CHAPTER 2

NONLINEAR OPTICS THEORY

This chapter presents the fundamental theory of nonlinear optical interactions from harmonic generation processes to parametric oscillations. The main concepts for the physics of nonlinear optical processes, namely three-wave mixing, phase matching conditions and calculations of phase matching angles are given. The aim of this chapter is to introduce and outline the important aspects of frequency conversion and parametric interactions in nonlinear materials with KD*P and BBO as examples .

2.1 Three-Wave Interactions

In nonlinear optics(NLO), the relationship between the polarization and electric field can be expressed in the form

$$P = \varepsilon_0\chi^{(1)}E + \varepsilon_0\chi^{(2)}E + \varepsilon_0\chi^{(3)}E + \ldots\ldots \qquad (2.1)$$

where $\chi^{(2)}$ is the second order nonlinear polarization term and require two input fields. In three-wave mixing (3WM) these two input fields at frequencies ω_1 and ω_2 are mixed in the second order nonlinear medium and generate a third (ω_3) at the difference frequency or at the sum frequency. These processes are termed DFG and SFG, respectively. For this to occur the phase matching condition, which will be discussed in the next section, must be satisfied.

In the two cases, propagating monochromatic waves have to satisfy the energy relation

$$\omega_3 = \omega_1 + \omega_2 \qquad \text{SFG} \qquad (2.2a)$$

$$\omega_3 = \omega_1 - \omega_2 \qquad \text{DFG} \qquad (2.2b)$$

Second harmonic generation (SHG) is a special case of SFG whereby $\omega_1 = \omega_2$. Equation (2.2a) becomes

$$\omega_3 = 2\omega_1 \qquad (2.3)$$

Parametric oscillation is a special case of DFG which involves the generation of two light waves with frequencies ω_1 and ω_2 in the intense radiation of a pump wave with frequency ω_3.

2.2 Phase matching

Propagation of light through nonlinear crystals can produce observable effects when the phase matching conditions are satisfied. The wave vector expression for phase matching is given as

$$\Delta k = k_3 - k_2 - k_1 = 0 \qquad (2.4)$$

where k_i are the wave vectors corresponding to the waves with frequencies ω_i (i = 1,2,3)

For general 3WM the phase matching condition requires

$$\Delta k = 0 \qquad (2.5)$$

Therefore

$$\omega_3 n_3(\omega_3) = \omega_2 n_2(\omega_2) + \omega_1 n_1(\omega_1) \qquad (2.6)$$

or

$$n_3(\lambda_3)/\lambda_3 = n_2(\lambda_2)/\lambda_2 + n_1(\lambda_1)/\lambda_1 \qquad (2.7)$$

where the quantities n_i and λ_i are the refractive index and wavelength corresponding to the frequency ω_i , respectively.

The physical sense of phase matching conditions (2.4, 5) is a space resonance of the interacting light waves. Condition (2.5) is fulfilled only in anisotropic crystals with interaction of differently polarized waves. [1]

2.3 Optics of Uniaxial Crystals

We give attention to uniaxial crystals because the crystals involved in this work are uniaxial. In these crystals, a special direction exists called the optic axis or z axis. The plane containing the z axis and the wave vector k of the light wave is termed the principal plane. The wave whose polarization (i.e the directions of the vector E oscillations) is normal to the principal plane is called the ordinary wave or o-wave. The wave polarized parallel to the plane is the extraordinary wave or e-wave. The refractive index of the o-wave does not depend on the propagation direction, whereas for the e-wave it does. Their refractive indices are termed n_o and n_e respectively. For negative uniaxial crystals $n_o > n_e$ and $n_o < n_e$ for positive crystals. Type I phase matching is realized in mixing of waves having the same polarization. In negative crystals this is called *ooe* interaction and *eeo* interaction in positive crystals. Type II phase matching occurs when orthogonally polarized waves are mixed resulting in *oee, eoe* interaction in negative crystals and *oeo, eoo* interaction in positive crystals. This interactions also refers to parametric oscillations.

2.4 Optical Parametric Process

2.4.1 Parametric Oscillator Tuning

In the optical parametric process, a pump photon of frequency, ω_3 propagating in a nonlinear optical crystal, spontaneously or by stimulated emission, breaks down into smaller photons of frequencies, ω_1 and ω_2 . Conservation of energy predicts (as in equation 2.2a) , $\omega_3 = \omega_1 + \omega_2$.

The emitted photons cannot be uniquely determined on the basis of this condition alone. For a given ω_3 , there exists a continuous range of ω_1 and ω_2 - the source of tunability of OPOs. The specific pair of frequencies, ω_1 and ω_2 , that will result in a given condition is dictated by the phase matching condition (equation 2.4), that must also be satisfied.

In a normally isotropic medium, the material dispersion is such that the momentum or magnitude of the k-vector of the pump photon is always too large to satisfy the phase matching condition. However, this is compensated in anisotropic medium by the birefringence. The index of refraction, and hence, the vector magnitude of an extraordinary wave varies with the direction of propagation.

Fig. 2.1 shows the parametric tuning curve in 532nm pumped optical parametric oscillator with calculated values corresponding to wavelength range between 0.8μm - 2.5μm. A wider spectral coverage can be obtained with 266nm pumped BBO. The theoretical curve is shown in Fig. 2.2.

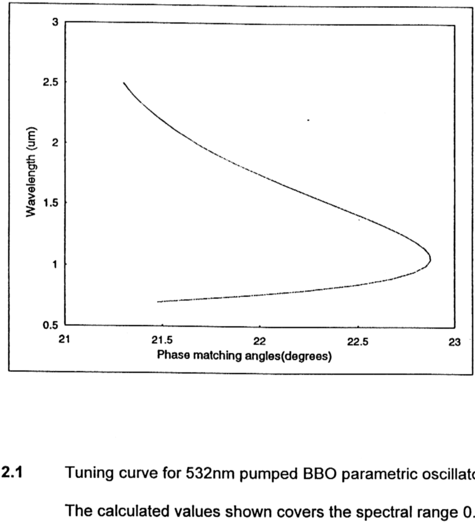


Fig. 2.1 Tuning curve for 532nm pumped BBO parametric oscillator. The calculated values shown covers the spectral range 0.7μm to 2.5μm

$$G = \cosh^2 (\Gamma L) \qquad (2.8)$$

where $\Gamma L = 6 \times 10^{-5}\, d_{eff} (P_p)^{1/2}\, L / w_0$

and

d_{eff} = effective nonlinear coefficient

L = crystal length

P_p = pump power

w_0 = beam radius

2.4.3 Threshold

In order to achieve oscillation some form of feedback is required. In an OPO this is accomplished by enclosing the nonlinear crystal in an optical resonator. Two types of oscillators are possible. In one, the feedback is provided provided for both the signal and idler, called the doubly resonant oscillator or DRO . In another, feedback is provided for either the signal or idler but not both, referred to as a singly resonant oscillator. In this work we are using the latter . In either type of oscillator, there is a threshold condition defined as the condition that the round-trip gain experienced by the resonated wave or waves equal the round-trip loss or losses. Below the threshold there is a small amount of spontaneous parametric emission but above threshold the OPO experience a net round-trip gain and useful output can be obtained. In the ways mentioned above, the OPO is similar to the laser.

2.4.4 Pump Characteristics

In order that the phase matching condition be satisfied it is necessary that the pump frequency have a bandwidth less than the sum of the bandwidth of the signal and idler cavities to be effective in pumping the oscillator. If the bandwidth of the pump is broader, then only a fraction of the pump is effective in pumping the OPO. For a SRO where only the signal or idler is resonated the restrictions on the pump spectrum are expected to be less severe since the nonresonant wave may itself become spectrally broad and adjust phase to optimize the parametric interaction [9].

2.5 Calculation of Phase Matching Angles

Prior to calculating the phase matching angles in nonlinear 3WM it is therefore important to determine the type of interaction. Only then the dispersion relations given by the Sellmeier equations be used to find the values of refractive indices at the particular wavelengths. The following equations [1, 8] have been found to be most practical in calculating phase matching angles θ_m for three-wave interactions in negative and positive crystals.

Negative uniaxial crystals

Type I (*ooe*)

$$\tan^2\theta_m = (1-U)/(W-1)$$

Type II (*eoe*)

$$\tan^2\theta_m = (1-U)/(W-R)$$

(*oee*)

$$\tan^2\theta_m = (1-U)/(W-Q)$$

Positive uniaxial crystals

Type I (*eeo*)

$$\tan^2\theta_m = (1-U)/(U-S)$$

Type II (*oeo*)

$$\tan^2\theta_m = (1-V)/(V-Y)$$

(*eoo*)

$$\tan^2\theta_m = (1-T)/(T-Z)$$

Notation:

$U = (A+B)^2/C^2$ $W = (A+B)^2/F^2$

$R = (A+B)^2/(D+B)^2$ $Q = (A+B)^2/(A+E)^2$

$S = (A+B)^2/ (D+E)^2$ $V = B^2/(C-A)^2$

$Y = B^2/E^2$ $\qquad$ $T = A^2/(C-B)^2$

$Z = A^2/D^2$

$A = n_{o1}/\lambda_1$; $B = n_{o2}/\lambda_2$; $C = n_{o3}/\lambda_3$

$D = n_{e1}/\lambda_1$; $E = n_{e2}/\lambda_2$; $F = n_{e3}/\lambda_3$

The formulas were used to compute values of phase matching angles in second harmonic generation (SHG) and fourth harmonic generation (FOHG) as well as optical parametric oscillation (OPO) in BBO. They were found to be in agreement with work reported earlier[2,3,4]. Sellmeier equations for BBO are given in a number of publications [3,5,6,7,8]. The dispersion relations [2] below gives the best fit for our calculations (Appendix 2A) .

$$n_o^2 = 2.7405 + 0.0184/(\lambda^2 - 0.0179) - 0.0155.\lambda^2$$

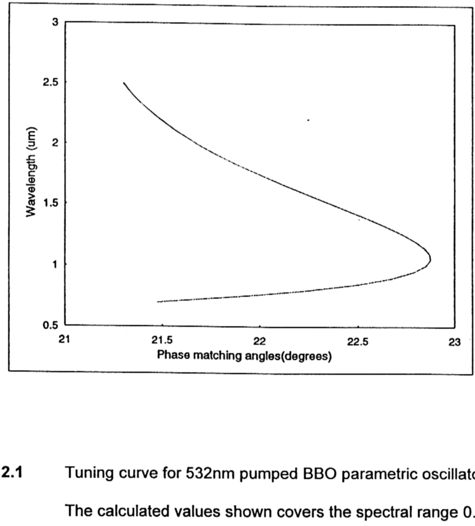
$$n_e^2 = 2.3730 + 0.0128/(\lambda^2 - 0.0156) - 0.0044.\lambda^2$$

where λ are given in μm. Table 2.1 gives the calculated values of phase matching angles for Type I SHG and Type I FOHG in BBO. The values are compared with the work before this.

Table 2.1 Calculated phase matching angles for BBO SHG and FOHG

Process	θ_m(Kato)[5]	θ_m(Chen et.al)[3]	θ_m(Eimerl et.al)[6]	θ_m(this work)
Type I SHG	22.8^0	19.8^0	22.9^0	22.9^0
Type I FOHG	47.5^0	49.5^0	47.5^0	47.4^0

Fig. 2.1 shows the parametric tuning curve in 532nm pumped optical parametric oscillator with calculated values corresponding to wavelength range between 0.8μm - 2.5μm. A wider spectral coverage can be obtained with 266nm pumped BBO. The theoretical curve is shown in Fig. 2.2.



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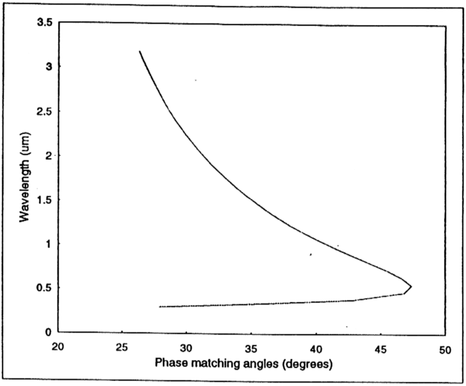

Fig. 2.2 Parametric tuning curve for 266nm pumped BBO OPO

2.6 Discussion

The basic concepts of three-wave interaction in nonlinear crystals have been briefly reviewed. The relevant theories led to a simple and accurate determination of phase matching angles in frequency conversion and parametric oscillation. Although the calculations reported are specifically for the negative uniaxial crystal BBO, they are, however easily modified for other anisotropic materials. SHG, FOHG and OPO are merely extensions of SFG and DFG. The equations for the calculation of phase matching angles given are in their general forms for negative and positive uniaxial crystals. In obtaining the correct angular values, one only requires to know the dispersion relations for the crystal and the type of interaction involved. It then becomes a simple matter of numerical substitution in the chosen equations.

Phase matching of nonlinear crystals has always been a primary concern for the experimentalist. Calculations of phase matching angles with proper orientation of the crystal often seem a formidable task. However, by beginning with the fundamental understanding of the optic behind nonlinear three-wave interaction , it would be easier to experiment with nonlinear crystals, either for SFG, SHG or OPO. Experimental configurations of SHG and OPOs are not different from a laser system but one needs a very clear understanding of the way the nonlinear crystals are cut, the principles of ordinary and extraordinary waves, propagation of these waves in the nonlinear medium and their interaction that would produce useful results.

APPENDIX 2A

Computation of Phase Matching Angles in BBO

Type I SHG

$\lambda1 := 1.064 \quad \lambda2 := 1.064 \quad \lambda3 := 0.532 \quad \lambda := \lambda2$

$$no(\lambda) := \sqrt{2.7405 + \frac{0.0184}{(\lambda^2 - 0.0179)} - 0.0155 \cdot \lambda^2}$$

$$ne(\lambda) := \sqrt{2.3730 + \frac{0.0128}{(\lambda^2 - 0.0156)} - 0.0044 \cdot \lambda^2}$$

$\lambda3 = 0.532$ $\qquad$ $\lambda2 = 1.064$

$no(\lambda3) = 1.675 \qquad no(\lambda1) = 1.655 \qquad no(\lambda2) = 1.655$

$ne(\lambda3) = 1.556 \qquad ne(\lambda1) = 1.543 \qquad ne(\lambda2) = 1.543$

$$A := \frac{no(\lambda1)}{\lambda1} \qquad B := \frac{no(\lambda2)}{\lambda2} \qquad C := \frac{no(\lambda3)}{\lambda3} \quad F := \frac{ne(\lambda3)}{\lambda3}$$

$A = 1.556 \quad B = 1.556 \qquad C = 3.148 \qquad F = 2.924$

$$U := \frac{(A + B)^2}{C^2} \qquad W := \frac{(A + B)^2}{F^2}$$

$U = 0.976 \qquad W = 1.132$

$$\alpha := \operatorname{atan}\left(\sqrt{\frac{1 - U}{W - 1}}\right) \qquad \theta(\alpha) := \frac{\alpha}{3.142} \cdot 180$$

$\alpha = 0.399 \qquad \lambda1 = 1.064 \qquad \theta(\alpha) = 22.877$

Type I FOHG

$\lambda 1 := 0.532 \quad \lambda 2 := 0.532 \quad \lambda 3 := 0.266 \quad \lambda := \lambda 2$

$$no(\lambda) := \sqrt{2.7405 + \frac{0.0184}{(\lambda^2 - 0.0179)} - 0.0155\cdot\lambda^2}$$

$$ne(\lambda) := \sqrt{2.3730 + \frac{0.0128}{(\lambda^2 - 0.0156)} - 0.0044\cdot\lambda^2}$$

$\lambda 3 = 0.266$ $\quad no(\lambda 1) = 1.675$

$no(\lambda 3) = 1.757$ $\quad ne(\lambda 1) = 1.556$ $\quad \lambda 2 = 0.532$

$ne(\lambda 3) = 1.614$ $\quad no(\lambda 2) = 1.675$ $\quad ne(\lambda 2) = 1.556$

$A := \frac{no(\lambda 1)}{\lambda 1} \quad B := \frac{no(\lambda 2)}{\lambda 2} \quad C := \frac{no(\lambda 3)}{\lambda 3} \quad F := \frac{ne(\lambda 3)}{\lambda 3}$

$A = 3.148 \quad B = 3.148 \quad C = 6.606 \quad F = 6.067$

$U := \frac{(A + B)^2}{C^2} \quad W := \frac{(A + B)^2}{F^2}$

$U = 0.909 \quad W = 1.077$

$\alpha := \operatorname{atan}\left(\sqrt{\frac{1 - U}{W - 1}}\right) \quad \theta(\alpha) := \frac{\alpha}{3.142}\cdot 180$

$\alpha = 0.828 \quad \lambda 1 = 0.532 \quad \theta(\alpha) = 47.423$

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