Third-Harmonic Generation of Intense Laser Pulses

by

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CURRICULUM VITAE

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PUBLICATIONS


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**ABSTRACT**

In this thesis, third harmonic generation of ultrashort pulses and the related phenomena have been studied experimentally and theoretically. The third harmonic conversion of 1.6 ps pulses at 1053 nm were performed with the Chirped-Pulse-Amplification (CPA) laser system using two type II KDP crystals, one for frequency doubling and another one for frequency tripling. Conversion efficiencies of 60-65% was achieved from frequency doubling and that of 45-50% was achieved for frequency tripling at input pulse intensities of 50-100 GW/cm².

Our experimental study also showed the importance of the self- and cross-phase modulation effects in the frequency tripling process. The self-phase modulation coefficients in KDP crystals at wavelengths of 1.053 μm, 0.527 μm and 0.351 μm were measured using single beam Z-scan technique. The cross-phase modulation coefficients between 1.053 μm and 0.527 μm were also measured by a two color z-scan. These coefficients are used in the theoretical simulation of frequency conversion of ultrafast pulses.

It is important to know the direction of the crystals axis relative to the propagation direction of the laser to optimize the phase matching for harmonic generations. The orientation of the optical axis in the crystal determines the spatial and temporal walk-off between the ordinary (o) and extraordinary (e) waves. Frequency domain interferometry has been used to measure the group velocity walk-off (GVW) between the ordinary and extraordinary waves in uniaxial crystals. The dependence of the interference fringe spacing and the walk-off on incident angle with respect to the birefringent crystal were measured in KDP and the direction of the optical axis was determined.
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Chapter I
Introduction

1.1 INTRODUCTION

Nonlinear optics has been studied for more than thirty years since the harmonic generation of light was first observed by Franken and coworkers in 1961.\textsuperscript{1} It is a study of the interaction of intense laser light with material and includes such problems as light induced changes of optical properties of media. Nonlinear effects are important in applications such as laser frequency conversion, optical switching, optical communications and high resolution atomic and molecular spectroscopy.\textsuperscript{2,3,4}

Frequency conversion is an important nonlinear effect associated with the second order nonlinearity in a medium. It involves the generation of radiation at wavelengths other than those contained in the incident radiation. It includes second and higher harmonic generation,\textsuperscript{2} frequency mixing,\textsuperscript{5} optical parametric amplification (OPA) and oscillation (OPO).\textsuperscript{6,7,8} It provides a powerful tool to extend laser radiation over a wide frequency range. Frequency up-converted high power lasers have become the mainstay of the laser fusion program during the past decade, since it become apparent that the physics of laser-plasma interaction favors short wavelengths.\textsuperscript{9,10,11} Currently, the OPO and OPA are of particular interest for the generation of intense ultra-short tunable pulses. Such ultrashort pulses are in demand for picosecond and shorter spectroscopy. Picosecond spectroscopic technique have found a unique place in the investigation of many ultrafast processes in physics, chemistry and biology.\textsuperscript{12,13}

In the frequency conversion process, the intense incident laser fields cause the polarization of the medium to develop new frequency components not present in the incident field. These new frequency components of the polarization act as sources of new frequency components of the electromagnetic field. For sum frequency generation, the incident electric field at frequency $\omega_1$ and $\omega_2$ couple through a
nonlinear medium and generate the new frequency component at frequency \( \omega \). Note that \( \omega = \omega_1 + \omega_2 \) because of photon energy conservation. Even though momentum conservation \( \mathbf{k} = \mathbf{k}_1 + \mathbf{k}_2 \) is not required, it maximizes the wave coupling. This photon momentum matching condition is known as the phase matching condition. Second harmonic generation (SHG) is the special case when \( \omega_1 = \omega_2 \).

Third harmonic generation (THG) can use two successive second order nonlinear steps. This indirect conversion process requires lower input laser intensity and can obtain higher conversion efficiency compared with the direct conversion which utilizes the third order nonlinearity of the medium. The generated second harmonic field (2\( \omega_1 \)) in the first step will couple with unconverted fundamental field (\( \omega_1 \)) and generate new frequency component at 3\( \omega_1 \) as shown in Fig. 1.1. The conversion efficiency varies with the different frequency processes, the nonlinear material and high-power laser sources. In the case of a long pump pulse, for example 100-ps and longer with narrow bandwidth, phase matching is the primary requirement for efficient conversion. Overall energy efficiencies of 80 percent were obtained both for second and third harmonic generation.\textsuperscript{14,15}

![Diagram](image)

**Fig. 1.1.** Schematical setup for third harmonic generation

For ultrashort pulses with broad bandwidth, the wave vector dependence on the frequency is, \( k = k_0 + \frac{\partial k}{\partial \omega} (\omega - \omega_0) + \frac{1}{2} \frac{\partial^2 k}{\partial \omega^2} (\omega - \omega_0)^2 + \cdots \), where \( k_0 \) is the wave vector at the carrier frequency \( \omega_0 \), \( \partial \omega / \partial k \) and \( \partial^2 \omega / \partial k^2 \) are group velocity and group velocity dispersion respectively. It is difficult to fulfill the phase matching and group
velocity matching simultaneously in a given direction within the nonlinear medium.\textsuperscript{5,16,17} The group velocity mismatch between three mixing pulses causes the three pulses spatially separate from each other after propagating a certain distance within the crystal. To overcome this difficulty and increase the up-conversion efficiency for ultrashort pulses, three schemes have been reported. One scheme is to introduce spectral angular dispersion by using gratings (or prisms) and lenses to match all wave vectors.\textsuperscript{18,19,20} Another is to introduce a predelay between two pump waves at the entrance of type II doubling crystal to compensate for the group velocity delay.\textsuperscript{21,22,23,24} High intensity ultrashort pulses also allow short interaction lengths to be used efficiently which reduces the effect of group velocity mismatch. Second-harmonic conversion efficiencies of \(~75\%\) for 4mm thick KDP crystal were reported with intensities up to several hundreds of GW/cm\textsuperscript{2}.\textsuperscript{25} At high intensities, third-order nonlinear effects such as self- and cross-phase modulation may lead to spectral broadening and influence the third harmonic conversion efficiency.

This thesis presents an approach to optimum third harmonic generation of 1-\textmu{}m light of ultrashort pulses in widely used KDP crystals and a study of related nonlinear effects. This is a special case of a more general class of short pulse nonlinear frequency upconversion processes. A formulation of the optical wave interaction in the nonlinear media based on the Maxwell equations is described to introduce the conditions required for optimizing the conversion process. The important considerations for efficient harmonic conversion such as group velocity dispersion for short pulses and self- and cross-phase modulation associated with high order nonlinearity are examined in the theoretical description. An experimental investigation is conducted to achieve high efficiency frequency conversion to the third harmonic in a KDP crystal at high intensity. At ultra-high intensities, the effects of phase modulation on THG are discussed.

It is important to know the direction of the crystal axis relative to the propagation direction of the laser to optimize the phase matching for frequency conversion processes. The wave polarized perpendicular to the plane containing the propagation vector and optical axis is called ordinary wave (o), and the wave polarized
in the plane containing the propagation vector and optical axis is called extraordinary
wave (e). The orientation of the optical axis in the crystal determines the spatial and
temporal walk-off between the o- and e-waves. The spatial and temporal walk-off limit
the useful crystal length for the nonlinear interaction process. A frequency-domain
interferometer method (FDI), was developed to locate the orientation of the optical
axis of the crystal and also to measure the group velocity walk-off of ordinary wave
(o-wave) and extraordinary wave (e-wave) when a short-pulse laser propagates
through a nonlinear crystal. Frequency-domain interferometry utilizes
interference fringes in the frequency domain to measure changes in a probe pulse. The
basic idea is that the e- and o- wave components of broad bandwidth, short pulses
passing through a birefringent medium undergo group velocity walk-off. These two
temporally separated pulses are combined in a spectrometer to create an interference
pattern. The spacing of the interference fringes depends on the group velocity
dispersion which depends on the propagation angle with respect to the optical axis. If
the refractive index and its dispersion are known, the angle and orientation of the
optical axis in the uniaxial crystal can be determined by further measuring the
dependence of the GVD on the incident angle with respect to the birefringent crystal.
Compared with the usual x-ray diffraction method which is used to determine the
optical axis of the crystal, the x-ray diffraction method requires an x-ray source and
sophisticated analysis which may not be readily available, but the FDI technique can be
used “in situ”, for example, with the ultrafast laser system to be frequency converted.

During frequency up conversion of intense pulses which involves two or more
optical waves, nonlinear phase changes caused by self-phase modulation (SPM) and
cross-phase modulation (XPM) can destroy the phase coherence required for efficient
conversion. It is important to measure the third order nonlinear susceptibility which
is responsible for SPM, XPM. A Z-scan technique was used to determine self -
phase modulation coefficients at 1 μm, 0.53 μm and 0.35 μm, and cross-phase
modulation coefficients between 1 μm and 0.53 μm in KDP crystals due to the third
order susceptibility (χ(3)).
In the single Gaussian beam Z-scan\textsuperscript{30,31} as depicted in Fig. 1.2, a sample is moved along the z direction of a focused Gaussian beam. The transmittance of the beam (D2/D1 in Fig. 1.2) through an aperture in the far field is measured as a function of the sample position z with respect to the focal plane. The intense beam induces both absorption and refraction nonlinearities in the nonlinear medium. For the refractive nonlinearity, the nonuniform refractive index is induced in the sample in the transverse direction of the propagation due to the Gaussian intensity profile of the laser beam. Therefore the refractive index nonlinearity induces a weak lensing effect in the sample. The nature of the lens depends on the sign of n\textsubscript{2}. For positive n\textsubscript{2} there is a positive lens and self-focusing; a negative n\textsubscript{2} leads to a negative lens and a self-defocusing.

When the sample is far from the focus (-z), the beam irradiance is low and negligible nonlinear refraction occurs; hence, the transmission remain relatively constant. As the sample moves close to the focus, the increased irradiance leads to self-lensing in the sample. A negative self-lensing prior to the focus will tend to collimate the beam, increasing the aperture transmittance. When the sample at the +z side of the focus, the negative lensing tends to increase the beam divergence and the aperture transmittance is decreased. The approximate null at z = 0 is analogous to placing a thin lens at the focus that results in a minimum far field pattern change. For larger +z position the irradiance is reduced and transmittance returns to the original linear value. For positive lensing effect, the aperture transmittance decreases when the sample at the -z side of the focus and increase when the sample at the +z side of the

![Diagram](image-url)
focus. Its behavior is opposite from that of negative lensing effect. Thus, from the resultant Z-scan curve, the sign and magnitude of $n_2$ of the sample can be determined.

If the sample also has an absorptive nonlinearity, the absorptive nonlinearity can be measured by an open aperture Z-scan. When the aperture is removed, Z-scan will only be sensitive to the nonlinear absorption. Fitting the results yields an accurate value of the nonlinear absorption coefficient.

These measured third-order nonlinearities, including the first measurement of self-phase modulation coefficients at 0.53 μm, 0.35 μm and cross-phase modulation coefficients between 1 μm and 0.53 μm in KDP were used as input for an improved theoretical model and the resulting conversion efficiency is compared with the experiment result.

1.2 HISTORY OF RESEARCH ON HARMONIC GENERATION

Since the first observation of second-harmonic generation (SHG) by Franken and co-workers in 1961,¹ many theoretical and experimental developments have been reported. In the first experiments the amount of harmonic light observed was small. With the advent of phase matching techniques,²³ the conversion efficiency increased. The phase matching techniques also stimulated numerous applications including the creation of such important devices as optical parametric oscillators, optical image and signal converters, difference frequency generators, etc. Armstrong et al. found exact solutions of SHG by plane waves in 1962,⁵ while shortly thereafter, in 1965, Maker and Terhune reported their study of electric-field-induced SHG.⁴ This was followed by Boyd and Kleinman’s analysis in 1968, which considered the effects of diffraction and double refraction.³⁵ Based on this ground work, a lot of research has been done to study additional mechanisms associated with the conversion processes. These mechanisms include inhomogeneity of the nonlinear media,³⁶ temperature-dependent phase-matching,³⁷ the effects of inhomogenous laser radiation,³⁸ the group velocity dispersion³⁹ and the diffraction shift of the optimal phase relationship.

Conversion efficiencies exceeding 50% and higher are commonplace for long pump pulse with narrow bandwidth. For example, radiation from neodymium laser has
been converted from 1064 nm to 532 nm with an efficiency of 92%,\textsuperscript{40} from 1054 to 355 nm with an efficiency of 80%,\textsuperscript{14} and from 532 to 266 nm with an efficiency of 85%.\textsuperscript{41} For ultrashort pulses, it has long been known that a finite matching bandwidth limits the up-conversion efficiency.\textsuperscript{42,43} This limited phase matching bandwidth appears due to the group velocity mismatch between the fundamental and the harmonic.\textsuperscript{44} As the use of high-power ultrafast laser became common, several techniques were developed to overcome this problem. Volosov\textsuperscript{20} and other workers\textsuperscript{18,20,45} introduced a spectral angular dispersion by using gratings (and lenses to match wave vectors for all frequencies. Using this method, frequency doubling efficiency of 15% was achieved for 300-fs, 496-nm laser pulses generated by an excimer laser pumped cascaded distributed feedback dye laser system,\textsuperscript{19} and the conversion efficiency of second harmonic of Nd:glass laser radiation in a 28 mm-thick LiIO\textsubscript{3} crystal was increased from 0.05% to 0.55%. Wang et al.\textsuperscript{22,24} and Pronko\textsuperscript{21} et al. introduce a predelay between two pump waves at the entrance of the type II doubling crystal to compensate for the group velocity delay. The frequency doubling efficiency increased from 40% to 75% in a 1-cm long KDP for picosecond pulses.\textsuperscript{24} High intensity ultrashort pulses also allow short interaction lengths to be used efficiently with reduced group velocity mismatch. Second harmonic generation efficiencies of \textasciitilde75% for 4mm thick KDP crystal were reported with intensities up to several hundreds of GW/cm\textsuperscript{2}.\textsuperscript{25}

1.3 Outline

Chapter II is theoretical description of optical wave propagation and interaction in the nonlinear medium. The goal is to optimize the frequency conversion efficiency. The influences of phase matching, birefringent walk-off, group velocity dispersion, self- and cross-phase modulations are described.

Chapter III is devoted to the experimental investigation of second and third harmonic generation of ultra-intense picosecond pulses. Self- and cross-phase modulation were observed by measuring the spectrum and polarization state of the pulse at different intensities after the frequency conversion process. The importance of
the phase modulation is shown. The third harmonic conversion efficiency in KDP was reduced by placing glass windows between the crystals to enhance the SPM and XPM. Theoretical calculations are compared with the experiment results and discussed.

Chapter IV shows results of measurements of self- and cross-phase modulation in KDP crystals at different wavelength. Self-phase-modulation coefficients at wavelengths of 1.053 μm, 0.527 μm and 0.351 μm were measured by single beam z-scan and cross-phase-modulation coefficients between 1.053 μm and 0.527 μm were measured by a two color z-scan. These coefficients are important in frequency conversion of intense pulses. Most of the results were measured for the first time and are compared with a simple theoretical model. The SPM coefficient at 1.053 μm is in good agreement with experimental results reported by other authors.

Chapter V describes a technique based on frequency domain interferometer to determine the orientation of optical axis and group velocity walk off of a uniaxial crystal. The amount of group velocity walk off, which is determined by the orientation of the axis, has strong effects on ultrafast frequency conversion. Group velocity walk off has been measured in several crystals, and good agreement between theory and experiment has been found.
REFERENCES


Chapter II
The General Description of Frequency Up-Conversion

2.1 Introduction

The interaction between an electromagnetic field and an atomic system was studied perturbatively in the early years of modern quantum mechanics.\textsuperscript{1,2,3} In recent years with the development of lasers, great interest has been shown in the interaction of optical radiation with dielectric media which show an induced polarization. In 1962 Armstrong et al.\textsuperscript{4} first derived the formal theory of sum-frequency generation for three interacting waves. Thereafter, other workers have studied the additional frequency conversion processes associated with the nonlinear interaction processes.

In this chapter, we will review the formalism which has been established for the nonlinear optical interaction, especially the three wave interaction. We first provide, in section 2.2, a brief description of the physical origin of some of the nonlinear optical effects. Various factors limiting the energy conversion efficiency in the nonlinear interaction between optical beams are described. In section 2.3.1 we will discuss the phase matching requirement for efficient frequency conversion. Section 2.3.2 discusses the effect of birefringent walk off, group delay during interaction between short pulses, and the influence of the cubic nonlinearity of a noncentrosymmetric medium on these frequency conversion processes. In section 2.4, we discuss the material considerations for the process. The frequency tripling method is introduced in the last section.

2.2 Nonlinear Polarization

In the presence of an electromagnetic field, a dielectric material will become polarized. The electrons in the medium are polarized with respect to the nuclei. The
induced polarization $\mathbf{P}(\mathbf{r},t)$ can be expanded in a power series in the electric field of the form

$$\mathbf{P}(\mathbf{r},t) = \mathbf{\chi}^{(1)}: \mathbf{E}(\mathbf{r},t) + \mathbf{\chi}^{(2)}: \mathbf{E} \mathbf{E} + \mathbf{\chi}^{(3)}: \mathbf{E} \mathbf{E} \mathbf{E} + \ldots$$

(2.1)

Here $\mathbf{E}(\mathbf{r},t)$ is the electric field of the optical wave, and $\mathbf{\chi}^{(1)}$ is the linear susceptibility which describes the linear optical effects. $\mathbf{\chi}^{(2)}$ and $\mathbf{\chi}^{(3)}$ are the nonlinear susceptibilities of the medium which govern three- and four-wave interactions respectively. Tensor notations are used in Eq. (2.1) to described anisotropic media adequately. In such media, $\mathbf{P}$ and $\mathbf{E}$ are, in general, not parallel and are connected through tensor relationships. The susceptibilities $\mathbf{\chi}^{(1)}$, $\mathbf{\chi}^{(2)}$, $\mathbf{\chi}^{(3)}$, ... are tensors of the second, third, and fourth ranks, respectively. In general, $\mathbf{\chi}^{(1)}$, $\mathbf{\chi}^{(2)}$, and $\mathbf{\chi}^{(3)}$ are dependent on the frequencies of the optical radiation. In the case where the electric field is small, only the first linear term needs to be considered. This is the regime for linear optics including the effects of refraction, linear absorption, dispersion, and birefringence of the medium.

The scalar relation between linear polarization $\mathbf{P}^{(1)}$ and electric field $\mathbf{E}$ is

$$P_i = \sum_j \chi_{ij} E_j$$

(2.2)

where $i,j$ refer to the Cartesian components of the fields ($E_x$, $E_y$, and $E_z$). the magnitude of the $\chi_{ij}$ depends on the choice of the $x$, $y$ and $z$ axes relative to the crystal structure. For lossless, non-optically active material, it is always possible to choose $x$, $y$, $z$ in such a way that the off-diagonal elements vanish, leaving

$$P_i = \chi_{ij} E_j$$

(2.3)

These directions are called the principle dielectric axes of the crystal. $\chi_{ij}$ is related to the dielectric permittivity tensor $\varepsilon_{ij}$ as $\varepsilon_{ij} = 1 + 4\pi\chi_{ij}$. The principal refractive indices $n_1$, $n_2$, $n_3$ along the principal dielectric axes of the crystal are defined as $n_i = \sqrt{\varepsilon_{ii}}$. If $\chi_{ii}$ is complex, the complex refractive index $n_i$ which is denoted as $n_{0i} + i\kappa_i$ is related to the linear susceptibilities by $n_{0i} = [1 + 4\pi \text{Re}(\chi_{ii})]^{1/2}$ and $\kappa = 2\pi \text{Im}(\chi_{ii})/n_{0i}$. When $\varepsilon_{11} = \varepsilon_{22} = \varepsilon_{33}$, all orthogonal axes of the system are principal axes, and the crystal is said to be optically isotropic. When $\varepsilon_{11}$, $\varepsilon_{22}$, $\varepsilon_{33}$ are not all equal, the crystal is said to be
birefringent. Birefringent crystals are generally classified into two categories: biaxial and uniaxial. In the general case that the three principle indices of refraction are all different $n_1 \neq n_2 \neq n_3$ (or $\varepsilon_1 \neq \varepsilon_2 \neq \varepsilon_3$), there are two optical axes, hence the crystal is called biaxial. Along these two directions, the two allowed orthogonal polarizations travel with the same phase velocity. In many optical materials two of the principal indices are equal, $n_1 = n_2$. There is a unique direction ($z$) along which the two allowed orthogonal polarizations travel with the same phase velocity. This is called a uniaxial crystal.

In the common case in which the nonlinearity is electronic in origin, the lowest-order nonlinear terms $P^{(2)}$ (=$\chi^{(2)}$:EE) would be comparable to the linear response $P^{(1)}$ (=$\chi^{(1)}$:E ) when the optical field strength is comparable to the characteristic intratomic electric field $E_{at} = e/a_0^2$, where $-e$ is the charge of the electron and $a_0 = h^2/m_e^2$ is the Bohr radius of the hydrogen atom. Numerically $E_{at} = 2 \times 10^7$ esu. The second order nonlinearity is responsible for the phenomenon of harmonic generation, sum- and difference frequency generation and parametric generation, while the third order nonlinearity is responsible for nonlinear refraction, phase modulation, stimulated Raman scattering, and phase conjugation. The magnitude of the tensor components rapidly decreases with the increasing rank of the nonlinearity ($\chi^{(1)}: \chi^{(2)} : \chi^{(3)} = 1 : 1/E_{at} : 1/E_{at}^2 = 1:10^8 : 10^{15}$; $\chi^{(1)}: \chi^{(2)} : \chi^{(3)}$ are all in electric static units). It defines the intensity region in which nonlinear processes will occur.

When the field contains more than one frequency component, the relationship between second order nonlinear polarization and the nonlinear susceptibility $\chi^{(2)}$ can be expressed in component form as\(^5\)

$$P_i^{(2)}(\omega_n + \omega_m) = \sum_{j,k} \sum_{(nm)} \chi_{ijk}(\omega_n + \omega_m) E_j(\omega_n) E_k(\omega_m) \quad (2.4)$$

where the indices $i,j,k$ refer to the Cartesian components of the fields. The notation (nm) indicates that, in performing the summation over $n$ and $m$, the sum $\omega_n + \omega_m$ is to be held fixed, although $\omega_n$ and $\omega_m$ are each allowed to vary. The nonlinear coupling coefficients $d_{ijk}$ are commonly used instead of $\chi_{ijk}\(^6\)$ and are defined as, $d_{ijk} = \chi_{ijk}/2.\(^5\)

The third order nonlinear polarization can be expressed as
\[ P_i^{(3)}(\omega_o + \omega_n + \omega_m) = \sum_{j \neq i} \sum_{\phi} \chi_{j\phi}^{\text{oo}}(\omega_o + \omega_n + \omega_m, \omega_o, \omega_n, \omega_m) E_j(\omega_o) E_k(\omega_n) E_l(\omega_m) \] (2.5)

In general, the number of independent elements of the nonlinear susceptibility tensor is determined by the symmetry of the crystal. For example, in a lossless medium, second order nonlinear susceptibility tensor \( \chi^{(2)} \) has 27 elements. However, any crystalline symmetries of the nonlinear material reduce the number of independent elements.\(^7\)\(^8\) For crystal class 42m such as KDP, \( \chi^{(2)} \) has six elements of which three are independent. For a centro-symmetric media, the second order nonlinearity (\( \chi^{(2)} \)) is zero.

### 2.3 Three-Wave Interaction

The propagation of the optical field in a dielectric medium without free charges can be generally described by Maxwell’s equation:

\[ \nabla^2 \mathbf{E}(r, t) - \frac{1}{c^2} \frac{\partial^2 \mathbf{E}(r, t)}{\partial t^2} = \frac{4\pi}{c^2} \frac{\partial^2 \mathbf{P}(r, t)}{\partial t^2}. \] (2.6)

Many nonlinear effects involve the interaction of two or more optical fields with different wavelengths. In order to calculate the optical fields involved in the nonlinear interactions, we assume that the electric field can be expressed in the form

\[ \mathbf{E}(r, t) = \frac{1}{2} \sum_i \left[ \mathbf{E}_i(r, t)e^{i(k_i - \omega_i)t} + \text{c.c.} \right] \] (2.7)

Here the summation \( i \) is over the fields incident on the medium and the generated fields, the wave number is \( k_i = n_i\omega/c \), where \( n_i \) is the refractive index with \( n_i^2 = \varepsilon_i = (1 + 4\pi \chi^{(1)}(\omega_i)) \), where \( \varepsilon_i \) is the dielectric permittivity and \( c \) is the speed of light.

The polarization is often written as

\[ \mathbf{P}(r, t) = \mathbf{P}^{(1)}(r, t) + \mathbf{P}^{\text{NL}}(r, t) \]

\[ \mathbf{P}^{(1)}(r, t) = \frac{1}{2} \sum_i \left[ \chi^{(1)}(\omega_i) \cdot \mathbf{E}_i(r, t)e^{i(k_i - \omega_i)t} + \text{c.c.} \right] \]

\[ \mathbf{P}^{\text{NL}}(r, t) = \frac{1}{2} \sum \left[ \mathbf{P}^{\text{NL}}(r, t)e^{i(k_f - \omega_f)t} + \text{c.c.} \right] \] (2.8)
Here $P_{i}^{NL}$ is the nonlinear polarization with frequency $\alpha$. It is determined by an appropriate combination of optical fields and nonlinear susceptibilities according to Eq.(2.4). $P_{i}^{NL}$ acts as the source term for the optical field with frequency $\alpha$. The wave vector of the nonlinear polarization $k_{i}$ is, in general, different from the wave vector of the optical field at the same frequency since wave momentum conservation is not strictly required in a finite medium. This difference in wave vectors (phase mismatch) plays a very important role in determining the effectiveness of many nonlinear optical interactions.

A simple assumption is usually made to calculate the amplitudes of the various waves. The slowly varying envelope approximation assumes that the field envelopes vary slowly in time compared to the optical frequency and slowly in space compared to the optical wavelength. It is generally valid for all of the interactions that are encountered in the laboratory, but not for pulse duration of tens of femtoseconds and less. It enables us to neglect the second derivatives of the field amplitudes with respect to $z$ and $t$ when Eqs. (2.7) and (2.8) are substituted into Eq. (2.6). If we further assume the wave is a plane wave propagating along $z$, we obtain the following expression for the three wave interaction such as sum frequency generation in which waves 1 and 2 combine to generate wave 3 in a nonlinear medium, $^{9}$

$$\frac{\partial E_1}{\partial z} + \frac{1}{v_{g1}} \frac{\partial E_1}{\partial t} = -\frac{1}{2} \gamma_1 E_1 - iK_1 E_2 E_2^* \exp(-i\Delta k z) + i(\alpha_{11}|E_1|^2 + \alpha_{12}|E_2|^2 + \alpha_{13}|E_3|^2) E_1$$

(2.9)

$$\frac{\partial E_2}{\partial z} + \frac{1}{v_{g2}} \frac{\partial E_2}{\partial t} = -\frac{1}{2} \gamma_2 E_2 - iK_2 E_3 E_3^* \exp(-i\Delta k z) + i(\alpha_{21}|E_1|^2 + \alpha_{22}|E_2|^2 + \alpha_{23}|E_3|^2) E_2$$

(2.10)

$$\frac{\partial E_3}{\partial z} + \frac{1}{v_{g3}} \frac{\partial E_3}{\partial t} = -\frac{1}{2} \gamma_3 E_3 - iK_3 E_1 E_2 \exp(i\Delta k z) + i(\alpha_{31}|E_1|^2 + \alpha_{32}|E_2|^2 + \alpha_{33}|E_3|^2) E_3$$

(2.11)
where \( v_\phi = (\partial k / \partial \omega)^{-1} \) (i = 1, 2, 3) is the group velocity, \( \gamma_i (= 2 \pi \omega \text{Im}(\chi^{(1)})/n_0 c) \) is the linear absorption coefficient. \( \Delta k = k_3 - k_1 - k_2 \) is the wave vector mismatch between three optical fields. \( K_i = 2 \pi \omega_i^2 \chi^{(2)} / k_i c^2 \) is the constant of nonlinear coupling and \( \omega_i \) gives the change of the refractive index for wave \( i \) that is due to the intensity of wave \( j \) resulting from the third-order susceptibility \( \chi^{(3)} \).

Second harmonic generation occurs when \( \omega_1 = \omega_2 = \omega \), and \( \omega_3 = 2\omega \). The third harmonic can be generated through two consecutive processes. The first step is to make the second harmonic generation, and the output from the first step will become input optical wave in the second step. The generated second harmonic field (\( \omega_1 = 2\omega \)) in the first step will couple with unconverted fundamental fields (\( \omega_2 = \omega \)) and generate a new frequency at \( \omega_3 = 3\omega \).

Eq. (2.9)-(2.11) are the fundamental equations describing the propagation and interaction of the optical fields involved in three-wave mixing processes. The frequencies of the fields satisfy \( \omega_3 = \omega_1 + \omega_2 \). The energy conversion efficiency is defined as the ratio of the generated pulse energy of wave 3 to the total input pulse energy. The optimum conditions for the interaction process can be determined with respect to the intensity of the pulse, the energy, the pulse width as a function of the crystal thickness.

### 2.3.1 Phase matching

Physically, the three wave interaction processes can be viewed as resulting from a distortion in the electric charges in the medium driven by the optical fields at frequencies \( \omega_1, \omega_2 \) and \( \omega_3 \). These distortions lead to an oscillating polarization at any combination of the frequencies, which in turn radiates optical waves at those frequencies. Of all the waves radiated at the different frequency combinations, only those with correct phase relationship will experience gain. In fact, only those interactions which satisfy the phase matching condition \( \Delta k = 0 \) will undergo macroscopic amplification as they propagate through the medium.
For general case of a three coupled wave interaction, \( \omega_3 = \omega_1 + \omega_2 \), the phase matching condition may be written as:
\[
\Delta k = k_1 + k_2 - k_3 = 0
\]  
(2.12)
where the wave vectors \( k_1, k_2 \) may be different not only in magnitude but also in direction (in an anisotropic media). The wave vector can be written
\[
k_i = \frac{2\pi n_i}{\lambda_i} \hat{k}_i
\]  
(2.13)
where \( \hat{k}_i \) is the unit vector parallel to \( k_i \), \( n_i \) is the refractive index and \( c \) is the velocity of light. Eq.(2.12) can be used to formulate the requirements for dispersion in the media to obtain the phase matching necessary for efficient frequency conversion. Collinear phase matching is often required to avoid reduction of effective beam interaction length caused by the finite cross section of the beam. Collinear phase matching can be achieved by utilizing the birefringence of anisotropic crystals.\(^{10} \)

In a birefringent medium, the index of refraction (and thus the phase velocity) for a wave at a given frequency depends on its state of polarization as well as the direction of propagation in the crystal. For an arbitrary propagation direction in a birefringent crystal, two orthogonal linear polarization directions are permitted. The general procedures for determining the orientation, refractive indices, and phase velocities of the two allowed orthogonal linear polarizations are described in detail in Refs. [11, 12]. For biaxial crystals in which the three principal indices of refraction are all different, there are two optical axes, Along these two directions, the two allowed orthogonal polarizations travel with the same phase velocity. In uniaxial crystals, the two of the principal indices are equal, \( n_1 = n_2 \). There is a unique direction (\( z \)) along which the two allowed orthogonal polarizations travel with the same phase velocity. Phase matching can be realized in both biaxial and uniaxial crystals. Uniaxial crystals are more commonly used.

In uniaxial crystals, the ordinary wave is polarized perpendicular to the plane containing the propagation vector and the optical axis (Figure 2.1). This wave experiences the ordinary refractive index. The extraordinary wave is polarized in the plane containing \( k \) and optical axis. Unlike the o-wave, e-wave experiences a refractive
index $n_e(\theta)$ that depends on the angle $\theta$ between the optical axis and propagation direction. The value of $n_e(\theta)$ can be expressed as:

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

(2.14)

where $n_e$ is the principal refractive index of the extraordinary wave. It can be seen that for $\theta = 0^\circ$, $n_e(0^\circ) = n_o$, and for $\theta = 90^\circ$, $n_e(90^\circ) = n_e$.

If $n_o < n_e$, the crystal is considered to be positive, whereas if $n_o > n_e$, it is considered to be negative. The normal index surfaces\textsuperscript{13} of a positive and a negative uniaxial crystal are shown in Fig. 2.2.

For sum frequency generation, two types of collinear phase matching are commonly used in which suitably polarized waves satisfy $\omega_3 = \omega_2 + \omega_1$ and have the same refractive index, thus satisfying $\Delta k = 0$. For the case of a negative uniaxial crystal ($n_e < n_o$) with normal dispersion in which $n_e(\omega)$ and $n_o(\omega)$ increase monotonically with $\omega$, the wave at $\omega_3$ needs to have extraordinary polarization.\textsuperscript{5} The two waves at $\omega_1$ and $\omega_2$ can either be both ordinary waves, or one ordinary wave and another extraordinary wave. Midwinter and Warner\textsuperscript{14} define type I phase-matching

Fig. 2.1. The optical axis is along z axis, the wave vector $k$ is on the yz (or xz) plane with angle $\theta$ to the optical axis. The extraordinary wave is polarized in the plane containing the wave vector $k$ and the optical axis, while the ordinary wave has a polarization perpendicular to the kz plane.
Fig. 2.2. Cross-section of the normal index surfaces (a) a positive uniaxial crystal and (b) a negative uniaxial crystal in the yz (or xz) plane. The index surface for the ordinary wave is a sphere, while that for the extraordinary wave is a spheroid. The index of a wave propagating along Θ is the distance from the origin of the intersection between the wave normal and the respective index surface.

to be the case where the lower frequency waves have the same polarization (the polarization of ω₁, ω₂ are parallel to each other) and type II phase-matching to be the case where they are orthogonal. Table 2.1 summarized the possible phase-matching schemes in uniaxial crystals.

Table 2.1. Phase-matching methods for Uniaxial Crystals

<table>
<thead>
<tr>
<th></th>
<th>Positive Uniaxial</th>
<th>Negative Uniaxial</th>
</tr>
</thead>
<tbody>
<tr>
<td>(nₑ &gt; n₀)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Type I</td>
<td>n₁ω₁ = n₁ω₁ + n₁₂ω₂</td>
<td>n₁ω₁ = n₁ω₁ + n₁₂ω₂</td>
</tr>
<tr>
<td>Type II</td>
<td>n₁ω₁ = n₁ω₁ + n₁₂ω₂</td>
<td>n₁ω₁ = n₁ω₁ + n₁₂ω₂</td>
</tr>
</tbody>
</table>

For a collinear three-wave mixing process, the phase-matching condition is: n₁ = (ω₁/ω₀)n₂+(ω₁/ω₀)n₀. The type II phase matching in a negative uniaxial crystal can be written as:
\[ n_z^{\omega_1}(\theta_m) = \left[ \left( \frac{\cos \theta_m}{n_o^{\omega_1}} \right)^2 + \left( \frac{\sin \theta_m}{n_r^{\omega_1}} \right)^2 \right]^{-1/2} = \left( \frac{\omega_2}{\omega_1} \right) n_z^{\omega_2}(\theta_m) + \left( \frac{\omega_1}{\omega_3} \right) n_o^{\omega_1} \] (2.15)

The frequencies of the optical waves are known. The refractive index of the crystals \( n_o \) and \( n_r \) at a given frequency can be obtained from the dispersion relation of the material. Therefore the phase matching angle \( \theta_m \) can be calculated using Eq. (2.15). In the experiment, this is realized by rotation of the crystal (angular tuning) to orient the wave vectors at an angle \( \theta_m \) to the optical axis in the crystal.

2.3.2 Effect of phase mismatch

If the phase between the optical waves is mismatched as they propagate through the nonlinear medium, \( \Delta k \neq 0 \), the interacting waves periodically step out of phase and interfere constructively as they travel through the medium. This causes the waves to exchange energy back and forth, and the generated wave undergoes amplitude oscillations along the propagation direction. Consider a simple case in which plane waves interact in a lossless medium, the Eqs. (2.9) - (2.1) can be simplified as

\[ \frac{\partial E_1}{\partial z} = -iK_1 E_2 E_2^* \exp(-i\Delta k z) \] (2.16)

\[ \frac{\partial E_2}{\partial z} = -iK_2 E_3 E_1^* \exp(-i\Delta k z) \] (2.17)

\[ \frac{\partial E_3}{\partial z} = -iK_3 E_1 E_2 \exp(i\Delta k z) \] (2.18)

In the nondepletion condition in which the magnitude of the generated wave \( \omega_3 \) is much smaller than the pump waves, the amplitude of the pump waves \( E_1 \) and \( E_2 \) can be taken as constants. Therefore the amplitude of the sum frequency (\( \omega_3 \)) field at the exit of the nonlinear medium is given by

\[ E_3(L) = -iK_3 E_1 E_2 \int \exp(i\Delta k z) dz = -iK_3 E_1 E_2 \left[ \frac{\exp(\Delta k L)}{i\Delta k} - 1 \right] \] (2.19)

and its intensity is proportional to
\[ I_{\omega_1} \propto I_{\omega_1} I_{\omega_2} K_1^2 L^2 \left( \frac{\sin(\Delta k L / 2)}{\Delta k L / 2} \right)^2 \]  

where \( L \) is the crystal length. The nondepletion condition is \( I_{\omega_1} I_{\omega_2} K_1^2 L^2 \ll 1 \). \( L_c = 2\pi/\Delta k \) is known as the coherence length. The phase relationship between the three waves is destroyed in a coherence length. It can be seen from Eqs. (2.10) and (2.12) that \( \Delta k \) depends on the propagation angle and, \( \Delta k \) will increase if the propagation angle deviates from the phase matching angle. Thus, the conversion efficiency will drop dramatically as a function of propagation angle. The angle here is measured inside the crystal and called the internal angle. Fig. 2.3 shows the effects of wavevector mismatch \( \Delta k(\theta)L \) on the energy conversion efficiency. The maximum conversion occurs at \( \theta_m (\Delta k(\theta_m) = 0) \). The degree of sensitivity of phase-matching on angular deviations is often measured in terms of \( \partial(\Delta k)/\partial \theta \) which is defined as the angular acceptance bandwidth of the crystal and can be calculated from the phase-matching geometry and the refractive index data. The smaller the magnitude of the \( \partial(\Delta k)/\partial \theta \) at certain angle, the lower the sensitivity of the phase matching to angular variations and the larger the acceptance bandwidth.

![Graph](image)

Fig. 2.3. The conversion efficiency as a function of wavevector mismatch (or tuning angle). From Ref. 5.

### 2.3.3 Effect of birefringent walk-off

For any direction of phase propagation, the direction of energy flow (Poynting vector \( \mathbf{S} \)) is given by the normal vector to the index surface\(^{11} \) as illustrated in Fig. 2.4. In a birefringent crystal, the phase propagation direction and the Poynting vector are
parallel for the ordinary wave. However, the propagation direction generally deviates from the Poynting vector for the e-wave. If the e-wave propagates at an angle \( \theta \) to the optical axis, the extraordinary wave will experience Poynting vector walk-off. The double refraction angle \( \rho \) which defined as the angle between Poynting vector and phase propagation direction is given by:\(^{15}\)

\[
\tan \rho = \frac{n_e^2(\theta)}{2} \left[ \frac{1}{n_e^2} - \frac{1}{n_0^2} \right] \sin 2\theta
\]  

(2.15)

The main consequence of this Poynting vector walk-off is that, in a collinear phase-matched interaction, although the waves of the coupled fields are propagating in the same direction, the corresponding waves walk off from one another as they propagate through the crystal. This results in a reduction of the interchange of energy between the interacting fields with a finite aperture along the direction of propagation. This limits the maximum useful crystal length for the nonlinear process with a small beam aperture.

From Eq.(2.15), the walk-off angle is maximum for propagation at 45° to the optical axis and is zero for propagation either along or at 90° to the optical axis. Thus, in order to overcome the deteriorous effects of walk-off, either a beam with large aperture or the 90° phase matching is used. The 90° phase matching is called noncritical phase matching (NCPM) and can sometimes be obtained by temperature tuning of the refractive index.\(^5\)
2.3.4 Effect of Frequency conversion of ultrashort pulse

An ultrashort pulse has a broad bandwidth because the spectrum of the pulse is the Fourier transform of the temporal shape. For pulses with appreciable spectral width and high peak intensities, the effects of group velocity walk off and third order nonlinear susceptibility need to be considered.

2.3.4.1 Effect of group velocity walk off

For ultrashort pulses, the frequency dependence of the wave vector is

\[ k = k_0 + (\partial k / \partial \omega)(\omega - \omega_0) + (\partial^2 k / \partial \omega^2)(\omega - \omega_0)^2/2 + ... \]  (2.16)

and \((\partial^2 k / \partial \omega^2)\) is the group velocity dispersion. The conversion efficiency for such pulses usually decreases due to the mismatch of the group velocities of the pulses even though the central-frequency wave vectors of the three mixing waves are well matched. Consider three waves that are mixed in a type II negative uniaxial crystal, the group velocities of the fundment e-, o-wave and generated e-wave are defined as \(v_{te}\), \(v_{to}\) and \(v_{3e}\), respectively with the corresponding pulse duration of \(\tau_1\), \(\tau_2\) and \(\tau_3\). The three waves may spatially separate from one another after propagating a certain distance in the crystal as shown in Fig. 2.5. At the distance \(z = L \tau = \tau / |1/v_{te} - 1/v_{to}|^{-1}\), the retardation \(\tau\) between wave 1 and wave 2 equals the pulse duration. The interaction length is limited by the condition \(\tau \ll \tau_{1,2,3}\).

![Fig. 2.5. Illustration of the group velocity mismatch effect in the frequency conversion process. In normal dispersion negative uniaxial crystal, fundamental e-wave travels fast than the fundamental o-wave and generated e-wave moves in between.](image-url)
The refractive index of the e-wave is a function of propagation direction in uniaxial crystals, and thus the group velocity of the e-wave is as well. When $\theta = 90^\circ$, the group velocity walk off between o- and e-wave will be a maximum. For subpicosecond pulses, group velocity dispersion is small and can be ignored in most of the crystals. Introducing a predelay between the two orthogonally polarized fundamental pulses at the entrance of the interacting crystal to compensate for the group velocity mismatch, the frequency conversion efficiency can be increased. A thin crystal ($L \ll \tau_{\phi2} \left\{ 1/v_{1e} - 1/v_{2o} \right\}^{-1}$) is also used to minimize the group velocity mismatch and increase the conversion efficiency.

In the frequency domain, the finite spectral bandwidth of the conversion crystal limits the frequency conversion efficiency of short pulses with broad bandwidth because the phase matching condition cannot be satisfied over the whole spectral range. By introducing angular spectral dispersion by using gratings (or prisms) it is possible to achieve phase matching for all wave vectors.

### 2.3.4.2 Effect of third order nonlinearity

When the light applied to the nonlinear medium is intense enough, the nonlinear polarization due to the third order nonlinearity becomes significant. The third order nonlinearity contribution comes from different physical mechanisms within the medium such as electronic polarization, molecular orientation, electrostriction, and thermal effects. It is responsible for the nonlinear phenomena such as direct third harmonic generation, four wave mixing and nonlinear refraction and absorption. Among them, nonlinear refraction and absorption do not require any phase matching. They only depend on the incident light intensity and can have a strong effect on the propagation characteristics of the light wave, including both absorption and changing the focusing properties. They can also change the spatial, temporal, and spectral distribution of the incident waves, as well as their polarization state. Nonlinear refraction is caused by the real part of the nonlinear susceptibility and nonlinear absorption caused by the imaginary part of the susceptibility. Different waves can undergo different nonlinear phase changes, leading to a phase mismatch.
At high intensities the total intensity dependent refractive index $n$ is equal to $n_0 + \Delta n(I)$, where $n_0$ is the linear refractive index and $\Delta n$ can be expanded as $\Delta n = n_2 I + n_4 I^2 + \ldots$, where $n_2 I \gg n_4 I^2$. The nonlinear refractive index $n_2$ is related to the real part of the third order nonlinearity $\chi_{1111}^{(3)}$ by the relation, $n_2 = 3/8 n_0 \chi_{1111}^{(3)}$, where $\chi_{1111}^{(3)}$ is the real part of the third order nonlinearity $\chi_{1111}^{(3)}$. The nonlinear index of refraction can be either positive or negative depending on characteristics of the medium. In dielectric materials, the nonlinear index is due to electronic polarization. The optical field interacts with the electronic energy levels and causes a distortion of the electron cloud, which results in a change in the refractive index. Nonlinearities due to the molecular orientation usually occur in a liquid with anisotropic molecules. They are caused by the tendency of molecules to become aligned to the electric field of an applied optical wave. The optical wave then experiences a modified value of the refractive index because the average polarizability per molecule has changed. A nonlinear index can also arise from electrostriction, in which the molecules of the medium move into the most intense regions of the electric field. The resulting increase in density causes an increase in the refractive index. The response time of the nonlinearities depends on the physical processes, which is in the order of $10^{-15}$ sec for electronic polarization, $10^{-12}$ sec for molecular orientation and $10^{-9}$ sec for electrostriction.\(^5\)

The intensity dependent nonlinear refractive index can affect the beam propagation in a large number of ways. Self-focusing occurs in a medium with positive $n_2$. The incident beam such as an optical beam with Gaussian spatial profile is more intense in the center than at the edge. The central part of the beam with a higher intensity experience a larger refractive index and, therefore, travel with a slower phase velocity than at the edge. In this case the intense light beam creates its own positive lens in the nonlinear medium. Self-focusing occurs only when the focusing is strong enough that it overcomes the tendency of the beam to increase in size due to the diffraction. This requirement leads to an existence of the critical power defined by $P_c = c\lambda^2/8\pi^2 n_2$.\(^{21}\) For incident powers above the critical power, the self-focusing overcomes diffraction and the beam will be focused. For incident powers below the critical power,
the self-focusing cannot overcome the spreading due to the diffraction and the beam will not be focused although it spreads more slowly than it would in the absence of the nonlinear index.

Self-phase modulation (SPM) and cross-phase modulation (XPM)\textsuperscript{22} are two other commonly observed effects due to the nonlinear refraction. SPM refers to the self-induced phase shift experienced by an optical field during its propagation and XPM refers to the nonlinear phase shift due to an optical field at a different wavelength or polarization. If we consider optical fields at \(\omega_1\) and \(\omega_2\) propagating in a nonlinear medium, the nonlinear phase shift for the field at \(\omega_1\) is given by

\[
\Delta \Phi_1(t) = -\frac{2\pi}{\lambda} \int \left[ n_{\text{spm}}(z)f_1(z,t) + 2n_{\text{xpm}}f_2(z,t) \right] dz
\]

where the first term is SPM and the second term is the contribution of XPM. \(n_{\text{spm}}\) and \(n_{\text{xpm}}\) are related to tensor components of \(\chi^{(3)}\). For the simple case when the optical fields at \(\omega_1\) and \(\omega_2\) are orthogonal, \(n_{\text{spm}} = 3/8n_0\chi^{(3)}\) and \(n_{\text{xpm}} = 3/8n_0\chi^{(3)}\)\textsuperscript{22}.

In a birefringent medium, if the initial polarization state of the incident beam is not one of the polarization eigenstates, this beam can be decomposed by two linearly and orthogonal polarized beams. The effects of SPM and XPM generate phase difference between these two orthogonal polarized components and change the beam polarization state during its propagation through the medium.

During second order nonlinear interaction processes such as frequency up-conversion, third order nonlinearities can have effects on the process if the interacting waves are at high intensities. Efficient frequency conversion is achieved through phase matching (\(\Delta k = 0\)). In the presence of a nonlinear phase shift, it is possible, in principal to impose a linear phase mismatch to compensate for this nonlinear detuning. Since generally the optical beam has a non-uniform profile in both time and space, it will experience a different nonlinear phase shift at each position. It is impossible to compensate for the full range of intensities in space and time simultaneously. As a result, the net phase will be mismatched and the conversion efficiency will be decreased.
2.4 **NONLINEAR MATERIAL CONSIDERATIONS**

The basic requirement for a material to be used in second order nonlinear processes such as frequency conversions is that it should be noncentrosymmetric. In practical devices, the performance of the nonlinear crystals depends critically on material characteristics such as transparency, ability to support phase-match, high optical quality, nonlinearity, and availability in bulk form.

The material to be selected must be transparent and exhibit low optical losses at the wavelengths of interest. The losses which are usually due to absorption or scattering can severely affect the conversion efficiency of the nonlinear process or the threshold for oscillation in parametric devices. Moreover, absorption losses can cause thermal distortions in the material, which dramatically limit the device performance. The optical damage threshold of a material for a nonlinear application is also an important factor to be considered. Because of the high optical intensities involved in the nonlinear processes, the material must be able to withstand power densities corresponding to the experimental conditions. The optical damage may be surface or bulk damage which can be caused by thermal heating, induced absorption, self-focusing, dielectric breakdown\textsuperscript{23} or other mechanisms.

For efficient frequency conversion, the nonlinear crystal must possess sufficient birefringence in order to allow phase matching over the range of interaction frequencies. Material dispersion is normally defined by the Sellmeier or Zernike equations.\textsuperscript{11,24} The degree of the birefringence of the crystal and its ability to support phase-match can be determined from the refractive index at the appropriate wavelength. Another important factor to ensure high efficiency is that the material possesses a large enough effective nonlinear coefficient in the phase matching direction. The effective nonlinear coefficient depends on the symmetry group of the material, type of the crystal (negative uniaxial or positive uniaxial), and the type of the phase-matching.

The choice of the material for any conversion process also depends on many specific details. For example, crystals with small spatial walk off between o- and e-
waves or non-critical phase-matching are preferred if the laser beam quality is poor. The beam spatial walk-off can be ignored and the beam divergence can be made much less than the angular acceptance of the crystal if the optical beam is well collimated and has a large aperture with a uniform profile.

2.5 FREQUENCY TRIPLING METHOD

For third harmonic generation, a two step three wave interaction (type II-type II) was proposed to optimize the frequency tripling efficiency. The input fundamental pulse is linearly polarized at 35.3° with respect to the “o” direction of the first doubling crystal. This ensures that two o-photons are input for every e-photon. By properly choosing the thickness of doubling crystal, the e-photon will combine with one o-photon to give one e-photon at 2ω, leaving one o-photon unconverted. The plane of the optical axis of the tripler is perpendicular to that of the doubler. The e-wave (o-wave) output from the doubler will be the o-wave (e-wave) of the input for the tripling. Thus, the unconverted fundamental and the converted second harmonic pulses are mixed in the second tripling crystal and generate an e-wave at 3ω. By adjusting the orientation of the polarization plane of the fundamental input beam, the conversion of the first crystal can be controlled and overall efficiency can be optimized.

Fig. 2.6. Experimental setup for third harmonic generation. From Ref. 25.
In the frequency tripling experiment described here using 1 ps, 1 μm laser pulses, we used KDP crystals as the nonlinear conversion medium.\textsuperscript{26} Large, high optical quality KDP crystals can be grown easily. They have moderately high birefringence and adequate nonlinear, and are resistant to high intensity optical damage.\textsuperscript{27} Also, KDP offers an excellent match of group velocities between the interacting optical waves in broad bandwidth frequency conversion.

KDP is a negative uniaxial material which belong to point group 42m. The refractive index indices of KDP were measured accurately in the region of transparencies by Zernike.\textsuperscript{24} These data fit a five-parameter relation of the form

\[ n^2 = A + Bv^2/(1-v^2/C) + D/(E-v^2) \]  

(2.18)

where \( v = 1/\lambda \) in cm\(^{-1}\). The parameters A, B, C, D and E are given in Table 2.2.

| \( n_e \) | 2.132668 | 8.637494\times10^{-11} | 8.142631\times10^9 | 8.069981\times10^5 | 2.5\times10^5 |
| \( n_o \) | 2.259276 | 1.008956\times10^{-10} | 7.726408\times10^9 | 3.251305\times10^5 | 2.5\times10^5 |

Table. 2.2. Values of the constants A, B, C, D, and E for indices with respect to air\textsuperscript{24}

The index of refraction at different wavelengths can be obtained from Eq. (2.18). Based on the dispersion, the phase matching condition for type II doubling and tripling is:

Type II doubling: \[ \Delta k = \alpha/c(2n_e^2\theta_0-n_e^2\theta_0-n_0) \]  

Type II tripling: \[ \Delta k = \alpha/c(3n_e^3\theta_0-2n_e^6\theta_0-n_e^6) \]  

(2.19)  

(2.20)

Solutions for \( \Delta k = 0 \) in the above equations result in the phase matching angle \( \theta_m = 59^\circ 13' \) for type II doubling, and \( \theta_m = 59^\circ 00' \) for type II tripling respectively.

The nonlinear coupling coefficient K in Eq. (2.7)-(2.9) is related to the effective nonlinear coefficient \( d_{\text{eff}} \) by

\[ K = \frac{\omega_1}{2c\varepsilon_0}(n_{1o}n_{2e}n_{3e})^{-1/2}d_{\text{eff}} \]  

(2.21)
where $d_{\text{eff}}$ for type II interaction is given by $d_{\text{eff}} = d_{36}\sin 2\theta \cos 2\phi$. The crystal axes are $x$, $y$ and optical axis is $z$. The angle $\theta$ is measured relative to the $z$-axis (optical axis) and $\phi$ is measured in the $xy$-plane from the $x$-axis as shown in Fig. 2.7.

![Diagram](image)

Fig. 2.7. The locus of phase-matching direction in optically uniaxial crystal. The phase matching angle $\theta$ is measured from optical ($z$-) axis and $\phi$ is measured in the $xy$ plane from the $x$ axis.

2.6 SUMMARY

The physical origin of some nonlinear optical effects has been discussed in this chapter. A brief qualitative description of nonlinear optical interaction, especially the three wave interaction has been given. Various factors limiting the energy conversion efficiency in the nonlinear interaction between optical beams such as the effect of birefringent walk off, group velocity delay during interaction between short pulses in the birefringent crystals and the influence of the cubic nonlinearity are described. The phase matching requirement and material considerations for efficient frequency conversion is also discussed.
REFERENCES


Chapter III
Third-Harmonic Generation of Intense Laser Pulses

Third-harmonic conversion efficiencies as high as 50% have been demonstrated from type II KDP crystals using 1.053-μm picosecond laser pulses at intensities of 120 GW/cm². Self-phase and cross-phase modulation are important in the frequency conversion process. The nonlinear refractive index \(n_2\) used in the calculation is based on our experimental measurements described in Chap. IV.

3.1 INTRODUCTION

Harmonic generation has been studied extensively in the past twenty years. Some of the experiments were mentioned in the first two chapters. In particular for third harmonic generation, R. S. Craxton has thoroughly studied frequency tripling of laser radiation at wavelength of 1.054 μm for intensity up to 10 GW/cm²\(^2\).\(^1\) Several frequency tripling schemes were proposed and demonstrated.\(^2\)\(^3\)\(^4\)\(^5\)\(^6\) Among them, a scheme called polarization mismatching required two type II KDP crystals in series: a doubling crystal followed by a tripling crystal in which the second-harmonic pulse after the doubler is mixed with the unconverted fundamental pulse to produce a third-harmonic pulse.

In a dispersive nonlinear birefringent medium, the optical waves with different wavelengths or polarizations have different group velocities. They will walk away from each other after propagating certain distance in the crystal and limit the effective interaction length. The group velocities of fundamental e- and o-wave are defined as \(v_e\) and \(v_o\), respectively, with the corresponding pulse duration of \(\tau_e\) and \(\tau_o\) \((\tau_e = \tau_o\) when the e- and o-wave are at the same wavelength\). If \(\tau_{e(0)} >> L / |1/v_e - 1/v_o|\), where \(L\) is the crystal thickness, the pulse is long compared to the crystal length and the group-
velocity mismatch can be ignored. In this case, the polarization mismatching scheme is advantageous in that it can be easily configured to produce efficient second or third harmonic generation. This scheme is insensitive to small angular errors in the phase-matching angle and to intensity fluctuations as it operates at the phase-matching peaks of the two Type II KDP crystals. It is, however, sensitive to the polarization of the beam. Overall energy conversion efficiencies up to 80% to third harmonic were achieved in KDP with long pulses.\(^3\)

For short pulses where \(\tau_{\omega_0} = L |1/v_e-1/v_o|\), the group-velocity mismatch becomes important. The frequency conversion efficiency is usually low with such laser pulses even though the central-frequency wave vectors of the three mixing waves are well matched. This is because these pulses contain wave vectors with different group velocities in the dispersive crystals.\(^7,8,9\) The group velocity mismatch causes a substantial decrease in the efficiency of the frequency doubling and tripling processes for short pulses, for example, of 1 ps pulse duration. Several methods have been proposed to increase the frequency conversion efficiency of ultrashort pulses. One is to introduce angular spectral dispersion using gratings (or prisms) to achieve phase matching for all wave vectors.\(^10,11,12\) Another is to introduce a predelay between the two fundamental pulses at the entrance of the interacting crystal to compensate the group velocity mismatch.\(^13,14\) A third is to use very high intensities in thin crystals.

With the development of chirped-pulse amplification lasers,\(^15\) output laser intensities up to \(10^{18} \text{ W/cm}^2\) with pulse duration of 1 ps have been obtained in the Nd:Glass laser systems.\(^16,17,18\) Such high intensity ultrashort pulses allow short interaction lengths to be used efficiently with reduced group velocity mismatch.\(^19\) Also thin crystals have larger spectral bandwidth acceptance than thick crystals. Second harmonic generation efficiencies of ~75% for 4mm thick KDP crystal were reported with intensities up to several hundreds of GW/cm\(^2\).\(^19\)

In this chapter, we used the polarization mismatch scheme\(^1\) with two thin Type II KDP crystals for efficient frequency tripling of picosecond pulses. KDP crystals were used here because they can be grown in large sizes with qualities required for efficient frequency conversion including high optical quality, moderately high
birefringence, and adequate nonlinearity.\textsuperscript{3,20,21,22} They are also quite resistant to high intensity optical damage.\textsuperscript{23} The thickness of KDP crystals were chosen to be thin (2.5 mm each) in order to satisfy the long pulse condition $\tau_{e(o)} \gg L\left|1/v_e-1/v_o\right|$. The intensities of the laser pulses need to be up to 100 GW/cm$^2$ to optimize the conversion process in such thin crystals. To complete the study of frequency tripling for picosecond pulses, we did the investigation in the following order. First, we measured the third harmonic conversion efficiency using two thin KDP crystals and the energy conversion efficiency of 60-65\% was achieved for frequency doubling and 45-50\% were achieved for tripling for input pulse intensities of 50-100 GW/cm$^2$. We then investigated the higher order nonlinear effects such as self- and cross-phase modulations associated with the frequency conversion process. The experimental results of frequency conversion were also compared with theoretical simulations. The third order nonlinearities used in simulations were obtained by $Z$-scan measurements in Chapter IV.

3.2 Experimental Method

The polarization mismatch scheme for the third harmonic generation is shown in Fig. 3.1.(a) Two thin type-II KDP crystals, one for doubling and another for tripling\textsuperscript{3} were used. The input fundamental pulse was linearly polarized at 35° with respect to the "o" direction of the doubling crystal. This ensures that two o-photon are input for every e-photon. By properly choosing the thickness of doubling crystal, for every three photons, an e-photon will combine with an o-photon to give one e-photon at 2\omega, leaving one o-photon unconverted. The plane of the optical axis of the tripler is perpendicular to that in the doubler. The e-wave (o-wave) output from the doubler will be an o-wave (e-wave) of the input for the tripling. Thus, the unconverted fundamental and the converted second harmonic pulses mix in the second, tripling, crystal and generate an e-wave at 3\omega.

The crystals used here were chosen to be 2.5 mm thick to optimize frequency tripling efficiency over the 50-100 GW/cm$^2$ intensity range. Solutions of tripling equations scale as $KL\sqrt{I}$ (see Eqs. (2.20)) where K is a crystal constant, L is the
Fig. 3.1. (a) The experiment setup for harmonic generations. (b) the schematic layout of frequency tripling experiments.
crystal thickness, and I is the incident beam intensity. Therefore, the same overall tripling efficiency will result if similar crystal types are used and the product $KL\sqrt{I}$ is held constant. These crystals were mounted in a housing with 1/4"-thick glass windows for part of the experiments because they are easily damaged by moisture. The windows for the doubling crystal have an AR coating on input surface for 1.053 µm and its output face for both 1.053 µm and 0.527 µm. The windows for the tripling crystal have an AR coating on their input face for 0.527 µm and 1.053 µm and their output face for 0.351 µm.

From the refractive index data reported in Ref. [24], the group velocities of the fundamental o- and e-wave, and second harmonic e-wave are calculated as shown in Table 3.1. The crystal cut angle and refractive indices are also shown in the table. The o-wave will have 0.33 ps group velocity delay relative to the e-wave after propagating through the doubling KDP crystal, and the e-wave at 2ω will move 0.15 ps ahead of o-wave. The o-wave at ω and e-wave at 2ω become opposite polarizations in the tripling crystal and e-wave at ω moves faster than o-wave at 2ω. Thus, the fundamental e-wave in the tripling crystal will catch up the o-wave at 2ω. The calculated electric field envelope of laser pulses at different wavelengths at the exit of the doubling and tripling crystal are shown in Fig. 3.2. In the figure, $\omega_0$ is the electric field envelope of the input fundamental wave to the doubling crystal. We can see that using relatively long pulses and thin crystals can reduce the temporal walk-off induced in the crystals. The type-II -type-II setup used in our experimental setup can also utilize the second crystal to

<table>
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<th>$n_o$</th>
<th>$n_e$</th>
<th>$v_o \times 10^5$ cm/s</th>
<th>$v_e \times 10^5$ cm/s</th>
<th>$\theta_m$</th>
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<th>$v_e(\theta_m) \times 10^5$ cm/s</th>
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<td></td>
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<td>59°00</td>
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Fig. 3.2. Electric field outputs at exit of the doubling and tripling crystal. \( \omega_0 \) is the input electric fields to the doubling crystal.

(a) Type II KDP doubling crystal. Crystal thickness = 2.48 mm.

(b) Type II KDP tripling crystal. Crystal thickness = 2.37 mm.

The input electric fields are the outputs from doubling crystal.
compensate for the temporal walk-off induced in the doubling crystal.\textsuperscript{25} This is similar to the method to increase frequency doubling efficiency of short pulses by using predelay between the two fundamental pulses at the entrance of the interacting crystal to compensate for the group velocity mismatch.\textsuperscript{25}

The input laser pulses for frequency tripling experiment were generated from a chirped-pulse-amplification laser system.\textsuperscript{15,18} The schematic layout of the system is shown in Fig. 3.1.(b) The laser produces energies up to 2J with a wavelength of 1.053 \( \mu \text{m} \) and a temporal full width at half maximum (FWHM) of 1.6 ps. The energy of the fundamental pulses (\( \lambda = 1.053 \, \mu \text{m} \)) was monitored by a calibrated PIN diode. A down collimator (3.4:1) was used to reduce the beam size to fit the aperture of the KDP crystals and increase the beam intensity. A half-wave plate before the KDP crystal was used to control the polarization angle. The laser pulse energies were controlled by tuning a half-wave plate polarizer combination. The energies of the fundamental (\( \lambda = 1.053 \, \mu \text{m} \)), second (\( \lambda = 0.527 \, \mu \text{m} \)), and third harmonic (\( \lambda = 0.351 \, \mu \text{m} \)) pulses were monitored by an energy meter. Fig.3.3 shows the total output from the frequency doubling crystal for different input laser energies. By considering the Fresnel reflection losses at all surface (4\% at each surface), we can see that the energy is balanced. This indicated that there was no nonlinear absorption.

The temporal pulsewidth of the fundamental incident beam was monitored by an off-line single-shot autocorrelator, as shown in Fig. 3.4 (a). The polarizer is used to allow only the s-polarization light to be incident on the crystal (LiIO\(_3\)). The half wave plate is used to control the input laser intensity. The autocorrelation signal is measured by a linear detector array with a BG-18 filter used to block the IR background.

The pulse width of the second harmonic signal is measured by a separate autocorrelator from the autocorrelation signal at \( 4\omega \). The setup is shown in Fig. 3.4. (b). A 2 mm--thick BBO crystal (60\degree cut) is used. In this setup, two slits are used in front of the crystal to prevent all secondary reflections within the autocorrelator from reaching the detector. The autocorrelation signal is measured by a photomultiplier with a UV interference filter used to block the \( 2\omega \) background. Multiple shots with different delays in one arm are needed to obtain the trace.
Fig. 3.3. The total energy out of doubling crystal with different input fundamental pulse energy.
Fig. 3.4. Setup for autocorrelator (a) 2ω autocorrelator. (b) 4ω autocorrelator.
Fig. 3.5. Laser beam profile.

(a) Intensity contours relative to the maximum intensity (contour level 167). x, y axis are in the unit of centimeter.

(b) An equivalent one dimensional profile based on relative intensity groups.
A CCD camera was used to record the near field beam patterns in front of the doubling crystal. The images were analyzed and converted to intensity data. Fig. 3.5 (a) is an example which shows the intensity contours of the laser beam used. Contour levels are lower with respect to the peak (contour level 167). In order to simplify the theoretical calculation, a pixel distribution function which relates the number of pixels in each intensity groups was generated. Then an equivalent one dimensional profile based on relative intensity groups was generated for use by MIXER\(^1\) and is shown in Fig. 3.5 (b). MIXER is described below.

We used crystals to generate second and third harmonic at low intensity. A PIN diode with a interference filter for 0.527 or 0.351 μm was used to measured the harmonic signals with different tuning angles. The phase matching angle was optimized by tuning the crystals. Conversion scans for doubling and tripling as a function of tuning angle at low laser incident intensity for doubling and tripling crystals respectively were carried out before the efficiency scan. Angular scans were carried out again when laser intensity reached 50 GW/cm\(^2\). Because nonlinear phase shifts usually detune the interaction at high intensity as discussed in Chapter II, it is necessary to compensate for the nonlinear detuning by imposing a linear phase mismatch. For single intensity we can choose a Δθ to fully compensate the nonlinear phase error,\(^{26}\) but we cannot simultaneously compensate full range of intensities in the beam in space and time.

### 3.3 Theoretical simulations

The theoretical predictions for frequency tripling were provided by Dr. R. S. Craxton. The experiments were simulated with the program, MIXER,\(^1\) which was enhanced to model group-velocity dispersion and \(\chi^{(3)}\) effects. The theoretical calculation is based on the following differential equations:

\[
\frac{\partial E_1}{\partial z} + \frac{1}{V_{g1}} \frac{\partial E_1}{\partial t} = \frac{1}{2} \gamma_1 E_1 - iK_1 E_2 E_2^* \exp(i\Delta k z) + i(\alpha_{11}|E_1|^2 + \alpha_{12}|E_2|^2 + \alpha_{33}|E_2|^2)E_1
\]

(3.1)
\[
\frac{\partial E_2}{\partial z} + \frac{1}{V_{g2}} \frac{\partial E_2}{\partial t} = -\frac{1}{2} \gamma_2 E_2 - iK_2 E_1 E_1^* \exp(i\Delta k z) + i(\alpha_{21} |E_1|^2 + \alpha_{22} |E_2|^2 + \alpha_{23} |E_2|^2) E_2
\]

(3.2)

\[
\frac{\partial E_3}{\partial z} + \frac{1}{V_{g3}} \frac{\partial E_3}{\partial t} = -\frac{1}{2} \gamma_3 E_3 - iK_3 E_2 E_2^* \exp(i\Delta k z) + i(\alpha_{31} |E_1|^2 + \alpha_{32} |E_2|^2 + \alpha_{33} |E_2|^2) E_3
\]

(3.3)

where for wave i (waves 1 and 2 combining to generate wave 3), \(E_i\) is the electric field, \(V_{gi}\) is the group velocity, \(\gamma_i\) is the linear absorption coefficient, \(K_i\) is the nonlinear coefficient, and \(\alpha_{ij}\) gives the change of the refractive index that is due to the intensity of wave j [resulting from the third-order susceptibility \(\chi^{(3)}\)]. The quantity \(\Delta k\) is the wave-vector mismatch, \(z\) indicates the propagation distance, and \(t\) denotes time. The coefficients \(\alpha_{ij}\) are related to the coefficients \(\gamma_{ij}\) by \(\alpha_{ij} = \pi \gamma_{ij} \lambda_0 \sigma_0 \). \(\gamma_i\) are defined by the equation \(n_i = n_{i0} + \sum \gamma_{ij} I_j\). This gives the refractive index \(n_i\) of wave \(i\) in terms of its linear index \(n_{i0}\) and a nonlinear correction over the intensities \(I_j\) of wave \(j\). The nonlinear refractive index used here were measured by z-scan technique\(^{27}\) as described in Chap. IV. The measured nonlinear coefficients are listed in Table 3.2, \(i = j\) for self-phase modulation and \(i \neq j\) for cross-phase modulation. The propagation and \(K_i\) terms of Eqs. (3.1), (3.2) and (3.3) are solved in the frequency domain, and the \(\chi^{(3)}\) term in the time domain. (In the frequency domain, the group-velocity term is replaced by a simple translation of each Fourier mode with its phase velocity, thus including dispersion to all orders). Predictions for the experimental energy conversion efficiencies are

<table>
<thead>
<tr>
<th>Wavelength</th>
<th>(\gamma_i) (10(^{-20}) m(^2)/W)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.053 (\mu m) (o)</td>
<td>2.3±0.5</td>
</tr>
<tr>
<td></td>
<td>2.5±0.5</td>
</tr>
<tr>
<td>0.527 (\mu m) (o)</td>
<td>4±1</td>
</tr>
<tr>
<td></td>
<td>3.5±0.9</td>
</tr>
<tr>
<td>0.351 (\mu m) (e)</td>
<td>3±1</td>
</tr>
<tr>
<td>0.527 (\mu m) (e), 1.053 (\mu m) (e)</td>
<td>0.10±0.03</td>
</tr>
<tr>
<td>0.527 (\mu m) (o), 1.053 (\mu m) (e)</td>
<td>0.06±0.02</td>
</tr>
</tbody>
</table>
obtained by averaging solutions of Eq. (3.3) over the measured two-dimensional spatial profile of the IR laser beam (see Fig. 3.5). For THG, it takes two steps. After SHG (wave 3) is generated in the first crystal, the unconverted waves 1 and 2 and SHG are as the input for the second step.

In frequency conversion experiments the input polarization angle $\theta_p$ (the angle of the input electric-field vector to the o direction of the first crystal) was controlled by a half-wave plate (in Fig. 3.1 (b)). Following Ref. [1], consider an input ray of fundamental intensity $I_0 = I_e + I_o$, where $I_o (=\alpha I_0$) and $I_e (=1-\alpha)I_0$ are the o- and e-input intensities. The second and third harmonic output intensity $I_{2o}$ and $I_{3o}$ can be expressed as:

$$I_{2o} \propto I_0^2 \cdot \alpha(1-\alpha)(\sin^2 \varphi_o/\varphi_o^2)$$  \hspace{1cm} (3.4)

and

$$I_{3o} \propto I_0^3 \cdot \alpha^2(1-\alpha)(\sin^2 \varphi_o/\varphi_o^2) \cdot (\sin^2 \varphi_e/\varphi_e^2)$$  \hspace{1cm} (3.5)

where $L_d$ is the doubler thickness, and $\varphi_o = \Delta k_d L_d/2$ with $\Delta k_d$ is the value of $\Delta k$ in the doubler. $L_o$, $\varphi_e$ and $\Delta k_e$ are defined similarly for tripler. $\alpha$ related to the polarization angle $\theta_p$ as, $\alpha = \cos^2 \theta_p$. $I_{2o}$ is maximum when $\alpha = 1/2$, $\theta_p = 45^\circ$ (conventional type II doubling), and $I_{3o}$ is maximum when $\alpha = 2/3$, $\theta_p = 35.3^\circ$ (type II-type II tripling).

The frequency conversion efficiency was also optimized by tuning the crystal to obtain the phase matching condition. In the small signal and low power case, the relationship between second harmonic output intensity and phase mismatching is as Eq. (3.4), $\Delta k_d$ is the phase mismatch. For type II doubling, $\Delta k$ is defined as:

$$\Delta k_d = \frac{2\alpha}{c} \left(2n_e^{2\omega}(\theta) - n_e^o(\theta) - n_e^o \right)$$  \hspace{1cm} (3.6)

$\Delta k_d$ will increase if the propagation angle deviate from the phase matching angle. When $\Delta k$ increases, the conversion efficiency drops dramatically.

### 3.4 Experiment Results and Analysis

Conversion scans were carried out as a function of the polarization angle $\theta_p$ at low intensity for type II frequency doubling and type II frequency tripling respectively.
The result is close to the theoretical calculations (solid lines) using Eq. (3.4) and (3.5) as shown in Fig. 3.6.

The angular tuning curves of second harmonic generation for the two crystals are shown in Fig. 3.7. The dotted data with error bar are the experiment data and the solid lines are the theoretical fitting based on Eq. (3.6). The angles in the figure are relative to the air side of the interface, and are approximately 1.5 times larger than the corresponding angles in the crystal. The FWHM ($\Delta \theta_{FW}$) of the angle-tuning curves are $13.0 \pm 0.1$ mrad and $13.5 \pm 0.1$ mrad. These correspond to crystal lengths of 2.48 mm and 2.37 mm using Eq. (3.6).

The conversion efficiency of frequency doubling in a 2.5-mm thick type II crystal and frequency tripling in two 2.5-mm thick type II crystals, are shown in Fig. 3.8 and Fig. 3.9. The polarization of the incident beam was tuned optimum for SHG and THG respectively. Conversion efficiencies of 60-65% was achieved for frequency doubling and 45-50% (without windows) were achieved for frequency tripling with an input pulse intensity of 50-100 GW/cm². The simulations were carried out assuming perfect tuning and no phase modulation. The SHG conversion efficiencies are well fit by the theoretical calculation using MIXER. The THG conversion efficiencies are lower than the theoretical calculation in which no phase modulation was considered.

In order to understand the cause of the inconsistency between the theoretical calculation and the experiment results for third harmonic generation, a series experiments were carried out to investigate the amount of SH and TH generated by tuning the tripler, the polarization state of the beam after the doubler, and the spectrum and pulse shape of the fundamental and SH pulse after propagating through the doubler.

Angular tuning the frequency tripling crystal was carried out by measuring the SHG and THG signal at the same time. Fig. 3.10 shows the tuning curve at the intensity around the peak of the conversion curve ($\sim 40$ GW/cm²). Since the phase matching angle of frequency doubling crystal (59°13' cut) is very close to that of the frequency tripling crystal (59°00' cut) and the thin crystals have a wide tuning angle, the angular tuning range for the third harmonic generation is overlapping with that of
Fig. 3.6. Dependence of the SHG and THG efficiency on the polarization angle of the incident laser. The cross data represent the polarization tuning for SHG after the doubling crystal and the dot data represent the polarization tuning for THG. The solid lines are the theoretical calculations using Eqs. (3.4) and (3.5).
Fig. 3.7. The dependence of SHG efficiency on the crystal tuning angle at low intensity. (a) doubling crystal (b) tripling crystal.
Fig. 3.8. The SHG conversion efficiency as a function of input laser intensity for a 2.5-mm thick type II crystal. The cross dotted data was in the case that the crystal was mounted without window and diamond dotted data was in the case that the crystal was mounted in a housing with windows. The solid line is the theoretical simulations with $\Delta \theta = 0$. 
Fig. 3.9. THG conversion efficiency as a function of input laser intensity for a 2.5-mm thick type-II and a 2.5-mm thick KDP type II tripler. The cross dotted experimental data was obtained with no windows present. The star dotted data was the case with only one window in front of the doubler. The diamond dotted data was the case that both crystals have front and back windows.
Fig. 3.10. Measured dependence of the SHG and THG efficiency on the tripler tuning angle at input laser intensity of 40 GW/cm².

Fig. 3.11. Theoretical calculation of dependence of the SHG and THG efficiency on the tripler tuning angle at input laser intensity of 10 GW/cm². From Ref. 29.
the second harmonic generation. At peak intensity of 40 GW/cm², the peak conversion efficiency is 34% for tripling and 5% for doubling. Two peaks are separated by 10 mrad. Compared with the theoretical calculation using Eqs. (3.1)-(3.3) in Fig. 3.11, the peak positions are well matched. At the frequency tripling tuning peak, 33% energy was converted to TH and 6% was converted to SHG. This experiment confirmed that, during the THG, a small portion of the energy is consumed to generate the SH.

The spectra of the transmitted fundamental and SHG pulses after the frequency doubling are shown in Fig. 3.12. At low input intensity (5 GW/cm²), the spectra of the transmitted IR and SHG pulses were Gaussian and bandwidth limited. The spectra were broadened and had side structure at high input intensity (50 GW/cm²) due to the self- and cross-phase modulation. This experiment showed that, during the frequency up conversion of intense pulses which involves two or more optical waves, nonlinear phase changes caused by SPM and XPM occurred.

The polarization state of the fundamental pulses was measured before and after the doubler as shown in Fig. 3.13. Nonlinear phase changes in the doubler change the polarization state of the transmitted fundamental. The beam before the crystal is linearly polarized. At low intensity, the polarization state of the transmitted IR wave changed due to the linear birefringence of the crystal. At high intensity, the polarization state changed even more when incident beam was polarized at 35° to the optical axis. This was because the o-wave and e-wave had unequal intensities and experienced different amounts of nonlinear phase shift. The polarization state of the SH beam at different intensities is shown in Fig. 3.14. The polarization remains the same. The dashed curve was measured at low intensity and the solid curve was measured at high intensity. This is because the generated SH beam is purely p-polarized (e-wave). The nonlinear phase modulation it experienced will not change its polarization. The input laser intensity fluctuation were about 8% in all cases. These experiments indicated that there are nonlinear phase modulation effects under high intensity laser pulses.
Fig. 3.12. The spectra of transmitted fundamental and SHG pulses at different intensities.
Fig. 3.13. The polarization state of the transmitted fundamental at different intensities.

Fig. 3.14. The polarization state of the transmitted SHG at two different intensities.
Phase modulation has an impact on the conversion efficiency of THG. This was confirmed by introducing several 0.25” thick windows into our system. The windows caused extra phase modulation when the intense laser pulse propagates through them. The cross dotted experimental data in Fig. 3.8 were obtained with no windows present. The star dotted data was the case only one window in front of the doubling crystal. The diamond dotted data was the case both crystals have front and back windows. It can be seen that although phase modulation induced by the windows does not affect SHG conversion efficiency, the modulated output from the doubler will be the input for the tripler. It decreases the THG conversion efficiency because the phase modulation generated in the pulse affect the perfect phase matching at other times in the presence of group velocity walk-off.

We have investigated the pulse width of the second harmonic generation using the multi-shot autocorrelator as shown in Fig. 3.4. An experimental fourth-harmonic trace with FWHM of 2.0±0.2 ps is shown in Fig. 3.15 (a) for the an IR intensity of 5 GW/cm². The corresponding SH pulse width is 1.4±0.2 ps which is close to the theoretical prediction of a transform limited pulse. At 40 GW/cm², the FWHM of the autocorrelation trace broadened to 2.7 ps as shown in Fig. 3.15 (b). This is possibly caused by the combined effects of phase modulation and group-velocity dispersion within the crystal.

Including the effect of phase modulation, the calculated THG conversion efficiency curve is closer to the experimental data (curve 1,2 and 3 in Fig. 3.16). These calculations used our measurement value of SPM coefficients (see chapter IV) at 1 μm, 0.527 μm, 0.531 μm and the XPM coefficients between laser radiation at 1 μm and 0.527 μm. Curve 1 assumed that all other cross terms are zero are one order magnitude smaller than the SPM coefficients. Curve 2 assume all other cross terms as Chiens results¹⁹ (see Table. 4.3) and curve 3 (a) and (b) assume all other terms equal 4γ₁₁ (γ₁₁ is the SPM coefficients of IR) and 8γ₁₁ respectively. We can see that larger XPM phase modulations coefficients make the calculated results closer to the experimental data (curve 3 (b) in Fig. 3.16).
Fig. 3.15. Experimental $4\omega$ autocorrelation traces at different input laser intensity. (a) SHG pulse width was measured at IR input intensity of 5GW/cm$^2$. (b) SHG pulse width was measured at IR input intensity of 40GW/cm$^2$. 
Fig. 3.16 Theoretical calculation of TH conversion efficiency curves. The crosses are experimental data. Curves 1, 2 and 3 used the SPM and XPM coefficients measured in Chap. IV. Assume all other cross terms Curve 1 zero.

2 using results from Ref. 18.

3 (a) assuming $4\gamma_{11}$ ($\gamma_{11}$ is the SPM coefficient of IR), (b) assuming $8\gamma_{11}$. 
In our theoretical calculation, only the influence on the frequency tripling process arising from the nonlinear phase shift due to third order nonlinear susceptibility has been considered. However, the high-intensity laser beam may suffer from small-scale self-focusing and whole beam self-focusing. Self-focusing involves adding to the left-hands sides of Eqs. (3.1) - (3.3) the second derivatives of the appropriate amplitudes with respect to the transverse spatial coordinates. The influence of the whole beam self-focusing on the beam propagation depends on the relationship between the crystal thickness and the focal length of the resultant nonlinear lens. We measured the focal position using a lens (f = 2 m) at 1 GW/cm² and 50 GW/cm² respectively. No noticeable focal position change was observed. For a nonlinear crystal several millimeters long the contribution of whole-beam self-focusing can be neglected for the laser beam intensity around 200 GW/cm².

Small-scale self-focusing depends on nonlinear phase shift B (B-integral) is given by

\[ B = \frac{2\pi}{\lambda} \int_0^L \gamma f(z) dz = \Phi(t) \] (3.7)

In the frequency doubling crystals, the contribution of self focusing from different wavelength or polarization to the B-integral needs to be considered. At a laser beam intensity of 50 GW/cm², the B-integral induced by the doubling crystal is about 0.4 rad. The two \( \frac{1}{2} \) thick windows induce B-integral about 1.6 rad. Here the nonlinear coefficients \( \gamma \) we used for KDP were measured in Chap. IV, and the for glass windows we use \( \gamma = 2.9 \times 10^{-16} \) cm²/W for all wavelengths and polarizations. Due the poor spatial uniformity of the laser beam intensity (see Fig. 3.17), the B-integral would be larger. This could distort the beam wave front after propagation through the doubling crystal and affect the interaction in the tripling crystal.

Spatial walk-off may also affect the THG conversion efficiency. By using Eq. 3.8 in which the refractive indexes are taken from Table 3.1, the walk-off angle \( \rho \) between the second harmonic e-wave and the fundamental o-wave is calculated to be 24 mrad.
\[
\tan \rho = \frac{1}{2} \left( n_\omega^2 \right)^2 \left[ \frac{1}{(n_2\omega)^2} - \frac{1}{(n_3\omega)^2} \right] \sin 2\Theta_m
\] (3.8)

The second harmonic e-wave can shift 62 µm away from the fundamental o-wave in the plane of optical axis and the propagation direction. The near field beam profile measured at the x direction (Fig. 3.17) showed the existence of fine structures with an spatial scale of \(~50\) µm. It is speculated that the spatial walk-off shifted the spatial energy distribution of the SH beam in the SHG crystal and may unbalance the number of photons required to generate TH. Spatial walk-off in the THG crystal may shift the spatial energy distribution in y direction and may further reduce the THG efficiency.

### 3.5 Conclusion

In conclusion, we have performed systematic studies of THG using intense, 1.6-ps laser pulses. Two type II KDP crystals were used in the experiment, one for frequency doubling and another for frequency tripling. A conversion efficiency of 60-65% was achieved from frequency doubling and 45-50% was achieved for frequency tripling at input pulse intensities of 50-100 GW/cm². Our experimental study also showed the importance of the self- and cross-phase modulation effects in the frequency tripling process. By reducing the phase modulation induced by the windows, conversion efficiency achieved at high input pulse intensities was increased. The poor spatial uniformity of the incident beam is the possible source of the disagreement between the experimental results and theoretical predictions.
Fig. 3.17 Line-out of the beam profiles in (a) x-direction and (b) y-direction.
REFERENCES


Chapter IV
Measurements of Self- and Cross-Phase Modulation Coefficients

In this chapter, we show measurements of the self-phase modulation coefficients in KDP crystals at wavelengths of 1.053 μm, 0.527 μm and 0.351 μm using single beam Z-scan technique. The cross-phase modulation coefficients between 1.053 μm and 0.527 μm were also measured by a two color z-scan. These coefficients are important in frequency conversion of ultrafast pulses as discussed in Chapter II.

4.1 Introduction

When an intense laser pulse propagates through matter, it causes refractive index changes in the material due to the third order nonlinear susceptibility $\chi^{(3)}$. This, in turn, changes the phase, amplitude, and frequency of the incident laser pulses. Self-phase modulation (SPM) refers to the self-induced phase shift experienced by an optical field during its propagation in the materials. The main effect is to broaden the spectrum. Cross-phase modulation (XPM) refers to the nonlinear phase shift of the optical field induced by a second field at a different wavelength. XPM is always accompanied by SPM.

SPM was first observed by Shimizu as a modulated spectrum extending both above and below the laser frequency after self-focusing had occurred in liquid CS$_2$. SPM leads to a wide spectral broadening of picosecond and femtosecond pulses which can cover a frequency band up to 10000 cm$^{-1}$ wide, from UV to IR. This ultrafast super continuum pulse source has been used to probe nonlinear optical effects, for pulse compression, and for fiber diagnostics. The frequency-broadened pulses can play an important role in ranging, imaging, remote sensing, communications, and other fields. Spectral effects attributed to XPM were first observed in 1985, when it was reported that intense picosecond pulses could be used to enhance the spectral broadening of weaker pulses copropagating in bulk glasses. It has also been
observed in fiber Raman soliton lasers\textsuperscript{14,15} and has been seen to be important in optical parametric oscillation, optical parametric amplification\textsuperscript{16} and harmonic generation in bulk nonlinear crystals.\textsuperscript{17,18} XPM induced nonlinear birefringence has practical applications through Kerr shutters\textsuperscript{19} and intensity discriminators.\textsuperscript{20} SPM and XPM in optical fibers have been used in pulse compression to produce ultrashort laser pulses.\textsuperscript{21} They could also be a source of noise on signals in multiband communications and signal processing applications due to interference.

During frequency up-conversion of intense pulses, which involves two or more optical waves, nonlinear phase changes caused by SPM and XPM can destroy the phase coherence required for efficient conversion. It is important to measure the third order nonlinear susceptibility $\chi^{(3)}$ which is responsible for SPM and XPM. Z-scan,\textsuperscript{1,2} Four wave mixing,\textsuperscript{22,23} ellipse rotation\textsuperscript{24} and nonlinear interferometric\textsuperscript{25,26} techniques have been used to measure the nonlinear refractive index $n_2$ ($n_2 = 3/8n \chi^{(3)}$) associated with SPM. The nonlinear refractive index coefficients associated with SPM in KDP, widely used in frequency conversion,\textsuperscript{27,28} were measured by degenerate three-wave mixing\textsuperscript{29} and time resolved interferometry\textsuperscript{30} at $1 \mu$m. In this chapter we used single beam Z-scan technique\textsuperscript{1,31} to measure the nonlinear refractive index associated with SPM at wavelength of 1.053 $\mu$m, 0.527 $\mu$m and 0.351 $\mu$m and two-color Z-scan\textsuperscript{2} to measure the nonlinear coefficients of XPM between laser radiation at 1.053 $\mu$m and 0.527 $\mu$m in a KDP crystal. To our knowledge, the SPM measurements at 0.527 $\mu$m and 0.351 $\mu$m and the XPM measurements are the first reported measurements in KDP.

### 4.2 Z-SCAN TECHNIQUE

In the single Gaussian beam Z-scan,\textsuperscript{1,31} as depicted in Fig. 4.1, a sample is moved along the z direction of a focused Gaussian beam. The transmittance of the beam through an aperture in the far field is measured as a function of the sample position z with respect to the focal plane. While the input energy is kept constant, the sample experiences different incident fields (amplitude and phase) at different z
Fig. 4.1. The Gaussian beam Z-scan experiment setup in which the ratio $D_2/D_1$ is recorded as a function of the sample position $z$. 
positions. If the sample is thin (with a thickness much less than the beam Rayleigh range (depth of focus)), has $n_2 > 0$, and a purely refractive nonlinearity, it can be regarded as a thin lens of variable focal length. Starting the scan far from the focus ($-z$), the beam irradiance is low and negligible nonlinear refraction occurs; hence, the transmission remains relatively constant. As the sample moves close to the focus, the increased irradiance leads to a positive lensing effect that tends to augment diffraction, decreasing the aperture transmittance. When the sample is on the $+z$ side of the focus, the positive lensing tends to collimate the beam and the aperture transmittance is increased. The lack of significant transmission change at $z = 0$ is analogous to placing a thin lens at the focus that results in a minimum far field pattern change. For larger $+z$ position the irradiance, and hence, the lensing, are reduced and the transmittance returns to its original linear value. From the resultant Z-scan curve, the sign and magnitude of $n_2$ of the sample can be determined. If the sample also has an absorptive nonlinearity arising from either direct multiphoton absorption, saturation of the single photon absorption, or dynamic free-carrier absorption, it will strongly affect the measurement of the nonlinear refraction. The absorption suppresses the peak and enhances the valley, while saturation produces the opposite effect. The maximum absorption or saturation occurs when the sample is at the focal point of the lens, since here the on-axis intensity of the beam is highest. The sensitivity to nonlinear refraction is entirely due to the aperture. When the aperture is removed, Z-scan will be sensitive only to the nonlinear absorption. Hence, from the two Z-scans with and without an aperture, the nonlinear absorption and refraction can be separately determined.

In the two-color Z-scan measurement, two collinear beams with different wavelengths are used, a weak probe beam can be defocused by the action of the strong pump beam in a thin sample. The far field transmission variation of the probe beam through an aperture, as the sample is moved along the propagation direction ($Z$ axis) of the focused beams, is used to determine the optical nonlinearity from XPM.

A detailed theoretical analysis has been carried out for Gaussian beam z-scan in Ref. [31]. For laser beams with non-Gaussian beam profile, a Z-scan with top-hat beam has the advantage of simplifying the analysis. A top-hat beam can be readily
obtained by taking a small area of the expanded beam from lasers with arbitrary beam profile, for example by using an aperture.\(^{32}\)

A schematic of the setup is shown in Fig. 4.2. A small portion of infrared (IR) laser pulses (\(\lambda_1 = 1.053 \, \mu\text{m}\)), second harmonic (SH) pulses (\(\lambda_2 = 0.527 \, \mu\text{m}\)) or third harmonic (TH) pulses (\(\lambda_3 = 0.351 \, \mu\text{m}\)) are transmitted through an aperture A1 (with radius \(a\)) and can be treated as top-hat beams. Second harmonic pulses are generated using a KDP type-I crystal and third harmonic pulses are generated using two KDP type-II crystals (see Chap. III).\(^{33}\) We use the top-hat spatial profile because it increases the measurement sensitivity.\(^{32}\) This top-hat beam then copropagates through a lens with focal length of \(f_i\) for IR, \(f_2\) for SH and \(f_3\) for TH. \(f_1\), \(f_2\) and \(f_3\) are slightly different due to the dispersion of the lens. The electric field distribution near the focal point \(E_i(r,z,t)\) (\(i = 1, 2, 3\)) is described by Lommel functions.\(^{34}\) It has Airy radius at the focal spot of \(1.22\lambda_i F_i\), where \(F_i = f_i/2a\). The beam waist (\(w_{0i}\)) is defined as \(w_{0i} = \lambda_i F_i\). The Rayleigh range (\(z_0\)) is \(\pi w_{0i}^2/\lambda_i\). The electric field distribution of the top-hat beam near the focal spot is described by:

\[
E_i(u_i,v_i,t) = 2E_{0i}(t)e^{i\left(\frac{\lambda_i}{a}\right)^2} \int_0^1 J_0(v_i \rho) e^{-\frac{1}{2} \lambda_i \rho^2} \rho dp, \quad i = 1, 2, 3
\]  \(\text{(4.1)}\)

where \(u_i = 2\pi\lambda_i(a/f_i)^2 z\), and \(v_i = 2\pi\lambda_i(a/f_i) r\), where \(r\) is the coordinate perpendicular to the \(z\) axis. \(z\) is position of the sample, and \(E_{0i}(t)\) is the electric field of the top-hat beam at wavelength \(\lambda_i\) at aperture A1. It is normalized so that the light intensity \(I_i = |E_i(u_i,v_i,t)|^2\). For a Gaussian temporal pulse shape, \(E_{0i}(t) = \sqrt{\frac{\pi \eta_0}{4 w_{0i}^2}} e^{-\left(\frac{t}{\tau_i}\right)^2}\), and \(\tau_i\) is the optical power of the top hat-beam passing through the aperture.

A nonlinear crystal located in the focal region will introduce phase modulation proportional to the intensity. The nonlinear refraction coefficient \(n_{2i}\) (esu) or \(\gamma_i (m^2/W)\) is defined by

\[
n_i = n_{0i} + \frac{n_{2i}}{2} |E_i|^2 = n_{0i} + \gamma_i I_i
\]  \(\text{(4.2)}\)
Fig. 42. The experimental setup for one and two beam Z-scan. A1, A2: aperture; L1: lens with focal length of $f_1$ at $\lambda_1$, $f_2$ at $\lambda_2$ and $f_3$ at $\lambda_3$; D1, D2, D3: photon detectors; BS: beam splitter.
where \( n_{0i} \) is the linear index of refraction, \( E_i \) is the peak electric field (cgs), and \( I_i \) denotes the irradiance (MKS) of the laser beam within the sample. \( n_{2i} \) and \( \gamma_i \) are related through the conversion formula \( n_{2i} \text{(esu)} = (cn_{0i}/40\pi)\gamma_i \text{ (m}^2/\text{W}) \) where \( c \) (m/s) is the speed of light in vacuum. The nonlinear absorption coefficient \( \beta_i \) is defined as

\[
\alpha_i(I) = \alpha_{0i} + \beta_i I_i
\]

(4.3)

where \( \alpha_{0i} \) is the linear absorption coefficient, and \( \alpha_i(I) \) is the intensity dependent absorption.

The single beam Z-scan occurs when only one wavelength beam passes through A1. Changes in the beam diameter within the sample due to either diffraction or nonlinear refraction can be neglected if the sample thickness is much less than the Rayleigh range \( z_0 \). The amplitude and nonlinear phase change \( \Delta \phi_i \) of the electric field within the sample can be simplified to:

\[
\frac{d\Delta \phi_i}{dz'} = k_i \gamma_i I_i
\]

(4.4)

\[
\frac{dI_i}{dz'} = -\alpha_i(I_i) I_i
\]

(4.5)

From Eqs. (4.2) - (4.5), the complex field distribution at the exit surface of the sample, can be derived as

\[
E_{ei}(r,z,t) = E_i(r,z,t) e^{-\alpha u L/2 (1 + \psi_i |E_i(r,z,t)|^2)^{(k_{ri}/c_{ri})^{-1/2}}} \]

(4.6)

where \( \psi_i = \beta_i I_i(r,z,t)L_{eff}, \) is the imaginary phase shift due to nonlinear absorption, \( k_i = 2\pi/\lambda_i, \) and \( L_{eff} = [1-\exp(-\alpha_0 L)]/\alpha_0, \) is the effective sample thickness. When nonlinear absorption can be ignored, Eq. (4.6) reduces to

\[
E_{ei}(r,z,t) = E_i(r,z,t) \exp[ik_iL_{eff}(\gamma_i |E_i(r,z,t)|^2)]
\]

(4.7)

In the two-color Z-scan, we measured the cross phase modulation coefficients between optical wave at \( \lambda_1 \) and \( \lambda_2 \). The output unconverted IR (pump beam) and SH pulses (probe beam) from frequency doubler co-propagate through A1, the field distribution at the exit surface of the sample is as follows:

\[
E_{ei}(r,z,t) = E_i(r,z,t) \exp[i k_i L_{eff}(\gamma_1 |E_i(r,z,t)|^2 + 2\gamma_2 |E_2(r,z,t+\tau)|^2)]
\]

(4.8)
\[ E_{e_2}(r, z, t + \tau) = E_2(r, z, t + \tau) \exp[i k_2 L_{\text{eff}}(\gamma_2) E_2(r, z, t + \tau)]^2 + 2 \gamma_{12} |E_1(r, z, t)|^2 \] 

(4.9)

where \( \tau \) is the time delay between IR and SH pulses introduced in the KDP type I frequency doubling crystal. In the exponent of Eq. (4.8) and (4.9), the first term reflects the impact of the self-phase nonlinearity and the second term reflects the phase modulation induced by an optical wave of the other wavelength. If the optical wave at wavelength \( \lambda_2 \) is weak enough that \( |E_2|^2 \ll \pi \), the second term in the exponential of Eq. (4.8) and the first term in Eq. (4.9) can be ignored. Therefore, as the nonlinear crystal is moved along the \( z \) axis, the transmittance of the electric field at wavelength \( \lambda_1 \) through a finite aperture in the far field is determined by the self-phase modulation of the \( \lambda_1 \) optical wave, while the transmittance of the electric field at wavelength \( \lambda_2 \) is determined by the cross phase modulation due to the \( \lambda_1 \) optical wave.

When the Fresnel number \( w_0/\lambda_1 D \) is much smaller than unity, where \( D \) is the distance from the sample to the aperture \( A2 \), the field distribution \( E_{A2}(\rho, z, t) \) at the sampling aperture \( A2 \) (Fig. 4.1) is proportional to the Fourier transform of field at the exit surface of the sample. The electric field at aperture \( A2 \) can be expressed by

\[ E_{A2}(\rho, z, t) = \frac{e^{i k_2 z}}{i \lambda_2 z_{12}} e^{i \rho^2} \text{FFT}(E_{e_1}(u_1, v_1, t)) \] 

(4.10)

where \( z_{12} \) is the distance between the sample and the aperture, and \( \rho \) is the radial coordinate at the aperture.

Having the electric field distribution at the aperture, we can obtain the normalized Z-scan power transmittance:

\[ T(z) = \frac{\int_{-r_a}^{r_a} \int_{-r_a}^{r_a} \left| E_{A2}(\rho, z, t) \right|^2 \rho d\rho dt}{\int_{-r_a}^{r_a} \int_{-r_a}^{r_a} \left| E_{iA2}(\rho, z, t) \right|^2 \rho d\rho dt} \] 

(4.11)

where \( r_a \) is the radius of aperture \( A2 \) and \( E_{iA2} \) is the electric field at \( A2 \) without nonlinear crystal. Eq. (4.11) gives the Z-scan fluence transmittance \( T(z) \) as a function of crystal position.
Figure 4.3 shows numerical examples of the normalized transmittance as a function of sample position in the presence of SPM or XPM. For all of the curves the on-axis nonlinear phase (either self- or cross-phase) the maximum phase shift is chosen to be $\Phi_0 = 0.4$, where $\Phi_0 = k_1 L \gamma I_0$ and $I_0$ is the on-axis peak intensity. The different curves compare the effects of self- and cross-phase modulation on the transmittance of the two beams through the aperture. Even though the phase shifts are the same, the different focusing of the two beams means that transmittance as a function of crystal position will differ for the different physical processes. This can be beneficial in distinguishing the different effects, in particular, eliminating the contamination of SPM in the XPM measurement.

Curves (a), (b) and (c) in Fig. 4.3 show the effects of self-phase modulation on the transmittance for the beam at wavelength of 1.053 $\mu$m ($\lambda_1$), 0.527 $\mu$m ($\lambda_2$) and 0.351 $\mu$m ($\lambda_3$) respectively. Since the f-number of the system at $\lambda_1$ and $\lambda_2$ are about the same, the Rayleigh range $z_{01}$ is twice $z_{02}$. The distance between peak and valley corresponding to the $\lambda_2$ optical wave is half that of the $\lambda_1$ optical wave. Curve (d) shows the effects of XPM of $\lambda_1$ on the transmittance of weak $\lambda_2$ beam. The asymmetry in the relative decrease or increase in transmittance is mainly due to the dispersion of the focusing lens. The focal length is slightly longer for the $\lambda_1$ optical wave. The irradiance of electric field at 1.053 $\mu$m induces a positive lens for the $\lambda_2$ wave in the thin sample near its focus since $n_{21} > 0$. With the sample on the -z side of the $\lambda_1$ focus (Fig. 4.2), the positive lensing effect tends to augment diffraction. Therefore, the aperture transmittance is reduced. When the sample moves on the +z side of the $\lambda_1$ focus, the positive lensing effects tend to collimate the beam and increase the transmittance through the aperture. The transmittance reaches a maximum when the sample is located approximately at the focal point of $\lambda_1$ (i.e., $F_{pl}$ in Fig. 4.2) because the maximum intensity dependent phase distortion takes place at $F_{pl}$. Curve (e) shows the effects of XPM of $\lambda_2$ on the transmittance of a weak $\lambda_1$ beam which is opposite to curve (d). The asymmetry in the relative decrease or increase in transmittance is similar to curve (d) but the sensitivity is much smaller in this case. Because the pump beam
Fig. 4.3. The effect of SPM and XPM on the transmittance of the beams at different wavelengths through the aperture. In all cases, the nonlinear phase shift is $\Phi_0 = 0.4$. Curves (a), (b) and (c) are the transmittance of the single color Z-scan at wavelength of $\lambda_1, \lambda_2$ and $\lambda_3$. Curve (d) is the transmittance of the two color ($\lambda_1, \lambda_2$) Z-scan at $\lambda_2$. Curve (e) is the transmittance of the two color ($\lambda_1, \lambda_2$) Z-scan at wavelength of $\lambda_1$. $z/z_{02}$ is the position in terms of the Rayleigh range of the second harmonic beam.
spot size is half that of the probe beam, only the center portion of the probe beam will experience nonlinear phase distortion.

4.3 Experimental results

In the experiment, 2.0-ps, 1-μm laser pulses are generated from a chirped pulse amplification laser system. These 3 cm diameter (1/e²) pulses are incident on a 1-cm thick type I KDP frequency doubling crystal to measure n₂ at 0.527 μm, and XPM coefficient between 1.053 μm and 0.527 μm. These pulses are incident on two 1.6-cm thick type II KDP crystals to generate TH when n₂ at 0.351 μm is measured. A half wave plate is placed before the doubler to tune the polarization of the IR in order to control the amount of SH or TH wave generated. A BG18 filter after doubler and UG11 filter after tripler were used respectively to block light at other wavelengths. A 6.8-mm diameter aperture (A1 in Fig. 4.2) is placed after the crystal to select a small portion of the IR, SH or TH waves. The spatial profile of the pulse passing through the aperture can be regarded as a top hat pulse. The focal lengths of the lens after the A1 aperture were determined by a far field spot size scan using a CCD camera. The measured focal lengths (f₁, f₂ and f₃), the resulting beam waists (w₀₁, w₀₂ and w₀₃) and the Rayleigh ranges (z₀₁, z₀₂ and z₀₃) are listed in Table 4.1. For crystals with a small n₂, a longer crystal is preferred as long as the thickness is less than one third of the corresponding Rayleigh range. In all cases, the intensity is kept well below the damage threshold. Samples with different thickness and cuts were used for different

Table 4.1. The measured focal lengths, the resulting beam waists and Rayleigh ranges.

<table>
<thead>
<tr>
<th>Wavelength λᵢ (μm)</th>
<th>Focal length fᵢ (cm)</th>
<th>Beam waist w₀ᵢ (μm)</th>
<th>Rayleigh range z₀ᵢ (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.053</td>
<td>76.5±0.5</td>
<td>118±2</td>
<td>4.2±0.1</td>
</tr>
<tr>
<td>0.527</td>
<td>74.3±0.5</td>
<td>57.6±0.8</td>
<td>2.0±0.1</td>
</tr>
<tr>
<td>0.351</td>
<td>65.4±0.5</td>
<td>33.8±0.5</td>
<td>1.02±0.03</td>
</tr>
</tbody>
</table>
wavelengths. The crystals were mounted on a translation stage. To simplify the experimental analysis, a 7.5 mm thick KDP sample cut at 90° to the wave propagation direction was used for measuring XPM coefficients to avoid generating additional for second and third harmonic during the interaction. For other axis orientations, the k vector spread due to focusing will allow part of the beam to satisfy the second or third harmonic phase matching condition. The transmittance through the aperture would then be due to the combined effects from refractive index changes, second and third harmonic generation. The beam splitter after the aperture (A1 in Fig. 4.2) sends a small portion of the beam to a PIN diode (D1 in Fig. 1) which used to monitor the top hat IR energy. Part of the probe beam is reflected by the beam splitter before the analyzing aperture (A2 in Fig. 4.2) to PIN diode (D2 in Fig. 4.1) and gave the open-aperture Z-scan curve. From the open aperture transmission the nonlinear absorption is measured. The change of transmission due purely to the nonlinear index of refraction is determined by dividing the closed aperture transmittance by the open aperture transmittance. This also has the advantage of compensating for the energy fluctuations during the experiment which could lead to systematic transmittance changes with z. The linear (low intensity) transmittance of the aperture A2, defined as the ratio of power transmitted through A2 to the total power incident on the plane of the aperture, is 0.03. The incident IR temporal full width at half-maximum (τ_{FWM}) was 2.0±0.2 ps as measured by a single-shot autocorrelator. The SH and TH pulse widths were calculated to be 1.41 ps and 1.45 ps respectively in the small signal gain region. The energy of the incident IR pulse ε was measured with an energy meter. For a Gaussian temporal profile, the on-axis peak intensity I₀ within the sample is

\[ I_0 = \sqrt{\pi \ln 2} \varepsilon / 2w_0^2 \tau_{FWM}. \]

Fig. 4.4 shows typical Z-scan results for measuring SPM and XPM coefficients of KDP crystals. The parameters for each case is listed in Table. 4.2. The peak-to-valley configuration of all these Z-scan indicates a positive nonlinearity. The solid line in each of the figures is the least squares fit to the experiment data using Eq. (4.11) to determine the Φ₀. We use a temporal separation induced in the frequency doubling
Fig. 4.4. Experimental one and two-beam z-scan as a function of $z/z_n$, $i = 1, 2, 3$. In all cases, the solid line is fit to determine the peak phase shift $\Phi_0$. (a) Single beam, $\lambda_1 = 1.053 \mu m$, $\Phi_0 = 2.3$. (b) Single beam, $\lambda_2 = 0.527 \mu m$, $\Phi_0 = 1.8$. (c) Single beam, $\lambda_1 = 0.351 \mu m$, $\Phi_0 = 2.1$. (d) Two color ($\lambda_1, \lambda_2$) beams, $\Phi_i = 0.51$. 
Table 4.2. The parameters of Z-scans for measuring SPM and XPM.

<table>
<thead>
<tr>
<th></th>
<th>λ (µm)</th>
<th>Pulse width (ps)</th>
<th>Energy (µJ)</th>
<th>Φ₀</th>
</tr>
</thead>
<tbody>
<tr>
<td>(a)</td>
<td>1.053 (e) (SPM)</td>
<td>2.0</td>
<td>91±5</td>
<td>2.3±0.1</td>
</tr>
<tr>
<td>(b)</td>
<td>0.527 (o) (SPM)</td>
<td>1.41</td>
<td>3.6±0.4</td>
<td>1.8±0.1</td>
</tr>
<tr>
<td>(c)</td>
<td>0.351 (o) (SPM)</td>
<td>1.45</td>
<td>2.9±0.4</td>
<td>2.1±0.1</td>
</tr>
<tr>
<td>(d)</td>
<td>1.053 (e), 0.527 (e) (XPM)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>(pump) (probe)</td>
<td>2.0 (IR)</td>
<td>111±7 (IR)</td>
<td>0.51±0.04</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1.41 (SH)</td>
<td>&lt; 0.2 (SH)</td>
<td></td>
</tr>
</tbody>
</table>

crystal (KDP I), τ = 0.73-ps, in Eq. (4.9) in the case of XPM based on the predicted temporal walk-off induced in KDP type I doubler between pulses at different wavelengths. The extraordinary IR wave moves 0.73-ps ahead of extraordinary SH wave at the exit of the 1-cm KDP type I doubler.

The nonlinear coefficient γ₂ can then be calculated from Φ₀ = kLₐeffγ₁₂I₀. There are several error sources in the measurement. The error in the curve fit, the error in measuring the crystal thickness, the error in measuring the pulse width and energy which determine beam intensity. The least squares fit for the experiment data yields an error of 5%. The error for the crystal thickness is 1%. The largest error comes from the measurement of the pulse width and beam energy. The resultant error of the on-axis intensity is about 30%. The nonlinear coefficients of SPM and XPM with different polarization were measured, with the results presented in Table 4.3. Both n₂ and γ are presented. They are related by n₂(ESU) = (cm²/40πγγ) (m²/W) where c (m/s) is the speed of light in vacuum. Our results for the nonlinear coefficient at wavelength 1.053 µm are in good agreement with the work reported in Ref. [29,30] as shown in the fourth and fifth columns of Table 4.3. The last column lists the theoretical estimation used in Ref. [28]. To our knowledge, the nonlinear SPM coefficients at wavelength of 0.527 µm, 0.351 µm and XPM coefficients are the first data set reported.

In the two color Z-scan, there is a further temporal walk-off between two colors with different wavelengths in the KDP sample to be measured during the measurement of XPM coefficient. To measure n₂(o-o), in which both probe and pump
beam are ordinary waves, the optical axis of the sample KDP (90° cut) is perpendicular to the polarization of IR and SH pulses. The pump and probe beam both are o-waves in the sample and the pump pulse (IR) moves 0.51 ps further ahead of the probe pulse (SH) after the sample. For measuring $n_2(e-e)$, the pump (IR) and probe beam (SH) both are e-waves in the sample and the pump pulse moves 0.59 ps further ahead of the probe pulse. We include the walk-off effects in the theoretical fit by dividing the sample into segments and integrating the nonlinear phase experienced in each of the pieces. In each of the segments, the probe beam will experience different nonlinear phase shift which is due to the different time delay between the pump and probe. Thus the XPM can be well determined. We could improve the sensitivity of our measurement by putting an predelay crystal before or after the frequency doubling crystal to compensate for the walk-off introduced in the sample to be measured.

4.4 Conclusion

In conclusion, a top hat Z-scan method to was used to measure the phase shift caused by the self- and cross phase nonlinearity in KDP crystals. The third order nonlinear coefficient of KDP at different polarizations at wavelengths of 1.053 μm, 0.527 μm and 0.351 μm were obtained. By considering the temporal walk-off between the pump and probe beam in the crystals, we are also able to estimate nonlinear index of refraction due to the cross phase modulation. We are applying these results to calculations of third harmonic generation with ultrafast pulses as described in Chap. III.
Table 4.3 Nonlinear refractive index coefficients of KDP measured by Z-scan method.

<table>
<thead>
<tr>
<th></th>
<th>$n_2$ ($10^{-13}$ esu)</th>
<th>$\gamma$ ($10^{16}$ cm$^2$/W)</th>
<th>$n_2$ ($10^{-13}$ esu)</th>
<th>$\gamma$ ($10^{16}$ cm$^2$/W)</th>
<th>$\gamma$ ($10^{16}$ cm$^2$/W)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>theoretical$^{28}$</td>
</tr>
<tr>
<td>1.053 $\mu$m (o) (e)</td>
<td>0.8±0.2  0.8±0.2</td>
<td>2.3±0.5  2.5±0.5</td>
<td>0.72$^{29}$  0.76$^{29}$  1.0±0.3$^{30}$</td>
<td>2.9±0.9$^{10}$</td>
<td>2.7  2.7</td>
</tr>
<tr>
<td>0.527 $\mu$m (o) (e)</td>
<td>1.4±0.4  1.3±0.3</td>
<td>4±1  3.5±0.9</td>
<td></td>
<td></td>
<td>4.1  4.1</td>
</tr>
<tr>
<td>0.351 $\mu$m (o) (e)</td>
<td>2.4±0.7  1.2±0.4</td>
<td>7±2  3±1</td>
<td></td>
<td></td>
<td>6.1  6.1</td>
</tr>
<tr>
<td>0.527(e); 1.053 (e) (weak; strong)</td>
<td>0.03±0.01</td>
<td>0.10±0.03</td>
<td></td>
<td></td>
<td>6.6</td>
</tr>
<tr>
<td>0.527(o); 1.053 (o) (weak; strong)</td>
<td>0.023±0.007</td>
<td>0.06±0.02</td>
<td></td>
<td></td>
<td>2.2</td>
</tr>
</tbody>
</table>
REFERENCES

19 M. A. Duguay, in Progress in Optics, 14, ed. by E. Wolf (North-Holland, Amsterdam, 1976) p. 163.
37 Optical Glass Filters, Scott Glass Technologies Inc. 400 York Avenue, Duryea, PA. 18642.

Chapter V

Measurement of the Group Velocity Walk-off and Optical Axis Orientation in a Uniaxial Crystal

Frequency domain interferometry has been used to measure the group velocity walk-off (GVW) between the ordinary and extraordinary waves in a uniaxial crystal and to locate the crystal optical axis. This technique is based on the spectral interference of two short pulses in the frequency domain. The ordinary (o) and extraordinary (e) waves, which have different group delays in a birefringent crystal, are used as the two arms of the interferometer. The dependence of the interference fringe spacing and the walk-off on incident angle with respect to the birefringent crystal were measured in KDP and the direction of the optical axis was determined.

5.1 Introduction

Uniaxial nonlinear crystals have been widely used in frequency conversion of high power laser beams. The application of the frequency conversion includes second- and higher-harmonic generation,\textsuperscript{1,2,3} optical parametric oscillators and amplifiers (OPO, OPA)\textsuperscript{4,5} and autocorrelators. It is important to know the direction of the crystal's axis relative to the propagation direction of the laser to optimize the phase matching for these processes. The orientation of the optical axis in the crystal determines the spatial and temporal walk-off between the ordinary (o) and extraordinary (e) waves. For intracavity frequency conversion in OPOs and OPAs where the beam size is small, spatial walk-off limits the conversion efficiency. In addition to the spatial walk-off, the temporal walk-off between the o- and e-waves due
to group velocity dispersion for the two polarizations is a major limitation for ultra-
short pulse frequency conversion.\textsuperscript{2,6}

The plane of the optical axis or the plane perpendicular to it can be easily
found using two crossed polarizers. It is difficult, however, to measure the angle of the
axis relative to the laser propagation direction or its orientation with respect to the
tuning axis. The optical axis of the crystal is usually determined by x-ray diffraction.\textsuperscript{7}
This requires an x-ray source and sophisticated analysis which may not be readily
available. In this chapter, we introduce a simple direct method to measure the GVW
between the \( o \)- and \( e \)-waves in a birefringent crystal. The GVW is directly related to
the axis orientation. In the measurement, the same ultrashort pulse to be frequency
converted, or more exactly its spectrum, can be used for these measurements. If the
refractive index and its dispersion is known, the angle and orientation of the optical
axis in the uniaxial crystal can be determined by further measuring the dependence of
the GVW on the incident angle with respect to the birefringent crystal. The refractive
indices have been measured by minimum-deviation methods and are accurate to the
fifth decimal place.\textsuperscript{8} Extensive measurements of refractive indices of nonlinear crystals
isomorphic to \( \text{KH}_2\text{PO}_4 \) were made by Kirby and DeShazer.\textsuperscript{9,10} The dispersion
properties are usually obtained by a fitting to the Sellmerier or Zernike formulae.\textsuperscript{10,11}

Our technique is based on frequency-domain interferometry,\textsuperscript{12,13,14} utilizing
interference fringes in the frequency domain. The basic idea is that the \( e \)- and \( o \)- wave
components of broad bandwidth, short pulses passing through a birefringent medium
undergo group velocity walk-off. These two temporally separated pulses are combined
in a spectrometer to create an interference pattern. The spacing of the interference
fringes depends on the temporal separation. The group velocity dispersion which
depends on the propagation angle with respect to the optical axis determines the
temporal separation. One advantage of this technique is that it can be used "in situ",
for example, with an ultrafast laser system.
5.2 Method of Frequency-Domain Interferometer

The configuration of the frequency-domain interferometer used is shown schematically in Fig. 5.1. We define Cartesian coordinate system in which \( z \) is the laser propagation direction, the \( x \) and \( z \) axes form the vertical plane and the \( y \) and \( z \) axes form the horizontal plane. The birefringent axes are along the \( x \) and \( y \) directions. A \( \lambda/2 \) wave plate was used to control the polarization direction of the linearly polarized incident pulse, aligning at \( 45^\circ \) with respect to the \( x \) or \( y \) axis. An analyzer placed after the nonlinear crystal was also aligned to \( 45^\circ \) to combine the components from the \( x \) and \( y \) directions. The output light was then sent to a spectrometer with an optical multichannel analyzer.

At the incident surface of the crystal \( (z = 0) \), the electric fields of the components polarized along \( x \) and \( y \) direction can be expressed as

\[
E_x(t, z = 0) = \frac{1}{\sqrt{2}} E(t) \exp(i\omega_0 t)
\]

(5.1)

\[
E_y(t, z = 0) = \frac{1}{\sqrt{2}} E(t) \exp(i\omega_0 t)
\]

(5.2)

where \( E(t) \) is the slowly varying envelope of the laser pulse before the polarizer and \( \omega_0 \) is the carrier frequency. Since the pulses interfere in the frequency domain, the Fourier transformation is used to describe the electric field at the exit face of the crystal:

\[
E_x(\omega, z = L) = \frac{1}{\sqrt{2}} E(\omega) \exp[-ik_x(\omega)L]
\]

(5.3)

\[
E_y(\omega, z = L) = \frac{1}{\sqrt{2}} E(\omega) \exp[-ik_y(\omega)L]
\]

(5.4)

where \( E(\omega) \) is the Fourier transformation of \( E(t) \) and \( k_x(\omega) \) and \( k_y(\omega) \) are the propagation constants of the \( x \) and \( y \) components.

A polarizer placed after the birefringent crystal was aligned to \( 45^\circ \) to combine the two components.

\[
E_{out}(\omega, z = L) = \frac{1}{2} \left[ E_x(\omega, z = L) + E_y(\omega, z = L) \right]
\]

(5.5)
Fig. 5.1. Experimental setup: P, polarizer; M's, mirrors. The linearly polarized light is coupled into the nonlinear crystal with the polarization direction aligned at 45° with respect to birefringent axis.
The output light was sent to a spectrometer with an optical multichannel analyzer. The power spectrum detected in the spectrometer takes the following form:

$$I(\omega) = \frac{1}{4}[E(\omega)]^2 [1 + V \cos(\phi_{oe}(\omega))]$$  \hspace{1cm} (5.6)

where $E(\omega)$ is the spectrum of the incident pulse, $V$ is the visibility of the interference patterns, $\phi_{oe}(\omega) = ((k_x(\omega) - k_y(\omega))L = \frac{2\pi}{\lambda} [n_o(\omega)\cos\Psi_o - n_e(\omega,\theta)\cos\Psi_e]L$ is the phase retardation between o-wave and e-wave and $\Psi_o, \Psi_e$ are the refraction angles of o-wave and e-wave in the birefringent crystal respectively.\(^{15}\) The phase retardation, $\phi_{oe}(\omega)$, can be expanded as follows:

$$\phi_{oe}(\omega) = \phi_{oe}(\omega_o) + \frac{d\phi_{oe}(\omega_o)}{d\omega} \Delta \omega + \frac{1}{2} \frac{d^2\phi_{oe}(\omega_o)}{d\omega^2} \Delta \omega^2 + \cdots$$  \hspace{1cm} (5.7)

where $\Delta \omega = \omega - \omega_o$, $\phi_{oe}(\omega_o)$ is a constant. The second term in Eq. (5.7) is the group velocity walk off. The third term is the difference of the group velocity dispersion, which describes the difference in pulse spreading for o-wave and e-wave. For picosecond pulses, it can be ignored. Therefore from Eqs. (5.6) and (5.7), the periodicity of the interference pattern in the frequency domain is given by:

$$\Delta \omega = \frac{2\pi}{\frac{d\phi_{oe}(\omega_o)}{d\omega}}$$  \hspace{1cm} (5.8)

and the group walk-off of the crystal is

$$\frac{d\phi_{oe}(\omega_o)}{d\omega} = \frac{2\pi}{\Delta \omega}$$  \hspace{1cm} (5.9)

Since the refractive index of the extraordinary wave $n_e(\omega,\theta)$ depends on the propagation angle relative to the optical axis ($\theta$), $n_e(\omega,\theta) = [\sin^2 \theta / n^2_x(\omega) + \cos^2 \theta / n^2_z(\omega)]^{1/2}$, the group velocity walk off $d\phi_{oe}(\omega_o)/d\omega$ is also a function of propagation direction.\(^{15}\) It approaches 0 when $\theta = 0^\circ$, and approaches a maximum when $\theta = 90^\circ$. Therefore the interference fringe spacing will be minimized when the optical axis is perpendicular to the propagation direction.

From Eq. (5.9), the interference spacing $\Delta \omega$ gives the group walk off
\( \frac{d\phi_{\infty}(\omega)}{d\omega} \). If the crystal dispersion data and cut angle are known, the crystal length can also be determined.

Temporaly separated pulses can interfere owing to the linear dispersion of the grating in a spectrometer. Different frequency components propagate along different paths, which results in a frequency-dependent time delay. Therefore, two temporally separated pulses can physically overlap on the detector surface of the spectrometer. A simplified version of a spectrometer is shown in Fig. 5.2. The incident beam is collimated and has a diameter of \( D \). The incident angle to the grating is \( i \), while the diffracted angle is \( \alpha \). Assuming two pulses with pulse width \( \tau_p \) that are separated by \( T \), the amplitude fronts of these two pulses are no longer parallel to the phase fronts after the grating. At the focus of the image lens, each pulse is temporally stretched to a duration of \( DN/\cos(i)c \), where \( N \) is the number of grooves per millimeter of the grating, \( \lambda \) is the wavelength of the pulses, and \( c \) is the speed of the light. The two separate pulses can physically overlap for a time \( t_0 \) at the focal (frequency) plane, provided that the original separation \( T \) is less than the grating-induced stretching \( DN/\cos(i)c \) shown in Fig 5.2.

The requirement for the two pulses overlapping in the spectrometer places a limit for the crystal length \( L \).

\[
L < \frac{DN\lambda}{[c \cos(i)] \frac{d(k_x(\omega) - k_y(\omega))}{d\omega}].
\]  

(5.10)

Since the incident laser pulses have finite spectrum width \( \Delta \Omega \), the interference fringe spacing \( \Delta \omega \) needs to be smaller than \( \Delta \Omega/2 \) for the fringes to be distinct. This gives the lower limit for the crystal length.

\[
L > \frac{4\pi}{[\Omega \frac{d(k_x(\omega) - k_y(\omega))}{d\omega}]}.
\]  

(5.11)

If the crystal length falls outside of this range, the delay between the two polarizations can be adjusted using a delay line or a crystal which adds a known delay \( \tau \). Then, the group walk-off to be measured can be expressed as

\[
\frac{\partial \phi_{\infty}(\omega_0)}{\partial \omega} = \frac{2\pi}{\Delta \omega} - \tau
\]  

(5.12)
Fig. 5.2  Temporal stretching of Short pulses in a spectrometer.
When the crystals to be measured is thin, a delay line with positive $\tau$ needs to be used, the fast axis of the delay line needs to be aligned parallel to that of the crystal so that the pulses move out of the delay line with delay $\tau$ will move further apart after propagating through the crystal. If a crystal with known group walk-off is used as a predelay, its fast axis is aligned parallel to that of the crystal to be measured. When crystals to be measured are long, $\tau$ needs to be negative. It will be the opposite of the previous situation.

The disadvantage of this single pass scheme is that the spatial walk off decreases the visibility. The visibility is not 100% because the pulses are not completely overlapped in time and space. It is the complex function of size of the slit, the magnification of the spectrometer and the group delay of the pulses. A double pass configuration used for thin crystals automatically compensates for the spatial walk off as shown in Fig. 5.3. Results are presented for both schemes.

### 5.3 Experimental Results

The laser beam originated from an actively mode-locked Nd:YLF oscillator that produces a 50-ps pulse train at a wavelength of 1054 nm with a repetition rate of 100 MHz. The pulse train passed through an 800-m single mode optical fiber that increases the bandwidth from 0.03 to 4 nm through the combined effects of self-phase modulation and group-velocity dispersion. The shape of the spectrum was controlled by a $\lambda/4$ plate-polarizer combination before the fiber. The pulses were then temporally compressed to 1 ps in a double-pass grating pair. A polarizer was placed in front of the nonlinear crystal. A polarizer was used instead of half wave plate to avoid introducing extra delay between the e- and o-waves. A second polarizer placed at the exit end of the crystal was used to mix the electric-field components of the fast and slow modes. In both cases, the polarizer was placed at 45° to the e- and o- axis of the crystal. Finally, the collimated output beam was sent to a 1/4 m spectrometer with an optical multichannel analyzer with a 1024 element linear array.
Fig. 5.3. Experimental setup for double pass crystal axis measurements: P, polarizer; M's, mirrors. The linearly polarized light is coupled into the nonlinear crystal with the polarization direction aligned at 45° with respect to birefringent axis.
In the experiment, the plane of optical axis of the crystal is located using a pair of crossed polarizers. The linear polarized light transmitted through the first polarizer is completely absorbed by the second polarizer (analyzer). When the nonlinear crystal is placed between the crossed polarizers, no light emerges from the analyzer when the optical axis is parallel to either the polarizer or analyzer. The polarizers are then adjusted so that $o$- and $e$- waves propagating through the crystal have equal energies and the waves are combined by the second polarizer. The spectral interference formed in the spectrometer is recorded. The fringe spacing is minimized when the optical axis is perpendicular to the propagation direction.

The phase retardation (fringe spacing) can be varied by adding a reference crystal with known optical axis orientation to the experimental configuration. The interference spacing is minimized when the optical axis of crystal to be measured is parallel to that of the reference crystal.

### 5.3.1 Measurements of group velocity walk-off

The measurements is based on the frequency domain interferometry described above. The intrinsic phase delay between the fast and slow modes of a birefringent medium is used. In the frequency domain, these two temporally separated pulses interfere in the same way that two waves with different frequencies beat in the time domain. The measurement of the modulation period of the interference fringes in the frequency domain gives the group velocity walk-off.

The input spectrum is shown in Fig. 5.4. The power spectrum has nearly a square-top shape with a width $31.60 \, \text{Å}$. Typical frequency domain interference fringes of a $1.9$ cm long KDP type II crystal and a $2.5$ cm thick CDA type I crystal are shown in Fig. 5.5 (a) and (b). In both cases, the incident beam propagation direction is normal to the crystal surface. A least squares method is used to fit the data in Fig. 5.5 using Eq. (5.6) as shown in dashed curve. There are three sources of error in the measurements, the error in measuring the lengths of the crystals, the error in calibrating the spectrometer, and the error in determining the spacing of the interference fringes. The uncertainty of the thickness measurements is $1\%$. The
Fig. 5.4. The spectrum of the incident pulses. The spectrum shape is typical of the combined effects of SPM and GVD. The peak-peak width is $31.60 \pm 0.06 \text{ Å}$. 
Fig. 5.5. Frequency-domain interference fringes of KDP type II and CDA type I crystals. The solid lines are the experimental data, while the dashed lines are the theoretical fit. (a) 1.9-cm thick KDP type II crystal. The fringe spacing is measured to be 15.10±0.05 Å and (b) 2.5-cm thick CDA type I crystal. The fringe spacing is measured to be 6.71±0.02 Å.
calibration was performed using five spectral lines of Rubidium lamp ranging from 1053 nm to 1073 nm. The spectral lines were fitted with Lorentzian line shapes, and the overall uncertainty in the calibration was found to be 0.2%. The least square fit for the interference fringes gives an error of 0.3%. The largest uncertainty comes from the measurement of thickness of the crystals. After taking these three error sources into account, we found that the error in determining the temporal walk-off is about 1%. The group walk-off parameters of several commonly used nonlinear crystals have been measured, the results are listed in Table 5.1. The cut angles which are defined as the angles between the optical axis and the normal of the crystal surface and lengths of the tested crystals are also listed. The last column of Table 5.1 shows the calculated values of the GVW based on the dispersion data of Ref. [9]. The calculated values are very close to the measured ones, as can be seen from Table 5.1. This method can also be used to measure the length of a birefringent crystal if its GVW parameter is known. The last entry in Table 5.1 shows the length of a KDP type I crystal determined by this method using the calculated result of the walk off. The resolution is about 50 μm.

<table>
<thead>
<tr>
<th>Crystal</th>
<th>Cut angle (°)</th>
<th>Length x2 (cm)</th>
<th>Walk-off(a) (ps/cm)</th>
<th>Walk-off(b) (ps/cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CDA I</td>
<td>85</td>
<td>2.50</td>
<td>1.00±0.01</td>
<td>1.01</td>
</tr>
<tr>
<td>KDP*II</td>
<td>53.7</td>
<td>1.50</td>
<td>0.94±0.02</td>
<td>0.97</td>
</tr>
<tr>
<td>KDP II</td>
<td>59.2</td>
<td>1.90</td>
<td>1.35±0.02</td>
<td>1.33</td>
</tr>
<tr>
<td>KDP I</td>
<td>41.2</td>
<td>1.029±0.005</td>
<td></td>
<td>0.79</td>
</tr>
</tbody>
</table>

(a) Measured results. (b) calculated results

It is well known that the refractive index of extraordinary wave is a function of propagation direction, which means that the GVW is also a function of propagation direction. Fig. 5.6 shows the dependence of walk-off on the propagation angle. The angle is measured with respect to the phase-matching angle of the crystal
(KDP-II) in the YZ plane. The scattered triangles are experimental data, the solid curve is the theoretical prediction based on material dispersion. The experimental data fits the theory, with an accuracy of 1%. It should be mentioned that the angle in Fig. 5.6 is the angle inside the crystal, that is, Snell’s law has been used to obtain the internal angle. Since the crystal is cubic, the propagation distance is also a function of angle, which has been taken into account in Fig. 5.6.

5.3.2 Optical axis determination

Frequency domain interferometer can also be used to determine the orientation of the optical axis. The experimental setup is similar to that shown in Fig. 5.1, except the l/2 wave plate before the crystal is replaced by a polarizer which was used to control the polarization direction of the linearly polarized incident pulse to 45° with respect to the x or y axis. The extra delay introduced by the l/2 wave plate was investigated as in follows: First, we recorded the interference spacing when the KDP crystal is normal to the propagation direction. Then we inserted the half wave plate. We record the interference spacing again when the fast axis or the slow axis of the l/2 wave plate was aligned to the optical axis of KDP. If the slow axis is parallel to the optical axis of KDP, the interference spacing is bigger. Fig. 5.7 shows the interference spacing for the different cases. In each case we take ten shots. The closed dot and open dot represent the case when the slow axis and fast axis of l/2 wave plate are aligned parallel to the crystal axis respectively. The solid line are the average of the ten shots. It can be seen that the wave plate changed the interference spacing about 0.018Å. This corresponded to a 36±20 fs predelay.

A 4.6-cm thick KDP crystal was mounted in an rotation mount with 1’ resolution after the plane of its optical axis was located. We measured the interference spacing as a function of propagation angle. The angle is measured with respect to the normal of the crystal. The angle is zero when the propagation direction is perpendicular to the crystal surface and the angle is positive when the crystal is rotated clockwise. Fig. 5.8 shows the 37 Å wide spectra before (long dashed) and after (dashed) the pulse passes through the crystal. A least squares method was used to fit
Fig. 5.6. Dependence of the group-velocity walk off on the propagation direction. The angle is measured with respect to the phase-matching angle of the sample (KDP II, 1.9-cm thick and 59.2° cut)
Fig. 5.7. The influence of the $\lambda/2$ wave plate on the interference fringe spacing. The dashed line is the interference fringe spacing of the KDP crystal which is normal to the propagation direction and no wave plate in the beam path. The close dot and open dot are represent the case when the slow axis and fast axis of $1/2$ wave plate are aligned parallel to the crystal axis respectively. In each case we take ten shots. The corresponding straight line are the average of the ten shots.
the data to determine the $\Delta \omega$ by use of Eq. 5.6 and the results are shown by the solid line. There are several error sources in the measurement. The error of the spectrometer calibration is 0.2%. The least square fit for the interference fringes yields an error of 0.2%. The largest error comes from the shot to shot fluctuation which is approximately 1%. Therefore the overall error in determine the frequency spacing is 1%.

Figure 5.9 shows the dependence of the interference fringe spacing on the propagation angle measured outside the crystal and defined as the external angle. ‘+’ represents the experiment data when crystal optical axis lay on the yz plane in Fig. 5.10 and ‘0’ represents the experiment data when the optical axis is on the xz plane. The geometry is shown in Fig 5.10. In the first case, the interference fringe spacing is sensitive to the crystal rotation. When the crystal was rotated clockwise (positive angle), the interference fringe spacing increased. This means that the group walk-off between o- and e-wave and the angle between wavevector and optical axis were increased. When the crystal was rotated counterclockwise, the phenomena is opposite. We can conclude that the crystal axis lies with an angle to the left of the z-axis (see Fig. 5.10). In the case when the optical axis is on the xz plane, rotating the crystal the same amount as the first case will have less effect on the propagation direction in the crystal to the optical axis. The interference fringes spacing decreased symmetrically around 0° angle. Since the angle between optical axis and wavevector increases, the group velocity walk-off are always increased.

The error bars in Fig. 5.9 represent the error analysis results mentioned above. The solid lines are the least squares fit to the experiment data based on Eq. (5.6) and material dispersion data. The resultant crystal cut angle $\theta_0$ defined as the angle between the optical axis and the normal of the surface (towards y axis) is 82.2°. The standard deviation of this least square fitting is 0.02°. When $\theta_0$ is changed from 82.2° to 82.2°±0.8°, the chi square of the curve fitting will be doubled as shown in Fig. 5.11.
Fig. 5.8. Spectrum of the frequency domain interference. Fig. 2a is the spectrum of the incident pulse applied to the crystal. In fig. 2b, the dashed line is the frequency domain interference fringe and the solid line is the least square fit. The fringe spacing is $4.61 \pm 0.04 \ \text{Å}$. 
Fig. 5.9. Dependence of the interference fringe spacing on the propagation direction. ‘+’ represents the experiment data when the optical axis is in the yz plane. ‘◊’ represents the experiment data when the optical axis is in the xy plane. The angle is the external angle which is measured with respect to the normal of the crystal. The solid line is the theoretical fit.
Fig. 5.10. The propagation of the pulse respective to crystal optical axis. (a) is the case when the crystal optical axis lay on the yz plane (rotation in the plane containing the optical axis). The angle (θ) between wave propagation direction and optical axis in the crystal is, \( \theta = \theta_0 + \Psi_o \) when \( \Psi_o \) is to the right side of the z axis and \( \theta = \theta_0 - \Psi_o \) when \( \Psi_o \) is to left side of the z axis. (b) is the case when the crystal optical axis lay on the xz plane, \( \theta = \arccos (\cos \Psi_o \cos \theta_0) \) (rotation in a plane not containing the optical axis).
Fig. 5.11. The variance of the fit in Fig. 5.9 for different $\theta_0$. 

Normalized error

Cut Angle ($^\circ$)

1.0 1.2 1.4 1.6 1.8 2.0

81.2 81.6 82.0 82.4 82.8 83.2
5.4 CONCLUSION

In conclusion, a new technique based on frequency domain interference has been used to measured the group velocity walk-off and to find the optical axis of a uniaxial crystal. In contrast to the usual interferometric methods used to measure the interference visibility as a function of optical delay between two interference arms, we used two optical axes of the birefringent media as two interference arms and measured the interference fringes in the frequency domain. The experimental setup is simple allowing good accuracy and resolution using the same ultrashort pulses which are to be frequency converted. No curve fitting is needed to find the GVW since the measured modulation period of the fringes is directly related to GVW. The resolution of this technique to determine the optical axis is limited by how parallel the two crystal surfaces are. The angular difference between the two surfaces should be less than the beam divergence.
REFERENCES

Chapter VI
Summary

In this thesis, frequency up-conversion and the related phenomena have been studied experimentally and theoretically. It has been confirmed that various nonlinear process such as the effect of birefringent walk-off, group velocity walk-off and the influence of the self- and cross-phase modulations limit the efficient energy. The spatial and temporal walk-off between the fundamental ordinary (o) and extraordinary (e) wave will limit the maximum useful interacting crystal length for the nonlinear process. The phase modulations which happens at high laser intensities can destroy the phase coherence required for efficient conversion.

The third harmonic conversion of 1.6 ps pulses at 1053 nm were performed with the Chirped-Pulse-Amplification (CPA) laser system using two type II KDP crystals, one for frequency doubling and another for frequency tripling. Conversion efficiency of 60-65% was achieved from frequency doubling and 45-50% was achieved for frequency tripling at input pulse intensities of 50-100 GW/cm².

Our experimental study also showed the importance of the self- and cross-phase modulation effects in the frequency tripling process. The self-phase modulation coefficients in KDP crystals at wavelengths of 1.053 µm, 0.527 µm and 0.351 µm were measured using single beam Z-scan technique. The cross-phase modulation coefficients between 1.053 µm and 0.527 µm were also measured by a two color z-scan. These coefficients are used in the theoretical simulation of frequency conversion of ultrafast pulses.

It is important to know the direction of the crystals axis relative to the propagation direction of the laser beam to optimize the phase matching for harmonic generations. The orientation of the optical axis in the crystal determines the spatial and temporal walk-off between the o- and e- waves. Frequency domain interferometry has been used to measure the group velocity walk-off (GVW) between the o- and e- waves in uniaxial crystals. The dependence of the interference fringe spacing and the walk-off on incident angle with
respect to the birefringent crystal were measured in KDP and the direction of the optical axis was determined.

Ultrashort pulse laser sources are enjoying a period of significant development and application. The short pulse characteristics of these lasers make them important tools for spectroscopy, remote sensing and many other scientific uses. The high intensity of short pulses make them particularly useful in situations where high peak power is required. However, lasers usually operate at fixed wavelengths or have limited tuning over the technologically important spectral regions. Nonlinear frequency conversion, including difference/sum (i.e. harmonic generations), optical parametric oscillators and amplifications, provides unique high power sources with wide tunability.