Chapter 3
Optical Three-Wave Coupling Processes

This chapter uses a reformed first-order frequency-domain wave equation for the isotopic medium to approximately describe the second-order nonlinear optics effects in the anisotropic medium. At first, the three-wave coupling equations are deduced, then based on these equations, several typical second-order nonlinear optics effects are studied: optical frequency doubling, sum frequency, difference frequency, and optical parameter amplification and parameter oscillation. The power conversion efficiency formulas for these effects are given. Finally, the basic concepts of phase matching are introduced based on the frequency doubling effect.

3.1 Three-Wave Coupled Equations

3.1.1 Review of Second-Order Nonlinear Optics Effects in Isotopic Medium

Firstly we discuss the second-order nonlinear optics effect in general, it contains what specific effects, and we will give the polarizations of these effects in the isotopic material.

Assuming that the incident light electrical fields consisted by two monochromatic light fields at the different frequencies and with same propagation direction, the total electrical field strength can be expressed as

$$\boldsymbol{E}(t) = \sum_{n=1,.2} \boldsymbol{E}_n e^{-i\omega_n t} + c.c. = \boldsymbol{E}_1 e^{-i\omega_1 t} + \boldsymbol{E}_2 e^{-i\omega_2 t} + c.c. \tag{3.1.1}$$

In the isotopic medium without center symmetry, the second-order nonlinear polarization is

C. Li, *Nonlinear Optics*, DOI 10.1007/978-981-10-1488-8_3

$$\boldsymbol{P}^{(2)}(t) = \varepsilon_0 \chi^{(2)} \boldsymbol{E}^2(t). \tag{3.1.2}$$

Substituting Eq. (3.1.1) into Eq. (3.1.2), after combination of the items with same frequency component, we obtain:

$$\begin{aligned}\boldsymbol{P}^{(2)}(t) = \varepsilon_0 \chi^{(2)} [(&\boldsymbol{E}_1^2 e^{-i2\omega_1 t} + \boldsymbol{E}_2^2 e^{-i2\omega_2 t} + 2\boldsymbol{E}_1\boldsymbol{E}_2 e^{-i(\omega_1+\omega_2)t} + 2\boldsymbol{E}_1\boldsymbol{E}_2^* e^{-i(\omega_1-\omega_2)t}) \\ &+ 2(\boldsymbol{E}_1\boldsymbol{E}_1^* + \boldsymbol{E}_2\boldsymbol{E}_2^*)] + c.c.\end{aligned} \tag{3.1.3}$$

The Eq. (3.1.3) can be summarized by a simple formula, that is

$$\boldsymbol{P}^{(2)}(t) = \sum_i \boldsymbol{P}^{(2)}(\omega_i) e^{-i\omega_i t} + c.c, \tag{3.1.4}$$

where i takes the positive integer. The polarization $\boldsymbol{P}^{(2)}(\omega_i)$ corresponds to the different second-order nonlinear optics effect with different susceptibility $\chi^{(2)}(\omega_i)$, which is

$$\boldsymbol{P}^{(2)}(\omega_i) = D\varepsilon_0 \chi^{(2)}(\omega_i) \boldsymbol{E}(\omega_1)\boldsymbol{E}(\omega_2), \tag{3.1.5}$$

where ω_i is the frequency of polarization field composed by two original monochromic fields at frequencies of ω_1 and ω_2 in different modes. Form Eq. (3.1.3) we can see that ω_i has five modes: $2\omega_1$, $2\omega_2$, $\omega_1 + \omega_2$, $\omega_1 - \omega_2$ and 0. For second-order nonlinearity, $n = 2$, the degeneration factor is $D = n!/m! = 2/m!$. When $m = 1$, $D = 2$; when $m = 2$, $D = 1$. Therefore, corresponding to the different ω_i, the second-order nonlinear optics effects and corresponding polarizations are respectively:

Optical frequency doubling

$$\boldsymbol{P}(2\omega_1) = \varepsilon_0 \chi^{(2)}(2\omega_1) \boldsymbol{E}_1^2, \tag{3.1.6}$$

Optical frequency doubling

$$\boldsymbol{P}(2\omega_2) = \varepsilon_0 \chi^{(2)}(2\omega_2) \boldsymbol{E}_2^2, \tag{3.1.7}$$

Optical sum frequency

$$\boldsymbol{P}(\omega_1 + \omega_2) = 2\varepsilon_0 \chi^{(2)}(\omega_1 + \omega_2) \boldsymbol{E}_1\boldsymbol{E}_2, \tag{3.1.8}$$

Optical difference frequency

$$\boldsymbol{P}(\omega_1 - \omega_2) = 2\varepsilon_0 \chi^{(2)}(\omega_1 - \omega_2) \boldsymbol{E}_1\boldsymbol{E}_2^*, \tag{3.1.9}$$

Optical rectification

$$\boldsymbol{P}(0) = 2\varepsilon_0\chi^{(2)}(0)(\boldsymbol{E}_1\boldsymbol{E}_1^* + \boldsymbol{E}_2\boldsymbol{E}_2^*). \quad (3.1.10)$$

3.1.2 Approximate Description of Second-Order Nonlinear Optics Effect in Anisotropic Medium

In general, the medium with the second-order nonlinear optical effect is not isotopic, it is anisotropic, such as the crystal medium. However, for describing the second-order nonlinear effect in the crystal medium should using tensor calculation method, it is complicated, and needs take up a larger space. This chapter we present a method that using the slowly-varying-amplitude approximated frequency-domain first-order wave equation for the isotropic medium to approximately describe the second-order nonlinear optical effects in the anisotropic medium. For simplicity, we suppose that the medium is far from the resonance area, and the absorption loss can be neglected.

The characteristic of light propagation in the anisotropic medium is: the propagation direction of light wave ($\boldsymbol{k}$) is different with the direction of energy flow ($\boldsymbol{I} = \boldsymbol{E} \times \boldsymbol{H}$), there is an included angle α between both. Because the electric induction strength $\boldsymbol{D}$ in the medium is perpendicular to the propagation direction of the light; and the electrical field strength $\boldsymbol{E}$ is perpendicular the direction of energy flow, so there is an included angle α between $\boldsymbol{D}$ and $\boldsymbol{E}$. Actually α is small, $\alpha < 3°$ for the most of crystals.

Considering a monochromic plane wave propagates along z direction in an anisotropic medium, Suppose its wave vector $\boldsymbol{k}$ is along z direction; and there is an included angle α between $\boldsymbol{k}$ and the energy flow $\boldsymbol{I} = \boldsymbol{E} \times \boldsymbol{H}$. The electric induction strength $\boldsymbol{D}$ is along x direction; the magnetic field strength $\boldsymbol{H}$ is along y direction, which is perpendicular to the plane consisted by $\boldsymbol{D}$, $\boldsymbol{E}$ and $\boldsymbol{k}$, as shown in Fig. 3.1 [1].

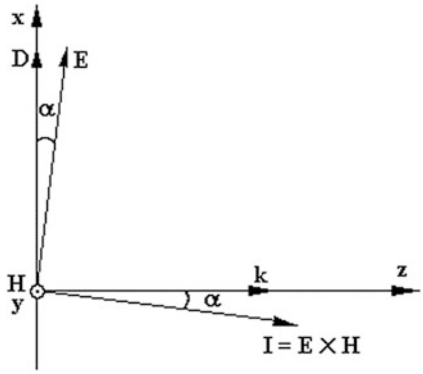


Fig. 3.1 Relationship among electromagnetic wave vectors E, D, H, k, and $\boldsymbol{I} = \boldsymbol{E} \times \boldsymbol{H}$, when the monochromic plane light wave propagates in an anisotropic medium

Suppose the frequency of above monochromic plane wave is ω, the light electrical field strength is expressed as a product of amplitude and phase:

$$\boldsymbol{E}(z,\omega) = \boldsymbol{E}(z)e^{i(kz-\omega t)} = \hat{\boldsymbol{e}}E(z)e^{i(kz-\omega t)}, \tag{3.1.6}$$

where $\hat{\boldsymbol{e}}$ is the unit vector along the electrical field direction. The polarization corresponding to the light electrical field strength is

$$\boldsymbol{P}_{NL}(z,\omega) = \boldsymbol{P}_{NL}(z)e^{i(k'z-\omega t)}. \tag{3.1.7}$$

The each of $\boldsymbol{E}(z,\omega)$ and $\boldsymbol{P}_{NL}(z,\omega)$ can be written to the vector sum of two orthogonal components, i.e., the horizontal component perpendicular to $\boldsymbol{k}$ (noted by $\boldsymbol{T}$) and the longitudinal component parallel to $\boldsymbol{k}$ (noted by S):

$$\boldsymbol{E}(z,\omega) = \boldsymbol{E}_T(z,\omega) + \boldsymbol{E}_S(z,\omega), \tag{3.1.8}$$

$$\boldsymbol{P}_{NL}(z,\omega) = \boldsymbol{P}^T_{NL}(z,\omega) + \boldsymbol{P}^S_{NL}(z,\omega). \tag{3.1.9}$$

The horizontal component of field amplitude abides by following frequency-domain wave equation for isotropic medium in the condition of slowly-varying-amplitude approximation:

$$\frac{\partial \boldsymbol{E}_T(z)}{\partial z} = \frac{i\omega}{2\varepsilon_0 cn}\boldsymbol{P}^T_{NL}(z)e^{i\Delta kz}. \tag{3.1.10}$$

To make the dot product of the unit vector $\hat{\boldsymbol{e}}$ in the two sides of Eq. (3.1.10) respectively, and using

$$\hat{\boldsymbol{e}} \cdot \boldsymbol{E}_T = E_T \cos\alpha = E\cos^2\alpha$$

and

$$\boldsymbol{P}^T_{NL} \approx \boldsymbol{P}_{NL},$$

then we obtain

$$\frac{\partial E(z)}{\partial z} = \frac{i\omega}{2\varepsilon_0 cn\cos^2\alpha}\hat{\boldsymbol{e}} \cdot \boldsymbol{P}_{NL}(z)e^{i\Delta kz}. \tag{3.1.11}$$

If take $\cos^2\alpha \approx 1$ approximately, Eq. (3.1.11) becomes

$$\frac{\partial E(z)}{\partial z} = \frac{i\omega}{2\varepsilon_0 cn}\hat{\boldsymbol{e}} \cdot \boldsymbol{P}_{NL}(z)e^{\mathrm{i}\Delta kz}, \tag{3.1.12}$$

where $\Delta k = k' - k$, k is the wave vector of original light field, k' is the wave vector of polarization field. This is the slowly-varying-amplitude-approximation frequency-domain wave equation for propagation of the light field amplitude of the monochromic plane wave in the anisotropic medium. The difference between this equation and the slowly-varying-amplitude- approximation frequency-domain wave equation in isotropic medium is replaced $P_{NL}(z)$ by $\hat{\boldsymbol{e}} \cdot \boldsymbol{P}_{NL}(z)$.

3.1.3 Three-Wave Coupled Equations in Anisotropic Medium

In general case, two light wave fields $\boldsymbol{E}(\omega_1, \boldsymbol{k}_1)$ and $\boldsymbol{E}(\omega_2, \boldsymbol{k}_2)$ with different incident directions interact with a nonlinear crystal medium, to induce a new light wave field $\boldsymbol{E}(\omega_3, \boldsymbol{k}_3)$. The three-wave coupling process is shown in Fig. 3.2.

This three-waves coupling process can use the photon concept to describe. The three photons at different frequency ω_1, ω_2 and ω_3 should meet the following energy conservation law:

$$\hbar\omega_3 = \hbar\omega_1 + \hbar\omega_2. \tag{3.1.13}$$

If we want to realize the optimum coupling of three photons, the three photons also need satisfy the momentum conservation law as follows

$$\hbar\boldsymbol{k}_3 = \hbar\boldsymbol{k}_1 + \hbar\boldsymbol{k}_2. \tag{3.1.14}$$

To describe this process by using optical wave concept, the frequencies of three waves should satisfy the relationship: $\omega_3 = \omega_1 + \omega_2$. Here we just talk about sum frequency process. Actually there are difference processes, which satisfy the relationship $\omega_1 = \omega_3 - \omega_2$ and $\omega_2 = \omega_3 - \omega_1$.

Assuming that three monochromic plane waves at frequencies ω_1, ω_2, ω_3 propagate in the anisotropic medium, they all along z direction, to generate a sum frequency or two difference frequency nonlinear effects, their second-order nonlinear polarizations can be expressed as respectively:

$$\boldsymbol{P}^{(2)}(z, \omega_1) = D\varepsilon_0\boldsymbol{\chi}^{(2)}(\omega_1; -\omega_2, \omega_3) : \boldsymbol{E}^*(z, \omega_2)\boldsymbol{E}(z, \omega_3), \tag{3.1.15}$$

$$\boldsymbol{P}^{(2)}(z, \omega_2) = D\varepsilon_0\boldsymbol{\chi}^{(2)}(\omega_2; \omega_3, -\omega_1) : \boldsymbol{E}(z, \omega_3)\boldsymbol{E}^*(z, \omega_1), \tag{3.1.16}$$

$$\boldsymbol{P}^{(2)}(z, \omega_3) = D\varepsilon_0\boldsymbol{\chi}^{(2)}(\omega_3; \omega_1, \omega_2) : \boldsymbol{E}(z, \omega_1)\boldsymbol{E}(z, \omega_2), \tag{3.1.17}$$

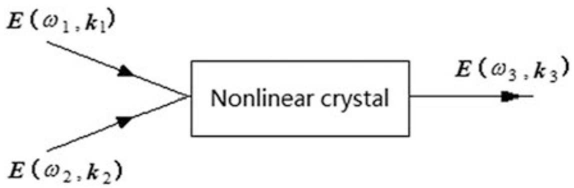


Fig. 3.2 Schematic diagram of the three-wave coupling process

where D is the degeneration factor, if two original lights are at different frequencies, $D = 2$; if two original lights are at same frequency, $D = 1$.

If denoting the three light fields respectively to be $\boldsymbol{E}(z, \omega_1) = \hat{\boldsymbol{e}}_1 E_1$, $E(\mathrm{z}, \omega_2) = \hat{\boldsymbol{e}}_2 E_2$, and $E(\mathrm{z}, \omega_3) = \hat{\boldsymbol{e}}_3 E_3$, then we have

$$\boldsymbol{P}_1^{(2)}(z) = D\varepsilon_0 \boldsymbol{\chi}^{(2)}(\omega_1; -\omega_2, \omega_3) : \hat{\boldsymbol{e}}_2 \hat{\boldsymbol{e}}_3 E_2^* E_3, \tag{3.1.18}$$

$$\boldsymbol{P}_2^{(2)}(z) = D\varepsilon_0 \boldsymbol{\chi}^{(2)}(\omega_2; \omega_3, -\omega_1) : \hat{\boldsymbol{e}}_3 \hat{\boldsymbol{e}}_1 E_3 E_1^*, \tag{3.1.19}$$

$$\boldsymbol{P}_3^{(2)}(z) = D\varepsilon_0 \boldsymbol{\chi}^{(2)}(\omega_3; \omega_1, \omega_2) : \hat{\boldsymbol{e}}_1 \hat{\boldsymbol{e}}_2 E_1 E_2. \tag{3.1.20}$$

In the frequency-domain wave Eq. (3.1.12), if setting $\boldsymbol{P}_{NL} = \boldsymbol{P}^{(2)}$, then substituting Eqs. (3.1.18)–(3.1.20) into Eq. (3.1.12), then we obtain following equations:

$$\frac{\partial E_1(z)}{\partial z} = \frac{i\omega_1}{2cn_1} D\hat{\boldsymbol{e}}_1 \cdot \boldsymbol{\chi}^{(2)}(\omega_1; -\omega_2, \omega_3) : \hat{\boldsymbol{e}}_2 \hat{\boldsymbol{e}}_3 E_2^* E_3 e^{\mathrm{i}\Delta kz}, \tag{3.1.21}$$

$$\frac{\partial E_2(z)}{\partial z} = \frac{i\omega_2}{2cn_2} D\hat{\boldsymbol{e}}_2 \cdot \boldsymbol{\chi}^{(2)}(\omega_2; \omega_3, -\omega_1) : \hat{\boldsymbol{e}}_3 \hat{\boldsymbol{e}}_1 E_3 E_1^* e^{\mathrm{i}\Delta kz}, \tag{3.1.22}$$

$$\frac{\partial E_3(z)}{\partial z} = \frac{i\omega_3}{2cn_3} D\hat{\boldsymbol{e}}_3 \cdot \boldsymbol{\chi}^{(2)}(\omega_3; \omega_1, \omega_2) : \hat{\boldsymbol{e}}_1 \hat{\boldsymbol{e}}_2 E_1 E_2 e^{-\mathrm{i}\Delta kz}. \tag{3.1.23}$$

According to the frequency substitution symmetry of susceptibility, three nonlinear susceptibilities are equal, namely

$$\begin{aligned} \hat{\boldsymbol{e}}_1 \cdot \boldsymbol{\chi}^{(2)}(\omega_1; -\omega_2, \omega_3) : \hat{\boldsymbol{e}}_2 \hat{\boldsymbol{e}}_3 &= \hat{\boldsymbol{e}}_2 \cdot \boldsymbol{\chi}^{(2)}(\omega_2; \omega_3, -\omega_1) : \hat{\boldsymbol{e}}_3 \hat{\boldsymbol{e}}_1 \\ &= \hat{\boldsymbol{e}}_3 \cdot \boldsymbol{\chi}^{(2)}(\omega_3; \omega_1, \omega_2) : \hat{\boldsymbol{e}}_1 \hat{\boldsymbol{e}}_2 = \boldsymbol{\chi}_{eff}^{(2)}, \end{aligned} \tag{3.1.24}$$

here $\chi_{eff}^{(2)}$ is a real number, it is called the efficient nonlinear susceptibility, which is used for measurement of the coupling strength among three waves. To omit the corner mark *eff*, i.e., $\chi_{eff}^{(2)} = \chi^{(2)}$, the three components of susceptibility can be written to the following scalar forms:

$$\chi^{(2)}(\omega_1; -\omega_2, \omega_3) = \hat{\boldsymbol{e}}_1 \cdot \boldsymbol{\chi}^{(2)}(\omega_1; -\omega_2, \omega_3) : \hat{\boldsymbol{e}}_2 \hat{\boldsymbol{e}}_3, \tag{3.1.25}$$

$$\chi^{(2)}(\omega_2; \omega_3, -\omega_1) = \hat{\boldsymbol{e}}_2 \cdot \boldsymbol{\chi}^{(2)}(\omega_2; \omega_3, -\omega_1) : \hat{\boldsymbol{e}}_3 \hat{\boldsymbol{e}}_1, \tag{3.1.26}$$

$$\chi^{(2)}(\omega; \omega_1, \omega_2) = \hat{\boldsymbol{e}}_3 \cdot \boldsymbol{\chi}^{(2)}(\omega_3; \omega_1, \omega_2) : \hat{\boldsymbol{e}}_1 \hat{\boldsymbol{e}}_2. \tag{3.1.27}$$

Therefore, the wave Eqs. (3.1.21)–(3.1.23) can be written to

$$\frac{\partial E_1(z)}{\partial z} = i\frac{D\omega_1}{2cn_1}\chi^{(2)}(\omega_1; -\omega_2, \omega_3)E_2^*(z)E_3(z)e^{i\Delta kz}, \tag{3.1.28}$$

$$\frac{\partial E_2(z)}{\partial z} = i\frac{D\omega_2}{2cn_2}\chi^{(2)}(\omega_2; \omega_3, -\omega_1)E_3(z)E_1^*(z)e^{i\Delta kz}, \tag{3.1.29}$$

$$\frac{\partial E_3(z)}{\partial z} = i\frac{D\omega_3}{2cn_3}\chi^{(2)}(\omega_3; \omega_1, \omega_2)E_1(z)E_2(z)e^{-i\Delta kz}, \tag{3.1.30}$$

where Δk is phase mismatch factor, which can be expressed as

$$\Delta k = k_1 + k_2 - k_3. \tag{3.1.31}$$

The meaning of Δ k for different processes are different: for difference frequency process described by Eq. (3.1.28) is $\Delta k = k_1 - (k_3 - k_2)$; for difference frequency process described by Eq. (3.1.29) is $\Delta k = k_2 - (k_3 - k_1)$; for sum frequency process described by Eq. (3.1.30) is $-\Delta k = k_3 - (k_1 + k_2)$. If the three wave is phase matched, then $\Delta k = 0$, it is equivalent to that the three photons satisfy the momentum conservation law.

3.2 Optical Second-Harmonic Generation

Optical second harmonic generation, i.e., optical frequency doubling, is a special case of three wave mixing processes. That is one of nonlinear optical phenomena, which found at the earliest. The experimental setup for studying the frequency doubling in 1961 by Franken et al. is shown in Fig. 3.3 [2]. The ruby laser (at the wavelength λ_1 = 694.3 nm) passed through a quartz crystal to produce the frequency doubling light (at the wavelength λ_2 = 347.15 nm). Two light beams are separated by a prism.

Now the application of optical frequency doubling has been mature. For example, it is used to transform from infrared laser at the wavelength of 1.06 μm generated by a Nd:YAG laser to the green laser at the wavelength of 532 nm.

Considering a monochromic plane light wave at frequency ω passes through a nonlinear crystal with length of L to produce a frequency doubling light at frequency 2ω, as shown in Fig. 3.4.

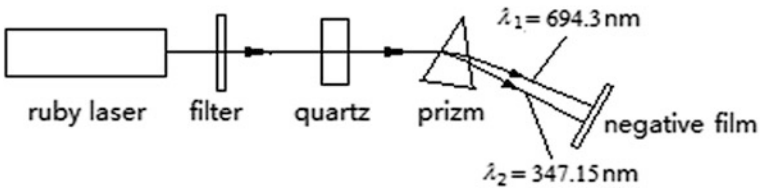


Fig. 3.3 Experimental facility of frequency doubling by Franken et al.

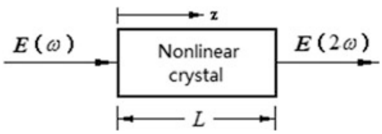


Fig. 3.4 Schematic diagram of the frequency doubling process

Suppose the crystal does no absorption for these two lights, now we are going to calculate the intensity of the frequency doubling light transmitted from crystal and the efficiency of frequency doubling conversion, i.e., the ratio between the power of frequency doubling light and the power of fundamental frequency light.

We can use three wave coupled Eqs. (3.1.28)–(3.1.30) to deal with second harmonic generation problems. We suppose the fundamental frequency is $\omega_1 = \omega_2 = \omega$, and the second harmonic frequency is $\omega_3 = 2\omega$. When establishing the wave equation of fundamental frequency light filed, we should set the degeneration factor to be $D = 2$; and when establishing the wave equation of frequency doubling optical field, setting the degeneration factor to be $D = 1$.

We will study following two kinds of optical frequency doubling effects: one is the case of low conversion efficiency without the fundamental frequency light loss (the small signal approximation); another one is the case of high conversion efficiency having the fundamental frequency light loss.

3.2.1 Small Signal Approximation

If the fundamental frequency light is very weak, only a small part of incident fundamental frequency light energy convert to the frequency doubling light energy, i.e., the frequency doubling conversion efficiency is very low. In this case the outputted frequency doubling light power is much smaller than the incident fundamental frequency light power, it can be regard that the amplitude of fundamental frequency light is a constant approximately:

$$E_2(z) = E_1(z) \approx E_1(0). \tag{3.2.1}$$

So when applying Eqs. (3.1.28)–(3.1.30), we can write that

$$\frac{\partial E_1(z)}{\partial z} = 0, \tag{3.2.2}$$

$$\frac{\partial E_2(z)}{\partial z} = 0, \tag{3.2.3}$$

$$\frac{\partial E_3(z)}{\partial z} = \frac{i(2\omega)}{2cn_3}\chi^{(2)}(2\omega;\, \omega,\, \omega)E_1^2(0)e^{-i\Delta kz}, \tag{3.2.4}$$

where

$$\Delta k = 2k_1 - k_3. \tag{3.2.5}$$

We assume that the length of crystal is L, and use boundary condition: $E_1(0)$ = constant, $E_3(0) = 0$, directly taking integral of Eq. (3.2.4), and using $\int_0^L e^{-i\Delta kz}dz = \frac{i}{\Delta k}(e^{-i\Delta kL} - 1)$ to obtain the frequency doubling optical field amplitude outputted from the crystal:

$$E_3(L) = -\frac{\omega\chi^{(2)}}{cn_3\Delta k}E_1^2(0)(e^{-i\Delta kL} - 1). \tag{3.2.6}$$

According to usual practice, we introduce the frequency doubling coefficient d to replace the second-order nonlinear susceptibility $\chi^{(2)}$ [3]:

$$d = \frac{\chi^{(2)}}{2}, \tag{3.2.7}$$

and set $n_1 = n_\omega$, $n_3 = n_{2\omega}$, then Eq. (3.2.6) becomes

$$E_3(L) = -\frac{2\omega d}{cn_{2\omega}\Delta k}E_1^2(0)(e^{-i\Delta kL} - 1). \tag{3.2.8}$$

Using frequency multiplication formula of trigonometric function, from Eq. (3.2.8) we can obtain

$$\begin{aligned}|E_3(L)|^2 &= E_3(L)\cdot E_3(L)^* = \left(\frac{2\omega d}{cn_{2\omega}\Delta k}\right)^2 |E_1(0)|^4 \cdot 4\sin^2(\Delta kL/2)\\ &= \frac{4\omega^2 d^2 L^2}{c^2 n_{2\omega}^2}|E_1(0)|^4 \cdot \frac{\sin^2(\Delta kL/2)}{(\Delta kL/2)^2} = \frac{4\omega^2 d^2 L^2}{c^2 n_{2\omega}^2}|E_1(0)|^4 \operatorname{sinc}^2\left(\frac{\Delta kL}{2}\right).\end{aligned}$$

Using the relation between the intensity and the amplitude for the fundamental frequency light and the frequency doubling light:

$$I_1(0) = \frac{1}{2}\varepsilon_0 cn_\omega |E_1(0)|^2, \tag{3.2.9}$$

$$I_3(L) = \frac{1}{2}\varepsilon_0 cn_{2\omega}|E_3(L)|^2, \tag{3.2.10}$$

we can obtain the relationship between the intensity of outputted frequency doubling light and the intensity of incident fundamental light:

$$I_3(L) = \frac{8\omega^2 d^2 L^2}{\varepsilon_0 n_{2\omega} n_\omega^2 c^3} I_1^2(0) \mathrm{sinc}^2\left(\frac{\Delta kL}{2}\right), \tag{3.2.11}$$

where the relationship between the function $\mathrm{sinc}^2(\Delta kL/2)$ and $\Delta kL/2$ is shown in Fig. 3.5.

The optical frequency doubling efficiency is defined as the ratio of the outputted frequency doubling light power $P_3(L)$ with the inputted fundamental frequency light power $P_1(0)$:

$$\eta = \frac{P_3(L)}{P_1(0)} = \frac{I_3(L)}{I_1(0)} = \frac{8\omega^2 d^2 L^2}{\varepsilon_0 n_{2\omega} n_\omega^2 c^3} \frac{P_1(0)}{S} \mathrm{sinc}^2\left(\frac{\Delta kL}{2}\right). \tag{3.2.12}$$

The above equation has been used $I_1 = P_1/S$, were S is the cross sectional area of incident fundamental frequency light beam.

From Eq. (3.2.12) we can see that, under small signal condition (the loss of fundamental wave is omitted), the frequency doubling process has following properties:

1. When $\Delta k = 0$, $\sin \mathrm{c}^2(\Delta kL/2) = 1$, $\mathrm{n}_{2\omega} = \mathrm{n}_\omega = \mathrm{n}$, the frequency doubling conversion efficiency η takes the maximum:

$$\eta = \frac{8\omega^2 d^2 L^2}{\varepsilon_0 n^3 c^3} \cdot \frac{P_1(0)}{S}. \tag{3.2.13}$$

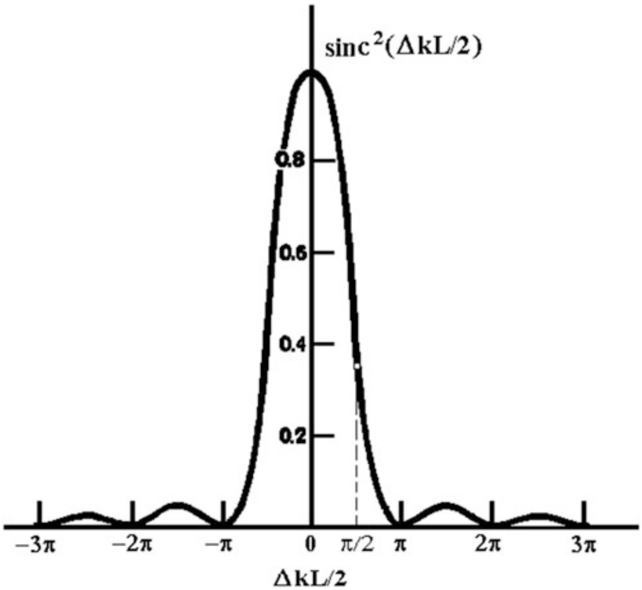


Fig. 3.5 Relationship between function $\mathrm{sinc}^2(\Delta kL/2)$ and $\Delta kL/2$

As you see that the frequency doubling conversion efficiency is proportional to the power of incident fundamental frequency light; is proportional to the frequency doubling coefficient of crystal d and the length of crystal L, and is reverse proportional to the cross sectional area of incident fundamental frequency light S. Therefore, we can used following measures to enhance the frequency doubling efficiency: to select the nonlinear crystal with high frequency doubling coefficient and a longer size; to use the high power fundamental frequency light and the focused fundamental frequency light beam.

2. When $\Delta k \neq 0$, for a certain wave vector mismatch factor Δk, the length of crystal L is equal to so called the coherence length:

$$L_c = \frac{\pi}{\Delta k}, \tag{3.2.14}$$

we have $\frac{\Delta k L_c}{2} = \frac{\pi}{2}$ at the location of imaginary line in Fig. 3.5. If $L < L_c$, there is higher frequency doubling conversion efficiency; if $L > L_c$, the frequency doubling conversion efficiency goes down quickly, after that making a periodic variation with a small amplitude.

3.2.2 High Fundamental Wave Consumption

In high conversion efficiency case, the foundational frequency wave amplitude cannot regard as a constant, the small signal approximation is not applicable. Below we consider the case that the phase matching condition is satisfied ($\Delta k = 0$), i.e., $n_1 = n_2 = n_3 = n$. For the foundational frequency light at $\omega_1 = \omega_2 = \omega$, and $E_1 = E_2$, the degeneration factor is $D = 2$; for the frequency doubling light at $\omega_3 = 2\omega$, the degeneration factor is $D = 1$; replacing the frequency doubling coefficient d to the susceptibility $\chi^{(2)} = 2d$, in this case the three coupling Eqs. (3.1.28)–(3.1.30) becomes the two coupling equations:

$$\frac{\partial E_1(z)}{\partial z} = \frac{i2\omega d}{cn} E_1^*(z) E_3(z), \tag{3.2.15}$$

$$\frac{\partial E_3(z)}{\partial z} = \frac{i2\omega d}{cn} E_1^2(z). \tag{3.2.16}$$

Taking conjugate complex number and multiplying by $E_1(z)$ on the two sides of Eq. (3.2.15), and then multiplying by $E_3^*(z)$ on the two sides of Eq. (3.2.16), finally adding the two equations, then we obtain:

$$\frac{\partial}{\partial z}(|E_1(z)|^2 + |E_3(z)|^2) = 0. \tag{3.2.17}$$

Namely $|E_1(z)|^2 + |E_3(z)|^2 = |E_1(0)|^2 + |E_3(0)|^2$. Because when $z = 0$, $E_3(0) = 0$ and $E_1(0) \neq 0$, then we have

$$|E_1(z)|^2 + |E_3(z)|^2 = |E_1(0)|^2 = \text{constant}. \tag{3.2.18}$$

Visible, at any z-coordinate point in the crystal, the sum of the intensity of foundational frequency light and the intensity of frequency doubling light is equal to the intensity of foundational frequency light at the start point, that is to say, the production of frequency doubling light comes at the expense of the consumption of fundamental frequency light.

Taking module on the both sides of Eq. (3.2.16), then substituting the $|E_1(z)|^2$ obtained from Eq. (3.2.18) into it, and giving a definition of

$$\kappa = \frac{2\omega d}{cn}, \tag{3.2.19}$$

then we obtain

$$\frac{d(|E_3(z)/E_1(0)|)}{dz} = \kappa|E_1(0)|\left[1 - (|E_3(z)/E_1(0)|)^2\right]. \tag{3.2.20}$$

Setting $|E_3(z)/E_1(0)| = v$, making the separation of variables to Eq. (3.2.20), integral on both sides of it, and using integral formula

$$\int \frac{dv}{1 - v^2} = \tanh^{-1} v, \tag{3.2.21}$$

then we obtain the formula for the frequency doubling light field amplitude:

$$|E_3(z)| = |E_1(0)| \tanh(\kappa|E_1(0)|z). \tag{3.2.22}$$

Now we set the hyperbolic tangent function $\tanh(\kappa|E_1(0)|z) = \tanh x = a$, substitute Eq. (3.2.22) into Eq. (3.2.18), and use the relation between the hyperbolic secant function and the hyperbolic tangent function $\mathrm{sech} x = \sqrt{1 - a^2}$, then obtain the formula for the fundamental frequency light field amplitude:

$$|E_1(z)| = |E_1(0)| \mathrm{sech}(\kappa|E_1(0)|z). \tag{3.2.23}$$

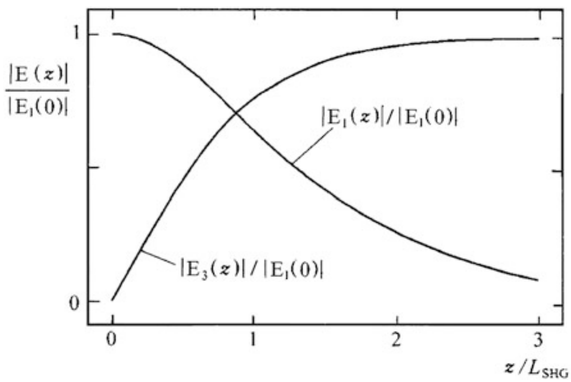


Fig. 3.6 Under the phase matched condition, the curves of the frequency doubling light relative amplitude $|E_3(z)|/|E_1(0)|$ and the foundational frequency light relative amplitude $|E_1(z)|/|E_1(0)|$ versus the relative coordinate z/L_{SHG} along the length direction of crystal

Making a definition of the efficient frequency doubling length:

$$L_{SHG} = [\kappa|E_1(0)|]^{-1} = \left[\frac{2\omega d}{cn}|E_1(0)|\right]^{-1}, \tag{3.2.24}$$

then Eqs. (3.2.22) and (3.2.23) can be written as

$$|E_3(z)| = |E_1(0)|\tanh(z/L_{SHG}), \tag{3.2.25}$$

$$|E_1(z)| = |E_1(0)|\mathrm{sech}(z/L_{SHG}). \tag{3.2.26}$$

Figure 3.6 shows the curves of $|E_3(z)|/|E_1(0)|$ and $|E_1(z)|/|E_1(0)|$ as the function of z/L_{SHG}.

We can see that, under the phase matching condition, the foundational frequency light continually transforms to the frequency doubling light with the increase of the crystal length coordinate. In the theory, when the frequency doubling crystal length achieves to the two times of efficient frequency doubling length, $E_3(z)$ will tend to $E_1(0)$, namely near the frequency doubling conversion efficiency of 100 %. However, in practice, there are many restrictions, such as the absorption and the diffraction of the materials, the reflection from crystal end face, and the laser beam is not monochromic plane wave, etc. For the KDP crystal with length of $L = 2L_{SHG} = 2\,cm$, the frequency doubling conversion efficiency is less than 60 %.

Utilizing the amplitude-intensity relation of Eqs. (3.2.9) and (3.2.10), from Eq. (3.2.25) we can obtain the frequency doubling conversion efficiency formula under the condition of high fundamental wave consumption:

$$\eta = \frac{P_3(L)}{P_1(0)} = \frac{n_{2\omega}}{n_\omega}\frac{|E_3(L)|^2}{|E_1(0)|^2} = \frac{n_{2\omega}}{n_\omega}\tanh^2\frac{L}{L_{SHG}}. \tag{3.2.27}$$

According to Eq. (3.2.24), $(L/L_{SHG})^2 \propto I_1(0)$, in the case of small signal approximation, there is the approximate relation:

$$\tanh^2(L/L_{SHG}) \approx (L/L_{SHG})^2. \tag{3.2.28}$$

For $\Delta k = 0$, $\mathrm{n}_{2\omega} = \mathrm{n}_\omega = \mathrm{n}$ is required, then Eq. (3.2.27) changes to Eq. (3.2.13), which is the frequency doubling conversion efficiency formula for the small signal approximation.

3.2.3 Phase Matching Technology

When the frequency doubling light and the fundamental frequency light are collinear, the phase matching condition is $\Delta k = k_3 - 2k_1 = 0$, or $2k_1 = k_3$, i.e., $2k_\omega = k_{2\omega}$. From wave vector formulas $k_\omega = (\omega/c)n_\omega$ and $k_{2\omega} = (2\omega/c)n_{2\omega}$ we obtain

$$\mathrm{n}_\omega = \mathrm{n}_{2\omega}. \tag{3.2.29}$$

Namely, the phase matching condition requires that the refractive index for the frequency doubling light is equal to the refractive index for the fundamental frequency light in the crystal.

How to realize that the fundamental frequency light and the frequency doubling light induce the same refractive index in an identical crystal? In general, we can use the birefringence characteristic of the anisotropic crystal to realize it.

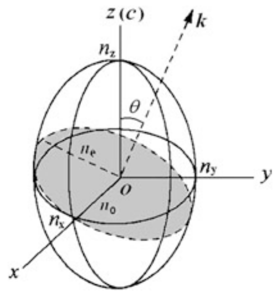
The light at any frequency ω propagates in the anisotropic crystal, besides the direction of optical axis, generally there exists two orthogonal polarization directions with different refractive index, namely $n_\perp(\omega) \neq n_\parallel(\omega)$, such as the situation of o light and e light. However, for two light beams at different frequency (such as the fundamental frequency light at ω and the frequency doubling light at 2ω), it is possible to find a propagation direction that its two orthogonal polarization directions have same refractive index, i.e., $n_\perp(\omega) = n_\parallel(\omega')$, in this way the phase matching condition can be satisfied.

Previous we have talk about that there are 7 crystal systems in the nature, in which 6 crystal systems are belong to anisotropic crystal; only one - cubic crystal system is belong to isotropic crystal. The 6 anisotropic crystals can be divided into two kinds: the uniaxial crystal and the biaxial crystal. The trigonal crystal system, the tetragonal crystal system, and the hexagonal crystal system are belong to the uniaxial crystal; the triclinic crystal system, the monoclinic crystal system and the orthorhombic crystal system are belong to the biaxial crystal. The classification of crystal is shown in Table 3.1.

Table 3.1 Classification of crystal

Classification of crystal		Crystal system
Anisotropic crystal	Uniaxial crystal	Trigonal, tetragonal, hexagonal
	Biaxial crystal	Triclinic, monoclinic, orthorhombic
Isotropic crystal		Cubic

Fig. 3.7 Refractive index ellipsoid of the positive uniaxial crystal



In order to describe the law of light wave propagation in the anisotropic medium, one can use the refractive-index ellipsoid method. We can establish an equation in the principal axis coordinate system O*xyz*:

$$\frac{x^2}{n_x^2} + \frac{y^2}{n_y^2} + \frac{z^2}{n_z^2} = 1. \tag{3.2.30}$$

The index ellipsoid described by this equation is shown in Fig. 3.7. In which n_x, n_y, n_z are the half axis length of rectangular coordinates axis x, y, z, respectively, which are called the principal refractive index.

For the isotropic cubic crystal, $n_x = n_y = n_z = n_0$, Eq. (3.2.30) becomes

$$x^2 + y^2 + z^2 = n_0^2. \tag{3.2.31}$$

This is a ball with radius of n_0, so no matter the light propagation along any direction, the refractive index always is same.

For anisotropic crystal, in general there exists one or two special optical axis directions, the light wave propagates along the optical axis direction without birefringence. The crystal only has one optical axis, which is called the uniaxial crystal. If selecting z-axis as the optical axis c, then $n_x = n_y = n_o$, $n_z = n_e \neq n_o$, the refractive index ellipsoid equation is

$$\frac{x^2}{n_o^2} + \frac{y^2}{n_o^2} + \frac{z^2}{n_e^2} = 1, \tag{3.2.32}$$

where n_o is the refractive index of ordinary light (o light); n_e is the refractive index of extraordinary light (e light). This is an index ellipsoid, its rotation axis is z-axis.

There are two kinds of uniaxial crystals: if $n_o > n_e$, it is a negative uniaxial crystal; if $n_e > n_o$, it is a positive uniaxial crystal. Figure 3.7 shows a refractive index ellipsoid of positive uniaxial crystal.

In Fig. 3.7, wave vector $\boldsymbol{k}$ denotes the propagation direction of light wave, the intersection angle between $\boldsymbol{k}$ and optical axis c is θ. A cross section passing through the original point and perpendicular to $\boldsymbol{k}$ intersects with the ellipsoid to form a ellipse-shape intersecting line. The length of two half axis of the ellipsoid are just refractive indexes n_o and n_e corresponding to two orthogonal polarization states. Visible, the polarization direction of the ordinary light (0 light) is perpendicular to the plane consisted with the optical axis c and wave vector $\boldsymbol{k}$; and the polarization direction of the extraordinary light (e light) is just in that plane.

The refractive index of extraordinary light n_e is a function of intersection angle θ of the optical axis c and the wave vector $\boldsymbol{k}$, It is not difficult to prove that the following formula is satisfied:

$$\frac{1}{n_e^2(\theta)} = \frac{\sin^2\theta}{n_e^2} + \frac{\cos^2\theta}{n_o^2}. \tag{3.2.33}$$

When $\boldsymbol{k}$ is along the optical axis direction, $\theta = 0$, $n_e(0^0) = n_o$; when k is perpendicular to the optical axis direction, $\theta = 90^0$, $n_e(90^0) = n_e$.

We can through adjusting the interaction angle θ between the crystal optical axis c and the incident light wave vector $\boldsymbol{k}$ to change the refractive index of crystal $n_e(\theta)$, to lead the frequency doubling effect satisfying the phase matching condition $\Delta\boldsymbol{k} = 0$. This is called as angle phase matching method [3, 4]. Meanwhile, we also can through varying the temperature of crystal to change refractive index of crystal $n_e(\theta)$, to realize $\Delta\boldsymbol{k} = 0$, this is called the temperature phase matching method.

Now we discuss the so called first phase matching condition, in this condition, two fundamental frequency lights take the same polarization direction. In general, the polarization state of fundamental frequency light is selected with higher refractive index. Namely for negative uniaxial crystal ($n_o > n_e$), taking the fundamental frequency light as o polarization state, the frequency doubling light as e polarization state; for positive uniaxial crystal ($n_e > n_o$), taking the fundamental frequency light as e polarization state, the frequency doubling light as o polarization state.

The phase matching condition for the negative uniaxial crystal is

$$n_{\mathrm{o}}^{\omega} = n_e^{\omega}(\theta_m). \tag{3.2.34}$$

Figure 3.8 draws a refractive index surface diagram in the negative uniaxial crystal when the phase matching between the fundamental frequency light and the frequency doubling light are sufficed. The solid line are the o light index ball and the e light index ellipsoid for the fundamental frequency light at frequency of ω; the imaginary line are o light index ball and the e light index ellipsoid for the frequency doubling light at frequency of 2 ω. We can see that there is a point of intersection between the o light index surface of fundamental wave and the e light index surface

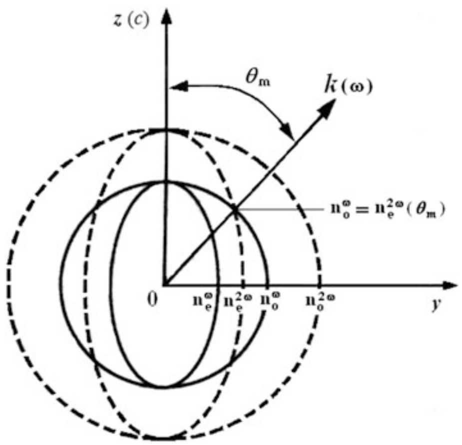


Fig. 3.8 In negative uniaxial axis crystal, the fundamental frequency light and the frequency doubling light matched refractive-index surface diagram. The *solid line* are the o light index ball and the e light index ellipsoid for the fundamental frequency light at frequency ω; the *imaginary line* are the o light index ball and the e light index ellipsoid for the frequency doubling light at frequency 2ω

of frequency doubling wave, the intersection point corresponded the intersection angle between the wave vector and optical axis θ_m satisfies Eq. (3.2.34), θ_m is just phase matching angle. In the figure, n_o^ω, n_e^ω and $n_o^{2\omega}$, $n_e^{2\omega}$ are two principal refractive indexes of the fundamental frequency light and the frequency doubling light, respectively.

Substituting Eq. (3.2.34) into Eq. (3.2.33), we can obtain the formula of the phase matching angle of negative uniaxial crystal θ_m:

$$\sin^2\theta_m = \frac{\left(n_o^\omega\right)^{-2} - \left(n_o^{2\omega}\right)^{-2}}{\left(n_e^{2\omega}\right)^{-2} - \left(n_o^{2\omega}\right)^{-2}}. \tag{3.2.35}$$

For example, in negative uniaxial crystal KDP, using the ordinary light of ruby laser at frequency of 0.6943 μm as the fundamental frequency, under the phase matching condition, the matching angle is $\theta_m = 50.4°$, the extraordinary light of frequency doubling wave at frequency of 0.3471 μm can be obtained.

The phase matching condition of the positive uniaxial crystal is

$$n_o^{2\omega} = n_e^\omega(\theta_m). \tag{3.2.36}$$

Figure 3.9 draws a refractive index surface diagram in positive uniaxial crystal when the phase matching between the fundamental frequency light and the frequency doubling light are sufficed. Visible, there is an intersection point between the fundamental wave e light refractive-index surface and the frequency doubling wave o light refractive-index surface. The intersection point corresponded intersection angle between the wave vector and the optical axis θ_m satisfies Eq. (3.2.34), θ_m is just the phase matching angle.

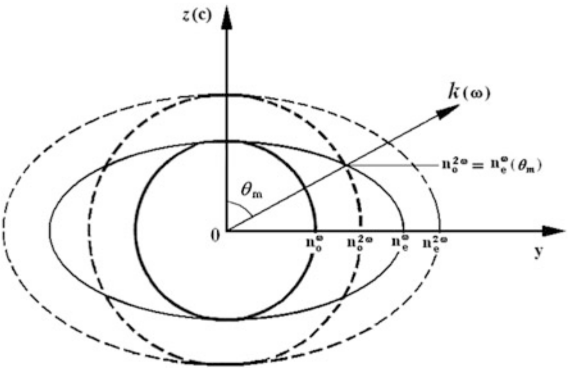


Fig. 3.9 In positive uniaxial crystal, the fundamental frequency light and the frequency doubling light matched refractive-index surface diagram. The *solid line* are the o light index ball and the e light index ellipsoid for the fundamental frequency light at frequency ω; the *imaginary line* are the o light index ball and the e light index ellipsoid for the frequency doubling light at frequency 2ω

Substituting Eq. (3.2.36) into Eq. (3.2.33), in same way the possible uniaxial crystal phase matching angle θ_m formula can be obtain:

$$\sin^2 \theta_m = \frac{\left(n_o^{2\omega}\right)^{-2} - \left(n_o^{\omega}\right)^{-2}}{\left(n_e^{\omega}\right)^{-2} - \left(n_o^{\omega}\right)^{-2}}. \tag{3.2.37}$$

In above phase matching condition, two fundamental frequency lights take same polarization direction, we call the first class phase match. In which, the polarization characteristic of negative uniaxial crustal is denoted by symbol o + o → e; the polarization characteristic of positive uniaxial crustal is denoted by symbol e + e → o. their phase matching condition are list in the Table 3.2.

Actually, the phase matching condition also exists second class phase matching scheme, that is taking perpendicular propagation directions of two foundational frequency lights: one is o light, another one is e light. Its polarization characteristics for negative uniaxial crystal is o + e → e; for positive uniaxial crystal is o + e → o. Their phase matching condition is also listed Table 3.2. About the

Table 3.2 Phase matching condition of uniaxial crystal

Crystal type	First class phase match		Second class phase match	
	Polarization characteristic	Phase matching condition	Polarization characteristic	Phase matching condition
Negative uniaxial crystal	o + e → e	$n_o^{\omega} = n_e^{2\omega}(\theta_m)$	o + o → e	$\frac{1}{2}[n_o^{\omega}(\theta_m) + n_e^{\omega}] = n_o^{2\omega}(\theta_m)$
Positive uniaxial crystal	o + e → o	$n_o^{2\omega} = n_e^{\omega}(\theta_m)$	e + e → o	$\frac{1}{2}[n_o^{\omega} + n_e^{\omega}(\theta_m)] = n_o^{2\omega}$

deduction of second class phase matching condition, reader can refer to the related reference material, here we do not give unnecessary details.

Except for the uniaxial crystal phase matching condition, there is the biaxial crystal phase matching condition, if you want to know the knowledge in this aspect, please also refer to the related reference material.

3.2.4 Experimental Facilities for Second Harmonic Generation

Figure 3.10 gives several schematic diagrams of typical experimental facilities, these facilities are composed by three parts: the nonlinear optical crystal, the fundamental wave source, and the phase matching system.

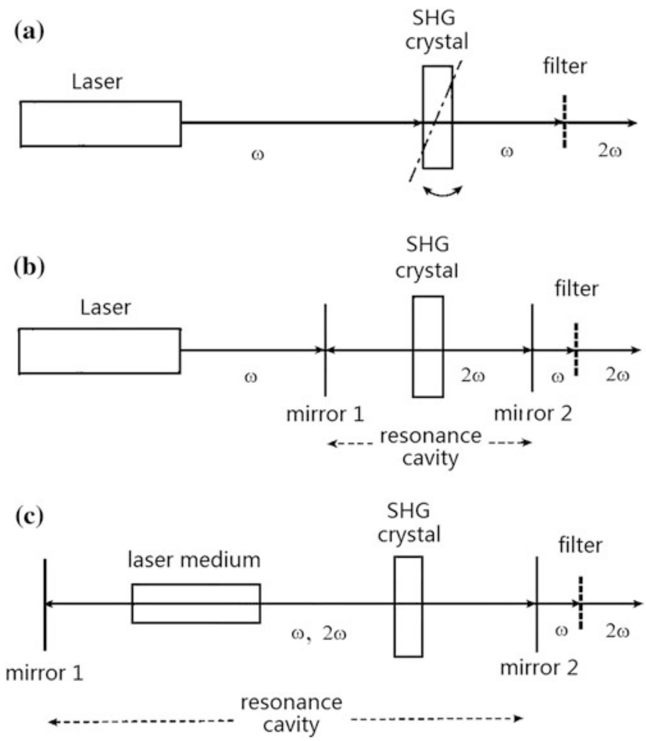


Fig. 3.10 Typical experimental facilities for the optical second harmonic generation **a** single-pass mode; **b** external cavity mode; **c** intracavity mode

1. **Nonlinear Optical Crystal**

Generally people use the artificial growth bulk or flake-like high quality crystal, the optical axis of crystal relative to the incident fundamental wave has a certain matching angle. To reduce the reflection loss, it can plate the antireflection coating on crystal two parallel end surface. According to the different methods of phase matching, the nonlinear crystal is divided into the following three categories:

(1) Angle tuning phase matching crystal: KDP, ADP, KTP, $LiIO_3$, BBO, LBO etc. Frequency doubling of the incidence laser at the near infrared or visible wavelength, to produce the visible light or near ultraviolet light, the efficiency can reach 30 and 50 %.
(2) Temperature tuning phase matching crystal: $LiNbO_3$, $KNbO_3$, Ba_2NaNbO_{15} etc. They have better optical transmission property in the spectrum region 0.4-5μm, it can be used to produce near-infrared frequency doubling light, the efficiency is higher than 50 %.
(3) Semiconductor crystal for producing infrared second harmonic wave, such as Ag_3AsS_3, $AgGaSe_2$, $CdGeAs_2$, CdSe, GaSe, etc. These crystals have high second-order nonlinear susceptibility, in the wide infrared spectrum region have better transmittance.

2. **Fundamental Wave Optical Source**

Mostly people adopt the solid pulse lasers as the optical sources, such as Nd-glass laser, Nd-doped Gamet laser, Ruby laser, etc. The CW solid and liquid laser can also be used for continuous frequency doubling light output. In order to enhance the frequency doubling conversion efficiency, the focusing light passing through the crystal also can be used.

3. **Phase Matching System**

According to the different nonlinear crystals and experimental conditions, the different phase matching, exciting and coupling methods can be used. Figure 3.10a shows a usually used the mode that the light beam single-pass through the crystal, it is suitable to the angle phase matching method. Figure 3.10b showed equipment is suitable to temperature phase matching frequency doubling crystal, the crystal inserts into the resonance cavity, let's lower power fundamental light multiple-pass through the crystal, to enhance the convention efficiency. It also can insert the frequency doubling crystal into the fundamental wave laser cavity, as shown in Fig. 3.10c, because the light intensity inside laser cavity is much stronger than outputted light intensity outside the cavity, in benefit of enhancement of conversion efficiency.

3.3 Optical Sum Frequency, Difference Frequency and Parameter Amplification

3.3.1 Optical Sum Frequency and Frequency Up-Conversion

Now we discuss the coupled Eqs. (3.1.28)–(3.1.30), which will be used for the sum frequency and difference frequency processes. In order to simplify the equations, we define a set of new light electric field amplitudes A_i and nonlinear coefficient κ, which are

$$A_i(z) = \sqrt{\frac{n_i}{\omega_i}} E_i(z) \quad (i = 1, 2, 3), \tag{3.3.1}$$

$$\kappa = \frac{\chi^{(2)}}{2c}\sqrt{\frac{\omega_1\omega_2\omega_3}{n_1 n_2 n_3}} = \frac{d}{c}\sqrt{\frac{\omega_1\omega_2\omega_3}{n_1 n_2 n_3}}, \tag{3.3.2}$$

where d is the frequency doubling coefficient. So the coupled Eqs. (3.1.28)–(3.1.30) are simplified as

$$\frac{\partial A_1(\mathrm{z})}{\partial \mathrm{z}} = iD\kappa A_2^*(z)A_3(z)e^{i\Delta kz}, \tag{3.3.3}$$

$$\frac{\partial A_2(\mathrm{z})}{\partial \mathrm{z}} = iD\kappa A_3(z)A_1^*(z)e^{i\Delta kz}, \tag{3.3.4}$$

$$\frac{\partial A_3(\mathrm{z})}{\partial \mathrm{z}} = iD\kappa A_1(z)A_2(z)e^{-i\Delta kz}, \tag{3.3.5}$$

where $\Delta k = k_1 + k_2 - k_3 = 0$.

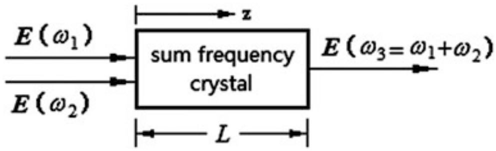
Now we discuss the sum frequency process. There are three photons at different frequency joining this process. According to Eqs. (3.1.13) and (3.1.14), they satisfy following the energy and momentum conservation relations, respectively:

$$\omega_3 = \omega_1 + \omega_2, \tag{3.3.6}$$

$$\boldsymbol{k}_3 = \boldsymbol{k}_1 + \boldsymbol{k}_2. \tag{3.3.7}$$

The optical sum frequency can be used for the frequency upconversion [5, 6], that is an effective means to produce shorter wavelength coherent radiation. For example, using crystal Ag_3AsS_3 as a sum frequency crystal, a 1.06-wavelength YAG laser as the pumping light (ω_2), to convert the 10.6 μm-wavelength CO_2 infrared light (ω_1) into the 96 μm-wavelength visible light (ω_3). That is because the detection of middle and far infrared light must use the refrigerant detector, the detection of visible light can use the room temperature fast detector.

Fig. 3.11 Schematic diagram of the collinear optical sum frequency process



Assuming that the three waves at ω_1, ω_2 and ω_3 participated in sum frequency process collinearly propagate in the nonlinear crystal, all along z-direction, as shown in Fig. 3.11, we will used coupled Eqs. (3.3.3)–(3.3.5) to calculate the variation of field amplitudes of the sum frequency light (ω_3) and the signal light (ω_1) with the coordinate axis z.

We suppose that the intensity of pump light ω_2 is strong enough, so that its amplitude does not change in the sum frequency process, i.e., it is a constant:

$$A_2(z) \approx A_2(0) = \text{constant}. \tag{3.3.8}$$

Thus the three coupled Eqs. (3.3.3)–(3.3.5) becomes two. Considering the frequency without degeneration, taking $D = 2$; and to defied a sum-frequency nonlinear coefficient κ_{SF}, it is two times of the original nonlinear coefficient κ, i.e.,

$$\kappa_{SF}=2\kappa, \tag{3.3.9}$$

Therefore the two coupled equations for sum frequency process are given by

$$\frac{\partial A_1(z)}{\partial z} = i\kappa_{SF}A_2(0)A_3(z)e^{i\Delta kz}, \tag{3.3.10}$$

$$\frac{\partial A_3(z)}{\partial z} = i\kappa_{SF}A_1(z)A_2(0)e^{-i\Delta kz}. \tag{3.3.11}$$

We further define g_{SF} is the sum frequency gain factor (suppose it is a real number):

$$g_{SF} = \kappa_{SF}A_2(0) = \frac{2d}{c}\sqrt{\frac{\omega_1\omega_3}{n_1n_3}}\boldsymbol{E}_2(0), \tag{3.3.12}$$

Under phase matching condition, i.e., $\Delta k = 0$, Eqs. (3.3.10) and (3.3.11) are simplified to

$$\frac{\partial A_1(z)}{\partial z} = ig_{SF}A_3(z), \tag{3.3.13}$$

$$\frac{\partial A_3(z)}{\partial z} = ig_{SF}A_1(z). \tag{3.3.14}$$

Then making derivation of two side of Eq. (3.3.14), and substituting Eq. (3.3.13) into it, we obtain

$$\frac{d^2A_3(z)}{dz^2} + g_{SF}^2 A_3(z) = 0. \tag{3.3.15}$$

The general solution of Eq. (3.3.15) is

$$A_3(z) = C_1 \cos(g_{SF}z) + C_2 \sin(g_{SF}z). \tag{3.3.16}$$

To take $z = 0$ in Eq. (3.3.16), and use boundary condition $A_3(0) = 0$, obtain $C_1 = 0$. Equation (3.3.16) becomes

$$A_3(z) = C_2 \sin(g_{SF}z). \tag{3.3.17}$$

Then we substitute Eq. (3.3.17) into (3.3.14), after derivation, set $z = 0$ to get $C_2 = iA_1(0)$. So from Eq. (3.3.16) to obtain

$$A_3(z) = iA_1(0) \sin(g_{SF}z). \tag{3.3.18}$$

Further substituting Eq. (3.3.18) into (3.3.14), after derivation to obtain

$$A_1(z) = A_1(0) \cos(g_{SF}z), \tag{3.3.19}$$

To take mode square of above two amplitudes $A_3(z)$ and $A_1(z)$ respectively, then add together, we obtain

$$|A_1(z)|^2 + |A_3(z)|^2 = |A_1(0)|^2. \tag{3.3.20}$$

Using relations

$$I_1 = \frac{1}{2}\varepsilon_0 c n_1 |E_1|^2 = \frac{1}{2}\varepsilon_0 c \omega_1 |A_1|^2, \tag{3.3.21}$$

$$I_3 = \frac{1}{2}\varepsilon_0 c n_3 |E_3|^2 = \frac{1}{2}\varepsilon_0 c \omega_3 |A_3|^2, \tag{3.3.22}$$

we obtain

$$\left(\frac{\omega_1}{\omega_3}\right) I_3(z) + I_1(z) = I_1(0). \tag{3.3.23}$$

We can see that because the intensity of incident signal $I_1(0)$ is a constant, the increase of light intensity $I_3(z)$ is the price of the decrease of light intensity $I_1(z)$. From Eq. (3.3.19) to obtain $|\boldsymbol{A}_1|^2$, then use Eqs. (3.3.21) and (3.3.23), we obtain

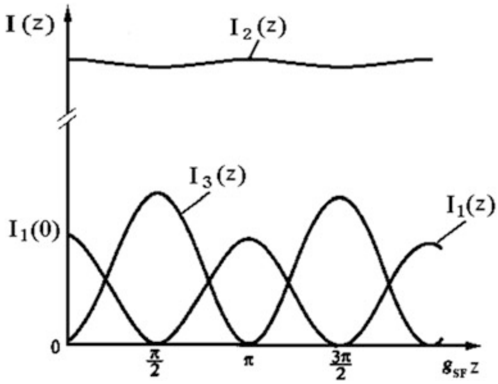


Fig. 3.12 In the phase match sum frequency process the *curves* of variation of intensities of two light beams at ω_1 and ω_3 with the distance $g_{SF}z$

$$I_3(z) = \frac{\omega_3}{\omega_1} I_1(0)\sin^2(g_{SF}z). \tag{3.3.24}$$

If the length of crystal is L, the conversion efficiency of sum frequency is

$$\eta = \frac{I_3(L)}{I_1(0)} = \frac{\omega_3}{\omega_1}\sin^2(g_{SF}L). \tag{3.3.25}$$

Figure 3.12 shows that in sum frequency process, under phase matching condition, the curves of variation of light intensities of two beams at frequency ω_1 and ω_3 with distance z.

Figure 3.12 can be explained as follows:in the beginning the intensity of light wave at ω_1 drop off gradually, its energy transfers to the light wave at ω_3 . When the propagation distance increases to $g_{SF}z = \pi/2$, the conversion efficiency reaches to maximum. In this case, $\eta > 1$, that is due to except all $I_1(0)$ converts to $I_3(z)$ at this point, actually there is a small part light coming from pump light $I_2(z)$ at ω_2 . It can be proved that the sum of three light intensities remains unchanged, i.e., $I_1 + I_2 + I_3 = constant$. After the intensity of light wave at ω_3 reaches the peak, it will pass through the difference frequency with the pump light at ω_2 to send its energy back to the light at $\omega_1 = \omega_3 - \omega_2$. Therefore, the periodic oscillation situation will appear.

If the intensity of pump light $I_2(z)$ at frequency ω_2 is very small, from gain factor definition Eq. (3.3.12) we can know that g_{SF} is also very small, so in the efficiency Eq. (3.3.25) we have

$$\sin^2(g_{SF}L) \approx (g_{SF}L)^2, \tag{3.3.26}$$

This is the case of small signal approximation. Therefore we obtain the frequency conversion efficiency formula in the case of the small signal approximation and $\Delta k = 0$:

$$\eta \approx \frac{\omega_3}{\omega_1} g_{SF}^2 L^2 = \frac{8\omega_3^2 d^2 L^2 I_2(0)}{\varepsilon_0 n_1 n_2 n_3 c^3}. \tag{3.3.27}$$

It can be proved, in the case of the small signal approximation and $\Delta k \neq 0$, the frequency conversion efficiency formula becomes

$$\eta \approx \frac{\omega_3}{\omega_1} g_{SF}^2 L^2 = \frac{8\omega_3^2 d^2 L^2 I_2(0)}{\varepsilon_0 n_1 n_2 n_3 c^3} \mathrm{sinc}^2(\Delta kL/2). \tag{3.3.28}$$

We can see that Eq. (3.3.28) is only an oscillation factor more than Eq. (3.3.27).

3.3.2 Optical Difference Frequency and Frequency Down-Conversion

In the optical difference frequency process, according to the energy and momentum conservation laws, the frequencies and wave vectors are required to satisfy the following relations:

$$\omega_2 = \omega_3 - \omega_1, \tag{3.3.29}$$

$$\boldsymbol{k}_2 = \boldsymbol{k}_3 - \boldsymbol{k}_1. \tag{3.3.30}$$

Figure 3.13 is a schematic diagram of the optical difference frequency process in the z-direction collinear propagation case. To utilize this process can realize frequency down-conversion: using the difference frequency of two visible laser (ω_3 and ω_1) to obtain an infrared laser ($\omega_2 = \omega_3 - \omega_1$) output. For example using $LiNbO_3$ as a difference frequency crystal, and a 532 nm-wavelength YAG frequency doubled laser (ω_3) as a pump laser, it makes difference frequency with a tunable dye laser (ω_1) with the wavelength range of 575–650 nm, the result of difference frequency can obtain a tunable infrared laser (ω_2) output with the wavelength range of 3.40–5.65 μm [7].

Assuming that the intensity of the pump light at frequency ω_3 is strong enough, so that its intensity can be regarded do not change in the difference frequency process, we have

$$A_3(z) \approx A_3(0), \tag{3.3.31}$$

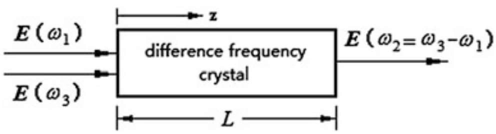


Fig. 3.13 Schematic diagram of optical difference frequency or frequency down-conversion process

Suppose $A_3(0)$ is a real number, and taking $D = 2$ for the difference frequency process, then three coupled Eqs. (3.3.3)–(3.3.5) become two equations:

$$\frac{\partial A_1(z)}{\partial z} = i\kappa_{DF} A_2^*(z) A_3(0) e^{i\Delta kz}, \tag{3.3.32}$$

$$\frac{\partial A_2(z)}{\partial z} = i\kappa_{DF} A_1^*(z) A_3(0) e^{i\Delta kz}, \tag{3.3.33}$$

where κ_{DF} is the nonlinear coupling coefficient of difference frequency, the relationship between κ_{DF} and κ is

$$\kappa_{DF} = 2\kappa. \tag{3.3.34}$$

Under the case of $\Delta k = 0$, Eqs. (3.3.32) and (3.3.33) are simplified as

$$\frac{\partial A_1(z)}{\partial z} = ig_{DF} A_2^*(z), \tag{3.3.35}$$

$$\frac{\partial A_2(z)}{\partial z} = ig_{DF} A_1^*(z), \tag{3.3.36}$$

where g_{DF} is defined as the gain coefficient of difference frequency:

$$g_{DF} = \kappa_{DF} A_3(0) = \frac{2d}{c}\sqrt{\frac{\omega_1 \omega_2}{n_1 n_2}} \boldsymbol{E}_3(0). \tag{3.3.37}$$

Making derivation of two side of Eq. (3.3.35), and substituting the conjugate Eq. (3.3.36) into it, then we obtain

$$\frac{d^2 A_1(z)}{dz^2} - g_{DF}^2 A_1(z) = 0. \tag{3.3.38}$$

The general solution of Eq. (3.3.38) is

$$A_1(z) = D_1 \sinh(g_{DF} z) + D_2 \cosh(g_{DF} z). \tag{3.3.39}$$

To utilize the bounder condition at $z = 0 : A_2(0) = 0$ and $A_1(0) \neq 0$, from Eqs. (3.3.39) to (3.3.36), we obtain the following field amplitudes:

$$A_1(z) = A_1(0) \cosh(g_{DF} z), \tag{3.3.40}$$

$$A_2^*(z) = -iA_1(0) \sinh(g_{DF} z). \tag{3.3.41}$$

Figure 3.14 draws the variation of two field amplitudes with z. from the figure we can see that the difference frequency generation field at frequency ω_2 and the

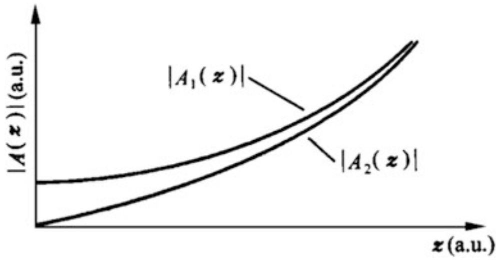


Fig. 3.14 Characteristics of the variation of field amplitude $|A_1(z)|$ and $|A_2(z)|$ with z

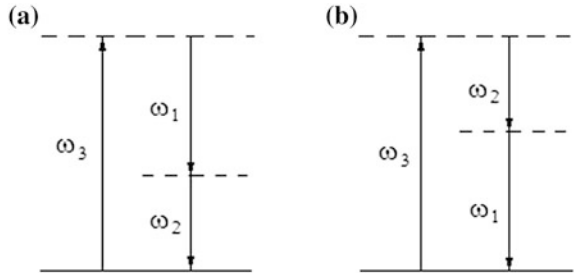


Fig. 3.15 Explanation of difference frequency characteristics: **a** the signal filed ω_1 excites to generate the difference frequency filed $\omega_2 = \omega_3 - \omega_1$; **b** the difference frequency filed ω_2 excites to generation the new signal field ω_1

signal field at ω_1 monotonously increase at the same time in the nonlinear interaction, it is total different with the sum frequency appeared oscillation.

Following figure of the energy level transition is used for explaining the reason of the difference frequency generation field and the signal filed monotonously increase at the same time.

Figure 3.15a shows the signal field ω_1 excites to generate the difference frequency field $\omega_2 = \omega_3 - \omega_1$, Fig. 3.15b shows the difference frequency field ω_2 excites to generate the signal field ω_1, the new signal field ω_1 again enhances the generation of the new difference frequency field ω_2, such repetition, to lead the two fields exponentially growth.

From Eqs. (3.3.40) to (3.3.41) we obtain the amplitude square formulas:

$$|A_1(z)|^2 = |A_1(0)|^2 \cosh^2(gz), \tag{3.3.42}$$

$$|A_2(z)|^2 = |A_1(0)|^2 \sinh^2(gz). \tag{3.3.43}$$

If the length of crystal is L, from Eq. (3.3.43) and relationships

$$I_1 = \frac{1}{2}\varepsilon_0 c n_1 |E_1|^2 = \frac{1}{2}\varepsilon_0 c \omega_1 |A_1|^2, \tag{3.3.44}$$

$$I_2 = \frac{1}{2}\varepsilon_0 c n_2 |E_2|^2 = \frac{1}{2}\varepsilon_0 c \omega_2 |A_2|^2, \tag{3.3.45}$$

we obtain the difference frequency conversion efficiency formula under $\Delta k = 0$:

$$\eta = \frac{I_2(L)}{I_1(0)} = \frac{\omega_2}{\omega_1}\sinh^2(g_{DF}L). \tag{3.3.46}$$

In small signal case, the pump light $E_3(0)$ is small, from Eq. (3.3.37) g_{DF} is also small, so that in Eq. (3.3.46) we have

$$\sinh^2(g_{DF}L) \approx (g_{DF}L)^2, \tag{3.3.47}$$

Therefore, under the small signal and $\Delta k = 0$ case the difference frequency conversion efficiency is

$$\eta \approx \frac{\omega_2}{\omega_1}(g_{DF}L)^2 = \frac{8\omega_2^2 d^2 L^2 I_3(0)}{\varepsilon_0 n_1 n_2 n_3 c^3}. \tag{3.3.48}$$

3.3.3 *Optical Parametric Amplification*

In the process similar to the difference frequency, the pump light energy gradually transfers to the signal light with the increase of propagation distance, leads the signal light to amplify, and in the same time generates the idler frequency light, this process is similar to the parametric amplification in microwave waveband, so it is called the optical parametric amplification (OPA) [8]. Suppose that the pump light at the frequency of $\omega_3 = \omega_p$ with the amplitude $E_3 = E_p$; the signal light at the frequency of $\omega_1 = \omega_s$ with the amplitude $E_1 = E_s$; the idler light at the frequency of $\omega_2 = \omega_i$ with the amplitude $E_2 = E_i$, the optical parametric amplification process is shown in Fig. 3.16.

We can regard the gain coefficient of difference frequency g_{DF} as the gain coefficient of parametric amplification g, which is described by Eq. (3.3.37).

In the beginning, $I_1(0) \neq 0$ and $I_2(0) = 0$. If the pump light filed $E_3(0)$ is very strong, we have $gz > > 1$, so that

$$\sinh gz = \frac{e^{gz} - e^{-gz}}{2} \approx \frac{1}{2}e^{gz}, \quad \text{and } \cosh gz = \frac{e^{gz} + e^{-gz}}{2} \approx \frac{1}{2}e^{gz},$$

then Eqs. (3.3.42) and (3.3.43) becomes

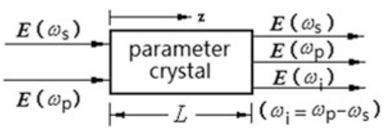


Fig. 3.16 Schematic diagram of optical parametric amplification process

$$|A_1(z)|^2 \approx \frac{1}{4}|A_1(0)|^2 e^{2gz}, \tag{3.3.50}$$

$$|A_2(z)|^2 \approx \frac{1}{4}|A_1(0)|^2 e^{2gz}. \tag{3.3.51}$$

That means that in this case, the intensity of idler light is equal to the intensity of signal light.

According to Eq. (3.3.50), the magnification of parametric amplification M for signal light is defined as

$$M = \frac{I_1(z)}{I_1(0)} = \frac{|A_1(z)|^2}{|A_1(0)|^2} \approx \frac{1}{4}e^{2gz}. \tag{3.3.45}$$

Because the gain coefficient of parametric amplification g is proportional to the pump light filed amplitude $E_3(0)$, form Eq. (3.3.45) we can see that the magnification of parametric amplifier exponentially enhances with increase of $E_3(0)$. Due to g is proportional to $d \propto \chi^{(2)}$, so that the second-order nonlinear susceptibility decides the ability of parametric amplification.

3.3.4 Comparison of Four Kinds of Three-Wave Mixing Processes and Experimental Facilities

1. **Comparison of Characteristics of Four Kinds of Three-Wave Mixing Processes**

Previous we introduced 4 kinds of three-wave mixing processes: the second harmonic generation (SHG), the sum frequency generation (SFG), the difference frequency generation (DFG) and the optical parametric amplification (OPA), as shown in Fig. 3.17.

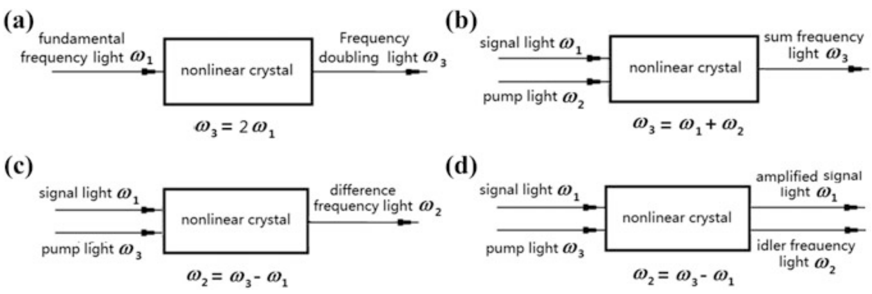


Fig. 3.17 Three-wave mixing processes: **a** second harmonic generation; **b** sum frequency generation; **c** difference frequency generation; **d** optical parametric amplification

The second harmonic is a particular case of the sum frequency $\omega_3 = \omega_1 + \omega_2$, both are belong to the case that the energy of two low-frequency light fields transfer to that of a high-frequency light field, i.e., the frequency up-conversion; the optical parametric amplification is particular case of the difference frequency, both are belong to the case that the energy of two high-frequency light fields transfer to that of a low-frequency light field, i.e., the frequency down-conversion. The difference of both is that the difference frequency pays attention to the generation of the difference frequency light at $\omega_2 = \omega_3 - \omega_1$, however the optical parametric amplification pays attention to the amplification of the signal light at ω_1 (the light at ω_2 regards as the idler-frequency light). For the three processes: the frequency doubling, the sum frequency and the difference frequency, the power conversion efficiency η needs to be studied. For the parametric amplification, the magnification M is instead of η. The pump light is different in above 4 different processes: in second harmonic process, it is the fundamental frequency light at ω_1; in sum frequency process, it is the light at ω_2; in difference frequency process and parametric amplification process, it is the light at ω_3.

2. **Experimental Facilities of Three-Wave Mixing Process**

Above four kinds of optical three-wave mixing processes have similar generation mechanism, the requirements of the nonlinear crystal materials and the phase matching condition are the same.

The common requirements of nonlinear crystals are (1) piezo-electric crystal without center symmetry; (2) the phase match in certain way is satisfied, for example the angle match or the temperature match. The propagation directions of three waves can be different, but should satisfy the momentum conservation condition; (3) the crystal materials have good optical transparency for the two incident lights and one generated light.

To three-wave mixing experimental systems we have following same requirements: (1) two incident light sources at different frequencies; (2) the facilities to realize angle match or temperature match; (3) the dispersion element and absorption element (prism, grating and filter, etc.) used for separating the transmitted lights at different frequencies.

As an example, Fig. 3.18 gives a typical three-wave mixing (such as sum frequency) experimental setup.

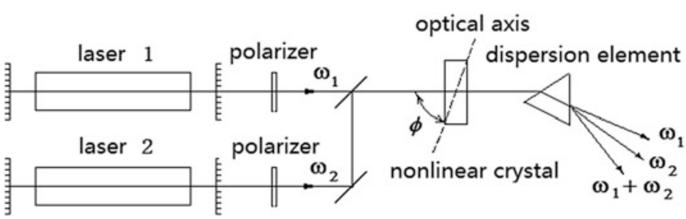


Fig. 3.18 Typical experimental setup for three-wave mixing (sum frequency)

3.4 Optical Parametric Oscillator

Because the amplification factor of the single pass through the nonlinear crystal in the optical parametric amplification is small, in order to enhance the energy conversion efficiency, we can place the parametric amplifier into a resonant cavity, the lights at frequency ω_s (and ω_i) oscillates in the cavity to be enhanced, when the energy of pump light at frequency ω_p over a certain threshold value, the gain of the nonlinear interaction overcomes the intracavity loss, then a stable light beam at frequency of ω_s (and ω_i) can be outputted from the cavity, this device is called the optical parametric oscillator (OPO) [9, 10].

In comparison of the parametric oscillator with the laser oscillator, the similarities is that both can generate the coherent light output; the difference is that the gain in the cavity of optical parametric oscillator is generated by the nonlinear effect, not by the population inversion; and the gain is in one way, the returning light cannot be enhanced, only be wastage.

3.4.1 Threshold Value Equations of Optical Parametric Oscillation

In order to deduce the optical parametric oscillation threshold value equation, suppose the length of crystal is L, two ends of crystal is fabricated to be spherical mirrors with the equal radius of curvature, their amplitude reflectivity are r_1 and r_2 for the signal light at frequency ω_1 and the idler light at frequency ω_2, respectively; the intensity reflectivity are $R_1 = |r_1|^2$ and $R_2 = |r_2|^2$, respectively; and the pump light at frequency ω_3 is transparent, as shown in Fig. 3.19.

Suppose the pump light intensity in the cavity is independent of propagation distance, the signal light electrical field and the idler light electrical field on the plane at any position z in the cavity can be expressed by a matrix $\tilde{A}(z)$:

$$\tilde{\boldsymbol{A}}(z) = \begin{vmatrix} A_1(z)e^{ik_1 z} \\ A_2^*(z)e^{-ik_2 z} \end{vmatrix}. \tag{3.4.1}$$

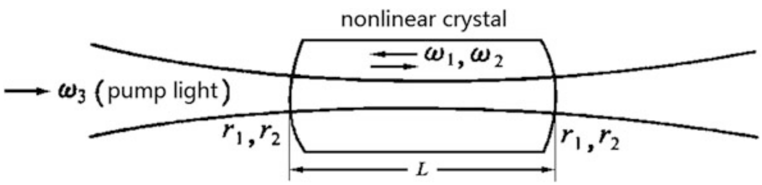


Fig. 3.19 Schematic diagram of the parametric oscillator with crystal structure

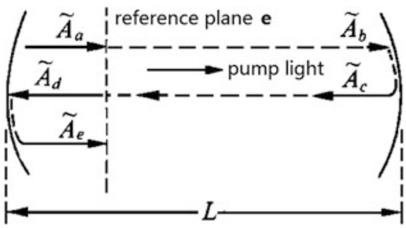


Fig. 3.20 The signal light and the idler light satisfy the self-consistent condition

Considering that under the excitation of pump light ω_3, at $z = 0$ the spontaneous radiation of the signal light at ω_1 and the idler light at ω_2 are produced in the meantime, namely $A_1(0) \neq 0$ and $A_2(0) \neq 0$, the solutions of coupled Eqs. (3.3.32) and (3.3.33) in difference frequency process are

$$A_1(z) = A_1(0)\cosh(gz) + iA_2^*(0)\sinh(gz), \tag{3.4.2}$$

$$A_2^*(z) = A_2^*(0)\cosh(gz) - iA_1(0)\sinh(gz). \tag{3.4.3}$$

The optical field amplitude at $z = L$ is

$$\tilde{\boldsymbol{A}}(L) = \begin{vmatrix} e^{ik_1L}\cosh(gL) & ie^{ik_1L}\sinh(gL) \\ \text{-}\ ie^{-k_2L}\sinh(gL) & e^{-k_2L}\cosh(gL) \end{vmatrix}\tilde{\boldsymbol{A}}(0). \tag{3.4.4}$$

The stable oscillation requires satisfying the self-consistent condition that after the light propagation for a round trip in the cavity $\tilde{\boldsymbol{A}}(z)$ is invariable, as shown in Fig. 3.20.

At the reference plane e, it should have

$$\tilde{\boldsymbol{A}}_e(z) = \tilde{\boldsymbol{A}}_a(z), \tag{3.4.5}$$

$\tilde{\boldsymbol{A}}_e(z)$ is obtained from $\tilde{\boldsymbol{A}}_a(z)$ multiplies the following 4 matrixes: the parametric amplification matrix for the light propagation from left to right, the reflection matrix at the end of right, the propagation matrix from right to left without gain, and the reflection matrix at the end of left, namely

$$\tilde{\boldsymbol{A}}_e = \begin{vmatrix} r_1 & 0 \\ 0 & r_2^* \end{vmatrix}\begin{vmatrix} e^{\mathrm{i}k_1L} & 0 \\ 0 & e^{-\mathrm{i}k_2L} \end{vmatrix}\begin{vmatrix} r_1 & 0 \\ 0 & r_2^* \end{vmatrix}\begin{vmatrix} e^{\mathrm{i}k_1L}\cosh(gL) & ie^{\mathrm{i}k_1L}\sinh(gL) \\ -ie^{-\mathrm{i}k_2L}\sinh(gL) & e^{-\mathrm{i}k_2L}\cosh(gL) \end{vmatrix}\tilde{\boldsymbol{A}}_a. \tag{3.4.6}$$

That is

$$\tilde{\boldsymbol{A}}_e = \begin{vmatrix} r_1^2\cosh(gL)e^{\mathrm{i}2k_1L} & ir_1^2\sinh(gL)e^{\mathrm{i}2k_1L} \\ -i(r_2^*)^2\sinh(gL)e^{-\mathrm{i}2k_2L} & (r_2^*)^2\cosh(gL)e^{-\mathrm{i}2k_2L} \end{vmatrix}\tilde{\boldsymbol{A}}_a = \boldsymbol{M}\tilde{\boldsymbol{A}}_a \tag{3.4.7}$$

The following self-consistent condition should be satisfied:

$$\tilde{\boldsymbol{A}}_e = \tilde{\boldsymbol{A}}_a = \boldsymbol{I}\tilde{\boldsymbol{A}}_a = \boldsymbol{M}\tilde{\boldsymbol{A}}_a$$

or

$$(\boldsymbol{M} - \boldsymbol{I})\tilde{\boldsymbol{A}}_a = 0, \tag{3.4.8}$$

where $\boldsymbol{I}$ is an unit matrix. If $\tilde{\boldsymbol{A}}_a$ has nonzero solution, it requires the determinant $|\boldsymbol{M} - \boldsymbol{I}| = 0$, so we obtain

$$[r_1^2 \cosh(gL)e^{i2k_1L} - 1][(r_2^*)^2 \cosh(gL)e^{-i2k_2L} - 1] = r_1^2(r_2^*)^2 \sinh^2(gL)e^{-i2(k_2-k_1)L}. \tag{3.4.9}$$

Equation (3.4.9) is called the parametric oscillation threshold equation, i.e. the starting oscillation condition of the parametric oscillator.

There are two kinds of optical parameter oscillators: one parameter oscillator allows signal light (ω_s) and idler light (ω_i) together oscillation and output, which is called the Double Resonant Oscillator (DRO); another one only allows the signal light (ω_s) oscillation and output, which is called the Singly Resonant Oscillator (SRO). Below we will introduce their working principles respectively.

3.4.2 Double Resonant Parametric Oscillator

Figure 3.21 shows the schematic diagram of the double resonant oscillator, in which the three light beams are collinear. The nonlinear crystal is inserted into the optical cavity consisted of two spherical reflectors. The signal light and the idler light are two longitudinal modes of resonant cavity, and the resonant cavity for the

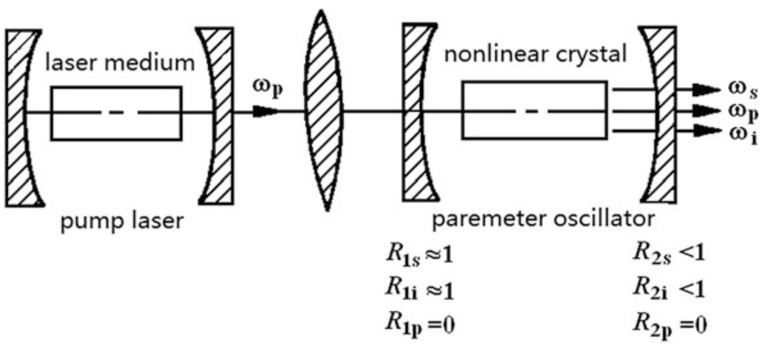


Fig. 3.21 Schematic diagram of collinear double resonant oscillator

pump light is transparent. So that the reflectivity of two spherical reflectors at front and back of the cavity for the signal light, the idler light and the pump light are $R_{1s} \approx 1$, $R_{1i} \approx 1$, $R_{1p} = 0$ and $R_{2s} < 1$, $R_{2i} < 1$, $R_{2p} = 0$, respectively.

Considering two cavity mirrors have the reflection loss and phase shift at the same time for the two lights at $\omega_1 = \omega_s$ and $\omega_2 = \omega_i$, we set

$$r_1^2 = R_1 e^{-i\phi_1}, \tag{3.4.10}$$

$$(r_2^*)^2 = R_2 e^{i\phi_2}, \tag{3.4.11}$$

where ϕ_1 and ϕ_2 are two cavity mirrors induced phase shifts. Substituting Eqs. (3.4.10) and (3.4.11) into Eq. (3.4.9), we obtain the threshold equation:

$$\begin{aligned} &[R_1 \cosh(gL) e^{\mathrm{i}(2k_1L - \phi_1)} - 1][R_2 \cosh(gL) e^{-\mathrm{i}(2k_2L-\phi_2)} - 1] \\ &\quad = R_1 R_2 \sinh^2(gL) e^{-\mathrm{i}[2(k_2-k_1)L - (\phi_2 - \phi_1)]}. \end{aligned} \tag{3.4.12}$$

When satisfying the phase condition:

$$\begin{aligned} 2k_1L - \phi_1 &= 2\,m\pi \\ 2k_2L - \phi_2 &= 2n\pi \end{aligned} \quad (m, n \,\text{is the integer}), \tag{3.4.13}$$

the exponents in two factors on left side of Eq. (3.4.12) are positive real numbers, in this case the corresponding gain is minimum, i.e., the threshold gain is $g = g_t$. The Eq. (3.4.13) denoted two light beams at frequency of ω_1 and ω_2 are laser longitudinal modes of the resonant cavity.

Using $\cosh^2 x - \sinh^2 x = 1$ and phase condition Eq. (3.4.13), the threshold Eq. (3.4.12) becomes

$$(R_1 + R_2)\cosh(g_tL) - R_1R_2 = 1. \tag{3.4.14}$$

When g_tL is smaller, After series expansion of $\cosh(g_tL)$ and approximately taking the front two items, we obtain

$$\cosh(g_tL) \approx 1 + \frac{(g_tL)^2}{2}, \tag{3.4.15}$$

Substituting Eq. (3.4.15) into Eq. (3.4.14), we obtain

$$(g_tL)^2 = \frac{2(1-R_1)(1-R_2)}{R_1+R_2}. \tag{3.4.16}$$

Assuming $R_1 \approx R_2 \approx 1$, $R_1 + R_2 \approx 2$, then

$$(g_t L)^2 = (1 - R_1)(1 - R_2). \tag{3.4.17}$$

Therefore, the threshold condition of double resonant parametric oscillator is

$$(g_t L)_{DRO} = \sqrt{(1 - R_1)(1 - R_2)}. \tag{3.4.18}$$

Substituting Eq. (3.4.18) into Eq. (3.3.48) for the difference frequency conversion efficiency, then we obtain the pump light intensity threshold of double resonant parametric oscillator:

$$(I_{3t})_{DRO} = \frac{\varepsilon_0 n_1 n_2 n_3 c^3}{8\omega_1 \omega_2 d^2 L^2}(1 - R_1)(1 - R_2). \tag{3.4.19}$$

As an example, the $LiNbO_3$ crystal based double resonant parametric oscillator, to take the single pass loss of cavity is 2%, $\lambda_1 = \lambda_2 = 1\,\mu\text{m}$, $(1 - R_1) = (1 - R_2) = 2 \times 10^{-2}$, $d = 5 \times 10^{-12}\,\text{m/V}$, $n_1 \approx n_2 \approx n_3 \approx 2$, the estimated oscillation threshold intensity is $I_{3t} = 1.2 \times 10^3\,\text{W/cm}^2$. It is equivalently the output intensity of a common CW laser.

Although the pump light intensity threshold of the double resonant parametric oscillator is lower, its requirement for the stability of the resonant cavity is very high, and the length of cavity is easy effected by temperature variation and vibratory.

3.4.3 Singly Resonant Parametric Oscillator

The use of non-collinear phase matching technology to separate the directions of three light beams is shown in Fig. 3.22. It only allows the wave vector $\boldsymbol{k}_s$ of signal light at ω_s along the cavity axis direction, and making the signal light resonance with the resonant cavity. But the $\boldsymbol{k}_p$ of pump light and $\boldsymbol{k}_i$ of idler light are not along the cavity axis direction. The wave vectors of three light beams must satisfy following phase matching condition:

$$\boldsymbol{k}_p = \boldsymbol{k}_s + \boldsymbol{k}_i. \tag{3.4.20}$$

Now we start from the threshold Eq. (3.4.9) to deduce the threshold condition of the singly resonant parametric oscillator. For the singly resonant parametric oscillator, $r_2^* = 0$, Eq. (3.4.9) is simplified as

$$r_1^2 \cosh(gL) e^{i2k_1 L} = 1. \tag{3.4.21}$$

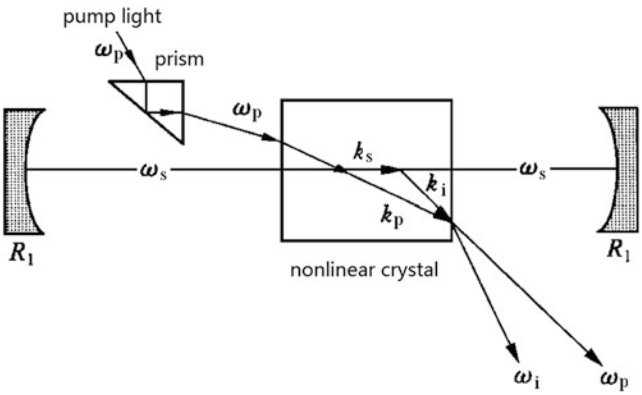


Fig. 3.22 Schematic diagram of non-collinear phase match in the singly resonant parametric oscillator

Setting $r_1^2 = R_1 e^{-i\phi_1}$, and substituting it into Eq. (3.4.21), then we obtain the phase condition:

$$2k_1 L - \phi_1 = 2\,m\pi, \tag{3.4.22}$$

Equation (3.4.21) then is

$$R_1 \cosh(g_t L) = 1. \tag{3.4.23}$$

Because $g_t L$ is very small, Eq. (3.4.23) can approximately simplify to

$$R_1\left(1 + \frac{g_t^2 L^2}{2}\right) = 1. \tag{3.4.24}$$

Taking $R_1 \approx 1$, then

$$(g_t L)_{SRO} = \sqrt{2(1 - R_1)}. \tag{3.4.25}$$

We substitute Eq. (3.4.24) into Eq. (3.3.48) for the difference frequency conversion efficiency, then obtain the pump light intensity threshold of singly resonant parametric oscillator:

$$(I_{3t})_{SRO} = \frac{\varepsilon_0 n_1 n_2 n_3 c^3}{8\omega_1 \omega_2 d^2 L^2} 2(1 - R_1). \tag{3.4.26}$$

To compare the pump light threshold intensity formula (3.4.26) for the singly resonant parametric oscillator and the formula (3.4.19) for the double resonant parametric oscillator, we obtain

$$\frac{(I_{3t})_{SRO}}{(I_{3t})_{DRO}} = \frac{[(g_t L)_{SRO}]^2}{[(g_t L)_{DRO}]^2} = \frac{2}{1 - R_2}. \tag{3.4.27}$$

where $1 - R_2$ can be regard as the cavity mirror loss of idler light at frequency ω_2. If the loss is 2 %, then the threshold of singly resonant parametric oscillator is higher than the threshold of double resonant parametric oscillator about 100 times. Even though the starting threshold of singly resonant parametric oscillator is higher, however its requirement in respect of resonant cavity stability is much lower.

Optical parametric oscillation (OPO) can be used to obtain wavelength tunable laser in a wide wavelength region. It not only can obtain the visible and infrared steady-state continuous wave, but also can obtain the picosecond or femtosecond ultrashort pulse laser, it has extensive application in the optical spectrum technology. There are many kinds of nonlinear crystal used for OPO, more good crystals mainly include KTP, BBO and LBO, they have not only larger second-order nonlinear coefficient and much higher optical damage threshold, but also wide transparent wavelength range, for example, BBO can reach 2500–190 nm; LBO can reach 3000–160 nm.

Review Questions of Chapter 3

1. Please deduce slowly-varying-amplitude approximation wave equation for describing the propagation of the monochromic plane wave in anisotropic medium and the three-wave mixing equations for describing the second-order nonlinear optics processes.
2. In fundamental wave small signal approximation condition, from three-wave mixing equations, please find out the frequency doubling wave intensity conversion efficiency formula. In order to enhance the frequency doubling efficiency, what measures you can adopt?
3. In high fundamental wave consumption condition, from three-wave mixing equations, please find out the law of variation of the frequency doubling field amplitude and fundamental frequency field amplitude with the propagation distance, and find out frequency doubling wave intensity conversion efficiency formula.
4. When the frequency doubling light and fundamental frequency light collinearly propagate, what is the refractive-index phase matching condition of frequency doubling crystal? Please discuss the phase matching conditions of negative uniaxial crystal and positive uniaxial crystal in the first- class phase matching condition and the second-class phase matching condition.
5. From three-wave mixing equations to deduce the coupling equations between the sum frequency light field and the signal light field, and the intensity conversion efficiency formula.

6. From three-wave mixing equations to deduce the coupling equations between the difference frequency light field and the signal light field, and the intensity conversion efficiency formula.
7. What is optical parameter amplification? Please deduce the magnification formula of parameter amplification. The parameter amplification process has what different physical meaning in comparison with the sum frequency and difference frequency processes?
8. What is optical parametric oscillator? Please discuss the working principles of the double resonant parametric oscillator and the singly resonant parametric oscillator, please deduce the pump light intensity threshold formula of these two parameter oscillators, and point out the advantages and disadvantages of both parameter oscillators.

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