

การศึกษาเชิงทฤษฎีของการเกิดแสงเช็คกันฮาร์โมนิกตามแนวหักเหในผลึก  
แอมโมเนียมไดไฮโดรเจนฟอสเฟต

นายทรงฤทธิ์ คำยอด

วิทยานิพนธ์นี้เป็นส่วนหนึ่งของการศึกษาตามหลักสูตรปริญญาวิทยาศาสตรมหาบัณฑิต

สาขาวิชาเทคโนโลยีเลเซอร์และโฟตอนิกส์

มหาวิทยาลัยเทคโนโลยีสุรนารี

ปีการศึกษา 2543

**ISBN 974 739 561**

**THEORETICAL STUDY OF TRANSMITTED  
SECOND HARMONIC GENERATION IN  
AMMONIUM DIHYDROGEN PHOSPHATE  
(ADP) CRYSTAL**

**Mr. Songrit Kamyod**

**A Thesis Submitted In Partial Fulfillment of the Requirements  
for the Degree of Master of Science in Laser Technology and  
Photonics**

**Suranaree University of Technology**

**Academic Year 2000**

**ISBN 974-7359-56-1**

**Thesis Title**

**Theoretical study of transmitted second harmonic generation in  
Ammonium Dihydrogen Phosphate (ADP) crystal**

Suranaree University of Technology Council has approved this thesis submitted in partial fulfillment of the requirements for a Master's Degree

Thesis Examining Committee

.....

(Prof. Dr. Vutthi Bhanthumnavin)  
Chairman

.....

(Prof. Dr. Vutthi Bhanthumnavin)  
Thesis Advisor

.....

(Asst. Prof. Dr. Joewono Widjaja)  
Member

.....

(Asst. Prof. Dr. Arjuna Chaiyasena)  
Member

.....

(Assoc. Prof. Dr. Kasem Prabripataloong)  
Vice Rector for Academic Affairs

.....

(Assoc. Prof. Dr. Tassanee Sukosol)  
Dean / Institute of Science

นายทรงฤทธิ์ คำยอด: การศึกษาเชิงทฤษฎีของการเกิดแสงเช็คกันฮาร์โมนิคตามแนวหัก  
เหในผลึกแอมโมเนียมไดไฮโดรเจนฟอสเฟต

(THEORETICAL STUDY OF TRANSMITTED SECOND HARMONIC  
GENERATION IN AMMONIUM DIHYDROGEN PHOSPHATE (ADP) CRYSTAL)

อ.ที่ปรึกษา : ศ. ดร. วุฑฒิ พันธุมนาวิน, 90 หน้า. ISBN 974-7359-56-1

การศึกษาเชิงทฤษฎีของการเกิดแสงเช็คกันฮาร์โมนิคในแนวส่งผ่านโดยใช้ทฤษฎีของ  
บลอมเบอร์เกนและเพอร์ชาน ในผลึกแอมโมเนียมไดไฮโดรเจนฟอสเฟตที่มีการวางตัวของแกน  
ผลึกที่แตกต่างกันโดยให้ผลึกจมอยู่ในของเหลว วัน-โบรโมเนพธาไลน์ ในการศึกษาที่ลำแสง  
เลเซอร์ช่วงสั้นมากโดยมีโพลาไรเซชัน  $[1\bar{1}0]$  ที่ความยาวคลื่น 900 นาโนเมตรถูกใช้เป็นตัวกระตุ้น  
ให้เกิดแสงเช็คกันฮาร์โมนิคในแนวส่งผ่าน ค่าของความเข้มแสงเช็คกันฮาร์โมนิคในแนวส่งผ่านได้  
ถูกคำนวณเป็นฟังก์ชันกับค่ามุมตกกระทบ ( $\theta'$ ) มีการใช้เทคนิคของลิเนียร์ และ นอนโคลิเนียร์ เฟส  
แมชชิงในการทำให้เกิดค่าความเข้มแสงเช็คกันฮาร์โมนิคในแนวส่งผ่าน นอกจากนี้ได้แสดงการเกิด  
ความเข้มแสงเช็คกันฮาร์โมนิคในแนวส่งผ่านเท่ากับศูนย์ที่มุมของการตกกระทบแบบตั้งฉาก ผล  
ของการศึกษาเชิงทฤษฎีพบว่าสอดคล้องเป็นอย่างดีกับผลวิจัยก่อนหน้านี้ของการทดลองการเกิด  
แสงเช็คกันฮาร์โมนิคในแนวส่งผ่านในผลึกโพแทสเซียมไดไฮโดรเจนฟอสเฟต (พันธุมนาวิน และ  
ลี, 1990; 1994)

สาขาวิชาเทคโนโลยีเลเซอร์และ	ลายมือชื่อนักศึกษา.....
โฟตอนิกส์	ลายมือชื่ออาจารย์ที่ปรึกษา.....
ปีการศึกษา 2543	ลายมือชื่ออาจารย์ที่ปรึกษาร่วม.....
	ลายมือชื่ออาจารย์ที่ปรึกษาร่วม.....

Mr. SONGRIT KAMYOD: THEORETICAL STUDY OF TRANSMITTED SECOND HARMONIC GENERATION IN AMMONIUM DIHYDROGEN PHOSPHATE (ADP) CRYSTAL.

THESIS ADVISOR: PROF. VUTTHI BHANTHUMNAVIN. Ph. D.

90 pp. ISBN 974-7359-56-1

The theoretical investigation, based on the Bloembergen and Pershan theory, of transmitted second harmonic generation in Ammonium Dihydrogen Phosphate (ADP) crystal immersed in the optically denser fluid 1-bromonaphthalene was performed at different orientations of crystal. The transmitted second harmonic intensity has been theoretical calculated as a function of the incident angle ( $\theta^i$ ) of the ultrashort pulse laser which has the polarization in  $[1\bar{1}0]$  direction with respect to the ADP crystallographic axes at wavelength equal 900 nm. The colinear and noncolinear phase matching of second harmonic generation in transmission and also null transmitted second harmonic intensity at normal incidence are shown. The theoretical results agree well with the previous experimental work of the transmitted second harmonic generation in KDP crystal (Bhanthumnavin and Lee, 1990; 1994)

สาขาวิชาเทคโนโลยีเลเซอร์และ ลายมือชื่อนักศึกษา.....

โฟตอนิกส์ ลายมือชื่ออาจารย์ที่ปรึกษา.....

ปีการศึกษา 2543 ลายมือชื่ออาจารย์ที่ปรึกษาร่วม.....

ลายมือชื่ออาจารย์ที่ปรึกษาร่วม.....

# Acknowledgments

First, I am very grateful to my supervisor, Professor Dr. Vutthi Bhanthumnavin, for his help, long-term guidance, patience and many discussions of the thesis. His constant encouragement always kept me pursuing the course of research until completion. Without his kind support, this thesis would not have been possible.

Second, I am deeply indebted to Assistant Professors Dr. Joewono Widjaja and Dr. Arjuna Chaiyasena for their patience in reading several chapters of this thesis and making suggestions for correction and improvement.

I am gratefully indebted to all who taught me graduate courses at the School of Laser Technology and Photonics of Suranaree University of Technology (SUT) and especially Assistant professor Dr. Eckart Schulz for his advice.

I am also thankful to several graduate students at SUT: Mr. Jessada Tanthanuch and Mr. Sampart Cheedket for computer programming, Miss Ubon Suripon for her kindness and some of her documents that were useful for writing the thesis, and Miss Jarin Osaklung for her help on many occasions during this study.

Finally, I am grateful to my parents and relations for encourage and providing all supports throughout the course of study.

Mr. Songrit Kamyod

# Contents

	Page
<b>Abstract (Thai)</b> .....	I
<b>Abstract (English)</b> .....	II
<b>Acknowledgments</b> .....	III
<b>Contents</b> .....	IV
<b>Lists of Figures</b> .....	VII
<b>Lists of Tables</b> .....	X
<b>Lists of Symbols</b> .....	XI
<b>Chapter I. Introduction</b> .....	1
1.1 Literature Review.....	1
1.2 Purpose.....	4
1.3 Hypothesis.....	5
<b>Chapter II. Theory</b> .....	6
2.1 Introduction.....	6
2.2 Theory of Second Harmonic Generation.....	7
2.2.1 Nonlinear Polarization, $\bar{P}^{NLS}$ .....	7
2.2.2 Maxwell Equations of Nonlinear Medium.....	8
2.2.3 Nonlinear Polarization and Second Harmonic Generation.....	14
2.2.4 Theoretical Calculation for Transmitted Second Harmonic Intensities.....	19
2.2.5 The Criteria of Optimization of Second Harmonic Generation...	25

## Contents (Continued)

	<b>Page</b>
<b>Chapter III. Procedure</b> .....	33
3.1 Introduction.....	33
3.2 Ammonium Dihydrogen Phosphate, ADP Crystal.....	33
3.3 Fluid 1-Bromonaphthalene.....	35
3.4 Computer Program.....	36
<b>Chapter IV. Results and Discussion</b> .....	41
4.1 Introduction.....	41
4.2 Minimum Transmitted Second Harmonic Generation.....	41
4.2.1 Null Transmitted Second Harmonic Generation.....	42
4.2.2 Minimum Transmitted Second Harmonic Generation when $\bar{P}^{NLS}(2\omega)$ Makes Phase Matching Angle with the Crystal Surface..	44
4.3 Transmitted Second Harmonic Generation under Phase Matching Condition.....	48
4.3.1 The Maximum Transmitted Second Harmonic Generation under Colinear Phase Matching Condition when $P^{NLS}(2\omega)$ Lies Along Face Normal.....	48
4.3.2 The Maximum Transmitted Second Harmonic Generation under Colinear Phase Matching Condition when $P^{NLS}(2\omega)$ Makes Phase Matching Angle with the Crystal Surface.....	50

## Contents (Continued)

	Page
4.3.3 The Maximum Transmitted Second Harmonic Generation under Colinear Phase Matching Condition, Two Beam Spatial Mixing (TBSM).....	53
<b>Chapter V. Conclusion</b> .....	56
<b>References</b> .....	59
<b>Appendixes</b> .....	63
<b>Appendix A. The Calculation of Refractive Indices of Ammonium Dihydrogen Phosphate and Liquid 1-Bromonaphthalene at Wavelength Equal 900 nm and 450 nm</b> .....	
	64
<b>Appendix B. Computer Programming (C++)</b> .....	70
<b>B1. Colinear Incidence when <math>\vec{P}^{NLS}(2\omega)</math> Lies Along Face Normal..</b>	71
<b>B2. Colinear Incidence when <math>\vec{P}^{NLS}(2\omega)</math> Makes Phase Matching Angle with the Crystal Surface</b> .....	77
<b>B3. Noncolinear Incidence when <math>\vec{P}^{NLS}(2\omega)</math> Parallel to the Crystal Surface</b> .....	84
<b>Biography</b> .....	90

# Lists of Figures

Figure	Page
2.1 The incident, reflected and refracted rays near the boundary between vacuum and ADP crystal at the fundamental and second harmonic frequencies .....	10
2.2 Two incident rays at frequencies $\omega_1$ and $\omega_2$ caused the reflected, homogeneous and inhomogeneous transmitted wave at the sum frequency $\omega_3 = \omega_1 + \omega_2$ eliminate from the boundary between the linear and nonlinear medium .....	11
2.3 The noncolinear incident case or Two Beam Spatial Mixing [for $k_1^i(\omega) \neq k_2^i(\omega)$ ] .....	14
2.4 The second harmonic and electric and magnetic field when nonlinear polarization $\vec{P}^{NLS}(2\omega)$ perpendicular to the plane of incidence .....	15
2.5 The second harmonic electric and magnetic field when nonlinear polarization $\vec{P}^{NLS}(2\omega)$ parallel to the plane of incidence .....	17
2.6 The geometry of wave vector of the incident wave, the refracted waves, and the reflected wave at the interface between liquid 1-bromanaphthalene and ADP crystal .....	21
2.7 The construction for finding the value of index of refraction when vector $s$ is the direction of propagation of wave in crystal (Yariv, 1989) .....	27

## Lists of Figures (Continued)

<b>Figure</b>	<b>Page</b>
2.8 The condition for the occurrence of colinear phase matching .....	29
2.9 The occurrence of noncolinear phase matching .....	30
2.10 The crystallographic orientation of the theoretical study, which caused noncolinear phase matching .....	32
3.1 The flowchart of C++ program for the calculation of transmitted second harmonic intensity .....	38
3.2 The computing process of the C++ program for calculation of transmitted second harmonic intensity .....	39
4.1 The null transmitted second harmonic intensity (SHI) at the angle of incidence ( $\theta^i$ ) equal $0^\circ$ . The nonlinear polarization $\vec{P}^{NLS}(2\omega)$ lies along face normal .....	43
4.2 The minimum transmitted second harmonic intensity (SHI) at the angle of incidence ( $\theta^i$ ) equal $42.88^\circ$ . The nonlinear polarization $\vec{P}^{NLS}(2\omega)$ makes phase matching angle with the crystal surface .....	45
4.3 The transmitted second harmonic intensity (SHI) as a function of the angle of incidence. The nonlinear polarization $\vec{P}^{NLS}(2\omega)$ lies along face normal.	49
4.4 The transmitted second harmonic intensity (SHI) as a function of the angle of incidence. The nonlinear polarization $\vec{P}^{NLS}(2\omega)$ makes phase matching angle with the crystal surface .....	52

## Lists of Figures (Continued)

Figure	Page
4.5 The transmitted second harmonic intensity (SHI) at the noncolinear phase matching condition. The phase matching angle ( $\theta^i$ ) equal $9.63^\circ$ . The nonlinear polarization $\vec{P}^{NLS}(2\omega)$ parallel to the crystal surface .....	54

## Lists of Tables

Table	Page
5.1 The summarized results of the transmitted second harmonic generation at different crystallographic orientation .....	57
A1 The value of the refractive index of ADP crystal and liquid 1-bromonaphthalene at wavelength equal 450 nm and 900 nm. ....	65
A2 The value of constant $A, B, C, D$ , and $E$ with respect to air. ....	66
A3 The four set of the value of $n_{liq}$ for calculating the new value of $n_{liq}$ at wavelength equals 900 nm. ....	68
A4 The four set of the value of $n_{liq}$ for calculating the new value of $n_{liq}$ at wavelength equals 450 nm. ....	69

## Lists of Symbols

$\bar{P}^L$  = linear polarization

$\bar{P}^{NLS}$  = nonlinear polarization

$n_o$  = ordinary ray index

$n_e$  = extraordinary ray index

$n_e^{(2\omega)}$  = refractive index of transmitted homogeneous wave vector

$n_o^{(\omega)}$  = refractive index of transmitted source wave vector

$l_c$  = the crystal length

nm = nanometer

$\bar{P}^{total}$  = total polarization

$\chi^L$  = linear susceptibility

$\chi^{NL}$  = nonlinear susceptibility

$\omega$  = frequency

$\epsilon$  = permittivity of crystal

$\mu$  = permeability of crystal

$\bar{D}$  = electric flux density

$\bar{J}$  = electric current density

$\rho$  = electric charge density

$\bar{B}$  = magnetic flux density

## Lists of Symbols (Continued)

$\vec{H}$	= magnetic field
$\nabla^2$	= laplacian operator
$c$	= speed of light which is equal to $3 \times 10^8 \text{ m/s}$
$t$	= time
$\vec{E}$	= electric field
$\vec{E}_1^T$	= transmitted electric field in crystal
$\vec{k}^i$	= the incident wave vector
$\vec{k}^T$	= the transmitted homogeneous wave vector
$\vec{k}^S$	= the transmitted inhomogeneous source wave vector which equal to $2\vec{k}^T$
$\vec{k}^R$	= the reflected harmonic wave vector
$\vec{E}_2^R$	= electric field of the reflected second harmonic light
$\vec{E}_2^T$	= electric field of the transmitted second harmonic light
$\vec{H}_2^R$	= magnetic field of the reflected second harmonic light
$H_2^T$	= magnetic field of the transmitted second harmonic light
$\mathcal{E}_2^T$	= amplitude of the electric field of the transmitted second harmonic light
$\mathcal{E}_2^R$	= amplitude of the electric field of the reflected second harmonic light
$\vec{P}_\perp^{NLS}$	= nonlinear polarization perpendicular to the plane of incidence
$\vec{P}_\parallel^{NLS}$	= nonlinear polarization parallel to the plane of incidence

## Lists of Symbols (Continued)

- $E_{//}^R$  = reflected harmonic field
- $\mathcal{E}_{//}^T$  = amplitude of the transmitted second harmonic wave
- $F_L^T$  = the linear fresnel factor
- $F_T^{NL}$  = the nonlinear fresnel factor of the transmitted homogeneous wave
- $F_R^{NL}$  = the nonlinear fresnel factor of the reflected harmonic wave
- $F_S^{NL}$  = the nonlinear fresnel factor of the transmitted inhomogeneous wave
- $I_{R,S,T}$  = the power of the reflected, transmitted inhomogeneous and homogeneous wave respectively
- $A_{R,S,T}$  = the cross section area of the reflected, transmitted inhomogeneous and homogeneous light beam respectively
- $I_{total}^T$  = total transmitted second harmonic intensity

# Chapter I

## Introduction

### 1.1 Literature Review

After its invention by Gordon (1954), the new theory of the Maser was presented by Bloembergen (1956). Later, the theory of optical Masers that was presented by Schawlow and Townes (1958) and following the first invention of laser (Ruby laser) by Maiman (1960) appeared. This laser light source not only has one frequency (monochromatic) at wavelength equals 694.3 nm, but also high coherence and higher intensity than any other light source known at that time. Physicists have been able to utilize the properties of the laser light above them for the study of nonlinear optic phenomena, since an ordinary light source is not monochromatic and has low intensity. The nonlinear optic effects occur when the intensity of the induced electric field of light sources have high value (*about*  $10^8 \text{ V/m}$ ). Second harmonic generation in transmission of laser light was performed for the first time by using quartz by Franken and Weinreich (1961). The experiment was conducted by using ruby laser at wavelength equal 694.3 nm incident on the quartz and it was found that the emerging transmitted light had double frequencies of the incident laser. The nonlinear optical experiment carried out by Franken et al. was regarded as the beginning of nonlinear optical harmonic generation. Thereafter, the laser was frequently used for studies the nonlinear optic effects. The improvement of efficiency of second harmonic generation was performed by Maker, Terhune, Nieoff, and

Savage (1962) and Giordmaine (1962) by using a phase matching technique. Furthermore, Hellwarth (1961,1966) had achieved a generation of giant laser pulses, called Q-switched laser which became the standard laser pulse for second harmonic generation. Later on, the output pulsewidth of laser was improved to a narrow size of femtosecond by Shank, Fork, Yen, Stolen, and Toomlinson (1982). The theory of the interactions between light waves in nonlinear dielectric have been studied by Armstrong, Bloembergen, Ducuing, and Pershan (1962). Light waves at the boundary of nonlinear media have been studied by Bloembergen and Pershan (1962). Bloembergen and Pershan (BP) theory became the main theory for second harmonic generation and has been verified experimentally for a variety of geometrical situations. The first second harmonic generation in reflection by using ruby laser incident on GaAs crystal was demonstrated by Ducuing and Bloembergen (1963) and as a consequence the generalized Snell's law was established. Further, the values of reflected second harmonic light were varying relative with the incident angle and nonlinear polarization,  $\vec{P}^{NLS}(2\mathbf{w})$  in GaAs crystal. Later, Chang and Bloembergen (1966) used the same crystal in their experiment of second harmonic generation and successfully demonstrated the first minimum reflected second harmonic light at the condition of the nonlinear Brewster angle. The phenomenon was not clearly seen in their experiment because of the complex values of nonlinear susceptibility,  $\mathbf{C}_{ijk}$  of the GaAs crystal. Later Lee and Bhanthumnavin (1976) studied the reflected second harmonic light by using picosecond pulse of Nd: Glass laser incident on KDP crystal for achieve phenomena of nonlinear brewster angle from transparent medium was firstly discovered. In addition, the nonlinear brewster angle of ADP crystal was theoretically predicted by Bhanthumnavin and Ampole (1990). In former times, the

studying of second harmonic generation almost always involved the case of incident light normal to the crystal for observing the maximum transmitted second harmonic light. In 1969, Bolembergen, Simon, and Lee has demonstrated second harmonic generation in reflection and transmission when the incident light make an oblique angle of incidences with  $\text{NaClO}_3$  crystal. They also presented the first second harmonic generation at the noncolinear incidence or two beam spatial mixing (TBSM) condition. However the maximum second harmonic light at noncolinear phase matching condition was not achieved from this experiment. Later, Bhanthumnavin and Lee (1990) have demonstrated TBSM of transmitted second harmonic generation at the noncolinear phase matching condition from KDP crystal by using Nd: Glass laser at wavelength equals 1064 nm. Moreover, the maximum transmitted second harmonic light and the first null transmitted second harmonic intensity at normal incident angle when  $\vec{P}^{NLS}(2\mathbf{w})$  lies parallel to face normal were illustrated. Later Dirr, Hildebrand, Marowsky, and Stolle (1997) use the idea of null transmitted intensity for the demonstration of the nonlinear brewster angle in transmission in liquid crystal cell. Later in 1994, Bhanthumnavin and Lee have performed the maximum and minimum second harmonic generation in reflection and transmission from KDP crystal at different orientations of  $\vec{P}^{NLS}(2\mathbf{w})$  by using Mode-locked Nd: Glass laser at wavelength equal 1064 nm.

However, the field of transmitted second harmonic generation is interesting and not fully investigated. In order to improve the knowledge of second harmonic generation, this thesis theoretically studies transmitted second harmonic generation from Ammonium Dihydrogen Phosphate (ADP) crystal immersed in the optically denser liquid 1-Bromonaphthalene. This theoretical study will be based on the theory

of Bloembergen and Pershan by using an ultrashort laser pulse (Shapiro, 1977) at wavelength equals 900 nm.

## **1.2 Purpose**

The main purpose of this thesis is the theoretical study of second-harmonic intensity in transmission from ADP crystals as a function of incident angle by using an ultra short pulse laser at wavelength 900 nm. The ADP crystal has real susceptibility so that second harmonic light will not be absorbed. An ultrashort pulse laser is used as a source at wavelength 900 nm because its high peak power output can help to generate second-harmonic beam in crystal more efficiently than a low peak power laser. The study is involved with two cases of the characteristics of the incident light: the colinear and noncolinear incident cases. The colinear incident case means there are two incident lights having the same direction of propagation and also the same angle of incidence. In this case of incidence, both maximum and minimum transmitted second harmonic generation at different orientations of ADP crystal will be searched for. The maximum transmitted second harmonic generation at the noncolinear phase matching condition will be studied. This technique had been theoretical studied and was demonstrated by the theory of Bloembergen and Pershan (1962) furthermore it had been first verified experimentally by Bhanthumnavin and Lee (1990).

### **1.3 Hypothesis**

According to the purpose, it is expected that second-harmonic light in transmission will be obtained by using ultrashort pulse laser at wavelength equals 900 nm. For the colinear incidence case, the maximum and minimum of transmitted second-harmonic intensity in case of colinear phase matching are expected to occur at new crystallographic orientation of ADP crystal. For noncolinear incidence case, the maximum transmitted second harmonic intensity in case of noncolinear phase matching is expected to occur from ADP crystal.

# Chapter II

## Theory

### 2.1 Introduction

When an electromagnetic wave is incident on crystalline materials, the electron is induced by an electric field creating dipole and linear polarization  $\vec{P}^L$ . As a consequence, there are waves could be classified as reflected and transmitted waves. However, if the incident light is a monochromatic wave with high intensity such as laser light not only linear polarization but also nonlinear polarization  $\vec{P}^{NLS}$  will be created.  $\vec{P}^L$  is a source of linear optic effects and  $\vec{P}^{NLS}$  is a nonlinear optical source that caused transmitted and reflected harmonic light in material. In this theoretical study, only second harmonic generation will be considered. Further, if the nonlinear medium is a uniaxial crystal, there will be two beams of transmitted light due to different values of refractive indices, which are the ordinary ray index  $n_o$  and extraordinary ray index  $n_e$ . The theory of second harmonic generation has been developed (Bloembergen and Pershan, 1962) after the first discovery of second harmonic light by Franken et al. (1961). The theory of light waves at the boundary of nonlinear media (Bloembergen and Pershan, 1962) has successful predictions for various phenomena of second harmonic generation. Therefore in this thesis, the theory of Bloembergen and Pershan will be employed as the main theoretical analysis

tool for transmitted second harmonic generation in Ammonium Dihydrogen Phosphate or ADP crystal.

## 2.2 Theory of Second Harmonic Generation

The second harmonic light, generated by  $\vec{P}^{NLS}$ , is created in the material, by using high intensity of light source such as laser light. The occurrence of  $\vec{P}^{NLS}$  can be theoretically created as of following.

### 2.2.1 Nonlinear Polarization, $\vec{P}^{NLS}$

When laser light source with high peak power output is incident on the crystal, the polarization in the crystal will not only have the linear polarization term, ( $\vec{P}^L$ ) but also have nonlinear polarization term, ( $\vec{P}^{NLS}$ ). The relationship between total polarization,  $\vec{P}^{Total}$  and electric field,  $\vec{E}$  of the incident light can be expressed as the following.

$$\vec{P}^{NLS} = \mathbf{c}^L \vec{E}(\mathbf{w}) + \mathbf{c}^{NL}(\mathbf{w}_2 = 2\mathbf{w}) \vec{E}(\mathbf{w}) \vec{E}(\mathbf{w}) + \dots, \quad (2.1)$$

where  $\mathbf{c}^L$  is the linear susceptibility,  $\mathbf{c}^{NL}$  is the nonlinear susceptibility tensor, and  $\mathbf{w}$  is the frequency. From equation (2.1), the nonlinear polarization is not only proportional to  $\vec{E}$  but also proportional to  $|E|^2$ . The term  $\mathbf{c}^{NL} \vec{E} \vec{E}$  of equation (2.1) is the nonlinear source term which second harmonic light ( $\mathbf{w}_2 = 2\mathbf{w}_1$ ) is generated. This second order polarization is given by

$$\vec{P}^{NLS}(\mathbf{w}_2) = \mathbf{c}^{NL} \vec{E}(\mathbf{w}_1) \vec{E}(\mathbf{w}_1). \quad (2.2)$$

The coefficient  $\mathbf{c}^{NL}$  is a tensor so that the vector  $\bar{\mathbf{P}}^{NL}$  is not necessary parallel to vector  $\bar{\mathbf{E}}$ . The nonlinear polarization,  $\bar{\mathbf{P}}^{NLS}(\mathbf{2}\mathbf{w})$  could be expressed in term of summation of its components as.

$$\bar{P}_i^{NLS}(\mathbf{2}\mathbf{w}) = \mathbf{a} \underset{j,k}{c}_{ijk} \bar{E}_j \bar{E}_k, \quad (2.3)$$

where  $i, j, k$  represent the coordinates  $x, y, z$ . The  $\bar{\mathbf{P}}^{NLS}(\mathbf{2}\mathbf{w})$  occurs only in certain type of crystals that lack inversion symmetry or noncentrosymmetric crystals.

### 2.2.2 Maxwell Equations of Nonlinear Medium

In order to describe the behavior of second harmonic light at the boundary of nonlinear medium, Maxwell equations are considered and employed as follows

$$\tilde{\mathbf{N}} \times \bar{\mathbf{E}} = -\frac{1}{c} \frac{\partial \tilde{\mathbf{m}} \bar{\mathbf{H}}}{\partial t}, \quad (2.4)$$

$$\tilde{\mathbf{N}} \cdot \bar{\mathbf{H}} = \frac{1}{c} \frac{\mathbf{1} \bar{\mathbf{D}}}{\mathbf{1} t} + \frac{4\mathbf{p}}{c} \bar{\mathbf{J}}, \quad (2.5)$$

$$\tilde{\mathbf{N}} \times \bar{\mathbf{D}} = 4\mathbf{p} \bar{\mathbf{r}}, \quad (2.6)$$

$$\tilde{\mathbf{N}} \times \bar{\mathbf{B}} = 0, \quad (2.7)$$

where  $\bar{\mathbf{D}} = \mathbf{e} \bar{\mathbf{E}} + 4\mathbf{p} \bar{\mathbf{P}}^{NLS}$ ,  $\bar{\mathbf{E}}$  is the electric field,  $\bar{\mathbf{H}}$  is the magnetic field,  $\bar{\mathbf{D}}$  is the electric flux density,  $\bar{\mathbf{B}}$  is the magnetic flux density,  $\bar{\mathbf{P}}^{NLS}$  is the nonlinear polarization,  $\mathbf{e}$  and  $\mathbf{m}$  are the permittivity and permeability of crystal respectively. For a noncentrosymmetric crystal, consider  $\mathbf{e}$  as a scalar and nonmagnetic material, so that  $\mathbf{m}=1$ . The wave equation is given by

$$\tilde{\mathbf{N}} \cdot \tilde{\mathbf{N}} \cdot \bar{\mathbf{E}} + \frac{\mathbf{e}(2\mathbf{w}_1)}{c^2} \frac{\mathbf{1}^2 \bar{\mathbf{E}}}{\mathbf{1} t^2} = -\frac{4\mathbf{p}}{c^2} \frac{\mathbf{1}^2 \bar{\mathbf{P}}^{NLS}(\mathbf{2}\mathbf{w})}{\mathbf{1} t^2}, \quad (2.8)$$

$$\text{or} \quad \nabla^2 \bar{E} - \frac{\mathbf{e}(2\mathbf{w})}{c^2} \frac{\partial^2 \bar{E}}{\partial t^2} = \frac{4\mathbf{p}}{c^2} \frac{\partial^2 \bar{P}^{NLS}(2\mathbf{w})}{\partial t^2}, \quad (2.9)$$

where  $\bar{P}^{NLS}(2\mathbf{w})$  is the nonlinear polarization.

The nonlinear source term on the right hand side of (2.9) will yield an inhomogeneous solution of the wave equation. The nonlinear source term at frequency  $\mathbf{w}_2 = 2\mathbf{w}_1$  is given by

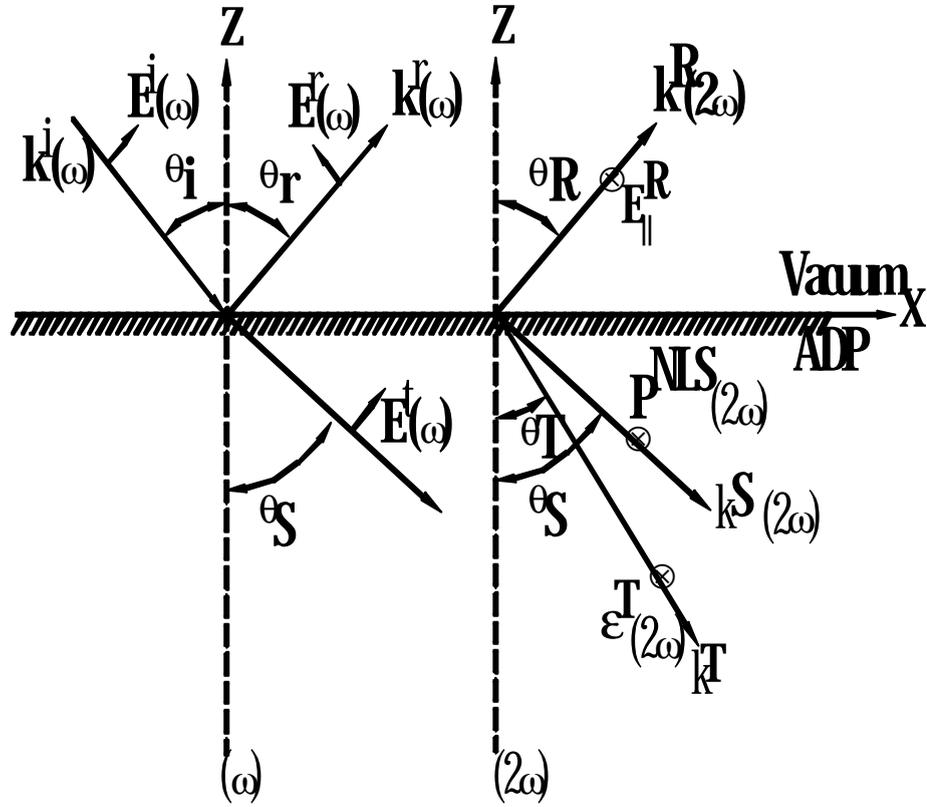
$$\bar{P}^{NLS}(2\mathbf{w}_1) = \mathbf{c}_2(\mathbf{w}_2 = 2\mathbf{w}_1) \bar{E}_1^T \bar{E}_1^T \exp i(\bar{k}^S \cdot \bar{r} - 2\mathbf{w}_1 t), \quad (2.10)$$

where  $\bar{E}_1^T$  is the transmitted electric field in the crystal,  $\bar{k}^S$  is the source wave vector ( $\bar{k}^S = 2\bar{k}_1^T$ ),  $t$  is the time that wave propagates along distance wave vector  $\bar{r}$ . By using equation (2.9) and (2.10), the electric ( $\bar{E}_2^T$ ) and magnetic ( $\bar{H}_2^T$ ) fields of the transmitted second harmonic light and the electric ( $\bar{E}_2^R$ ) and magnetic ( $\bar{H}_2^R$ ) fields of the refracted second harmonic light, of the incident light from vacuum to the nonlinear medium are given by

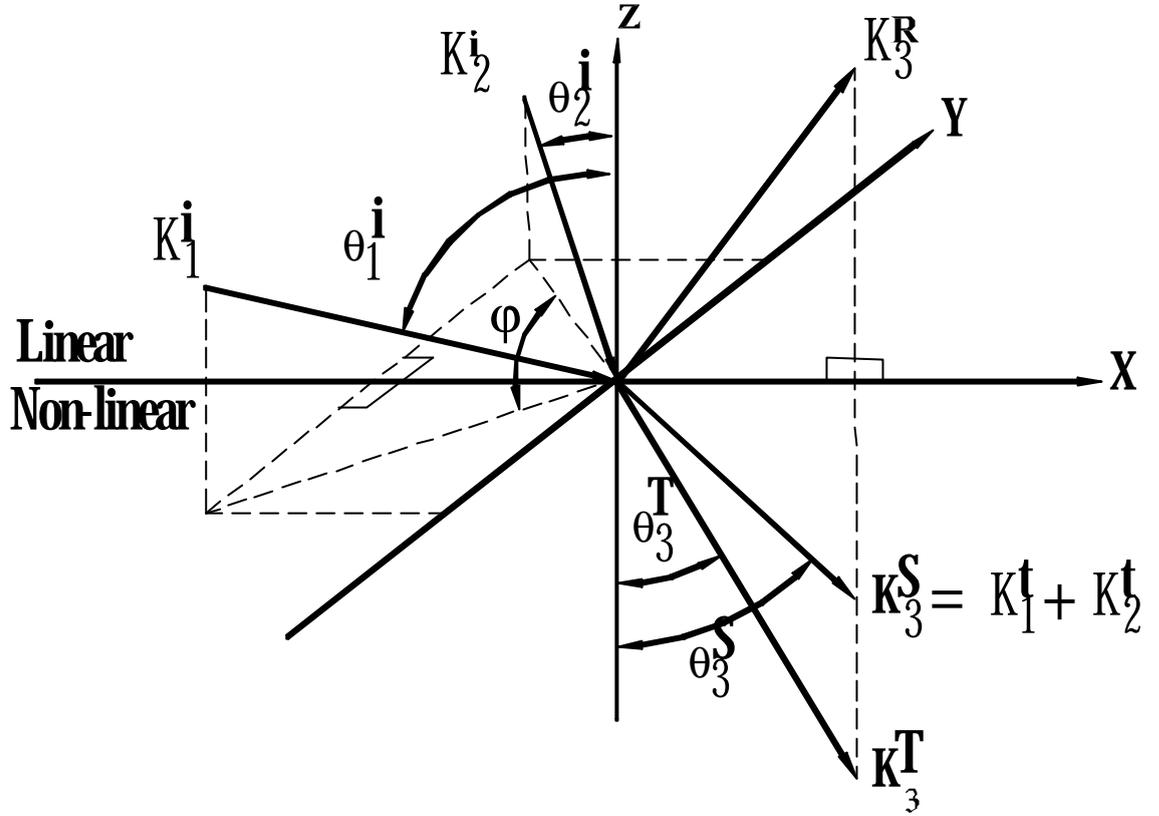
$$\begin{aligned} \bar{E}_2^T &= \hat{e}_T \mathbf{e}_2^T \exp i(\bar{k}_2^T \cdot \mathbf{x}\hat{r} - 2\mathbf{w}_1 t) - \frac{4\mathbf{p}^{NLS}(\frac{4\mathbf{w}_1^2}{c^2})}{(k_2^T)^2 - (k^S)^2} \\ &\times \left[ \hat{p} - \frac{\bar{k}^S (\bar{k}^S \cdot \hat{p})}{(k^T)^2} \right] \exp i(\bar{k}^S \cdot \bar{r} - 2\mathbf{w}_1 t), \\ \bar{H}_2^T &= \frac{c}{2\mathbf{w}_1} (\bar{k}_2^T \wedge \hat{e}_T) \mathbf{e}_2^T \exp i(\bar{k}_2^T \cdot \mathbf{x}\hat{r} - 2\mathbf{w}_1 t) \\ &- \frac{4\mathbf{p}^{NLS}(\frac{4\mathbf{w}_1^2}{c^2})}{(k_2^T)^2 - (k^S)^2} \frac{c}{2\mathbf{w}_1} (\bar{k}^S \times \hat{p}) \exp i(\bar{k}^S \cdot \bar{r} - 2\mathbf{w}_1 t), \\ \bar{E}_2^R &= \hat{e}_R \mathbf{e}_2^R \exp i(\bar{k}_2^R \cdot \mathbf{x}\hat{r} - 2\mathbf{w}_1 t), \end{aligned} \quad (2.11)$$

$$\vec{H}_2^R = \left(\frac{c}{2\omega_1}\right)(\vec{k}_2^R \times \hat{e}_R) \mathbf{e}_2^R \exp i(\vec{k}_2^R \cdot \hat{r} - 2\omega_1 t), \quad (2.12)$$

where  $\mathbf{e}_2^T, \mathbf{e}_2^R$  are the amplitude of the electric field of the transmitted and refracted second harmonic light respectively,  $\vec{k}_2^R$  and  $\vec{k}_2^T$  are the wave vector of the reflected wave and homogeneous transmitted wave which have the unit vector  $\hat{e}_R$  and  $\hat{e}_T$  respectively and  $\hat{p}$  is the unit vector of  $\vec{P}^{NLS}(2\omega)$  as shown in figure 2.1.



**Figure 2.1.** The incident, reflected and refracted rays, near the boundary between vacuum and ADP crystal at the fundamental and second harmonic frequencies.



**Figure 2.2.** Two incident rays at frequencies  $\omega_1$  and  $\omega_2$  caused the reflected wave, a homogeneous and inhomogeneous transmitted wave at the sum frequency  $\omega_3 = \omega_1 + \omega_2$  eliminate from the boundary between the linear and nonlinear medium.

From figure 2.2, the fundamental incident beams are the two incident waves represent by  $\vec{E}_1 \exp i(\vec{k}_1^i \cdot \vec{r} - \omega_1 t)$  and  $\vec{E}_2 \exp i(\vec{k}_2^i \cdot \vec{r} - \omega_2 t)$ . The incident, reflected, homogeneous transmitted and inhomogeneous transmitted wave vectors are represented by  $\vec{k}_1^i$ ,  $\vec{k}_2^i$ ,  $\vec{k}_3^R$ ,  $\vec{k}_3^T$ ,  $\vec{k}_3^S$  respectively. These two waves are incident at the interface between linear and nonlinear medium. The angle of incidence of wave

vectors  $\bar{k}_1^i$  and  $\bar{k}_2^i$  are represented by  $\mathbf{q}_1^i$  and  $\mathbf{q}_2^i$  respectively. The planes of incident waves  $\bar{k}_1^i$  and  $\bar{k}_2^i$  make an angle  $\mathbf{j}$  with each other. Consider  $x$  and  $y$  direction of the coordinate system at plane  $z = 0$  (the interface between linear and nonlinear medium), the  $x$ ,  $y$  components of the momentum wave vector are conserved. The summation of the wave vector leads to the condition

$$\begin{aligned} k_{3x}^R &= k_{3x}^T = k_{3x}^S = k_{1x}^T + k_{2x}^T = k_{1x}^i + k_{2x}^i, \\ k_{3y}^R &= k_{3y}^T = k_{3y}^S = k_{1y}^T + k_{2y}^T = k_{1y}^i + k_{2y}^i = 0. \end{aligned} \quad (2.13)$$

From (2.13), it is found that the inhomogeneous wave, the transmitted and reflected waves and the boundary normal lie in the  $xz$  plane called the plane of sum reflection. The propagation of the inhomogeneous wave is at the sum frequency proportional to  $\bar{P}^{NLS}(\mathbf{2}\mathbf{w})$  and given by  $\exp i \{ (\bar{k}_1^T + \bar{k}_2^T) \cdot \mathbf{r} - (\mathbf{w}_1 + \mathbf{w}_2)t \}$ . The angle between the normal and its direction  $\mathbf{q}_3^S$  is determined by

$$\sin \mathbf{q}_3^S = |k_{1x}^T + k_{2x}^T| / |\bar{k}_1^T + \bar{k}_2^T|. \quad (2.14)$$

The propagation directions of wave vectors  $k_1^T$  and  $k_2^T$  are given by Snell's law for refraction. From figure 2.2, the value of the angle  $\mathbf{j}$  varies from 0 to  $\mathbf{p}$  and the values of the incident angle  $\mathbf{q}_i$  vary from 0 to  $2\mathbf{p}$ . The wave vectors  $k_3^T, k_1^i$  and  $k_2^i$  are related by following equation

$$\begin{aligned} |\bar{k}_3^T|^2 \sin^2 \mathbf{q}_3^T &= |\bar{k}_3^R|^2 \sin^2 \mathbf{q}_3^R \\ &= |\bar{k}_1^i|^2 \sin^2 \mathbf{q}_1^i + |\bar{k}_2^i|^2 \sin^2 \mathbf{q}_2^i + 2|\bar{k}_1^i|^2 |\bar{k}_2^i|^2 \sin \mathbf{q}_1^i \sin \mathbf{q}_2^i \cos \mathbf{j}. \end{aligned} \quad (2.15)$$

The permittivity is defined by  $\mathbf{e} = k^2 \frac{c^2}{\mathbf{w}^2}$ . Equation (2.15) can be rewritten as the following

$$\begin{aligned}
\mathbf{e}_3^T \mathbf{w}_3^2 \sin^2 \mathbf{q}_3^T &= \mathbf{e}_3^R \mathbf{w}_3^3 \sin^2 \mathbf{q}_3^R \\
&= \mathbf{e}_1^R \mathbf{w}_1^2 \sin^2 \mathbf{q}_1^i + \mathbf{e}_2^R \mathbf{w}_2^2 \sin^2 \mathbf{q}_2^i + 2(\mathbf{e}_1^R \mathbf{e}_2^R)^{1/2} \mathbf{w}_1 \mathbf{w}_2 \sin \mathbf{q}_1^i \sin \mathbf{q}_2^i \cos \mathbf{j} . \quad (2.16)
\end{aligned}$$

From equation (2.13) and (2.16) we get

$$\mathbf{e}_3^S \sin^2 \mathbf{q}_3^S = \mathbf{e}_3^T \sin^2 \mathbf{q}_3^T = \mathbf{e}_3^R \sin^2 \mathbf{q}_3^R . \quad (2.17)$$

If two incident beams have the same angle of incidence  $\mathbf{q}_1^i = \mathbf{q}_2^i$ , and also  $\mathbf{j} = 0$ ,  $\mathbf{e}_1^R = \mathbf{e}_2^R = \mathbf{e}_3^R$ ,  $\mathbf{w}_1 = \mathbf{w}_2 = \mathbf{w}$ , equation (2.16) becomes

$$\mathbf{e}_3^T \sin^2 \mathbf{q}_3^T = \mathbf{e}_1^R \sin^2 \mathbf{q}_1^i . \quad (2.18)$$

Thus equation (2.17) can be written as

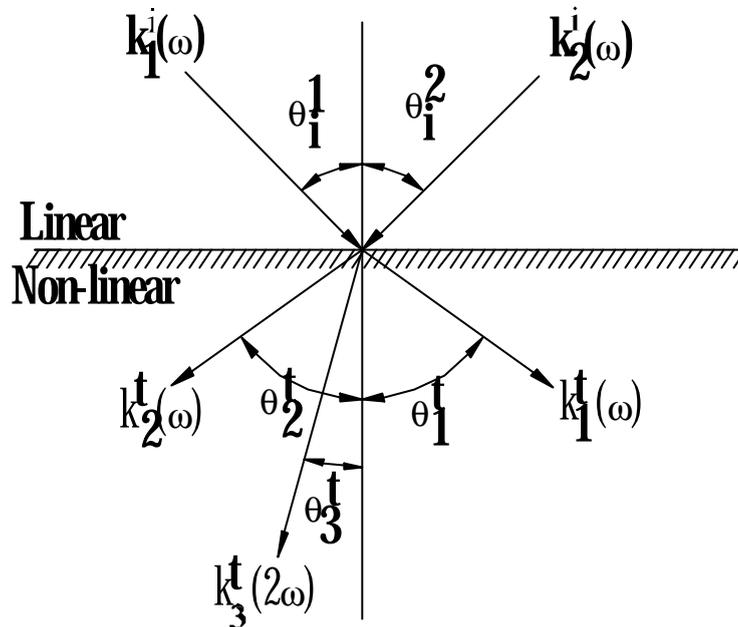
$$\sqrt{\mathbf{e}_1^R} \sin \mathbf{q}_1^i = \sqrt{\mathbf{e}_3^T} \sin \mathbf{q}_3^T = \sqrt{\mathbf{e}_3^R} \sin \mathbf{q}_3^R = \sqrt{\mathbf{e}_3^S} \sin \mathbf{q}_3^S , \quad (2.19)$$

where the refractive index is,  $n = \sqrt{\mathbf{e}}$ . This is the case of colinear incidence. figure 2.6.

Now if two incident beams are incident on the crystal at this case is opposite directions and make the same angle of incidence with the face normal, that called two beam special mixing, (TBSM),  $\mathbf{q}_1^i = \mathbf{q}_2^i$ . The angle  $\mathbf{j} = 180^\circ$ , means that two incident beams lie on the same plane of incidence. From (2.16) and (2.17), it can be shown that

$$\mathbf{q}_3^T = \mathbf{q}_3^S = \mathbf{q}_3^R = 0^\circ \quad \left[ \text{if } k_1^i(\mathbf{w}) = k_2^i(\mathbf{w}) \right]. \quad (2.20)$$

This means the wave vectors  $\bar{k}_3^T$ ,  $\bar{k}_3^R$  and  $\bar{k}_3^S$  propagate along the direction of face normal. The noncolinear incident in general case as showed in figure 2.3.



**Figure 2.3.** The noncolinear incident case of Two Beam Spatial Mixing

[ for  $k_1^i(\omega) \neq k_2^i(\omega)$  ].

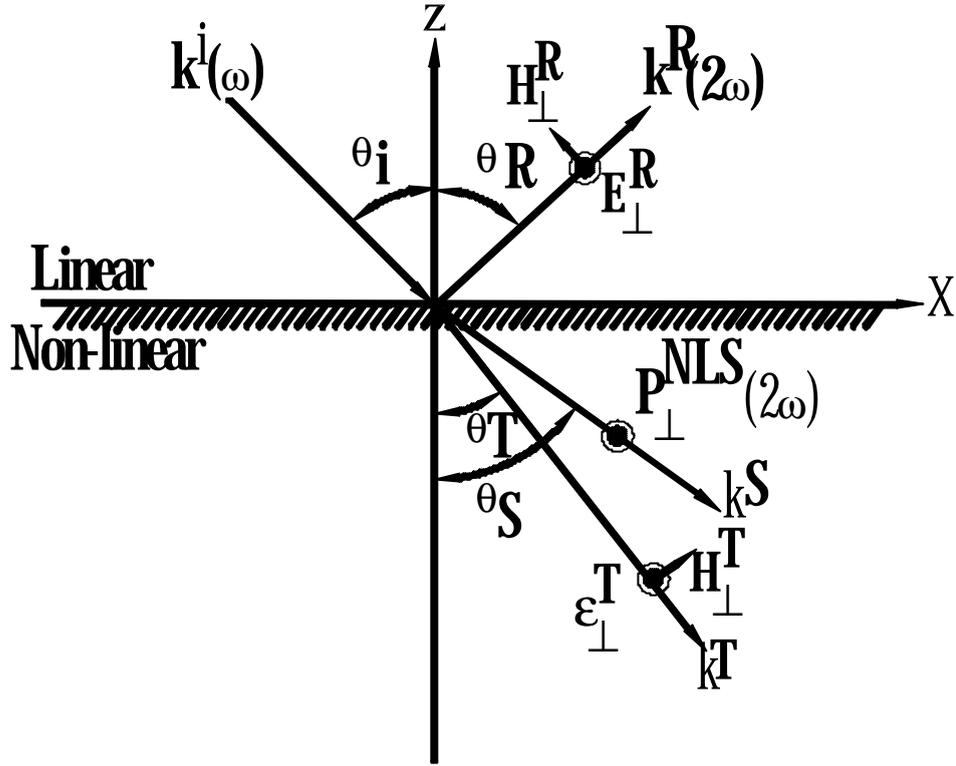
### 2.2.3 Nonlinear Polarization and Second Harmonic Generation

In order to analyze second harmonic generation, the direction of polarization of the incident light could be classified into two types: perpendicular and parallel to the plane of incidence.

#### A. Nonlinear Polarization Perpendicular to the Plane of Incidence,

$$(\bar{P}_{\perp}^{NLS}(2\omega))$$

The beam of frequency ( $\omega$ ) is incident on the crystal that lies on the  $xy$  plane with boundary at plane  $z=0$ . The direction of nonlinear polarization occurred in the  $y$  direction perpendicular to the plane of incidence,



**Figure 2.4** The second harmonic and electric and magnetic field when nonlinear polarization  $\bar{P}^{NLS}(2\omega)$  perpendicular to the plane of incidence.

$\bar{P}_Y^{NLS} = \bar{P}_\perp^{NLS}(2\omega)$ . The directions of polarization of second harmonic waves are parallel to the plane of incidence as shown in figure 2.4. In this case, using equation (2.11) and (2.12), the component of the electric and magnetic fields have the following form

$$E_y = E_\perp^R = \mathbf{e}_\perp^T + 4\mathbf{p}P_\perp^{NLS}(\mathbf{e}_S - \mathbf{e}_T)^{-1}, \quad (2.21)$$

$$H_y = -\mathbf{e}_R^{y/2} E_\perp^R \cos \mathbf{q}_R = \mathbf{e}_T^T \mathbf{e}_\perp^T \cos \mathbf{q}_T + 4\mathbf{p}e_S^{y/2}(\mathbf{e}_S - \mathbf{e}_T)^{-1} P_\perp^{NLS} \cos \mathbf{q}_S. \quad (2.22)$$

Combining equations (2.21) and (2.22),  $E_\perp^R$  can be written as

$$E_{\wedge}^R = - \frac{4\mathbf{p}^{\wedge NLS} \hat{\mathbf{e}} \mathbf{e}_T^{1/2} \cos \mathbf{q}_T - \mathbf{e}_S^{1/2} \cos \mathbf{q}_S \dot{\mathbf{u}}}{\mathbf{e}_T - \mathbf{e}_S \hat{\mathbf{e}} \mathbf{e}_T^{1/2} \cos \mathbf{q}_T + \mathbf{e}_R^{1/2} \cos \mathbf{q}_R \dot{\mathbf{u}}} \dot{\mathbf{u}}. \quad (2.23)$$

By substituting equation (2.23) into equation (2.21), the amplitude of transmitted second harmonic wave,  $\mathbf{e}_{\wedge}^T$  has the following form

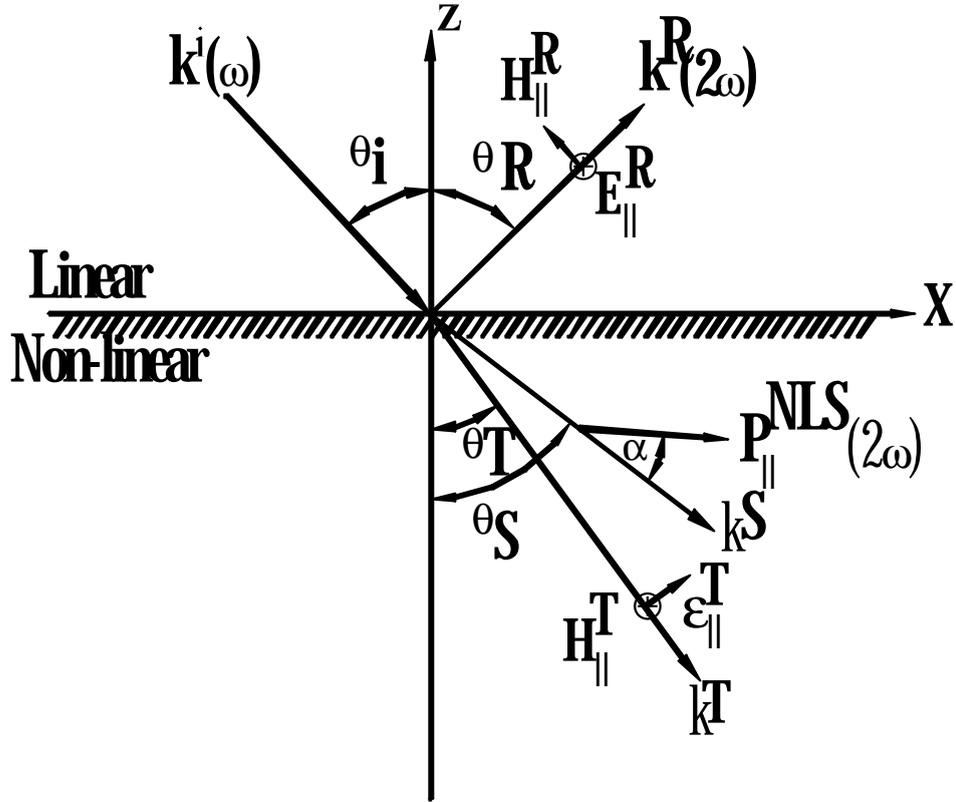
$$\begin{aligned} \mathbf{e}_{\wedge}^T &= E_{\wedge}^R - 4\mathbf{p}^{\wedge NLS} (\mathbf{e}_S - \mathbf{e}_T)^{-1} \\ &= - \frac{4\mathbf{p}^{\wedge NLS} \hat{\mathbf{e}} \mathbf{e}_T^{1/2} \cos \mathbf{q}_T - \mathbf{e}_S^{1/2} \cos \mathbf{q}_S \dot{\mathbf{u}}}{\mathbf{e}_T - \mathbf{e}_S \hat{\mathbf{e}} \mathbf{e}_T^{1/2} \cos \mathbf{q}_T + \mathbf{e}_R^{1/2} \cos \mathbf{q}_R \dot{\mathbf{u}}} - \frac{4\mathbf{p}^{\wedge NLS}}{\mathbf{e}_S - \mathbf{e}_T}. \end{aligned} \quad (2.24)$$

### B. Nonlinear Polarization Parallel to the Plane of Incidence, ( $P_{//}^{NLS}(2\mathbf{w})$ )

When the polarization of the incident wave is parallel to the  $xy$  plane, the plane of incidence, the values of the  $y$  components of the incident electric field,  $E_y^i$  and nonlinear polarization,  $\bar{P}_y^{NLS}$ , are definitely equal to zero. The nonlinear polarization parallel to the  $xz$  plane make an angle alpha,  $\mathbf{a}$  with the direction of propagation of the transmitted source wave vector,  $\bar{k}^S$  as shown in figure 2.5. From equation (2.11) and (2.12), the tangential components at  $z=0$  of the electric and magnetic field become

$$\begin{aligned} E_x &= -E_{//}^R \cos \mathbf{q}_R = \mathbf{e}_{//}^T \cos \mathbf{q}_T + \frac{4\mathbf{p}^{\wedge NLS} \sin \mathbf{a} \cos \mathbf{q}_S}{\mathbf{e}_S - \mathbf{e}_T} \\ &\quad - \frac{4\mathbf{p}^{\wedge NLS} \cos \mathbf{a} \sin \mathbf{q}_S}{\mathbf{e}_T}, \end{aligned} \quad (2.25)$$

$$H_y = -\mathbf{e}_R^{1/2} E_{//}^R = -\mathbf{e}_T^{1/2} \mathbf{e}_{//}^T - \mathbf{e}_S^{1/2} \frac{4\mathbf{p}_{//}^{\wedge NLS} \sin \mathbf{a}}{\mathbf{e}_S - \mathbf{e}_T}. \quad (2.26)$$



**Figure 2.5** The second harmonic electric and magnetic field when nonlinear polarization  $\bar{P}^{NLS}(2\omega)$  parallel to the plane of incidence.

The amplitude of the reflected harmonic field,  $E_{//}^R$ , is given by substituting

$\mathbf{e}_{//}^T$  of (2.26) into (2.25) and has the following form

$$E_{//}^R = \frac{4\mathbf{p}_{//}^{NLS} \sin \mathbf{a}}{\mathbf{e}_R^{1/2} \cos \mathbf{q}_T - \mathbf{e}_T^{1/2} \cos \mathbf{q}_R} \frac{\hat{\mathbf{e}}_1 - (\mathbf{e}_S + \mathbf{e}_T)^{-1} \mathbf{e}_R \sin^2 \mathbf{q}_R}{\hat{\mathbf{e}} \mathbf{e}^{1/2} \cos \mathbf{q}_T + \mathbf{e}_T^{1/2} \cos \mathbf{q}_S} \frac{\hat{\mathbf{u}}}{\hat{\mathbf{u}}} + \frac{4\mathbf{p}_{//}^{NLS} \cos \mathbf{a} \sin \mathbf{q}_S}{\mathbf{e}_T^{1/2} \mathbf{e}_R^{1/2} \cos \mathbf{q}_T - \mathbf{e}_T \cos \mathbf{q}_R} \quad (2.27)$$

Using (2.16), (2.27) can be rewritten as

$$E_{//}^R = \frac{4\mathbf{p}_{//}^{NLS} \sin \mathbf{q}_S \sin^2 \mathbf{q}_T \sin(\mathbf{a} + \mathbf{q}_T + \mathbf{q}_S)}{\mathbf{e}_S \sin \mathbf{q}_R \sin(\mathbf{q}_T + \mathbf{q}_S) \sin(\mathbf{q}_T + \mathbf{q}_R) \cos(\mathbf{q}_T - \mathbf{q}_R)}. \quad (2.28)$$

Substituting (2.28) into (2.26), the amplitude of the transmitted second harmonic wave,  $\mathbf{e}_{//}^T$  is given by

$$\mathbf{e}_{//}^T = \frac{\mathbf{e}_R^{1/2} E_{//}^R}{\mathbf{e}_T^{1/2}} - \frac{\mathbf{e}_S^{1/2} 4\mathbf{p}_{//}^{NLS} \sin \mathbf{a}}{\mathbf{e}_T^{1/2} (\mathbf{e}_S - \mathbf{e}_T)}. \quad (2.29)$$

From (2.23), (2.24), (2.27), and (2.29), the electric fields of second harmonic waves compose with the term of nonlinear polarization,  $\bar{\mathbf{P}}^{NLS}(2\mathbf{w})$ . These relations show and confirm that  $\bar{\mathbf{P}}^{NLS}(2\mathbf{w})$  was a source term of second harmonic generation so the electric fields of second harmonic wave could be written in term of  $\bar{\mathbf{P}}^{NLS}(2\mathbf{w})$ .

$$\bar{\mathbf{E}}_R(2\mathbf{w}) = 4\bar{\mathbf{p}}^{NLS} F_R^{NL}, \quad (2.30)$$

$$\bar{\mathbf{E}}_T(2\mathbf{w}) = 4\bar{\mathbf{p}}^{NLS} F_T^{NL}, \quad (2.31)$$

$$\bar{\mathbf{E}}_S(2\mathbf{w}) = 4\bar{\mathbf{p}}^{NLS} F_S^{NL}, \quad (2.32)$$

where  $\bar{\mathbf{E}}_R(2\mathbf{w})$  is the electric field of the reflected harmonic wave,  $\bar{\mathbf{E}}_T(2\mathbf{w})$  and  $\bar{\mathbf{E}}_S(2\mathbf{w})$  are the electric fields of the transmitted harmonic wave of the wave vectors  $\bar{k}^R$ ,  $\bar{k}^T$ , and  $\bar{k}^S$  respectively. The nonlinear fresnel factors  $F_R^{NL}$ ,  $F_S^{NL}$ , and  $F_T^{NL}$  are the ratio of the amplitude of the incident field on the amplitude of the reflected, transmitted inhomogeneous and transmitted homogeneous wave respectively. The subscripts  $\wedge$ , and  $//$  will be used to represent the variables that occur in case of nonlinear polarization perpendicular and parallel to the plane of incidence respectively. From (2.23) and (2.24), the nonlinear fresnel factors when nonlinear polarization perpendicular to the plane of incidence are given by

$$F_{R, \wedge}^{NL} = \frac{1}{\mathbf{e}_S - \mathbf{e}_T} \frac{\hat{\mathbf{e}} \mathbf{e}_T^{1/2} \cos \mathbf{q}_T - \mathbf{e}_S^{1/2} \cos \mathbf{q}_S \hat{\mathbf{u}}}{\hat{\mathbf{e}} \mathbf{e}_T^{1/2} \cos \mathbf{q}_T + \mathbf{e}_R^{1/2} \cos \mathbf{q}_R \hat{\mathbf{u}}}, \quad (2.33)$$

$$F_{S, \wedge}^{NL} = \frac{1}{\mathbf{e}_S - \mathbf{e}_T}, \quad (2.34)$$

$$F_{T, \wedge}^{NL} = -\frac{1}{\mathbf{e}_T - \mathbf{e}_S} + F_{R, \wedge}^{NL}. \quad (2.35)$$

Consider (2.27) and (2.29). The nonlinear fresnel factors when the nonlinear polarization is parallel to the plane of incidence are given by

$$F_{R, //}^{NL} = \frac{\sin \mathbf{q}_S \sin^2 \mathbf{q}_T \sin(\mathbf{a} + \mathbf{q}_S + \mathbf{q}_T)}{\mathbf{e}_R \sin \mathbf{q}_R \sin(\mathbf{q}_T + \mathbf{q}_S) \sin(\mathbf{q}_T + \mathbf{q}_R) \cos(\mathbf{q}_T - \mathbf{q}_R)}, \quad (2.36)$$

$$F_{S, //}^{NL} = \frac{\sin \mathbf{a}}{\mathbf{e}_S - \mathbf{e}_T}, \quad (2.37)$$

$$F_{T, //}^{NL} = -\frac{\mathbf{e}_S^{1/2}}{\mathbf{e}_T^{1/2}} \frac{\sin \mathbf{a}}{\mathbf{e}_S - \mathbf{e}_T} + \frac{\mathbf{e}_R^{1/2}}{\mathbf{e}_T^{1/2}} F_{R, //}^{NL}, \quad (2.38)$$

where  $\mathbf{a}$  is the angle between  $\bar{\mathbf{P}}^{NLS}(2\mathbf{w})$  and  $\bar{\mathbf{k}}^S$ .

#### 2.2.4 Theoretical Calculation for Transmitted Second Harmonic Intensities

The transmitted second harmonic intensity will be theoretically studied as a function of the angle of incidence,  $\mathbf{q}_i$  from an ADP crystal, the nonlinear medium, that is immersed in the optically denser liquid 1-Bromonaphthalene, a linear medium, by the using ultra short pulse laser at wave length equal 900nm.

The ADP crystal is a uniaxial crystal, with a crystallographic point group  $\bar{4}2m$ . The elements of the nonlinear susceptibility tensor of the crystal,  $\mathbf{c}^{NL}$ , can be represented as a  $3 \times 6$  matrix so that relation between nonlinear polarization and the incident electric field is given by

$$\begin{bmatrix} \bar{P}_x^{NLS}(\mathbf{2}\mathbf{w}) \\ \bar{P}_y^{NLS}(\mathbf{2}\mathbf{w}) \\ \bar{P}_z^{NLS}(\mathbf{2}\mathbf{w}) \end{bmatrix} = \begin{bmatrix} 0 & 0 & 0 & \mathbf{c}_{14} & 0 & 0 \\ 0 & 0 & 0 & 0 & \mathbf{c}_{25} & 0 \\ 0 & 0 & 0 & 0 & 0 & \mathbf{c}_{36} \end{bmatrix} \begin{bmatrix} \bar{E}_x^2 \\ \bar{E}_y^2 \\ \bar{E}_z^2 \\ 2\bar{E}_z\bar{E}_y \\ 2\bar{E}_z\bar{E}_x \\ 2\bar{E}_x\bar{E}_y \end{bmatrix}, \quad (2.39)$$

where  $\mathbf{c}_{14}$ ,  $\mathbf{c}_{25}$ , and  $\mathbf{c}_{36}$  are the nonlinear susceptibilities of the crystal and the value of  $\mathbf{c}_{14}$  is equal to  $\mathbf{c}_{25}$ . From (2.39), the components of nonlinear polarization,  $P^{NLS}(\mathbf{2}\mathbf{w})$  at each point inside the crystal are represented by

$$\begin{aligned} \bar{P}_x^{NLS}(\mathbf{2}\mathbf{w}) &= 2\mathbf{c}_{14}\bar{E}_z\bar{E}_y, \\ \bar{P}_y^{NLS}(\mathbf{2}\mathbf{w}) &= 2\mathbf{c}_{25}\bar{E}_z\bar{E}_x, \\ \bar{P}_z^{NLS}(\mathbf{2}\mathbf{w}) &= 2\mathbf{c}_{36}\bar{E}_x\bar{E}_y. \end{aligned} \quad (2.48)$$

In this case, the incident laser pulses have the polarization in  $[\bar{1}\bar{1}0]$  direction with respect to ADP crystallographic axes so the electric field has direction of polarization perpendicular to the plane of incidence and causes the nonlinear polarization to be parallel to the plane of incidence. Since it has a component in x and y direction, equation (2.48), indicates that the nonlinear polarization will occur in z direction. (Fig. 2.6)

$$\bar{P}_z^{NLS}(\mathbf{2}\mathbf{w}) = 2\mathbf{c}_{36}\bar{E}_x\bar{E}_y, \quad (2.49)$$

where  $\bar{E}_x$  and  $\bar{E}_y$  are the component of the transmitted electric field in x and y direction respectively.



inhomogeneous transmitted wave vector  $(\mathbf{w})$ ,  $\bar{k}^S$  make the angle of incidence  $(\mathbf{w})$ ,  $\mathbf{q}_i$ , the angle of reflection  $(2\mathbf{w})$ ,  $\mathbf{q}_R$ , the angle of reflection  $(\mathbf{w})$ ,  $\mathbf{q}_t$ , the angle of transmission  $(2\mathbf{w})$ ,  $\mathbf{q}_T$  and  $\mathbf{q}_S$  with the normal line respectively. The driven polarization,  $\bar{P}^{NLS}(2\mathbf{w})$  which represents the particular solution of the inhomogeneous wave, has the same direction of propagation as the transmitted laser beam and has a wave vector  $\bar{k}^S = 2\bar{k}^L(\mathbf{w})$ .

In order to know the directions of the incident light wave, the reflected waves and the transmitted waves, the relation of the angle  $\mathbf{q}_i$ ,  $\mathbf{q}_R$ ,  $\mathbf{q}_S$ , and  $\mathbf{q}_T$  or the nonlinear Snell's law will be derived by using (2.16) when the value of index of refraction of the crystal is  $n = \mathbf{e}^{1/2}$ .

$$n_{liq}(\mathbf{w}) \sin \mathbf{q}_i = n_{liq}(2\mathbf{w}) \sin \mathbf{q}_R = n(\mathbf{w}) \sin \mathbf{q}_S = n(2\mathbf{w}) \sin \mathbf{q}_T, \quad (2.50)$$

where  $n_{liq}$ ,  $n(\mathbf{w})$ , and  $n(2\mathbf{w})$  are the refractive index of liquid and the refractive indices of ADP crystal respectively.

In practice, the transmitted electric field in the crystal can not be measured inside the crystal. Therefore, the linear fresnel factor, the ratio of the amplitude of the incident wave with the transmitted wave, will be used together with  $E_o$  instead of the term of transmitted electric field. Then (2.49) can be written as

$$\bar{P}_z^{NLS}(2\mathbf{w}) = \mathbf{c}_{36}^{NL} \mathbf{h} (F_T^L E_o)^2, \quad (2.51)$$

where  $E_o$  is the amplitude of the incident wave and  $\mathbf{h}$  is a geometrical factor. This factor depends on the polarization of the incident electric field and the direction of nonlinear polarization component with respect to the crystallographic axes of the ADP crystal.  $F_L^T$  is the linear fresnel factor and is given as

$$F_L^T = \frac{2 \sin \mathbf{q}_S \cos \mathbf{q}_i}{\sin(\mathbf{q}_S + \mathbf{q}_i)}. \quad (2.52)$$

In this case, the fundamental beam is transmitted parallel to the surface in the nonlinear crystal ADP. According to Bloembergen and Pershan theory, the nonlinear fresnel factors in case of nonlinear polarization  $\bar{P}^{NLS}(\mathbf{2w})$  parallel to the plane of incidence follow equation (2.36), (2.37), and (2.38) and are given by

$$F_{R||}^{NL} = \frac{\sin \mathbf{q}_S \sin^2 \mathbf{q}_T \sin(\mathbf{a} + \mathbf{q}_S + \mathbf{q}_T)}{\mathbf{e}_R(\mathbf{2w}) \sin \mathbf{q}_R \sin(\mathbf{q}_T + \mathbf{q}_R) \cos(\mathbf{q}_T - \mathbf{q}_R) \sin(\mathbf{q}_S + \mathbf{q}_T)}, \quad (2.36)$$

$$F_{S||}^{NL} = \frac{\sin \mathbf{a}}{(\mathbf{e}_R - \mathbf{e}_T)}, \quad (2.37)$$

$$F_{T||}^{NL} = \frac{\mathbf{e}_S^{1/2} \sin \mathbf{a}}{\mathbf{e}_T^{1/2} (\mathbf{e}_R - \mathbf{e}_T)} + \frac{\mathbf{e}_R^{1/2}}{\mathbf{e}_T^{1/2}} F_{R||}^{NL}, \quad (2.38)$$

where  $\mathbf{e}_R^{1/2}(\mathbf{2w})$ ,  $\mathbf{e}_S^{1/2}(\mathbf{2w})$ , and  $\mathbf{e}_T^{1/2}(\mathbf{2w})$  are equal to  $n_{liq}(\mathbf{2w})$ ,  $n_{KDP}(\mathbf{2w})$ , and  $n_{KDP}(\mathbf{2w})$  respectively.  $\mathbf{a}$  is the angle between  $\bar{P}^{NLS}(\mathbf{2w})$  and  $\bar{k}^S$ . The power of second harmonic beam is given by

$$I_{R,S,T}(\mathbf{2w}) = \left(\frac{c}{8\mathbf{p}}\right) \mathbf{e}_{R,S,T}^{1/2}(\mathbf{2w}) |E_{R,S,T}(\mathbf{2w})|^2 A_{R,S,T}. \quad (2.53)$$

The subscripts  $R, S$ , and  $T$  mean the reflected, transmitted inhomogeneous, and transmitted homogeneous respectively. The term  $A_{R,S,T}$  is the cross section area of the beam and is given by

$$A_{R,S,T} = (dd' \cos \mathbf{q}_{R,S,T}) / \cos \mathbf{q}_i, \quad (2.54)$$

where  $dd'$  is the width of the rectangular slit. In the experiment, the laser beam is incident on this slit before incident to the ADP crystal. Then  $dd'$  used to define the cross section area of the beam. Substituting equations (2.30), (2.31), (2.32), (2.36),

(2.37), (2.38), (2.54) into equation (2.53), the second harmonic intensities are given by

$$I_R(2\mathbf{w}) = \left(\frac{c}{8\mathbf{p}}\right) \mathbf{e}_R^{\frac{1}{2}} |E_0|^4 (4\mathbf{pc}_{36}^{NL})^2 \mathbf{h}^2 |F_L|^4 |F_{R,\downarrow}^{NL}|^2 \times \cos \mathbf{q}_R (\cos \mathbf{q}_i)^{-1}, \quad (2.55)$$

$$I_S(2\mathbf{w}) = \left(\frac{c}{8\mathbf{p}}\right) \mathbf{e}_R^{\frac{1}{2}} |E_0|^4 (4\mathbf{pc}_{36}^{NL})^2 \mathbf{h}^2 |F_L|^4 |F_{S,\downarrow}^{NL}|^2 \times \cos \mathbf{q}_S (\cos \mathbf{q}_i)^{-1}, \quad (2.56)$$

$$I_T(2\mathbf{w}) = \left(\frac{c}{8\mathbf{p}}\right) \mathbf{e}_R^{\frac{1}{2}} |E_0|^4 (4\mathbf{pc}_{36}^{NL})^2 \mathbf{h}^2 |F_L|^4 |F_{T,\downarrow}^{NL}|^2 \times \cos \mathbf{q}_T (\cos \mathbf{q}_i)^{-1}. \quad (2.57)$$

The intensities of the transmitted homogeneous wave and transmitted source will be obtained from equation (2.57) and (2.56) respectively. The two transmitted beams are not in the same direction, therefore, in general, the total transmitted second harmonic beam,  $I_{total}^T(2\mathbf{w})$  is calculated using

$$I_{total}^T(2\mathbf{w}) = I^S(2\mathbf{w}) + I^T(2\mathbf{w}) + 2I^S(2\mathbf{w})I^T(2\mathbf{w})\cos \mathbf{f}. \quad (2.58)$$

The third term of (2.58) is the interference term that occurs by the interference of two transmitted waves. When an experiment is performed, the third term of (2.58) is neglected (Savage, 1965; Bhanthumnavin and Lee, 1994) because of the incident surface of the ADP crystal and the exit surfaces of the transmitted harmonic wave of the crystal are not precisely parallel. Therefore there exists phase difference among the two waves. Then the integration along the optical path length of two transmitted harmonic waves in the crystal caused the average value of the interference pattern of the two transmitted beams to vanish. So the total transmitted second harmonic intensity is equal to the sum of homogeneous and inhomogeneous intensity.

$$I_{total}^T(2\mathbf{w}) = I^S(2\mathbf{w}) + I^T(2\mathbf{w}). \quad (2.59)$$

From equation (2.55), (2.56), and (2.57) the value of the term

$(\frac{c}{8\rho})e_R^{1/2}|E_0|^4(4\mathbf{p}\mathbf{c}_{36}^{NL})^2\mathbf{h}^2$  can be considered as a constant, therefore, the theoretical

value of the total transmitted second harmonic intensity is given by

$$I_{total}^T(2\mathbf{w}) \approx |F_L|^4 |F_{S,||}^{NL}|^2 \times \cos \mathbf{q}_S (\cos \mathbf{q}_i)^{-1} + |F_L|^4 |F_{T,||}^{NL}|^2 \times \cos \mathbf{q}_T (\cos \mathbf{q}_i)^{-1}. \quad (2.60)$$

### 2.2.5 The Criteria of Optimization of Second Harmonic Generation

One of the purposes of this theoretical study is searching for the appropriate condition that caused the maximum transmitted second harmonic intensity and also the minimum transmitted second harmonic intensities. We now discuss the relevant conditions.

From equation (2.10), the nonlinear polarization term that causes second harmonic generation in the crystal is

$$\bar{P}^{NLS}(2\mathbf{w}_1) = \mathbf{c}_2(\mathbf{w}_2 = 2\mathbf{w}_1)\bar{E}_1^T \bar{E}_1^T \exp i(\bar{k}^S \cdot \bar{r} - 2\mathbf{w}_1 t). \quad (2.10)$$

The electric field of second harmonic field is proportional to the following term (Yariv, 1989).

$$\bar{E}(2\mathbf{w}) \propto E_o^2 \left( \frac{\exp(i\Delta k l) - 1}{i\Delta k} \right), \quad (2.61)$$

$$\text{where } \Delta k = k^{(2w)} - 2k^{(w)} = 2\mathbf{w} \times [n^{(2w)} - n^{(w)}] / c. \quad (2.62)$$

Here,  $\Delta k$  is the difference of wave vector at  $(2\mathbf{w})$  and the wave vector at  $(\mathbf{w})$ .  $n^{(2w)}$  and  $n^{(w)}$  are the refractive index of crystal of the second-harmonic beam and the refractive index of crystal of the fundamental beam respectively.  $c$  is the velocity of light and  $l$  is the optical path length and  $E_o$  is the electric field of fundamental beam.

The intensity of second harmonic light is proportional to the following term (Yariv, 1989).

$$I(2\mathbf{w}) \propto \bar{E}^2(2\mathbf{w}) \propto (\mathbf{C}^{NL})^2 \left( \frac{\sin^2(\Delta kl/2)}{(\Delta kl/2)^2} \right) \times E_o^4, \quad (2.63)$$

where  $I(2\mathbf{w})$  is the intensity of second harmonic light. The coherence length,  $l_c$  is a measurement of the maximum crystal length that is useful in producing the second-harmonic power (Yariv, 1989).

$$l_c = \frac{2\mathbf{P}}{\Delta k} = \frac{\mathbf{P}}{\mathbf{w}[n^{2w} - n^w]} = \frac{\mathbf{I}}{2[n^{2w} - n^w]}. \quad (2.64)$$

The conditions for producing the enhancement value of  $I(2\mathbf{w})$  and  $l_c$ , by considering equations (2.63) and (2.64), could be summarized as follows

(1) From (2.63), by using the crystal which has high value of nonlinear susceptibility,  $\mathbf{C}^{NL}$ , the value of nonlinear susceptibility of ADP crystal that is used in this theoretical study is as high as the KDP crystal to generate second harmonic light.

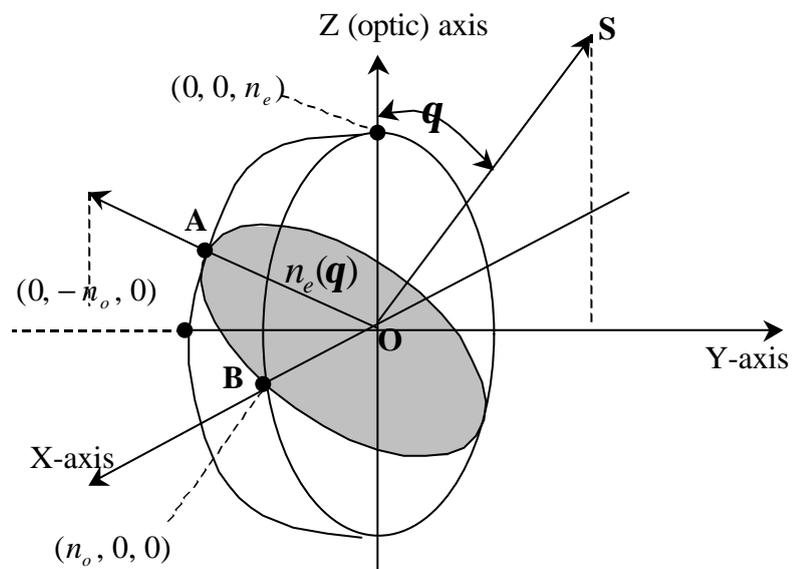
(2) From (2.63), the second harmonic intensity is proportional to fourfold of the incident electric field, so increase the amplitude or intensity of the incident light to increase the second harmonic intensity

(3) From (2.62) and (2.63), when the value of  $\Delta k$  is set to equal to zero, the value of second harmonic intensity will increase to maximum value. This technique was first demonstrated by Giordmine (1962) and Maker et al. (1962).

$$\Delta k = k^{(2w)} - 2k^{(w)} = 2\mathbf{w} \left[ n^{(2w)} - n^{(w)} \right] / c = 0. \quad (2.62)$$

Then the value of  $n^{(2w)}$  is equal to the value of  $n^{(w)}$ . This is the condition for the occurrence of phase matching and is the very important phase matching condition.

The ADP crystal used in the theoretical study is a negative uniaxial crystal. When a wave is incident on the uniaxial crystal, there are two refracted waves in the crystal. These waves depend on two refractive indices of the uniaxial crystal. The first is the extraordinary wave whose refractive index is independent of the direction of propagation ( $n_o$ ). The second is the ordinary wave whose refractive index depends on the direction of propagation ( $n_e(\mathbf{q})$ ). The characteristic of the refractive index is described by using index of ellipsoid. Figure 2.7 shows the construction for finding the value of index of refraction when the vector  $\mathbf{s}$  is the direction of propagation of wave in crystal.



**Figure 2.7.** The construction for finding the value of index of refraction when vector  $\mathbf{s}$  is the direction of propagation of wave in crystal (Yariv, 1989).

The propagation of the wave vector inside a crystal makes an angle  $\mathbf{q}$  with the optic axis (z-axis). The intersection, the ellipse shaded in figure 2.7 and perpendicular

to the direction of propagation is used to describe the characteristic of the index of refraction. The polarization of the wave along the segment OA is the extraordinary wave and corresponds to the refractive index  $n_e(\mathbf{q}) = |OA|$ . The polarization of wave along the segment OB is the ordinary wave and corresponds to the refractive index propagation  $n_o = |OB|$ . From figure 2.7, the equation of index of ellipsoid on z-y plane is.

$$\frac{y^2}{n_o^2} + \frac{z^2}{n_e^2} = 1. \quad (2.65)$$

From figure 2 we have following relations

$$n_e^2(\mathbf{q}) = z^2 + y^2, \quad (2.66)$$

$$\frac{z}{n_e(\mathbf{q})} = \sin \mathbf{q}, \quad \frac{y}{n_e(\mathbf{q})} = \cos \mathbf{q}. \quad (2.67)$$

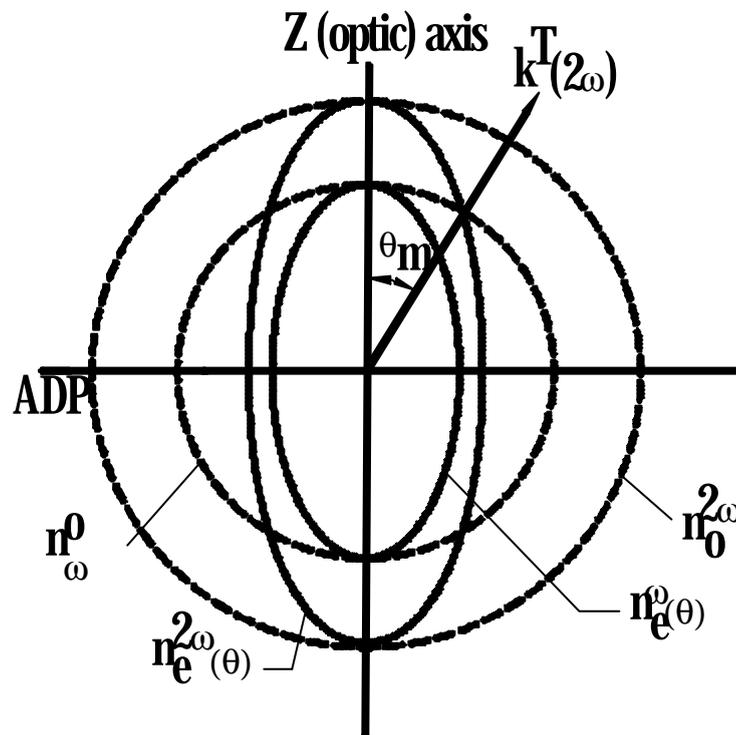
From equation (32), (33), and (34) the value of  $n_e(\mathbf{q})$  is given by

$$\frac{1}{n_e^2(\mathbf{q})} = \frac{\cos^2(\mathbf{q})}{n_o^2} + \frac{\sin^2(\mathbf{q})}{n_e^2}. \quad (2.68)$$

For this case, consider the negative uniaxial crystal with refractive index of ordinary wave greater than the refractive index of the extraordinary wave ( $n_e < n_o$ ). The incident wave will propagate along the direction of ordinary wave and the second harmonic wave will propagate along the direction of extraordinary wave. In order to yield the condition of phase matching, the value of  $n_o^w$  must be equal to the value of  $n_e^{2w}(\mathbf{q}_m)$ . The phase matching angle,  $(\mathbf{q}_m)$  was the angle between homogeneous transmitted second harmonic wave,  $\vec{k}^T(2\mathbf{w})$  and the optic axis of the crystal that

caused the value of  $n_e(\mathbf{q}_m) = n_o$ . Figure 2.8. From (2.68), the phase matching angle is given by

$$\mathbf{q}_m = \sin^{-1} \frac{\hat{\mathbf{e}}(n_o^w)^{-2} - (n_o^{2w})^{-2} \hat{\mathbf{u}}}{\hat{\mathbf{e}}(n_e^{2w})^{-2} - (n_o^{2w})^{-2} \hat{\mathbf{u}}} \cdot \hat{\mathbf{u}} \quad (2.69)$$



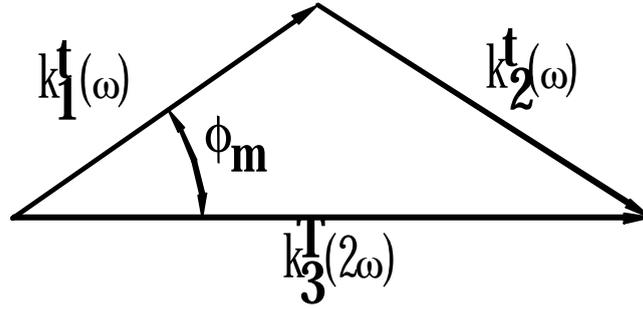
**Figure 2.8.** The condition for the occurrence of colinear phase matching

For the case of noncolinear phase matching, two incident waves, lie in the same plane of incidence, incident on the crystal at the opposite directions and make the same angle of incidence with the face normal (Figure 2.3). The same two incident wave vectors are represented by  $\bar{k}_1$  and  $\bar{k}_2$ . The incident angle of two incident wave are  $\mathbf{q}_1^i$  and  $\mathbf{q}_2^i$  respectively. This incident case is the case of TBSM. From (2.20) and

figure 2.5, if  $\mathbf{q}_1^i = \mathbf{q}_2^i$  the results of the summation of two waves vector,  $\bar{k}_3$  will propagate along the direction of face normal. The condition of noncolinear phase matching occur when the summation of two transmitted waves vector,  $\bar{k}_1^t(\boldsymbol{\omega}) = \bar{k}_2^t(\boldsymbol{\omega})$ , is equal to  $\bar{k}_3^T(2\boldsymbol{\omega})$  and is given by, (Figure 2.9)

$$\bar{k}_3^T(2\boldsymbol{\omega}) = \bar{k}_1^t(\boldsymbol{\omega}) + \bar{k}_2^t(\boldsymbol{\omega}),$$

$$\text{or } \Delta \vec{k} = \bar{k}_1^t(\boldsymbol{\omega}) + \bar{k}_2^t(\boldsymbol{\omega}) - \bar{k}_3^T(2\boldsymbol{\omega}) = 0. \quad (2.70)$$



$$k_1^t(\omega) = k_2^t(\omega)$$

**Figure.2.9** The occurrence of noncolinear phase matching

From figure 2.3, in order to find out the incident angle that caused noncolinear phase matching angle, the same method for analysis for the colinear phase matching angle will be used. In the negative uniaxial crystal (ADP crystal), the incident wave will propagate along the direction of ordinary wave and the second harmonic wave

will propagate along the direction of extraordinary wave. From (2.70), The results of the summation of two wave vectors is given by the normal components of the results:

$$\frac{n_e^{2w}}{c} 2\mathbf{w}\cos \mathbf{q}_3 = \frac{n_o^w}{c} \mathbf{w}\cos(\mathbf{q}_1 + \mathbf{q}_3) + \frac{n_o^w}{c} \mathbf{w}\cos(\mathbf{q}_2 - \mathbf{q}_3), \quad (2.71)$$

and the tangential component of the results

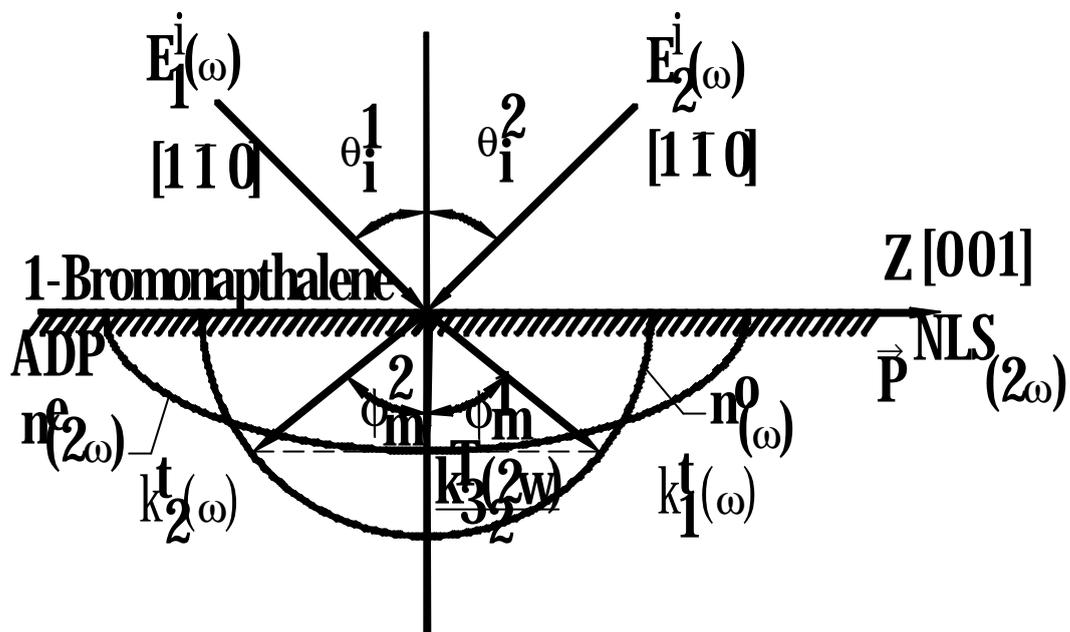
$$\frac{n_e^{2w}}{c} 2\mathbf{w}\sin \mathbf{q}_3 = \frac{n_o^w}{c} \mathbf{w}\sin(\mathbf{q}_1 + \mathbf{q}_3) + \frac{n_o^w}{c} \mathbf{w}\sin(\mathbf{q}_2 - \mathbf{q}_3), \quad (2.72)$$

where  $k(\mathbf{w}) = \frac{n_o^w \mathbf{w}}{c}$  and  $k(2\mathbf{w}) = \frac{n_e^{2w} 2\mathbf{w}}{c}$ . From (2.20), if the value of the angle  $\mathbf{q}_3$  is equal to zero, the results of the summation of two waves vector will propagate along the direction of face normal. Therefore from (2.40), the result is given by

$$n_e^{2w} = n_o^w \cos \mathbf{f}_m, \quad (2.73)$$

where  $\mathbf{f}_m$  is the noncolinear phase matching angle and is refracted angle of the wave vectors  $\bar{k}_1^t(\mathbf{w})$  and  $\bar{k}_2^t(\mathbf{w})$ .

From equation (2.42), the crystallographic orientation of the negative uniaxial crystal (ADP crystal) for the noncolinear phase matching condition of this theoretical study to occur is shown in figure 2.10.



**Figure 2.10** The crystallographic orientation of the theoretical study, which caused noncolinear phase matching.

# Chapter III

## Procedure

### 3.1 Introduction

The transmitted second harmonic generation of wavelength 450 nm in Ammonium Dihydrogen Phosphate, ADP crystal immersed in an optically denser liquid 1-Bromonaphthalene will be theoretically studied and analyzed by using ultra short pulse laser at wavelength equal 900 nm as the incident beam. The Bloembergen and Pershan theory (1962) is used as the main theory for analysis. The results of transmitted second harmonic intensity, as a function of the incident angle will be calculated by the application of C++ language and simulated by the Microsoft Excel Version 7. In order to perform the theoretical study, the technical data of the ADP crystal and the liquid 1-Bromonaphthalene are necessary for investigation of  $I^T(2\mathbf{w})$  and  $I^S(2\mathbf{w})$ .

### 3.2 Ammonium Dihydrogen Phosphate, ADP Crystal

The ADP crystal has transparent range at wavelength from 184 nm to 1500 nm. (Johnson and Duordo, 1967; Cimerl, 1987), thus there are transparencies at the fundamental and second harmonic wavelengths. In addition the crystal has high damage threshold then so that it is suitable for the high peak power of laser pulse. The crystal is a negative uniaxial crystal with two values of refractive index: the ordinary

index,  $n_o$  and extraordinary ray index,  $n_e$ , for which  $n_e < n_o$ . The refractive indices of ADP crystal at the fundamental and second harmonic wavelengths of 900 nm and 450 nm respectively, are calculated to be the following: (Zernike ,1964a; 1964b).

At the wavelength equal 900 nm:

$$n_o^w = 1.5120, \quad n_e^w = 1.4709$$

At the wavelength equal 450 nm:

$$n_o^{2w} = 1.5343, \quad n_e^{2w} = 1.4870$$

(The details of calculations are given in appendix A.)

The optimization of second harmonic generation to obtain the maximum transmitted second harmonic intensities in the ADP crystal was successfully performed by the phase matching technique (Giordmaine, 1962; Maker et al., 1962, Bloembergen and Lee, 1967). The occurrence of the phase matching condition could be considered in the case of colinear incidence and noncolinear incidence.

For colinear incidence, the colinear phase matching angle,  $\mathbf{q}_m$ , causing the condition of colinear phase matching ( $n_e^{2w} = n_o^w$ ) is given by (2.68) and (2.69)

$$\frac{1}{n_e^{2w}(\mathbf{q}_m)} = \frac{1}{n_o^w} = \frac{\cos^2 \mathbf{q}_m}{(n_o^{2w})^2} + \frac{\sin^2 \mathbf{q}_m}{(n_e^{2w}[\mathbf{p}/2])^2}, \text{ and} \quad (2.68)$$

$$\mathbf{q}_m = \sin^{-1} \left[ \frac{(n_o^w)^{-2} - (n_o^{2w})^{-2}}{(n_e^{2w}[\mathbf{p}/2])^{-2} - (n_o^{2w})^{-2}} \right]. \quad (2.69)$$

In case of noncolinear incidence, the noncolinear phase matching angle,  $\mathbf{f}_m$  is given by (2.42) as the following

$$n_e^{2w} = n_o^w \cos \mathbf{f}_m, \text{ and}$$

$$\mathbf{f}_m = \cos^{-1} \left[ \frac{n_e^{2w}}{n_o^w} \right]. \quad (2.42)$$

The ADP crystal is a piezoelectric crystal with the same crystal point group,  $4\bar{2}m$  as the KDP crystal. Therefore, theoretical results could be compared with the previous experimental work of transmitted second harmonic generation from KDP crystal (Bhanthumnavin and Lee, 1990; 1994a; 1994b; Lee and Bhanthumnavin, 1976). Furthermore, the ADP crystal has high values of the nonlinear susceptibility tensor, which could increase second harmonic intensity, and the crystal is readily available. Thus, these crystal properties will be useful when the experiment will verify the theoretical study. From the mentioned properties, ADP crystal is the suitable one of the negative uniaxial crystals to be used for both theoretical and experimental studies of transmitted second harmonic generation based on the theory of Bloembergen and Pershan (1962).

### 3.3 Fluid 1-Bromonaphthalene

The theoretical study of transmitted second harmonic generation in ADP crystal could be directly performed from ADP crystal in air. When the incident light wave propagate from optically denser medium to the lower medium, the total reflection phenomenon could be observed. In order to establish a connection with preliminary research of reflected second harmonic generation at total reflection as mentioned in chapter one, the optically denser fluid 1-bromonaphthalene will be used in this theoretical study. The liquid is not corrosive to the crystal and is transparent at the wavelength of fundamental and second harmonic beams. Further it is stable when the temperature is varied and its vapor is not dangerous to human health. Furthermore,

the liquid will prevent the humidity in air to dissolve the crystal surface. Therefore, the fluid 1-bromonaphthalene was chosen because of its properties above.

The refractive indices of the optically denser liquid 1-bromonaphthalene,  $n_L$  at wavelength equal 900 nm and 450 nm are calculated by using Cauchy equation (Jenkins and White, 1976) and are given by

At wavelength equal 900 nm:

$$n_{liq}^w = 1.6355$$

At wavelength equal 450 nm:

$$n_{liq}^{2w} = 1.6952$$

(The details of calculations are given in appendix A.)

### 3.4 Computer Program

In the study, C++ (Turbo C++ Version 3) is used to calculate the theoretical value of total transmitted second harmonic intensity. The results are plotted and illustrated by Excel Version 7. The procedures of the program are shown by the flowchart and illustrated in figure 3.1

From flowchart, the upper part of the program will give the total information used in the program such as the crystallographic orientation of the crystal, the values and the definition of the variables used in the program. The main procedure for calculation of total transmitted second harmonic generation of colinear incidence is the same as noncolinear incidence shown in the flowchart. At the beginning of the program, the initial values of the variables such as the starting transmitted angle, the ending transmitted angle, and the increment of the transmitted angle will be inputted.

Then the data of the initial values will be used in the computing process (Figure 3.2). The process of calculation begins by using the value of starting transmitted angle,  $\mathbf{q}_T$  to discover the angle between homogeneous transmitted wave vector,  $\bar{k}^T(2\mathbf{w})$  and the optic axis of the crystal,  $\mathbf{q}$ . Because of the wave vector,  $\bar{k}^T(2\mathbf{w})$  is defined to propagate along the direction of the extraordinary ray, thus  $n_e^{2w}$  is depends on the angle,  $\mathbf{q}$ . The refractive index,  $n_e^{2w}$  is calculated as given by (2.68)

$$n_e^{2w}(\mathbf{q}) = \left[ \frac{1}{\frac{\cos^2 \mathbf{q}}{(n_e^{2w})^2} + \frac{\sin^2 \mathbf{q}}{(n_e^{2w}(\mathbf{p}/2))^2}} \right]^{1/2}. \quad (2.68)$$

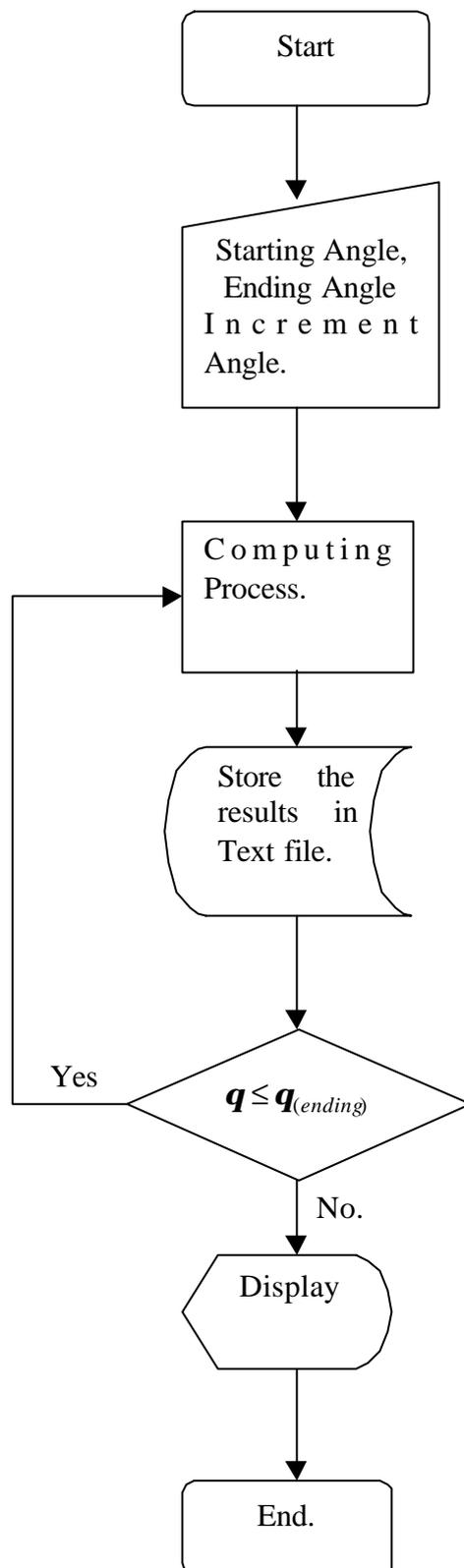
The transmitted angle of inhomogeneous wave vector ( $\bar{k}^S(2\mathbf{w})$ ),  $\mathbf{q}_S$ , the reflected harmonic wave vector ( $\bar{k}^R(2\mathbf{w})$ ),  $\mathbf{q}_R$ , and the fundamental incident angle beam  $\mathbf{q}_i$  are then calculated by the relation of nonlinear Snell's law as given by (2.50).

$$n_e^{2w}(\mathbf{q}) \sin \mathbf{q}_T = n_L^w(\mathbf{q}) \sin \mathbf{q}_i = n_L^w(\mathbf{q}) \sin \mathbf{q}_R = n_e^w(\mathbf{q}) \sin \mathbf{q}_S. \quad (2.50)$$

Now, the values of the angles and the refractive index will be used to calculate the linear fresnel factor,  $F_{\perp}^L$  and nonlinear fresnel factors,  $F_{//}^{NL}$  by using equation (2.52), (2.36), (2.37), and (2.38), respectively.

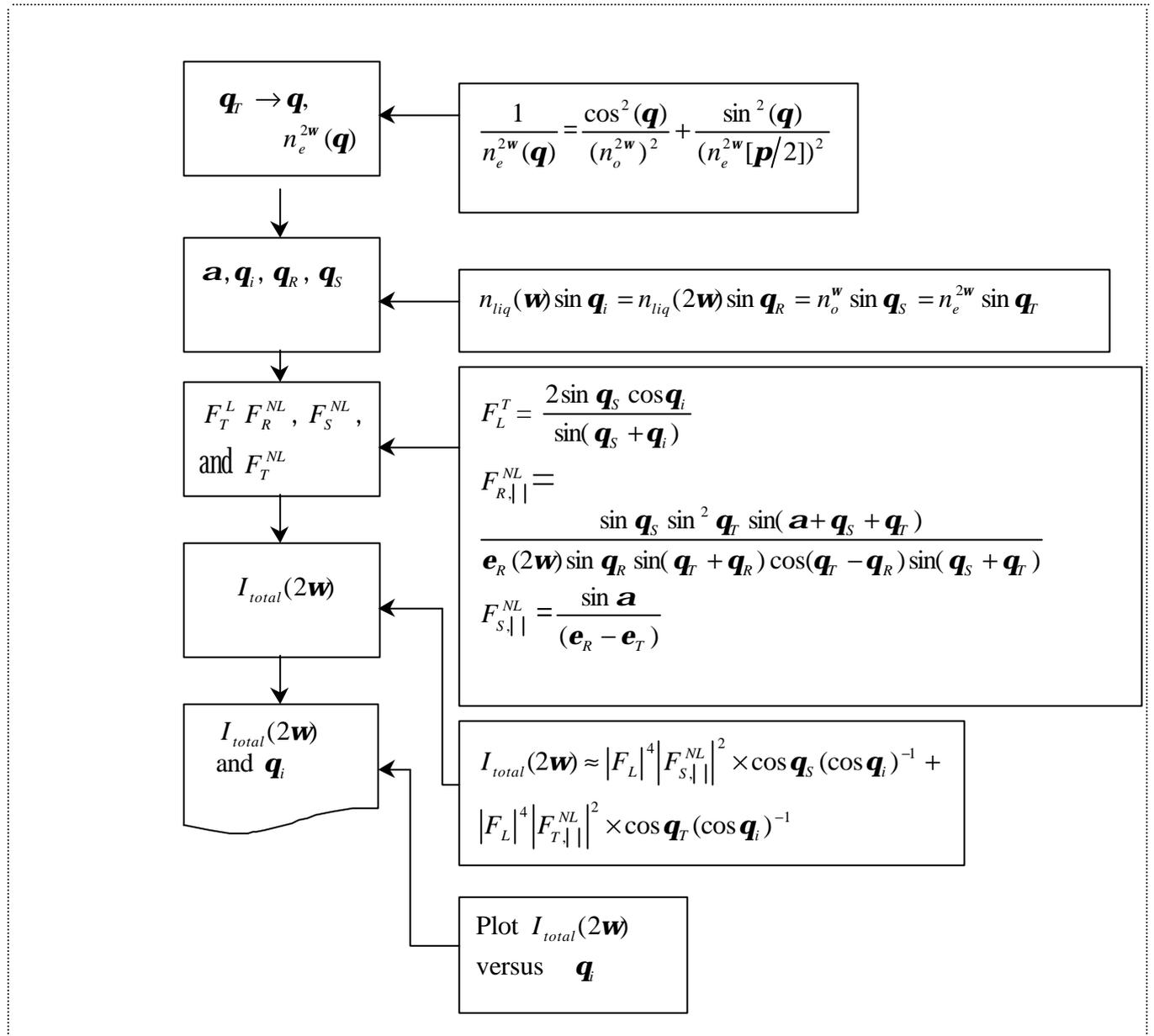
$$F_{L,\perp}^T = \frac{2 \sin \mathbf{q}_s \cos \mathbf{q}_i}{\sin(\mathbf{q}_s + \mathbf{q}_i)}, \quad (2.52)$$

$$F_{R, //}^{NL} = \frac{\sin \mathbf{q}_s \sin^2 \mathbf{q}_T \sin(\mathbf{a} + \mathbf{q}_s + \mathbf{q}_T)}{\mathbf{eR} \sin \mathbf{q}_R \sin(\mathbf{q}_s + \mathbf{q}_T) \sin(\mathbf{q}_T + \mathbf{q}_R) \cos(\mathbf{q}_T - \mathbf{q}_R)}, \quad (2.36)$$



**Figure 3.1.** The Flowchart of C++ program for the calculation of transmitted second harmonic intensity.

### Computing Process



**Figure 3.2.** The computing process of the C++ program for calculation of transmitted second harmonic intensity.

$$F_{S, //}^{NL} = \frac{\sin \mathbf{a}}{\mathbf{e}_s - \mathbf{e}_t}, \quad (2.37)$$

$$F_{T, //}^{NL} = -\frac{\mathbf{e}_S^{1/2}}{\mathbf{e}_T^{1/2}} \frac{\sin \mathbf{a}}{\mathbf{e}_S - \mathbf{e}_T} + \frac{\mathbf{e}_R^{1/2}}{\mathbf{e}_T^{1/2}} F_{R, //}^{NL}, \quad (2.38)$$

where  $\mathbf{e}_s = (n_o^w)^2$ ,  $\mathbf{e}_t = (n_e^{2w}(\mathbf{q}))^2$ , and  $\mathbf{e}_R = (n_{liq}^{2w})^2$ .

All data will then be used to calculate the total transmitted second harmonic intensity as a function of specified incident angle for the colinear incident case by:

$$I_{total}^T(2\mathbf{w}) \approx |F_L|^4 |F_{S, //}^{NL}|^2 \times \cos \mathbf{q}_s (\cos \mathbf{q}_i)^{-1} + |F_L|^4 |F_{T, //}^{NL}|^2 \times \cos \mathbf{q}_t (\cos \mathbf{q}_i)^{-1}. \quad (2.60)$$

In the case of noncolinear incidence or Two Beam Spatial Mixing, the total transmitted second harmonic intensity can also be calculated from equation (2.60) used in the colinear incident case. The process of calculation will only differ at the values of ordinary ray index,  $n_o^w$  and the permittivity,  $\mathbf{e}_s$  equal to  $n_o^w \cos \mathbf{q}_s$  and  $(n_o^w \cos \mathbf{q}_s)^2$  respectively as indicated in chapter two.

Data of the total transmitted second harmonic intensities at each incident angle will then be plotted by using Microsoft Excel Version 7. (The detailed of C++ is given in appendix B.).

# Chapter IV

## Results and Discussion

### 4.1 Introduction

In the study, the incident laser is polarizing in  $[1\bar{1}0]$  direction with respect to crystallographic axis of ADP crystal and perpendicular to the plane of incidence. The nonlinear polarization  $\bar{P}^{NLS}(2\mathbf{w})$  will occur in the  $z$  direction, is the optic axis and parallel to the plane of incidence. The results of second harmonic generation in transmission at any orientation of the ADP crystal will be illustrated in this chapter. The occurrence of maximum transmitted second harmonic intensity at colinear and noncolinear phase matching conditions and also the occurrence of minimum and null transmitted second harmonic intensity will be shown here.

### 4.2 Minimum Transmitted Second Harmonic Generation

The first nonlinear optical phenomenon was second harmonic generation in transmission (Franken et al., 1961). The condition that causes the minimum transmitted second harmonic intensity will be illustrated instead of the phase matching condition that caused the maximum value of transmitted second harmonic intensity as shown in chapter two. As generally understood, when laser light is incident on the nonlinear crystal, the transmitted second harmonic light is usually always generated in the crystal. But for some specific crystallographic orientation of

the nonlinear crystal and the angle of incidence, the null transmitted second harmonic light was experimentally observed (Bhanthumnavin and Lee, 1990; 1994).

#### 4.2.1 Null Transmitted Second Harmonic Generation

The ADP crystal is prepared with  $\bar{P}^{NLS}(2\mathbf{w})$  lying along the optic axis, which is in the direction of the face normal as shown in the inset of figure 4.1. The variation of total transmitted second harmonic intensity is observed near the zero value of the incident angle and the null transmitted second harmonic intensity occurred at this normal incident angle. The value of total transmitted second harmonic intensity is given by (2.60)

$$I_{total}^T(2\mathbf{w}) \approx |F_L|^4 |F_{S,||}^{NL}|^2 \times \cos \mathbf{q}_s (\cos \mathbf{q}_i)^{-1} + |F_L|^4 |F_{T,||}^{NL}|^2 \times \cos \mathbf{q}_t (\cos \mathbf{q}_i)^{-1}, \quad (2.60)$$

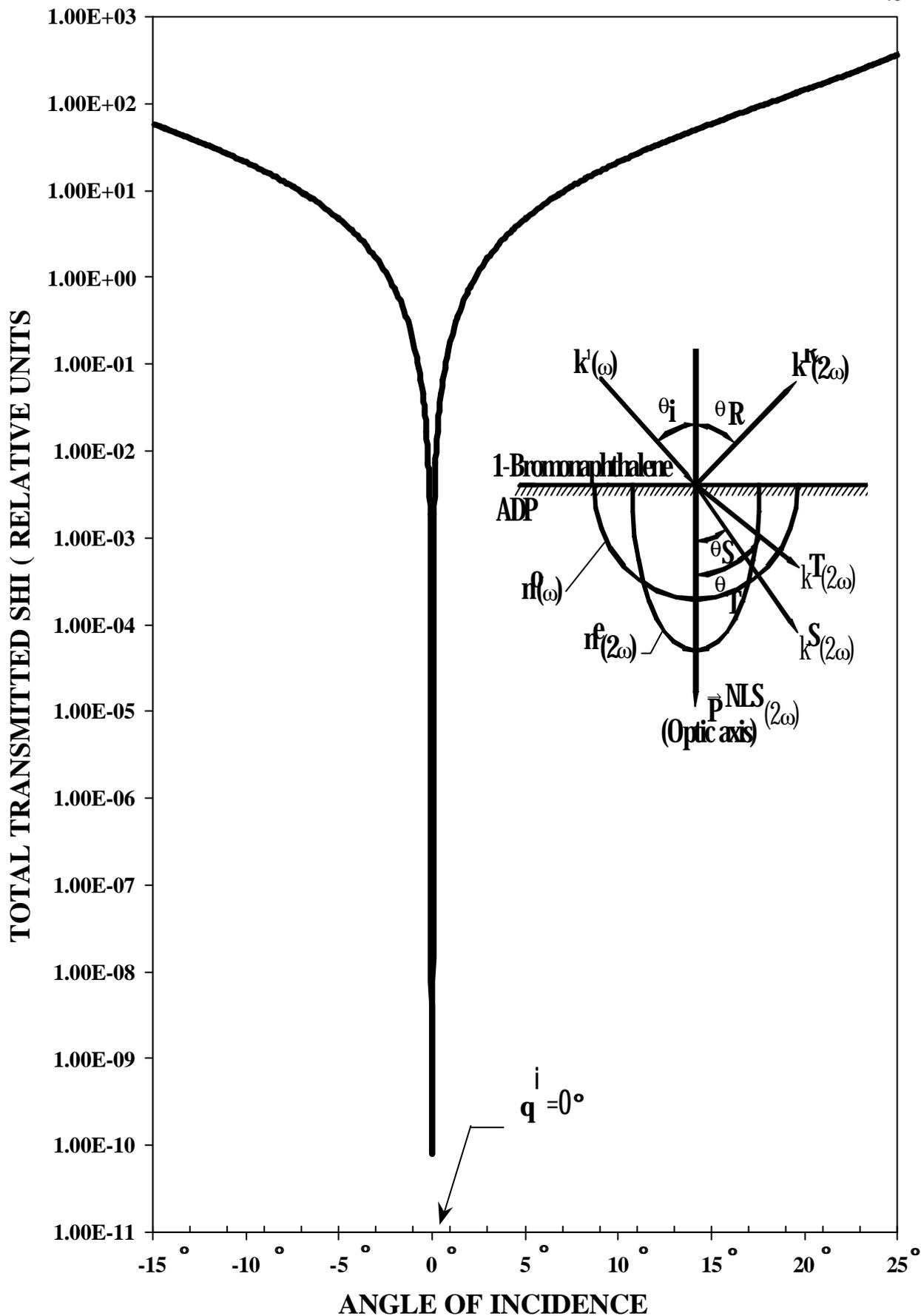
where  $|F_{L,\perp}^T|$ ,  $|F_{S,||}^{NL}|$ ,  $|F_{T,||}^{NL}|$  are linear, nonlinear fresnel factors of inhomogeneous and homogeneous wave vector, which are given by (2.52), (2.37), and (2.38) respectively.

$$F_L^T = \frac{2 \sin \mathbf{q}_s \cos \mathbf{q}_i}{\sin(\mathbf{q}_s + \mathbf{q}_i)}, \quad (2.52)$$

$$F_{S,||}^{NL} = \frac{\sin \mathbf{a}}{(\mathbf{e}_R - \mathbf{e}_T)}, \quad (2.37)$$

$$F_{T,||}^{NL} = \frac{\mathbf{e}_S^{\frac{1}{2}} \sin \mathbf{a}}{\mathbf{e}_T^{\frac{1}{2}} (\mathbf{e}_R - \mathbf{e}_T)} + \frac{\mathbf{e}_R^{\frac{1}{2}}}{\mathbf{e}_T^{\frac{1}{2}}} F_{R,||}^{NL}, \quad (2.38)$$

$$\text{where } F_{R,||}^{NL} = \frac{\sin \mathbf{q}_s \sin^2 \mathbf{q}_t \sin(\mathbf{a} + \mathbf{q}_s + \mathbf{q}_t)}{\mathbf{e}_R(2\mathbf{w}) \sin \mathbf{q}_R \sin(\mathbf{q}_T + \mathbf{q}_R) \cos(\mathbf{q}_T - \mathbf{q}_R) \sin(\mathbf{q}_s + \mathbf{q}_T)}. \quad (2.39)$$



**Figure 4.1** The null transmitted second harmonic intensity (SHI) at the angle of incidence ( $\theta^i$ ) equal  $0^\circ$ . The nonlinear polarization  $\vec{P}^{NLS}(2\omega)$  lies along face normal.

The values of reflected, transmitted angle at normal incident angle and the angle  $\mathbf{a}$ , the angle between the source transmitted wave vector,  $k^S$  and  $\bar{P}^{NLS}(2\mathbf{w})$  are equal to zero. Then equation of nonlinear fresnel factor is rewritten as

$$F_{T,||}^{NL} = \frac{\mathbf{e}_S^{1/2} \sin \mathbf{a}}{\mathbf{e}_T^{1/2} (\mathbf{e}_R - \mathbf{e}_T)} \quad ; (\mathbf{a}=0)$$

$$+ \frac{\mathbf{e}_R^{1/2}}{\mathbf{e}_T^{1/2}} \frac{\sin \mathbf{q}_X \sin^2 \mathbf{q}_X \sin(\mathbf{q}_X + \mathbf{q}_X)}{\mathbf{e}_R(2\mathbf{w}) \sin \mathbf{q}_X \sin(\mathbf{q}_X + \mathbf{q}_X) \cos(\mathbf{q}_X - \mathbf{q}_X) \sin(\mathbf{q}_X + \mathbf{q}_X)} = 0$$

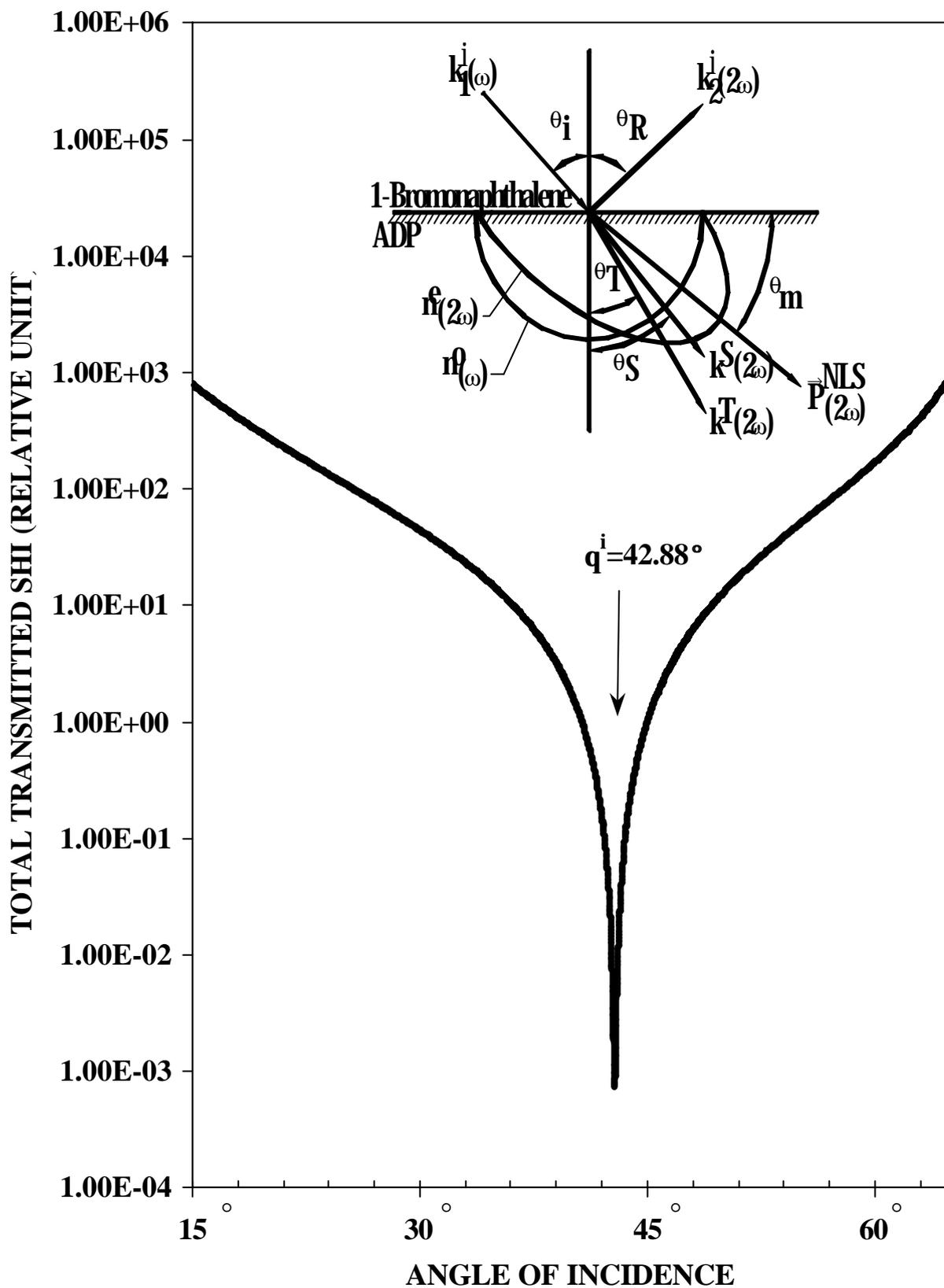
$$F_{T,||}^{NL} = \frac{\mathbf{e}_R^{1/2} \sin \mathbf{q}_X}{\mathbf{e}_T^{1/2} \cos \mathbf{q}_X} = 0 \quad ; (\mathbf{q}_X = 0)$$

From above, the values of  $F_{S,||}^{NL}$  and  $F_{T,||}^{NL}$  are equal to zero respectively. Therefore the value of total transmitted second harmonic intensity as given by (2.60) is equal to zero at normal incident angle. This result agrees well when compared with the result of the previous experimental work in the KDP crystal, which has the same crystal point group as ADP crystal, under the same crystal orientation (Bhantumnavin and Lee, 1990; 1994).

#### 4.2.2 Minimum Transmitted Second Harmonic Generation when $\bar{P}^{NLS}(2\mathbf{w})$

##### Makes Phase Matching Angle with the Crystal Surface.

At this condition, the ADP crystal has nonlinear polarization,  $\bar{P}^{NLS}(2\mathbf{w})$  lying along optic axis and makes phase matching angle with the boundary as shown by the inset of figure 4.2. The minimum value of total transmitted second harmonic intensity that occurs, instead of the null transmitted harmonic intensity as shown in figure 4.1, at the angle of incidence equal to  $42.88^\circ$ . From (2.59), the value of total transmitted



**Figure 4.2** The minimum transmitted second harmonic intensity (SHI) at the angle of incidence ( $\theta^i$ ) equal  $42.88^\circ$ . The nonlinear polarization  $\bar{P}^{NLS}(2\omega)$  makes phase matching angle with the crystal surface.

second harmonic intensity is the summation of  $I^S(2\mathbf{w})$  and  $I^T(2\mathbf{w})$  so implying that the minimum value of  $I^S(2\mathbf{w})$  and  $I^T(2\mathbf{w})$  does not occur at the same angle of incidence. By means of dipole radiation, there is no radiation of waves in the direction of oscillation of polarization. If consider the condition of the values of  $I^S(2\mathbf{w})$  will be equal to zero by (2.56) and (2.37). The value of the angle  $\mathbf{a}$  must equal to zero. This means that the direction of the source wave vector  $\vec{k}^S$  is same as the direction of nonlinear polarization  $\vec{P}^{NLS}(2\mathbf{w})$ . This condition agrees well with the dipole radiation theory. Further, the value of transmitted angle  $\mathbf{q}_s$  is equal to  $47.31^\circ$ . Its value is not equal to the transmitted angle  $\mathbf{q}_t$  because of the different values of the refractive index. The direction of homogeneous wave vector  $\vec{k}^T(2\mathbf{w})$  is not parallel to the direction of  $\vec{P}^{NLS}(2\mathbf{w})$ . However the small value of  $I^T(2\mathbf{w})$  occurs because of the first term of the right side of (2.38) is equal to zero. So from (2.57), the value of  $I_{total}^T(2\mathbf{w})$  is small but not equal to zero at this condition. Then the incident angle that causes the zero values of  $I^S(2\mathbf{w})$  is calculated by Snell's law:

$$\begin{aligned}
 n_{iq}^w \sin \mathbf{q}_i &= n_o^w \sin \mathbf{q}_s, \\
 \mathbf{q}_i &= \sin^{-1} \left( \frac{n_o^w \sin \mathbf{q}_s}{n_{iq}^w} \right), \\
 \mathbf{q}_i &= \sin^{-1} \left( \frac{1.512 \times \sin 47.31}{1.6355} \right) = 42.80^\circ. \tag{4.1}
 \end{aligned}$$

Later consider the value of  $I^T(2\mathbf{w})$  by using (2.57). By means of dipole radiation, the minimum or zero values of  $I^T(2\mathbf{w})$  should occur when the direction of the homogeneous wave vectors  $\vec{k}^T$  is parallel to the direction of  $\vec{P}^{NLS}(2\mathbf{w})$ . Under

this condition the direction of the source wave vector,  $\bar{k}^s$  is not parallel to  $\bar{P}^{NLS}(2\mathbf{w})$  because of the different value of the index of refraction. Thus the value of the angle  $\mathbf{a}$  is not equal to zero. Therefore, from (2.57) and (2.38), the zero value of  $I^T(2\mathbf{w})$  can not occur. However, the small value of the angle  $\mathbf{a}$  leads to the small value of  $I^T(2\mathbf{w})$ . The value of the angle  $\mathbf{q}_T$  at this condition is equal to  $47.31^\circ$  and the value of  $n_e^{2w}(\mathbf{q}) = n_e^{2w}(0^\circ) = n_o^{2w}$  is equal to 1.5343. So the incident angle is given by

$$n_{iq}^w \sin \mathbf{q}_i = n_e^{2w}(\mathbf{q}) \sin \mathbf{q}_T,$$

$$\mathbf{q}_i = \sin^{-1} \left( \frac{n_e^{2w}(0^\circ) \sin \mathbf{q}_T}{n_{iq}^w} \right),$$

$$\mathbf{q}_i = \sin^{-1} \left( \frac{1.5343 \times \sin 47.31}{1.6355} \right) = 43.59^\circ. \quad (4.2)$$

Therefore, from (4.1) and (4.2), the values of the angle of incidence that caused the minimum values of total harmonic intensity is found to occur between the value of  $\mathbf{q}_i$  equal  $42.80^\circ$  to  $43.59^\circ$ . Further, the tendency to decrease of the values of  $I_{total}^T(2\mathbf{w})$  is slowly than the results of the occurrence of null transmitted harmonic intensity of item 4.2.1. The result agrees well with the result of the experimental work that performed with the KDP crystal under the same crystallographic orientation (Bhanthumnavin and Lee, 1994).

### 4.3 Transmitted Second Harmonic Generation under Phase

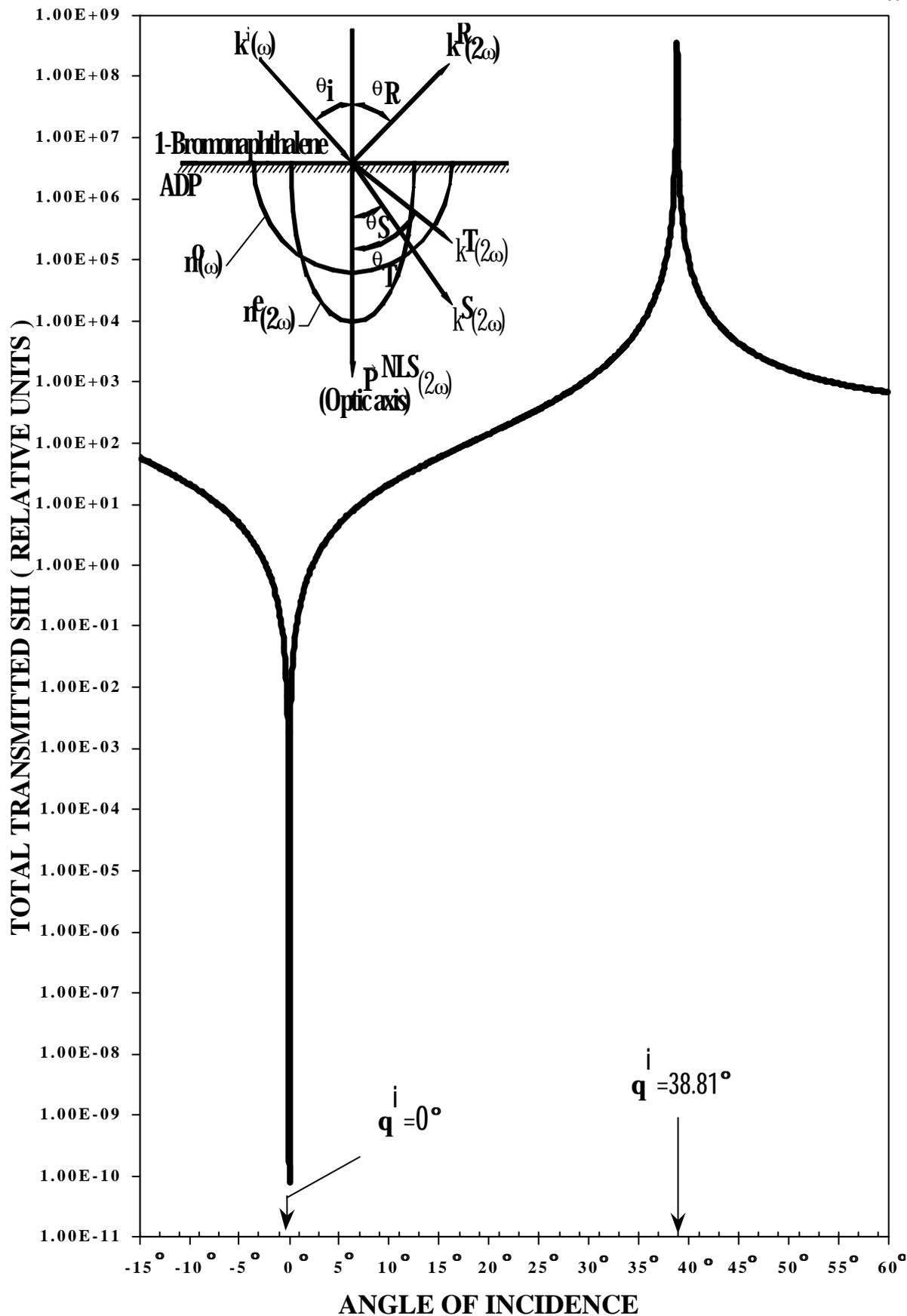
#### Matching Condition.

The occurrence of maximum transmitted second harmonic intensity could be achieved under phase matching condition as shown in chapter two. In order to yield this condition, the values of the refractive index of  $n_e$  must equal to  $n_o$ . This study, the transmitted inhomogeneous source wave vector  $\vec{k}^S(2\mathbf{w})$  and the homogeneous transmitted harmonic wave vector  $\vec{k}^T(2\mathbf{w})$  are the ordinary and extraordinary light that depend on the refractive index  $n_o^w$  and  $n_e^{2w}$  respectively. Therefore, the same values of  $n_o^w$  and  $n_e^{2w}$  is considered at phase matching condition. The result of total transmitted second harmonic intensity under colinear and noncolinear phase matching at any orientations of ADP crystal will be illustrated in this topic.

#### 4.3.1 The Maximum Transmitted Second Harmonic Generation under

##### Colinear Phase Matching Condition when $\vec{P}^{NLS}(2\mathbf{w})$ Lies Along Face Normal.

The ADP crystal was cut to have the direction of the optic axis and the nonlinear polarization  $\vec{P}^{NLS}(2\mathbf{w})$  lie along face normal as same as the orientation of the ADP crystal of item 4.2.1. At this orientation not only providing the null but also the maximum transmitted second harmonic intensity as shown by figure 4.3. The tendency of the values of total transmitted second harmonic intensity is increase near the region of the values of the incident angle equal  $38.81^\circ$  and reach to the maximum



**Figure 4.3** The transmitted second harmonic intensity (SHI) as a function of the angle of incidence. The nonlinear polarization  $\vec{P}^{NLS}(2\omega)$  lies along face normal.

value at this angle. The maximum values of  $I_{total}^T(2\mathbf{w})$  must be occurred under phase matching condition where the value of the refractive index  $n_e^{2w}$  is equal  $n_o^w$  as the propagation direction of wave vector  $\bar{k}^s$  is the same as  $\bar{k}^T$ . This condition lead to the equivalence of the value of the angle  $\mathbf{q}_s = \mathbf{q}_T$ . Its value equal to the value of phase matching angle inside the crystal that equal to  $42.69^\circ$ . So the value of the refractive indices equal to  $n_e^{2w}(\mathbf{q}_m) = n_e^{2w}(42.69) = n_o^w = 1.5120$ . Then from Snell's law, the value of the incident angle is given by

$$n_{liq}^w \sin \mathbf{q}_i = n_e^{2w}(\mathbf{q}_m) \sin \mathbf{q}_T$$

$$\mathbf{q}_i = \sin^{-1} \left( \frac{[n_e^{2w}(\mathbf{q}_m) = n_o^w] \times \sin \mathbf{q}_T}{n_L^w} \right)$$

$$\mathbf{q}_i = \sin^{-1} \left( \frac{1.5120 \times \sin 42.69^\circ}{1.6355} \right) = 38.81^\circ$$

From above, the result confirmed that the maximum values of transmitted second harmonic intensity would be occurred at colinear phase matching condition. This agrees well with the BP theory and the experimental work that performed under the same orientation with KDP crystal (Bhanthumnavin and Lee, 1990; 1994).

#### 4.3.2 The Maximum transmitted Second Harmonic Generation under

##### Colinear Phase Matching Condition when $\bar{P}^{NLS}(2\mathbf{w})$ Makes Phase Matching Angle with the Crystal Surface.

In this case, the orientation of the crystal is the same as was discussed in item 4.2.2, so that the direction of nonlinear polarization  $\bar{P}^{NLS}(2\mathbf{w})$  makes phase matching angle with the boundary. Not only the minimum but also the maximum transmitted

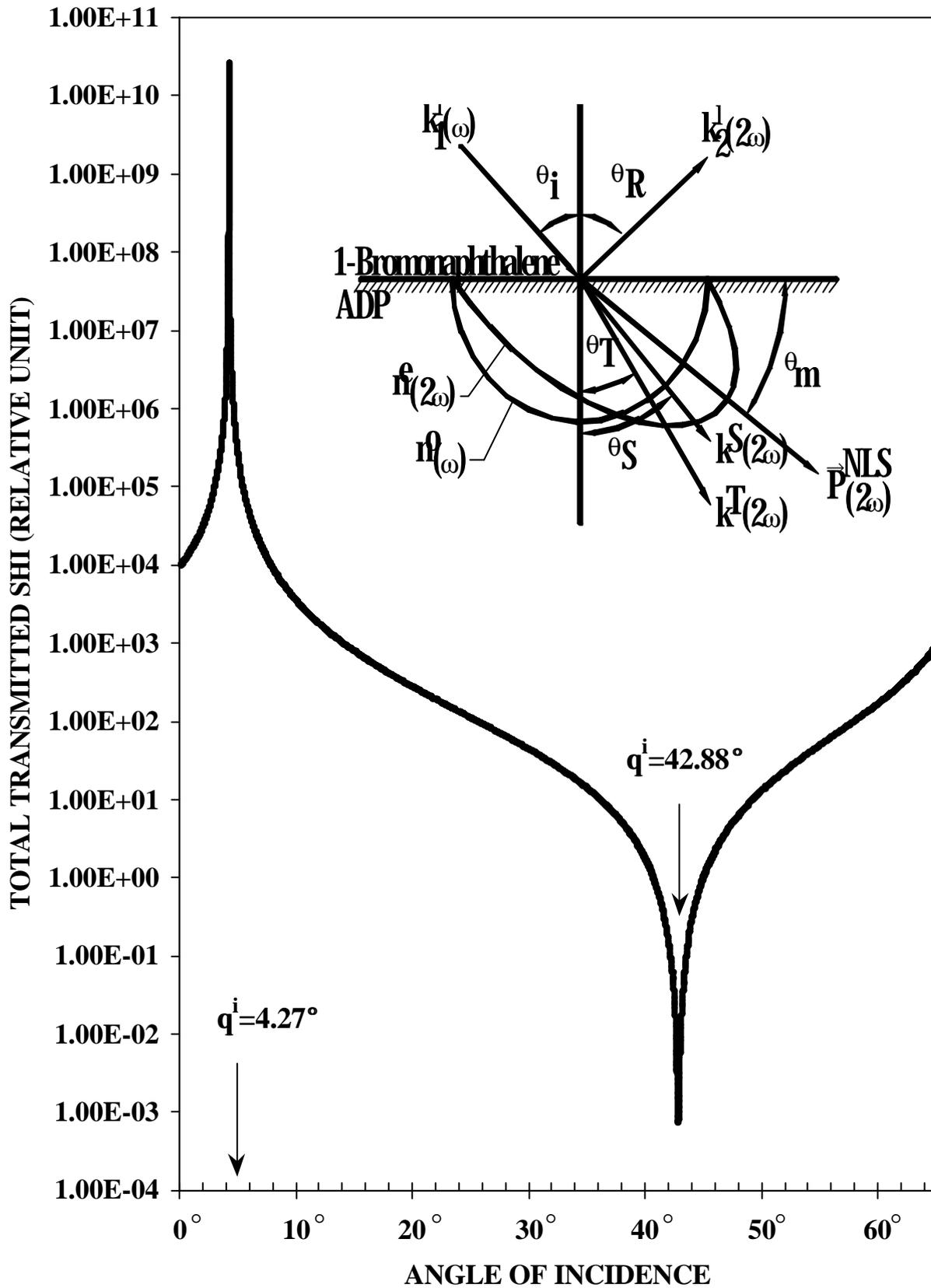
second harmonic occurred. This phenomenon is shown by figure 4.4. The tendency of the values of total transmitted second harmonic intensity increases from zero value of the incident angle and reach to the maximum values at the value of incident angle equal  $4.27^\circ$ . The incident angle that yield the colinear phase matching condition could be discussed as case of the direction of nonlinear polarization  $\vec{P}^{NLS}(2\mathbf{w})$  parallel to face normal of item 4.3.1. At phase matching condition, the value of  $n_e^{2w}$  equal  $n_o^w$  and also the value of the transmitted angle  $\mathbf{q}_s$  equal  $\mathbf{q}_r$  which equal to 4.61. Then the value of the refractive indices  $n_e^{2w}(\mathbf{q}) = n_e^{2w}(\mathbf{q}_m) = n_o^w$  equals to 1.5120. So the value of the incident angle is given by Snell's law as

$$n_{liq}^w \sin \mathbf{q}_i = n_e^{2w}(\mathbf{q}_m) \sin \mathbf{q}_r$$

$$\mathbf{q}_i = \sin^{-1} \left( \frac{n_e^{2w}(\mathbf{q}_m) \sin \mathbf{q}_r}{n_{liq}^w} \right)$$

$$\mathbf{q}_i = \sin^{-1} \frac{1.5120 \cdot \sin 4.61^\circ}{1.6355} = 4.27^\circ$$

The result agrees well with BP theory and the experimental work that performed under the same crystal orientation with KDP crystal (Bhantumnavin and Lee, 1994). From the occurrence of maximum transmitted second harmonic intensity as shown by figure 4.3 and 4.4, it's clear that this phenomenon will be occurred at phase matching condition.



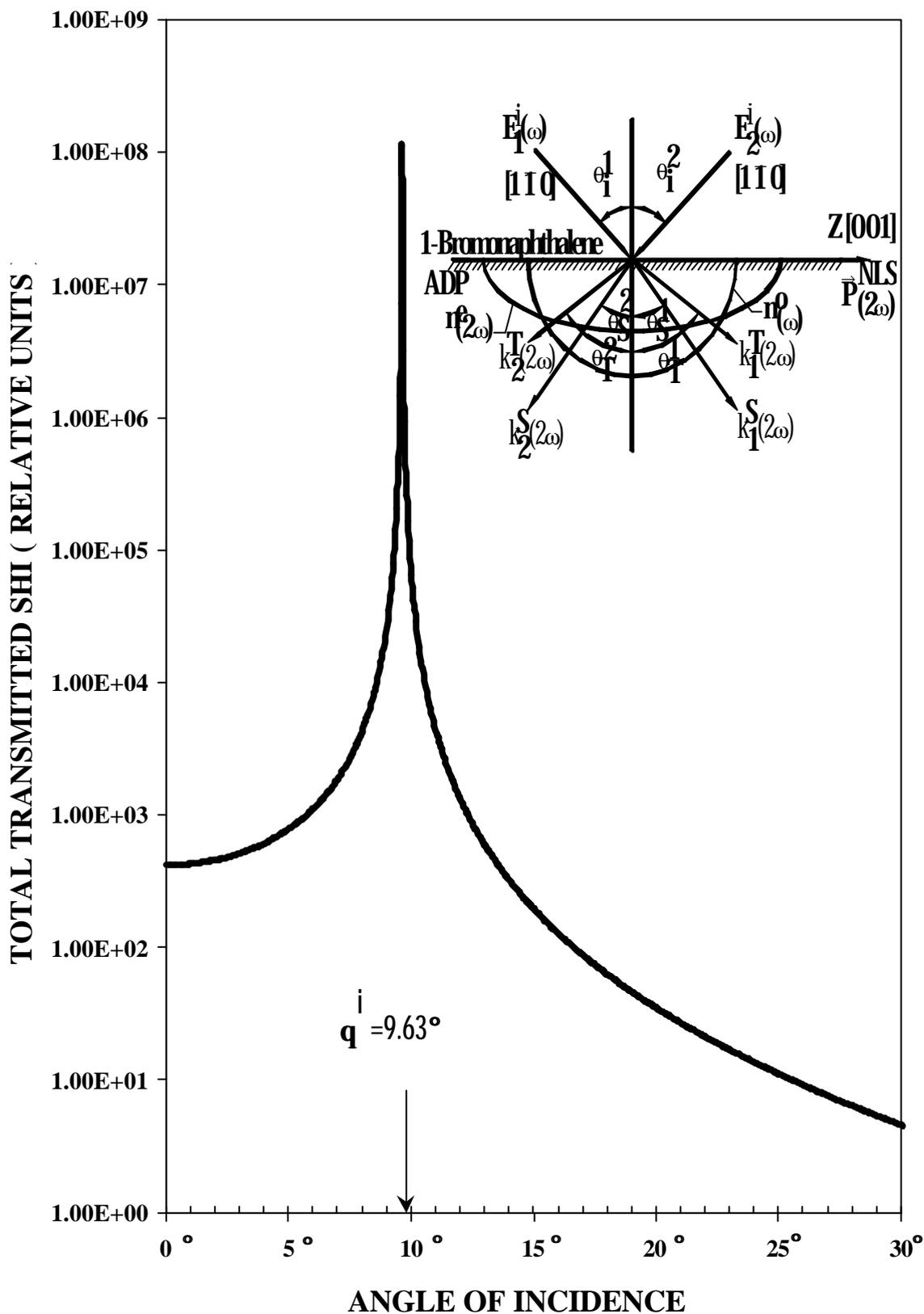
**Figure 4.4** The transmitted second harmonic intensity (SHI) as a function of the angle of incidence. The nonlinear polarization  $\bar{P}^{NLS}(2\omega)$  makes phase matching angle with the crystal surface.

### 4.3.3 The Maximum Transmitted Second Harmonic Generation under Noncolinear Phase Matching Condition, Two Beam Spatial Mixing (TBSM).

For the case of colinear incidence, the second harmonic light beam occurred from the participation of two photons of one laser light beam. However the combination of each photon of two incident laser light beam of different direction could generate second harmonic light at the condition of noncolinear incidence or Two Beam Spatial Mixing (TBSM). The ADP crystal is cut to have the direction of nonlinear polarization  $\vec{P}^{NLS}(2\mathbf{w})$  and the optic axis lie at the interface as shown by the inset of figure 4.5. The tendency of the value of total transmitted intensity begin to increase from zero value of the incident angle and reach to the maximum value at the angle of incidence equal  $9.63^\circ$ . The maximum value of the harmonic intensity under noncolinear phase matching condition is explained in chapter two. From figure 2.10, the linear transmitted wave vector  $\vec{k}^t(\mathbf{w})$  is considered as the inhomogeneous wave vector  $\vec{k}^s(\mathbf{w})$  and the results of the harmonic wave vector will be occurred in the direction of face normal. Therefore, the value of the wave vector  $\vec{k}^s(2\mathbf{w})$  as shown by the inset of figure 4.5 is given by

$$\vec{k}^s = \frac{2\mathbf{w}}{c} n_o^w \cos \mathbf{q}_s \hat{a}$$

where the angle  $\mathbf{q}_s$  is the transmitted angle of  $\vec{k}^s(2\mathbf{w})$  and  $\hat{a}$  is the unit vector in the direction of face normal. Then the value of the refractive index  $n_e^{2w}(\mathbf{q}) = n_e^{2w}(\mathbf{p}/2)$  equal to 1.4870. So (2.70) could be rewritten as



**Figure 4.5** The transmitted second harmonic intensity (SHI) at the noncolinear phase-matching condition. The phase matching angle ( $\theta^i$ ) equal  $9.63^\circ$ . The nonlinear polarization  $\vec{P}^{NLS}(2\omega)$  parallel to the crystal surface.

$$\left[ \frac{n_e^{2w}(\mathbf{p}/2)}{c} 2\mathbf{w} \right] \hat{\mathbf{a}} = \left[ \frac{n_o^w}{c} \mathbf{w} \cos(\mathbf{q}_s) + \frac{n_o^w}{c} \mathbf{w} \cos(\mathbf{q}_s) \right] \hat{\mathbf{a}}$$

$$n_e^{2w}(\mathbf{p}/2) = n_o^w \cos \mathbf{q}_s$$

$$\mathbf{q}_s = \mathbf{f}_m = \cos^{-1} \left[ \frac{n_e^{2w}(\mathbf{p}/2)}{n_o^w} \right] = \cos^{-1} \left[ \frac{1.4870}{1.5120} \right] = 10.43^\circ$$

where  $\mathbf{f}_m$  is the phase matching angle of noncolinear phase matching condition. Then the incident angle under noncolinear phase matching condition is given by Snell's law as

$$\mathbf{q}_i = \sin^{-1} (n_o^w \sin \mathbf{f}_m)$$

$$\mathbf{q}_i = \sin^{-1} (1.5120 \sin 10.43^\circ) = 9.63^\circ$$

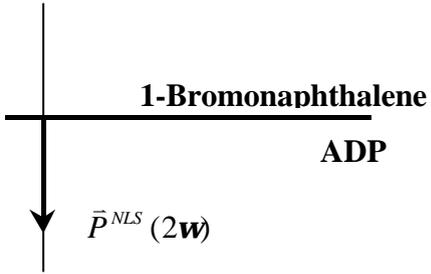
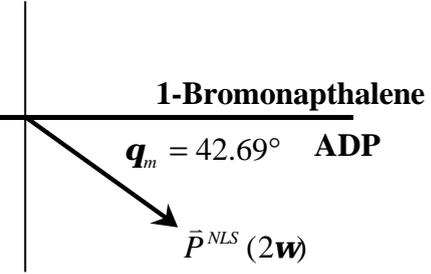
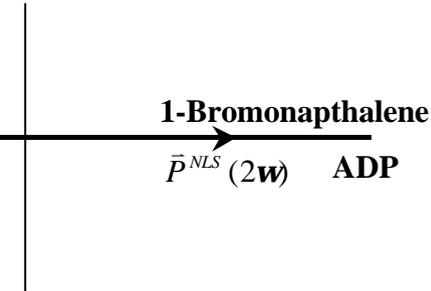
The result agrees well with the BP theory and the result of the experimental work that performed under the same crystal orientation from KDP crystal (Bhanthumnavin and Lee, 1990).

# Chapter V

## Conclusion

Transmitted second harmonic generation in Ammonium Dihydrogen Phosphate, ADP crystal immersed in the optically denser fluid 1-bromonaphthalene by using ultra short laser pulse laser at wavelength equal 900 nm. is investigated at different orientations of  $\bar{P}^{NLS}(2\mathbf{w})$  of the crystal. The theoretical results are compared with the results of the previous experimental work with KDP crystal immersed in the optically denser liquid 1-bromonaphthalene. (Bhanthumnavin and Lee, 1990; 1994) with very good agreement. The results of the maximum transmitted second harmonic intensity of colinear and noncolinear incident cases at the phase matching condition are shown. Besides, null transmitted second harmonic intensity at normal incidence is found to occur at a specific orientation of  $\bar{P}^{NLS}(2\mathbf{w})$  of the crystal. The occurrence of the minimum and maximum values of the transmitted second harmonic intensity are differentiated when the crystal orientation is changed, concluding that the transmitted second harmonic generation is dependent on the orientation of the crystal as shown by the conclusion of the theoretical results (Table 5.1). These agree well with the preliminary experimental work (Bhanthumnavin and Lee, 1990; 1994) and the theory of Bloembergen and Pershan (1962).

**Table 5.1** The summarized results of the transmitted second harmonic generation at different crystallographic orientations.

Crystallographic orientation	$q_i$	$I_S(2\omega)$	$I_T(2\omega)$	$I_{total}^T(2\omega)$
<u>Colinear</u> 	Null = 0°	Min	min	min
	38.81°	Max	max	max
<u>Colinear</u> 	42.80°	Min	-	very low
	43.59°	-	min	very low
	42.88°	-	-	min
	4.27°	max	max	max
<u>Noncolinear</u> 	9.63°	max	max	max

A further experiment to be performed for verification of this theoretical study is suggested as the following:

1. The laser light source should be pico or femto second pulsewidth because a very short duration of pulsewidth will produce less crystal damage. Besides, it has a very high peak power which is useful for generating the transmitted second harmonic intensity. It is especially useful for observing the small signal at the condition of minimum values of the harmonic intensity. Therefore, the ultrashort pulse laser should be used as the incident light source for both theoretical and experiments.

2. A very sensitive detector should be used for detecting the second harmonic signal so that it could easily detect the small value of the second harmonic signal, especially at the occurrence of the null signal and minimum transmitted second harmonic intensity. Nevertheless the spurious signal and light generated by other sources should be avoided by using the analyzer or the filter technique.

3. The entrance and exit surface of ADP should be polished optically flat to  $\lambda/10$  for protection of the scattering of laser light. If the crystal is surrounded by air, the humidity could damage the crystal surface. Therefore, the crystal should be immersed in the fluid 1-bromonaphthalene for protection the crystal surface and the results could be compared with the previous experimental works (Bhanthumnavin and Lee, 1990; 1994).

## **References**

# References

- Armstrong, J.A., Bloembergen, N., Ducuing, J., and Pershan, P.S. (1962). Interactions between light waves in nonlinear dielectric. **Phys. Rev.** 127:1918.
- Bhanthumnavin, V., and Ampole, N. (1990). Theoretical prediction of nonlinear brewster angle in ADP. **Microw. Opt. Technol. Lett.** 3:239.
- Bhanthumnavin, V., and Lee, C.H. (1990). Reflection and transmission in second harmonic generation of light in KDP crystal. **Microw. Opt. Technol. Lett.** 3:279.
- Bhanthumnavin, V., and Lee, C.H. (1994). Optical second-harmonic generation at total reflection in a potassium dihydrogen phosphate crystal. **Phys. Rev.** 50:2579.
- Bhanthumnavin, V., and Lee, C.H. (1994). Optical second-harmonic generation of oblique incident light in transmission in potassium dihydrogen phosphate crystal. **J. Appl. Phys.** 75:3294.
- Bloembergen, N. (1956). Proposal for a new type of solid state maser. **Phys. Rev.** 104:324.
- Bloembergen, N., and Pershan, P.S. (1962). Light waves at the boundary of nonlinear media. **Phys. Rev.** 128:606.
- Bloembergen, N., and Lee, C.H. (1967). Total reflection in second-harmonic generation. **Phys. Rev. Letters.** 19:835.

- Bloembergen, N., Simon, H.J., and Lee, C.H. (1969). Total reflection phenomena in second-harmonic generation of light. **Phys. Rev.** 181:1261.
- Chang, R.K., and Bloembergen, N. (1966). Experiment verification of the laws for the reflected intensity of second-harmonic light. **Phys. Rev.** 144:775.
- Ducing, J., and Bloembergen, N. (1963). Observation of reflected light harmonics at the boundary of piezoelectric crystals. **Phys. Rev. Letters.** 10:474.
- Dürr, T., Hildebrandt, R., Marowsky, G., and Stolle R. (1997) Nonlinear brewster angle in transmission. **Phys. Rev.** 56:4139.
- Franken, P.A., Hill, A.E., Peters, C.W., and Weinreich, G. (1961). Generation of optical harmonics. **Phys. Rev. Letters.** 7:118.
- Giordmaine, J.A. (1962). Mixing of light beams in crystal. **Phys. Rev. Letters.** 8:19.
- Gordon, J.P. (1984). Molecular microwave oscillator and new hyperfine structure in the microwave spectrum of NH<sub>3</sub>. **Phys. Rev.** 95:282.
- Hellwarth, R.W. (1961). **Advances in quantum electronics.** New York: Columbia University Press.
- Hellwarth, R.W. (1966). **Lasers.** New York: Marcel Dekker.
- Javan, A., W.R., Bennett, J.R., and Herriott, D.R. (1961). Population inversion and continuous optical maser oscillation in a gas discharge containing a He-Ne mixture. **Phys. Rev. Letters.** 6:106
- Jenkis, F.A., and White, H.E. (1976). **Fundamental of Optics.** 4ed. Singapore: Mc Graw Hill
- Lee, C.H., and Bhanthumnavin, V. (1976). Observation of nonlinear brewster angle in KDP. **Opt. Commun.** 18:326.

- Maker, P.D., Terhune, R.W., Nieoff, M., and Savage, C.M. (1962). Effects of dispersion and focusing on the production of optical harmonics. **Phys. Rev. Letters.** 8:21.
- Maiman, T.H. (1960). Stimulated optical radiation in ruby masers. **Nature** 487:493.
- Patel, C.K.N. (1964). Interpretation of CO<sub>2</sub> optical maser experiments. **Phys Rev. Letters.** 12:588
- Savage, A. (1965). Improved geometry for quantitative measurements of optical frequency doubling. (1965). **J. Appl. Phys.** 36:1496.
- Shank, C. V., Fork, R.L., Yen, R., Stolen, R.H., and Toomlinson, W.J. (1982). Compression of femto-second optical pulses. **Appl. Phys. Letters.** 40:761.
- Shapiro, S.L. (ed.). (1977). **Ultrashort light pulses.** Berlin New York:Springer-Verlag
- Shawlow, A. L., and Townes, C.H. (1958). Infrared and optical masers. **Phys. Rev.** 112:1940.
- Yariv, A. (1989). **Quantum electronics.** 3<sup>rd</sup> ed. Singapore: John Wiley & Sons.
- Zernike, F., Jr. (1964) Refractive indices of ammonium dihydrogen phosphate and potassium dihydrogen phosphate between 2000 Å and 1.5 μm, **J. Opt. Soc. Am.**, 54:1215
- Zernike, F., Jr. (1965). Errata of Zernike, F., Jr. (1964), **J. Opt. Soc. Am.** 54:1215

# **Appendixes**

## **Appendix A.**

**The Calculation of Refractive Indices of Ammonium  
Dihydrogen Phosphate and Liquid 1-Bromonaphthalene at  
Wavelength Equal 900 nm and 450 nm.**

The conclusion of the refractive indices that used in the theoretical study transmitted second harmonic intensity of Ammonium Dihydrogen Phosphate, (ADP) crystal and liquid 1-Bromonaphthalene is shown by table A1.

**Table A1** The value of the refractive index of ADP crystal and liquid 1-bromonaphthalene at wavelength equal 450 nm and 900 nm.

Wavelength, $\lambda$ (nm)	The refractive index, $n$		
	$n_e$	$n_o$	$n_{liq}$
900	1.4709	1.5120	1.6355
450	1.4870	1.5343	1.6952

**Where**  $n_e$  = **The refractive index of extraordinary ray**

$n_o$  = The refractive index of ordinary ray

$n_{liq}$  = The refractive index of liquid 1-Bromonaphthalene

**The calculation of**  $n_e$  **and**  $n_o$

The values of refractive index of ADP crystal at wavelength 900 nm and 450 nm has been calculated by using the method of known incidence used by Rydberg (1828), Tilton, Plyler, and Stephens (1949) and by using the work of Zernike (1964, 1965). From the work of Zernike, the refractive indices of ADP crystal and KDP crystal have been measured at 25 wavelengths between 2138 Å and 1.529 μm. The following equations are used to calculate the values of index of refraction:

$$n^2 = A + Bn^2/(1-n^2/C) + D/(E-n^2), \text{ or} \quad (1)$$

$$n^2 = A + B/(I^2 - 1/C) + (DI^2/E)/(I^2 - 1/E), \quad (2)$$

where  $n = \frac{1}{I}$  ( $\text{cm}^{-1}$ ) and  $A, B, C, D, E$  are constants.

**Table A2** The value of constant  $A, B, C, D,$  and  $E$  with respect to air.

ADP, e ray		ADP, o ray
$A$	2.163510	2.302842
$B$	$9.616676 \times 10^{-11}$	$1.1125165 \times 10^{-10}$
$C$	$7.698751 \times 10^9$	$7.5450861 \times 10^9$
$D$	$1.479974 \times 10^6$	$3.775616 \times 10^6$
$E$	$2.5 \times 10^5$	$2.5 \times 10^5$

[ Zernike (1965) ]

Equation (2) was used to calculate the value of index of refraction  $n_e$  and  $n_o$  at wavelength 900 nm and 450 nm by substituting the values of constant A,B,C,D,E from table 2 and the value of wavelength into equation(2). The unit of wavelength has been change from  $cm$  . to  $mm$  . by multiplying by  $10^4$  .

The refractive indices at wavelength equal 900 nm or 0.9  $mm$ .

$$n_e^2 = 2.163512 + \frac{0.009616676}{[(0.9)^2 - 0.01298912]} + \frac{5.919896 \cdot (0.9)^2}{[(0.9)^2 - 400]} = 2.163563814$$

$$n_e = (2.163563814)^{\frac{1}{2}} = 1.4709$$

$$n_o^2 = 2.302842 + \frac{0.011125165}{[(0.9)^2 - 0.013253659]} + \frac{15.102464 \cdot (0.9)^2}{[(0.9)^2 - 400]} = 2.286160701$$

$$n_o = (2.286160701)^{\frac{1}{2}} = 1.5120$$

The refractive indices at wavelength equal 450 nm or 0.45  $\mu\text{m}$ .

$$n_e^2 = 2.163512 + \frac{0.009616676}{[(0.45)^2 - 0.01298912]} + \frac{5.919896 \cdot (0.45)^2}{[(0.45)^2 - 400]} = 2.211256252$$

$$n_e = (2.211256252)^{\frac{1}{2}} = 1.4870$$

$$n_o^2 = 2.302842 + \frac{0.011125165}{[(0.45)^2 - 0.013253659]} + \frac{15.102464 \cdot (0.45)^2}{[(0.45)^2 - 400]} = 2.35397919$$

$$n_o = (2.35397919)^{\frac{1}{2}} = 1.5343$$

Therefore, we conclude that the value of refractive indices at wavelength equal 900 nm  $n_e = \underline{1.4709}$  and  $n_o = \underline{1.5120}$ . At wavelength equal 450 nm, the refractive indices  $n_e = \underline{1.4870}$  and  $n_o = \underline{1.5343}$

### **The calculation of $n_{liq}$**

The refractive index of liquid 1-bromonaphthalene,  $n_{liq}$  at wavelengths equal 900 nm and 450 nm, is calculated by using the Cauchy's equation (Jenkins and White, 1976).

$$n = A + B/I^2 + C/I^4, \quad (3)$$

where  $A$ ,  $B$ , and  $C$  are constants.

In order to know the values of constants  $A$ ,  $B$ , and  $C$ , it necessary to know at least three values of  $n_{liq}$  at different values of  $I$ . Then equations could be set up and

will be solved for finding the constant's values. In this calculation, four sets of three different wavelengths are used. The result is the average value of each set. The refractive index of  $n_{liq}$  of four sets of three different wavelengths will be used to calculate  $n_{liq}$  at wavelength equals 900 nm and 450 nm. Further average values of  $n_{liq}$  are shown in Table A3 and Table A4 respectively.

**Table A3** The four set of the value of  $n_{liq}$  for calculating the new value of  $n_{liq}$  at wavelength equals 900 nm.

1 <sup>st</sup> Set		2 <sup>nd</sup> Set		3 <sup>rd</sup> Set		4 <sup>th</sup> Set	
$\lambda$ (nm)	$n_{liq}$	$\lambda$ (nm)	$n_{liq}$	$\lambda$ (nm)	$n_{liq}$	$\lambda$ (nm)	$n_{liq}$
1064	1.6262	1064	1.6262	1064	1.6262	977	1.6340
532	1.6701	532	1.6701	977	1.6340	532	1.6701
486.1	1.6817	434	1.7041	532	1.16701	434	1.7041
$n_{liq} = 1.6320$		$n_{liq} = 1.631548$		$n_{liq} = 1.642057$		$n_{liq} = 1.636248$	
The average values of $n_{liq}$ at wavelength equal 900 nm. is <u>1.6355</u>							

Table A3 shows the refractive index of  $n_{liq}$  of four sets of three different wavelengths that used to calculate  $n_{liq}$  at wavelength equals 900 nm and the average value of  $n_{liq}$ .

**Table A4** The four set of the value of  $n_{liq}$  for calculating the new value of  $n_{liq}$  at wavelength equals 450 nm.

1 <sup>st</sup> Set		2 <sup>nd</sup> Set		3 <sup>rd</sup> Set		4 <sup>th</sup> Set	
$\lambda$ (nm)	$n_{liq}$	$\lambda$ (nm)	$n_{liq}$	$\lambda$ (nm)	$n_{liq}$	$\lambda$ (nm)	$n_{liq}$
1064	1.6262	1064	1.6262	1064	1.6262	977	1.6340
532	1.6701	532	1.6701	977	1.6340	532	1.6701
486.1	1.6817	434	1.7041	488.5	1.6810	434	1.7041
$n_{liq} = 1.69350$		$n_{liq} = 1.69669$		$n_{liq} = 1.69408$		$n_{liq} = 1.696435$	
The average values of $n_{liq}$ at wavelength equal 450 nm is <u>1.6952</u>							

Table A4 shows the refractive index of  $n_{liq}$  of four sets of three different wavelengths that used to calculate  $n_{liq}$  at wavelength equals 450 nm and the average value of  $n_{liq}$ . In order to average out for good accuracy, the different values of three wavelengths of the third set were used to calculate  $n_{liq}$  at wavelength equals 450 nm.

# **Appendix B.**

## **Computer Programming (C++)**

### B1. Colinear Incidence when $P^{NLS}(2\mathbf{w})$ Lies Along Face Normal.

Program for calculation relative transmission intensity of ADP at wavelength equal 900nm. in case of  $\vec{P}^{NLS}(2\mathbf{w})$  and Optic axis lie along face normal. The refractive indexes of ADP crystal  $n_o(\mathbf{w})$  equal 1.5120,  $n_o(2\mathbf{w})$  equal 1.5343, and  $n_e(2\mathbf{w})$  equal 1.4870. The refractive indexes of liquid 1-bromonaphthalene  $n_{liq}(\mathbf{w})$  equal 1.6355,  $n_{liq}(2\mathbf{w})$  equal 1.6952.

```
#include <iostream.h>

#include <stdio.h>

#include <iomanip.h>

#include <stdlib.h>

#include <math.h>

#include <conio.h>

#include <complex.h>

double nzeta2w1(double);

double nzeta2w2(double);

double It(double,double,complex);

int show(double,double,double,double,double,int);

void main()

{

    float AngS,AngE,AngEE,Add;

    float OiD;
```

```

double zetai,zetas1,zetat1,OTT,OSS,nzeta2w,Itotal;

const float now=1.5120;

const float nlqw=1.6355;

clrscr();

FILE *f;

f=fopen("data3.txt","w+");

cout<<"\n\nStaring Incident Angle at: ";cin>>AngS;

cout<<"\n\nEnding Incident Angle at: ";cin>>AngE;

cout<<"\n\nIncreasing Angle equal: ";cin>>Add;

OiD=AngS;

AngEE=AngE;

int i=0;

while (OiD<=AngEE)

{ if(OiD<0)

{ while(OiD<0)

{ double OiD1=(OiD*(-1));

zetai=OiD1 *M_PI/180;

nzeta2w=nzeta2w1(zetai);

complex zs=nlqw*sin(zetai)/now;

complex zs1=asin(zs);

complex zeta1=zs1;

Itotal=It(zetai,nzeta2w,zeta1);

OTT=(nlqw*sin(zetai)/nzeta2w);

complex OT=asin(OTT);

```

```

OSS=(nlqw*sin(zetai)/now);
complex OS=asin(OSS);
zetat1=real(OT)*180/M_PI;
zetas1=real(OS)*180/M_PI;
fprintf(f," %10.6f %20.20lf\n",
        OiD,Itotal);
i=show(OiD,zetas1,zetat1,nzeta2w,Itotal,i);
OiD=OiD+Add;
}
}
if (OiD<65.40)
{
zetai=OiD*M_PI/180;
nzeta2w=nzeta2w1(zetai);
complex zs=nlqw*sin(zetai)/now;
complex zs1=asin(zs);
complex zeta1=zs1;
Itotal=It(zetai,nzeta2w,zeta1);
OTT=(nlqw*sin(zetai)/nzeta2w);
complex OT=asin(OTT);
OSS=(nlqw*sin(zetai)/now);
complex OS=asin(OSS);
zetat1=real(OT)*180/M_PI;
zetas1=real(OS)*180/M_PI;

```

```

    }

    if (OiD>65.40)
    { const float ne2w=1.4870;

      zetai=OiD*M_PI/180;

      nzeta2w=ne2w;

      complex zs=nlqw*sin(zetai)/now;

      complex zs1=asin(zs);

      complex zeta1=zs1;

      Itotal=It(zetai,nzeta2w,zeta1);

      OTT=(nlqw*sin(zetai)/nzeta2w);

      complex OT=asin(OTT);

      OSS=(nlqw*sin(zetai)/now);

      complex OS=asin(OSS);

      zetat1=real(OT)*180/M_PI;

      zetas1=real(OS)*180/M_PI;

    }

    fprintf(f," %10.6lf %20.20lf\n",

           OiD,Itotal);

    i=show(OiD,zetas1,zetat1,nzeta2w,Itotal,i);

    OiD+=Add;

  }

  fclose(f);

  cout<<"Completely Calculation";

  getch();

```

```

}

double nzeta2w1(double zetai)
{
    const float nlqw=1.6355;
    const float no2w=1.5343;
    const float ne2w=1.4870;
    double termn1 =pow( (nlqw*sin(zetai)),2);
    double termn2 = 1+( termn1*(1/pow(no2w,2)-1/pow(ne2w,2)) );
    double termn3 =termn2*pow(no2w,2);
    double nzeta2w =pow(termn3,0.5);
    return nzeta2w;
}

double It(double zetai,double nzeta2w,complex zeta1)
{
    const float now=1.5120;
    const float nlqw=1.6355;
    const float nlq2w=1.6952;
    complex alpha=zeta1;
    double zetar = asin(nlqw*sin(zetai)/nlq2w);
    complex zs=(nlqw*sin(zetai)/now);
    complex zetas = asin(zs);
    complex zt=(nlqw*sin(zetai)/nzeta2w);
    complex zetat = asin(zt);
    complex fl = ( 2*cos(zetai)*sin(zetas) )/
                ( sin(zetai+zetas) );
}

```

```

complex fnlr = (sin(zetas)*pow(sin(zetat),2)*sin(alpha+zetas+zetat))/
               (pow(nlq2w,2)*sin(zetar)*sin(zetat+zetar)
               *cos(zetat-zetar)*sin(zetas+zetat) );

complex fnls = sin(alpha)/( pow(now,2)-pow(nzeta2w,2) );

complex fnlt = ( -now*fnls/nzeta2w )+( nlq2w*fnlr/nzeta2w );

complex Its = pow(fl,4)*pow(fnls,2)*cos(zetas)/cos(zetai);

complex Itt = pow(fl,4)*pow(fnlt,2)*cos(zetat)/cos(zetai);

complex Irr = pow(fl,4)*pow(fnlr,2)*cos(zetar)/cos(zetai);

double Is =abs(Its);

double It =abs(Itt);

double Itotal= (Is+It);

return Itotal;

}

int show(double OiD, double zetas1,double zetat1,double nzeta2w, double
Itotal,int i)

{printf("% 10.5lf|",OiD);

printf("% 10.6lf|",zetas1);

printf("% 10.6lf|",zetat1);

printf("% 10.4lf|",nzeta2w);

printf("% 15.8lf\n|",Itotal);

return i;

}

```

## B2. Colinear Incidence when $P^{NLS}(2\mathbf{w})$ Makes Phase Matching Angle with the Crystal Surface.

Program for calculation Relative Transmission intensity of ADP at wavelength equal 900nm. (Note: when  $q_r > 128.76$ ,  $q_i > 90$ ). The refractive indexes of ADP crystal  $n_o(\mathbf{w})$  equal 1.5120,  $n_o(2\mathbf{w})$  equal 1.5343, and  $n_e(2\mathbf{w})$  equal 1.4870. The refractive indexes of liquid 1-bromonaphthalene  $n_{liq}(\mathbf{w})$  equal 1.6355 and  $n_{liq}(2\mathbf{w})$  equal 1.6952. The direction of  $P^{NLS}(2\mathbf{w})$  makes phase matching angle with the surface crystal.

```
#include <iostream.h>
```

```
#include <stdio.h>
```

```
#include <iomanip.h>
```

```
#include <stdlib.h>
```

```
#include <math.h>
```

```
#include <conio.h>
```

```
#include <complex.h>
```

```
double nzeta2w1(double);// with the interface of two media.
```

```
double nzeta2w2(double);
```

```
double It(double,double,double,double,complex);
```

```
int show(double,double,double,double,double,int);
```

```
void main()
```

```

{

float AngS,AngE,AngEE,Add;

double zetai,zeta1,OiT,zetas1,zetat,zetat1,OTT,OSS,nzeta2w,Itotal;

const float now=1.5120;

const float nlqw=1.6355;

clrscr();

FILE *f;

f=fopen("dataA1.txt","w+");

cout<<"\n\nStaring Transmit Angle at: ";cin>>AngS;

cout<<"\n\nEnding Transmit Angle at: ";cin>>AngE;

cout<<"\n\nIncreasing Angle equal: ";cin>>Add;

OiT=AngS;

AngEE=AngE;

int i=0;

while (OiT<=AngEE)

{

if(OiT<=47.31)

{

zetat=OiT*M_PI/180;

nzeta2w=nzeta2w1(zetat);

double zetai=asin(nzeta2w*sin(zetat)/nlqw);

complex zs=nlqw*sin(zetai)/now;

complex zs1=asin(zs);

double zeta1=((47.31-real(zs1)*180/M_PI))*M_PI/180;

```

```

double zeta2=47.31;

complex zeta3=0;

Itotal=It(zetai,nzeta2w,zeta1,zeta2,zeta3);

OSS=(nlqw*sin(zetai)/now);

complex OS=asin(OSS);

zetai1= zetai*180/M_PI;

zetat1=OiT;

zetas1=real(OS)*180/M_PI;

}

if (OiT<=90)

{

zetat=OiT*M_PI/180;

nzeta2w=nzeta2w2(zetat);

double zetai=asin(nzeta2w*sin(zetat)/nlqw);

complex zs=nlqw*sin(zetai)/now;

complex zs1=asin(zs);

double zeta1=((real(zs1)*180/M_PI)-47.31)*M_PI/180;

double zeta2=(2*(real(zs1)*180/M_PI)+47.31)*M_PI/180;

complex zeta3=0;

Itotal=It(zetai,nzeta2w,zeta1,zeta2,zeta3);

OSS=(nlqw*sin(zetai)/now);

complex OS=asin(OSS);

zetai1=zetai*180/M_PI;

zetat1=OiT;

```

```

zetas1=real(OS)*180/M_PI;

}else

{

zetat=OiT*M_PI/180;

nzeta2w=nzeta2w2(zetat);

double zetai2=asin(nzeta2w*sin(zetat)/nlqw);

double zetai=((67.591457-(zetai2*180/M_PI))+67.591457)*M_PI/180;

complex zs=nlqw*sin(zetai)/now;

complex zs1=asin(zs);

double zeta1=((arg(zs1)*180/M_PI)+42.68)*M_PI/180;

double zeta2=0;

complex zeta3((((arg(zs1)*180/M_PI)+42.68)*M_PI/180)+zs1);

Itotal=It(zetai,nzeta2w,zeta1,zeta2,zeta3);

complex OSS=(nlqw*sin(zetai)/now);

complex OS=asin(OSS);

zetai1=zetai*180/M_PI;

zetat1=OiT;

zetas1=real(OS)*180/M_PI;

}

fprintf(f," %6.3lf %15.8lf\n", zetai1,Itotal);

i=show(zetai1,zetas1,zetat1,nzeta2w,Itotal,i);

OiT+=Add;

}

fclose(f);

```

```

cout<<"Completely Calculation";

getch();

}

double nzeta2w1(double zetat)

{const float no2w=1.5343;

const float ne2w=1.4870;

double zeta=47.31-(zetat*180/M_PI);

double zeta1=zeta*M_PI/180;

double termn= (pow(cos(zeta1),2)/pow(no2w,2))+

               (pow(sin(zeta1),2)/pow(ne2w,2));

double nzeta2w = pow(termn,-0.5);

return nzeta2w;

}

double nzeta2w2(double zetat)

{const float no2w=1.5343;

const float ne2w=1.4870;

double zeta=(zetat*180/M_PI)-47.31;

double zeta1=zeta*M_PI/180;

double termn= (pow(cos(zeta1),2)/pow(no2w,2))+

               (pow(sin(zeta1),2)/pow(ne2w,2));

double nzeta2w = pow(termn,-0.5);

return nzeta2w;

```

```

}

double It(double zetai,double nzeta2w,double zeta1,double zeta2,complex
zeta3)

{ const float now=1.5120;

const float nlqw=1.6355;

const float nlq2w=1.6952;

int imp=1;

if (zetai*180/M_PI>90)

imp=0;

double alpha=zeta1;

complex zt=(nlqw*sin(zetai)/nzeta2w);

complex zetat= asin(zt);

double zetar = asin(nlqw*sin(zetai)/nlq2w);

complex zs=(nlqw*sin(zetai)/now);

complex zetas = asin(zs);

complex fl = ( 2*cos(zetai)*sin(zetas) )/

( sin(zetai+zetas) );

complex fnlr = (sin(zetas)*pow(sin(zetat),2)*sin(zeta2+zetat+zeta3))/

(pow(nlq2w,2)*sin(zetar)*sin(zetat+zetar)

*cos(zetat-zetar)*sin(zetas+zetat) );

complex fnls = sin(alpha)/( pow(now,2)-pow(nzeta2w,2) );

complex fnlt = ( -now*fnls/nzeta2w )+( nlq2w*fnlr/nzeta2w );

complex Its = pow(fl,4)*pow(fnls,2)*cos(zetas)/cos(zetai);

complex Itt = pow(fl,4)*pow(fnlt,2)*cos(zetat)/cos(zetai);

```

```
complex Irr = pow(f1,4)*pow(fnlr,2)*cos(zetar)/cos(zetai);  
double Is =abs(Its);  
double It =abs(Itt);  
double Itotal= (It+Is)*imp;  
return Itotal;  
}
```

```
int show(double OiT, double zetas1,double zetat1,double nzeta2w, double  
Itotal,int i)  
{printf("% 10.5lf|",OiT);  
printf("% 10.6lf|",zetas1);  
printf("% 10.6lf|",zetat1);  
printf("% 10.4lf|",nzeta2w);  
printf("% 15.8lf\n|",Itotal);  
return i;  
}
```

### B3. Noncolinear Incidence when $P^{NLS}(2\mathbf{w})$ Parallel to the Crystal Surface

Program for calculation relative transmission intensity of ADP at wavelength equal 900nm. at the noncolinear phase-matching condition (Note: when  $q_x > 124.95$ ,  $q_y > 90$ ). The refractive indexes of ADP crystal  $n_o(\mathbf{w})$  equal 1.5120,  $n_o(2\mathbf{w})$  equal 1.5343, and  $n_e(2\mathbf{w})$  equal 1.4870. The refractive indexes of liquid 1-bromonaphthalene.  $n_{liq}(\mathbf{w})$  equal 1.6355 and  $n_{liq}(2\mathbf{w})$  equal 1.6952.  $\bar{P}^{NLS}(2\mathbf{w})$  parallel to the crystal surface.

```
#include <iostream.h>
```

```
#include <stdio.h>
```

```
#include <iomanip.h>
```

```
#include <stdlib.h>
```

```
#include <math.h>
```

```
#include <conio.h>
```

```
#include <complex.h>
```

```
double nzeta2w2(double);
```

```
double It(double,double,double);
```

```
int show(double,double,double,double,double,int);
```

```
void main()
```

```
{
```

```

float AngS,AngE,AngEE,Add;

double zeta,no1,zetai,zetai1,OiT,zetas1,zetat,zetat1,OTT,OSS,nzeta2w,Itotal;

const float now=1.5120;

const float nlqw=1.6355;

clrscr();

FILE *f;

f=fopen("data7.txt","w+");

cout<<"\n\nStarting Transmit Angle at: ";cin>>AngS;

cout<<"\n\nEnding Transmit Angle at: ";cin>>AngE;

cout<<"\n\nIncreasing Angle equal: ";cin>>Add;

OiT=AngS;

AngEE=AngE;

int i=0;

while (OiT<=AngEE)

{

    if (OiT<=90)

    {

        zetat=OiT*M_PI/180;

        zeta=(M_PI/2)-zetat;

        nzeta2w=nzeta2w2(zeta);

        double zetai=asin(nzeta2w*sin(zetat)/nlqw);

        complex zs=nlqw*sin(zetai)/now;

        complex zs1=asin(zs);

        double zeta1=(90.0-(real(zs1)*180/M_PI))*M_PI/180;

```

```

Itotal=It(zetai,nzeta2w,zeta1);

OSS=(nlqw*sin(zetai)/now);

complex OS=asin(OSS);

zetai1=zetai*180/M_PI;

zetat1=OiT;

zetas1=real(OS)*180/M_PI;

}else

{

zetat=OiT*M_PI/180;

zeta=zetat-(M_PI/2);

nzeta2w=nzeta2w2(zeta);

double zetai2=asin(nzeta2w*sin(zetat)/nlqw);

double zetai=((69.738675-(zetai2*180/M_PI))+69.738675)*M_PI/180;

complex zs=nlqw*sin(zetai)/now;

complex zs1=asin(zs);

double zeta1=arg(zs1);

Itotal=It(zetai,nzeta2w,zeta1);

complex OSS=(nlqw*sin(zetai)/now);

complex OS=asin(OSS);

zetai1=zetai*180/M_PI;

zetat1=OiT;

zetas1=real(OS)*180/M_PI;

}

fprintf(f," %10.6lf %20.20lf\n",

```

```

        zeta1,Itotal);

    i=show(zeta1,zetas1,zetat1,nzeta2w,Itotal,i);

    OiT+=Add;

}

fclose(f);

cout<<"Completely Calculation";

getch();

}

double nzeta2w2(double zeta)

{const float no2w=1.5343;

const float ne2w=1.4870;

double termn= (pow(cos(zeta),2)/pow(no2w,2))+

               (pow(sin(zeta),2)/pow(ne2w,2));

double nzeta2w = pow(termn,-0.5);

return nzeta2w;

}

double It(double zetai,double nzeta2w,double zeta1)

{ const float now=1.5120;

const float ne2w=1.4870;

const float nlqw=1.6355;

const float nlq2w=1.6952;

int imp=1;

if (zetai*180/M_PI>90)

    imp=0;

```

```

double alpha=zeta1;

complex zt=(nlqw*sin(zetai)/nzeta2w);

complex zetat= asin(zt);

double zetar = asin(nlqw*sin(zetai)/nlq2w);

complex zs=(nlqw*sin(zetai)/now);

complex zetas = asin(zs);

complex fl = ( 2*cos(zetai)*sin(zetas) )/
              ( sin(zetai+zetas) );

complex fnlr = (sin(zetas)*pow(sin(zetat),2)*sin(alpha+zetas+zetat))/
              (pow(nlq2w,2)*sin(zetar)*sin(zetat+zetar)
              *cos(zetat-zetar)*sin(zetas+zetat) );

complex fnls = sin(alpha)/( pow((now*cos(zetas)),2)-pow(ne2w,2) );

complex fnlt = ( -(now*cos(zetas))*fnls/ne2w )+( nlq2w*fnlr/ne2w );

complex Its = pow(fl,4)*pow(fnls,2)*cos(zetas)/cos(zetai);

complex Itt = pow(fl,4)*pow(fnlt,2)*cos(zetat)/cos(zetai);

complex Irr = pow(fl,4)*pow(fnlr,2)*cos(zetar)/cos(zetai);

double Is =abs(Its);

double It =abs(Itt);

double Itotal= (Is+It)*imp;

return Itotal;

}

int show(double OiT, double zetas1,double zetat1,double nzeta2w, double
Itotal,int i)

```

```
{printf("% 10.5lf|",OiT);  
printf("% 10.6lf|",zetas1);  
printf("% 10.6lf|",zetat1);  
printf("% 10.4lf|",nzeta2w);  
printf("% 15.8lf\n|",Itotal);  
return i;  
}
```

## **Biography**

Mr. Songrit Kamyod was born on 8<sup>th</sup> June 1975 in Changrai, Thailand. He graduated with High School Diploma from Phayao Pittayakom School in 1994. Later he went to study in the School of Telecommunication Engineering, Institute of Industrial Technology, Suranaree University of Technology (SUT), Nakhonratchasima. While studying, he participated in World Tech'95 Thailand Exhibition during 4 Nov 1995 to 16 Dec. 1995 as a Booth attendant, responsible for explaining and demonstrating about Technology of Telecommunication, for TelecomAsia Public Co., Ltd. According to B.Sc. Degree program, he participated in a cooperative education program at System Engineering and Management Consultant Co., Ltd., Bangkok during 23 Sep. 1997 to 19 Dec 1997. He worked as an assistant engineer for the position about the small satellite earth station research project by collecting, and analyzing data, and making the conclusion. He graduated with a B.Sc. Degree in telecommunication Engineering in 1997 then decided to study in the Master's Degree program in the School of Laser Technology and Photonics, Institute of science, Suranaree University of Technology. While he was studying for his Master's Degree, he was a teaching assistant (TA) attached to physics laboratory for four trimesters.