain above 3 processes by the 3 energy level diagrams as shown in Fig. 4.3 [4, 5].

4.2.3 Degenerated Four-Wave Mixing

The four-wave mixing process in the situation of 4 waves at same frequency is called the degenerated four-wave mixing [6], the relationship of 4 frequencies is

$$\omega_1 = \omega_2 = \omega_3 = \omega_4 = \omega. \tag{4.2.18}$$

The energy conservation condition for the degenerated four-wave mixing is

$$\omega = \omega - \omega + \omega. \tag{4.2.19}$$

In this process, the third-order susceptibility is denoted by $\boldsymbol{\chi}^{(3)}(\omega; \omega, -\omega, \omega)$, the degeneration factor is $D = 3$, and then the nonlinear polarization is expressed as

$$\boldsymbol{P}^{(3)}(\boldsymbol{r}, \omega) = 3\varepsilon_0 \boldsymbol{\chi}^{(3)}(\omega; \omega, -\omega, \omega)\boldsymbol{E}^2(\boldsymbol{r}, \omega)\boldsymbol{E}^*(\boldsymbol{r}, \omega). \tag{4.2.20}$$

In general case, 4 wave vectors of degenerated four-wave mixing could have different directions. However, it must obey the momentum conservation relation in the phase matching case:

$$\boldsymbol{k}_4 = \boldsymbol{k}_1 - \boldsymbol{k}_2 + \boldsymbol{k}_3. \tag{4.2.21}$$

Now we consider a special degenerated four-wave mixing case, that is existing two pairs of wave vectors, each of them has two wave vectors with reverse direction: one pair of $\boldsymbol{k}$ and $-\boldsymbol{k}$ are wave vectors of two pump lights with reverse direction; another one pair of $\boldsymbol{k}'$ and $-\boldsymbol{k}'$ are wave vectors of mutual conjugated signal lights, if the former is a probe light, the latter is called the phase conjugation light of the probe light, as shown in Fig. 4.4a. These four light waves satisfy the

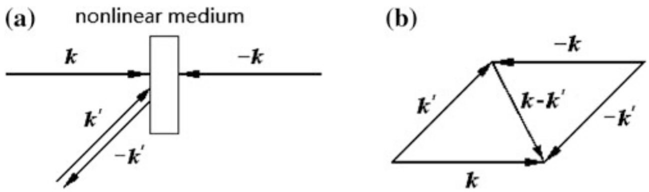

Fig. 4.4 Schematic diagram of degenerated four–wave mixing: **a** the configuration of the two pump lights, the probe light and the conjugated light; **b** the momentum-conservation vector relation

relation of momentum conservation as show in Eq. (4.2.21). If setting $\boldsymbol{k}_1 = \boldsymbol{k}, \boldsymbol{k}_2 = \boldsymbol{k}', \boldsymbol{k}_3 = -\boldsymbol{k}, \boldsymbol{k}_4 = -\boldsymbol{k}'$ Eq. (4.2.21) becomes

$$-\boldsymbol{k}' = \boldsymbol{k} - \boldsymbol{k}' + (-\boldsymbol{k}). \tag{4.2.22}$$

Then the wave vectors of these 4 light waves satisfy the momentum-conservation vector relation as shown in Fig. 4.4b. Equation (4.2.22) can be written to

$$\boldsymbol{k}' + (-\boldsymbol{k}') = \boldsymbol{k} + (-\boldsymbol{k}). \tag{4.2.23}$$

That is to say, no matter what the angle between signal light and pump light is, these two pairs of light automatically satisfy the phase matching condition.

It is worth noting that, such degenerated four-wave mixing nonlinear process is similar to typical holography process (or grating forming process), both can generate the phase conjugation light. The holography process is shown in Fig. 4.5.

In the degenerated four-wave mixing, $\boldsymbol{k}'$ can be regard as the object light, $\boldsymbol{k}$ is the reference light, two lights mutually interfere to generate and record a hologram (form a grating), as shown in Fig. 4.5a. If using the reference light $\boldsymbol{k}$ to irradiate the hologram, and blocking the object light $\boldsymbol{k}'$, from the reverse direction of the object light, i.e. the direction of $-\boldsymbol{k}'$, we can see the virtual image of object (the reflected light of grating), as shown in Fig. 4.5b; if under the irradiation of another reference light $-\boldsymbol{k}$, from the direction of $-\boldsymbol{k}'$, we can see the pseudoscopic image (the

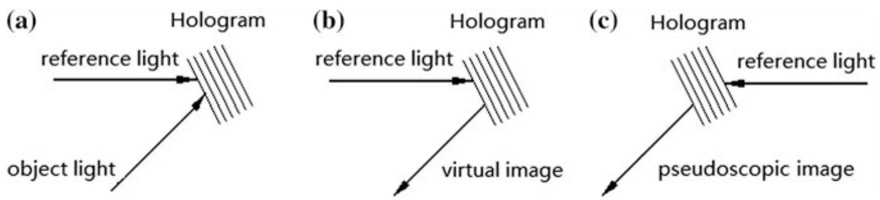

Fig. 4.5 The holography process: **a** the hologram records; **b** the virtual image reappears; **c** the pseudoscopic image reappears

scattered light of the grating), as shown in Fig. 4.5c. The pseudoscopic image is the phase conjugated light of the original object light.

Although both of the holography process and the degenerated four-wave mixing process all can generate the phase conjugated light, but the holography has the basic difference from the four-wave mixing: at first, the holography has the record and reproduce two processes. These two processes are paragraphing in the time. However the phase conjugated light and the signal light in the four-wave mixing are appeared at the same time in the same nonlinear process. Secondly, the frequency of the reference light and the frequency of the object light in the holography should be the same; otherwise it cannot obtain the stable hologram. But in the four-wave mixing, the pump light and the signal light can be different frequencies. In addition, in the holography, the polarization directions of the reference light and the object light cannot be mutual orthogonal; otherwise the hologram cannot be recorded. However, in the four-wave mixing, when the polarization directions of the pump light and the signal light are orthogonal, one can also obtain the phase conjugated light, due to the tensor characteristic of third-order susceptibility.

In next section we will give a strict definition of the optical phase conjugation.

4.3 Optical Phase Conjugation

4.3.1 Definition and Characteristic of Optical Phase Conjugation

A light wave at the frequency ω propagates along z direction, its field amplitude can generally be expressed as a complex number, which is an amplitude factor multiplied by a phase factor, and then adding its conjugation complex number, namely

$$\boldsymbol{E}(\boldsymbol{r},t) = \boldsymbol{E}(\boldsymbol{r})e^{i(kz-\omega t)} + c.c. \tag{4.3.1}$$

If this light field inputs into a system, its outputted-light-field amplitude $\mathbf{E}^*(\mathbf{r})$ is the conjugated complex number of the original inputted-light-field amplitude $\mathrm{E(r)}$, then the outputted light wave is called the phase conjugated wave of the inputted light wave, its field amplitude can be expressed as

$$\boldsymbol{E}_c(\boldsymbol{r},t) = \boldsymbol{E}^*(\boldsymbol{r})e^{i(\pm kz-\omega t)} + c.c. \tag{4.3.2}$$

When taking the negative sign at the front of the wave vector k, it is corresponding to the case of the backward phase conjugation of the original light wave, its light field amplitude is the complex phase conjugation of the original light field amplitude, but the propagation direction of phase conjugated wave is the reverse propagation direction of the original light wave, and its space distribution of the wavefront is totally the same as the space distribution of wavefront of the original light wave.

When taking the positive sign at the front of the wave vector k, it is corresponding to the case of the forward phase conjugation of the original light wave, its light field amplitude is the complex phase conjugation of the original light field amplitude, the propagation direction of phase conjugated wave is the same as the propagation direction of the original light wave, but its space distribution of the wavefront is relative to the space distribution of wavefront of the original light wave into the mirror symmetry [6, 7].

To utilize the characteristic of the backward phase conjugation can make the phase conjugated reflective mirror; and to utilize the characteristic of the forward phase conjugation can make the phase conjugated transmitting mirror,what is the difference between these phase conjugated mirrors and the common planar reflective and transmitting mirrors? Figure 4.6a, b draw up a spherical wave sent by a point light source, which is reflected by the phase conjugated reflective mirror based on the backward phase conjugation and transmitted by the phase conjugated transmitting mirror based on the forward phase conjugation, respectively. The propagation direction of former is contrary to the propagation direction of the original wave, but the wave fronts of two waves are the same. The propagation direction of latter is consistent with the propagation direction of the original wave, but the wave fronts of two waves are made a relation of mirror image symmetry. Figure 4.1c, d also draw up the action of corresponding common reflective mirror and transmitting mirror to the spherical wave sent by a point light source, it is clear that they have a big difference in compared with the phase conjugated reflective mirror and the phase conjugated transmitting mirror.

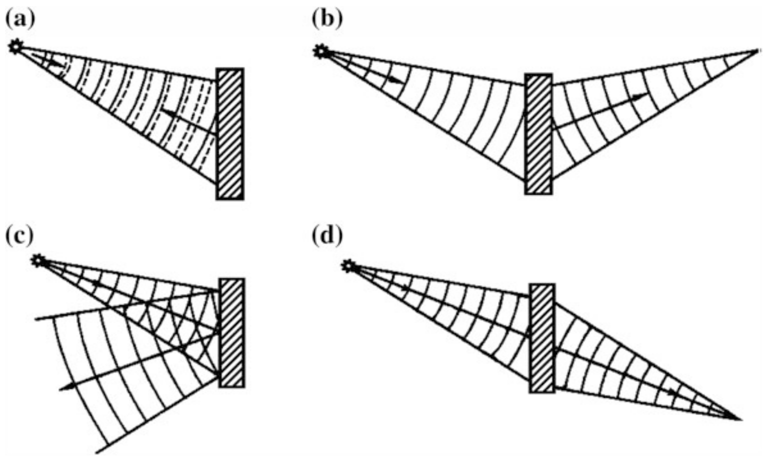

Fig. 4.6 Comparison of the action of phase conjugation mirrors and common planar mirrors to the light wave of point light source: **a** the phase conjugation reflective mirror; **b** the phase conjugation transmitting mirror; **c** the common planar reflective mirror; **d** the common planar transmitting mirror

Now we go back to discuss the backward phase conjugation [taking negative sign in front of the k in the phase conjugation light field expression (4.3.2)]. Now we write down the complete expressions of the original wave field and the phase conjugation wave field, respectively:

$$\boldsymbol{E}(\boldsymbol{r},t) = \boldsymbol{E}(\boldsymbol{r})e^{i(kz-\omega t)} + \boldsymbol{E}^*(\boldsymbol{r})e^{i(-kz+\omega t)}, \tag{4.3.3}$$

$$\boldsymbol{E}_c(\boldsymbol{r},t) = \boldsymbol{E}^*(\boldsymbol{r})e^{i(-kz-\omega t)} + \boldsymbol{E}(\boldsymbol{r})e^{i(kz+\omega t)}. \tag{4.3.4}$$

To compare above two equations, we can see that

$$\boldsymbol{E}_c(\boldsymbol{r},t) = \boldsymbol{E}(\boldsymbol{r},-t). \tag{4.3.5}$$

Therefore the backward phase conjugation light wave is called the time reversal of the original light wave, the space distribution of wavefront of the back phase conjugation light wave is the same as that of the original light wave, and only the propagation direction of the backward phase conjugation light wave is opposite to the original light wave.

4.3.2 *Optical Phase Conjugation in Four-Wave Mixing Process*

1. **Backward Phase Conjugation and Forward Phase Conjugation in Four-Wave Mixing**

Though the four-wave mixing to realize the phase conjugation, in general, there are a pair of pump lights and a pair of signal lights. The pump light $\boldsymbol{E}_1$ or $\boldsymbol{E}_2$ has stronger power in comparison with the signal light. Two signal lights are the probe light $\boldsymbol{E}_{\mathrm{p}}$ and the phase conjugation light $\boldsymbol{E}_c$. The four-wave-mixing phase conjugation satisfies following the energy conservation and momentum conservation relations:

$$\omega_c = \omega_1 + \omega_2 - \omega_p, \tag{4.3.6}$$

$$\boldsymbol{k}_c = \boldsymbol{k}_1 + \boldsymbol{k}_2 - \boldsymbol{k}_p. \tag{4.3.7}$$

Under the conditions of energy conservation and momentum conservation, the polarization expression of the phase conjugation signal light field $\boldsymbol{E}_c(\boldsymbol{r},\omega_c)$ is (D = 6):

$$\boldsymbol{P}_c^{(3)}(\boldsymbol{r},\omega_{\mathrm{c}}) = 6\varepsilon_0\boldsymbol{\chi}^{(3)}(\omega_c;\omega_1,\omega_2,-\omega_p)\boldsymbol{E}_1(\boldsymbol{r},\omega_1)\boldsymbol{E}_2(\boldsymbol{r},\omega_2)\boldsymbol{E}_P^*(\boldsymbol{r},\omega_p). \tag{4.3.8}$$

The 4 light fields joined four-wave mixing phase conjugation have two geometry configurations: the backward phase conjugation and the forward phase conjugation.

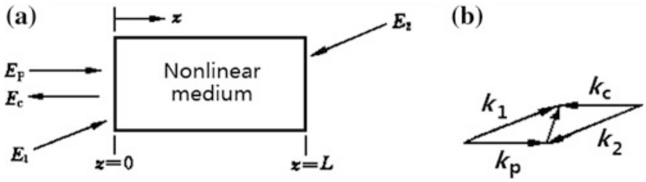

Fig. 4.7 The backward four-wave mixing phase conjugation: **a** the geometry configuration of 4 light fields; **b** the relation of wave vectors under the momentum conservation

For the backward phase conjugation, the pump lights $\boldsymbol{E}_1$ and $\boldsymbol{E}_2$ are separated in the two sides of the nonlinear medium, with reversed propagation directions (with reversed wave vectors); the probe signal light $\boldsymbol{E}_p$ and the conjugate signal light $\boldsymbol{E}_c$ are located at one side of the medium, also with reversed propagation directions (with reversed wave vectors), the geometry configuration of 4 light fields is shown in Fig. 4.7a. The wave vectors of 4 light fields satisfy the momentum conservation relation, namely $\boldsymbol{k}_c = (\boldsymbol{k}_1 - \boldsymbol{k}_p) + \boldsymbol{k}_2$, as shown in Fig. 4.7b.

For the forward phase conjugation, the pump light $\boldsymbol{E}_1$ and $\boldsymbol{E}_2$ are located at the same side of the nonlinear medium, with the same propagation direction; the probe signal light $\boldsymbol{E}_p$ and conjugated signal light $\boldsymbol{E}_c$ are located at two sides of the medium, into a mirror image each other, its geometry configuration of 4 light fields is shown in Fig. 4.8a. The wave vectors of 4 light fields satisfy the momentum conservation principle, namely $\boldsymbol{k}_c = (\boldsymbol{k}_1 + \boldsymbol{k}_2) - \boldsymbol{k}_p$, as shown in Fig. 4.8b.

2. **Backward Phase Conjugation in Degenerated Four-Wave Mixing**

Now we study the backward phase conjugation based on the degenerated four-wave mixing. In the case of the degenerated four-wave mixing, the probe light $\boldsymbol{E}_p(\boldsymbol{r})$, the conjugation light $\boldsymbol{E}_c(\boldsymbol{r})$, the pump lights $\boldsymbol{E}_1(\boldsymbol{r})$ and $\boldsymbol{E}_2(\boldsymbol{r})$ are at the same frequency:

$$\omega_p = \omega_c = \omega_1 = \omega_2 = \omega. \tag{4.3.9}$$

Thus the degenerated four-wave mixing based backward phase conjugation has the relations of wave vectors: $\boldsymbol{k}_2 = -\boldsymbol{k}_1$ and $\boldsymbol{k}_c = -\boldsymbol{k}_p$. From the momentum

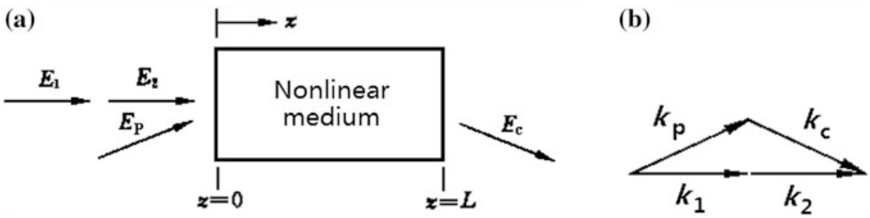

Fig. 4.8 The forward four-wave mixing phase conjugation: **a** the geometry configuration of 4 light fields; **b** the relation of wave vectors under the momentum conservation

conservation formula (4.3.7) we can see that the phase conjugation condition of this nonlinear process is automatically satisfied.

Now we start discuss the propagation characteristic of the conjugation light in the nonlinear medium. We assume that the nonlinear medium is isotropic, non-absorption, and far from the resonance; the process satisfies the phase matching condition, $\Delta \boldsymbol{k} = 0$; This is in small signal approximation case, the pump light is much stronger than the signal light, so $\boldsymbol{E}_1(\boldsymbol{r})$ and $\boldsymbol{E}_2(\boldsymbol{r})$ have no decay in z direction, we can only consider the coupling equations for two signal lights. And assuming $\boldsymbol{E}_p(\boldsymbol{r})$ propagates along the z direction; $\boldsymbol{E}_c(\boldsymbol{r})$ propagates along the –z direction, using polarization Eq. (4.3.8), we obtain following two nonlinear coupling wave equations:

$$\frac{dE_p(z)}{dz} = i\frac{3\omega}{nc}\chi^{(3)}E_1E_2E_c^*(z), \tag{4.3.10}$$

$$\frac{dE_c(z)}{dz} = -i\frac{3\omega}{nc}\chi^{(3)}E_1E_2E_p^*(z), \tag{4.3.11}$$

where $\chi^{(3)} = \chi^{(3)}(\omega_c; \omega_1, \omega_2, -\omega_p) = \chi^{(3)}(\omega_p; \omega_1, \omega_2, -\omega_c)$ is third-order nonlinear susceptibility of medium, in the far from resonance case it is a real number.

To define the gain coefficient:

$$g = \frac{3\omega}{nc}\chi^{(3)}E_1E_2. \tag{4.3.12}$$

g is a real number. Equations (4.3.10) and (4.3.11) are simplified as

$$\frac{dE_p(z)}{dz} = igE_c^*(z), \tag{4.3.13}$$

$$\frac{dE_c(z)}{dz} = -igE_p^*(z). \tag{4.3.14}$$

Setting the length of the medium is L, and considering the following boundary condition: the light field E_p is inputted from the surface at $z = 0$ where $E_p(0) \neq 0$, and outputted from the surface at $z = L$ where $E_p(L) \neq 0$; the light field E_c is reflected by the surface at $z = L$ where $E_c(L) = 0$, then outputted from the surface at $z = 0$ where $E_c(0) \neq 0$. The solutions of Eqs. (4.2.13) and (4.2.14) are

$$E_p(z) = \frac{\cos[g(z-L)]}{\cos(gL)}E_p(0), \tag{4.3.15}$$

$$E_c(z) = -i\frac{\sin[g(z-L)]}{\cos(gL)}E_p^*(0). \tag{4.3.16}$$

So the light field amplitudes at the two end surfaces of medium are respectively:

$$E_p(L) = \frac{1}{\cos(gL)} E_P(0), \tag{4.3.17}$$

$$E_c(0) = i\,\tan(gL) E_p^*(0), \tag{4.3.18}$$

where $E_p(L)$ is the outputted probe light amplitude from the surface at $z = L$; $E_c(0)$ is the outputted conjugation light amplitude from the surface at $z = 0$. They all are produced from $E_p(0)$, the former is the transmitted light of $E_p(0)$; the latter is its reflected light, both of them are amplified in the nonlinear process. Form Eq. (4.3.18) we can see, at $z = 0$, the conjugated light amplitude $E_c(0)$ is proportional to the conjugate complex number of probe light amplitude $E_p^*(0)$, but their propagation directions are opposite.

We define the power transmitted coefficient and the power reflected coefficient of phase conjugation, respectively:

$$T = \frac{|E_p(L)|^2}{|E_p(0)|^2} = \sec^2(gL), \tag{4.3.19}$$

$$R = \frac{|E_c(0)|^2}{|E_p(0)|^2} = \tan^2(gL). \tag{4.3.20}$$

From Eqs. (4.2.17)–(4.2.20) we can obtain the following conclusions:

(1) The process produced signal light $E_c(0)$ is the phase conjugation wave of original probe light $E_p(0)$. The reflectivity R increases with the increase of gL. Form Eq. (4.2.12) we can see, $\chi^{(3)}$ of the medium and the pump light amplitudes E_1 and E_2 are larger, g is larger, in order to produce high phase conjugation reflectivity, we need select the material with large $\chi^{(3)}$ and the high power pump light.

(2) When $\frac{1}{4}\pi < gL < \frac{3}{4}\pi$, due to $\tan(gL) > 1$, $R > 1$, this means that through the four-wave mixing process, the phase conjugation light is amplified. At same time, the transmittance of the probe light $T \geq 1$, so the probe light is also amplified.

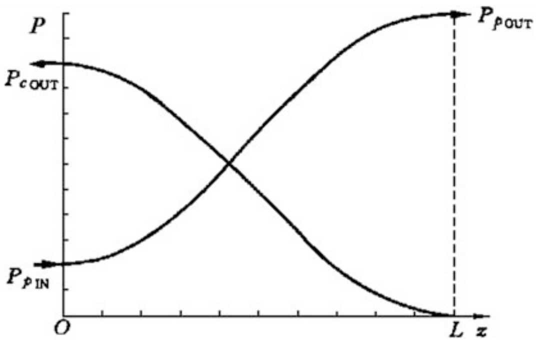

Fig. 4.9 Amplification action of the probe light and the phase conjugation light in degenerated four-wave mixing backward phase conjugation process

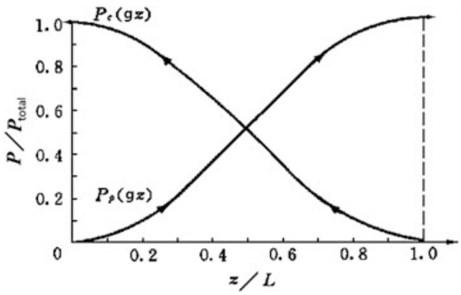

Fig. 4.10 Power distributions of the probe light and the phase conjugation light in the medium when the oscillation condition $gL = \pi/2$ is satisfied

Figure 4.9 gives the curves of that the probe light power and the phase conjugation light power vary with increase of z when R and T both are greater than 1.

(3) When $gL = \pi/2$, $R \to \infty$, $\mathrm{T} \to \infty$, the non-cavity self-oscillation in the medium is produced. In this case the signal light don't need input into the medium, the energy comes from the pump light, there are outputted signal light and phase conjugation light, it just likes a parameter oscillator. In this case the light power distribution is shown in Fig. 4.10.

Early stage the amplification and oscillation phenomena of degenerated four-wave mixing backward phase conjugation wave had observed in the CS_2 liquid and sodium steam. The experimental facility is shown in Fig. 4.11 [8].

3. **Other Phase Conjugation Situation**

Above study only consider the small signal situation. In the high conversion efficiency case, the variation of pump light with change of the distance has to consider, in this case we need simultaneous solve the 4 coupling equations. In above study we also ignored the pump light induced refractive-index change and absorption in the medium; moreover, the study is only based on the non-resonance condition. The strict study should consider these factors.

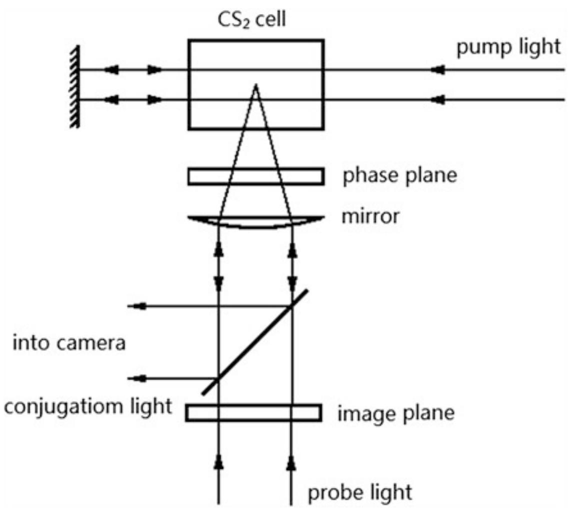

Fig. 4.11 Experimental facility for study degenerated four-wave mixing backward phase conjugation process

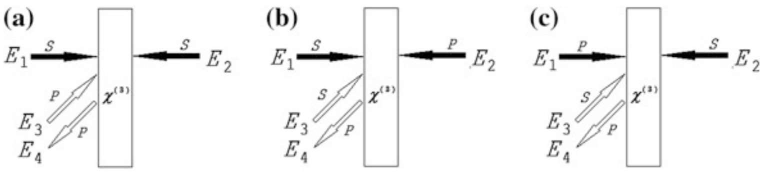

Fig. 4.12 3 Different light polarization combinations for degenerated four-wave mixing backward phase conjugation

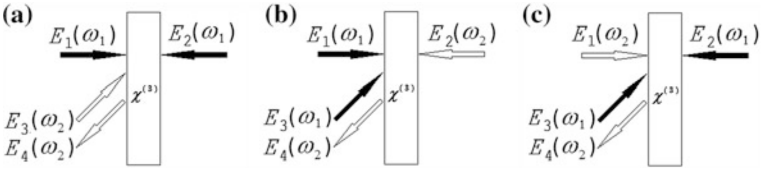

Fig. 4.13 Two group frequency configurations for the part degenerated four-wave mixing phase conjugation

It's worth noting that, above deduction of the formulas for the four-wave mixing phase conjugation implies the assumption that the 4 light waves participated in the process all have the same frequency and polarization direction. Actually, in the degenerated four-wave mixing phase conjugation, 4 light beams at the same frequency can have different polarization directions (the vertical polarization s or the parallel polarization p). Figure 4.12 shows the 3 kinds of combinations with the orthogonal polarization directions: (a) is the case that two pump lights have a same polarization direction, and two signal lights have another same polarization direction, these two polarization directions are orthogonal. (b) and (c) are the cases that each of pump light-signal light group has a same polarization direction, the two groups have the orthogonal polarization directions. All above three cases can realize the phase conjugation.

Not only complete degenerated four-wave mixing can realize the phase conjugation, but also the part degenerated four-wave mixing composited by two pair of lights at different frequency can realize the phase conjugation. Figure 4.13 shows three phase conjugation configurations at two different frequencies.

There are many methods to realize phase conjugation, except the four-wave mixing, the method of three-wave mixing, stimulated scattering, and backward laser emission etc. can also be used to generate the phase conjugation wave [9].

4.3.3 *Application of Optical Phase Conjugation*

1. **Wavefront Distortion Compensation**

The conjugate reflected mirror based on the backward phase conjugation principle can automatically compensate the wavefront distortion, which is formed when the light pass through the irregular disturbance medium [10]. Figure 4.14 gives a comparison of different actions to the wavefront distortion in the medium by a phase conjugation mirror and a common reflected mirror. After a parallel plane wave passing though the distortion medium, it is then reflected by a common reflected mirror or a phase conjugation mirror, respectively, the common reflected mirror plays a roll of increasing the wavefront distortion; but the phase conjugation mirror has an action that compensates or offsets the wavefront distortion. In the figure, the real line is the wavefront of incident wave; the dotted line is the wavefront of reflected wave.

2. **Measurement of Susceptibility**

Now we introduce a method for measuring third-order susceptibility by a degenerated four-wave mixing backward phase conjugation experiment [11]. If from the experiment to measure the data of light-wave amplitudes: E_1, E_2, E_c and E_p, and know the laser wavelength λ, the length of nonlinear medium L, and the refractive index of medium n, using the following reflectivity formula (4.3.20) and gain coefficient formula (4.3.12):

$$R = \frac{|E_c(0)|^2}{|E_p(0)|^2} = \tan^2(gL),$$

$$g = \frac{3\omega}{nc}\chi^{(3)}E_1E_2,$$

we can calculate the value of the third-order susceptibility $\chi^{(3)}$.

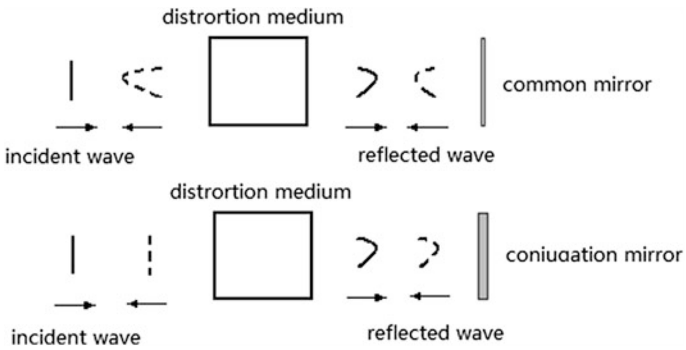

Fig. 4.14 Comparison of the distortion-compensation action of phase conjugation mirror and the distortion-enhancement action of common reflected mirror

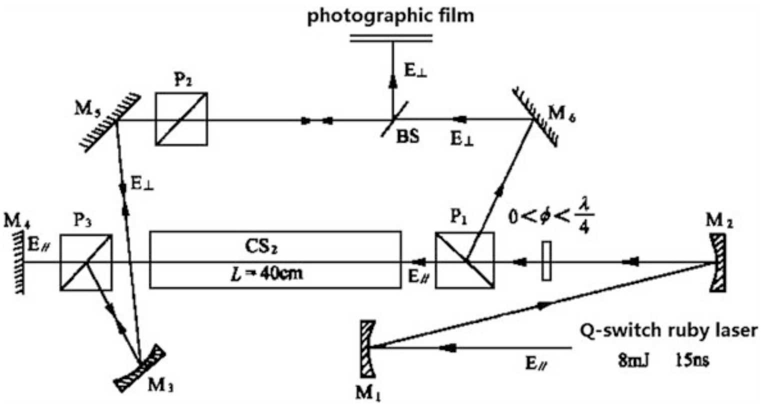

Fig. 4.15 Experimental setup for measuring $\chi^{(3)}$ based on degenerated four-wave mixing phase conjugation using a polarization separation technique

Here we introduce a degenerated four-wave mixing backward phase conjugation experiment based on the light polarization separation technique with a high measurement accuracy, in the experiment the 4 light beams collinearly propagate in the medium CS_2, because the polarization directions of signal lights and pump lights are orthogonal, we can separate out the conjugated signal lights from the pump lights. Figure 4.15 gives an experimental setup in detail.

A parallel polarization light send by a Q-switch pulsed ruby laser, it passes through the reflected mirrors M_1 and M_2, a wave pate, two Glan prisms (polarization beam splitter) P_1 and P_3, and the reflected mirror M_4, to form two pump lights E_1 and E_2 with the reversed propagation direction and the paralleled polarization direction. The light reflected from P_1 with the vertical polarization passes through M_6, P_2, M_5 and M_3, then reflected into the medium CS_2 by P_3, to form the incident signal light E_c with the vertical polarization. The conjugated signal light E_P with the reverse propagation direction relative to the signal light and the vertical polarization passes through P_3, M_3, M_5, P_2 and the beam splitter BS is finally measured by a photographic film.

Review Questions of Chapter 4

1. Which are passive third-order nonlinear optics effects? Write down their nonlinear susceptibility formulae.
2. Please derive the third harmonic effect power conversion efficiency formula in small signal approximation case.
3. What is four-wave mixing effect? Write down its polarization expression and slowly-varying amplitude-approximation frequency-domain wave equation
4. What is degenerated four-wave mixing? What kind phase matching condition it satisfies? To compare with the holography, what identical or different characteristics do they have?

5. How to define the phase conjugation? What characteristic does it has? Which methods can be used for realizing the phase conjugation?
6. Give out the polarization expression of four-wave-mixing phase conjugation. What is difference between back phase conjugation and forward phase conjugation? What kind energy and momentum conservation relations do they have to meet?
7. Establish the signal light and the conjugated light coupled wave equations for the back phase conjugation process based on the degenerated four-wave mixing. Discuss the transmission characteristic of phase conjugation wave in the medium.
8. What are applications of optical phase conjugation? Draw a picture to explain the action of phase conjugation in compensation of the wavefront distortion. How to use degenerated four-wave mixing phase conjugation method to measure the third-order susceptibility of nonlinear medium $\chi^{(3)}$?

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